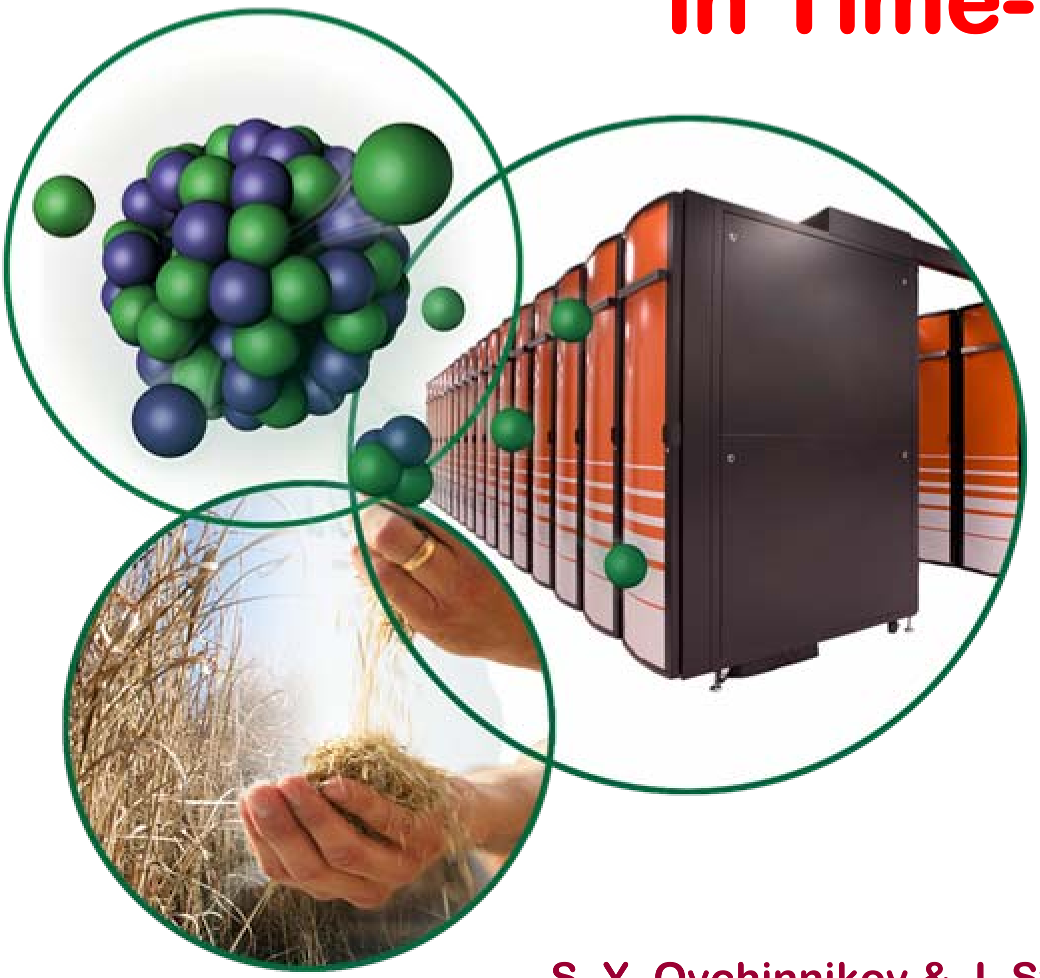
Quantum Treatment of Continuum Electrons in Time-Dependent Fields



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Motivations

1. **Evolution of electronic wavefunction from microscopic (atomic) to macroscopic (laboratory) scale.**

   - **Ion-atom collisions: Initial state is microscopic. Final state has phase difference $\delta \sim 16/(vR^{-1/2})$. For macroscopic $R \sim 10^6$ a.u. and $v = 0.5$ a.u., the $\delta \sim 0.03$. Neglecting δ can result in error of 6%. If R (~100 a.u) is microscopic, error is significantly larger.**
   - **Laser-atom interaction. One femtosecond pulse travels ~ 30 microns.**

2. **Long-interacting systems and long-time propagation, numerical difficulties at the boundary.**

   – **Phase problem:**

   ➢ **wavefunction has highly oscillatory divergent phase factor $\exp[ir^2/t]$ where $r^2/t \rightarrow v^2 t$ as $t \rightarrow \infty$.**

