Implementation of established algorithms to extend HELIUM

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Plan of Talk

- Introduction
- HELIUM
- Combining HELIUM methods with R-matrix technology
- Computational Details
- Progress to date - coding and testing
- Summary

Introduction

Two-electron ions in intense laser fields

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Two-electron ions in intense laser fields

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  - ab initio
  - full-dimensionality
  - accurate
  - good over a *wide* range of laser wavelength and intensity

Towards multi-electron atoms and molecules

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Towards multi-electron atoms and molecules

- There is a crucial need to handle the TDSE accurately for multi-electron atoms and molecules coupled to IR/visible/UV and VUV laser fields
- The R-matrix concept allows the carry over of HELIUM methods

HELIUM

Two-electron ion plus intense laser

$$\mathrm{i}\frac{\partial \Psi}{\partial t} = H\Psi(\mathbf{r}_1, \mathbf{r}_2, t) \qquad \text{TDSE}$$

$$\begin{aligned} H \;=\; & -\tfrac{1}{2}\boldsymbol{\nabla}_1^2 - \frac{Z}{r_1} - \frac{A(t)}{\mathrm{i}\,c}\frac{\partial}{\partial z_1} \\ & + \frac{1}{r_{12}} \\ & - \tfrac{1}{2}\boldsymbol{\nabla}_2^2 - \frac{Z}{r_2} - \frac{A(t)}{\mathrm{i}\,c}\frac{\partial}{\partial z_2} \end{aligned}$$

Atomic units $e = m = \hbar = 1$

The HELIUM code (1)

Reported in:

ES Smyth, JS Parker and KT Taylor 1998 *Comput. Phys. Comm.* **114** 1-14

- Finite-difference/ basis set method
- (6+1)D PDE – $\Psi(r_1, r_2, \theta_1, \theta_2, \phi_1, \phi_2, t)$
- Basis set for $\theta_1, \theta_2, \phi_1, \phi_2$

$$\Psi = \sum_{\ell_1\, \ell_2\, L\, M} \frac{1}{r_1 r_2} f_{\ell_1 \ell_2 L M}(r_1, r_2, t)\, |\ell_1\, \ell_2\, L\, M\rangle$$

The HELIUM code (2)

Reported in:

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- Through orthonormality of the angular basis $|\ell_1\, \ell_2\, L\, M\rangle$, a set of coupled differential equations for the functions $f_{\ell_1 \ell_2 L M}(r_1, r_2, t)$ is obtained

- Propagate over grid from $\Psi(\mathbf{r}_1, \mathbf{r}_2, t)$ to $\Psi(\mathbf{r}_1, \mathbf{r}_2, t + \Delta t)$ via an Arnoldi propagator

Combining HELIUM methods with R-matrix technology

R-matrix approach

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- Allows an accurate, genuinely multi-electron, representation of the multi-electron regions near nuclei through basis set methods (Inner Region)

- Allows only the one electron present at larger distances in single ionization processes to be represented there using HELIUM methods (Outer Region)

Combining HELIUM and R-matrix Methods

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- Split into a multi-electron inner region and a one-electron outer region

- Inner region handled with an R-matrix basis
  - Long-established computer codes available for time-independent calculations using basis set methods
  - These codes are being extended to handle the TDSE

Combining HELIUM and R-matrix Methods

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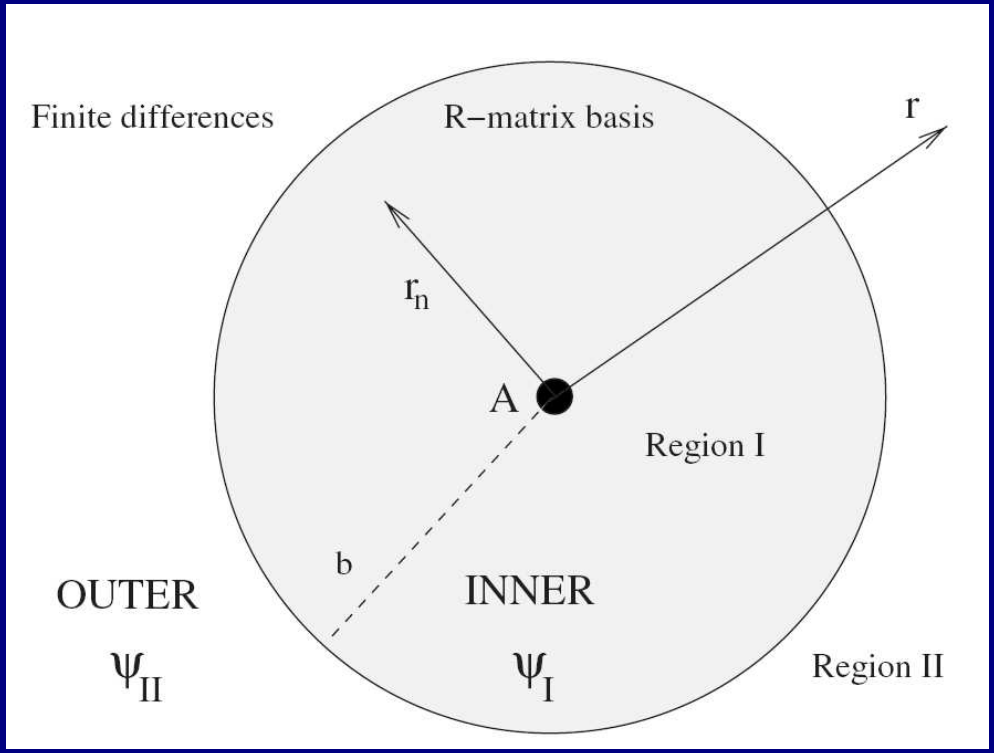
- Split into a multi-electron inner region and a one-electron outer region
- Outer region handled with a finite difference grid
  - Explicit high-order time-propagator techniques are crucial for accuracy, stability and efficiency
  - Implementation of HELIUM methods

