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ATTOSECOND PHYSICS

UNIT VI
NUMERICAL METHODS

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CLASSICAL TRAJECTORY MONTE CARLO

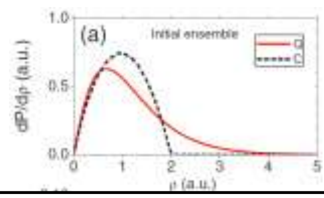
Classical Trajectory Monte Carlo (CTMC) is a non-perturbative method in which the classical equations of motion (Hamilton equations) are numerically solved. Computer experiment.

• The initial state is sampled by a set of points in the phase space (position and momentum) chosen at random fulfilling the microcanonical ensemble:

$$\rho_{\varepsilon_i}(\vec{r},\vec{p}) = C_1\delta(\varepsilon + I_p) = C_1\delta\left(\frac{p^2}{2} + V(r) + I_p\right)$$

C_1 : Normalization constant

• N_{traj} points in the phase space $(\vec{r},\vec{p})$ follow the Hamilton equations describing N_{traj} trajectories.



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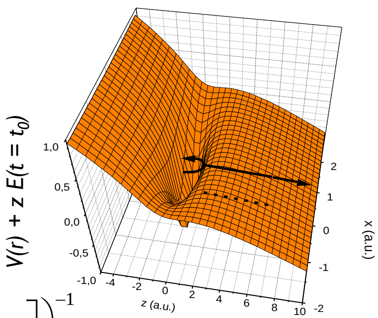
CTMC-T (cont.)

• The electron is allowed to tunnel out through the potential barrier each time that finds the classical turning point: $p_z = 0$, with $zE(t) < 0$ with a tunneling probability T given by the WKB approximation:



$$T = \left(1 + \exp\left[2\sqrt{2} \int_{\tilde{z}_1}^{\tilde{z}_2} d\tilde{z} \sqrt{V(\tilde{z}) - V(\tilde{z}_1)} \right] \right)^{-1}$$

The electron is tunneled from the turning point (x_1, y_1, z_1) to the other side of the barrier (x_1, y_1, z_2) in the same energy manifold. The tunneling path through the potential barrier chosen is the one which maximizes the tunneling probability.
Recapture by tunneling is neglected in the calculations.
If the tunneling probability is switched off ($T = 0$), then a pure classical calculation (CTMC) arises. In this case, the only ionization way is over the barrier ionization.

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CTMC-T (cont.)

• After tunneling, classical trajectories must be followed beyond the end of the laser pulse (τ) if one wants to calculate the momentum (or angle) because the interaction of the active electron with the core continues. If one wants to calculate the energy distribution this is not necessary since the energy is a constant of motion.
The energy and momentum distributions are calculated as

$$\frac{dP}{d\varepsilon} = \frac{N_i}{N_{\text{traj}} \Delta\varepsilon}; \quad \frac{d^2P}{dp_\rho dp_z} = \frac{N_i}{N_{\text{traj}} \Delta p_\rho \Delta p_z}; \quad \frac{dP}{dL} = \frac{N_i}{N_{\text{traj}} \Delta L}$$

N_i : number of trajectories with final energy in the interval $(\varepsilon, \varepsilon + \Delta\varepsilon)$

N_i : number of trajectories with final momentum in the interval $(p_\rho, p_\rho + \Delta p_\rho) \cap (p_z, p_z + \Delta p_z)$

N_i : number of trajectories with final angular momentum in the interval $(L, L + \Delta L)$

$$\text{CTMC} \xrightarrow[N_{\text{traj}} \to \infty]{} \text{classical mechanics}$$

$$\frac{\Delta\sigma}{\sigma} = \left(\frac{N_{\text{traj}} - N_i}{N_{\text{traj}} N_i} \right)^{1/2} \quad \text{standard deviation}$$

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CTMC-T (cont.)

Alternative Method:

- The electron dynamics before tunneling is neglected.

- Initial conditions for each trajectory according to quantum tunneling probabilities (ADK) are chosen with energy $-I_p$, postion $x_0(t_0) = y_0(t_0) = 0$ and $z_0(t_0)$ as the root of $-V(z) - zE(t) = -I_p$.

The tunneling probability is given by the ADK rate

$$w_0(t_0, v_{0\perp}) \propto e^{-\frac{2(2I_P)^{3/2}}{3F(t_0)}} e^{-\frac{\sqrt{2I_P}v_{0\perp}^2}{F(t_0)}} \qquad t_0 = \mathrm{Re}[t_s]$$

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CTMC-T (cont.)

$$P_{ADK}(t_0) = \int_0^{t_0} dt W_{ADK}(t),$$

which can be very high when considering a long pulse. In this case the depletion of the ground state must be considered:

$$P_{tun}(t_0) = P_{ADK}(t_0) e^{-P_{ADK}(t_0)}$$

- The momentum distributions in p_ρ and p_z are gaussian

This method is much less CPU time consumer than CTMC-T

Delone & Krainov, J. Opt. Soc. Am. B **8**, 1207 (1991).

