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A spectral method for integration of the time-dependent Schrödinger equation in hyperspherical coordinates

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Abstract

We describe a spectral method for the direct numerical calculation of the time-dependent Schrödinger equation described in hyperspherical coordinates. The method is based on the split-step technique where the wavefunction is expanded in the appropriate eigenfunctions for the partial operators, making the time integration efficient, accurate and simple. The fast Fourier transform is applied to produce the expansion in the hyperradial direction, and a general hyperspherical harmonics transformation is applied to create an expansion in the angular directions. The latter transformation is set up by a combination of spherical harmonics and Jacobi polynomials. The method is ideal to describe correlated ionization dynamics of two-electron systems in strong fields and other phenomena where a hyperradial expansion is efficient.

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1. Introduction

The three-body problem remains a challenge in a range of areas of theoretical and computational physics now more than 100 years after the monumental work of Poincaré, showing its non-separability [1]. For example, in classical mechanics the non-separability is one origin of chaos [2], while in quantum mechanics it plays a central role in the understanding of three-particle structure in atomic [3, 4], nuclear [5] and particle physics [6]. In atomic physics, recent research has been focused towards the understanding of the spectra of excited and loosely bounded states [7].

It is well known that any three-body problem can be described by three sets of Jacobi coordinates which decouple from the centre-of-mass motion, thus reducing the problem from

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a nine-dimensional problem to an effective six-dimensional problem. The advantage of the Jacobi coordinates is that the Laplacian of the total system is a simple sum over the Laplacians corresponding to each Jacobi coordinate. Hence, no polarization terms enter the description. In atomic physics, the six-dimensional problem has been solved numerically for fully correlated two-electron dynamics in the time domain based on a coarse Cartesian grid [8] or a spherical basis functions' expansion relating to each electron [9].

Alternatively, the problem may be addressed based on hyperspherical coordinates, i.e. a hyperradius $\rho = \left(\sum_{i=1}^{6} x_i^2\right)^{1/2}$ and a set of five independent angles [10]. In structure theory, this transformation has for more than 30 years been applied to classify excited and loosely bound states in nuclear and atomic physics [3–5]. The advantage of these coordinates is a near decoupling of the hyperradial coordinate from the angular part, which opens for Born–Oppenheimer-like separation methods. Correlated break-up processes, caused by a strong short laser pulse, are an example of a dynamical process which may be conveniently described by hyperspherical coordinates. In fact, this process has very recently been studied in the time domain based on a 'close coupling' expansion of the wavefunction in hyperspherical basis states [11].

In this paper we develop an algorithm for direct numerical calculation of the solution to the time-dependent Schrödinger equation in hyperspherical coordinates without reference to a precalculated spectrum of basis states. In addition, no time-consuming calculations of matrix elements of the operators are needed. The method is based on a splitting of the Hamiltonian operator and is a generalization of the time-propagation algorithms for spherical geometry first published by Hermann and Fleck for potentials with azimuthal symmetry [12], and recently generalized to potentials of arbitrary spatial dependence [13]. In the following section the mathematical problem is defined, followed by a section which describes the propagation scheme. In section 4 we demonstrate the method on two well-known static problems and in section 5 we conclude and comment on the application of the present method to time-dependent problems.

2. The Schrödinger equation in hyperspherical coordinates

We consider three particles in three dimensions, i.e., nine degrees of freedom. The masses, coordinates and momenta of the particles are m_i, $\mathbf{r}_i$ and $\mathbf{p}_i (i = 1, 2, 3)$, and the total mass is $M = m_1 + m_2 + m_3$. The Hamiltonian is given by

$$\hat{H} = \sum_{i=1}^{3} \frac{p_i^2}{2m_i} + \sum_{i=1;\, j>i}^{3} V_{ij}(|\mathbf{r}_i - \mathbf{r}_j|) + V_{\mathrm{E}}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, t), \tag{1}$$

where V_{ij} denotes the two-particle interactions between particles i and j, while V_{E} is a general external time-dependent interaction. We follow [14] and for each $i = 1, 2, 3$ we define the ith set of Jacobi coordinates $(\mathbf{x}_i, \mathbf{y}_i)$ as

$$\begin{aligned} \mathbf{x}_i &= \mu_{jk}(\mathbf{r}_j - \mathbf{r}_k), & \mu_{jk} &= \sqrt{\frac{m_j m_k}{m(m_j + m_k)}}, \\ \mathbf{y}_i &= \mu_i \left(\mathbf{r}_i - \frac{m_j \mathbf{r}_j + m_k \mathbf{r}_k}{m_j + m_k} \right), & \mu_i &= \sqrt{\frac{m_i (m_j + m_k)}{mM}}, \end{aligned} \tag{2}$$

where m is a normalization mass. Each of the sets $\{i, j, k\} = \{1, 2, 3\}, \{2, 3, 1\}, \{3, 1, 2\}$ combined with the centre-of-mass (CM) coordinate, $M\mathbf{R} = \sum_{i=1}^{3} m_i \mathbf{r}_i$, describes the system completely.

