

Numerical tensor method for atomic and exotic three-particle systems

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In this paper we derive a numerical tensor method to solve the many-body Schrödinger equation for three-particle systems. The Schrödinger equation is first represented in a canonical tensor format using a triple orthogonal Laguerre-based wave function in scaled perimetric coordinates. We demonstrate that the antisymmetry of the wave function can be explicitly included in the tensor method formulation by coupling the indices associated with the coordinates representing the identical fermions, e.g., electrons or muons. It is shown that energies accurate to the picohartree can be achieved in full tensor format in less than 3300 s for atomic and muonic molecular systems. Furthermore, this CPU time is reduced to less than 300 s, without compromising the accuracy of the energy or quality of the wave function, using a low-rank tensor-train decomposition.

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I. INTRODUCTION

Numerical methods that include the motion of all particles can provide high-accuracy energies and wave functions, facilitating new insights and fundamental understanding (see, for example, [1–9] and references therein). The direct solution of the full many-particle Schrödinger equation, without the conventional separation of the electronic and the nuclear motion, makes it possible to approach the nonrelativistic limit arbitrarily close [8]. Furthermore, systems where all the masses are similar can be investigated without further approximation. However, due to the exponential growth of dimension with the number of degrees of freedom, all-particle treatments are limited to few-particle systems. As with conventional quantum chemistry methods, the Schrödinger equation is usually solved as a generalized eigenvalue problem in matrix form, and the size of the matrices involved increases rapidly with basis and system size. This results in calculations becoming increasingly complex and computationally demanding. Therefore, a key challenge is the development of more efficient computational approaches and techniques.

An approach that has the potential to transform our ability to perform high-level quantum chemical calculations is based on exploiting the power of tensors. Tensors are multidimensional generalizations of matrices, and due to the effectiveness of multilinear algebra, researchers in many fields (scientific computing, mathematics, engineering, and more) have been actively working on tensor related problems, developing tensor representations, algorithms, and analysis to solve a range of problems. A key motivation in many fields is to overcome the exponential growth in complexity with the number of de-

grees of freedom, known as the curse of dimensionality. One particular technique to emerge is to express a full tensor using a low-rank tensor decomposition. This mathematical technique approximates a given matrix or tensor by expressing it as the product of two or more matrices or tensors with reduced dimensions. This approach reduces computational complexity (the amount of resource required), making it an invaluable tool for effectively solving high-dimensional problems. Examples of different tensor formats include the canonical format [10], the Tucker format [11], and the tensor-train (TT) format [12] [known as matrix product states (MPSs) in the physics and quantum chemistry communities [13]], all of which offer a compact representation of operators by expressing them as a product of smaller tensors or matrices.

In quantum chemistry (QC), to tackle the exponential growth cost in both computation and memory with increasing electronic excitations in configuration-interaction theory, the density-matrix renormalization-group (DMRG) method has emerged as a method of choice for strongly correlated quantum systems. Implementations of the QC DMRG method, built on the underlying tensor structure, in which both the MPS, representing the variational state, and the Hamiltonian operator converted to a matrix product operator are represented as tensor networks. The DMRG algorithm, pioneered by White [14], is an iterative optimization algorithm used to find the ground states of Hamiltonians with wave functions parametrized in terms of MPSs [15]. The DMRG method effectively addresses the challenge of the exponentially growing computational resources required for such complex systems when a suitable bond dimension (TT rank) is chosen [15–17].

More generally, the principle of solving eigenvalue problems in the TT format is based on the alternating linear scheme (ALS) [18], which utilizes the idea of the DMRG algorithm to optimize each core of the wave function iteratively by solving microequations. Gelß has implemented the ALS algorithm in PYTHON [19], based on the description in [18,20], following the algorithm described in [21]. This implementation has been successfully applied in [22] to solve the time-independent Schrödinger equation for chains of

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coupled excitons and phonons in the tensor-train format and in our previous work [23] where numerical tensor methods (NTMs) were used for all particle calculations on three-body systems using a finite-element approach. In [23] the wave function was approximated as a linear combination of Lagrange polynomials within a truncated domain $\Omega_c = (0, z_c)^3$, where z_c is a chosen cutoff parameter specific to each type of system (atomic, exotic, or molecular). Although the use of low-rank decompositions was shown to significantly reduce computational resources, the main bottleneck was the computation of the factor matrices due to their nonsparsity and the high-precision numerical integration required to achieve results accurate to the nanohartree or better.

In this paper we take a different approach and overcome both of these challenges. Here we derive a NTM to solve the many-body problem using a globally analytical wave function, i.e., we choose the wave function to be defined across the entire domain $\Omega = (0, \infty)^3$, demonstrating the validity of NTMs without discretization. To overcome the computational bottlenecks in [23], we approximate the wave function with Laguerre functions and exploit their orthogonality and recursion relations so that numerical integration is avoided and rounding errors are minimized. The method is applied to a range of three-particle systems. Many-particle Coulomb systems involving elementary particles such as electrons, positrons, muons, protons, and their antimatter counterparts are of topical interest to both the chemistry and physics communities. They are particularly prevalent in precision spectroscopy and particle physics; for example, muon spin resonance spectroscopy is used to probe defect states in semiconductors and ($e^-e^-\mu^+$) has been observed as an impurity [24]. Furthermore, μD_2^+ and μDT^+ are known to catalyze nuclear fusion [25–27]. It will be shown that this Laguerre-based NTM can provide energies for a range of electronic and muonic systems accurate to the picohartree in a vastly reduced computation time.

II. METHOD OF SOLUTION

The motion of three charged point particles with masses m_i and charges Z_i , $i = 1, 2, 3$, is considered. After separating off the center-of-mass motion and taking the origin to be

the location of particle 3, the time-independent Schrödinger equation (SE) $H\psi = E\psi$ in atomic units for three-particle systems in interparticle coordinates is given by (see [7] and references therein)

$$\left(-\frac{1}{2\mu_1}\nabla_1^2 - \frac{1}{2\mu_2}\nabla_2^2 - \frac{1}{m_3}\nabla_1 \cdot \nabla_2 + \frac{Z_1Z_3}{r_1} + \frac{Z_2Z_3}{r_2} + \frac{Z_1Z_2}{r_{12}}\right)\psi = E\psi, \quad \psi \neq 0, \quad (1)$$

where $\mu_i = m_i^{-1} + m_3^{-1}$ for $i \in \{1, 2\}$, and r_1 , r_2 , and r_{12} are the interparticle distances, with r_{12} the distance between the like-charged particles.

The SE is recast in perimetric coordinates [23], where the perimetric coordinates are defined as

$$z_i = r_j + r_k - r_i, \quad (2)$$

where i , j , and k denote cyclic permutation of 1, 2, and 12, respectively, and each variable ranges from 0 to ∞ . In this work we introduce scaled perimetric coordinates, defined as

$$u = az_1, \quad v = az_2, \quad w = 2az_3, \quad (3)$$

where $a > 0$ is a parameter. Following the work of [28,29], the wave function is chosen to be of the form

$$\psi(u, v, w) = e^{-(u+v+w)/2} F(u, v, w), \quad (4)$$

where $F \in \mathcal{C}^2(\Omega)$, i.e., F is at least twice continuously differentiable on $\Omega = (0, \infty)^3$ and will be defined later.

The variational form of (1) is given by

$$I_{\min} = \langle \psi | H - E | \psi \rangle, \quad (5)$$

where H is the Hamiltonian operator, E the energy of the system, and $\langle \cdot \rangle$ the inner product in $L^2(\Omega)$. The transformation from (r_1, r_2, r_{12}) to (u, v, w) results in a Jacobian given by

$$J = \frac{1}{4a^3}(u+v)(2v+w)(w+2u). \quad (6)$$

Definition 1. Let $F \in \mathcal{C}^2(\Omega)$. Choosing ψ as provided in Eq. (4), the variational form (5) of the SE in scaled perimetric coordinate (3) is explicitly given by

$$\begin{aligned} I_{\min} = & \int_0^\infty \int_0^\infty \int_0^\infty \left\{ - \left[\left(\frac{2}{am_2} + \frac{2}{am_3} \right) u^2 v + \left(\frac{2}{am_2} + \frac{2}{am_3} \right) uv^2 + \left(\frac{1}{am_1} + \frac{1}{am_2} \right) u^2 w \right. \right. \\ & + \left. \left(\frac{1}{2am_1} + \frac{1}{2am_2} \right) uw^2 + \frac{2}{am_2} uvw \right] F F_{uu} - \left[\left(\frac{2}{am_1} + \frac{2}{am_3} \right) u^2 v + \left(\frac{2}{am_1} + \frac{2}{am_3} \right) uv^2 \right. \\ & + \left. \left(\frac{1}{am_1} + \frac{1}{am_2} \right) v^2 w + \left(\frac{1}{2am_1} + \frac{1}{2am_2} \right) vw^2 + \frac{2}{am_1} uvw \right] F F_{vv} - \left[\left(\frac{4}{am_1} + \frac{4}{am_3} \right) u^2 w \right. \\ & + \left. \left(\frac{2}{am_1} + \frac{2}{am_3} \right) uw^2 + \left(\frac{2}{am_2} + \frac{2}{am_3} \right) vw^2 + \left(\frac{4}{am_2} + \frac{4}{am_3} \right) v^2 w + \frac{8}{am_3} uvw \right] F F_{ww} \\ & + \left[\frac{2u^2v}{am_2} - \left(\frac{1}{am_1} - \frac{1}{am_2} \right) u^2 w + \frac{2uv^2}{am_2} + \frac{2uvw}{am_2} - \left(\frac{1}{2am_1} - \frac{1}{2am_2} \right) uw^2 + \left(\frac{2}{am_1} + \frac{2}{am_3} \right) u^2 \right. \\ & - \left. \left(\frac{2}{am_2} + \frac{2}{am_3} \right) v^2 - \left(\frac{1}{2am_2} + \frac{1}{2am_1} \right) w^2 - \frac{4uv}{am_2} - \frac{2vw}{am_2} - \frac{2uw}{am_2} \right] F F_u \\ & + \left. \left[\frac{2u^2v}{am_1} - \left(\frac{1}{am_2} - \frac{1}{am_1} \right) v^2 w + \frac{2uv^2}{am_1} + \frac{2uvw}{am_1} - \left(\frac{1}{2am_2} - \frac{1}{2am_1} \right) vw^2 - \left(\frac{2}{am_1} + \frac{2}{am_3} \right) u^2 \right. \right. \end{aligned}$$

