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Attosecond chronoscopy of photoemission - an introduction

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XLIC
XUV/X-ray light and fast ions for ultrafast chemistry
cost
EUROPEAN COOPERATION IN SCIENCE AND TECHNOLOGY

APS

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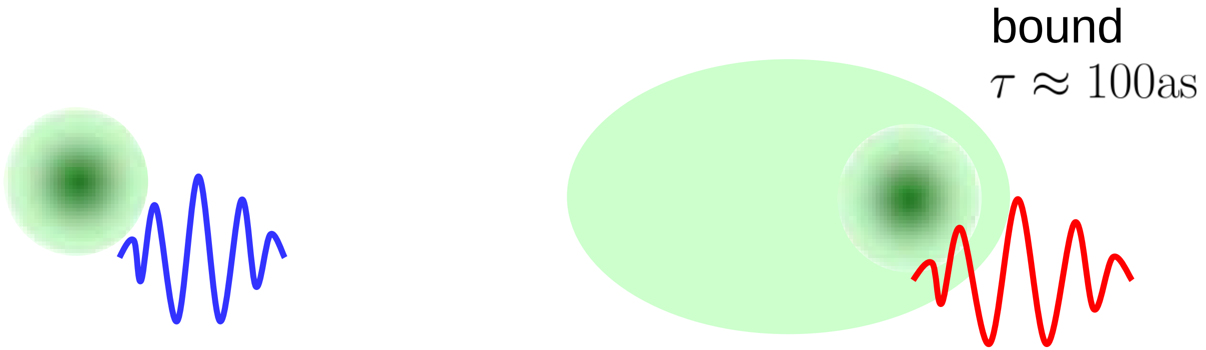
Vienna Computational Materials Laboratory
ViCoM

Timing electronic dynamics

Goal: Time-resolved electron wave packet



Overview

- **Time as observable**
- Clocks for electrons:
  - attosecond streaking
- Time-resolved photoionization: time delay and electron correlation
- Time ordering in double ionization
- Ionization delays in endohedral fullerenes
- Photoemission from surfaces and adlayers
- Attosecond streaking of tunneling time?
- Photoemission from nanotips

Time in quantum physics: parameter or observable?

- Position – momentum uncertainty:

$$\Delta p \Delta x \geq \frac{\hbar}{2} \leftrightarrow [x,p] = i\hbar$$

- Time – energy uncertainty:

$$\Delta E \Delta t \geq \frac{\hbar}{2} \leftrightarrow [t,H] = ?$$

„t is classical parameter with which no quantum mechanical operator is associated.“

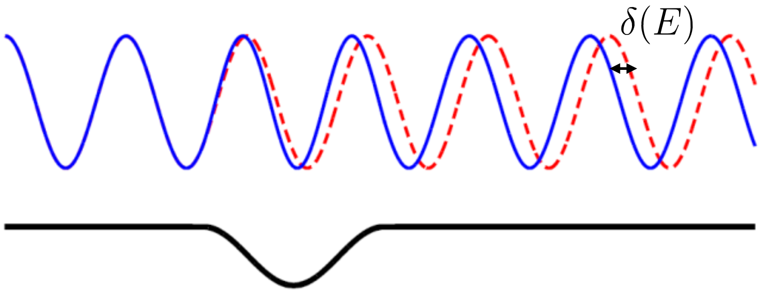
C. Cohen-Tannoudji

Time delay as observable in scattering

$$t \Leftrightarrow i\hbar \frac{\partial}{\partial E}$$

Eisenbud-Wigner-Smith time (delay) operator

$$t_{\text{EWS}} = i\hbar S^{\dagger}(E) \frac{\partial}{\partial E} S(E)$$



$$t_{\text{EWS}} = 2 \frac{d}{dE} \delta(E)$$

Eisenbud (1948), Wigner (1955), Smith (1960)