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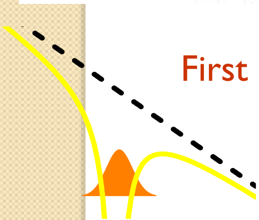
CAPE CONICET

Two-step Semiclassical Model

$$T = -i\int_{-\infty}^{+\infty} dt \left\langle \chi_{\vec{k}}^{-}(\vec{r},t) \middle| \vec{r}\cdot\vec{F}(t) \middle| \phi_i(\vec{r},t) \right\rangle$$

Saddle point approximation:

$$T = -i\sqrt{2\pi}\sum_{t_s} \frac{e^{i\Phi(\vec{k},t_s)}}{\left|\ddot{\Phi}\left(\vec{k},t_s\right)\right|^{1/2}} \left|\left\langle \chi_{\vec{k}}^{-}(\vec{r},t_s) \middle| \vec{r}\cdot F(\vec{t}_s) \middle| \varphi_i(\vec{r},t_s) \right\rangle\right| \qquad \left.\frac{d\Phi(t)}{dt}\right|_{t=t_s} = 0$$



First step: Tunneling through the potential barrier

$$w_0\left(t_0, v_{0\perp}\right) \propto e^{-\frac{2(2I_P)^{3/2}}{3F(t_0)}} e^{-\frac{\sqrt{2I_P}v_{0\perp}^2}{F(t_0)}} \qquad t_0 = \mathrm{Re}[t_s]$$

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Semiclassical Methods: Two-step Model

Second step: CTMC+phase

$$\dot{\vec{r}} = \frac{\partial H}{\partial \vec{p}}; \qquad -\dot{\vec{p}} = \frac{\partial H}{\partial \vec{r}}$$

$$\frac{dP}{d\vec{k}} = \left|\sum_j \sqrt{w_0\left(t_0^j, v_{0\perp}^j\right)} e^{i\Phi(\vec{k},t_0^j)}\right|^2$$

$$\frac{dP}{d\vec{k}} = \sum_j\sum_{j'} \sqrt{w_0(t_0^j, v_{0\perp}^j)}\sqrt{w_0(t_0^{j'}, v_{0\perp}^{j'})}\, e^{i[\Phi(\vec{k},t_0^j)-\Phi(\vec{k},t_0^{j'})]}$$

$$= \sum_j w_0(t_0^j, v_{0\perp}^j) \qquad \text{CTMC}$$

mising in QTMC

$$\Phi\left(t_0^j, \mathrm{v}_0^j\right) = \left[\vec{k}+\vec{A}(t)\right]\cdot\vec{r} + I_P t_0^j - \vec{v}_0^j\cdot\vec{r}_0^j - \int_{t_0^j}^{\infty} dt \left(\frac{\vec{p}^2(t)}{2} + V(r) - \vec{r}(t)\cdot\frac{\partial V(r)}{\partial \vec{r}}\right)$$

N. I. Shvetsov-Shilovski *et al.*, Phys. Rev. A **94**, 013415 (2016).

The detection process takes place at t >> τ . Energy and angular momentum are constant of motion for t > τ, unlike linear momentum **p**.
Projection of the phase space at t = τ to t → ∞ .

Numerical Solution of the Time-Dependent Schrödinger Equation (TDSE)

Several methods are usually employed to numerically solve the TDSE.
I will show only one: Pseudo-spectral method (Tong & Chu, Chem. Phys. **217**, 119 (1997)).

We will see the basics of the method for a hydrogenic system in the presence of a linearly polarized electric field.

$$i\frac{\partial\Psi}{\partial t} = \left(H_0 + H_{\text{int}}\right)\Psi$$

$$H_0 = \frac{p^2}{2} - \frac{1}{r} = -\tfrac{1}{2}\frac{d^2}{dr^2} + \frac{\hat{L}^2}{2r^2} - \frac{1}{r}$$

$$H_{\text{int}} = E(t)z$$

The problem has cylindrical symmetry => 2D problem with L_z constant of motion ➔ (magnetic quantum number) *m = const. = 0*

Numerical Solution of the TDSE (cont.)

We start solving the TISE

Pseudospectral method: (Tong & Chu, Chem. Phys. **217**, 119 (1997))

One chooses a grid (r_i,θ_j) in the coordinate space.

