Alternative sets of hyperspherical harmonics: Satisfying cusp conditions through frame transformations

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Abstract

By extending the concept of Euler-angle rotations to more than three dimensions, we develop the systematics under rotations in higher-dimensional space for a novel set of hyperspherical harmonics. Applying this formalism, we determine all pairwise Coulomb interactions in a few-body system without recourse to multipole expansions. Our approach combines the advantages of relative coordinates with those of the hyperspherical description. In the present method, each Coulomb matrix element reduces to the “1/r” form familiar from the two-body problem. Consequently, our calculation accounts for all the cusps in the wave function whenever an inter-particle separation vanishes. Unlike a truncated multipole expansion, the calculation presented here is exact. Following the systematic development of the procedure for an arbitrary number of particles, we demonstrate it explicitly with the simplest non-trivial example, the three-body system.

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I. INTRODUCTION

A system consisting of $N$ charged particles gives rise to $N(N - 1)/2$ pairwise Coulomb interaction terms in its Hamiltonian. Since only the two-body problem ($N = 2$) can actually be solved exactly, conventional atomic physics methods view the complete system of particles at the outset as a conglomerate of independent two-body systems, adding the interactions between these independent particles in a second step. This approach amounts to selecting a suitable subset of the $N(N - 1)/2$ Coulomb terms for which a solution in terms of “simultaneous two-body wave functions” can be given. For instance, the independent-particle model for atomic systems treats each electron as interacting primarily with the nucleus (or with the ionic core in the case of valence electrons). Each (valence) electron’s position introduces an independent spherical direction. Just as in the familiar solution of the hydrogen atom, each electron contributes to the angular part of the total system’s wave function a spherical harmonic $Y_{lm}$ of the angles specifying its direction in space. In the next step, all the electron-electron interactions are calculated by expanding the corresponding separations into Legendre polynomials of the inter-electronic angles, thus yielding the familiar multipole expansion. In more general terms, the independent-particle model first selects a specific particle #1 (on physical grounds, typically the nucleus or the ionic core) and solves for each of the remaining $N - 1$ particles the two-body problem \{1, $j$\}, $j = 2, \ldots, N$, thereby providing a basis for expanding the total wave function. In the next step, the interaction between particles $i$ and $j$ is calculated by adapting the reciprocal of their separation, $1/r_{ij}$, to the coordinate system pertaining to the “two-body” basis functions for $i$ and $j$ ($i, j = 2, \ldots, N$).

The Coulomb interaction is singular whenever an inter-particle separation vanishes. Since these singularities are isolated from one another, they do not pose fundamental difficulties in calculating the Hamiltonian matrix. However, they give rise to cusps (discontinuous derivatives) in the wave function $f$, thus slowing down the convergence of partial-wave expansions of the wave function in their vicinity $f$. The independent-particle wave functions can only account (through the corresponding s-wave components) for the cusps arising
from vanishing separations $r_{ij}$, $j = 2, \ldots, N$. Cusps due to vanishing $r_{ij}$ with $i \neq 1$ are not reproduced. Possible remedies to these shortcomings include the explicit use of coordinates like the inter-electronic angle $\theta_{ij}$ or even $r_{ij}$ besides $r_i$ and $r_j$; various approximations for $1/r_{ij}$ [4]; or replacing $r_i, r_j$ by $r_\prec = \min(r_i, r_j), r_\succ = \max(r_i, r_j)$ in the Hamiltonian [3]. All these approaches amount to adapting the interaction operator $1/r_{ij}$ to a single coordinate system.

The present investigation explores the alternative approach of adapting the basis functions to match the relevant inter-particle separations $r_{ij}$ by utilizing several coordinate systems simultaneously. We calculate a specific interaction term $\sim 1/r_{ij}$ in the coordinate system best suited for this particular purpose. Thus, the particle separations $r_{ij}$ dictate the choice of coordinate systems, the wave functions being transformed between the relevant reference frames to evaluate the different terms of the Hamiltonian. The success of this approach hinges on our ability to perform the numerous transformations between reference frames with high efficiency. Sec. II describes hyperspherical Jacobi coordinates appropriate for this task. Sec. III provides the main results by implementing the relevant transformations for a system with an arbitrary number of particles and by constructing basis functions (harmonics) suitable for extensive transformation between reference frames. The resulting set of hyperspherical harmonics, derived here in the context of calculating the Hamiltonian matrix for a system of $N$ charged particles, has in fact much wider applicability.

Hyperspherical coordinates and corresponding hyperspherical harmonics have been applied in various areas of physics since the 1950s, for instance, in three-body scattering [3, 8], nuclear [3, 10] and atomic [11, 13] physics, as well as in quantum chemistry [20, 21]. However, the sets of functions introduced in the present investigation are equivalent to, but much more flexible than, the hyperspherical harmonics discussed in [3, 21]. Beyond constituting a complete orthogonal set of functions appropriate for expansions, their frame-independence affords greater flexibility in analyzing selection rules and other relations between harmonics. These aspects are conveniently investigated through ladder operators; they are determined entirely by the symmetry properties of the $N$-particle system, independent of any coordinate
representation. Only calculating the non-vanishing matrix elements requires an appropriate coordinate representation of the generic basis functions. Sec. IV illustrates the relevant procedures with the simplest non-trivial example, the three-body problem. The concluding Sec. V discusses advantages and limitations of the new technique, as compared to the conventional multipole expansion.

II. HYPERSONICAL JACOBI COORDINATES

The inverse proportionality between pairwise interaction and inter-particle separation suggests replacing individual particles’ positions with relative (Jacobi) coordinates. Starting from one pair of particles, the construction of Jacobi coordinates proceeds hierarchically by joining (the centers of mass of) increasingly complex groups of particles. This hierarchical structure is commonly referred to as a “Jacobi tree” [10]. Alternative choices for the initial particle pair, as well as the ordering of successive particle groups, correspond to different Jacobi trees. In the next step, mass-scaling of the Jacobi coordinates fully exposes the symmetry of the kinetic energy operator for the multi-particle system. Let \( p, q \) denote two (groups of) particles with masses \( M_p, M_q \) and center of mass positions \( r_p \) and \( r_q \), respectively. Appropriate mass-scaling of the relative coordinate in the form \[ \xi_{p,q} = \frac{M_p M_q}{(M_p + M_q)} \left( r_p - r_q \right) \] removes the individual mass dependence from the expression for the kinetic energy:

\[
-\sum_{i=1}^{N} \frac{\hbar^2}{2M_i} \Delta r_i = -\frac{\hbar^2}{2} \sum_{k=1}^{N-1} \Delta \xi_k - \frac{\hbar^2}{2M_{\text{tot}}} \Delta r_{\text{CM}}. \tag{2.1}
\]

Setting the origin at the center of mass of the whole system allows discarding the CM’s position and motion. With the individual mass factors removed, the kinetic energy (generalized Laplacian) displays complete symmetry under rotations in \((3N - 3)\)-dimensional space.

Hyperspherical coordinates exploit this symmetry by separating the “shape” of the system (described by \(3N - 4\) angular coordinates) from its overall “size” \( R^2 = \sum_k \xi_k^2 \) (with the dimension of a moment of inertia). Eigenfunctions of the Laplacian’s angular part,
termed “hyperspherical harmonics” by generalizing the two- and three-dimensional cases, constitute a basis for expanding the complete wave function. In higher-dimensional spaces, the angular Laplacian’s eigenvalues $\Lambda(\Lambda + 3N - 5)$ are highly degenerate. In addition to the “grand angular momentum” $\Lambda$ \[8\], the hyperspherical harmonics thus require numerous labels—analogues of the single “magnetic” quantum number $m$ for three-dimensional spherical harmonics—for their unique specification. Different sets of hyperspherical harmonics, resulting from solving the Laplacian eigenvalue problem by separation of variables in alternative sets of coordinates, have been investigated extensively (see, e.g., \[10,17,21\] for systematic studies). However, their construction through separation of variables inevitably ties these harmonics to a specific coordinate system. They are thus not well suited for extensive transformation between different reference frames. The definition of harmonics based only on their behavior under the relevant transformations is the main result of this paper, to be derived in Sec. II.

The construction of each Jacobi tree starts with a pair of particles. Thus, each Jacobi tree contains at least one Jacobi vector joining two particles only (rather than centers of mass of particle groups). We denote such a vector as a “primary” coordinate. In any one Jacobi tree, up to $\lfloor N/2 \rfloor$ Jacobi vectors are directly proportional to actual inter-particle separations, $r_{ij}$, the remaining relative coordinates necessarily involving larger complexes of particles. (Here and in the following, $\lfloor x \rfloor$ denotes the largest integer not exceeding $x$.) We will calculate each of the $N(N - 1)/2$ Coulomb terms of the Hamiltonian in a Jacobi tree where it occurs as a “primary” coordinate. Obviously, all these terms then take the same form as the simple one-electron integral over $1/r$ in hydrogen, but the variables now refer to a multitude of different reference frames. Having thus eliminated all nested integrations, our next task consists in determining the transformations between different Jacobi trees.