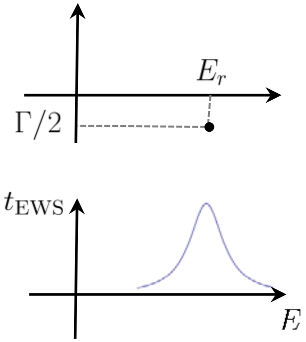
Time delay

$$t_{\mathrm{EWS}} = -i\hbar S^{\dagger}(E)\frac{\partial}{\partial E}S(E)$$

- Resonance:

$$S(E) = \frac{E-E_r-i\Gamma/2}{E-E_r+i\Gamma/2}$$

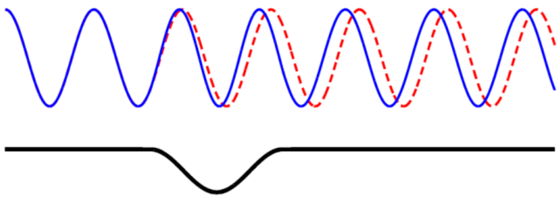
$$t_{\mathrm{EWS}}(E) = \frac{\Gamma}{(E-E_r)^2+\Gamma^2/4}$$



- Partial wave scattering phase shift:

$$S_\ell(E) = e^{2i\delta_\ell(E)}$$

$$t_{\mathrm{EWS}}^{(\ell)}(E) = 2\frac{\mathrm{d}}{\mathrm{d}E}\delta_\ell(E)$$



Eisenbud (1948), Wigner (1955), Smith (1960)

- Time delay of crest of wavepacket:

$$r(t) = v_g \left(t - t_{\mathrm{EWS}}(E)\right)$$

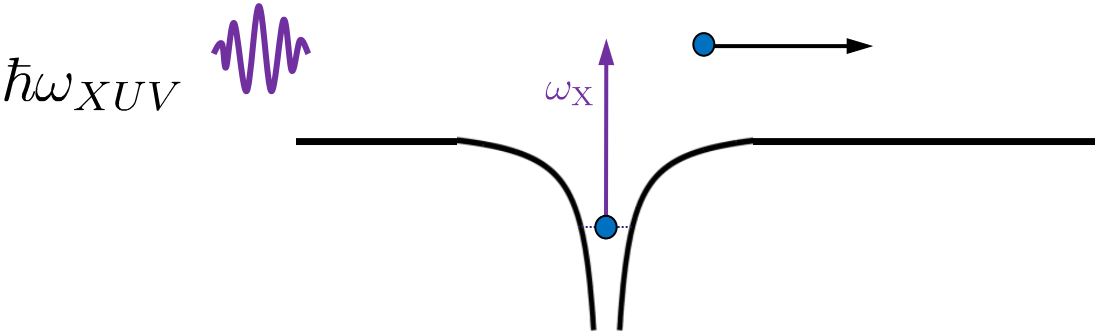
- Time delay of expectation value of wave packet:

$$\langle r(t) \rangle = v_g \left(t - t_{\mathrm{EWS}}(E)\right)$$

Brenig and Haag (1959)

- Well-defined only for short-ranged interaction (!)
- Extension to (in)coherent transport in complex systems