Partition of Configuration Space



Computational Details

Inner Region Multi-electron Wavefunction

$$\psi_I(\mathbf{r_1 r_2 ... r_{N+1}}, t) = \sum_{k=1}^{K} C_k(t)\psi_k(\mathbf{r_1 r_2 ... r_{N+1}})$$

for $0 \leq r_1, r_2, ..., r_{N+1} = r \leq b$

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- The $\psi_k, k = 1, K$ form a field-free, time-independent R-matrix basis for the $(N + 1)$ electrons within the Inner Region with outer boundary at $r = b$ in all electron radial co-ordinates.

Inner Region Hamiltonian

- Must be Hermitian over the Inner Region
- The Hermitian Inner Region Hamiltonian is $H_I = H_0 + \hat{L}_h$
- The Hermitian dipole operator is $D_I = D + g\hat{L}_d$ (where $g = 1$ in velocity gauge and $g = 0$ in length gauge)
- $\hat{L}_h$ and $\hat{L}_d$ are Bloch surface terms, and $\hat{L}_h$ has a spatial derivative to be evaluated at the boundary $r = b$

Inner Region TDSE (1)

$$\mathrm{i}\frac{d}{dt}\psi_I(\mathbf{r_1 r_2} \quad ... \quad \mathbf{r_{N+1}}, t) =$$

$$\left[H_I + D_I(t)\right]\psi_I(\mathbf{r_1 r_2 ... r_{N+1}}, t)$$

$$- \quad \left[\hat{L}_h + \hat{L}_d(t)\right]\psi(\mathbf{r_{N+1}}, t)$$

for $0 \leq r_1, r_2, ..., r_{N+1} = r \leq b$

Inner Region TDSE (2)

After algebra, and writing in matrix notation:

$$\mathrm{i}\frac{d}{dt}C_k(t) = \quad [\mathbf{H}.\mathbf{C}]_k(t) - \sum_p \frac{1}{2}\omega_{kp}\bar{f}_p(b,t)$$

where

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where

- ω_{kp} are known surface amplitudes
- $\bar{f}_p(b,t)$ contain spatial derivatives of the outer wavefunction on the boundary

Outer Region One-electron Wavefunction

$$\psi_{II}(\tilde{\mathbf{r}}_N, \mathbf{r}(i), t) = \sum_p \Phi_p(\tilde{\mathbf{r}}_N, \hat{r}) \frac{f_p(i,t)}{r(i)}$$

for $b \leq r(i)$

Outer Region One-electron Wavefunction

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for $b \leq r(i)$

- where $i = i_b, .., I$ and $r(i_b) = b$
- The radial motion of the ejected electron is described by the radial channel functions $f_p(i,t)$

Outer Region TDSE

$$\begin{aligned}\frac{d}{dt}f_p(i,t) = & - \mathrm{i}[\mathbf{H}\cdot\mathbf{F}]_p(i,t) \\ & + \delta_{ii_b}\left[B_{0p}(i_b-1,t)+B_{0p}(i_b-2,t)\right] \\ & + \delta_{ii_b+1}B_{1p}(i_b-1,t)\end{aligned}$$

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- The terms multiplying δ_{ii_b} and δ_{ii_b+1} require the B terms to be evaluated at grid points i_b-1 and i_b-2 on the *inner* side of the boundary at $r=b$.
- This reflects the use of 5-point difference operators on the FD grid.

Time Propagation

Time Propagation

- Taylor Propagator

$$\begin{aligned}\psi(t+\delta t) &= a_0\psi(t) + a_1 H\psi(t) \\ &+ a_2 H^2\psi(t) + \cdots + a_n H^n\psi(t)\end{aligned}$$

where $a_k = (-\mathrm{i}\delta t)^k/k!$, so that

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$$\psi(t+\delta t) = \exp(-\mathrm{i}\delta t H)\psi(t)$$

  - Explicit single-step propagator
  - Can easily monitor local truncation error
  - May not be efficient, especially in inner region

Time Propagation

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  - Q has columns $Q_0, Q_1, \cdots, Q_n$ where Q_k is obtained by calculating HQ_{k-1} and orthonormalizing with respect to $Q_0, Q_1, \cdots, Q_{k-1}$
  - h is the Krylov subspace Hamiltonian
  - More efficient than the Taylor propagator

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- Using Outer Region information (spatial derivative terms at $r = b$), solve the Inner Region TDSE for $C_k(t + \delta t)$.