The most general transformation from one Jacobi tree to another (for the same system of particles) resolves into a sequence of elementary operations. Each elementary step consists in “transplanting” a sub-complex $q$ from some particle complex $\{pq\}$ to a complex $\{qr\}$ \[10\]; it is achieved by a two-dimensional kinematic rotation \[8\] through the angle
$$\phi = \tan^{-1} \sqrt{M_q(M_p + M_q + M_r)/M_p M_r} \tag{2.2}$$

in the \((\xi_{p,q}, \xi_{pq,r})\)-plane of the \((N - 1)\)-dimensional space of mass-scaled Jacobi vectors:

$$\xi'_{q,r} = \cos \phi \xi_{p,q} - \sin \phi \xi_{pq,r} \quad (2.3a)$$

$$\xi'_{p,q} = \sin \phi \xi_{p,q} + \cos \phi \xi_{pq,r}. \quad (2.3b)$$

As each Jacobi vector is a vector in three-dimensional physical space, the basic rotation in the \((\xi_j, \xi_k)\)-plane implies three rotations through the same angle \(\phi\) in the \((x_j, x_k)\)-, the \((y_j, y_k)\)-, and the \((z_j, z_k)\)-planes, where \((x, y, z)\) denote the Cartesian components of \(\xi\). An elementary kinematic rotation through a finite angle \(\phi\) in the \((\xi_j, \xi_k)\)-plane then reads

$$T_{\xi_j, \xi_k}(\phi) = \exp(i\phi J_{x_j x_k}) \exp(i\phi J_{y_j y_k}) \exp(i\phi J_{z_j z_k}), \quad (2.4)$$

with infinitesimal rotation operators

$$J_{uv} \equiv -i \left( u \frac{\partial}{\partial v} - v \frac{\partial}{\partial u} \right), \quad (u, v) = (x_j, x_k), (y_j, y_k), (z_j, z_k). \quad (2.5)$$

(For a heuristic explanation of \(2.4\), recall the Taylor expansion of \(f(x + \Delta x)\) which may formally be written as \(\exp(i\Delta x p) f(x)\) with the infinitesimal “translation operator” \(p \equiv -i \frac{\partial}{\partial x}\) corresponding to the quantum mechanical linear momentum (with \(\hbar \equiv 1\). In the present context, the translation from \(x\) to \(x + \Delta x\) is replaced by a rotation through an angle \(\phi\) from some direction in multi-dimensional space to a new direction.) In order to determine \(T_{\xi_j, \xi_k}\)’s effect on the basis functions, we need to (i) express the generic rotation operators \(J_{uv}\) in terms of “ladder operators” whose action on the basis functions is straightforward to calculate, and (ii) construct basis functions suitable for such transformations.

This section concludes with the outline of a strategy for determining the most efficient sequence of two-dimensional rotations to achieve the transformation between arbitrary Jacobi trees. Note first that the individual terms in the Hamiltonian are referred to by means of particle labels, implying the need for appropriate antisymmetrization of the total wave function under interchange of identical particles. This consideration dominates the construction
of the first Jacobi tree to which all subsequent transformations are applied. In an atomic or molecular system, antisymmetrization concerns primarily the electronic part of the wave function. Thus the basic Jacobi tree starts out with a pair of electrons. It grows by adding one electron at a time, finishing with the addition of nuclei or ionic core(s). This procedure results in a “canonical” Jacobi tree, represented by the sequence of particle labels

\[(\cdots(((12)3)4)5)\ldots),\]

whose parentheses separate sub-complexes. Alternatively, one could start by first forming as many pairs of particles as possible, i.e., \([N/2]\) pairs, before joining these pairs into larger complexes. Although the antisymmetrization of an electron pair is particularly compact (namely, their relative angular momentum and their spin must add to an even value), antisymmetrization among different pairs requires breaking up all these pairs, in essence going back to a canonical tree. Since breaking up sub-complexes involves about as many elementary rotations as forming the new complexes, this procedure is not efficient. Nevertheless, a set of \(N\) (for odd particle number) or \(N - 1\) (even particle number) trees with the maximum number of \([N/2]\) pairs suffices to isolate all the \(N(N - 1)/2\) inter-particle separations. A successful strategy therefore aims at building these particular Jacobi trees from the canonical tree. The simplest of these “pair-trees”, \((\cdots((12)(34))(56)\ldots)\), is obtained from the canonical tree with only \([N/2] - 1\) rotations. In general, the pair \((jk)\) with \(j < k\) is \((k - j - \delta_{j,1})\) rotations away from the canonical tree, but many other “useful” pairs are formed in the course of these “transplantations”.

### III. TRANSFORMING THE BASIS FUNCTIONS

This section provides the tools required to rotate harmonics from one coordinate system to another. Our transformation matrices are analogous to the so-called Raynal-Revai coefficients. The reader familiar with Lie algebra will of course recognize the relevant aspects of \(so(3N - 3)\), the algebra of rotations in \((3N - 3)\) dimensional space. However,
such familiarity is not presumed, and we hardly use the terminology of group theory. Our presentation reflects a more pragmatic point of view, emphasizing both the technical implementation as well as its relation to the physical application at hand rather than full mathematical generality. For the latter aspects, the reader should turn to the mathematical literature [24].

A. Rotations in \( d \)-dimensional space

The most intuitive description of a rotation in three dimensions requires two elements: the (invariant) axis, and the angle of rotation. The rotation itself occurs in a plane perpendicular to the axis of rotation. In three dimensions, specifying the direction of the axis of rotation is equivalent to, but more economical than, describing the actual plane of rotation. For a higher dimensional space this is no longer the case, because there are several invariant directions perpendicular to a given plane. Thus, in \( d \)-dimensional space, a basic rotation is more appropriately characterized as occurring in the plane spanned by two coordinates rather than by an invariant axis orthogonal to it. The generic infinitesimal rotation operators then take the form (2.3). The number of different planes, \( \frac{1}{2}d(d-1) \) in \( d \) dimensions, equals the number of basic rotations. Because rotations occurring in non-intersecting planes affect different pairs of coordinates, they are evidently independent of each other; the corresponding rotation operators commute with one another. As there are \( [d/2] \) non-intersecting planes in a \( d \)-dimensional space, the largest set of simultaneously commuting operators \( J_{uv} \) contains \( \ell \equiv [d/2] \) elements. These commuting operators are commonly denoted as \( H_j, j = 1, \ldots, \ell \) [24]. For our application, the power of the Lie algebraic method derives from the structure and the properties of rotations being completely independent of any particular coordinate representation of the operators \( H_j \). In fact, the following manipulations are most efficiently carried out in generic Cartesian coordinates without any reference to a specific Jacobi tree.

Simultaneous eigenfunctions of the \( H_j \) with (integer) eigenvalues \( m_j, j = 1, \ldots, \ell \), provide a basis for building hyperspherical harmonics suitable for extensive rotations between refer-
ence frames. If the infinitesimal rotation corresponding to $H_j$ occurs in the $(u_j, v_j)$-plane, the simultaneous eigenfunction of the $\ell H_j$ with eigenvalues $m_j$, respectively, reads

$$F(\{u_k, v_k\}) \times \prod_{j=1}^{\ell} (u_j + i v_j)^m_j$$

(3.1)

with a function $F$ that bears further specification in Subsect. III C. At this point, we only require that $H_j F \equiv 0$ for all $j$. The set of eigenvalues $\mu = \{m_1, m_2, \ldots, m_\ell\}$ serves as a label identifying different harmonics with the same value of $\Lambda(\Lambda + d - 2)$ in the angular Laplacian’s eigenvalue equation. Additional labels required for a unique specification will be introduced in Subsect. III C.

Appropriate linear combinations of the remaining infinitesimal rotation operators act as ladder operators $E_\alpha$ satisfying

$$[H_j, E_\alpha] = \alpha_j E_\alpha, \quad j = 1, \ldots, \ell,$$

(3.2)

with an $\ell$-dimensional vector index $\alpha$ with components $\alpha_j = 0, \pm 1$. Here $\alpha_j = 0$ indicates that $E_\alpha$ does not change the part of the eigenfunction pertaining to $H_j$, whereas $\alpha_j = \pm 1$ means that it maps this part to the eigenfunction with eigenvalue $m_j \pm 1$. The ladder operators interrelate eigenfunctions with different $\mu$ but degenerate $\Lambda$. Raising and lowering operators form Hermitian conjugate pairs $E_{-\alpha} = E_\alpha^\dagger$.

In general (for $\ell > 1$), each ladder operator affects two of the $m_j$ simultaneously; for odd-dimensional spaces a subset of $\ell$ pairs of raising and lowering operators change one of the $m_j$ only [24]. Abbreviating the vector label $\alpha$ by giving only its two non-vanishing components $\alpha_j$ and $\alpha_k$, the ladder operator that raises $m_j$ and simultaneously lowers $m_k$ takes the form

$$E_{jk}^{+-} = -\frac{i}{2} \left( (u_j + i v_j) \frac{\partial}{\partial(u_k + i v_k)} - (u_k - i v_k) \frac{\partial}{\partial(u_j - i v_j)} \right),$$

(3.3a)

whereas the operator raising both $m_j$ and $m_k$ is represented by

$$E_{jk}^{++} = -\frac{i}{2} \left( (u_j + i v_j) \frac{\partial}{\partial(u_k - i v_k)} - (u_k + i v_k) \frac{\partial}{\partial(u_j - i v_j)} \right).$$

(3.3b)
(Straightforward application of these operators to eigenfunctions (3.1) verifies their behaving as ladder operators: they contribute to, or remove from, (3.1) factors \((u_j \pm iv_j)\) and \((u_k \pm iv_k)\) as appropriate for the intended “ladder operator action”.) For odd-dimensional spaces, a residual coordinate, denoted here by \(w\), does not occur in any of the \(H_j\). The ladder operators changing only \(m_j\) (rather than a pair \(m_j, m_k\)) read then

\[
E_j^\pm = -i \left( (u_j \pm iv_j) \frac{\partial}{\partial w} - w \frac{\partial}{\partial (u_j \mp iv_j)} \right), \quad j = 1, \ldots, \ell. 
\]  

(3.3c)

(For \(d = 3\), setting \((u, v, w) = (x, y, z)\) and transforming the derivatives reveals the familiar pair of ladder operators \(l_x \pm il_y\), the single \(H_j\) occurring in this case coinciding with \(l_z\).) This symbolic representation allows for efficient implementation on the computer.