EWS delay for photoionization



Photoionization: scattering with photon **in**

→ Half-scattering process

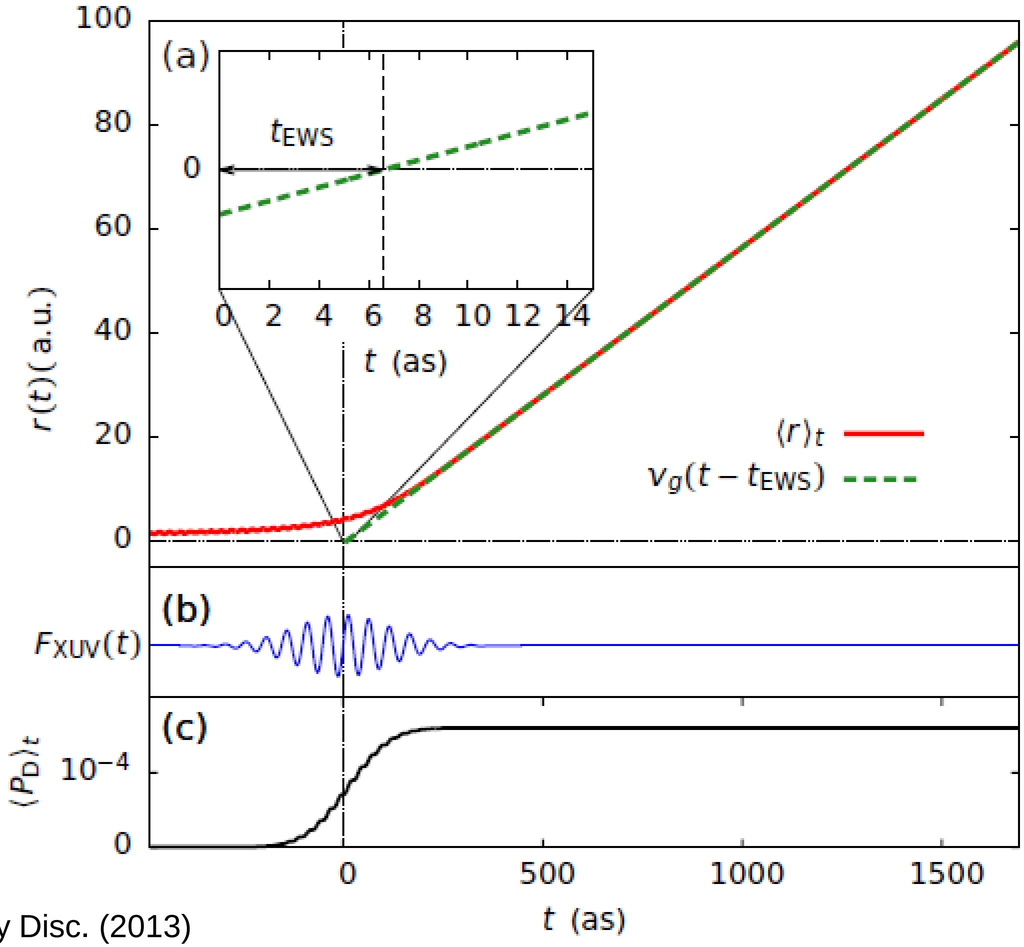
- No electronic reference wave packet
- Time-shift relative to XUV pulse

$$t_{\text{EWS}} = \frac{d}{dE}\delta(E) = \frac{d}{dE}\arg\langle\psi_f(E,\theta)|\hat{\mu}|\psi_i\rangle$$

L. Eisenbud, PhD thesis (Princeton, 1948); E.P. Wigner, Phys. Rev. **98**, 145 (1955); F. T. Smith, Phys. Rev. **119**, 349 (1960)