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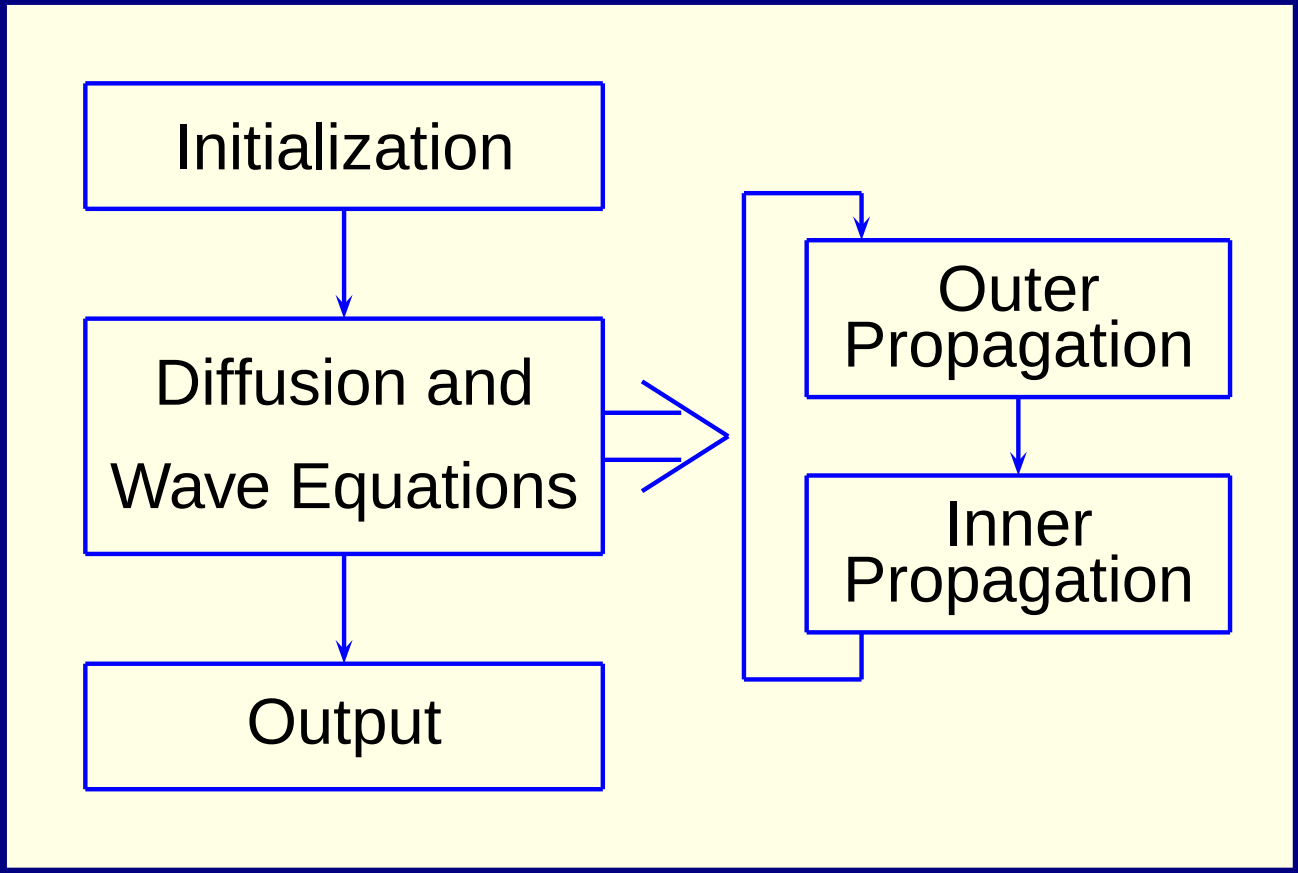
- Using Outer Region information (spatial derivative terms at $r = b$), solve the Inner Region TDSE for $C_k(t + \delta t)$.

Repeat the process for the next time step.

Progress to date

Code Schematic



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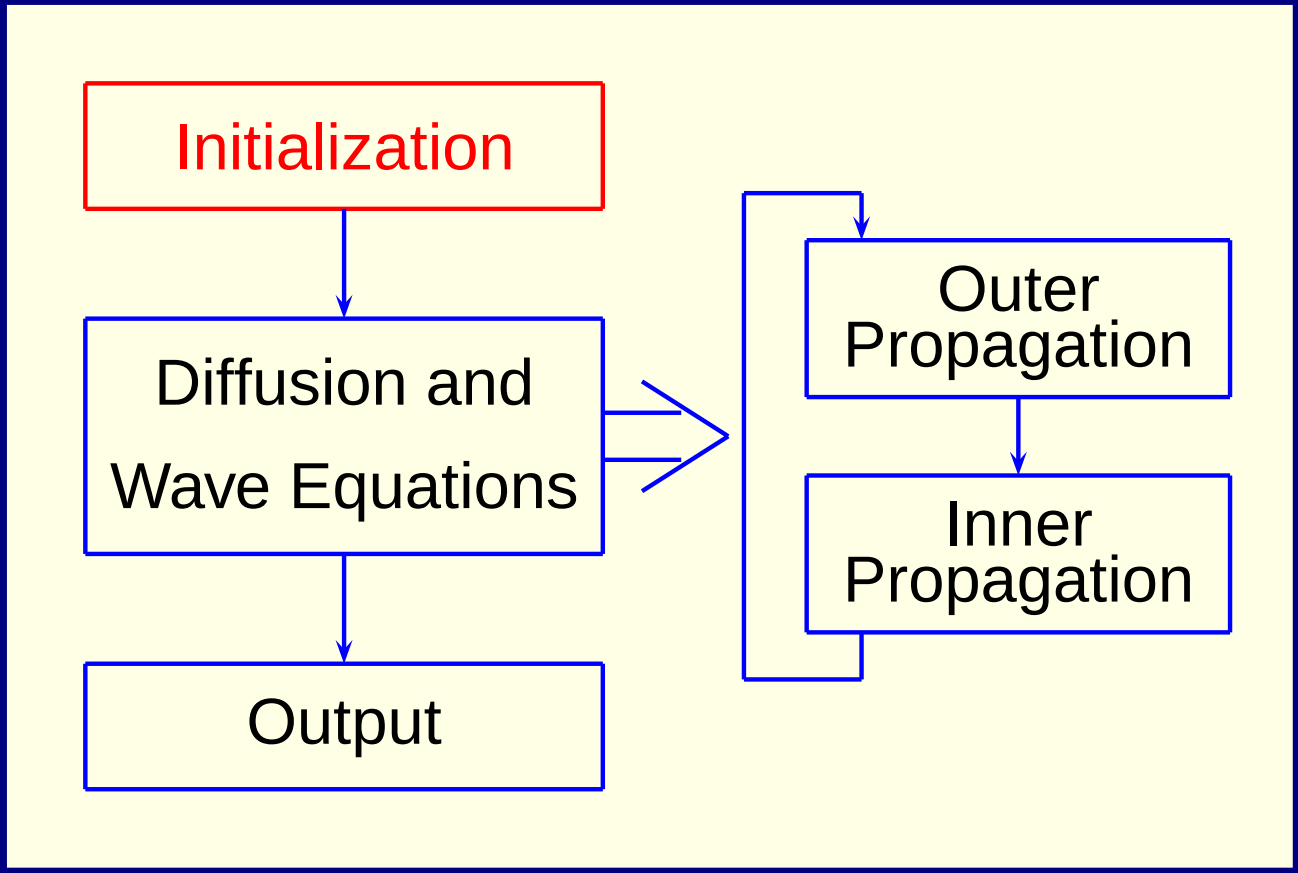