$$\begin{aligned}
& + \left(\frac{2}{am_2} + \frac{2}{am_3} \right) v^2 - \left(\frac{1}{2am_2} + \frac{1}{2am_1} \right) w^2 - \frac{4uv}{am_1} - \frac{2vw}{am_1} - \frac{2uw}{am_1} \Big] F F_v \\
& + \left[\frac{8uvw}{am_3} - \frac{8uv}{am_3} + \left(\frac{4}{am_3} + \frac{2}{am_1} \right) u^2 w + \left(\frac{2}{am_3} + \frac{1}{am_1} \right) u w^2 + \left(\frac{4}{am_3} + \frac{2}{am_2} \right) v^2 w + \left(\frac{2}{am_3} + \frac{1}{am_2} \right) v w^2 \right. \\
& - \left(\frac{4}{am_1} + \frac{4}{am_3} \right) u^2 - \left(\frac{4}{am_2} + \frac{4}{am_3} \right) v^2 + \left(\frac{1}{am_1} + \frac{1}{am_2} \right) w^2 - \frac{4vw}{am_3} - \frac{4uw}{am_3} \Big] F F_w \\
& + \left(\frac{4u^2 v}{am_3} + \frac{4uv^2}{am_3} \right) F F_{uv} + \left(\frac{4v^2 w}{am_2} + \frac{2vw^2}{am_2} \right) F F_{vw} + \left(\frac{4u^2 w}{am_1} + \frac{2uw^2}{am_1} \right) F F_{wu} \\
& - \left[\left(\frac{1}{2am_1} + \frac{1}{2am_2} + \frac{2}{am_3} \right) uvw + \left(\frac{1}{2am_1} + \frac{1}{2am_2} \right) u^2 v + \left(\frac{1}{2am_3} + \frac{1}{8am_2} + \frac{1}{8am_1} \right) u w^2 \right. \\
& + \left(\frac{1}{am_3} + \frac{1}{4am_2} + \frac{1}{4am_1} \right) u^2 w + \left(\frac{1}{am_3} + \frac{1}{4am_2} + \frac{1}{4am_1} \right) v^2 w + \left(\frac{1}{2am_1} + \frac{1}{2am_2} \right) v^2 u \\
& + \left(\frac{1}{2am_3} + \frac{1}{8am_2} + \frac{1}{8am_1} \right) v w^2 - \left(\frac{4}{am_3} + \frac{2}{am_1} + \frac{2}{am_2} \right) uv - \left(\frac{2}{am_3} + \frac{1}{am_1} + \frac{1}{am_2} \right) vw \\
& - \left(\frac{2}{am_3} + \frac{1}{am_1} + \frac{1}{am_2} \right) uw - \left(\frac{2}{am_1} + \frac{2}{am_3} \right) u^2 - \left(\frac{2}{am_2} + \frac{2}{am_3} \right) v^2 \Big] F^2 \\
& + \frac{1}{4a^3} (u+v)(2v+w)(w+2u) \left(\frac{2aZ_1 Z_2}{u+v} + \frac{4aZ_2 Z_3}{w+2u} + \frac{4aZ_3 Z_1}{2v+w} - E \right) F^2 \Big\} e^{-(u+v+w)} du dv dw, \tag{7}
\end{aligned}$$

where $F_x = \frac{\partial F}{\partial x}$, $F_{xx} = \frac{\partial^2 F}{\partial x^2}$, and $F_{xy} = \frac{\partial^2 F}{\partial x \partial y}$ for $x, y \in \{u, v, w\}$.

Remark 1. (a) The Jacobian defined in Eq. (6) is included in Eq. (7), and the nonconstant coefficients of F are polynomials depending on u, v , and w .

(b) A variational form equivalent to Eq. (7), obtained by applying integration by parts to reduce the order of differentiation, is also possible. However, we focus on the derivation using Eq. (7) directly, as it requires no intermediate steps.

Numerical tensor methods are based on the principle of low-rank tensor decomposition, which involves representing (or approximating) high-dimensional tensors using several smaller tensors. In this work we start by approximating each operator in canonical tensor format, i.e., we express each operator as the sum of a finite number of rank-1 tensors.

Definition 2 (canonical format of an operator). (a) A tensor operator $\mathcal{G} \in \mathbb{R}^{(m_1 \times n_1) \times \dots \times (m_d \times n_d)}$, of order d , is called a rank-1 tensor if it can be written as the tensor product of d matrices, i.e.,

$$\mathcal{G} = \bigotimes_{i=1}^d \mathcal{G}_{i,:} = \mathcal{G}_{1,:} \otimes \dots \otimes \mathcal{G}_{d,:}, \tag{8}$$

with $\mathcal{G}_{i,:} \in \mathbb{R}^{m_i \times n_i}$ for $i = 1, \dots, d$. In this work we are interested in the case when $m_i = n_i$, i.e., $\mathcal{G}_{i,:} \in \mathbb{R}^{m_i \times m_i}$ for $i = 1, \dots, d$.

(b) A tensor operator $\mathcal{G} \in \mathbb{R}^{(m_1 \times m_1) \times \dots \times (m_d \times m_d)}$ is said to be in the canonical format if it can be written as the sum of rank-1 tensors, i.e.,

$$\mathcal{G} = \sum_{k=1}^R \bigotimes_{i=1}^d \mathcal{G}_{i,:}^{(k)} = \sum_{k=1}^R \mathcal{G}_{1,:}^{(k)} \otimes \dots \otimes \mathcal{G}_{d,:}^{(k)}, \tag{9}$$

with $\mathcal{G}_{i,:}^{(k)} \in \mathbb{R}^{m_i \times m_i}$ for all $k \in \{1, \dots, R\}$ and $i \in \{1, \dots, d\}$, where R is called the canonical rank of the decomposition. Each matrix $\mathcal{G}_{i,:}^{(k)}$ arising in (9) is called a canonical factor [30,31]. In this work we are interested in second- and third-order tensors, i.e., $d \in \{2, 3\}$.

A. Canonical derivation for any three particles

Before deriving the canonical format of each operator, we need to approximate the infinite functional space $\mathcal{C}^2(\Omega)$ to a finite and suitable dimensional functional space. The basis functions are chosen to be normalized Laguerre polynomials $L_n(x)$, where $n \in \mathbb{N}$ is the degree of the polynomial [32]

$$L_n(x) = \sum_{k=0}^n \binom{n}{k} \frac{(-x)^k}{k!} \tag{10}$$

and set

$$V_N := \text{span}\{L_n(x), 0 \leq n \leq N-1\}, \tag{11}$$

where $N-1$ is the highest degree of the Laguerre polynomial. The dimension of V_N is N and $V_N \subset \mathcal{C}^2(0, \infty)$. We define the tensor space

$$\mathcal{V}_N := V_N \otimes V_N \otimes V_N \subset \mathcal{C}^2(\Omega), \tag{12}$$

which is a functional space of dimension N^3 representing the number of terms in the wave function. Here $N-1$ is the highest degree of the Laguerre polynomial in each direction (u, v , or w). The functional space $\mathcal{V}_N$ is then equivalent to

$$\mathcal{V}_N = \text{span}\{L_l(u)L_m(v)L_n(w), 0 \leq l, m, n \leq N-1\}. \tag{13}$$

We approximate the function F as a linear combination of the basis of $\mathcal{V}_N$, i.e.,

$$F(u, v, w) = \sum_{l=0}^{N-1} \sum_{m=0}^{N-1} \sum_{n=0}^{N-1} C_{lmn} L_l(u) L_m(v) L_n(w). \quad (14)$$

Therefore, the wave function is defined as

$$\psi(u, v, w) = e^{-(u+v+w)/2} \sum_{l=0}^{N-1} \sum_{m=0}^{N-1} \sum_{n=0}^{N-1} C_{lmn} L_l(u) L_m(v) L_n(w), \quad (15)$$

where $\mathcal{C} = \{C_{lmn}\}$ is a third-order tensor representing the coefficients of the eigenvector. Finally, to approximate each operator in finite dimensions in a canonical format, we first define the following square canonical factor matrices of size $N \times N$ which arise in the derivation:

$$\begin{aligned} \mathbf{A}_1 &= [\langle \exp(-x) L_i, L_l \rangle]_{i,l=0}^{N-1}, & \mathbf{A}_2 &= [\langle \exp(-x) x L_i, L_l \rangle]_{i,l=0}^{N-1}, & \mathbf{A}_3 &= [\langle \exp(-x) x^2 L_i, L_l \rangle]_{i,l=0}^{N-1}, \\ \mathbf{B}_1 &= [\langle \exp(-x) L_i, L_l' \rangle]_{i,l=0}^{N-1}, & \mathbf{B}_2 &= [\langle \exp(-x) x L_i, L_l' \rangle]_{i,l=0}^{N-1}, & \mathbf{B}_3 &= [\langle \exp(-x) x^2 L_i, L_l' \rangle]_{i,l=0}^{N-1}, \\ \mathbf{C}_1 &= [\langle \exp(-x) L_i, L_l'' \rangle]_{i,l=0}^{N-1}, & \mathbf{C}_2 &= [\langle \exp(-x) x L_i, L_l'' \rangle]_{i,l=0}^{N-1}, & \mathbf{C}_3 &= [\langle \exp(-x) x^2 L_i, L_l'' \rangle]_{i,l=0}^{N-1}. \end{aligned} \quad (16)$$

The matrix elements of the factor matrices (16) are evaluated using the orthogonality property of Laguerre polynomials with respect to the weight e^{-x} , i.e.,

$$\int_0^\infty e^{-x} L_i(x) L_l(x) dx = \delta_{i,l}, \quad (17)$$

and the recursion relations of Laguerre polynomials [32]

$$\begin{aligned} x L_n''(x) &= (x-1) L_n'(x) - n L_n(x), \\ x L_n'(x) &= n L_n(x) - n L_{n-1}(x), \\ x L_n(x) &= -(n+1) L_{n+1}(x) + (2n+1) L_n(x) - n L_{n-1}(x) \end{aligned} \quad (18)$$

for all $x \geq 0$ and $n \in \mathbb{N}$. Thus, the factor matrices are highly sparse and, contrary to [23], where highly accurate numerical integration was required, no numerical integration is necessary.

To derive the tensor format of each operator in the variational form given in Eq. (7), we take advantage of the fact that the scaled perimetric coordinates u , v , and w are independent, ranging 0 to ∞ , and their associated indices l , m , and n in the expansion of F in (15) are also independent, ranging from

0 to $N-1$. Moreover, given that the nonconstant coefficients in the variational form in (7) are polynomial, we can separate the triple integral as a product of three integrals. Specifically, F is expressed as a linear combination of a triple orthogonal product so that each term in the variational form (7) is of the form

$$\begin{aligned} & \int_0^\infty e^{-u} u^a L_i(u) L_l(u) du \int_0^\infty e^{-v} v^b L_j(v) L_m(v) dv \\ & \times \int_0^\infty e^{-w} w^c L_k(w) L_n(w) dw, \end{aligned} \quad (19)$$

with $a, b, c \in \{0, 1, 2\}$. Since the scaled perimetric coordinates are independent of each other, the triple integral of (19) is expressed as the product of integrals in $\mathbb{R}$. In our derivation, we use the factors $\mathbf{A}_{a+1}$, $\mathbf{B}_{a+1}$, and $\mathbf{C}_{a+1}$, as provided in (16), corresponding to cases where no derivative, second-order derivatives, and first-order derivatives of $L_l(x)$ are involved, respectively, for $x \in \{u, v, w\}$.