Photodetachment of Yukawa potential bound electron

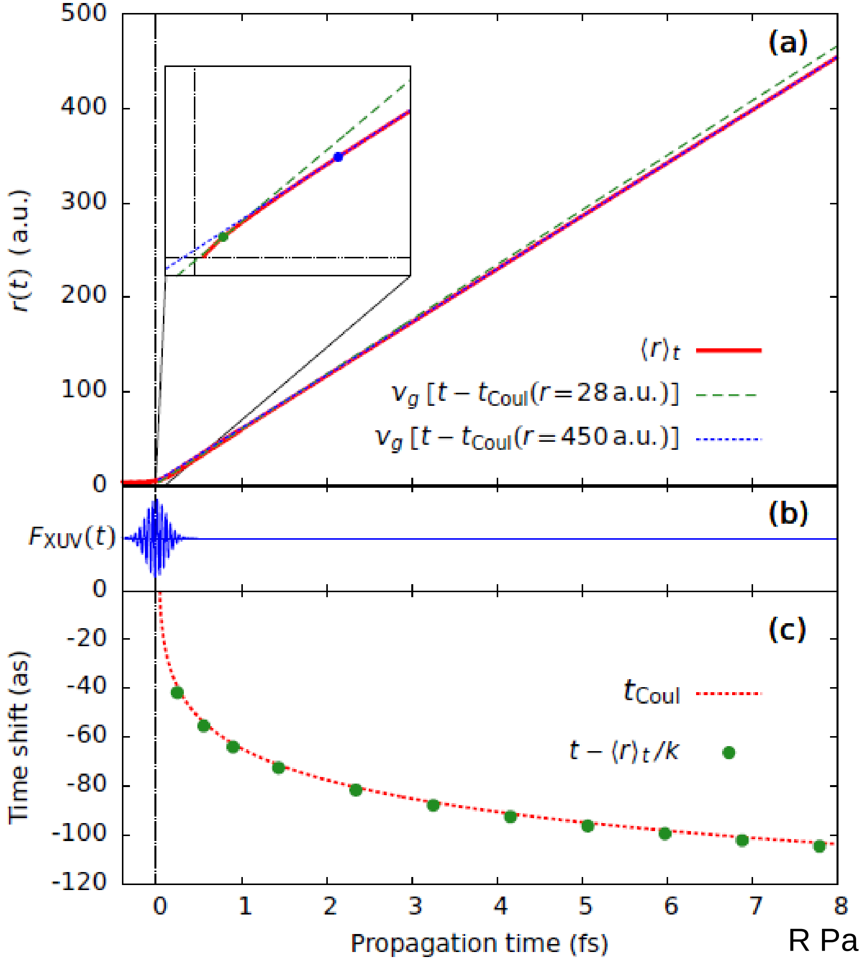
EWS delay = trajectory delay



Yukawa(1s)
200as 80eV XUV

R Pazourek, S Nagele, J Burgdörfer, Faraday Disc. (2013)

Photoionization: the problem of long-range interaction



$$t^{\mathrm{coul}}(k,r) = \frac{\partial}{\partial E}\left(\sigma_\ell^c(k) + \frac{Z}{k}\ln(2kr)\right)$$

$$= t^\ell_{\mathrm{EWS}}(k) + \frac{Z}{k^3}\left(1 - \ln(2kr)\right)$$

→ divergent

H(1s)
200as 80eV XUV

R Pazourek, S Nagele, J Burgdörfer., Faraday Discussions **163**, 353 (2013)
R Pazourek, S Nagele, J Burgdörfer, Rev.Mod.Phys. (submitted 2014)

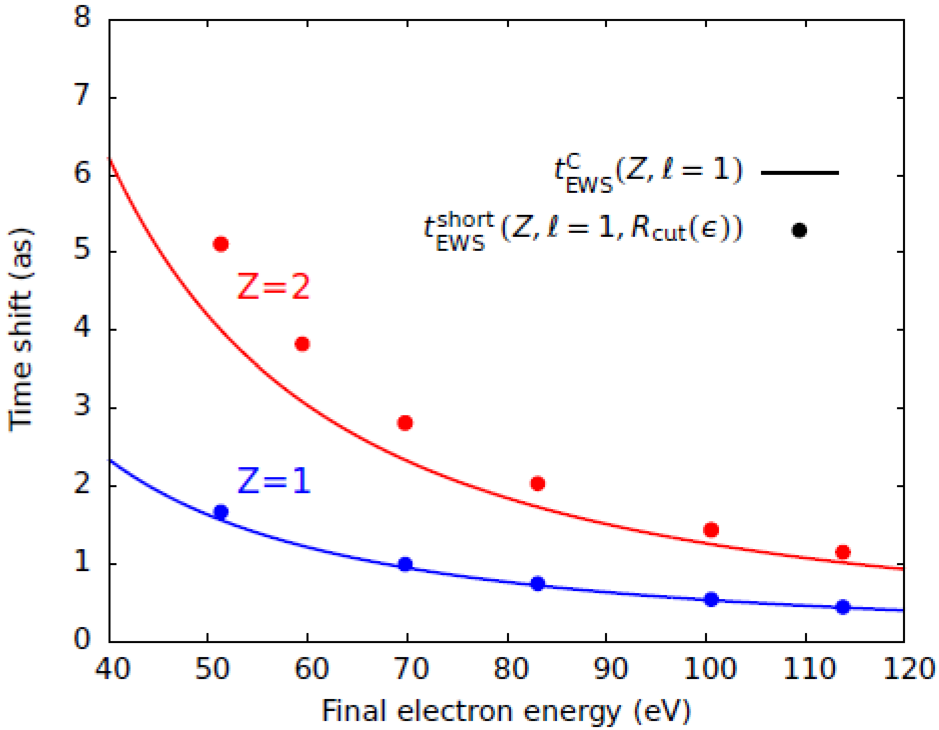
Separating long-range from short-range: Coulomb S-matrix and logarithmic phase distortion