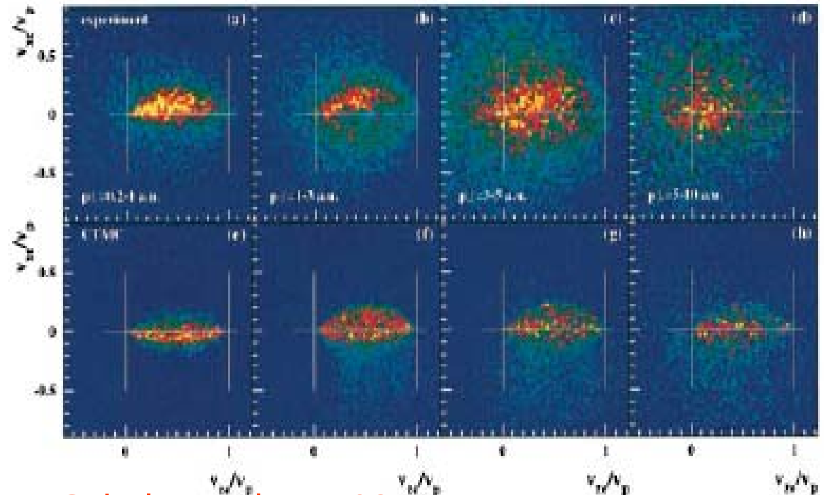
   – **Density-loss problem:**

   ➢ **The wavefunction is needed in regions where it is of the order ~ 10^{-10} relative to its value in more accessible regions.**

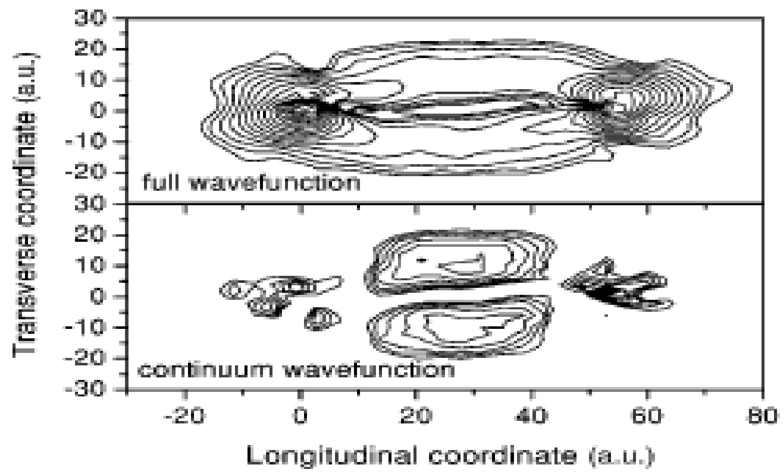
3. Ion-Atom Collisions: COLTRIMS experiment enables clear momentum analysis of both ejected electrons and ions thus unraveling the detailed information of collisions dynamics.
4. COLTRIMS reveals the rich structures of continuum electrons. These structures vary with impact parameters and collision velocities.

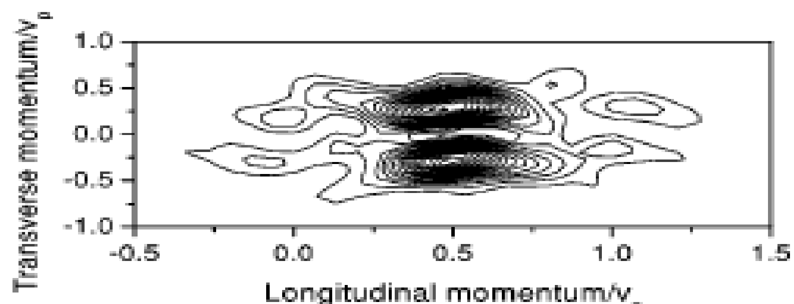
5. Concurrently, CTMC method, uncoupled two-center Sturmian, advanced adiabatic and TCMSD methods, all give contradictory explanations of the COLTRIMS' measurements for single ionization in H^+ + He collisions.
6. The ab-initio and direct numerical solution of TDSE on a lattice (LTDSE) attempted to resolve the controversies but ended time propagation at ~ 52 a.u.

Dorner et al PRL96



Schultz et al PRA02





Theoretical Approach

- Solve time-dependent Schrodinger Eqn (TDSE):

$$i\frac{\partial\Psi(\mathbf{r},t)}{\partial t} = H\Psi(\mathbf{r},t) = (T+V)\Psi(\mathbf{r},t)$$

- Removing the $\exp(ir^2/2t)$ from the continuum components of the wave function. We define our new coordinates and time as

$$\begin{aligned} \mathbf{r} &= \Omega\mathbf{q}\csc\theta/v \\ t &= -\Omega\cot\theta/v^2 \end{aligned}$$

and cast our total wave function in the form

$$\Psi(\mathbf{r},t) = \chi(\mathbf{q},\theta)\Phi(\mathbf{q},\theta)$$

where θ varies from 0 to π, and Ω and v are arbitrary parameters.