A subset of \(\ell\) ladder operators \(E_{\epsilon_j}\) (and their Hermitian conjugates) suffices to interrelate all harmonics with the same eigenvalue \(\Lambda\). A convenient choice [24] for these \(E_{\epsilon_j}\) is given by the ladder operators that raise \(m_j\) and simultaneously lower \(m_{j+1}\) for \(j = 1, \ldots, \ell - 1\). \(E_{\epsilon_j}\)'s form depends on whether \(d\) is even or odd. For \(d\) even, \(E_{\epsilon_{\ell}}\) raises both \(m_{\ell-1}\) and \(m_{\ell}\), and for odd \(d\), it raises \(m_{\ell}\) only, without changing any of the other \(m_j\). The set \(\{\epsilon_j\}, j = 1, \ldots, \ell\), then provides a basis for the \(\ell\)-dimensional space of the vector labels \(\alpha\) and \(\mu\). In an \(\ell\)-component vector notation, this basis reads (we assume \(\ell \geq 5\) in order to expose the generic structure clearly)

\[
\epsilon_1 = (1, -1, 0, 0, \ldots, 0) \\
\epsilon_2 = (0, 1, -1, 0, \ldots, 0) \\
\epsilon_3 = (0, 0, 1, -1, \ldots, 0) \\
\vdots \\
\epsilon_{\ell-1} = (0, 0, \ldots, 0, 1, -1) \\
\epsilon_\ell = \begin{cases} 
(0, 0, \ldots, 0, 1, 1) & \text{for } d = 2\ell \\
(0, 0, \ldots, 0, 0, 1) & \text{for } d = 2\ell + 1.
\end{cases}
\]  

(3.4)

In general these basis vectors are not orthogonal in \(\ell\)-dimensional space. Note, however, the following special cases: (i) In four-dimensional space (with \(\ell = 2\)), the two basis vectors \(\epsilon_1 = (1, -1)\) and \(\epsilon_2 = (1, 1)\) are orthogonal, indicating that the two basic ladder operator pairs,
$E_{\pm \epsilon_1}$ and $E_{\pm \epsilon_2}$, commute with each other. In group theoretical language, this feature reflects the direct product structure $SO(4) = SO(3) \otimes SO(3)$. However, the two $SO(3)$-components do not refer to $m_1$ and $m_2$ directly, but rather to $(m_1 - m_2)/2$ and $(m_1 + m_2)/2$. (ii) For rotations in three-dimensional space, we have $\ell = 1$, thus only one quantum number $m$ which is being changed by one pair of ladder operators $E_{\pm \epsilon_1} \equiv l_\pm$. (iii) In $d = 2$ dimensions, there are no ladder operators. Since all rotations occur in the same plane (the “only” plane of two-dimensional space), the order in which rotations through different angles are performed does not matter; they are all independent of each other. In the present formulation, the (single) rotation operator $H_1$ generates all rotations. Each rotation is associated with its own harmonic function, $\exp(i \phi)$, the phase functions for different rotations (i.e., for different rotation angles $\phi$) not being related to one another through linear operators.

B. Rotation of hyperspherical harmonics

We now turn to the analysis of the transformation described by (2.4). Note first that the three rotations occur in three non-intersecting planes, affording the more suitable representation $\exp(i \phi [J_{x_j x_k} + J_{y_j y_k}]) \exp(i \phi J_{z_j z_k})$. Arranging the Cartesian components of the $N - 1$ mass-scaled Jacobi vectors $\xi_k$ in the form

$$\{x_1, y_1, x_2, y_2, \ldots, x_{N-1}, y_{N-1}, z_1, z_2, \ldots, z_{N-1}\}, \quad (3.5)$$

we choose the $H_j$ by selecting pairs of coordinates from this list, starting from the left:

$$H_j = J_{x_j y_j}, \quad j = 1, \ldots, N - 1 \quad (3.6a)$$
$$H_{N-1+k} = J_{z_{2k-1} z_{2k}}, \quad k = 1, \ldots, \left\lfloor \frac{N - 1}{2} \right\rfloor, \quad (3.6b)$$

in the notation of (2.3). We have stressed repeatedly our aim of defining hyperspherical harmonics entirely through their behavior when acted upon by the operators $H_j$ and $E_{\epsilon_j}$, because these operators embody the structure and symmetry of the whole system in a frame-independent way. The explicit coordinate representation (3.6a, 3.6b) of the relevant opera-
tors appears at variance with our intended frame-independence. However, the spatial coordinates, \((x_j, y_j, z_j), j = 1, \ldots, N - 1,\) in (3.6a–3.6b) should be viewed as *generic Cartesian coordinates*; they are not tied to any particular Jacobi tree. Arranging the generic Cartesian coordinates as we did in (3.3), on the other hand, does reflect physical considerations beyond the purely mathematical structure of rotations in higher-dimensional space: The latter would instead label the coordinates most appropriately as \((X_1, X_{-1}, X_2, X_{-2}, \ldots, X_\ell, X_{-\ell}),\) supplemented possibly with an \(X_0\) for odd-dimensional spaces. Our arrangement takes into account that the dimension \((3N - 3)\) arises from a *product structure* of an \((N - 1)\)-dimensional particle space with the three-dimensional physical space of each Jacobi vector. In particular, the arrangement (3.3) affords attributing relevant physical meaning to the first \((N - 1)\) eigenvalues \(m_j, (j = 1, \ldots, N - 1):\) They represent the \(z\)-projections of physical angular momenta; their sum constitutes the \(z\)-projection \(L_z\) of the total orbital angular momentum, an invariant of the system.

As is evident from (3.6a), the coordinates \((x_j, x_k, y_j, y_k)\) occur in \(H_j\) and \(H_k.\) The \(x\)- and \(y\)-parts of the rotation (2.4) thus involve the ladder operators that change \(m_j\) and \(m_k\) only. A more detailed analysis shows that (3.3a–c) can be inverted to read:

\[
J_{x_jx_k} + J_{y_jy_k} = E_{jk}^{++} + E_{jk}^{--} \equiv E_{\alpha(jk)} + E_{-\alpha(jk)},
\]

(3.7)

for \(1 \leq j < k \leq N - 1,\) with \(\alpha(jk) = \sum_{s=j}^{k-1} \epsilon_s,\) i.e., the \(\ell\)-component vector with +1 as its \(j\)-th component and -1 as its \(k\)-th component, all other entries being 0.

The following consideration is central to our development, providing the crucial link between the ladder operator representation (3.7) for \((J_{x_jx_k} + J_{y_jy_k})\) and their transformation matrix elements, by means of Euler-angle rotations. In analogy to the relation \(l_x = \frac{1}{2}(l_+ + l_-)\) familiar from rotations in three dimensions, we view the sum of a ladder operator \(E_{\alpha(jk)}\) and its inverse (Hermitian conjugate) \(E_{-\alpha(jk)}\) as describing a rotation about an analogue of the \(x\)-axis \([25]\). In three dimensions, an arbitrary rotation conventionally resolves into a sequence of three rotations: about the \(z\)-axis, about the resulting \(y'\)-axis, and about the new \(z'\)-axis, through the Euler angles \((\varphi, \theta, \psi),\) respectively \([22]\). In terms of these three Euler-angle
rotations, a rotation about the $x$-axis through an angle $\phi$ results from the following sequence of operations: The first Euler-angle rotation about the $z$-axis through the angle $\varphi = -\pi/2$ rotates the $y$-axis onto the original $x$-axis; in the second step one rotates about this $y'$-axis (which is the original $x$-axis) through the angle $\theta = \phi$; the third Euler-angle rotation finally moves the $x'$-axis back to the original $x$-direction by rotating about the $z'$-axis (lying in the original $(yz)$-plane) through the angle $\psi = \pi/2$. Wigner’s $d$-symbol is the matrix element for the rotation of a spherical harmonic about the $y$-axis, $d^{(l)}_{m'm}(\phi) = \langle Y_{lm'} | \exp(i\phi l_y) | Y_{lm} \rangle$, whereas the two $z$-type rotations only contribute phase factors $\exp(i m'\pi/2 - i m\pi/2)$ [22], giving for the rotation of a spherical harmonic about the $x$-axis

$$
\langle Y_{lm'} | \exp(i\phi l_x) | Y_{lm} \rangle = e^{im'\frac{\pi}{2}} d^{(l)}_{m'm}(\phi) e^{-im\frac{\pi}{2}}.
$$

(3.8)

Generalization of the concept of Euler-angle rotations from three to more dimensions hinges on the following key observations: $m$ and $m'$ serve to distinguish between degenerate harmonics with the same $l$. In higher-dimensional spaces, the vector indices $\mu$ and $\mu'$ play the same role. However, the parameter $l$ in the $d$-symbol indicates not only the angular Laplacian’s eigenvalue $l(l+1)$, but also—more importantly—the range of possible $m$ values, $-l \leq (m, m') \leq l$. More precisely, it sets the upper limit of $2l$ for the number of times either one of the two ladder operators $l_+, l_-$ can act in direct succession before necessarily mapping any spherical harmonic $Y_{lm}$ to zero. (Since $l_+ Y_{lm} \rightarrow Y_{l,m+1}$, it takes $l - m \leq 2l$ “raising operator” steps to reach $Y_{ll}$ from $Y_{lm}$. Applying $l_+$ in succession $(2l + 1)$ times necessarily raises $m$ beyond its upper limit. The absence of a corresponding harmonic translates to $(l_+)^{2l+1} Y_{lm} \equiv 0$.). Viewed in this way, the harmonic’s parameter $m$ indicates its “position” along the string (of length $2l+1$) of degenerate harmonics interrelated by a ladder operator. We now extend these concepts to rotations in more than three dimensions.