As an example, the canonical representation arising from the first two terms in Eq. (7), for any two sets of triple indices (l, m, n) and $(i, j, k) \in \{1, \dots, N-1\}^3$, is derived as follows:

$$\begin{aligned} & -\frac{1}{a} \left(\frac{2}{m_2} + \frac{2}{m_3} \right) \int_0^\infty \int_0^\infty \int_0^\infty (u^2 v + uv^2) [L_i(u) L_j(v) L_k(w)] [L_l''(u) L_m(v) L_n(w)] e^{-(u+v+w)} du dv dw \\ & = -\frac{1}{a} \left(\frac{2}{m_2} + \frac{2}{m_3} \right) \int_0^\infty \int_0^\infty \int_0^\infty [e^{-u} u^2 L_i(u) L_l''(u) e^{-v} v L_j(v) L_m(v) e^{-w} L_k(w) L_n(w) \\ & \quad + e^{-u} u L_i(u) L_l''(u) e^{-v} v^2 L_j(v) L_m(v) e^{-w} L_k(w) L_n(w)] du dv dw \\ & = -\frac{1}{a} \left(\frac{2}{m_2} + \frac{2}{m_3} \right) [\mathbf{B}_3(i, l) \mathbf{A}_2(j, m) \mathbf{A}_1(k, n) + \mathbf{B}_2(i, l) \mathbf{A}_3(j, m) \mathbf{A}_1(k, n)]. \end{aligned} \quad (20)$$

By applying the same process to all terms in the variational form given in Eq. (7) and utilizing the distributive property of the tensor product $\otimes$ over addition, we can derive the tensor format for each operator. Specifically, for any matrices $\mathbf{A}$, $\mathbf{B}$, and $\mathbf{C}$, the distributive property is expressed as [33]

$$\underbrace{\mathbf{A} \otimes \mathbf{B} + \mathbf{C} \otimes \mathbf{B}}_{\text{rank 2}} = \underbrace{(\mathbf{A} + \mathbf{C}) \otimes \mathbf{B}}_{\text{rank 1}}. \quad (21)$$

Using this property, the canonical format arising from the kinetic energy operator, which initially had a canonical rank $R = 66$, can be reduced to 36 and has the following form:

$$\begin{aligned}
\mathcal{T} = & -\left(\frac{2}{am_2} + \frac{2}{am_3}\right)\mathbf{B}_3 \otimes \mathbf{A}_2 \otimes \mathbf{A}_1 - \left(\frac{2}{am_2} + \frac{2}{am_3}\right)\mathbf{B}_2 \otimes \mathbf{A}_3 \otimes \mathbf{A}_1 \\
& + \frac{2}{am_1}\mathbf{C}_2 \otimes \mathbf{A}_1 \otimes \mathbf{C}_3 + \frac{4}{am_1}\mathbf{C}_3 \otimes \mathbf{A}_1 \otimes \mathbf{C}_2 - \frac{2}{am_2}\mathbf{B}_2 \otimes \mathbf{A}_2 \otimes \mathbf{A}_2 - \frac{2}{am_2}\mathbf{C}_1 \otimes \mathbf{A}_2 \otimes \mathbf{A}_2 \\
& - \frac{8}{am_3}\mathbf{A}_2 \otimes \mathbf{A}_2 \otimes \mathbf{B}_2 - \frac{8}{am_3}\mathbf{A}_2 \otimes \mathbf{A}_2 \otimes \mathbf{C}_1 + \mathbf{A}_1 \otimes \left[\left(\frac{2}{am_2} + \frac{2}{am_3}\right)\mathbf{A}_3 + \left(\frac{2}{am_2} + \frac{2}{am_3}\right)\mathbf{C}_3\right] \otimes \mathbf{A}_1 \\
& + \mathbf{A}_1 \otimes \left[\frac{2}{am_2}\mathbf{C}_2 + \left(\frac{1}{am_1} + \frac{1}{am_2}\right)\mathbf{A}_1 + \left(\frac{1}{am_2} + \frac{2}{am_3}\right)\mathbf{A}_2\right] \otimes \mathbf{C}_3 \\
& + \mathbf{A}_1 \otimes \left[\frac{4}{am_2}\mathbf{C}_3 + \left(\frac{2}{am_2} + \frac{4}{am_3}\right)\mathbf{A}_3 - \frac{4}{am_3}\mathbf{A}_2\right] \otimes \mathbf{C}_2 + \mathbf{A}_1 \otimes \left[-\left(\frac{1}{8am_1} + \frac{1}{8am_2} + \frac{1}{2am_3}\right)\mathbf{A}_2\right. \\
& \left. - \left(\frac{1}{2am_1} + \frac{1}{2am_2}\right)\mathbf{B}_2 - \left(\frac{1}{2am_1} + \frac{1}{2am_2}\right)\mathbf{C}_1 + \left(\frac{1}{2am_1} - \frac{1}{2am_2}\right)\mathbf{C}_2\right] \otimes \mathbf{A}_3 \\
& + \mathbf{A}_1 \otimes \left[-\frac{2}{am_1}\mathbf{C}_2 + \left(\frac{2}{am_3} + \frac{1}{am_1} + \frac{1}{am_2}\right)\mathbf{A}_2 - \left(\frac{1}{4am_1} + \frac{1}{4am_2} + \frac{1}{am_3}\right)\mathbf{A}_3 - \left(\frac{1}{am_1} + \frac{1}{am_2}\right)\mathbf{B}_3\right. \\
& \left. - \left(\frac{1}{am_2} - \frac{1}{am_1}\right)\mathbf{C}_3\right] \otimes \mathbf{A}_2 + \mathbf{A}_2 \otimes \left(\frac{8}{am_3}\mathbf{A}_2 - \frac{4}{am_3}\mathbf{A}_1\right) \otimes \mathbf{C}_2 \\
& + \mathbf{A}_2 \otimes \left[-\frac{4}{am_1}\mathbf{C}_2 + \frac{2}{am_1}\mathbf{C}_3 + \left(\frac{4}{am_3} + \frac{2}{am_1} + \frac{2}{am_2}\right)\mathbf{A}_2 - \left(\frac{1}{2am_1} + \frac{1}{2am_2}\right)\mathbf{A}_3\right. \\
& \left. - \left(\frac{2}{am_1} + \frac{2}{am_3}\right)\mathbf{B}_3\right] \otimes \mathbf{A}_1 + \mathbf{A}_2 \otimes \left[-\frac{2}{am_1}\mathbf{B}_2 - \frac{2}{am_1}\mathbf{C}_1 + \frac{2}{am_1}\mathbf{C}_2 + \left(\frac{2}{am_3} + \frac{1}{am_1} + \frac{1}{am_2}\right)\mathbf{A}_1\right. \\
& \left. - \left(\frac{1}{2am_1} + \frac{1}{2am_2} + \frac{2}{am_3}\right)\mathbf{A}_2\right] \otimes \mathbf{A}_2 + \mathbf{A}_3 \otimes \left[\frac{2}{am_1}\mathbf{C}_2 + \left(\frac{2}{am_1} + \frac{2}{am_3}\right)\mathbf{A}_1 - \left(\frac{1}{2am_1} + \frac{1}{2am_2}\right)\mathbf{A}_2\right. \\
& \left. - \left(\frac{2}{am_1} + \frac{2}{am_3}\right)\mathbf{B}_2 - \left(\frac{2}{am_1} + \frac{2}{am_3}\right)\mathbf{C}_1\right] \otimes \mathbf{A}_1 + \mathbf{C}_2 \otimes \left(\frac{2}{am_2}\mathbf{A}_2 - \frac{2}{am_2}\mathbf{A}_1\right) \otimes \mathbf{A}_2 \\
& + \mathbf{C}_2 \otimes \left(-\frac{4}{am_2}\mathbf{A}_2 + \frac{2}{am_2}\mathbf{A}_3 + \frac{4}{am_3}\mathbf{C}_3\right) \otimes \mathbf{A}_1 + \mathbf{C}_3 \otimes \left[\frac{2}{am_2}\mathbf{A}_2 + \frac{4}{am_3}\mathbf{C}_2 + \left(\frac{2}{am_1} + \frac{2}{am_3}\right)\mathbf{A}_1\right] \otimes \mathbf{A}_1 \\
& - \left(\frac{2}{am_2} + \frac{2}{am_3}\right)\mathbf{A}_1 \otimes \mathbf{A}_2 \otimes \mathbf{B}_3 - \left(\frac{4}{am_2} + \frac{4}{am_3}\right)\mathbf{A}_1 \otimes \mathbf{A}_3 \otimes \mathbf{B}_2 - \left(\frac{4}{am_2} + \frac{4}{am_3}\right)\mathbf{A}_1 \otimes \mathbf{A}_3 \otimes \mathbf{C}_1 \\
& - \left(\frac{1}{2am_3} + \frac{1}{8am_1} + \frac{1}{8am_2}\right)\mathbf{A}_2 \otimes \mathbf{A}_1 \otimes \mathbf{A}_3 - \left(\frac{2}{am_1} + \frac{2}{am_3}\right)\mathbf{A}_2 \otimes \mathbf{A}_1 \otimes \mathbf{B}_3 \\
& + \left(\frac{2}{am_3} + \frac{1}{am_1}\right)\mathbf{A}_2 \otimes \mathbf{A}_1 \otimes \mathbf{C}_3 - \left(\frac{1}{4am_1} + \frac{1}{4am_2} + \frac{1}{am_3}\right)\mathbf{A}_3 \otimes \mathbf{A}_1 \otimes \mathbf{A}_2 \\
& - \left(\frac{4}{am_1} + \frac{4}{am_3}\right)\mathbf{A}_3 \otimes \mathbf{A}_1 \otimes \mathbf{B}_2 - \left(\frac{4}{am_3} + \frac{4}{am_1}\right)\mathbf{A}_3 \otimes \mathbf{A}_1 \otimes \mathbf{C}_1 + \left(\frac{4}{am_3} + \frac{2}{am_1}\right)\mathbf{A}_3 \otimes \mathbf{A}_1 \otimes \mathbf{C}_2 \\
& - \left(\frac{1}{2am_1} + \frac{1}{2am_2}\right)\mathbf{B}_2 \otimes \mathbf{A}_1 \otimes \mathbf{A}_3 - \left(\frac{1}{am_1} + \frac{1}{am_2}\right)\mathbf{B}_3 \otimes \mathbf{A}_1 \otimes \mathbf{A}_2 - \left(\frac{1}{2am_1} + \frac{1}{2am_2}\right)\mathbf{C}_1 \otimes \mathbf{A}_1 \otimes \mathbf{A}_3 \\
& - \left(\frac{2}{am_2} + \frac{2}{am_3}\right)\mathbf{C}_1 \otimes \mathbf{A}_3 \otimes \mathbf{A}_1 - \left(\frac{1}{2am_1} - \frac{1}{2am_2}\right)\mathbf{C}_2 \otimes \mathbf{A}_1 \otimes \mathbf{A}_3 - \left(\frac{1}{am_1} - \frac{1}{am_2}\right)\mathbf{C}_3 \otimes \mathbf{A}_1 \otimes \mathbf{A}_2. \quad (22)
\end{aligned}$$