Initialization Using R-matrix codes

- Read H and D files
  - 1st sweep reads problem parameters
  - 2nd sweep reads matrix elements

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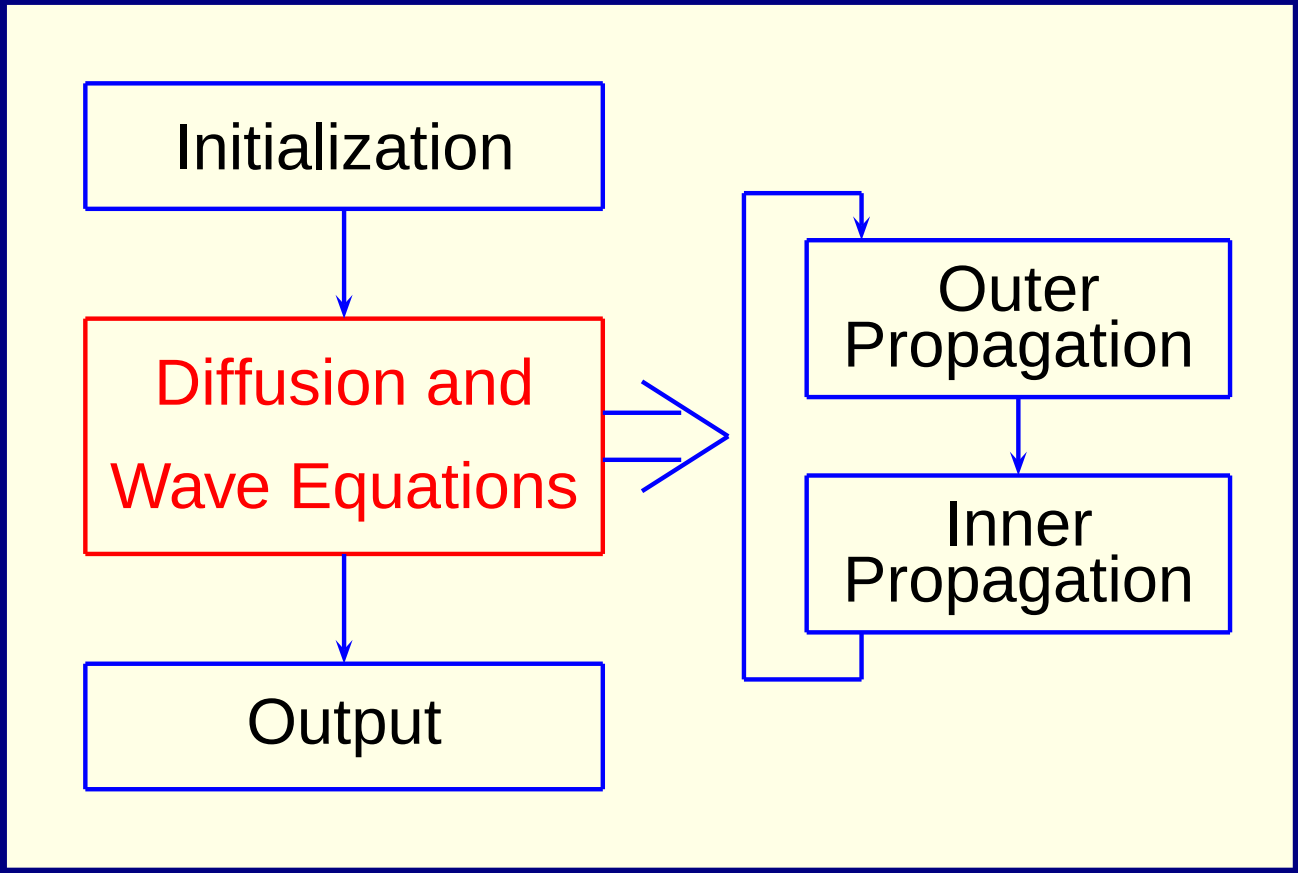
- Read H and D files
  - 1st sweep reads problem parameters
  - 2nd sweep reads matrix elements
- Set up inner Hamiltonian matrix
- Set up long range potentials for outer Hamiltonian
- Set up basis state arrays