The single $m$ ($l_z$’s eigenvalue) in three dimensions belongs to a string ranging from $-l$ to $+l$, accessed by the single pair of ladder operators $l_+, l_-$. In more than three dimensions, we replace it with an $\ell$-component vector $\mathbf{\mu}$ whose components are changed (typically in pairs) along different strings labeled by corresponding ladder operator pairs $E_\alpha$ and $E_{-\alpha}$. (Recall,
however, that the $\ell$ vectors $\epsilon_j$ provide a basis for the vectors $\alpha$; thus any ladder operator $E_\alpha$ can be expressed as products of $E_{\epsilon_j}$.) The role of $m$ as an indicator of the harmonic’s position along the single string in three dimensions extends therefore to higher dimensions if we project the “indicator” $\mu$ onto the “direction” of any particular string $\alpha$ of interest, i.e., if we define

$$m_\alpha = \frac{\mu \cdot \alpha}{\alpha \cdot \alpha}, \quad m'_\alpha = \frac{\mu' \cdot \alpha}{\alpha \cdot \alpha},$$

(3.9)

where the scalar product in the denominators accounts for $\alpha$’s two non-zero components in the case of a pairwise change of $m_j$’s. In analogy to $l$ in the three-dimensional case, a parameter $\lambda_\alpha$ determines how many times the ladder operators $E_{\pm \alpha}$ can be applied in direct succession. The modulus of $m_\alpha$ in (3.9) provides a lower limit for the relevant “string length” $\lambda_\alpha$. Since the $\epsilon_j$ form a basis for the $\alpha$, we anticipate that $\lambda_\alpha$ emerges from an $\ell$-component vector $\lambda = (\lambda_{\epsilon_1}, \ldots, \lambda_{\epsilon_\ell})$. The latter provides the additional parameters required for a unique specification of harmonics (besides $\mu$ and the eigenvalue $\Lambda$). Note that in the general case $\lambda_\alpha$ is not identical with the eigenvalue $\Lambda$, at variance with the situation in three dimensions.

To summarize the procedure so far, we note that the $x$- and $y$-parts of the finite rotation (2.4) affect only the two components $m_j$ and $m_k$ of the vector $\mu$, shifting $m_j$ by an integer amount $n$ while simultaneously changing $m_k$ by the same amount in the opposite direction. The projections (3.9) evaluate to

$$m_{\alpha(jk)} = \frac{1}{2}(m_j - m_k);$$  
$$m'_{\alpha(jk)} = \frac{1}{2}(m'_j - m'_k) = m_{\alpha(jk)} + n,$$

(3.10a)

(3.10b)

with non-vanishing matrix elements for the rotation of a hyperspherical harmonic $Y_{\Lambda,\mu,\lambda}$ into another harmonic $Y_{\Lambda',\mu',\lambda'}$ occurring only if $\mu$ and $\mu'$ lie along the same $\alpha$-string of harmonics with degenerate $\Lambda$, i.e., if an integral number $n$ of ladder operator steps $E_{\pm \alpha(jk)}$ separates $\mu'$ from $\mu$. With our previous considerations on Euler-angle rotations leading to (3.8), this matrix element reads
\[ \langle Y_{\Lambda',\mu',\lambda'} | \exp(i\phi[J_{xjzk} + J_{yjyk}]) | Y_{\Lambda,\mu,\lambda} \rangle = e^{i(m'_\alpha - m_\alpha)\pi/2} d^{(\lambda\alpha)}_{m'_\alpha m_\alpha} (2\phi) \delta'_{\Lambda',\Lambda} \delta_{\lambda',\lambda} \delta_{\mu',\mu + n\alpha}, \] (3.11)

where we omitted \( \alpha \)'s parameters \( j, k \) for brevity. The Kronecker symbols \( \delta \) ensure that only harmonics with the same eigenvalue \( \Lambda \), lying along the same \( \alpha \)-string, are connected. The factor 2 multiplying the rotation angle \( \phi \) reflects the absence of the factor \( 1/2 \) in the RHS of (3.7), as compared to the expression for \( l_x = \frac{1}{2}(l_+ + l_-) \), thereby effectively doubling the rotation angle in (3.8).

It remains to apply the same concepts to the \( z \)-part of the rotation (2.4). This part affects only \( \mu \)'s components that correspond to the \( H_\kappa \) involving \( z_j \) and \( z_k \). Because we chose to gather all the \( z \)-coordinates after all \((xy)\)-pairs in (3.3), the Jacobi vector indices \( j, k \) are shifted relative to \( \mu \)'s components for the \( z \)-coordinates. To simplify the notation, we use

\[ \iota \equiv N - 1 + \left\lfloor \frac{j + 1}{2} \right\rfloor; \quad \kappa \equiv N - 1 + \left\lfloor \frac{k + 1}{2} \right\rfloor, \] (3.12)

so that \( H_\iota \) and \( m_\iota \) now refer to the operator and quantum number pertaining to the coordinate \( z_j \), with the same connection between \( H_\kappa, m_\kappa, \) and \( z_k \). Depending on the indices \( j \) and \( k \), i.e., depending on the positions of the coordinates \( z_j \) and \( z_k \) in the sequence (3.5), \( J_{zjzk} \) takes alternative forms in terms of ladder operators or \( H_\kappa \). The possible cases are:

1. \( k \) even and \( j = k - 1 \). According to (3.6b), \( J_{zjzk} \) coincides with \( H_{N-1+k/2} \equiv H_\kappa \). The rotation matrix element is simply

\[ \langle Y_{\Lambda',\mu',\lambda'} | \exp(i\phi J_{zjzk}) | Y_{\Lambda,\mu,\lambda} \rangle = \exp(im_\kappa \phi) \delta'_{\Lambda',\Lambda} \delta_{\lambda',\lambda} \delta_{\mu',\mu}. \] (3.13)

2. \( k \) even and \( j < k - 1 \) even. We find

\[ J_{zjzk} = \frac{1}{4} \left( E^{++}_{l_\kappa} + E^{+-}_{l_\kappa} + E^{++}_{l_\kappa} + E^{--}_{l_\kappa} \right). \] (3.14)

Viewing again the sum of a ladder operator and its inverse (Hermitian conjugate) as proportional to an analogue of a rotation about the \( x \)-axis, we readily recognize
the last expression as the sum of two $x$-type rotations (compare with (3.7)’s central expression). With the explicit formulae (3.3a–3.3b), it is straightforward to verify that the ladder operator $E_{ik}^{-}$ raising $m_i$ and lowering $m_\kappa$ commutes with the “raising—raising” operator $E_{ik}^{++}$. The two $x$-type rotations are therefore independent of each other. As in the previous discussion of the $x$- and $y$-parts, an integer number of steps $E_{ik}^{++}$ must connect $(m_i, m_\kappa)$ to $(m'_i, m'_\kappa)$, i.e., $m'_i = m_i + n_1$, $m'_\kappa = m_\kappa - n_1$. However, the second rotation involves the same components of $\mu$, only this time changing both in the same direction: $m'_i = m_i + n_2$, $m'_\kappa = m_\kappa + n_2$. Compatibility of the two conditions requires $n_1 = n_2 = 0$, thus

\[ m_{\beta_-(\kappa)} = \frac{1}{2}(m_i - m_\kappa) = m'_{\beta_-(\kappa)} \]  
(3.15a)

\[ m_{\beta_+(\kappa)} = \frac{1}{2}(m_i + m_\kappa) = m'_{\beta_+(\kappa)}, \]  
(3.15b)

and the extra phase factors occurring in (3.11) drop out in this case:

\[ \langle Y_{\Lambda'}, \mu', \chi | \exp(i \phi \cdot J_{z_{jk}}) | Y_{\Lambda, \mu, \lambda} \rangle = d_{m_{\beta_-(\kappa)}, m_{\beta_-(\kappa)}}(\phi/2) d_{m_{\beta_+(\kappa)}, m_{\beta_+(\kappa)}}(\phi/2) \delta_{\Lambda', \Lambda} \delta_{\chi', \chi} \delta_{\mu', \mu}. \]  
(3.16)

The factor $1/2$ multiplying the rotation angle stems of course from the factor $1/4$ in (3.14).

3. $k$ even and $j < k - 1$ odd. In this case,

\[ J_{z_{jk}} = \frac{1}{4i} \left( E_{ik}^{-} - E_{ik}^{+} + E_{ik}^{++} - E_{ik}^{--} \right), \]  
(3.17)

obviously the sum of two $y$-type rotations. The “selection rules” for $m_i$ and $m_\kappa$ are the same as in the previous case, and since the extra phase factors distinguishing an $x$- from a $y$-type rotation cancel in (3.16), the matrix element is identical to the one in case 2.

4. $k < N - 1$ odd and $j$ even. $J_{z_{jk}}$ turns into

\[ J_{z_{jk}} = \frac{1}{4i} \left( E_{ik}^{++} - E_{ik}^{--} + E_{ik}^{++} - E_{ik}^{--} \right). \]  
(3.18)
Exactly the same considerations apply again, except that the difference of two $y$-type rotations corresponds to a rotation through the angle $-\phi/2$ for the $E^\pm$ part.

5. $k < N - 1$ odd and $j$ even, yielding

$$J_{z_j z_k} = \frac{1}{4}\left(E_{i\kappa}^+ + E_{i\kappa}^- - E_{i\kappa}^{++} - E_{i\kappa}^{--}\right), \quad (3.19)$$

i.e., the difference of two $x$-type rotations. The matrix element coincides with the one of case 4.

6. $k = N - 1$ odd and $j$ even. $z_k$ does not occur in any of the $H_\kappa$, and the relevant ladder operators act on $m_\iota$ only:

$$J_{z_j z_k} = \frac{1}{2}\left(E_i^+ + E_i^-\right). \quad (3.20)$$

This is an $x$-type rotation, exactly as in three dimensions. Projecting $\mu$ and $\mu'$ onto the appropriate direction $\gamma(i)$: $m_\gamma(i) = m_\iota$, $m'_\gamma(i) = m_\iota' = m_\gamma(i) + \nu$, we obtain for the matrix element

$$\langle Y_{\Lambda', \mu', \chi'} | \exp(i\phi J_{z_j z_k}) | Y_{\Lambda, \mu, \chi} \rangle = d_{m'_\iota, m_\gamma}^{(\lambda_\gamma)}(\phi) e^{i(m'_\iota - m_\gamma)\pi/2} \delta_{\Lambda', \Lambda} \delta_{\chi', \chi} \delta_{\mu', \mu + \nu'\gamma}. \quad (3.21)$$

7. $k = N - 1$ odd and $j$ odd.

$$J_{z_j z_k} = \frac{1}{2i}\left(E_i^+ - E_i^-\right), \quad (3.22)$$

a $y$-type rotation. Its matrix element differs from the one in case 6 only by the absence of the phase factor $\exp(i\nu\pi/2)$.