$$V_c(r) = -\frac{Z}{r}\left[\theta(R_{\mathrm{cut}}-r)+\theta(r-R_{\mathrm{cut}})\right]$$

$$= V_{\mathrm{short}}(r)+V_{\mathrm{asym}}(r)$$

$$t^{\mathrm{short}}_{\mathrm{EWS}}(\epsilon) = t^{\mathrm{C}}_{\mathrm{EWS}} + \frac{Z}{(2\epsilon)^{3/2}}\left[1-\ln\left(2\sqrt{2\epsilon}R_{\mathrm{cut}}\right)\right]$$

$$+\mathcal{O}\left[(Z/\epsilon^{1/2})^2\right]$$



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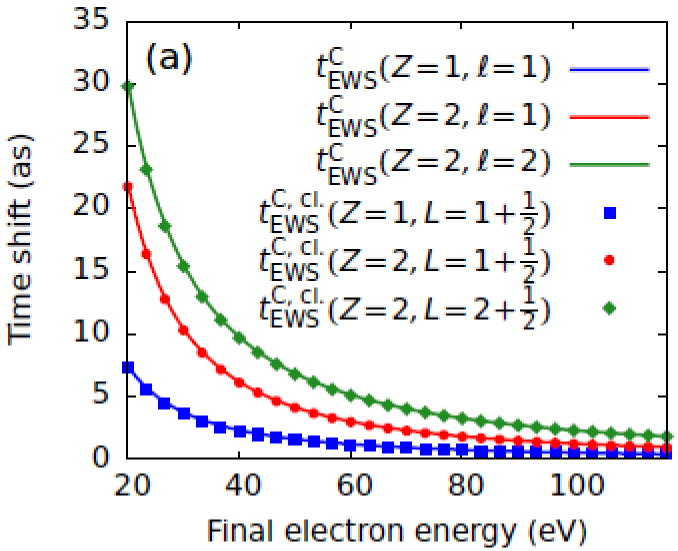
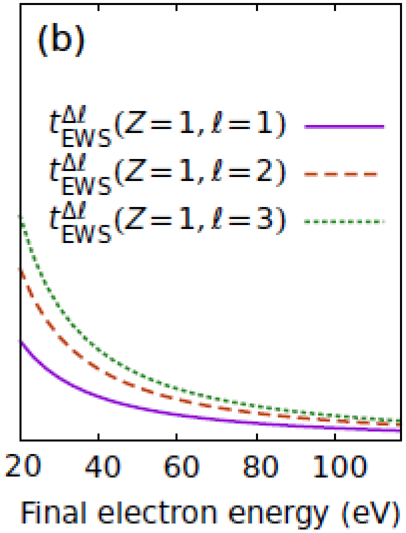
Coulomb EWS delay: quantum vs classical and centrifugal potential

$$t^{\mathrm{C}}_{\mathrm{EWS}}(\epsilon,\ell) = \frac{Z}{(2\epsilon)^{3/2}}\,\mathrm{Re}\left[\Psi(1+\ell-i\eta)\right]$$

$$t^{\mathrm{C}}_{\mathrm{EWS}}(\epsilon,\ell) \overset{|x|\gg 1}{=} \frac{Z}{(2\epsilon)^{3/2}}\ln\left(\sqrt{(1+\ell)^2+\eta^2}\right)$$