The space-fixed hyperspherical coordinates $\left(\rho, \alpha_i, \Omega_i^x, \Omega_i^y\right)$ are

$$x_i = \rho \sin\alpha_i, \qquad y_i = \rho \cos\alpha_i, \tag{3}$$

where ρ is the hyperradius and α_i is the hyperangle confined by $0 \leqslant \alpha_i \leqslant \pi/2$. For clarity, we suppress the i-index in the rest of this exposition.

The angular parts of $\mathbf{x}$ and $\mathbf{y}$ are denoted by Ω^x and Ω^y, each representing a pair of spherical angles (θ, ϕ). The total set of five angular coordinates α, Ω^x, Ω^y is denoted by Ω. The corresponding volume element is

$$\begin{aligned} \mathrm{d}\mathbf{x}\,\mathrm{d}\mathbf{y} &= \rho^5\,\mathrm{d}\rho \sin^2\alpha \cos^2\alpha\,\mathrm{d}\alpha\,\mathrm{d}\Omega^x\,\mathrm{d}\Omega^y \\ &\equiv \rho^5\,\mathrm{d}\rho\,\mathrm{d}\Omega. \end{aligned} \tag{4}$$

The kinetic energy operator in hyperspherical coordinates is given by

$$\hat{T} = \frac{\hbar^2}{2m}\left[-\frac{\partial^2}{\partial\rho^2} - \frac{5}{\rho}\frac{\partial}{\partial\rho} + \frac{\hat{\Lambda}^2}{\rho^2}\right], \tag{5}$$

$$\hat{\Lambda}^2 = -\frac{\partial^2}{\partial\alpha^2} - 4\cot(2\alpha_i)\frac{\partial}{\partial\alpha} + \frac{\hat{l}_x^2}{\sin^2\alpha} + \frac{\hat{l}_y^2}{\cos^2\alpha}, \tag{6}$$

where $\hat{l}_x^2$ and $\hat{l}_y^2$ are the angular momentum operators corresponding to $\mathbf{x}$ and $\mathbf{y}$, respectively [10].

Expressed in hyperspherical and CM coordinates, equation (1) becomes

$$\hat{H} = \frac{P^2}{2M} + \hat{T} + \sum_{i=1;\,j>i}^{3} V_{ij}(\rho, \Omega) + V_{\mathrm{E}}(\rho, \Omega, t), \tag{7}$$

where $\mathbf{P}$ is the centre-of-mass momentum which is decoupled from the internal hyperspherical coordinates. The solution for the centre-of-mass motion is thus trivial and the remaining challenge becomes to solve the internal problem. We note that the expression for the potentials in hyperspherical coordinates may be quite involved. For Coulombic interactions however, the expression is relatively simple and given explicitly in [19].

3. The propagation scheme

The time-dependent Schrödinger equation for the internal problem in the chosen set of hyperspherical coordinates is

$$\mathrm{i}\hbar\frac{\partial}{\partial t}\Psi = (\hat{T} + V)\Psi, \tag{8}$$

with $\Psi(\rho, \Omega, t)$ the wavefunction, $\hat{T}$ defined in equation (5) and the total potential is $V = \sum_{i=1;\,j>i}^{3} V_{ij}(\rho, \Omega) + V_{\mathrm{E}}(\rho, \Omega, t)$. Defining the reduced wavefunction $\Psi(\rho, \Omega, t) = \Phi(\rho, \Omega, t)/\rho^{5/2}$ and plugging in for $\hat{T}$, we arrive at the equation

$$\mathrm{i}\hbar\frac{\partial}{\partial t}\Phi = -\frac{\hbar^2}{2m}\left[\frac{\partial^2}{\partial\rho^2}\Phi - \frac{\hat{\Lambda}^2 + 15/4}{\rho^2}\Phi\right] + V\Phi, \tag{9}$$