We can expand the wave function in Legendre polynomials:

$$\Psi(r_i,\theta_j,t) = \sum_{l=0}^{l_{\max}} g_l(r_i) P_l\left(\cos\theta_j\right)$$

where $g_l(r_i)$ are determined by Gaussian quadratures (Gauss-Legendre)

$$g_l(r_i) = \sum_{k=1}^{L+1} w_k P_l\left(\cos\theta_k\right)\Psi(r_i,\theta_k,t)$$

$\{\cos\theta_k\}$ are the $L+1$ zeros of $P_{L+1}\left(\cos\theta_k\right)$ and w_k are the corresponding weights

Numerical Solution of the TDSE (cont.)

We search for an optimal (referring to the grid) discretization method which solves the eigenvalue and eigenfunction problem:

$$H_{0,l}(r)\chi_l(r) = \varepsilon\chi_l(r)$$

For the Coulombian case $V(r) = -\frac{1}{r}$, there are two problems:

a) The singularity at $r = 0$
b) Long range potential: $r \to \infty$

Therefore, we must restrict the semi-infinite domain:

$$(0,\infty) \to [r_{\min}, r_{\max}]$$

$$r_{\min} \ll 1 \text{ a.u.} \quad \text{and} \quad r_{\max} \gg 1 \text{ a.u.}$$

For what we need a great amount of grid points.

Numerical Solution of the TDSE (cont.)

We extend the generalized pseudo-spectral method:

$$(0,\infty) \text{ or } [r_{\min}, r_{\max}] \to [-1,1]$$

$$r(x) = L\frac{1+x}{1-x+\alpha}; \quad \alpha = \frac{2L}{r_{\max}}; \quad L \text{ mapping parameters}$$

$$\text{Example: } r = 0 \Rightarrow x = -1$$

$$r = r_{\max} \Rightarrow x = 1$$

This assure to have more density of grid points near $r = 0$.
The eigenvalue problem of last slide is equivalent to:

$$H_{0,l}(x)\Phi_l(x) = \varepsilon\,\Phi_l(x)$$

$$H_{0,l}(x) = -\tfrac{1}{2}\frac{1}{r'(x)}\frac{d^2}{dx^2}\frac{1}{r'(x)} + \frac{l(l+1)}{2r^2(x)} - \frac{1}{r(x)}$$

$$\Phi_l(x) = \sqrt{r'(x)}\chi_l\left(r(x)\right)$$

I do not continue with the details

Numerical Solution of the TDSE (cont.)

Time evolution

We make use of the Split Operator Method up to second order to cope with the time evolution of the TDSE.

$$\Psi(\vec{r},t+\Delta t) = e^{-iH_0\Delta t/2} e^{-iH_{\text{int}}(\vec{r},t+\Delta t/2)\Delta t} e^{-iH_0\Delta t/2}\Psi(\vec{r},t) + O(\Delta t^3)$$

Three steps:

i. The wave function is propagated during half time step in the energy space of H_0

$$\Psi_1(\vec{r},t) = e^{-iH_0\Delta t/2}\Psi(\vec{r},t)$$

$$\Psi_1(\vec{r},t) = e^{-iH_0\Delta t/2}\Psi(\vec{r},t) = \sum_{l=0}^{l_{\max}}\left[e^{-iH_{0,l}\Delta t/2} g_l(r_i)\right] P_l\left(\cos\theta_j\right)$$

In this equation each g_l must be propagated independently with each $H_{0,l}$

$$H_{0,l} = -\tfrac{1}{2}\frac{d^2}{dr^2} + \frac{l(l+1)}{2r^2} - \frac{1}{r}$$

Numerical Solution of the TDSE (cont.)

ii. The wave function Ψ_1 is transformed to the coordinate space and then a "kick" (delta function) is applied with all the force shrunk in t + Δt/2 with a momentum transfer given by

$$\Delta p = -\int_t^{t+\Delta t} dt' E(t') \simeq -E(t+\tfrac{\Delta t}{2})\Delta t$$

The evolution operator of a kick is: $e^{i\Delta\vec{p}\cdot\vec{r}} = e^{i\Delta p z}$ boost operator

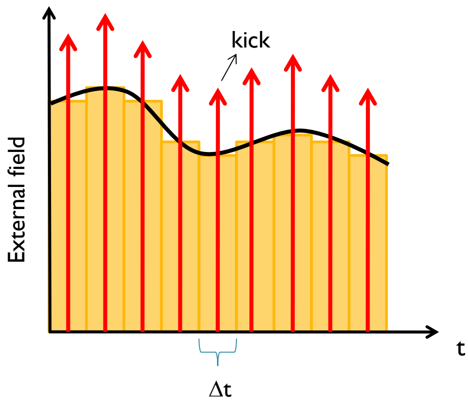
This implies a translation in the momentum space:

$$\Psi_2(\vec{r},t) = e^{-iE(t+\Delta t/2)\Delta t z}\Psi_1(\vec{r},t)$$

Numerical Solution of the TDSE (cont.)