Thus, the $z$-part of the rotation (2.4) takes essentially three different forms: (i) a phase factor $\exp(im_\kappa\phi)$, diagonal in the $\mu$, if $J_{z_j z_k}$ coincides with $H_\kappa$; (ii) an $x$- or $y$-type rotation through the angle $\phi$, changing $m_\iota$ into $m'_\iota = m_\iota + \nu$, as in three dimensions, if $z_k$ is the unpaired coordinate $z_{N-1}$ of an odd-dimensional space; (iii) the product of two rotations through the angles $\phi/2$ and $(-1)^k\phi/2$, respectively, but diagonal in $\mu$, if $z_j$ and $z_k$ belong to different $H_\iota$ and $H_\kappa$. 

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C. Harmonics suitable for rotation

In the previous subsection, we have completely determined the matrix elements for the rotations of our interest when applied to hyperspherical harmonics, before actually specifying these functions explicitly. This was possible because we expressed the matrix elements in terms of the labels (“quantum numbers”) \((\Lambda, \mu, \lambda)\) identifying the harmonics, rather than through integrals in coordinate space. So far, the harmonics are functions of the generic Cartesian coordinates \((x_j, y_j, z_j)\). Upon rotation from one Jacobi tree to another, these coordinates transform into another set \((x_j', y_j', z_j')\), \(j = 1, \ldots, N - 1\), but the harmonics, when expressed in terms of the new coordinates, retain their functional form. In this sense, these hyperspherical harmonics are frame-independent.

Direct solution of the angular Laplacian’s eigenvalue problem by separation of the \((3N - 4)\) coordinates in the second-order differential equation leads to hyperspherical harmonics represented by standard spherical harmonics and Jacobi or Gegenbauer polynomials, depending on the choice of hyperspherical coordinates \([10, 17, 21]\). However, these harmonics are not simultaneous eigenfunctions of all the \(H_j\). Consequently, they are not suitable for our purpose, because rotating the harmonics requires knowledge of the ladder operators’ effects on these functions, at least for the base set of ladder operators \(E_{\pm \epsilon_j}, j = 1, \ldots, \ell\), which in turn requires uniquely specifying the harmonics with labels \((\Lambda, \mu, \lambda)\). We now construct complete sets of functions defined exclusively by their behavior under the action of the first-order differential operators \(H_j\) and \(E_{\pm \epsilon_j}, j = 1, \ldots, \ell\). The resulting functions are “hyperspherical harmonics”, too, because they satisfy the generalized Laplacian’s symmetry under rotations.

The hyperspherical description separates the “hyperradial” momentum from the generalized angular momentum. Each of the Cartesian coordinates \(x_j, y_j, z_j, (j = 1, \ldots, N - 1)\), is proportional to the “hyperradius” \(R\). The angular Laplacian’s eigenvalue parameter \(\Lambda\) determines the harmonic’s degree by setting the radial scale as \(R^\Lambda\). Neither the \(H_j\) nor the ladder operators \(E_{\pm \epsilon_j}\) affect this radial factor, as is to be expected of angular momentum-like
operators. With \((3N - 4)\) angular coordinates, a complete specification of the harmonics requires \((3N - 5)\) labels in addition to \(\Lambda\). The vector \(\mu\) provides \(\ell = \lfloor(3N - 3)/2\rfloor\) of them in the form of the eigenvalues \(m_j\) of all the \(H_j\). The remaining labels are taken from the \(\ell\)-component vector \(\lambda\) consisting of the “string lengths” \(\lambda_{\epsilon_j}\) for the ladder operators \(E_{\pm \epsilon_j}\). The first \((\ell - 2)\) components of \(\lambda\) suffice to reach a total of \((3N - 4)\) labels if \(N\) is odd, as do the first \((\ell - 1)\) components for \(N\) even.

It remains to determine the function \(F\) introduced in (3.1). The requirement \(H_j F \equiv 0\) for all \(j\) implies that \(F\) depends only on \((u_j^2 + v_j^2)\) (and possibly on the single unpaired coordinate \(w\) in case \((3N - 3)\) is odd). Furthermore, if \(\sum_j |m_j| = \Lambda\) the product \(\prod_j (u_j + i v_j)^{m_j}\) already accounts for the radial factor \(R^\Lambda\), i.e., \(F = \text{const.}\) in this case. This circumstance suggests constructing complete sets of degenerate harmonics with the same \(\Lambda\) as follows: We set \(m_1\) to its maximum value and all other \(m_j = 0\), i.e., \(\mu = (\Lambda, 0, \ldots, 0)\) [26]. For \(j = 1, \ldots, \ell\), the modulus of the projection \(m_{\epsilon_j} = \mu \cdot \epsilon_j/(\epsilon_j \cdot \epsilon_j)\) sets the lower limit for the corresponding string length \(\lambda_{\epsilon_j}\). However, \(\lambda_{\epsilon_j}\) cannot be larger than \(m_{\epsilon_j}\) either, for applying any of the raising operators \(E_{+ \epsilon_j}\) necessarily results in a vector \(\mu' = \mu + \epsilon_j\) having \(\sum_j |m_j'| > \Lambda\), i.e., in a harmonic not belonging to the same set of degenerate functions. With \(\lambda\) completely specified, the first harmonic reads

\[
Y_{\Lambda, \mu, \lambda} = c_\Lambda (x_1 + iy_1)^\Lambda; \quad \lambda = (m_{\epsilon_1}, 0, \ldots, 0); \quad \mu = (\Lambda, 0, \ldots, 0),
\]

where \(c_\Lambda\) denotes a normalization constant. Starting from this first function, we generate harmonics by applying all the lowering operators \(E_{- \epsilon_j}\) first to (3.23), then applying the \(E_{- \epsilon_j}\) to the harmonics so obtained, and so on in a recursive procedure:

\[
Y_{\Lambda, \mu - \epsilon_j, \lambda'} = \frac{1}{\sqrt{(\lambda_{\epsilon_j} + m_{\epsilon_j})(\lambda_{\epsilon_j} - m_{\epsilon_j} + 1)}} E_{- \epsilon_j} Y_{\Lambda, \mu, \lambda}
\]

for all \(\epsilon_j\)-strings that have not yet terminated, i.e., the strings having \(m_{\epsilon_j} > -\lambda_{\epsilon_j}\). The recursive nature of the process suggests gathering together the harmonics in “levels”, with the level of a harmonic indicating the number of lowering operator steps separating it from the first harmonic (3.23). At level 0 with the harmonic (3.23), the \(\epsilon_1\)-string is the only string.
with non-zero length, and we can only generate one harmonic of level 1. But for \( \ell > 1 \), this level-1 harmonic will already have several non-vanishing \( m'_j \) and corresponding \( \lambda'_j \), giving rise to additional \( \epsilon_j \)-strings starting from this level. Repeating this procedure level by level, we work our way down until finally reaching the last harmonic with \( \mu = (-\Lambda, 0, \ldots, 0) \). The procedure stops automatically, because applying any of the lowering operators to this last harmonic maps it to zero (exactly as \( l^- Y_{l-1} \equiv 0 \) in three dimensions).

In \( d \) dimensions, the total number of different hyperspherical harmonics with the same “grand angular momentum” \( \Lambda \) is \([22, \text{p. 265}]\)

\[
\dim(\Lambda; \ d) = \frac{2\Lambda + d - 2}{\Lambda + d - 2} \left( \Lambda + d - 2 \right) - 2 \left( \Lambda - 2 \right).
\]  

(3.25)

For a given \( \Lambda \), the procedure outlined above generates exactly \( \dim(\Lambda, \ d) \) independent functions, i.e., a complete set of hyperspherical harmonics. However, some of these harmonics still require modifying for our purpose, as the following observations illustrate.

Note first that with our choice for \( E_{\epsilon_j} \), the first \((\ell - 1)\) ladder operators always change two \( m_j \) in opposite directions, thus leaving \( \sum_j |m_j| = \Lambda \) invariant when starting from the first harmonic. For all these harmonics, the function \( F \) reduces to a constant. At some stage during the above construction, however, an \( \epsilon_l \)-string will appear along which the sum \( \sum_j |m_j| \) no longer remains constant. Since the ladder operators do not change the overall radial factor \( R^\Lambda \), the function \( F \) accounts for any “missing powers” in \( \prod_j (u_j + iv_j)^{m_j} \) for harmonics having \( \mu \) with \( \sum_j |m_j| < \Lambda \). For instance, in three dimensions the lowering operator \( l^- \) (\( E_{-1} \) in (3.3c)’s notation) removes one power of \((x + iy)\) from the “highest” harmonic \( r^\ell Y_{\ell \ell} \sim (x + iy)^\ell = r^\ell \sin^\ell \theta \exp(i\ell \phi) \) while simultaneously adding a factor \( F = z = r \cos \theta \) to the next harmonic \( r^\ell Y_{l-1} \sim z(x + iy)^{\ell-1} = r^\ell \cos \theta \sin^{\ell-1} \theta \exp(i(\ell - 1)\phi) \). In the same way, repeated application of \( l_- \) generates the higher-degree Legendre polynomials in \( \cos \theta \) making up \( F \) in this case.

So far, \( \Lambda \) and the vector \( \mu \) provide enough information to uniquely specify the hyperspherical harmonics; we need their additional label \( \lambda \) only for the purpose of determining transformation matrix elements, not to distinguish the harmonics from one another. For
these harmonics, the construction described above provides the appropriate function $F$.