which is at the heart of the present method. The solution of this equation is based on a generalization of our recent split-operator algorithm for two-particle dynamics [13]. We introduce $A = -\frac{\hbar^2}{2m}\frac{\partial^2}{\partial\rho^2}$ and $B = \frac{\hbar^2}{2m\rho^2}(\hat{\Lambda}^2 + 15/4)$ and write the wavefunction at time $t + \Delta t$ with Δt small as

$$\Phi(t + \Delta t) = \mathrm{e}^{\frac{-\mathrm{i}\Delta t A}{2\hbar}}\,\mathrm{e}^{\frac{-\mathrm{i}\Delta t B}{2\hbar}}\,\mathrm{e}^{\frac{-\mathrm{i}\Delta t V}{\hbar}}\,\mathrm{e}^{\frac{-\mathrm{i}\Delta t B}{2\hbar}}\,\mathrm{e}^{\frac{-\mathrm{i}\Delta t A}{2\hbar}}\,\Phi(t) + \mathcal{O}(\Delta t^3). \tag{10}$$

The error term represents the splitting error and it disappears when the operators commute. For time-dependent potentials, the linear Hamiltonian we are working with has an error term $\left(\sim\frac{\partial V}{\partial t}\Delta t^2\right)$, i.e. of second order [15].

The spatial approximation is based on an expansion of the reduced wavefunction $\Phi(\rho,\Omega,t)$ in the hyperspherical harmonics of equation (19), defined on a finite set of points $\{\rho_i,\Omega_j\}$,

$$\Phi(\rho_i,\Omega_j,t)=\sum_{\mathcal{K}} f_{\mathcal{K}}(\rho_i,t)\mathcal{Y}_{\mathcal{K}}(\Omega_j), \tag{11}$$

with $\mathcal{K}$ denoting the set of hyperspherical quantum numbers $\mathcal{K}=\mathcal{K}(n,l_x,l_y)$. The functions $\mathcal{Y}_{(n,l_x,l_y)}(\Omega)$ are an orthonormal set of eigenfunctions of the 'grand' angular momentum operator of equation (6) with eigenvalues $K(K+4)$, i.e.

$$\hat{\Lambda}^2\mathcal{Y}_{(n,l_x,l_y)}(\Omega)=K(K+4)\mathcal{Y}_{(n,l_x,l_y)}(\Omega). \tag{12}$$

Here, K is the quantum number defined by

$$K=2n+l_x+l_y. \tag{13}$$

The application of the time operator of equation (10) involves first that each $f_{\mathcal{K}}(\rho,t)$ is calculated by the projection

$$f_{\mathcal{K}}(\rho_i,t)=\int \mathrm{d}\Omega\,\mathcal{Y}^*_{\mathcal{K}}(\Omega)\Phi(\rho_i,\Omega,t). \tag{14}$$

Then, each $f_{\mathcal{K}}(\rho_i,t)$ is represented in momentum space,

$$f_{\mathcal{K}}(\rho_i,t)=\sum_k g_{\mathcal{K},k}\,\mathrm{e}^{\mathrm{i}(\pi/\rho_{\max})k\rho_i}, \tag{15}$$

by applying a fast Fourier transform. Here, $\rho_{\max}$ denotes the grid size in the hyperradius ρ. Applying the kinetic energy operator then results in a multiplicative factor on each Fourier component, i.e.

$$\mathrm{e}^{\frac{-\mathrm{i}\Delta t A}{2\hbar}}g_{\mathcal{K},k}=\mathrm{e}^{\mathrm{i}(\pi/\rho_{\max})^2\hbar k^2/4m\Delta t}g_{\mathcal{K},k}. \tag{16}$$

The inverse Fourier transform is then applied, and the effect of the 'grand' angular momentum operator is evaluated as

$$\mathrm{e}^{-\mathrm{i}\Delta t B/2\hbar}f_{\mathcal{K}}(\rho_i,t)=\mathrm{e}^{-\mathrm{i}\Delta t(K(K+4)+15/4)\frac{\hbar}{4m\rho_i^2}}f_{\mathcal{K}}(\rho_i,t). \tag{17}$$

Then, the total wavefunction is reconstructed by equation (11) and the propagator of the external potential is evaluated by

$$\Phi(\rho_i,\Omega_j,t+\Delta t)=\mathrm{e}^{-\mathrm{i}\Delta t V(\rho_i,\Omega_j,t)/\hbar}\Phi(\rho_i,\Omega_j,t). \tag{18}$$

Finally, the operations of equations (14)–(17) are repeated in inverse order to complete the propagation of a single time step. The procedure continues a specified number of time steps to obtain the final wavefunction. The initial wavefunction can be found by propagation in imaginary time as described in section 5.

