Accurate computation of above threshold ionization spectra for stretched H_2^+ in strong laser fields

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Problem

Solving the time-dependent Schrödinger equation (TDSE) of H_2^+ in strong linearly polarized laser fields in the prolate spheroidal coordinates (ξ, η, ϕ) with the Born-Oppenheimer (BO) approximation at large internuclear distance R to get accurate photoelectron momentum distributions (PMD).

$$\mathrm{i}S\partial_t\Psi(t) = H(t)\Psi(t) = [H_0 + H_{\mathrm{int}}(t)]\Psi(t), \tag{1}$$

where the overlap term $S = (R/2)^3(\xi^2 - \eta^2)$, and the time-independent hamiltonian

$$\begin{aligned} H_0 = &-\frac{R}{4}\left[\frac{\partial}{\partial\xi}(\xi^2-1)\frac{\partial}{\partial\xi} + \frac{1}{\xi^2-1}\frac{\partial^2}{\partial\phi^2}\right] \\ &-\frac{R}{4}\left[\frac{\partial}{\partial\eta}(1-\eta^2)\frac{\partial}{\partial\eta} + \frac{1}{1-\eta^2}\frac{\partial^2}{\partial\phi^2}\right] - \frac{R^2}{2}\xi. \end{aligned} \tag{2}$$

For a linearly polarized pulse $\vec{A}(t) = A(t)\hat{\mathbf{y}}$ along the molecular axis, the interaction hamiltonian $H_{\mathrm{int}}(t)$ under the dipole approximation is respectively expressed as

$$\begin{aligned} H^V_{\mathrm{int}}(t) &= -\mathrm{i}A(t)\frac{R^2}{4}\left[\eta(\xi^2-1)\frac{\partial}{\partial\xi} + \xi(1-\eta^2)\frac{\partial}{\partial\eta}\right] \\ &= -\mathrm{i}A(t)\frac{R^2}{4}\left[\eta\sqrt{\xi^2-1}\frac{\partial}{\partial\xi}\sqrt{\xi^2-1} + \xi\sqrt{1-\eta^2}\frac{\partial}{\partial\eta}\sqrt{1-\eta^2}\right], \end{aligned} \tag{3}$$

in the velocity gauge, and as

$$H^L_{\mathrm{int}}(t) = E(t)(R/2)^4(\xi^2-\eta^2)\xi\eta, \tag{4}$$

in the length gauge, with $E(t) = -\partial_t A(t)$.

Method[1]

The angular variables (η, ϕ) are expanded using the spherical harmonics Y_ℓ^m, and the radial coordinates ξ is discretized by the finite element discrete variable representation (FE-DVR), i.e.

$$\Psi(\xi,\eta,\phi,t) = \sum_{I,\ell,m} a_{I\ell,m}(t)\chi_I(\xi)Y_\ell^m(\arccos\eta, \phi), \tag{5}$$

where $\chi_I(\xi)$ stands for $\chi_i^q(\xi)$, i.e., the i-th basis function on the q-th finite element

$$\chi_i^q(\xi) = f_i^q(\xi)\Theta(\xi-\xi_0^q)\Theta(\xi_{n-1}^q-\xi), \quad i = 1,\dots,n-2, \tag{6}$$

with the neighbouring finite elements connected by the bridge function

$$\begin{aligned} \chi_{n-1}^q(\xi) = &\frac{f_{n-1}^q(\xi)}{\sqrt{1+w_0^{q+1}/w_{n-1}^q}}\Theta(\xi-\xi_0^q)\Theta(\xi_{n-1}^q-\xi) \\ &+ \frac{f_0^{q+1}(\xi)}{\sqrt{w_{n-1}^q/w_0^{q+1}+1}}\Theta(\xi-\xi_0^{q+1})\Theta(\xi_{n-1}^{q+1}-\xi), \end{aligned} \tag{7}$$

where Q is the total number of finite elements, $f_i^q(\xi)$ is the DVR basis function of order n and $\Theta(x)$ is the Heaviside theta function. In practice, to overcome the singularity at $\xi = 1$, one uses the Gauss-Radau quadrature with the right end point fixed for the first finite element and the Gauss-Lobatto quadrature for the rest of the elements. For a linearly polarized pulse along the molecular axis, m is conserved and taken to be $\mathbf{0}$ in Eq. (5). For time evolution, we use Arnoldi propagator with an adaptive time step control. Finally, the wavefunction is projected onto scattering state of H_2^+ calculated by Killingbeck-Miller method to get physical observable. Splitting scheme in the asymptotic region is used to save the computation effort.