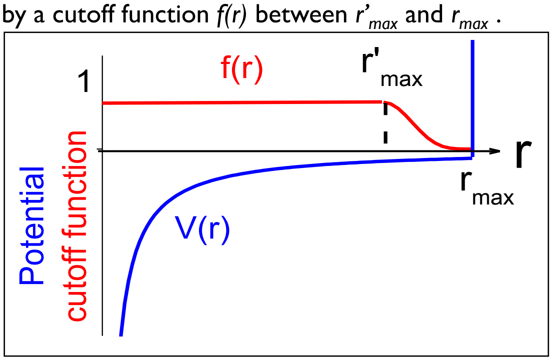
iii. Finally, the wave function Ψ_2 is transformed to the energy space of H_0 and then propagated (with no interaction) for another half time step

$$\Psi(\vec{r},t+\Delta t)=e^{-iH_0\Delta t/2}\Psi_2(\vec{r},t)$$



Numerical Solution of the TDSE (cont.)

The wave function eventually runs into the "wall of the box" $r = r_{max}$, or also $\varepsilon = \varepsilon_{max}$ or also $l = l_{max}$.
To do r_{max} ε_{max} l_{max} very high, it demands very much CPU time and we cannot avoid the wave function run into the "walls".
The wave packet will bounce against the walls which is an unphysical situation: We must avoid this unpleasant effect by including absorbing walls. For example one can chop the wave function many times along time by multiplying it by a cutoff function *f(r)* between r'_{max} and r_{max} .



We must be carefull of the quantum Zeno effect

Numerical Solution of the TDSE (cont.)

Quantum Zeno Effect

A quantum state evolves $\Psi(t) = e^{-iHt}\Psi(0)$

The decay probability is given by the correlation function:

$$P(t) = \left|\langle \Psi(0) | \Psi(t) \rangle\right|^2 = \left|\langle \Psi(0) | e^{-iHt} | \Psi(0) \rangle\right|^2$$

We use a series expansion of the evolution operator up to second order:

$$e^{-iHt} \simeq 1 - iHt + \tfrac{i^2}{2} H^2 t^2$$

$$P(t) = \left|1 - i\langle H\rangle t - \tfrac{1}{2}\langle H^2\rangle t^2\right|^2$$

$$= \underbrace{\left(1 - \tfrac{1}{2}\langle H^2\rangle t^2\right)^2}_{\text{Re}} + \underbrace{\left(\langle H\rangle t\right)^2}_{\text{Im}}$$

$$= 1 - \underbrace{\left(\langle H^2\rangle - \langle H\rangle^2\right)}_{\sigma^2 \geq 0} t^2$$

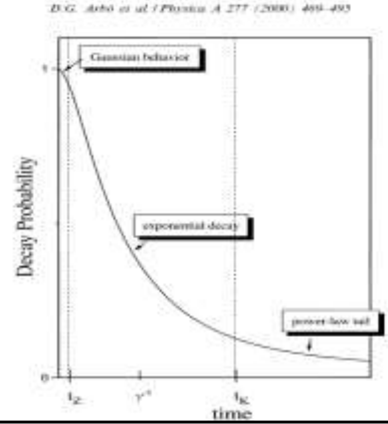
Numerical Solution of the Time-Dependent Schrödinger Equation (TDSE)

An exponential decay: $e^{-\Gamma t} \simeq 1 - \Gamma t + \tfrac{1}{2}\Gamma^2 t^2$ The linear term is absent in *P(t)*

=> exponential decay is forbidden by quantum mechanics

$\dot{P}(t=0) = 0$ The decay starts with null slope

$\left(\dot{e^{-\Gamma t}}\right)_{t=0} = -\Gamma$ The decay starts with slope $-\Gamma$



t_z has been measured for some systems.
But it is extremely difficult since $t_z \ll \Gamma^{-1}$

For $t \gg \Gamma^{-1}$ the exponential decay also fails:
For $t > t_K$ (Khalfin) it is polynomial:

$$P(t=0) \sim \frac{1}{t^q}$$

The value of *q* depends on the system

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Numerical Solution of the TDSE (cont.)