The canonical format arising from the Coulomb potential energy operator is given by

$$\begin{aligned}
\mathcal{V} = & \frac{Z_1Z_2 + Z_1Z_3 + Z_2Z_3}{a^2}(\mathbf{A}_1 \otimes \mathbf{A}_2 \otimes \mathbf{A}_2 + \mathbf{A}_2 \otimes \mathbf{A}_1 \otimes \mathbf{A}_2 + 2\mathbf{A}_2 \otimes \mathbf{A}_2 \otimes \mathbf{A}_1) \\
& + \frac{2Z_1Z_3}{a^2}\mathbf{A}_3 \otimes \mathbf{A}_1 \otimes \mathbf{A}_1 + \frac{2Z_2Z_3}{a^2}\mathbf{A}_1 \otimes \mathbf{A}_3 \otimes \mathbf{A}_1 + \frac{Z_1Z_2}{2a^2}\mathbf{A}_1 \otimes \mathbf{A}_1 \otimes \mathbf{A}_3 \quad (23)
\end{aligned}$$

and has a rank of 6. Finally, the canonical format arising from the identity operator is

$$\begin{aligned} \mathcal{S} = & \frac{1}{a^3} \mathbf{A}_2 \otimes \mathbf{A}_2 \otimes \mathbf{A}_2 + \frac{1}{a^3} \mathbf{A}_3 \otimes \mathbf{A}_2 \otimes \mathbf{A}_1 + \frac{1}{4a^3} \mathbf{A}_2 \otimes \mathbf{A}_1 \otimes \mathbf{A}_3 + \frac{1}{2a^3} \mathbf{A}_3 \otimes \mathbf{A}_1 \otimes \mathbf{A}_2 \\ & + \frac{1}{2a^3} \mathbf{A}_1 \otimes \mathbf{A}_3 \otimes \mathbf{A}_2 + \frac{1}{a^3} \mathbf{A}_2 \otimes \mathbf{A}_3 \otimes \mathbf{A}_1 + \frac{1}{4a^3} \mathbf{A}_1 \otimes \mathbf{A}_2 \otimes \mathbf{A}_3 \end{aligned} \quad (24)$$

and has rank 7.

For any three-particle system, in Definition 2, Eq. (9), $d = 3$, $m_i = N$, and $R \in \{36, 6, 7\}$ depending on whether $\mathcal{G}$ arises from the kinetic energy, the Coulomb potential energy, or the overlap operator, respectively.

B. Canonical derivation for systems with two identical fermions

For a three-body system with two identical fermions (e.g., He and H_2^+), the wave function must be antisymmetric to the exchange of identical fermions. Instead of choosing F in $\mathcal{V}_N$ provided in Eq. (12), we need to define a suitable functional space that includes the correct symmetry of the wave function. In this paper we focus on the singlet ground state, but in the derivation we will include the triplet state. For the singlet ground state, the spatial wave function must be symmetric, and for the triplet state it must be antisymmetric, under the interchange of the two fermions. To achieve this, for singlet states we set

$$\begin{aligned} F(u, v, w) = & \sum_{l=0}^{N-1} \sum_{m=0}^l \sum_{n=0}^{N-1} C_{lmn} [L_l(u) L_m(v) \\ & + L_m(u) L_l(v)] L_n(w), \end{aligned} \quad (25)$$

where $m \leq l$, and for triplet states we set

$$\begin{aligned} F(u, v, w) = & \sum_{l=1}^{N-1} \sum_{m=0}^{l-1} \sum_{n=0}^{N-1} C_{lmn} [L_l(u) L_m(v) \\ & - L_m(u) L_l(v)] L_n(w), \end{aligned} \quad (26)$$

where $m < l$ because of the symmetry with respect to the exchange of the identical fermions. The wave function in both cases is approximated as in (4), and we clearly have

$$\psi(u, v, w) = \pm \psi(v, u, w), \quad (27)$$

where the plus sign corresponds to the singlet state and the minus sign corresponds to the triplet state. We now define finite-dimensional functional spaces associated with F , to

allow for (25) or (26), as follows. For singlet states, we define

$$\begin{aligned} V_N^1 := & \text{span}\{L_l(u) L_m(v) + L_m(u) L_l(v) \mid 0 \leq l \leq N-1, \\ & 0 \leq m \leq l\}, \end{aligned} \quad (28)$$

which is a finite-dimensional subvector space of $\mathcal{C}^2(\Omega)$ with dimension $\frac{N(N+1)}{2}$, as $m \leq l$. For triplet states, we define

$$\begin{aligned} V_N^2 := & \text{span}\{L_l(u) L_m(v) - L_m(u) L_l(v) \mid 1 \leq l \leq N-1, \\ & 0 \leq m < l\}, \end{aligned} \quad (29)$$

which is a finite-dimensional subvector space of $\mathcal{C}^2(\Omega)$ of dimension $\frac{N(N-1)}{2}$, as $m < l$.

We then approximate $\mathcal{C}^2(\Omega)$ by

$$\mathcal{V}_{\text{sym}} := V_N^1 \otimes V_N, \quad \mathcal{V}_{\text{antisym}} := V_N^2 \otimes V_N, \quad (30)$$

with V_N defined in (11). They are finite-dimensional spaces of dimension $\frac{N^2(N+1)}{2}$ and $\frac{N^2(N-1)}{2}$ representing the number of terms in the wave function for the singlet and triplet states, respectively. To be more precise,

$$\begin{aligned} \mathcal{V}_{\text{sym}} = & \text{span}\{[L_l(u) L_m(v) + L_m(u) L_l(v)] L_n(w), \\ & 0 \leq l, n \leq N-1, 0 \leq m \leq l\} \end{aligned} \quad (31)$$

and

$$\begin{aligned} \mathcal{V}_{\text{antisym}} = & \text{span}\{[L_l(u) L_m(v) - L_m(u) L_l(v)] L_n(w), \\ & 1 \leq l \leq N-1, 0 \leq m < l, 0 \leq n \leq N-1\}. \end{aligned} \quad (32)$$

In this case, the indices l and m , in the expansion of F in Eqs. (25) and (26), are no longer independent for $L_l(u)$ and $L_m(v)$ as they were in Eq. (15). To address this, rather than working with a third-order tensor operator, we couple u and v and work with a second-order tensor operator where the first mode has a size of $N(N \pm 1)/2$, arising from the sum over l and m and the second mode has a size of N arising from the sum over n in (25) and (26).

To derive an example, we consider the same term we did in our previous example in (20). For any triplet of indices (l, m, n) and (i, j, k) we have

$$\begin{aligned} & - \left(\frac{2}{am_2} + \frac{2}{am_3} \right) \int_0^\infty \int_0^\infty \int_0^\infty (u^2 v + uv^2) [L_i(u) L_j(v) \pm L_j(u) L_i(v)] [L_l(u) L_m(v) \pm L_m(u) L_l(v)] L_k(w) L_n(w) \\ & \times e^{-(u+v+w)} du dv dw \\ = & - \left(\frac{2}{am_2} + \frac{2}{am_3} \right) \int_0^\infty \int_0^\infty \int_0^\infty (u^2 v + uv^2) [L_i(u) L_j(v) \pm L_j(u) L_i(v)] [L_l''(u) L_m(v) \pm L_m''(u) L_l(v)] L_k(w) L_n(w) \\ & \times e^{-(u+v+w)} du dv dw \end{aligned}$$

$$\begin{aligned}
&= - \left(\frac{2}{am_2} + \frac{2}{am_3} \right) [\mathbf{B}_3(i, l) \mathbf{A}_2(j, m) \pm \mathbf{B}_3(i, m) \mathbf{A}_2(j, l) \pm \mathbf{B}_3(j, l) \mathbf{A}_2(i, m) + \mathbf{B}_3(j, m) \mathbf{A}_2(i, l)] \mathbf{A}_1(k, n) \\
&\quad - \left(\frac{2}{am_2} + \frac{2}{am_3} \right) [\mathbf{B}_2(i, l) \mathbf{A}_3(j, m) \pm \mathbf{B}_2(i, m) \mathbf{A}_3(j, l) \pm \mathbf{B}_2(j, l) \mathbf{A}_3(i, m) + \mathbf{B}_2(j, m) \mathbf{A}_3(i, l)] \mathbf{A}_1(k, n).
\end{aligned} \tag{33}$$

Remark 2. (a) As shown in (33), we continue to employ the separation of variables to evaluate integrals in one dimension with respect to u , v , and w . However, unlike the previous section and the series of works where NTMs have been applied and indices are associated with the variables in a one-to-one manner [23,30,34,35], in this formulation it is not the case.

(b) To be able to construct its canonical format, we need to unfold the canonical factors associated with u and v so that the Hamiltonian becomes a product of a second-order tensor and a first-order tensor, rather than a third-order tensor as previously. To do this, we introduce the operator symbol $\square$ needed to make that unfold.

Definition 3. Let $\mathbf{A}$ and $\mathbf{B}$ in $\mathbb{R}^{N \times N}$ be any of the matrices defined in (16). We set $\mathbf{A} \square \mathbf{B} \in \mathbb{R}^{\frac{N(N+1)}{2} \times \frac{N(N+1)}{2}}$ to be the association of the first and second factors needed to include fermionic exchange in the formulation (27), defined by

$$\begin{aligned}
\mathbf{A} \square \mathbf{B}[i, j; l, m] &= \mathbf{A}(i, l) \mathbf{B}(j, m) \pm \mathbf{A}(i, m) \mathbf{B}(j, l) \\
&\quad \pm \mathbf{A}(j, l) \mathbf{B}(i, m) + \mathbf{A}(j, m) \mathbf{B}(i, l).
\end{aligned} \tag{34}$$

The plus or minus sign depends on whether the wave function is symmetric or antisymmetric, respectively.

