Non-adiabatic theories: orbital effects

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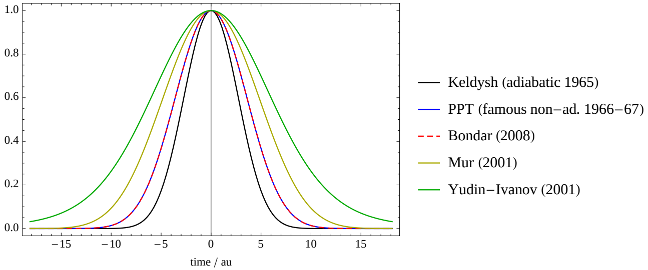
Recap

- take temporal changes of laser field into account
  $\rightarrow \omega$ and/or Keldysh parameter γ dependence of the ionisation probability

Keldysh: $\propto \exp\left\{-\frac{2(2I_p)^{3/2}}{3F(t)}\right\}$ $\leftrightarrow$ Bondar: $\propto \exp\left\{-\frac{2I_p}{\omega} f(\gamma, \vec{p}_{\text{final}}, t)\right\}$, [1]

PPT: $\propto \exp\left\{-\frac{2I_p}{\omega} g(\gamma, \epsilon, t)\right\} \cdot P(\vec{p}_{\text{final}})$, [2]

- in general:
  - wider spreads due to non-adiabatic effect
  - initial transverse momentum at tunnel exit (in elliptically polarised light)
- Various different treatments with different approximations



exit point

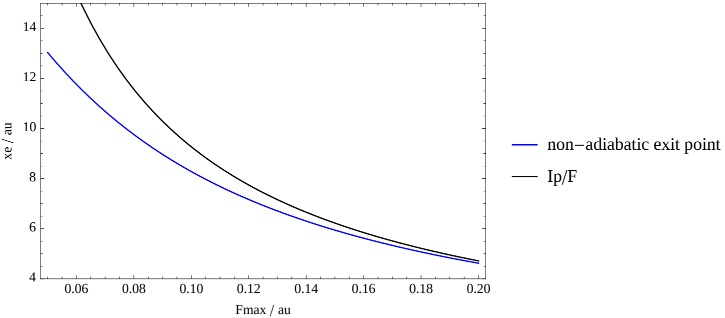
- non-adiabatic (PPT) exit point: [2]

$$x_e^{NA} = \frac{2I_p}{F\gamma^2}\left(\sqrt{\frac{1+\gamma^2}{1-s^2}} - 1\right)$$

where s is a paramater $s(\gamma, \epsilon) \in [0, \epsilon]$ from saddle point / transition point.

- compared to the simple triangular barrier exit point $x_e^{\triangle} = \frac{I_p}{F}$:

Out[38]=



Experiment

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Strong-Field Ionization Rate Depends on the Sign of the Magnetic Quantum Number

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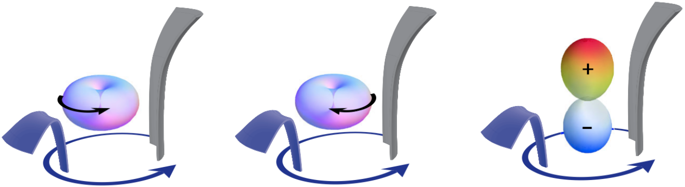
We report the first experimental observation of the dependence of strong-field ionization rate on the sign of the magnetic quantum number. We measure the strong-field sequential double ionization yield of argon by two time-delayed near-circularly polarized laser pulses. It is found that double-ionization yield is

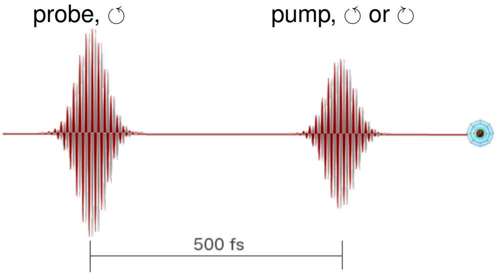
p orbitals: $l = 1$, $m_l = -1, 0, 1$



(a) P_{+1} orbital : Co-rotating (b) P_{-1} orbital : Counter-rotating (c) P_0 orbital