$$\simeq \frac{Z}{(2\epsilon)^{3/2}}\ln\left(\sqrt{L^2+\eta^2}\right) = t^{\mathrm{C,\,cl.}}_{\mathrm{EWS}}(\epsilon,L)$$





$$V(\ell,r) = \frac{\ell(\ell+1)}{2r^2}$$

$$t^{\Delta\ell}_{\mathrm{EWS}}(\epsilon,\ell) = t^{\mathrm{C}}_{\mathrm{EWS}}(\epsilon,\ell) - t^{\mathrm{C}}_{\mathrm{EWS}}(\epsilon,0)$$

$$= \sum_{j=1}^{\ell} \frac{1}{\sqrt{2\epsilon}}\frac{Zj}{2\epsilon Zj^2+Z^2}$$

R Pazourek, S Nagele, J Burgdörfer, Faraday Discuss. (2013)

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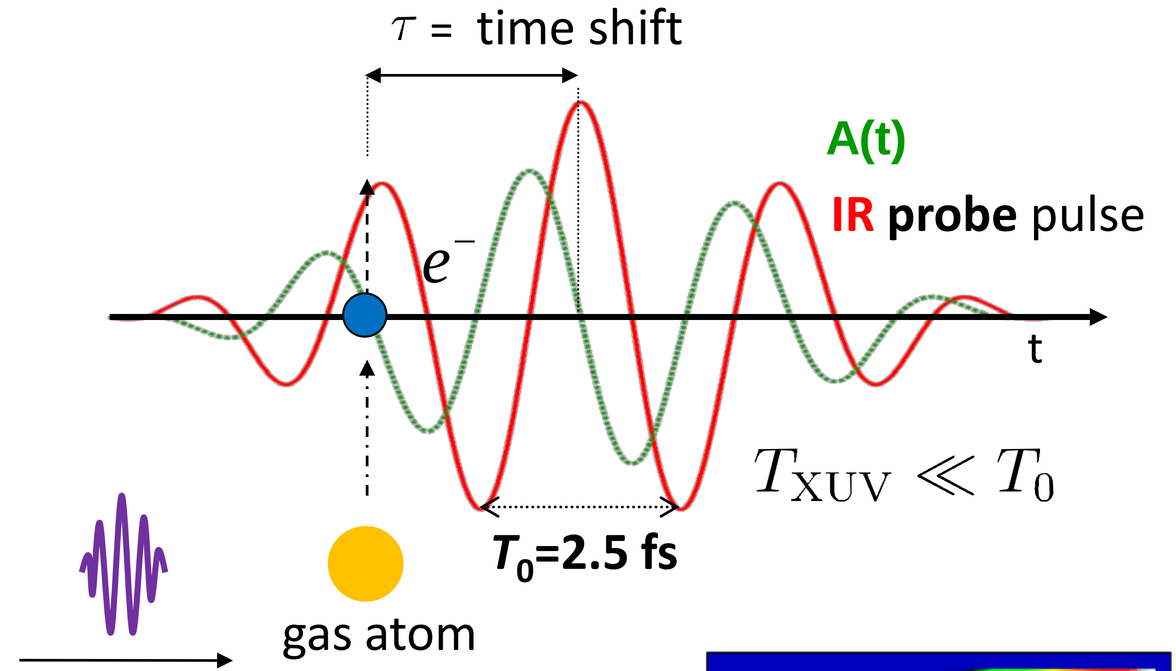
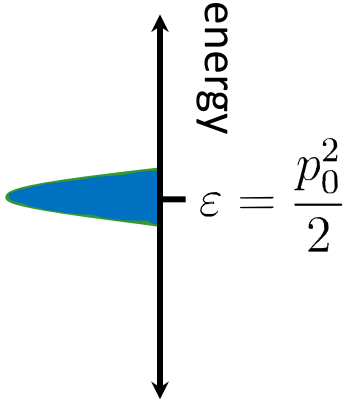
Attosecond streaking