The function

$$\chi(\mathbf{q},\theta) = \left(\frac{i\sin\theta}{\Omega}\right)^{3/2} \exp[-i\Omega q^2 \cot\theta/2]$$

is highly oscillatory as $q \longrightarrow \infty$.

The function $\Phi(\mathbf{q},\theta)$, however is slowly varying function and satisfies the regularized TDSE:

$$\left[-\frac{1}{2}\nabla_{\mathbf{q}}^2 + \frac{1}{2}\Omega^2 q^2 + V(\mathbf{q},\theta)\right]\Phi(\mathbf{q},\theta) = i\Omega\frac{\partial\Phi(\mathbf{q},\theta)}{\partial\theta}$$

To solve above RTDSE, the initial regularized wave function is given by

$$\Phi_0(\mathbf{q},\theta_0) = \chi(\mathbf{q},\theta_0)^{-1}\Psi_0(\mathbf{q}\,\Omega\csc\theta_0/v, \Omega\cot\theta_0/v^2)$$

Where Ψ_0 is the wavefunction at initial time $t_0 = \Omega\cot\theta_0$

In the limit as θ tends to π , the continuum components of the regularized wavefunction $\Phi(\mathbf{q}, \theta)$ is independent of Ω and directly gives the ejected electron momentum distribution

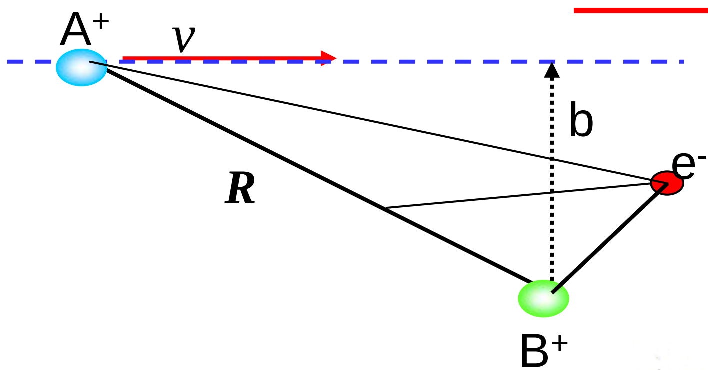
$$\lim_{\theta \to \pi} \Phi(\mathbf{q}, \theta) = A(\mathbf{k})$$

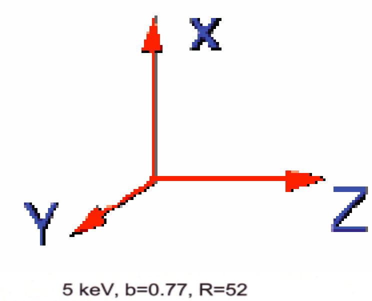
where $\mathbf{q} = \mathbf{k}/v$ and **k** is the momentum of the continuum electron.

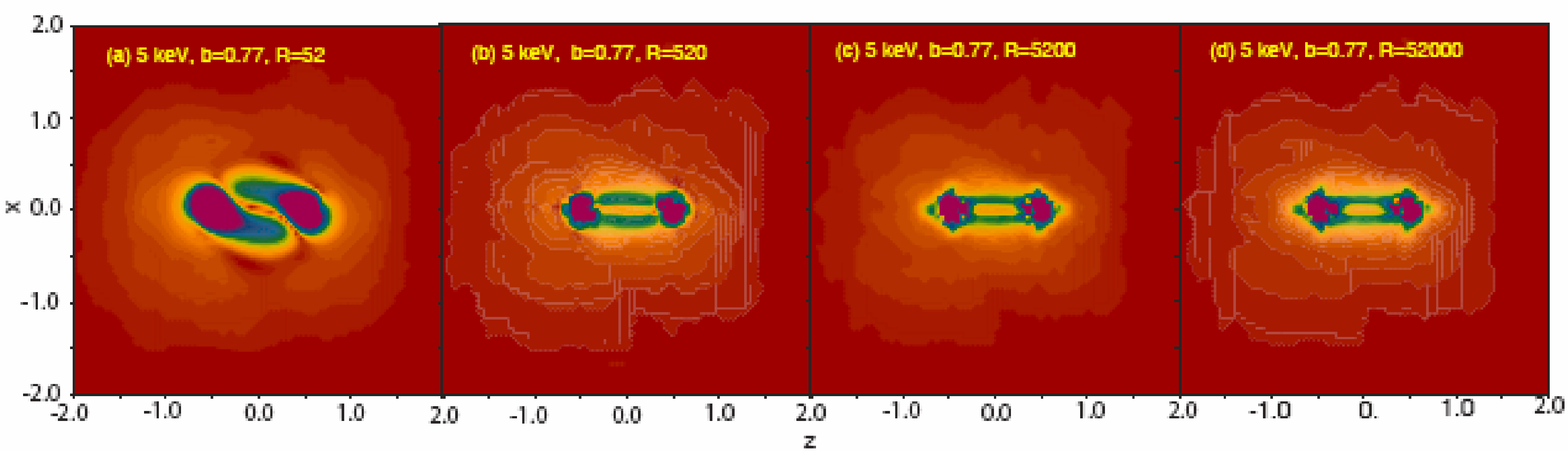
•The present approach does NOT require projection onto dynamic two-center continuum states at large times to extract the continuum components of the wavefunction since the bound states shrink to a minutely small region in the q-space.