Zeno's Paradox

- I suppose exact exponential decay and a quantum system measured N times at regular intervals of time Δt

$$P(t) = e^{-\Gamma t}$$

$$P(\Delta t) = (1 - \Gamma \Delta t)\, P(0)$$

$$P(2\Delta t) = (1 - \Gamma \Delta t)\, P(\Delta t) = (1 - \Gamma \Delta t)^2\, P(0)$$

$$\vdots \qquad \vdots \qquad \vdots$$

$$P(t = N\Delta t) = \left(1 - \frac{\Gamma t}{N}\right)^N P(0)$$

$$\lim_{N\to\infty} P(t) = e^{-\Gamma t}$$

The system decays in the same way as with no measurement.

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Numerical Solution of the TDSE (cont.)

- I suppose the system decays non-exponentially and is measured N times at regular intervals of time Δt with $\Delta t \ll t_Z$

$$P(\Delta t) = (1 - \sigma^2 \Delta t^2)\, P(0)$$

$$P(2\Delta t) = (1 - \sigma^2 \Delta t^2)\, P(\Delta t) = (1 - \sigma^2 \Delta t^2)^2\, P(0)$$

$$\vdots \qquad \vdots \qquad \vdots$$

$$P(t = N\Delta t) = (1 - \sigma^2 \Delta t^2)^N P(0) = \left(1 - \frac{\sigma^2 t^2}{N^2}\right)^N P(0)$$

$$\lim_{N\to\infty} P(t) = 1$$

The system does not decay!

Numerical Solution of the TDSE (cont.)

Using this pseudo-spectral method and the split operator method with the help of the masking function we find the wave function at the end of the pulse.

The detection process takes place at t >> τ .

As the energy is a constant of motion for t > τ ,

$$\frac{dP}{d\varepsilon} = \sum_l \left| \langle k,l | \Psi(\vec{r},\tau) \rangle \right|^2$$

$|k,l\rangle$ is the eigenstate of H_0 with eigenenergy $\frac{k^2}{2}$ and orbital quantum number l

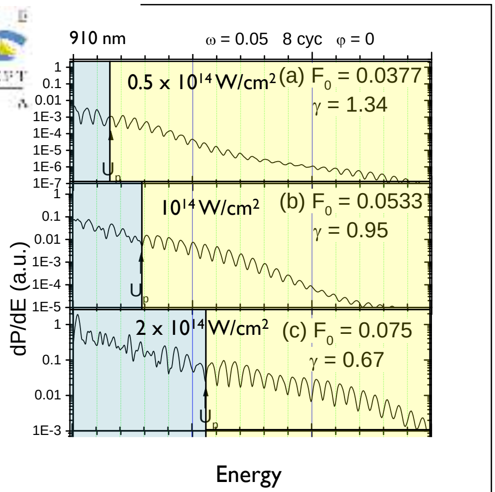
The process of detecting an electron with a specific momentum can be seen as the projection of the wave function right after the end of the pulse onto the Coulomb waves (solution of the continuum of H_0).

$$\frac{dP}{d\vec{k}} = \frac{1}{4\pi k} \left| \sum_l e^{i\delta_l(k)} \sqrt{2l+1} P_l(\cos\theta) \langle k,l | \Psi(\vec{r},\tau) \rangle \right|^2$$

$\delta_l(k)$: phase-shift (analytical for the case of hydrogenic atoms)

The projection is necessary for observables which are not constant of motion of the free evolution once the external field has been switched off.

Numerical Solution of the TDSE (cont.)



- Coexistence of multiphoton and tunneling.
- U_p delimits the two regions.
- Excited bound states are not responsible for the non-equidistance of photoelectron peaks.

Tunneling Regime: $\gamma < 1$

Multiphoton region

Numerical Solution of the TDSE (cont.)

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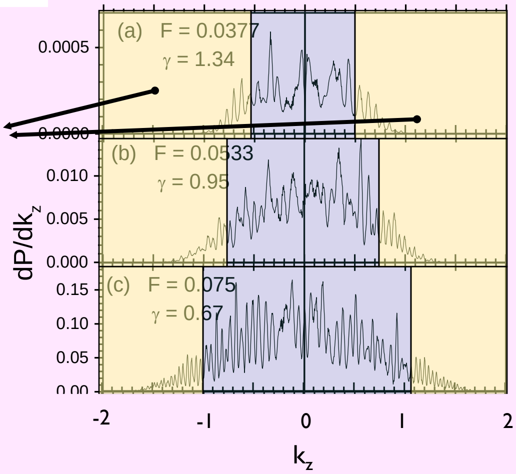

Multiphoton region

Ponderomotive momentum

$$k_p = \sqrt{2U_p}$$