4. The hyperspherical expansion

The complete set of normalized hyperspherical harmonics is given, in the product representation, as

$$\mathcal{Y}_{(n,l_x,l_y)}(\Omega)=\phi^{(n,l_x,l_y)}(\cos(2\alpha))\left[Y_{l_x,m_x}(\Omega^x)Y_{l_y,m_y}(\Omega^y)\right], \tag{19}$$

where $Y_{l_x m_x}(\Omega^x)$ and $Y_{l_y m_y}(\Omega^y)$, the standard orthonormal spherical harmonics, are the simultaneous eigenfunctions of $\hat{l}_x^2$ and $\hat{l}_y^2$, respectively. Here the function, $\phi^{(n,\gamma,\beta)}$, is the normalized nth degree Jacobi polynomial which, with $x = \cos(2\alpha)$, forms an orthogonal set under the inner product

$$(f, g) = \int_{-1}^{1} \mathrm{d}x\, w(x) f(x) g(x), \tag{20}$$

$w(x)$ being the weight function $w(x) = (1-x)^\gamma (1+x)^\beta$. In our computation, we apply the variable transformation $x = \cos(2\alpha)$, corresponding to equation (3).

The quantum numbers n, l_x and l_y are input parameters of the algorithm and for a given set of these it is important to reconstruct K. The solutions can be ordered into degenerated subspaces according to each value of $K = 0, 1, 2, \ldots$ with parity $(-1)^K$. In structure calculations, it is beneficial to apply linear combinations of these basis functions which directly relates to the symmetries of the various states [4]. In the present numerical implementation, there is no need to do this.

The main challenge of the above-outlined algorithm is to construct a discrete representation and the associated transformation for the hyperspherical harmonics which is unitary and thus conserves the norm. For any set of quantum numbers $\mathcal{K}$ in the expansion equation (11), we need a quadrature rule over the hypersphere which satisfies

$$\delta_{\mathcal{K}\mathcal{K}'} = \int_\Omega \mathrm{d}\Omega\, \mathcal{Y}^\star_{\mathcal{K}}(\Omega)\mathcal{Y}_{\mathcal{K}'}(\Omega) = \sum_{i=1}^{N_\Omega} w_i \mathcal{Y}^\star_{\mathcal{K}}(\Omega_i)\mathcal{Y}_{\mathcal{K}'}(\Omega_i), \tag{21}$$

with minimum number, N_Ω, of hyperangular mesh points Ω_i and associated weights w_i. The present construction rests on our recent procedure [13] for the three-dimensional problem and it is therefore instructive to review this procedure first.

For fixed m of the spherical harmonics, $Y_{l,m}(\theta, \phi)$, the discrete representation maintains the unitarian property if the θ_j's are chosen as the Gauss–Jacobi quadrature points with weight function determined by m [12]. However, when the azimuthal symmetry in V is broken, m is not conserved, and consequently the Gauss–Jacobi points and weights depend on m which again makes the method unattractive. This implies that a direct discretization of the orthonormalization relation for spherical harmonics,

$$\delta_{l,l'}\delta_{m,m'} = \int \sin\theta\, \mathrm{d}\theta\, \mathrm{d}\phi\, Y^*_{l'm'}(\Omega^x) Y_{lm}(\Omega^x), \tag{22}$$

cannot be constructed along these lines. The standard solution from related mathematical physics has been to re-expand the spherical harmonics in a basis of associated Legendre polynomials $P_l^m(\theta)$ and a Fourier basis in ϕ on a regular grid, since $Y_{lm}(\theta, \phi) = P_l^m(\theta)\, \mathrm{e}^{\mathrm{i}m\phi}$. Alternatively, we may take any interpolatory quadrature rule over the sphere which integrates exactly all polynomials of degree less than or equal to $2l_{\max}$. Recently, abscissas and weights for such rules were computed [16] and made available over the Internet [17]. Taking Ω_j^x, w_j as the set of abscissas and weights then ensures that the standard orthogonality property is fulfilled:

$$\delta_{l,l'}\delta_{m,m'} = \sum_{j=1}^{N_x} w_j Y^*_{l'm'}\left(\Omega_j^x\right) Y_{lm}\left(\Omega_j^x\right), \tag{23}$$

based on $N_x = (2l_{\max}+1)^2$ grid points.