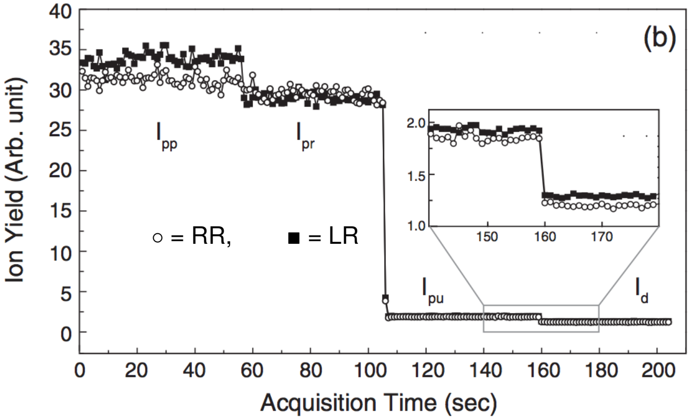
Experiment: Setup

- Argon atoms, [Ne] 3s2 3p6
- two strong IR pulses, near-circular polarisation, delayed long enough



- measurement goal: Ar $\rightarrow ϟ \rightarrow$ Ar^{+} $\rightarrow ϟ \rightarrow$ Ar^{2+}
- measurements: Ar^{2+} yield for
  - pump & probe, I_{pp}
  - probe alone, I_{pr}
  - pump alone, I_{pu}
  - dark, I_{d}

Experiment: Yield



[6]

Sequential double ionisation yield:
substract double ionisations from single pulses and dark counts

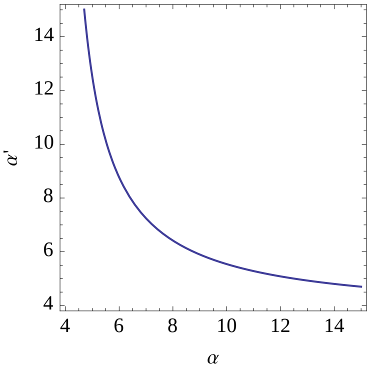
$$I_{\mathrm{SDI-LR}} = 4.21 \pm 0.98, \quad > \quad I_{\mathrm{SDI-RR}} = 1.16 \pm 0.92 \quad \text{arb. units}$$

Experiment: Result

$$\alpha := \frac{\mathcal{W}_{-R}}{\mathcal{W}_{+R}}$$

$$\alpha' = \frac{1 - 3.63 * \alpha}{3.63 - \alpha}$$

- symmetric function
- always > 1



$\Rightarrow$ co-rotoating and counter-rotating orbitals have different ionisation rate

ϟ the experiment does not conclusively show which direction is favoured! ϟ

$|m|$ or m dependence

- in the adiabatic limit, only the shape $|m|$ of the orbital has an influence on the ionisation rate:

into the corresponding formula of the adiabatic approximation:

$$w_{lm}(F) = \omega_0 |C_{\varkappa l}|^2 \left(\frac{6}{\pi}\right)^{1/2} \frac{(2l+1)(l+|m|)!}{2^{|m|}|m|!(l-|m|)!} \left(\frac{F}{2F_0}\right)^{|m|+3/2} \times \exp\left\{-\frac{2F_0}{3F}\left(1-\frac{1}{10}\gamma^2\right)\right\}. \quad (59)$$

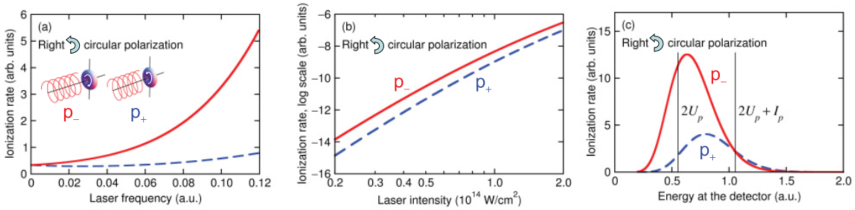
[7]

- in the non-adiabatic case, m dependence is visible

$$\frac{dw_n}{d\Omega} = \frac{A^2\omega^2}{8\pi^2}(2l+1)\frac{(l-|m|)!}{(l+|m|)!}\left|P_l^{|m|}\left(i\frac{p}{\kappa}\sin\theta\right)\right|^2 \times \frac{(\eta-\sqrt{\eta^2-1})^{2n}}{\sqrt{\eta^2-1}F\cos\theta}\left[\frac{\sqrt{\kappa^2+p^2\sin^2\theta}}{\frac{F}{\omega}(\eta+\sqrt{\eta^2-1})-p\cos\theta}\right]^{2m} \times \exp[2Fp\cos\theta\sqrt{\eta^2-1}/\omega^2], \quad (5)$$

[8]

Result


(a)
Right circular polarization
p−
p+
Ionization rate (arb. units)
Laser frequency (a.u.)
(b)
Right circular polarization
p−
p+
Ionization rate, log scale (arb. units)
Laser intensity (10^{14} W/cm^2)
(c)
Right circular polarization
p−
p+
$2U_p$
$2U_p + I_p$
Ionization rate (arb. units)
Energy at the detector (a.u.)

[9]

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