What's new[2]

Usually, the Gauss-quadrature approximation (GA) is used to evaluate matrix elements in DVR, i.e. for any operator D, we can compute $\langle i|D|j\rangle$ by

$$\langle i|D|j\rangle = \int_a^b f_i^*(x)[Df_j](x)\,\mathrm{d}x \approx \sum_k w_k f_i^*(x_k)[Df_j](x_k), \tag{8}$$

where $\{f_i(x)\}$ are often chosen to be Lagrange polynomials satisfying $f_i(x_k) = \delta_{ik}$. One can show, for differential operator $D = g^*(x)\,\mathrm{d}/\mathrm{d}x\, g(x)$, such kind of GA would result in a non-anti-Hermitian matrix except for few cases, i.e., $D_{ij} \neq -D_{ji}^*$, causing a non-Hermitian Hamiltonian and unstable time evolution of wavefunction. In the present work, we choose to approximate $\langle i|D|j\rangle$ as follows

$$\langle i|D|j\rangle = \int_a^b f_i^*(x)g(x)\frac{\mathrm{d}}{\mathrm{d}x}[g(x)f_j](x)\,\mathrm{d}x \approx w_i g(x_i)^* g(x_j) f_j'(x_i), \tag{9}$$

to get an anti-Hermitian matrix. To apply the new GA for the differential operator in the interaction hamiltonian of the velocity gauge, we look back on Eq. (3) and notice that $g(x) = \sqrt{x^2-1}$ in Eq. (9). Then, for the q-th finite element, one can derive

$$\begin{aligned} &\int f_i^q(\xi)\sqrt{\xi^2-1}\frac{\partial}{\partial\xi}\sqrt{\xi^2-1}f_j^q(\xi)\,\mathrm{d}\xi \approx \\ &\qquad \begin{cases} (-1)^{i-j}\dfrac{\sqrt{(\xi_i^2-1)(\xi_j^2-1)}}{\xi_i-\xi_j}, & i \neq j, \\ [\delta_{i,n-1} - \delta_{i,0}(1-\delta_{q,1})]\,\dfrac{\xi_i^2-1}{2w_i^q}, & i = j. \end{cases} \end{aligned} \tag{10}$$

As one can see, the anti-hermiticity is hold except for diagnal terms at boundaries of different finite elements, which would cancel out since the bridge function Eq. (7) is applied.