This integration procedure can be directly applied for the two spherical harmonics which is needed to construct $\mathcal{Y}_{\mathcal{K}}$, cf equation (19). In addition, a similar procedure for the Jacobi polynomials must be constructed. To evaluate the coefficients of an expansion in a specific

set of Jacobi polynomials, the appropriate integrals should be evaluated by a Gauss–Jacobi quadrature rule which takes as its abscissa set the N_J zeros of $P_{N_J}^{\gamma,\beta}(x)$. [18]. Software for calculating the appropriate abscissas and weights based on [18] is available online and is used in our routines. For a given specification of $N_J, l_{x_{\max}}, l_{y_{\max}}$, we here need a common set of grid points which is independent of the actual values of n, l_x and l_y. However, the different weight functions produce different abscissa sets. We circumvent this problem by taking as our fundamental set of Jacobi polynomials those corresponding to $\gamma = \beta = 1/2$. We then simply compute

$$g(x_j) = (1+x_j)^{l_x}(1-x_j)^{l_y}\Phi(\rho, x_j, \Omega^x, \Omega^y) \tag{24}$$

and evaluate

$$c_n^{l_x+1/2,l_y+1/2} = \sum_{j=0}^{N} w_j^{(n,lx,ly)} P_n^{l_x+1/2,l_y+1/2}(x_j), \tag{25}$$

where

$$w_j^{(n,lx,ly)} = g(x_j)(1+x_j)^{1/2}(1-x_j)^{1/2}. \tag{26}$$

While these coefficients will be evaluated exactly for any $\Phi(\rho, x_j, \Omega^x, \Omega^y)$ being a polynomial in x of degree less than or equal to N_x, this is no longer the case when using (25). To obtain the same accuracy and ensure orthogonality of the discrete basis functions when $l_x + l_y > 0$, we now need a quadrature rule which integrates polynomials of degree $2N_J - 1 + l_{x_{\max}} + l_{y_{\max}}$. This will be the one based on $P^{1/2,1/2}_{N_J+(l_{x\max}+l_{y\max})/2}(x)$.

Equation (21) can finally be expressed as

$$\delta_{\mathcal{K}\mathcal{K}'} = \int \mathrm{d}\Omega\, \mathcal{Y}^\star_{\mathcal{K}}(\Omega)\mathcal{Y}_{\mathcal{K}'}(\Omega) = \sum_{i=1}^{N_\Omega} w_i \mathcal{Y}^\star_{\mathcal{K}}(\Omega_i)\mathcal{Y}_{\mathcal{K}'}(\Omega_i) \tag{27}$$

$$= \sum_{h=1}^{N_\alpha}\sum_{j=1}^{N_x}\sum_{k=1}^{N_y} w_h w_j w_k \mathcal{Y}^\star_{\mathcal{K}}\left(\alpha_h, \Omega^x_j, \Omega^y_k\right)\mathcal{Y}_{\mathcal{K}'}\left(\alpha_h, \Omega^x_j, \Omega^y_k\right), \tag{28}$$

with the total number of points $N_\Omega = N_x N_y N_\alpha = \left(2l_{x_{\max}}+1\right)^2\left(2l_{y_{\max}}+1\right)^2\left(N + l_{x_{\max}} + l_{y_{\max}} + 1\right)$.

Each of the expansion coefficients of equation (14) is calculated by this summation with $\mathcal{Y}_{\mathcal{K}'}$ replaced by Φ. This calculation dominates the total cost of one time step, while the time propagation itself (equations (16) and (17)) is much less.

We note that the present algorithm is unitary and thus completely conserves the norm of the wavefunction for all real potentials.