Further Initialization Routines

- Set up surface amplitudes (ω_{kp})
- Initialize finite difference routines
- Initialize propagator coefficients
- Read a status file to determine starting point of run
- Initialize (or read in) the wavefunction itself

Code Schematic

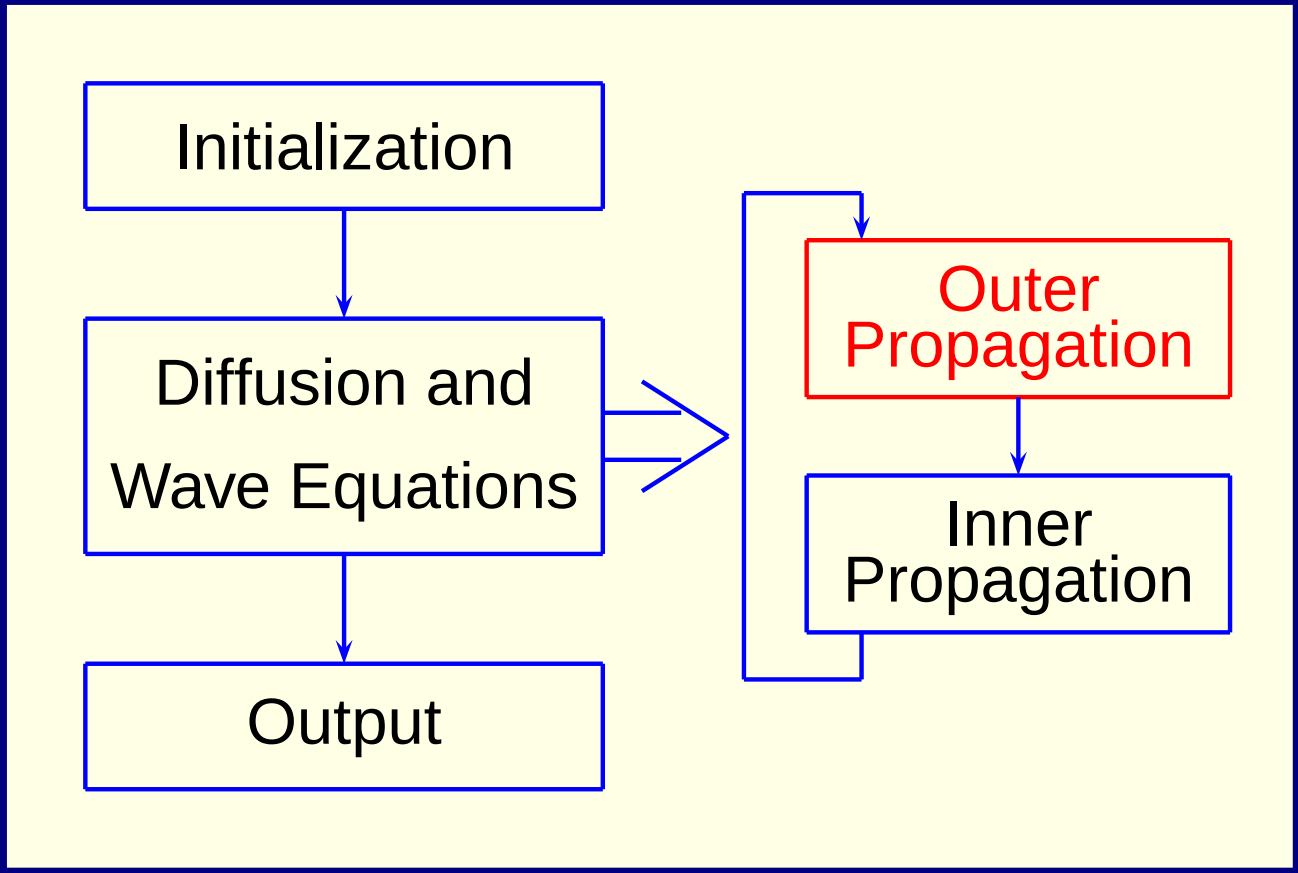


Diffusion and Wave Equations

For each timestep:

- Calculate f_p, $\dot{f}_p$, $\ddot{f}_p$, ... at $i_b - 1$ and $i_b - 2$
- Propagate outer wavefunction
- Calculate $\frac{df_p}{dr}$, $\frac{d\dot{f}_p}{dr}$, $\frac{d\ddot{f}_p}{dr}$, ... at $r = b$
- Propagate inner wavefunction
- Calculate populations and other observables

Code Schematic



Propagation of Outer Wavefunction

- Taylor and Arnoldi propagators coded
- Spatial derivatives $\frac{df_p}{dr}$, $\frac{d\dot{f}_p}{dr}$, $\frac{d\ddot{f}_p}{dr}$, ... at $r = b$ coded
- Full surface term $\sum_p \omega_{kp} \bar{f}_p(b,t)$ coded