In a higher-dimensional setting, however, the $E_{\epsilon_j}$ do not all commute with each other. It is thus possible to arrive at different functions with the same $\mu$ along different ladder operator sequences, starting from the first harmonic (3.23). This is exactly the situation of “degenerate eigenvalues”: in this case, $\mu$, the set of eigenvalues of the $H_j$, has multiplicity higher than one. Although our construction generates the appropriate number of independent functions, thus providing a basis system for the higher-dimensional eigenspace associated with $\mu$, we now need additional labels—to be taken from $\lambda$—to distinguish the different eigenfunctions with degenerate eigenvalue $\mu$. Suppose, therefore, the vector $\mu$ occurs with multiplicity $\nu > 1$. Our procedure generates $\nu$ independent functions $\Phi_\rho$, $\rho = 1, \ldots, \nu$, all having the same $\prod_j (u_j + iv_j)^{m_j}$. These functions differ only in their $F$-Harmonics suitable for the calculation of rotation matrix elements are expressed as linear combinations

$$\Phi_\sigma = \sum_{\rho=1}^{\nu} a_{\sigma \rho} \Phi_\rho,$$  

(3.26)

and the requirement

$$\left(E_{+\epsilon_j}\right)^{\lambda_{\epsilon_j}-m_{\epsilon_j}+1} \Phi_\sigma = 0, \quad \text{for } j = 1, \ldots, j_{\text{max}}$$  

(3.27)

determines the coefficients $a_{\sigma \rho}$ for $\sigma = 1, \ldots, \nu$ (and thus the functions $F$). In this way, appropriate sets of parameters ($\lambda_{\epsilon_1}, \ldots, \lambda_{\epsilon_{j_{\text{max}}}}$) serve to distinguish the harmonics by enforcing specific lengths for the different $\epsilon_j$-strings. As noted previously, the number of additional labels required is $j_{\text{max}} = \ell - 2$ (or $\ell - 1$) for even (or odd)-dimensional spaces, respectively. With this modification, even the harmonics corresponding to degenerate vectors $\mu$ show the desired behavior under rotations in $d$ dimensions, and their transformation matrix elements can be deduced from their “quantum numbers” $(\Lambda, \mu, \lambda)$ directly.

Finally, a remark concerning the degeneracy of $\mu$ seems in order. In three dimensions, it is impossible to arrive at the same $\mu \equiv m$ along different strings, because there is only one pair of ladder operators. Nevertheless, even in this case we note that the $m$-components arising for a given $l$ ($\equiv \Lambda$) occur again for $l' > l$. Due to the mostly pairwise change of
m_j in higher-dimensional spaces, we expect that a vector \( \mu \) occurring for a given \( \Lambda \) will appear again as a label for harmonics with \( \Lambda' = \Lambda + 2, \Lambda + 4, \ldots \). For \( d > 4 \) there are non-commuting ladder operators. Because the number of different pathways leading from the highest harmonic to a specific \( \mu \) increases with the length of this path, \( \mu \)'s multiplicity increases with \( \Lambda \). Furthermore, since different functions \( F \) imply contributions from different \((u_j^2 + v_j^2)\)-terms, higher multiplicity—even in spaces with more than three dimensions—can only arise for \( \Lambda - \sum_j |m_j| \geq 2 \). A detailed analysis of the recursion in \( \Lambda \) with \( \mu \) held fixed confirms these expectations, yielding for the multiplicity of \( \mu \)

\[
\text{mult}(\mu; \Lambda, d) = \binom{p+q}{q} \quad (3.28)
\]

with

\[
p = \left\lfloor \frac{\Lambda - \sum_j |m_j|}{2} \right\rfloor; \quad q = \left\lfloor \frac{d-3}{2} \right\rfloor,
\]

a useful result to test the implementation of the (recursive) procedure.

IV. EXAMPLE: TWO-ELECTRON SYSTEM

For the purpose of illustration, we apply the method outlined in the preceding sections to a Coulombic three-body system. Specifically, we consider a two-electron atom or ion with atomic number \( Z \), i.e., a system with only one heavy particle. The case of diatomic molecules (two-center Coulomb system) requires additional modifications of the hyperspherical approach [27].

A. Coulomb interactions among three particles

After elimination of the CM motion, a three-particle system requires two Jacobi vectors for its description. We use three different Jacobi trees, \( \xi_i, \eta_i, \zeta_i, (i = 1, 2) \). With \( r_1, r_2 \), and \( r_N \) denoting the positions of the two electrons and the nucleus, respectively, the relevant mass-weighted Jacobi vectors read

\[
\xi_1 = \sqrt{\frac{M_e}{2}}(r_1 - r_2), \quad (4.1a)
\]
\[ \xi_2 = \sqrt{\frac{2M_e M_N}{M_N + 2M_e}} \left( \frac{r_1 + r_2}{2} - r_N \right); \quad (4.1b) \]

\[ \eta_1 = \sqrt{\frac{M_N M_e}{M_N + M_e}} (r_N - r_1) = \cos \beta \xi_1 + \sin \beta \xi_2; \quad (4.2) \]

\[ \zeta_1 = \sqrt{\frac{M_N M_e}{M_N + M_e}} (r_2 - r_N) = \cos \gamma \xi_1 + \sin \gamma \xi_2; \quad (4.3) \]

where

\[ \cos \beta = -\sqrt{\frac{M_N}{2(M_N + M_e)}}, \quad (4.4a) \]

\[ \sin \beta = -\sqrt{\frac{M_N + 2M_e}{2(M_N + M_e)}}, \quad (4.4b) \]

and \( \gamma = -\beta \). \( M_N \) and \( M_e \) denote the nuclear and electron mass, respectively. The vectors \( \eta_2 \) and \( \zeta_2 \) will not be needed, since the Coulomb interaction among the three particles takes the form

\[ V_C = \sqrt{\frac{M_e}{2}} \frac{1}{|\xi_1|} - \sqrt{\frac{M_N M_e}{M_N + M_e}} \left( \frac{Z}{|\eta_1|} + \frac{Z}{|\zeta_1|} \right). \quad (4.5) \]

At this point, the familiar approach using the transformation of the coordinates would exploit (4.2–4.3) in a multipole expansion of the electron-nucleus interactions in terms of the coordinates \( \xi_1 \) and \( \xi_2 \). In our method, however, we apply the (kinematic) rotations (4.2–4.3) to the wave functions instead. This amounts to calculating the three interaction matrix elements in three different coordinate systems. The immediately obvious advantage of this method is that each of the three terms takes exactly the same form. Each of the integrals reduces to the same radial integral as in the textbook example of hydrogen. Higher-order multipoles and nested integrations over powers of the coordinates do not occur.

**B. Choice of \( H_j \) and ladder operators**

The kinetic energy has rotational symmetry in six dimensions. Partitioning the six coordinates into pairs defines three non-intersecting planes, and thus \( \ell = 3 \) mutually commuting rotations. We choose the corresponding first-order operators \( H_j \) as \( J_{z_1 y_1} \), \( J_{x_2 y_2} \), and \( J_{z_1 z_2} \).
Here \( \{x_1, y_1, z_1, x_2, y_2, z_2\} \) denote the Cartesian components of the mass-weighted Jacobi vectors. Accordingly, the harmonics—simultaneous eigenfunctions of the three \( H_j \) with eigenvalues \( \boldsymbol{\mu} = (m_1, m_2, m_3) \)—take the form

\[
F(x_1^2 + y_1^2, x_2^2 + y_2^2, z_1^2 + z_2^2) \prod_{j=1}^{2} (x_j + i \text{sign}(m_j) y_j)^{|m_j|} (z_1 + i \text{sign}(m_3) z_2)^{|m_3|},
\]

where we have rewritten (3.1) so as to yield non-negative powers of the hyperradius for either sign of the \( m_j \). The appropriate base set of three ladder operator pairs \( E_{\pm \epsilon_j} \) is then specified by the vector labels \( \epsilon_1 = (1, -1, 0) \), \( \epsilon_2 = (0, 1, -1) \), and \( \epsilon_3 = (0, 1, 1) \), corresponding to \( E_{12}^{\pm}, E_{23}^{\pm}, \) and \( E_{23}^{\pm} \) in (3.3a–3.3b).

C. Labeling the basis functions

Five angles specify each point on a sphere of fixed hyperradius \( R \) in six-dimensional space. Consequently, our hyperspherical harmonics require five labels—related to the numbers of nodes in the five angular variables—for their identification. Besides \( \Lambda \) and \( \boldsymbol{\mu} = (m_1, m_2, m_3) \) we need one more label, \( \lambda_{\epsilon_1} \), the string length along the ladder operator sequences spanned by \( E_{\pm \epsilon_1} \).

The set of harmonics so defined differs from the more familiar set labeled by quantum numbers \( (l_1, m_1, l_2, m_2, n_\alpha) \) \[17,22\]. However, the latter set of harmonics does not make use of the third eigenvalue \( m_3 \). Since rotating the harmonics is accomplished by acting on them with ladder operators which in turn act on all the \( m_j \), the conventional harmonics specified by \( (l_1, m_1, l_2, m_2, n_\alpha) \) are not suited for our purpose. They may, of course, still serve as a basis set in an application, being then expanded into the new set labeled by \( (\Lambda, \boldsymbol{\mu}, \lambda_{\epsilon_1}) \) prior to the actual rotation. A straightforward transformation links the two basis sets.