Remark 3. (a) Using (34), the canonical format of the first term in (7) given in (33) is

$$-\frac{1}{a} \left(\frac{2}{m_2} + \frac{2}{m_3} \right) (\mathbf{B}_3 \square \mathbf{A}_2 + \mathbf{B}_2 \square \mathbf{A}_3) \otimes \mathbf{A}_1. \tag{35}$$

(b) Given that each matrix element of $\mathbf{A}$ and $\mathbf{B}$ appearing in (34) evaluates to a real number, the product of matrix elements in each term commute and therefore

$$\mathbf{A} \square \mathbf{B}[i, j; l, m] = \mathbf{B} \square \mathbf{A}[i, j; l, m] \forall i, j, l, m.$$

We then have the property

$$\mathbf{A} \square \mathbf{B} = \mathbf{B} \square \mathbf{A}. \tag{36}$$

Combining the distributive property of the tensor product shown in Eq. (21) and the commutation property of Eq. (36), we obtain the canonical format arising from each operator. These properties are exploited in order to reduce the rank R of the operator. The canonical format arising from the kinetic energy operator can be written as

$$\begin{aligned}
\mathcal{T} &= \mathbf{T}_1 \otimes \mathbf{A}_1 + \mathbf{T}_2 \otimes \mathbf{A}_2 + \mathbf{T}_3 \otimes \mathbf{A}_3 + \mathbf{T}_4 \otimes \mathbf{B}_2 + \mathbf{T}_5 \otimes \mathbf{B}_3 \\
&\quad + \mathbf{T}_6 \otimes \mathbf{C}_1 + \mathbf{T}_7 \otimes \mathbf{C}_2 + \mathbf{T}_8 \otimes \mathbf{C}_3
\end{aligned} \tag{37}$$

and has a rank of 8. The matrices $\mathbf{T}_i$ for $i \in \{1, \dots, 8\}$ are sums of matrices of the general form $\mathbf{A} \square \mathbf{B}$ and are defined as follows:

$$\begin{aligned}
\mathbf{T}_1 &= \left(\frac{2}{am_1} + \frac{2}{am_2} \right) \mathbf{A}_2 \square \mathbf{C}_3 + \left(\frac{2}{am_1} + \frac{2}{am_2} \right) \mathbf{A}_3 \square \mathbf{C}_2 - \left(\frac{4}{am_3} + \frac{2}{am_1} + \frac{2}{am_2} \right) \mathbf{C}_1 \square \mathbf{A}_3 \\
&\quad + \left(\frac{4}{am_3} + \frac{2}{am_1} + \frac{2}{am_2} \right) \mathbf{C}_3 \square \mathbf{A}_1 - \left(\frac{4}{am_1} + \frac{4}{am_2} \right) \mathbf{C}_2 \square \mathbf{A}_2 + \left(\frac{4}{am_3} + \frac{2}{am_1} + \frac{2}{am_2} \right) \mathbf{A}_3 \square \mathbf{A}_1 \\
&\quad - \left(\frac{4}{am_3} + \frac{2}{am_1} + \frac{2}{am_2} \right) \mathbf{B}_2 \square \mathbf{A}_3 - \left(\frac{4}{am_3} + \frac{2}{am_1} + \frac{2}{am_2} \right) \mathbf{B}_3 \square \mathbf{A}_2 - \left(\frac{1}{am_1} + \frac{1}{am_2} \right) \mathbf{A}_2 \square \mathbf{A}_3 \\
&\quad + \left(\frac{4}{am_3} + \frac{2}{am_1} + \frac{2}{am_2} \right) \mathbf{A}_2 \square \mathbf{A}_2 + \frac{8}{am_3} \mathbf{C}_2 \square \mathbf{C}_3, \\
\mathbf{T}_2 &= - \left(\frac{2}{am_1} + \frac{2}{am_2} \right) \mathbf{A}_1 \square \mathbf{B}_3 - \left(\frac{2}{am_1} + \frac{2}{am_2} \right) \mathbf{C}_2 \square \mathbf{A}_1 - \left(\frac{2}{am_1} + \frac{2}{am_2} \right) \mathbf{C}_1 \square \mathbf{A}_2 \\
&\quad + \left(\frac{2}{am_1} + \frac{2}{am_2} \right) \mathbf{C}_2 \square \mathbf{A}_2 - \left(\frac{2}{am_1} + \frac{2}{am_2} \right) \mathbf{B}_2 \square \mathbf{A}_2 + \left(\frac{4}{am_3} + \frac{2}{am_1} + \frac{2}{am_2} \right) \mathbf{A}_1 \square \mathbf{A}_2 \\
&\quad - \left(\frac{2}{am_3} + \frac{1}{2am_1} + \frac{1}{2am_2} \right) \mathbf{A}_2 \square \mathbf{A}_2 - \left(\frac{2}{am_3} + \frac{1}{2am_1} + \frac{1}{2am_2} \right) \mathbf{A}_3 \square \mathbf{A}_1, \\
\mathbf{T}_3 &= - \left(\frac{1}{am_1} + \frac{1}{am_2} \right) \mathbf{C}_1 \square \mathbf{A}_1 - \left(\frac{1}{am_1} + \frac{1}{am_2} \right) \mathbf{B}_2 \square \mathbf{A}_1 - \left(\frac{1}{4am_1} + \frac{1}{4am_2} + \frac{1}{am_3} \right) \mathbf{A}_1 \square \mathbf{A}_2, \\
\mathbf{T}_4 &= - \left(\frac{8}{am_3} + \frac{4}{am_1} + \frac{4}{am_2} \right) \mathbf{A}_3 \square \mathbf{A}_1 - \frac{8}{am_3} \mathbf{A}_2 \square \mathbf{A}_2, \\
\mathbf{T}_5 &= \left(\frac{4}{am_3} + \frac{2}{am_1} + \frac{2}{am_2} \right) \mathbf{A}_1 \square \mathbf{A}_2,
\end{aligned}$$

$$\begin{aligned}
\mathbf{T}_6 &= \left(\frac{8}{am_3} + \frac{4}{am_1} + \frac{4}{am_2} \right) \mathbf{A}_1 \square \mathbf{A}_3 - \frac{8}{am_3} \mathbf{A}_2 \square \mathbf{A}_2, \\
\mathbf{T}_7 &= \frac{8}{am_3} \mathbf{A}_2 \square \mathbf{A}_2 - \frac{8}{am_3} \mathbf{A}_2 \square \mathbf{A}_1 + \left(\frac{8}{am_3} + \frac{2}{am_1} + \frac{2}{am_2} \right) \mathbf{A}_1 \square \mathbf{A}_3 + \left(\frac{4}{am_1} + \frac{4}{am_2} \right) \mathbf{C}_3 \square \mathbf{A}_1, \\
\mathbf{T}_8 &= \left(\frac{4}{am_3} + \frac{1}{am_1} + \frac{1}{am_2} \right) \mathbf{A}_1 \square \mathbf{A}_2 + \left(\frac{2}{am_1} + \frac{2}{am_2} \right) \mathbf{C}_2 \square \mathbf{A}_1 + \left(\frac{1}{am_1} + \frac{1}{am_2} \right) \mathbf{A}_1 \square \mathbf{A}_1.
\end{aligned}$$

The canonical format arising from the Coulomb potential energy has a rank of 3 and is given by

$$\begin{aligned}
\mathcal{V} &= \frac{2}{a^2} [(Z_1 Z_2 + Z_2 Z_3 + Z_3 Z_1) \mathbf{A}_2 \square \mathbf{A}_2 + (Z_2 Z_3 + Z_3 Z_1) \mathbf{A}_3 \square \mathbf{A}_1] \\
&\quad \otimes \mathbf{A}_1 + \frac{2}{a^2} (Z_1 Z_2 + Z_2 Z_3 + Z_3 Z_1) (\mathbf{A}_1 \square \mathbf{A}_2) \\
&\quad \otimes \mathbf{A}_2 + \frac{1}{2a^2} Z_1 Z_2 (\mathbf{A}_1 \square \mathbf{A}_1) \otimes \mathbf{A}_3.
\end{aligned} \tag{38}$$

The canonical format arising from the identity operator is also of rank 3 and is given by

$$\begin{aligned}
\mathcal{S} &= \frac{2}{a^3} (\mathbf{A}_2 \square \mathbf{A}_3) \otimes \mathbf{A}_1 + \frac{1}{a^3} (\mathbf{A}_2 \square \mathbf{A}_2 + \mathbf{A}_3 \square \mathbf{A}_1) \otimes \mathbf{A}_2 \\
&\quad + \frac{1}{2a^3} (\mathbf{A}_1 \square \mathbf{A}_2) \otimes \mathbf{A}_3.
\end{aligned} \tag{39}$$

Remark 4. (a) For systems having two identical fermions, in Definition 2 we have $d = 2$, $m_1 = N(N \pm 1)/2$, $m_2 = N$, and $R \in \{8, 3, 3\}$ depending on whether $\mathcal{G}$ is the kinetic energy, the Coulomb potential energy, or the overlap operator, respectively.

(b) The full format associated with each operator is straightforwardly obtained by evaluating the tensor product of its canonical format.

C. Tensor-train decomposition and the ALS algorithm

For systems with three different particles, an operator in canonical format is stored with a complexity of $O(3RN^2)$ [20], where R represents its canonical rank. While this format is efficient for representing high-dimensional tensors, it has a significant drawback: Optimization problems such as the eigenvalue problem (EVP) and systems of linear equations can be ill-posed, meaning that the best approximation of the solution may not exist within this format [36]. This limitation motivates us to rewrite each operator in a more suitable tensor format that allows us to solve the SE efficiently.

The TT format is a good candidate to address this issue [12,20,23,37]. The idea behind the TT format is to decompose a d -order tensor $\mathcal{G}$ into d component tensors $\mathcal{G}^{(i)}$, providing a more stable and tractable representation.

Definition 4. Given a tensor operator $\mathcal{G} \in \mathbb{R}^{(m_1 \times m_1) \times \dots \times (m_d \times m_d)}$, the TT decomposition of $\mathcal{G}$ has d factor tensors $\mathcal{G}^{(k)} \in \mathbb{R}^{R_{k-1} \times (m_k, m_k) \times R_k}$, where $(R_1, \dots, R_{d-1}) \in \mathbb{N}^{d-1}$ and $R_0 = R_d = 1$. The factor tensors $\mathcal{G}^{(k)}$ are known as TT cores.