5. Test examples

In this section we demonstrate the method by calculating the ground state on a chosen grid by the method of imaginary time propagation, i.e. by letting $t \to -\mathrm{i}\tau$ in equation (10). This method is an ideal starting point for obtaining the spectrum of any time-independent Hamiltonian. The propagation becomes

$$\Phi[\mathrm{i}(\tau + \Delta\tau)] = \mathrm{e}^{-\Delta\tau \hat{H}/\hbar}\Phi(\mathrm{i}\tau). \tag{29}$$

Thus, by starting the propagation at $\tau = 0$ with an arbitrary wavefunction, it evolves in terms of the spectrum of eigenstates ξ_n as

$$\Phi[\mathrm{i}(\tau + \Delta\tau)] = \sum_{n=0}^{\infty} c_n(\tau)\,\mathrm{e}^{-\Delta\tau\varepsilon_n/\hbar}\xi_n(\mathbf{r}). \tag{30}$$

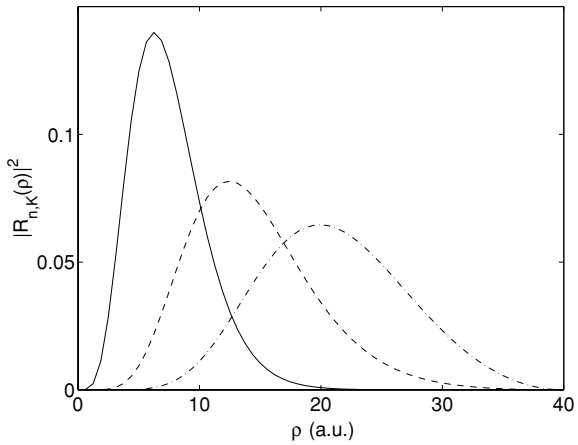


Figure 1. The three lowest squared hyperradial eigenstates of the six-dimensional Coulomb problem. Solid line: $K = 0$; dashed line: $K = 1$; dash-dotted line: $K = 2$.

The effect of the propagator is to exponentially damp all excited states compared with the ground state and thus leaving only the ground state, i.e. $\lim_{\tau\to\infty} \Phi(\tau) = \xi_0(\mathbf{r})\, \mathrm{e}^{-\Delta\tau\varepsilon_0/\hbar}$, with ε_0 being the ground-state energy. In the following we demonstrate the applicability of the algorithm by obtaining a set of known initial states on the grid when starting from a random initial state, in this case $\forall i, j : \Phi(\rho_i, \Omega_j, \tau = 0) = 1$. For simplicity, atomic units are used ($\hbar = m_e = e = 1$) in the remaining part of this section.

5.1. Six-dimensional hydrogen atom

The analytic Coulomb problem in d dimensions is well known [20]. For $d = 5$, it corresponds directly to the present method. To single out any state of the spectrum, we introduce the hyperradial projection Coulomb potential

$$V = V(\rho) = -\frac{\leftarrow[\int \mathrm{d}\Omega\, \mathcal{Y}_{\mathcal{K}}(\Omega)][\int \mathrm{d}\Omega\, \mathcal{Y}^{*}_{\mathcal{K}}(\Omega)]\rightarrow}{\rho}, \tag{31}$$

where the integral operators act to the left and right as indicated by subscript arrows. For this potential, the time-independent Schrödinger equation may be brought to the form

$$\left(-\frac{1}{2}\frac{\partial^2}{\partial\rho^2} + \frac{(K(K+d-2))}{2\rho^2} - \frac{1}{\rho}\right) R_{n,K}(\rho) = \varepsilon_{n,K} R_{n,K}(\rho), \tag{32}$$

where $R_{n,K}(\rho)$ is the hyperradial wavefunction. The energies are given by

$$\varepsilon_n = -\frac{1}{2[n + 1/2(d-3)]^2}, \tag{33}$$

with $n \geqslant K + 1$. The three lowest states are obtained by imaginary time propagation with $K = 1, 2$ and 3 for a hyperradial grid with 128 points as displayed in figure 1. For $\Delta\tau = 10^{-2}$, the ground state is obtained within 0.1% accuracy in energy after 10^3 time steps.

5.2. Helium ground state

As another example to describe the present methodology and to gauge the accuracy of the method, we consider the helium ground state. We note that correlated two-electron dynamics as reflected, e.g., in non-sequential double ionization of He by intense laser fields or heavy-ion

Table 1. Convergence to the exact ground-state energy of helium for different maxima in a number of α points and l_x, l_y. The exact ground-state energy is $E_{\text{exact}} = -2.904$ au.

$(n_{\max}, l_{x_{\max}}, l_{y_{\max}})$	Energy (au)
(4, 0, 0)	−1.0488
(4, 1, 1)	−2.4801
(4, 2, 2)	−2.7254
(6, 0, 0)	−1.2803
(6, 1, 1)	−2.5782
(6, 2, 2)	−2.8716
(8, 0, 0)	−1.4329
(8, 1, 1)	−2.8524
(8, 2, 2)	−2.8981

impact, are areas where the present method is expected to be very useful. Since reliable results on dynamics require an accurate atomic structure, it is therefore interesting to see how well the method works for the He ground state itself.