D. Explicit construction of harmonics with degenerate \( \boldsymbol{\mu} \)

As an example, we derive the expressions for some harmonics that are not completely characterized by \( \Lambda \) and \( \boldsymbol{\mu} \). Consider for instance harmonics with \( \Lambda = 4, \boldsymbol{\mu} = (1, 0, 1)\).
According to (3.28), there are two harmonics with these labels, to be distinguished by the additional parameter \( \lambda_{\epsilon_1} \). With the particular vector \( \mu \) of this example, we obtain
\[ m_{\epsilon_1} = \frac{1}{2}, \]
thus setting the lower limit for \( \lambda_{\epsilon_1} \). The two essentially different ways of arriving at the set of labels \((1, 0, 1)\) from harmonics of the next-lower level are given by an \( E_{-\epsilon_1} \)-step from \( \mu' = (2, -1, 1) \), and by an \( E_{-\epsilon_1} \)-step from \( \mu'' = (1, 1, 2) \). Both of these labels have multiplicity 1 since \( \sum_j |m_j| = \Lambda \); the function \( F(\ldots) \) in the corresponding harmonics reduces to a normalization constant. Up to these normalizing factors, \( c_1 \) and \( c_2 \), (4.6) gives these harmonics as
\[
\begin{align*}
Y_{4,(2,-1,1),\frac{3}{2}} &= c_1 (x_1 + iy_1)^2(x_2 - iy_2)(z_1 + iz_2), \\
Y_{4,(1,1,2),1} &= c_2 (x_1 + iy_1)(x_2 + iy_2)(z_1 + iz_2)^2.
\end{align*}
\]
(4.7a) (4.7b)

Applying the appropriate ladder operators to these harmonics provides a basis for the two-dimensional eigenspace of degenerate \( \mu = (1, 0, 1) \):
\[
\begin{align*}
\Phi_1 &= E_{-\epsilon_1} Y_{4,(2,-1,1),\frac{3}{2}} \\
&= c_3 (x_1 + iy_1)(z_1 + iz_2) \left( [x_1^2 + y_1^2] - 2[x_2^2 + y_2^2] \right) \\
\Phi_2 &= E_{-\epsilon_1} Y_{4,(1,1,2),1} \\
&= c_4 (x_1 + iy_1)(z_1 + iz_2) \left( 2[x_2^2 + y_2^2] - [z_1^2 + z_1^2] \right),
\end{align*}
\]
with constant factors \( c_3, c_4 \) to be ultimately absorbed into the normalization. Because \( \Phi_1 \) is obtained from \( Y_{4,(2,-1,1),\frac{3}{2}} \) by applying \( E_{-\epsilon_1} \), it behaves under the relevant rotations exactly as is required for \( Y_{4,(1,0,1),\frac{3}{2}} \). Apart from the normalization constant \( c_3 \), we thus find
\[
Y_{4,(1,0,1),\frac{3}{2}} = c_3 (x_1 + iy_1)(z_1 + iz_2) \left( [x_1^2 + y_1^2] - 2[x_2^2 + y_2^2] \right).
\]
(4.9)

However, the second harmonic with the same \( \mu = (1, 0, 1) \) does not simply coincide with \( \Phi_2 \), because the latter has the same string-length \( \lambda_{\epsilon_1} = \frac{3}{2} \) as \( \Phi_1 \): One easily verifies that
\[
E_{\epsilon_1} \Phi_2 \neq 0.
\]
(4.10)

We can, however, construct a harmonic \( Y_{4,(1,0,1),\frac{1}{2}} \) as a linear combination of \( \Phi_1 \) and \( \Phi_2 \) by requiring

We can, however, construct a harmonic \( Y_{4,(1,0,1),\frac{1}{2}} \) as a linear combination of \( \Phi_1 \) and \( \Phi_2 \) by requiring
yielding the condition \(3ac_3 - 2bc_4 = 0\) and thus

\[
Y_{4,(1,0,1),\frac{1}{2}} = c_5(x_1 + iy_1)(z_1 + iz_2) (2[x_1^2 + y_1^2] + 2[x_2^2 + y_2^2] - 3[z_1^2 + z_2^2]).
\]

(4.12)

The two harmonics with “degenerate” \(\mu = (1, 0, 1)\) are now distinguished by their respective \(\lambda_{\epsilon_1}\).

\section*{E. Rotating the harmonics}

Eq. (4.12) describes the coordinate transformation between the Jacobi trees \((\xi_1, \xi_2)\) and \((\eta_1, \eta_2)\). Accordingly, the hyperspherical harmonics transform as

\[
|\Lambda, \mu', \lambda_{\epsilon_1}\rangle_{\xi} = \sum_{\mu} D^{(\lambda_{\epsilon_1})}_{\mu', \mu}(\beta) |\Lambda, \mu, \lambda_{\epsilon_1}\rangle_{\eta}
\]

(4.13)

where the subscripts on the ket vectors indicate the respective Jacobi tree. The transformation matrix elements are given by

\[
D^{(\lambda_{\epsilon_1})}_{\mu', \mu}(\beta) = \langle \Lambda, \mu', \lambda_{\epsilon_1} | \exp(i\beta[J_{x_1}x_2 + J_{y_1}y_2 + J_{z_1}z_2])|\Lambda, \mu, \lambda_{\epsilon_1}\rangle
\]

\[
= \langle \Lambda, \mu', \lambda_{\epsilon_1} | \exp(i\beta[E_{\epsilon_1} + E_{-\epsilon_1} + H_3])|\Lambda, \mu, \lambda_{\epsilon_1}\rangle,
\]

(4.14)

according to Subsec. III B. Following the analysis given there, we find

\[
D^{(\lambda_{\epsilon_1})}_{\mu', \mu}(\beta) = e^{i|m' - m|\pi/2}d^{(\lambda_{\epsilon_1})}_{m', m}(2\beta) e^{im_3\beta} \delta_{\mu', \mu + n_{\epsilon_1}},
\]

(4.15)

with

\[
m = \frac{1}{2}(m_1 - m_2)
\]

(4.16a)

\[
m' = \frac{1}{2}(m'_1 - m'_2)
\]

(4.16b)

\[
n = m'_1 - m_1 = -m'_2 + m_2
\]

(4.16c)

in terms of \(\mu\)-components. Replacing the angle \(\beta\) with \(\gamma = -\beta\) yields the expressions for the transformation to Jacobi tree \(\zeta\).
F. Symmetry properties of basis functions

Due to the very high degeneracy of harmonics with the same grand angular momentum \( \Lambda \) as expressed in (3.25), it is important to exploit various symmetries of the functions in order to reduce the size of the hyperspherical basis. Reflection through the origin of the coordinate system transforms all six coordinates into their negatives. According to (4.6), the harmonic \( |\Lambda, \mu, \lambda, \epsilon_1\rangle \) picks up a factor \((-1)^{|m_1|+|m_2|+|m_3|} \) under this operation. Since all the ladder operators change two of the \( m_j \) at a time, the sum in the exponent has the same parity as \( \Lambda \). This first observation thus restricts the basis set by allowing only even \( \Lambda \) for even-parity states and odd \( \Lambda \) for odd-parity states.

Consider next the harmonics’ symmetry under interchange of identical particles (i.e., the two electrons). This interchange is most easily described in the Jacobi tree \( \{\xi_1, \xi_2\} \):

\[
\xi_1 \leftrightarrow -\xi_1; \quad \xi_2 \leftrightarrow \xi_2. \tag{4.17}
\]

Accordingly, the harmonic \( |\Lambda, m_1, m_2, m_3, \lambda, \epsilon_1\rangle \) turns into the harmonic \( |\Lambda, m_1, m_2, -m_3, \lambda, \epsilon_1\rangle \) under interchange of the electrons while acquiring a factor \((-1)^{m_1+m_3+S} \) (where \( S \) denotes the total spin of the two electrons). Antisymmetrized basis functions may thus be labeled by non-negative \( m_3 \) only, and \( m_3 = 0 \) is possible only for harmonics with \((m_1 + S)\) even.

Furthermore, when choosing the first-order operators \( H_j \) we have arranged the six coordinates in a way that allows us to identify \( m_1 \) and \( m_2 \) with the \( z \)-projections of the three-dimensional relative angular momenta \( l_1 \) and \( l_2 \). Therefore, the sum \( m_1 + m_2 = m_{\text{tot}} \) is the \( z \)-component of the coupled (total) orbital angular momentum \( L \). The fact that the Coulomb interaction does not couple states with different \( m_{\text{tot}} \) reduces the size of a basis consisting of antisymmetrized harmonics accordingly. This point is particularly interesting because the system’s invariant \( m_{\text{tot}} \) restricts our basis, even though the corresponding total orbital angular momentum \( L \) is not defined in this basis. The reason for this seemingly surprising fact lies in our use of first-order operators only; hence \( L_z \) can be identified, but not the second order operators \( L_j^2 \) or \( L^2 \). The absence of the invariant three-dimensional \( L \)
is the main trade-off we have to accept when treating all transformations as rotations in a
genuinely six-dimensional space [22, especially Sec. 10.2., p. 267ff].

G. Calculation of matrix elements

While Cartesian coordinates prove most appropriate for manipulating the hyperspherical
harmonics using ladder operators, hyperspherical coordinates lend themselves for the calcu-
lation of matrix elements. Specifically, the familiar representation of Cartesian coordinates
\begin{align*}
x_1 &= R \cos \alpha \sin \theta_1 \cos \varphi_1, \\
y_1 &= R \cos \alpha \sin \theta_1 \sin \varphi_1, \\
z_1 &= R \cos \alpha \cos \theta_1, \\
x_2 &= R \sin \alpha \sin \theta_2 \cos \varphi_2, \\
y_2 &= R \sin \alpha \sin \theta_2 \sin \varphi_2, \\
z_2 &= R \sin \alpha \cos \theta_2,
\end{align*}
transforms (4.18) into
\begin{equation}
R^\Lambda F \sin^{\left|m_1\right|} \theta_1 e^{im_1 \varphi_1} \sin^{\left|m_2\right|} \theta_2 e^{im_2 \varphi_2} (\cos \alpha \cos \theta_1 + \text{sign}(m_3) \sin \alpha \cos \theta_2)^{|m_3|},
\end{equation}
where \( F \) is a function of \((\cos^2 \alpha \sin^2 \theta_1), (\sin^2 \alpha \sin^2 \theta_2), \) and \((\cos^2 \alpha \cos^2 \theta_1 + \sin^2 \alpha \cos^2 \theta_2)\). Note that this form remains the same, regardless of whether the Cartesian components
\((x, y, z)\) refer to the Jacobi vectors in tree \(\xi\), in \(\eta\), or in \(\zeta\). The relevant interaction operator entering into the matrix element is always \(1/(R \cos \alpha)\), for each of the three pairwise Coulomb
interactions, with the angle \(\alpha\) referring to a different coordinate system in each case. The \(\theta\)-
and \(\alpha\)-integrals arising in the calculation are related to the Euler Beta function [28], namely,
\begin{align*}
\int_0^{2\pi} d\phi \ e^{i(m'-m)\varphi} &= 2\pi \delta_{m'm}, \\
\int_0^\pi d\theta \ \sin^p \theta \ \cos^q \theta &= (1 + (-1)^q) \frac{(p-1)!!(q-1)!!}{(p+q)!!} \ c_{pq}, \\
\int_0^{\pi/2} d\alpha \ \sin^p \alpha \ \cos^q \alpha &= \frac{(p-1)!!(q-1)!!}{(p+q)!!} \ c_{pq},
\end{align*}
where
\begin{equation}
c_{pq} = \begin{cases} 
\pi & p, q \text{ both even} \\
1 & \text{otherwise}.
\end{cases}
\end{equation}
A multipole expansion would have forced us to split the last integral into two parts, with
different integrands depending on whether \(| \cos \beta \xi |\) is greater or smaller than \(| \sin \beta \xi |\). The
resulting integral could only be expressed as a sum of factorial quotients, rather than a single
term, as in (4.20c).