A tensor operator $\mathcal{G} \in \mathbb{R}^{(m_1 \times m_1) \times \dots \times (m_d \times m_d)}$ is said to be in the TT format if

$$\mathcal{G} = \mathcal{G}^{(1)} \otimes \dots \otimes \mathcal{G}^{(d)}. \tag{40}$$

A tensor $\mathcal{T} \in \mathbb{R}^{m_1 \times \dots \times m_d}$ is said to be in the TT format if

$$\mathcal{T} = \mathcal{T}^{(1)} \otimes \dots \otimes \mathcal{T}^{(d)}. \tag{41}$$

Remark 5. The tensor product operation between TT cores in (40) and (41) can be regarded as a generalization of the standard matrix multiplication, where the cores contain matrices (of size $m_k \times m_k$ for an operator) or vectors (of size $m_k \times 1$ for eigenvectors) as elements instead of scalar values. Using standard matrix multiplication, we compute the tensor products of the corresponding elements and then sum over the columns and rows, respectively.

In this work, as in [23], we choose the TT ranks of the operators $R_1, \dots, R_{d-1} = R$ so that the TT cores are obtained from the canonical format straightforwardly. For example, the TT cores obtained from the canonical format of $\mathcal{S}$ can be written as (40) for systems with three different particles, from (24), as

$$\begin{aligned}
\mathcal{S}^{(1)} &= \frac{1}{4a^3} [4\mathbf{A}_2 \ 4\mathbf{A}_3 \ \mathbf{A}_2 \ 2\mathbf{A}_3 \ 2\mathbf{A}_1 \ 4\mathbf{A}_2 \ \mathbf{A}_1], \\
\mathcal{S}^{(2)} &= \text{diag}\{\mathbf{A}_2, \mathbf{A}_2, \mathbf{A}_1, \mathbf{A}_1, \mathbf{A}_3, \mathbf{A}_3, \mathbf{A}_2\}, \\
\mathcal{S}^{(3)} &= [\mathbf{A}_2 \ \mathbf{A}_1 \ \mathbf{A}_3 \ \mathbf{A}_2 \ \mathbf{A}_2 \ \mathbf{A}_1 \ \mathbf{A}_3]^T.
\end{aligned} \tag{42}$$

Remark 6. The definition of TT cores from the canonical format of an operator is not unique. For example, in systems with three different particles, a more compact representation of the TT cores of $\mathcal{S}$ can be expressed as

$$\begin{aligned}
\mathcal{S}^{(1)} &= \frac{1}{4a^3} [\mathbf{A}_1 \ \mathbf{A}_2 \ \mathbf{A}_3], \quad \mathcal{S}^{(2)} = \begin{bmatrix} \mathbf{0} & 2\mathbf{A}_3 & \mathbf{A}_2 \\ 4\mathbf{A}_3 & 4\mathbf{A}_2 & \mathbf{A}_1 \\ 4\mathbf{A}_2 & 2\mathbf{A}_1 & \mathbf{0} \end{bmatrix}, \\
\mathcal{S}^{(3)} &= \begin{bmatrix} \mathbf{A}_1 \\ \mathbf{A}_2 \\ \mathbf{A}_3 \end{bmatrix}.
\end{aligned} \tag{43}$$

We clearly have

$$\mathcal{S} = \mathcal{S}^{(1)} \otimes \mathcal{S}^{(2)} \otimes \mathcal{S}^{(3)}.$$

However, in our implementation, we use the simpler derivation of the TT cores defined in (42) as the size of the microsystems needed to solve the EVP in the TT format using the ALS does not depend on the rank R of the operator.

For systems having two identical fermions, from the canonical representation of $\mathcal{S}$ in (39), we can define TT cores as

$$\begin{aligned}
\mathcal{S}^{(1)} &= \frac{1}{a^3} [2\mathbf{A}_2 \square \mathbf{A}_3 \quad \mathbf{A}_2 \square \mathbf{A}_2 + \mathbf{A}_3 \square \mathbf{A}_1 \quad \frac{1}{2} \mathbf{A}_1 \square \mathbf{A}_2], \\
\mathcal{S}^{(2)} &= \begin{bmatrix} \mathbf{A}_1 \\ \mathbf{A}_2 \\ \mathbf{A}_3 \end{bmatrix}.
\end{aligned} \tag{44}$$

The TT cores of the wave function have the form provided in (41). For systems without identical fermions, each core of the

eigenvector $\mathcal{C}$ has the shape

$$\mathcal{C}^{(1)} \in \mathbb{R}^{1 \times N \times r}, \quad \mathcal{C}^{(2)} \in \mathbb{R}^{r \times N \times r}, \quad \mathcal{C}^{(3)} \in \mathbb{R}^{r \times N \times 1}, \quad (45)$$

whereas for systems with identical fermions, the TT cores of the eigenvector $\mathcal{C}$ have the shape

$$\mathcal{C}^{(1)} \in \mathbb{R}^{1 \times N(N \pm 1)/2 \times r}, \quad \mathcal{C}^{(2)} \in \mathbb{R}^{r \times N \times 1}. \quad (46)$$

In both cases, r represents the user-defined rank of the eigenvector, which is not known *a priori*, and is weakly bounded by N [37].

To solve the EVP in the TT format, we begin with an initial guess for the TT cores of $\mathcal{C}$, ensuring that the shapes of $\mathcal{C}^{(i)}$ are defined as in (45) and (46), with r specified. The ALS algorithm then optimizes each TT core $\mathcal{C}^{(i)}$ sequentially, first from left to right (forward sweep) and then from right to left (backward sweep), while keeping the other cores $\mathcal{C}^{(j)}$ fixed during each step. Each cycle of a complete forward sweep followed by a backward sweep is called an iteration. For systems with three different particles, a single sweep involves solving three EVPs corresponding to square matrices of size rN , r^2N , and rN , respectively [38]. In contrast, for systems with two identical fermions, two EVPs of sizes $rN(N \pm 1)/2$ and rN are required. This process of alternating sweeps continues until the desired accuracy is reached. During each optimization, a retraction operator is constructed to efficiently transition from higher-dimensional to lower-dimensional spaces. Further details on the ALS algorithm can be found in [18,21] and the references therein.

III. IMPLEMENTATION

In this work we consider only the ground state of each system. The factor matrices in (16), required to obtain the canonical tensor format of each operator, are evaluated by exploiting the Laguerre recursion relations and their orthogonality property. This ensures that matrix entries are integers and eliminates the need for numerical integration, which minimizes rounding errors. For systems with two identical fermions, an additional intermediate step is necessary to construct the unfolding as described in Definition 3. In both cases, the resulting factors are highly sparse because of the orthogonality property of the Laguerre functions. From these factor matrices, the full-format operators are constructed using the `sparse.kron` function from the `SCIPY` package. This function, designed for sparse matrices, efficiently executes the tensor Kronecker product, making the operation faster compared to `numpy.kron`. The resulting Hamiltonian and identity operators are square matrices, very sparse, and have the dimension of either N^3 or $N^2(N + 1)/2$, depending on whether the system has three different particles or two identical fermions. The resulting generalized eigenvalue problem is solved in full tensor format using the built-in `eigs` command in the `scipy.sparse.linalg` toolbox in `PYTHON`. The number of states (or eigenvalues) is set to 1 for the ground state, and the accuracy for solving the eigenvalue problem is set to $\epsilon = 10^{-16}$. There are a few schemes to obtain the TT representation. Using Algorithm 1 outlined in [12] or Fig. 4 in [37], it is possible to obtain a minimal-rank TT representation using a sequence of singular value decompositions on the operator in the full format to obtain an approximation of

the TT cores $\mathcal{G}^{(i)}$ for $i \in \{1, \dots, d\}$. However, as shown in Sec. II C, by first deriving the exact canonical format of each operator, converting to the TT format using the canonical rank of the operator is straightforward and does not involve any additional factorization or decomposition procedures [20]. Furthermore, it provides the exact TT cores of the operator directly from the canonical format; therefore, this is the approach that we have taken.

To solve the eigenvalue problem in the TT format, we use the ALS implementation available within the TT toolbox [19]. The convergence threshold ϵ , representing the accuracy of the energy, is set to 10^{-13} and the maximum number of iterations (or sweeps) is set to $\gamma = 100$. Indeed, setting the convergence threshold to less than 10^{-13} results in the calculation requiring the maximum number of iterations even when $\gamma > 100$ and numerical errors start to arise. Therefore, setting $\epsilon = 10^{-13}$ ensures that the energy is accurate to this level as the number of iterations is less than 100 and the energy remains variational. Given that the operators are sparse, we utilize the built-in `eigs` command to solve the micro-EVPs during the ALS sweeps. The optimal rank of the eigenvector is determined through a series of calculations whereby the rank is systematically increased until the accuracy achieved is that of the full format for a given N value.

Two versions of the code have been implemented, each corresponding to a specific choice of the parameter a in Eq. (3). In the first implementation, we set $a = 1$ such that $u + v + w = 2(r_1 + r_2)$, which leads to the generalized eigenvalue problem

$$(\mathcal{T} + \mathcal{V})\mathcal{C} = E\mathcal{S}\mathcal{C}. \quad (47)$$

In the second implementation, we set a equal to an energy parameter, specifically $a = \sqrt{-E}$, following [28,29], so that the right-hand side of (47) becomes $-a^2\mathcal{S}\mathcal{C}$. The generalized EVP is then multiplied by a^2 to eliminate the factor $\frac{1}{a}$ in $\mathcal{T}$ (22), the factor $\frac{1}{a^2}$ in $\mathcal{V}$ (23), and the factor $\frac{1}{a^3}$ in $\mathcal{S}$ (24), resulting in a new EVP of the form

$$\mathcal{V}\mathcal{C} = -a(\mathcal{T} + \mathcal{S})\mathcal{C}. \quad (48)$$

The CPU times presented in the results section represent the total time required for the entire calculation, including the computation of factor matrices (16) (using orthogonality and recursion relations), the construction of the canonical format, and the conversion to the full tensor format (via tensor product evaluation) and/or TT format, as well as the solving of the eigenvalue problem. All numerical calculations were performed using double precision in `PYTHON` 3.10.9 on a standard Precision 5820 Tower DELL computer.

IV. RESULTS AND DISCUSSION

A. Full format

To validate the formulations presented in Secs. II A and II B, calculations are performed on systems with and without two identical fermions. The latest CODATA mass values were used in these calculations and are provided in Table I. The electron, positron, muon, proton, and triton are fermions (half-integer spin particles) and deuteron and helion are bosons (integer spin particles).

TABLE I. Particle masses (from [39]) in atomic units where $m_e = m_{\text{positron}} = 1$.