•The lattice, TDSE is used to solve the RTDSE. The LTDSE solver is based on Split-Operator (time propagation) and Fast Fourier Transform (momentum space discretization) methods.

Results

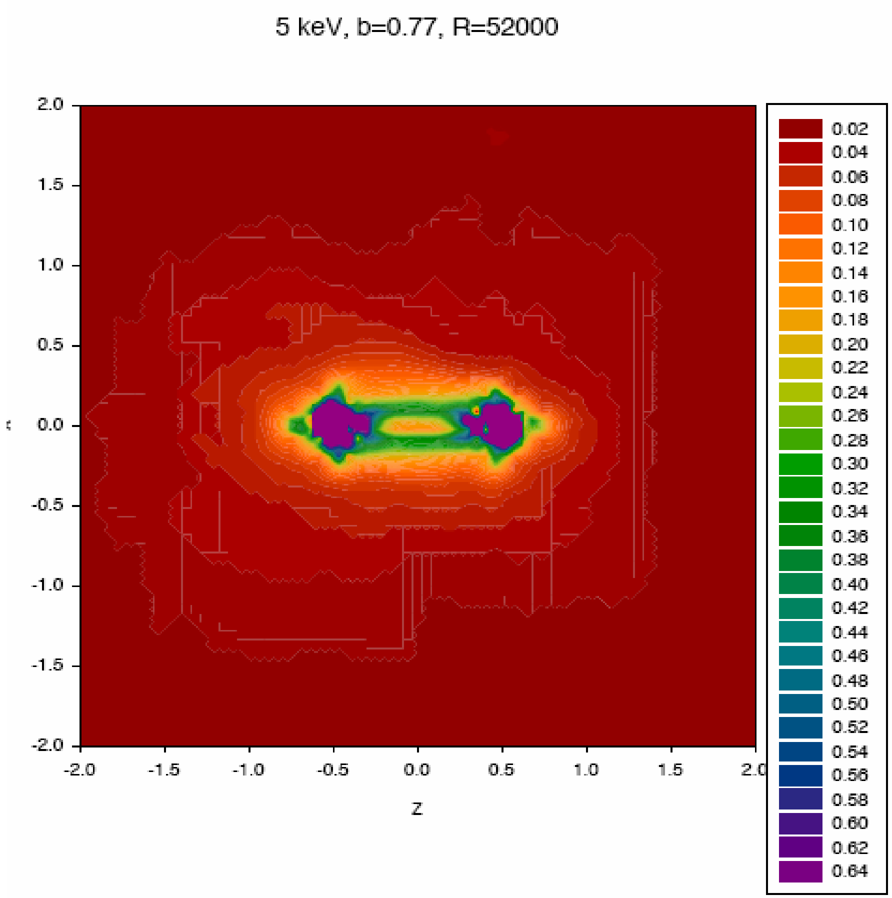


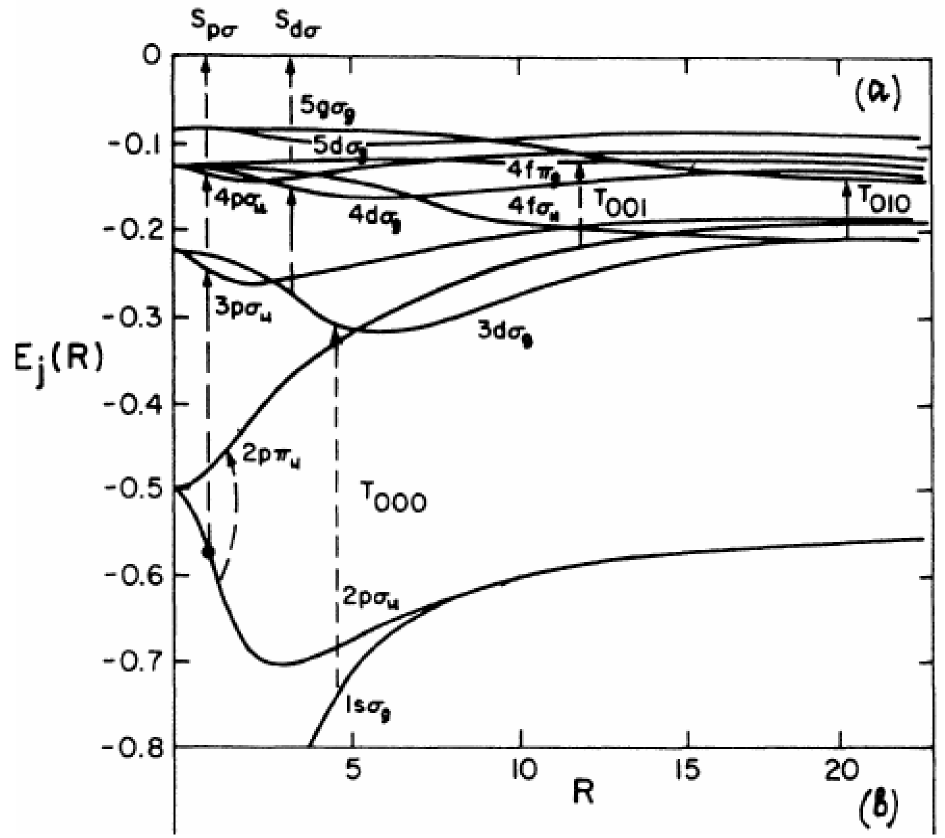




$H^+ + H \rightarrow H^+ + H^+ + e^-$ at $b = 0.77$ and $E = 5$ keV.

•Evolution of the electron momentum distributions in the scattering plane from internuclear distances R = 52 to 52,000 a.u. The distributions include bound target and projectile eigenstates localized near $v = \pm\hat{v}/2$.