Gathering together all the pieces developed in this section, we find for the angular part
of the matrix elements

\[
\langle \Lambda', \mu', \lambda', \epsilon | V_C | \Lambda, \mu, \lambda \rangle = \langle \Lambda', \mu', \lambda' | \frac{1}{r_{12}} - \frac{Z}{r_{1N}} - \frac{Z}{r_{2N}} | \Lambda, \mu, \lambda \rangle
\]

(4.22)

\[
= \frac{1}{R} \sqrt{\frac{M_e}{2}} \langle \Lambda', \mu', \lambda' | \frac{1}{\cos \alpha} | \Lambda, \mu, \lambda \rangle - \frac{Z}{R} \sqrt{\frac{M_N M_e}{M_N + M_e}} \times
\]

\[
\times \sum_{\mu_1, \mu_2} \left\{ \left( D^{(\lambda')}_{\mu_1, \mu} (\beta) \right)^\dagger D^{(\lambda')}_{\mu_2, \mu} (\beta) + \left( D^{(\lambda')}_{\mu_1, \mu} (\gamma) \right)^\dagger D^{(\lambda')}_{\mu_2, \mu} (\gamma) \right\} \times
\]

\[
\times \langle \Lambda', \mu_1, \lambda' | \frac{1}{\cos \alpha} | \Lambda, \mu_2, \lambda \rangle,
\]

(4.23)

with the further simplification \(\gamma = -\beta\). The double summation (transforming the bra and
ket vectors between Jacobi trees) seems to spoil the present approach’s advantage over a
multipole expansion. After all, the latter also leads to two summations, namely, a sum
over the multipole order and another summation stemming from the analytical evaluation
of the nested integral over \(\alpha\). Note, however, that the present method achieves significantly
more with two summations: it accounts for all the cusps in the wave functions whenever an
inter-particle separation vanishes.

Finally, the particular case of a three-body system involves two relevant Jacobi vectors
only. All possible Jacobi trees are thus related to one another by rotations in the same plane
\((\xi_1, \xi_2)\). This particularity of the three-body problem might suggest arranging the Cartesian
coordinates of the Jacobi vectors in the following way:

\[
\{ x_1, x_2, y_1, y_2, z_1, z_2 \},
\]

(4.24)

rather than our choice (3.3). The \(H_j\) resulting from the above arrangement of coordinates
coincide with \(J_{x_1 x_2}, J_{y_1 y_2}, \) and \(J_{z_1 z_2}\), thereby simplifying the rotation of harmonics: All
rotations reduce to the first case of \((z_1 z_2)\)-type rotations discussed in Sec. [1113] with matrix
This orthogonality relation virtually eliminates the double summation over $\mu_1, \mu_2$ in (4.23). However, the corresponding harmonics are now functions of $(x_1 \pm ix_2)$, $(y_1 \pm iy_2)$, and $(z_1 \pm iz_2)$. Transforming to hyperspherical coordinates using

$$
\begin{align*}
  x_1 &= R \sin \alpha_1 \cos \alpha_2 \cos \varphi_1, \\
  y_1 &= R \sin \alpha_1 \sin \alpha_2 \cos \varphi_2, \\
  z_1 &= R \cos \alpha_1 \cos \varphi_3, \\
  x_2 &= R \sin \alpha_1 \cos \alpha_2 \sin \varphi_1, \\
  y_2 &= R \sin \alpha_1 \sin \alpha_2 \sin \varphi_2, \\
  z_2 &= R \cos \alpha_1 \sin \varphi_3,
\end{align*}
$$

yields $H_j = -i \frac{\partial}{\partial \varphi_j}$, with $0 \leq \varphi_j \leq 2\pi$ and $0 \leq \alpha_k \leq \pi/2$. While the $H_j$, as well as the harmonics, now obviously attain their simplest form, the choice (4.24) has two serious drawbacks: (i) None of the three eigenvalues $m_j$ of the $H_j$ have physical significance, and (ii) $1/|\xi_1| = (x_1^2 + y_1^2 + z_1^2)^{-1/2}$ is a (complicated) function of all five angles ($\alpha_1, \alpha_2, \varphi_1, \varphi_2, \varphi_3$). The latter problem is solved by using hyperspherical coordinates (4.18) instead of (4.26), yielding for each of the combinations $(x_1 \pm ix_2)$, $(y_1 \pm iy_2)$, and $(z_1 \pm iz_2)$ a sum of two terms (as opposed to the single terms obtained for $(x_j \pm iy_j)$ in the previous sub-sections). Expanding powers of these binomials leads to two additional summations, leaving us with no net gain.

V. CONCLUSION

Recognizing the independent-particle model’s failure to account for cusps due to variables on which the wave function does not depend explicitly, we have developed a method that satisfies Kato’s cusp condition through reference frame transformations. By transforming the wave function to the appropriate reference frame, we expose the wave function’s cusp arising from vanishing of any given particle separation $r_{ij}$. In addition to satisfying the cusp condition on the wave function, this technique also simplifies the calculation of the pairwise Coulomb interaction $\sim 1/r_{ij}$, as compared to the conventional multipole expansion.
Implementation of the approach outlined above resolves into three major tasks, all addressed in the present investigation: (i) the systematic study of the relevant transformations between reference frames; (ii) the definition of functions suitable for such transformations; (iii) the determination of the transformation matrix. While the literature on Lie algebra provides ready-made solutions to problem (i), it usually fails to do so for (ii) and (iii). Furthermore, the mathematical literature does not exploit the particularity of an atomic or molecular $N$-body system.

More specifically, problem (i) is solved by using mass-scaled Jacobi coordinates, since the transformations between reference frames reduce then to generalized rotations in $(3N - 3)$ dimensions. The Lie algebra $\mathfrak{so}(3N - 3)$ describes these transformations completely, embodied in the sets of commuting rotation operators $\{H_j\}$ and ladder operators $\{E_{\pm \epsilon_j}\}$, $j = 1, \ldots, [(3N - 3)/2]$. To solve problem (ii) mentioned above, we have introduced basis functions defined entirely through their behavior under infinitesimal rotations, i.e., when acted upon by the operators $H_j$ and $E_{\pm \epsilon_j}$. Simultaneous eigenfunctions of all the $H_j$ constitute appropriate basis functions, classified further according to their matrix elements for the transformation between reference frames. The latter step removes possible ambiguities whenever the $H_j$ have degenerate eigenvalues. Finally, by extending the concept of Euler-angle rotations from three to higher dimensions, we have provided the solution to task (iii), the determination of the transformation matrix elements. For an arbitrary $N$-body system, each change of reference frames considered here resolves into a sequence of basic transformations described by these matrix elements.

In an $N$-body system, the high dimensionality arises from the product structure of the $(N - 1)$-dimensional particle-space and the three-dimensional physical space. The basic step in the transformation between reference frames reduces to a two-dimensional rotation in a plane of particle-space. Upon expansion of the particle-space variables into their physical-space components, the basic rotation induces three two-dimensional rotations in $(3N - 3)$-dimensional space that are analyzed using the Lie algebra $\mathfrak{so}(3N - 3)$. To take advantage of the efficiency offered by the Lie algebraic method, we treat the transformations as rotations.
in a genuine \((3N-3)\)-dimensional space. Thus, we work with the \emph{first-order} differential operators \(H_j\) and \(E_{\pm\epsilon_j}\) throughout. Arranging the generic Cartesian components \((x_j, y_j, z_j)\) of the particle-space variables \(\xi_j\) appropriately, we can \emph{partially} recover the product structure characteristic for the physical application at hand: The eigenvalues of \((N-1)\) among the \(H_j\) carry physical significance and we interpret them as \(z\)-projections of orbital angular momenta. Their sum represents the total angular momentum’s \(z\)-projection, an invariant of the system. However, neither the individual angular momenta, nor the coupled (total) angular momentum can appear in our treatment, as they are represented by second-order operators. Other methods exploit the product structure of the \((3N-3)\)-dimensional coordinate space to a larger extent by solving the Laplacian’s (second-order) eigenvalue problem through separation of variables. However, the resulting hyperspherical harmonics incorporate these features of the three-dimensional physical space at considerable cost: Transforming these functions between reference frames proves very inefficient, requiring essentially their expansion into the equivalent sets of harmonics introduced in the present investigation.

In conclusion, we have demonstrated by explicit construction the possibility of describing a system of \(N\) charged particles without recourse to multipole expansions. Kato’s cusp condition is satisfied implicitly through reference frame transformations of the \emph{wave function}. We introduced appropriate basis functions and discussed their symmetries under particle interchange and reflection through the origin. We derived the matrix elements for the transformation between reference frames for arbitrary numbers of particles, and we showed that the interaction matrix elements attain a simpler form as compared to the conventional multipole expansion. Because the formalism underlying our approach relies exclusively on first-order differential operators, it does not incorporate angular momenta, thereby limiting its usefulness for bound state problems. However, the technique introduced here provides significant simplifications in scattering problems where partial angular momenta are not resolved.
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[25] In the language of Lie algebra this procedure amounts to identifying $A_1$ sub-algebras in $B_\ell$ or $D_\ell$ (the complexifications of $so(d)$ for $d$ odd or even, respectively). See e.g. pp. 507ff, p. 538, and Appendices F and I in [24].
The procedure outlined in the text generates only weights of real, symmetric representations. As these representations account for all sets of harmonics needed in our applications, irreducible representations of $so(d)$ with highest weight other than $(\Lambda,0,\ldots,0)$ are not considered here.

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