Particle	Mass
${}^4\text{He}^{2+}$	7294.299 541 71
p	1836.152 673 426
d	3670.482 967 655
t	5496.921 535 51
μ	206.768 282 7

Table II provides the ground-state energies, calculated using the canonically derived full format. A range of systems, including atomic, muonic, exotic, and molecular, is considered. As in our previous work [23], the limiting factor in the calculations using the full format is N . Previously, the limit was $N = 42$. In this work we are able to go to larger N values due to the sparsity of the factor matrices. The maximum value for systems with identical fermions is $N = 85$. However, to obtain picohartree accuracy for atomic and exotic systems it was not necessary to go to the maximum value; $N = 65$ was sufficient, and beyond picohartree accuracy, rounding errors started to creep in. In the case of the molecular systems, at $N = 85$ the energy of H_2^+ with $a = 1$ is $-0.597\,13$ hartree and with $a = \sqrt{-E}$ it is -0.5971 hartree. However, this increases the CPU time significantly to more than 14 000 s. For systems without identical fermions the maximum value is $N = 45$. Therefore, in Table II, all calculations with identical fermions were performed with $N = 65$ and for systems without identical fermions $N = 45$.

The mass ratio between the heavier like-charged particle and the uniquely charged particle is also provided in Table II. This reveals that, in general, as the mass ratio becomes larger, the accuracy of the energy decreases. For systems with $m_2/m_3 < 10^2$, energies accurate to at least the picohartree were achievable in less than 20 min for atomic systems and 60 min for muonic molecular systems, demonstrating a significant improvement in the CPU time compared to our previous implementation using a finite-element

approach in combination with a low-rank tensor-decomposition ansatz [23]. Both formulations, i.e., using unscaled coordinates ($a = 1$) or using scaled coordinates ($a = \sqrt{-E}$) following the Pekeris approach to ensure the correct asymptotic behavior of the wave function for atoms [28], provide excellent energies for a range of atomic and exotic systems. We attribute the success of the unscaled coordinates to the large number of linear parameters in the wave function. For molecular systems with large mass ratios $m_2/m_3 > 10^3$, the rate of convergence is poor. This is not unexpected, as to obtain high-accuracy energies for molecular electronic systems it is necessary to ensure that the description of the interparticle distribution of the two heavier particles in the asymptotic region of the wave function is adequate [3,40].

B. Tensor train

In this section we explore the utility of the low-rank tensor-train decomposition in our goal to achieve high-accuracy energies and wave functions as a low computational cost. The ranks of the operators in the TT format are fixed at the canonical rank R as discussed in Sec. II C. To determine a suitable TT rank r for the wave function, the behavior of the relative errors in the energies and wave functions with respect to the full format are analyzed. The relative error in the energy ε is defined as

$$\varepsilon = \frac{|E_{\text{full}} - E_{\text{TT}}^r|}{E_{\text{full}}}, \quad (49)$$

where E_{full} is the energy calculated in the full tensor format and E_{TT}^r is the energy computed in the TT format with a specific rank r . To ensure an accurate choice of r , we select a rank such that $\varepsilon \leq 10^{-13}$, as the target precision for the energy in the calculations is set to 10^{-13} . To select an appropriate value of the TT rank r for the eigenvector, the relative error in the TT eigenvector, denoted by $\mathcal{C}_{\text{TT}}^r$, is compared with the full format eigenvector, denoted by $\mathcal{C}_{\text{full}}$. The error is defined as the relative difference of the Frobenius norm ε_1 [L^2 energy

TABLE II. Ground-state energy calculated using the full-tensor format using $a = \sqrt{-E}$ and $a = 1$ for systems with two identical fermions $N = 65$ and for systems without two identical fermions $N = 45$. The energy is reported in atomic units when an electron is the lightest particle and muon atomic units when a muon is the lightest particle.

System	Triad $\{m_1, m_2, m_3\}$	Mass ratio m_2/m_3 ($m_1 \leq m_2$)	Energy $a = \sqrt{-E}$	CPU time (s)	Energy $a = 1$	CPU time (s)
He	$(e^-, e^-, {}^4\text{He}^{2+})$	1.37×10^{-4}	-2.903 304 557 729 7	841	-2.903 304 557 72	1048
H^-	(e^-, e^-, p^+)	5.45×10^{-4}	-0.527 445 881 109 5	947	-0.527 445 881 109 5	1009
Mu^-	(e^-, e^-, μ^+)	4.84×10^{-3}	-0.525 054 806 511 8	1014	-0.525 054 806 511	1000
$\mu\mu\text{He}$	$(\mu^-, \mu^-, {}^4\text{He}^{2+})$	2.84×10^{-2}	-2.819 764 147 532	802	-2.819 764 147 5	834
Ps^-	(e^-, e^-, e^+)	1.00	-0.262 005 070 232 9	948	-0.262 005 070 232 9	1002
μH_2^+	(p^+, p^+, μ^-)	8.88	-0.494 386 812 813 4	956	-0.494 386 812 813 4	998
μHD^+	(p^+, d^+, μ^-)	1.78×10^1	-0.512 711 790 529 6	2021	-0.512 711 790 529 6	2071
μDT^+	(d^+, t^+, μ^-)	2.66×10^1	-0.538 594 970 860 5	3282	-0.538 594 970 860 5	3685
Mu_2^+	(μ^+, μ^+, e^-)	2.07×10^2	-0.585 126 098 1	957	-0.585 126 098 1	1000
H_2^+	(p^+, p^+, e^-)	1.84×10^3	-0.597	1003	-0.5971	965
HD^+	(p^+, d^+, e^-)	3.67×10^3	-0.597	2431	-0.597	2286
HT^+	(p^+, t^+, e^-)	5.50×10^3	-0.59	1831	-0.59	1811

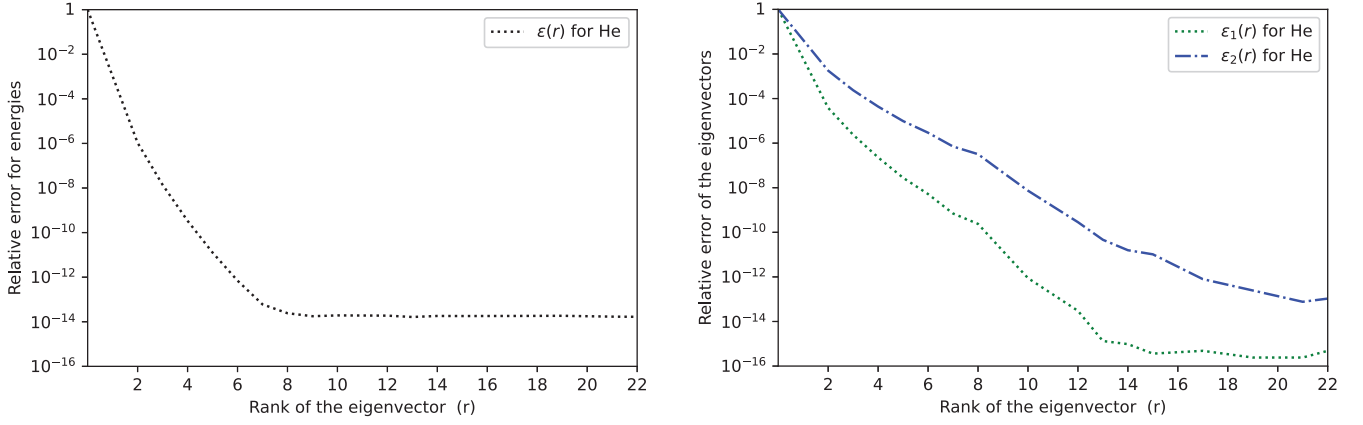


FIG. 1. Decay of the TT errors in energy ε and in the eigenvectors ε_1 and ε_2 for He as a function of rank r . The calculations are performed using $N = 65$ with $a = \sqrt{-E}$.

norm (see [35], p. 45)] and the relative error in the Frobenius norm ε_2 ,

$$\varepsilon_1(r) = \frac{\|\mathcal{C}_{\text{full}} - \mathcal{C}_{\text{TT}}^r\|}{\|\mathcal{C}_{\text{full}}\|}, \quad \varepsilon_2(r) = \frac{\|\mathcal{C}_{\text{full}} - \mathcal{C}_{\text{TT}}^r\|}{\|\mathcal{C}_{\text{full}}\|}, \quad (50)$$

where r is the rank for the eigenvector in the TT format, $r = 1, 2, \dots, N$, and $\|\mathcal{C}\|$ is the Frobenius norm of the tensor $\mathcal{C}$.

The behavior of the error in the energy and in the wave function of the helium atom as a function of r is shown in Fig. 1 and the behavior of the error in the energy and in the wave function of the molecular muonic system μHD^+ is provided in Fig. 2.

For both systems ^4He and μHD^+ , the error in the energy stabilizes around 10^{-13} , which matches the accuracy of the energy calculated using the full format (see Table II). The analysis indicates that $r \geq 9$ for the atomic system and $r \geq 10$ for the muonic molecular system is sufficient to achieve energies accurate to that obtained using the full format. The same approach has been applied to other systems to determine the optimal ranks, i.e., the smallest rank that gives the best possible energy. It was found that, in general, a rank of 9 was sufficient for systems with two identical fermions and a rank of 10 was sufficient for systems with three different particles.

The TT energies for systems that achieved at least pico-hartree accuracy using the full format with $a = \sqrt{-E}$ are summarized in Table III, along with the corresponding results for calculations performed with $a = 1$. It is shown that, regardless of the formulation used in terms of the value of a and the type of system, the TT format can provide energies as accurate as the full format at a fraction of the computation cost.