Results

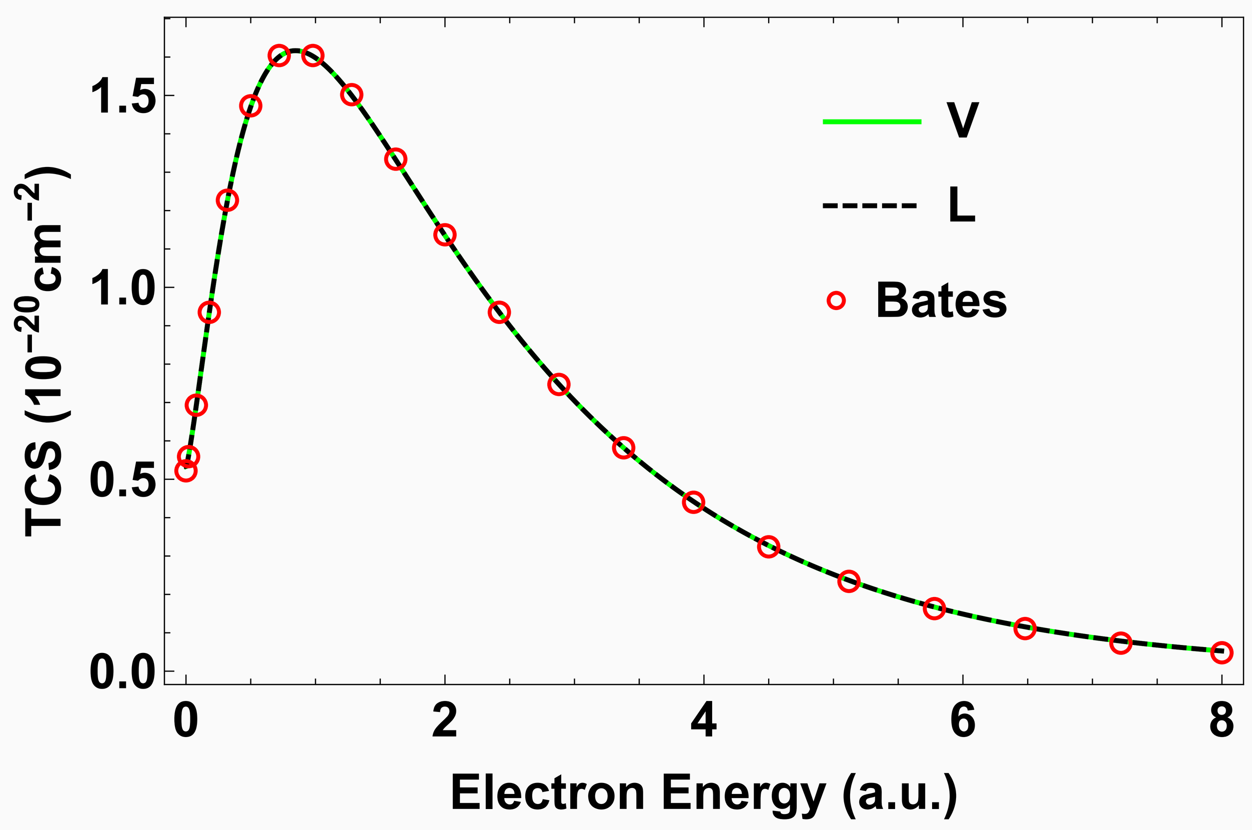

Figure 1: Total cross section (TCS) by one-photon ionization at equilibrium distance R = 2 a.u., from the present velocity gauge (green solid line) and length gauge (black dashed line), compared with Bates's data (red circle).

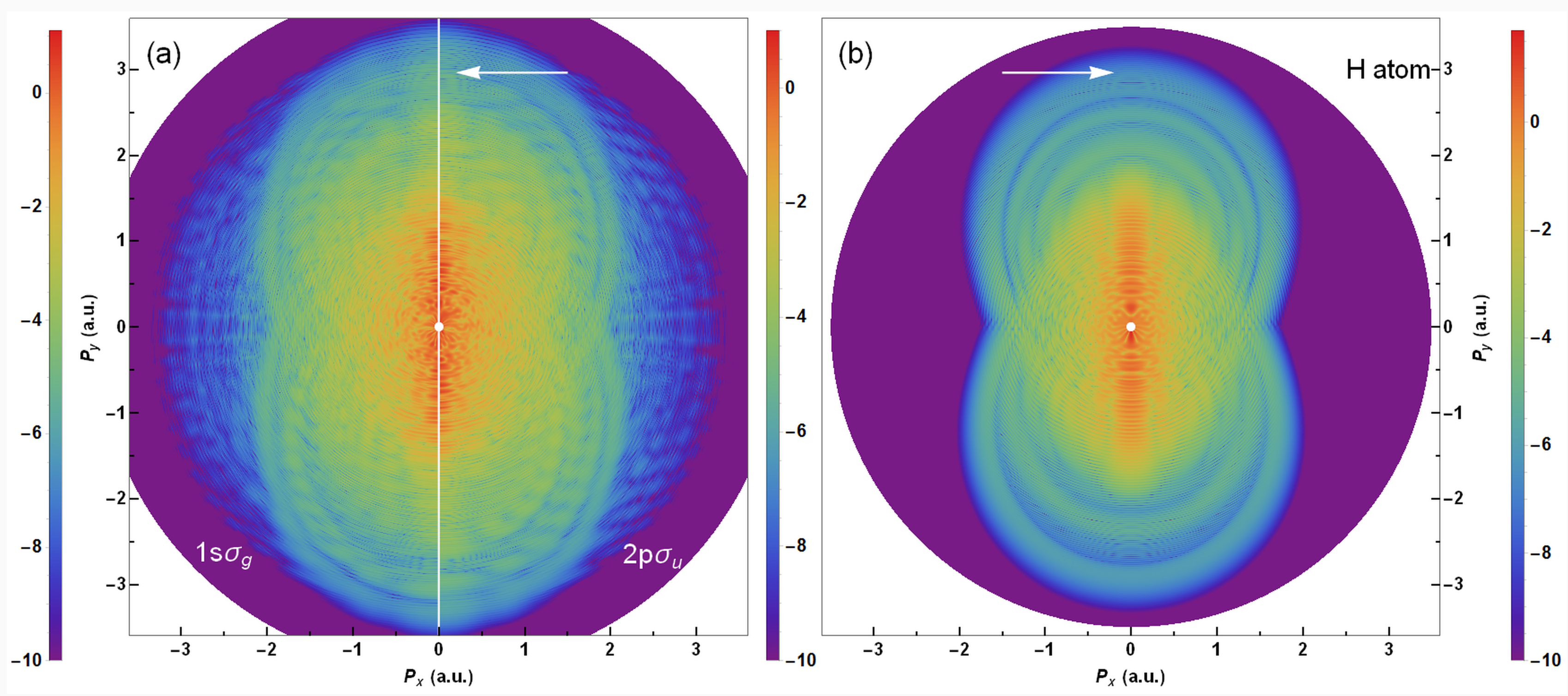

Figure 2: Electron momentum distributions by an **8**-cycle pulse at the wavelength of 800 nm and the peak intensity of $2\times10^{14}\,\mathrm{W\,cm^{-2}}$ for: (a) H_2^+ at $R = 23.3$ a u from $1s\sigma_g$ (left side) and $2p\sigma_u$ (right side); (b) H atom from the $1s$ state. White arrows indicate the position corresponding to energy of $10.007U_p$. Compared to the atom, one can clearly observe the extended cutoff for the molecular case due to the event that the electron is ionized from one nuclear but re-collides with the other.

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Reference

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