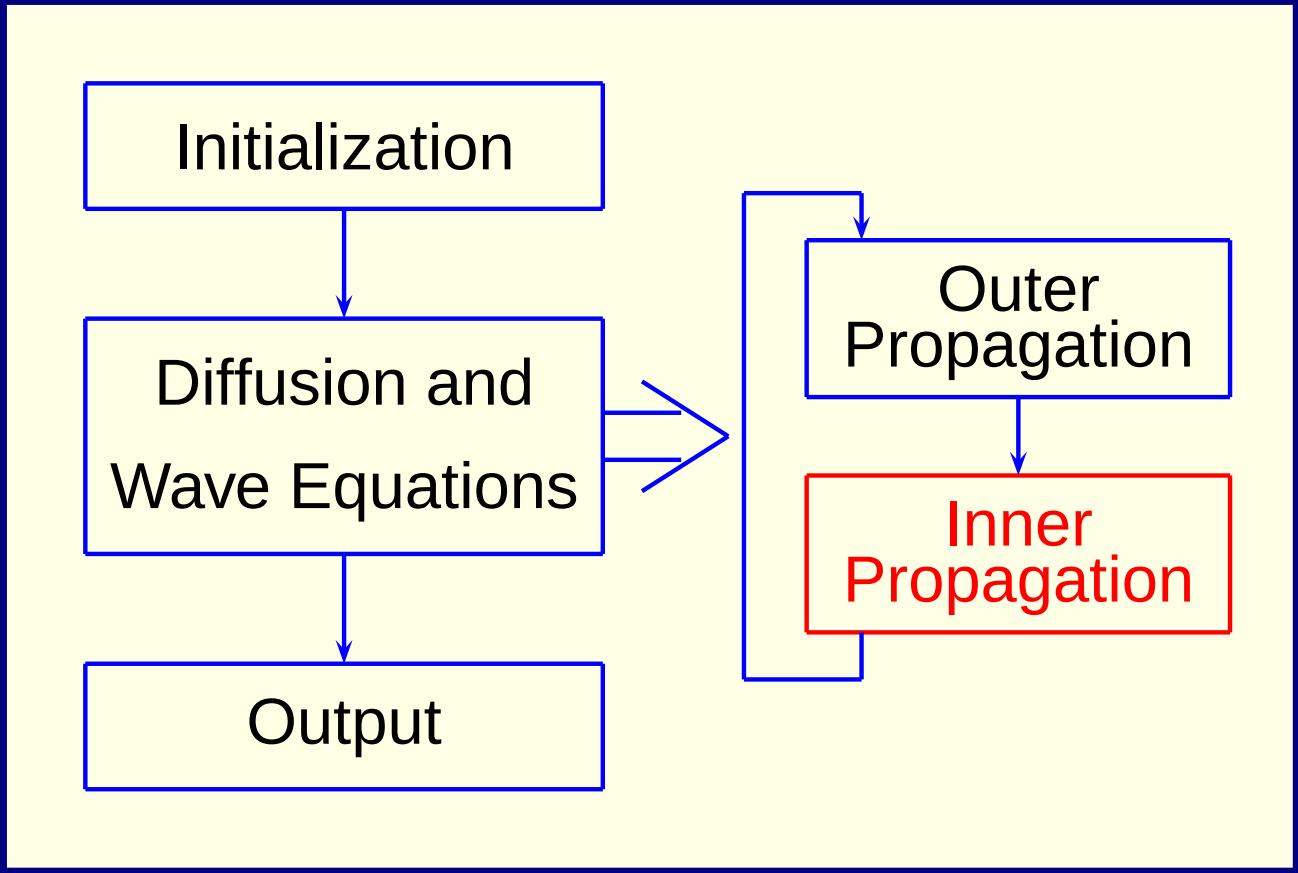
  where $\bar{f}_p(b,t) = \frac{df_p}{dr} + g\left(\frac{A(t)}{c}\right) \sum_{p'} K_{pp'} f_{p'}|_{r=b}$

Outer Wavefunction: $H f_p$ Calculation

- For each channel p:

$$H f_p = - \frac{1}{2}\nabla^2 f_p - \frac{Z-N}{r} f_p + \sum_{p'} (W_E + W_P + W_D) f_{p'}$$

Code Schematic



Propagation of Inner Wavefunction

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Dr Michael Lysaght has started development of Inner Region Code:

- Arnoldi propagator coded (without surface terms)
- Arnoldi propagator tested (without surface terms)
- Wavefunction values f_p, $\dot{f}_p$, $\ddot{f}_p$, ... at $i_b - 1$ and $i_b - 2$ for passing to outer region yet to be coded