For all types of systems and both formulations, the tensor-train calculations are at least three times faster than those in the full format. This highlights a key strength of the low-rank decomposition and the ALS algorithm: Rather than solving a large eigenvalue problem, of size $N^2(N+1)/2$ for systems with two identical fermions or N^3 for systems without two identical fermions, in a single step using classical eigensolvers, the ALS efficiently decomposes the task into smaller problems. The ALS algorithm performs a number of iterations γ until the desired accuracy (10^{-13}) in the energy is reached. Thus, in this work, the ALS algorithm sequentially solves γ EVPs of size $rN(N+1)/2$ for the optimization of $\mathcal{C}^{(1)}$ and γ EVPs of size rN for the optimization of $\mathcal{C}^{(2)}$ for systems with two identical fermions. In the case of systems with three different particles, the ALS sequentially solves γ EVPs of size rN , r^2N , and rN for the optimization of $\mathcal{C}^{(1)}$, $\mathcal{C}^{(2)}$, and $\mathcal{C}^{(3)}$,

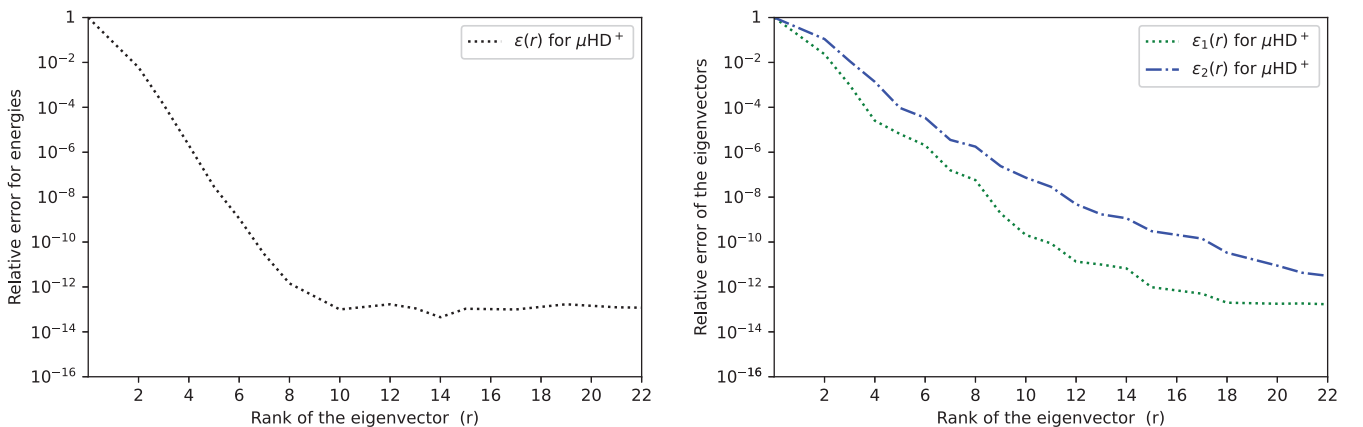


FIG. 2. Decay of the TT errors in energy ε and in the eigenvectors ε_1 and ε_2 for μHD^+ as a function of rank r . The calculations are performed using $N = 45$ with $a = \sqrt{-E}$.

TABLE III. Ground-state energy calculated using the tensor-train format with the ALS algorithm using $a = \sqrt{-E}$ and $a = 1$ for systems with two identical fermions $N = 65$ and $r = 9$ and for systems with three different particles $N = 45$ and $r = 10$. The energy is reported in atomic units when an electron is the lightest particle and muon atomic units when a muon is the lightest particle and the CPU time is reported in seconds.

System	Triad	Energy $a = \sqrt{-E}$	CPU time (s)	Energy $a = 1$	CPU time (s)
He	$(e^-, e^-, {}^4\text{He}^{2+})$	-2.903 304 557 729 7	264	-2.903 304 557 72	281
H ⁻	(e^-, e^-, p^+)	-0.527 445 881 109 5	277	-0.527 445 881 109 5	295
Mu ⁻	(e^-, e^-, μ^+)	-0.525 054 806 511 8	278	-0.525 054 806 511	293
$\mu\mu\text{He}$	$(\mu^-, \mu^-, {}^4\text{He}^{2+})$	-2.819 764 147 532	259	-2.819 764 147 5	278
Ps ⁻	(e^-, e^-, e^+)	-0.262 005 070 232 9	282	-0.262 005 070 232 9	345
μH_2^+	(p^+, p^+, μ^-)	-0.494 386 812 813 4	272	-0.494 386 812 813 4	282
μHD^+	(p^+, d^+, μ^-)	-0.512 711 790 529 6	22	-0.512 711 790 529 6	27
μDT^+	(d^+, t^+, μ^-)	-0.538 594 970 860 5	19	-0.538 594 970 860 5	27

respectively, where r is the TT rank of the eigenvector. For all of the systems in which $a = \sqrt{-E}$ the calculations converged to the desired energy in four iterations ($\gamma = 4$), and when $a = 1$ the energy converged in four or five iterations.

Furthermore, for systems with three different particles, such as (p^+, d^+, μ^-) , in which the tensor order is $d = 3$ and the matrix size of each full tensor operator is $N^3 \times N^3$, the TT calculations solve the problem at least 75 times faster. This highlights the efficiency of the ALS algorithm in tackling high-dimensional problems, where each mode size scales linearly in N for all cores. In contrast, for systems with two identical fermions, where the focus is on solving EVPs in lower dimensions (second-order tensors), where the mode size scales quadratically for the first TT core [$m_1 = N(N + 1)/2$] and linearly for the second TT core ($m_2 = N$), the increase in speed is not quite so dramatic. To further validate this finding, we solved the SE for $(e^-, e^-, \text{He}^{2+})$ using the TT code developed for any three particles, i.e., without directly accounting for the antisymmetry of the wave function with exchange of identical fermions. Remarkably, we achieved the same level of accuracy shown in Table III using $N = 65$ and a slightly

higher rank of 11, in just 25 s. This confirms the efficiency of low-rank decomposition for handling high-dimensional problems.

Accuracy of the wave function

To assess the quality of the TT wave function corresponding to the energy reported in Table III, the expectation values are reported in Table IV for three types of systems, atomic (He), exotic (Ps⁻), and molecular (μHD^+). The error analysis (Figs. 1 and 2) shows that the relative eigenvector errors converge more slowly with increasing rank than the error in the energy. Therefore, using a larger rank increases the accuracy relative to the expectation values obtained using the full format. However, for consistency, the expectation values corresponding to the rank used to obtain the energy in Table III, i.e., rank 9 for He and Ps⁻ and rank 10 for μHD^+ , are presented. For He and Ps⁻ the energy parameter is included in the coordinates, i.e., $a = \sqrt{-E}$, and for μHD^+ the energy parameter is not included. The data in Table IV are accurate to the number of digits reported. For (e^-, e^-, e^+) , the results are compared with those from [9]. The results for

TABLE IV. Expectation values $\langle X_{ij} \rangle$, the virial η , and the interparticle cusps v_{ij} , using the TT format with $a = \sqrt{-E}$ and rank 9 for He and Ps⁻ and using $a = 1$ and rank 10 for μHD^+ .

Property	He ($e^-, e^-, {}^4\text{He}^{2+}$)	Ps ⁻ (e^-, e^-, e^+)	μHD^+ (p^+, d^+, μ^-)
$\langle r_{13} \rangle$	0.929 607 915 012 8	5.489 633 252 3	2.451 487 669 669
$\langle r_{23} \rangle$	0.929 607 915 012 8	5.489 633 252 3	2.087 699 166 52
$\langle r_{12} \rangle$	1.422 247 512 607	8.548 580 655 0	3.100 710 490 8
$\langle 1/r_{13} \rangle$	1.688 076 584 66	0.339 821 023 059	0.641 146 354 615
$\langle 1/r_{23} \rangle$	1.688 076 584 66	0.339 821 023 059	0.753 373 610 431
$\langle 1/r_{12} \rangle$	0.945 697 223 219	0.155 631 905 652	0.369 096 383 988
$\langle r_{13}^2 \rangle$	1.193 834 894 42	48.418 937 22	8.033 494 743
$\langle r_{23}^2 \rangle$	1.193 834 894 42	48.418 937 22	5.896 526 406
$\langle r_{12}^2 \rangle$	2.517 061 842 48	93.178 633 84	10.829 021 78
$\langle \delta(r_{13}) \rangle$	1.809 672 3	0.020 733 19	0.117 709 725
$\langle \delta(r_{23}) \rangle$	1.809 672 3	0.020 733 19	0.173 456
$\langle \delta(r_{12}) \rangle$	0.106 (299) ^a	0.000 170 996	1.461×10^{-5}
η	4.3×10^{-13}	1.38×10^{-13}	1.2×10^{-12}
v_{12}	0.500 00	0.500 00	5.919
v_{13}	-1.999 72	-0.499 99	-0.946 671
v_{23}	-1.999 72	-0.499 99	-0.898 787

^aIn [23] this value is 0.106 300, but the value calculated here agrees with the value provided by Frolov [41] using a different nuclear mass.

(e^- , e^- , He^{2+}) and (p^+ , d^+ , μ^-) are compared with those in [41,42] and expectation values calculated using the series solution method described in [7], using the mass values provided in Table I. The virial factor η is determined by

$$\eta = \left| \frac{\langle V \rangle}{\langle T \rangle} + 2 \right| = 0. \quad (51)$$

This factor is a measure of the quality of the wave function obtained in the calculation. It is assumed that the closer the virial factor η is to zero, the better the wave function. The calculated cusp values v_{ij} are compared with the exact values given by [43]

$$v_{ij} = Z_i Z_j \frac{m_i m_j}{m_i + m_j}. \quad (52)$$

The exact value of v_{13} (to nine decimal places) is $-1.999\,725\,850$ for He, -0.5 for Ps^- , and $-0.946\,671\,425$ for μHD^+ . The exact value of v_{23} is $-1.999\,725\,850$ for He, -0.5 for Ps^- , and $-0.898\,787\,918$ for μHD^+ . The exact value of v_{12} is 0.5 for He and Ps^- and $5.919\,182\,636$ for μHD^+ .

The expectation distances $\langle r_{ij}^\alpha \rangle$ for $\alpha \in \{-1, 1, 2\}$ for all systems presented in Table IV show agreement with the reference values to at least ten significant figures. In contrast, contact properties between particles, such as $\delta(r_{ij})$ and v_{ij} , exhibit slightly lower accuracy compared to the average interparticle distances. This reduced accuracy is attributed to the fact that these properties describe the system at a single point where the particles coalesce. Similar observations have been reported in previous work dealing with three-particle quantum systems [9,23,44]. The virial factor η is accurate to at least 10^{-12} , which is two to three orders of magnitude more accurate than the results presented in [23]. These significant improvements highlight the better precision and reliability of the tensor methods presented here for these properties.

V. CONCLUSION

In this work we employed a wave function comprised of a triple orthogonal set of Laguerre functions to derive

formulations that accurately solve the Schrödinger equation for atomic and muonic systems using numerical tensor methods. Although the indices required to expand the wave function are naturally separable, as the coordinates are independent, we showed that it is possible to incorporate fermionic exchange in the wave function for systems with two identical fermions using a coupling of indices, demonstrating the feasibility of NTMs for this type of problem. Moreover, the sparsity of operators in full format, due to the properties of Laguerre functions, facilitates highly accurate energy calculations in less than an hour on a standard computer in the full tensor format. This approach addresses the key limitations of previous work [23], particularly the challenge of requiring high-precision factor matrix calculations. Additionally, we demonstrated that tensor-train decomposition significantly reduces CPU time by an order of magnitude without compromising the accuracy of the energies. Energies accurate to the picohartree can be obtained in less than 6 min for a range of electronic and muonic systems. However, the current formulation yields at best millihartree accuracy for the all-particle electronic molecular systems in a CPU time comparable to that required for picohartree accuracy for the atomic and exotic systems. Future work should focus on developing alternative formulations that offer greater flexibility in the wave function, ensuring an accurate representation of molecular properties at low computational cost.

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