Adiabatic theory of molecular ionization including nuclear motion

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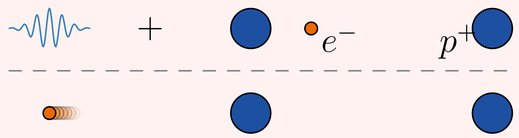
Introduction

- Adiabatic theory of tunneling ionization [1, 2]
- Works well in the adiabatic limit, when the time scale of the laser field is much larger than that of the electron
- Here we treat molecules including nuclear motion

Model

We consider a 1D model with electronic coordinate x and nuclear coordinate R interacting with a laser field $F(t)$



Adiabatic approximation (AAnf)

Starting with the Born-Oppenheimer ansatz

$$\Psi_{\mathrm{BO}}(x,R,t) = \Psi(R,t)\psi_e(x;R,F(t))$$

we can show that the nuclear wave function $\Psi(R,t)$ fulfills

$$i\frac{\partial}{\partial t}\Psi(R,t) = [H_R + E_e(R,F(t))]\Psi(R,t),$$

where $\psi_e(x;R,F(t))$ is an electronic Siegert state fulfilling

$$\left[-\frac{1}{2}\frac{d^2}{dx^2} + V(x,R) + Fx - E_e(R,F)\right]\psi_e(x;R,F) = 0.$$

A Siegert state is defined to have outgoing wave boundary conditions. As a consequence of this, the electronic energy $E_e(R,F(t))$ is complex, which leads to decay of $\Psi(R,t)$.

References

[1] O. I. Tolstikhin, and T. Morishita, Phys. Rev. A **86**, 043417 (2012)
[2] J. Svensmark, O. I. Tolstikhin, and Toru Morishita, Phys. Rev. A **101**, 053422, 2020.

Slides

Photo-electron momentum distribution (PEMD)

The (PEMD) in the adiabatic limit is given by

$$P_v^{\mathrm{nf}}(k) = \left|I_v^{\mathrm{nf}}(k)\right|^2,$$

where k is the momentum of the electron and

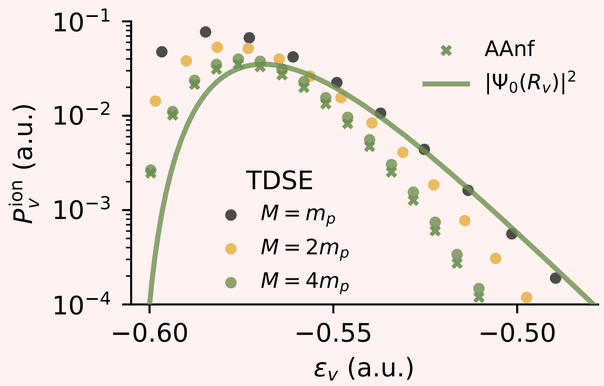
$$I_v^{\mathrm{nf}}(k) = I_v^a(k) + I_v^{r,I}(k) + I_v^{r,S}(k).$$

Rescattering contribution

$$I_v^{r,S}(k) = \sum_{\pm,i,r} p_\pm^{r,S}(t)\phi_{v,i}(t)\exp\left[i\mathcal{S}^{\mathrm{elec}}(t;k) + i\varepsilon_v t\right]\Big|_{t=t_{r,\pm}}$$
$$\phi_{v,i}(t) = \int_0^\infty \chi_v(R)S^{\pm\mathrm{sgn}(u_f(t))}(u_f(t);R)g^{\mathrm{wp}}(R,t)dR$$
$$g^{\mathrm{wp}}(R,t) = e^{-iH_R(t-t_i)}[f(R,F(t_i))\Psi(R,t_i)].$$

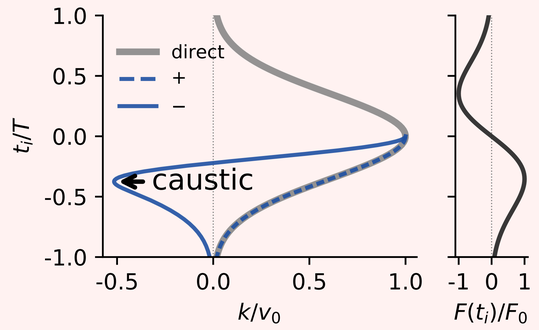
t_i and t_r is the ionization and rescattering time of an electron with final momentum k.
$\chi_v(R)$ is the vibrational state of the molecular ion.

Isotope effect



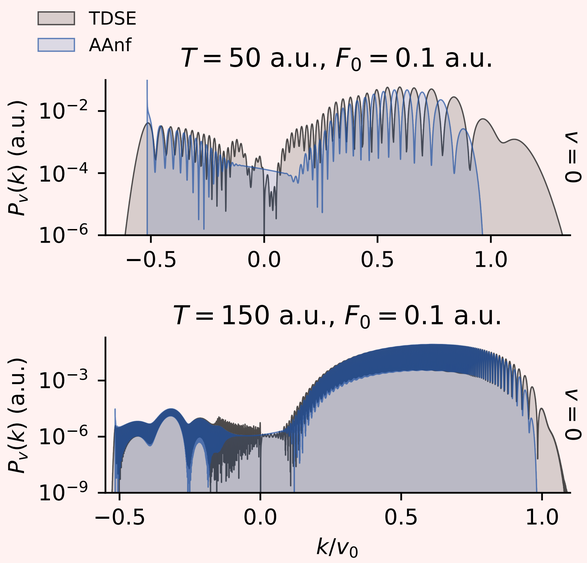
Energy distribution narrows with increasing nuclear mass. This reflects the nuclear wave function, and this connection can be explained using AAnf.

Trajectories (one-cycle pulse)



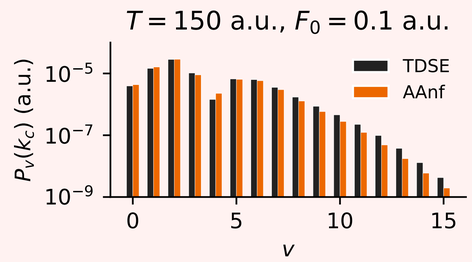
v_0: Maximal momentum
T: Pulse period
+, −: Forward and backward scattered electron trajectories

Convergence (PEMD)



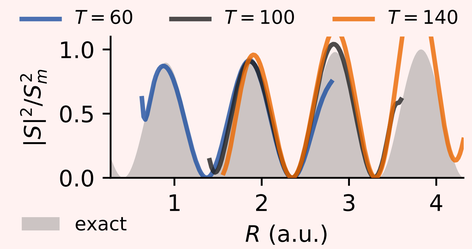
where $P(k) = \sum_v P_v(k)$. For longer pulse wavelengths the AAnf approaches the time-dependent Schrödinger equation (TDSE) results.

PEMD at the caustic



S-matrix reconstruction

Taking amplitudes and phases from the TDSE, the S-matrix can be reconstructed by assuming the nuclear wave packet to be known.



Wave packet reconstruction

Conversely, assuming S-matrix is known we can reconstruct the nuclear wave packet