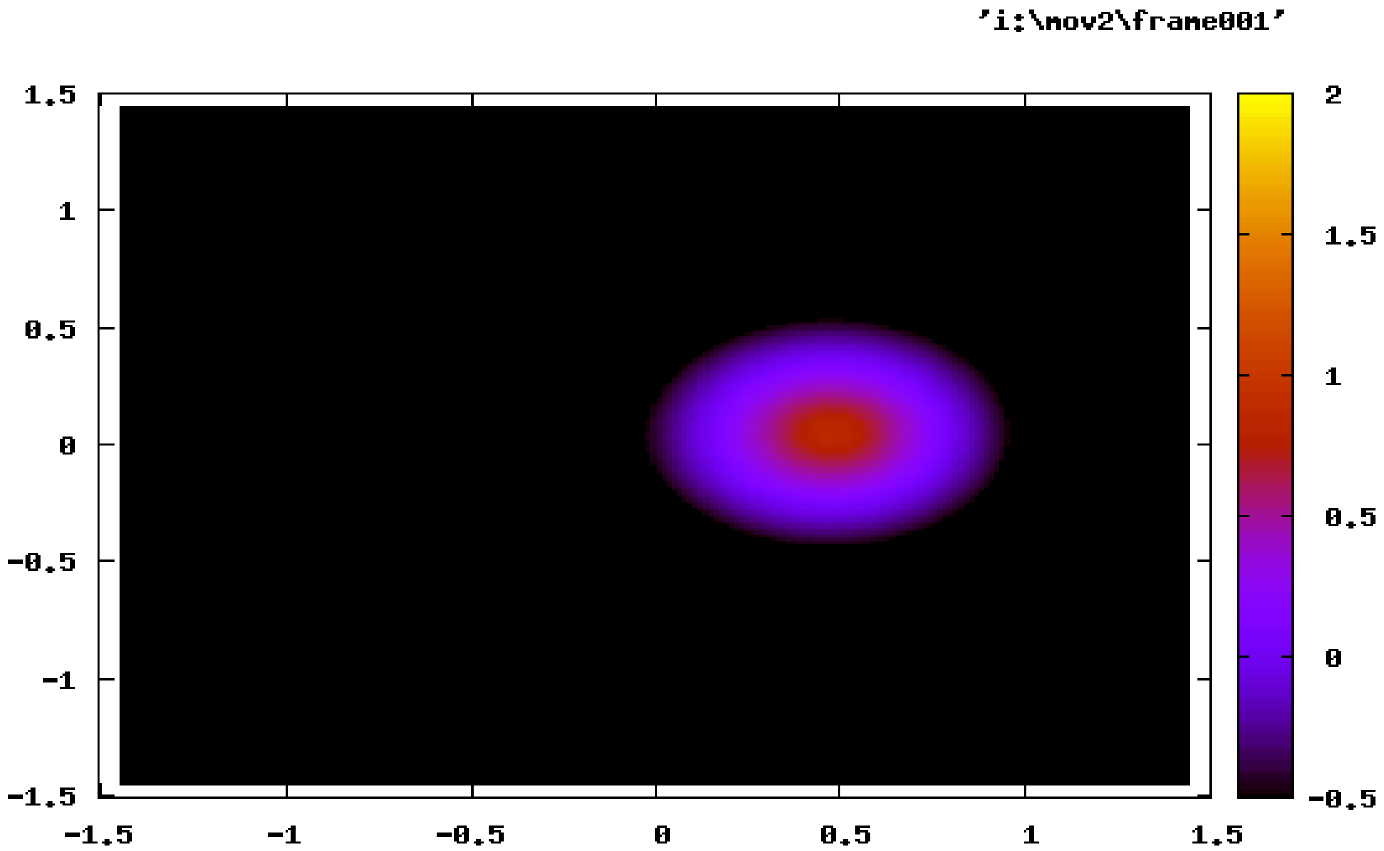


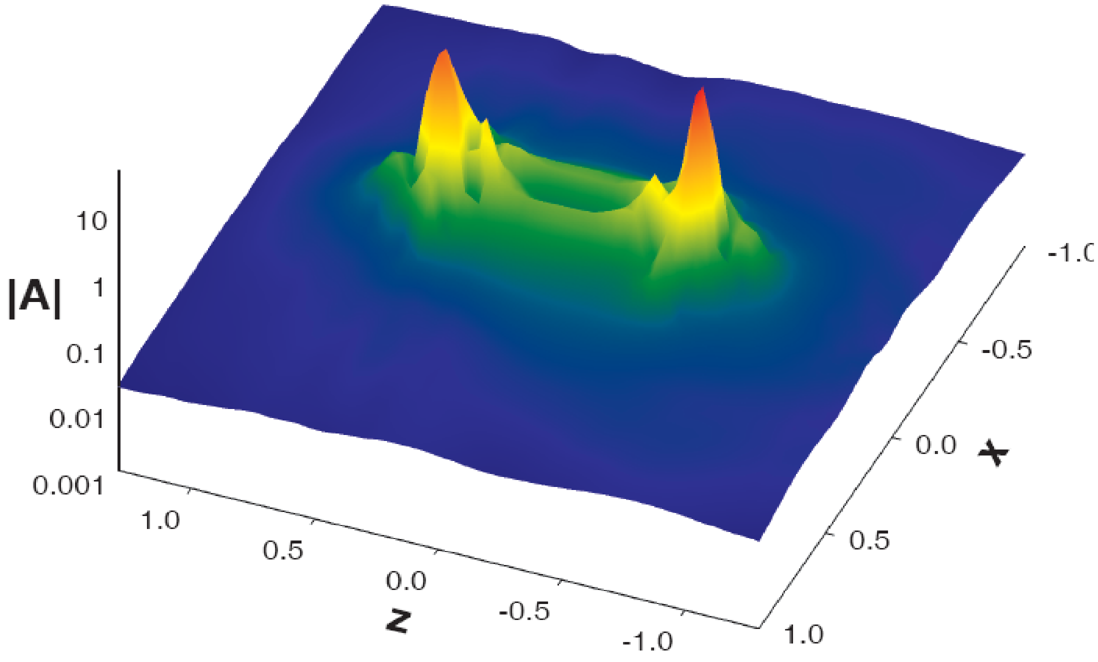
•Fast electrons are produced by S-promotion directly into the continuum near united-atom limits.

•Slow electrons are produced by **2pσ ->2pπ** united atom rotational coupling and the **1sσ ->3dσ** transitions followed by transitions to the continuum.

•Some of the electrons move to the direct and continuum capture cusps at large R.