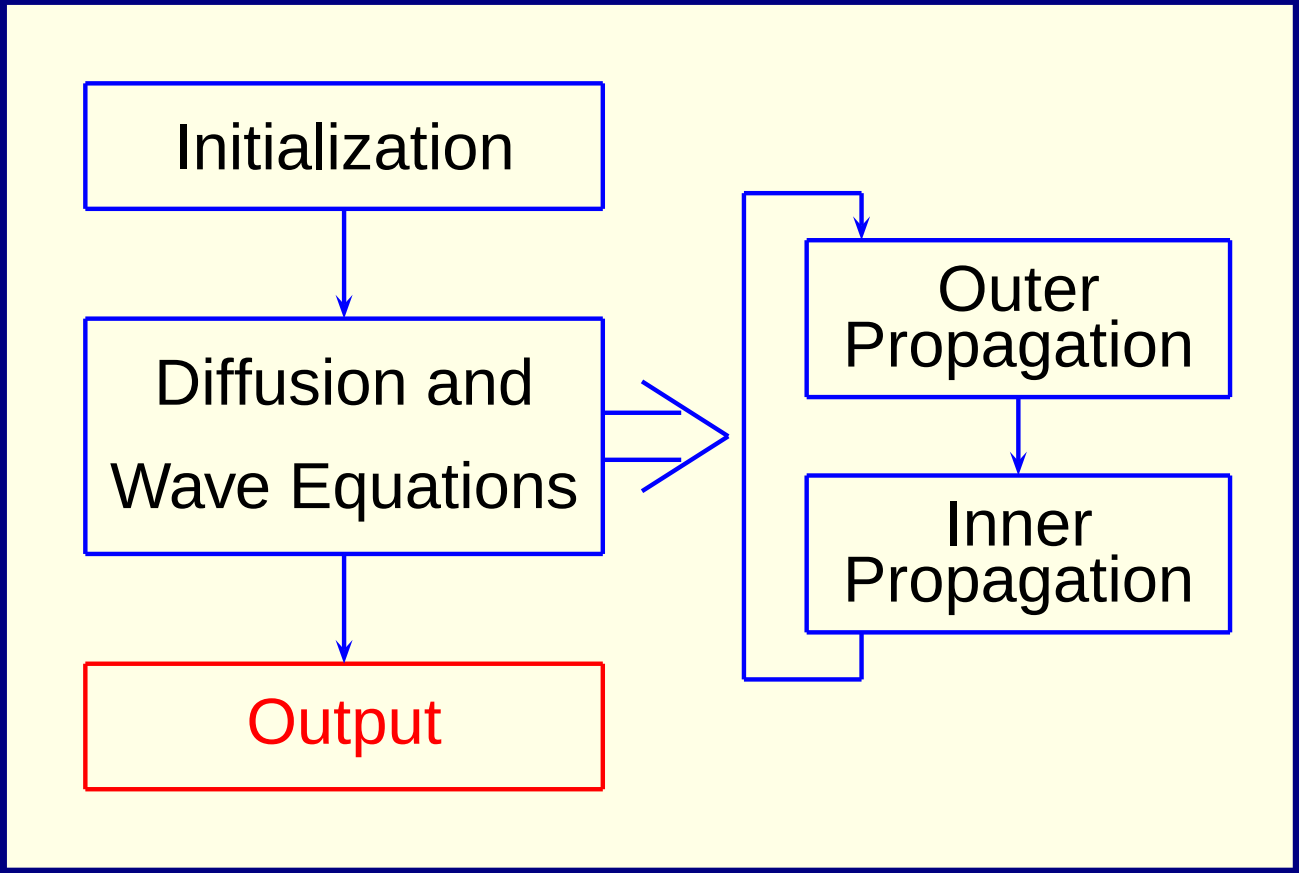
Inner Wavefunction: $\mathbf{HC}$ Calculation

- Ordering the eigenfunctions by symmetry means $\mathbf{H}$ is a block diagonal matrix:

H_S	D_{SP}			
D_{PS}	H_P	D_{PD}		
	D_{DP}	H_D	D_{DF}	
		D_{FD}	H_F	D_{FG}
			D_{GF}	H_G

- $\mathbf{HC}$ is calculated by a matrix multiplication

Code Schematic



Output Routines

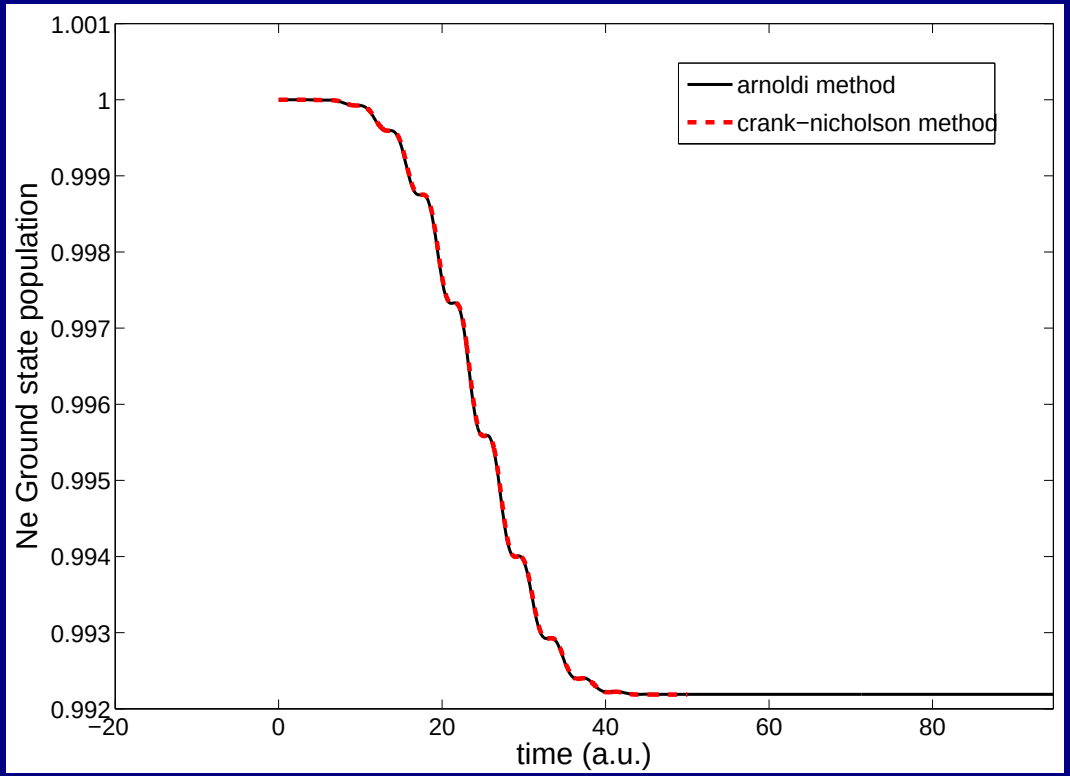
- Write the inner and outer wavefunctions to disk
- Write population arrays to disk
- Update the status file