For the present investigation, we choose a small radial grid of $\rho_{\max} = 6$ au, with 32 radial points, a time step of 0.03 au and a propagation time of 10 au. The exact ground-state energy excluding effects of the moving nuclei, radiative and relativistic effects is known to an accuracy of about one part in 10^{19} [21] and is obtained by a variational procedure involving sophisticated trial functions and a basis set containing 2114 terms. For our purposes, the value -2.904 au will be taken as the exact result. We assume an infinitely heavy mass. This means that the coordinates of the electrons are given directly in terms of the Jacobi coordinates as $\mathbf{r}_1 = \mathbf{x}_1$ and $\mathbf{r}_2 = \mathbf{y}_2$, respectively.

In table 1, we show the He ground-state energies found for different values of $n_{\max}$, $l_{x_{\max}}$ and $l_{y_{\max}}$. From the table, we note that the convergence is sensitive to all angular variables. We found that the results for the energies were much less sensitive to the radial grid, and therefore we do not consider the convergence behaviour in that variable. The calculational time ranged from a few seconds to a couple of minutes on an IBM Regatta using four processors.

6. Conclusion and outlook

In this paper we have described a spectral split-step algorithm for numerical solution of the time-dependent Schrödinger equation on a discrete hyperspherical grid. The method is a generalization and an extension of an analogous algorithm for the three-dimensional problems. It turns out that when the appropriate transformation rule based on the angular coordinates can be constructed, the six-dimensional, or in general N-dimensional, algorithm are identical. This implies that an optimized and parallel code in three dimensions can be directly extended to any dimension by providing the dimension-specific angular transformation. Research in this direction is in progress.

In the near future, we plan to apply the present algorithm to an investigation of two-electron effects in helium and molecular hydrogen when these systems are subject to intense coherent pulses of short duration and high frequency.

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References

[1] Poincare H 1899 *Acta Math.* **13** 1
[2] McCauley J L 1997 *Classical Mechanics, Transformations, Flows, Integrable and Chaotic Dynamics* (Cambridge: Cambridge University Press)
[3] Macek J 1968 *J. Phys. B: At. Mol. Phys.* **1** 831
[4] Lin C D 1986 *Adv. At. Mol. Opt. Phys.* **22** 77
[5] Zhukov M V, Danilin B V, Fedorov D V, Bang J M, Thompson I J and Vaagen J S 1994 *Phys. Rep.* **4** 151
[6] Dmitrasinovic V 2003 *Phys. Rev.* D **67** 114007
[7] Madsen L B 2003 *J. Phys. B: At. Mol. Opt. Phys.* **36** R223
[8] Parker J S, Doherty B J S, Meharg K J and Taylor K T 2003 *J. Phys. B: At. Mol. Opt. Phys.* **36** L393
[9] Lambropoulos P, Maragakis P and Zhang J 1998 *Phys. Rep.* **305** 203
[10] Avery J 2000 *Hyperspherical Harmonics and Generalized Sturmians* (Dordrecht: Kluwer)
[11] Tong X M and Lin C D 2005 *Phys. Rev.* A **71** 033406
[12] Hermann M R and Fleck J A Jr 1988 *Phys. Rev.* A **38** 6000
[13] Hansen J P, Sørevik T and Madsen L B 2003 *Phys. Rev.* A **68** 031401
[14] Nielsen E, Fedorov D V, Jensen A S and Garrido E 2001 *Phys. Rep.* **347** 373
[15] Birkeland T, Førre M, Hansen J P and Selstø S 2004 *J. Phys. B: At. Mol. Opt. Phys.* **37** 4205
[16] Wommersley R S and Sloan I H 2001 *Adv. Comput. Math.* **14** 195
[17] Wommersley R S and Sloan I H www.maths.unsw.edu.au/rsw/Sphere
[18] Gautchi W 1994 *TOMS* **20** 21–62
[19] Lin C D and Liu X 1988 *Phys. Rev.* A **37** 2749
[20] Nouri S 1999 *Phys. Rev.* A **60** 1702
[21] Drake G W F (ed) 1996 *Atomic, Molecular and Optical Physics Handbook* (New York: American Institute of Physics)