Animation of ejected electron momentum distributions

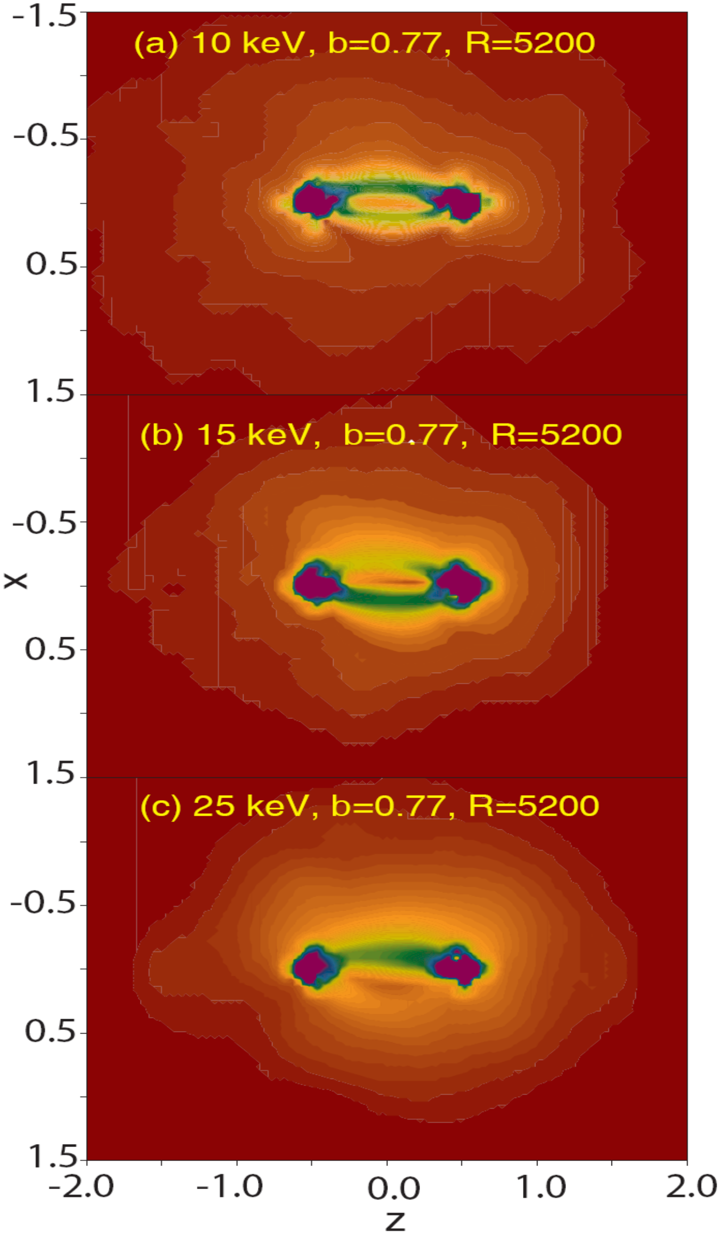




• **Plot of the ionization amplitude |A(k)| at R = 52,000 a.u., showing target and projectile cusps.**

•**The electrons in the cusps are a small fraction of the total that are ejected.**

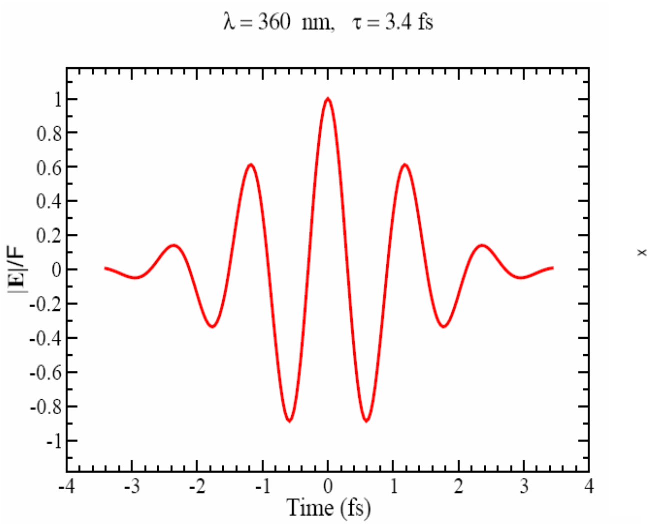
• **This component is small and generally negligible for most purposes, however, even this small component can be identified with the present theoretical approach.**