Testing of Coding

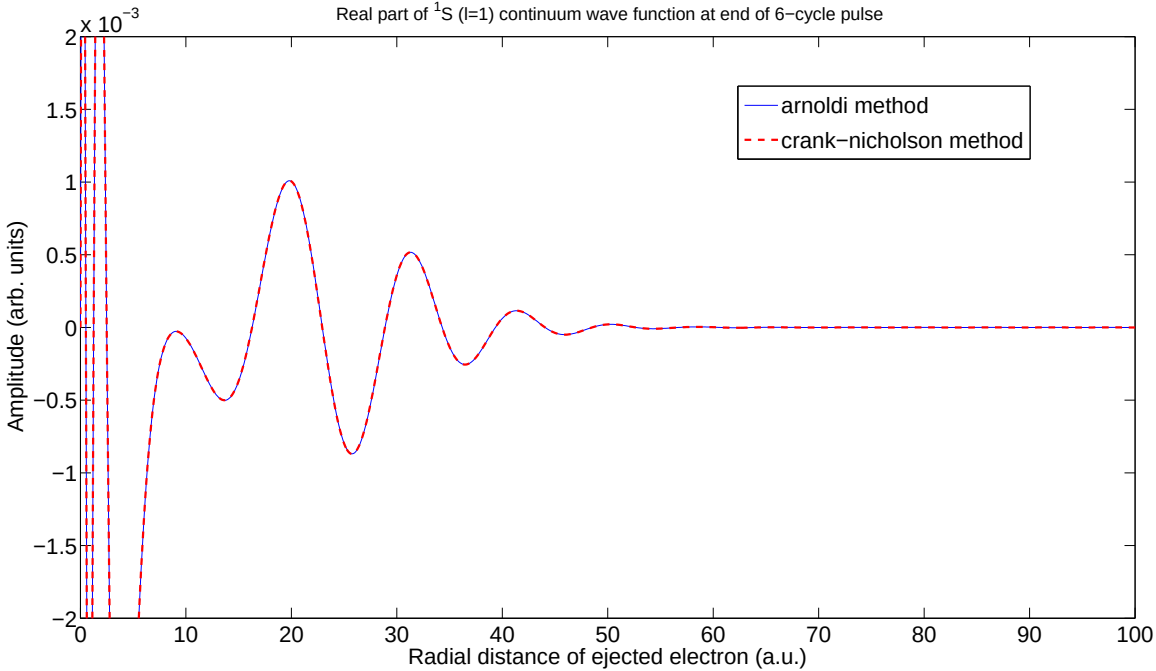
Inner Region Arnoldi Propagation

- Tested by extending the size of the inner region so that an outer region is not brought into play
- Results compared with those from an established R-matrix code that invokes an implicit time propagator
- The comparison is very satisfactory and certainly demonstrates the correctness of the new coding

Population in the ground state of Neon as a function of time



Real part of ^{1}S(ℓ=1) Neon continuum wavefunction after excitation with a 6 cycle laser pulse as a function of radial distance



Recap of Progress

Outer Region Coding

- Initialization routines
- Hamiltonian terms
- Diffusion and wave equations - Taylor and Arnoldi propagators
- Surface terms - to pass to inner region
- Population calculations
- Some I/O

Inner Region Coding

- Initialization routines
- Hamiltonian matrix
- Implementation and testing of the Arnoldi propagator without the Bloch surface terms
- Population calculations
- I/O Routines

Next Steps

- Incorporating surface terms in the Inner Region Arnoldi propagation
- Coding the surface terms at $i_b - 1$ and $i_b - 2$ required for propagating the Outer Region wavefunction
- Testing and re-testing!
- Parallelization ...

Summary - Goals

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- Inner Region code based on R-matrix basis set methods
- Outer Region code based on HELIUM finite-difference methods

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- Inner Region
  - Arnoldi propagator invoked and results agree very well with those obtained from an established R-matrix code
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- Inner Region
  - Arnoldi propagator invoked and results agree very well with those obtained from an established R-matrix code
  - Work continuing on surface terms
- Outer Region
  - Taylor propagator and Arnoldi propagator coded
  - Really need to start looking at tests

Other Tasks on Augmenting HELIUM: Dr Jonathan Parker starts 1st October

- Extensions to HELIUM to allow calculations with crossed laser fields

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- Extensions to HELIUM to allow calculations with crossed laser fields
- Implementation of a new final-state momentum-space code for HELIUM
- Implementation of new post-processing code to transform HELIUM output from spherical to cylindrical geometry
- Implementation of a hybrid MPI-OpenMP parallelism in HELIUM (Dr Ed Smyth, NAG)

Many thanks for your attention!