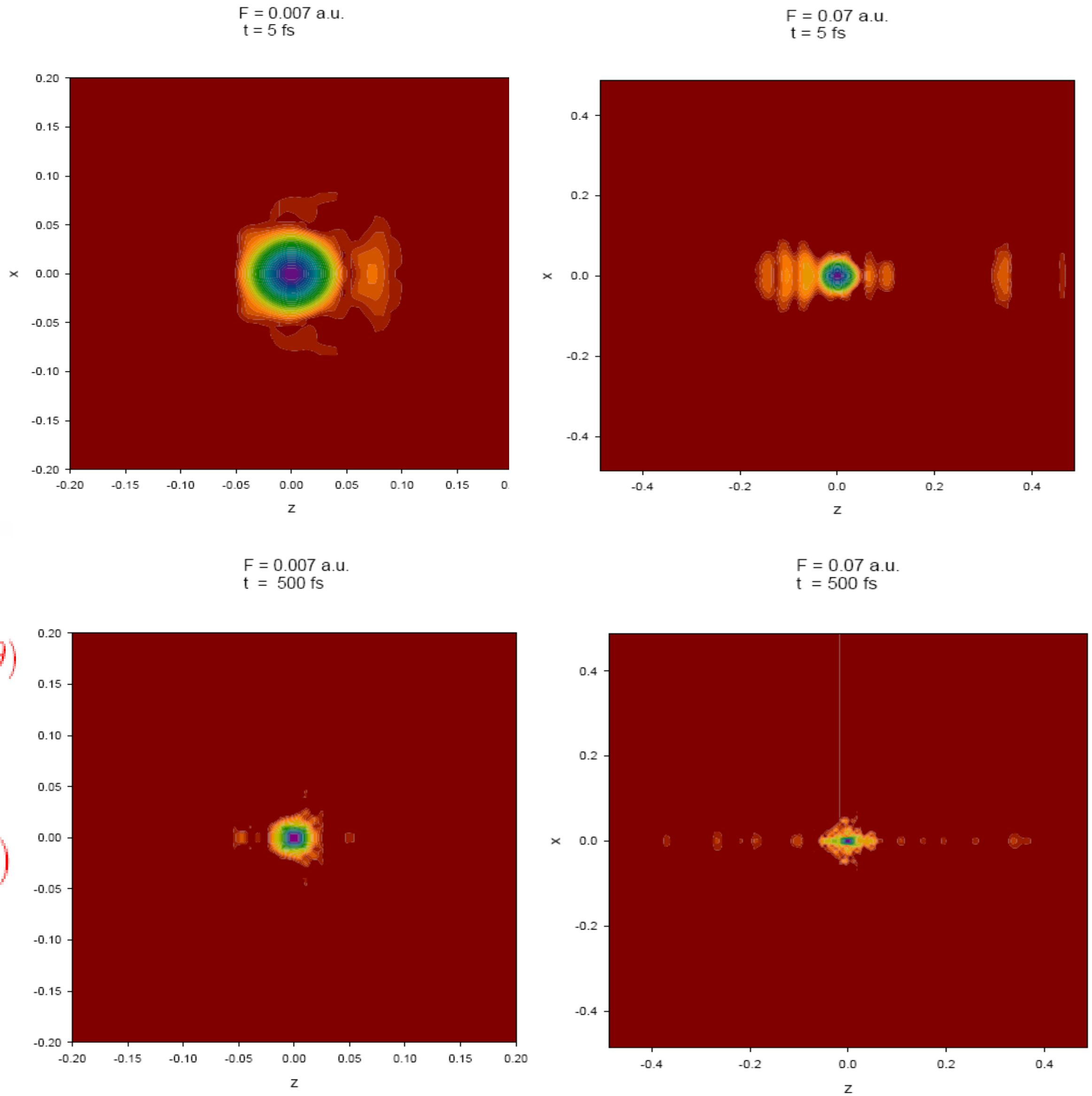


Plots of ejected electrons momentum distribution for p + H collisions at impact parameter b = 0.77 a.u.

- **Fast electron features shrink in extent but do not change shape.**
- **Alternatively, the slow electron distribution retains the same spatial extent but oscillates.**
- **In CONTRAST with that earlier approximate theory, we see some of the electrons actually move from the center to the target and projectile nuclei.**

laser + atom interactions





$$V(\mathrm{q},\theta) = -\frac{\Omega}{v\sin\theta}\frac{Z}{|\mathrm{q}|} + \left(\frac{\Omega}{v\sin\theta}\right)^3 \mathrm{q}\cdot\mathrm{E}(\mathrm{q},\theta)$$

with

$$\mathrm{E}(\mathrm{q},\theta) = F\,\hat{\mathrm{z}}\cos(\omega\, t(\theta))\,\exp(-t^2(\theta)/\tau^2)$$

Summary and Future Outlook

- Analytically removal of the highly oscillatory phase factor in the wave function.
- With RTDSE in the lattice representation, we can compute "converged" and accurate ejected electron momentum distribution for p + H(1s) with very low noise and visualize the dynamical structures of different components of a continuum electron in fine detail within a single theoretical method.
- Apply the method to one-electron atom involving collisions with multiply charged ions.
- Compute the population of high Rydberg states.
- Apply the method to laser+atom interactions.
- Possibly extending the method to treat two-electron processes in ion-atom and laser-atom interactions ???