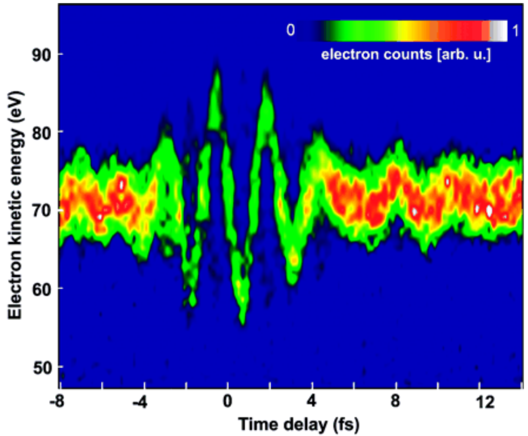


XUV pump pulse
~200 as



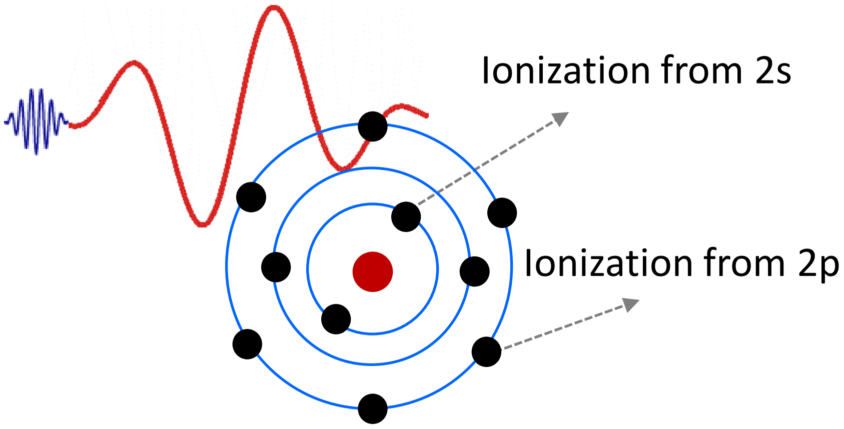
$$\vec{p}(\tau) \approx \vec{p}_0 - \vec{A}(\tau)$$

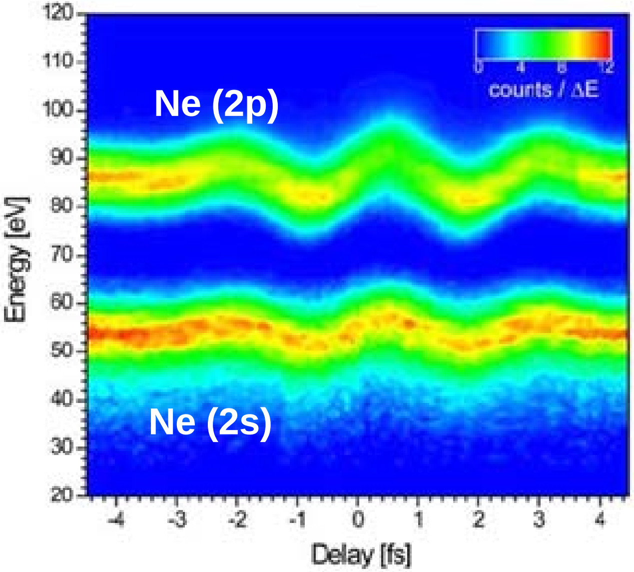
Mapping of (release) time to momentum



E. Goulielmakis *et al.*, *Science* **305**, 1267 (2004)

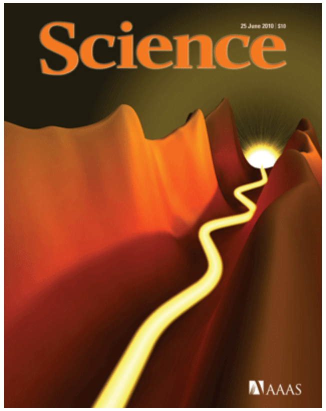
Streaking of Ne photoionization





$$p_0 - A(\tau + t_{\mathrm{S}}^{(2\mathrm{p})})$$

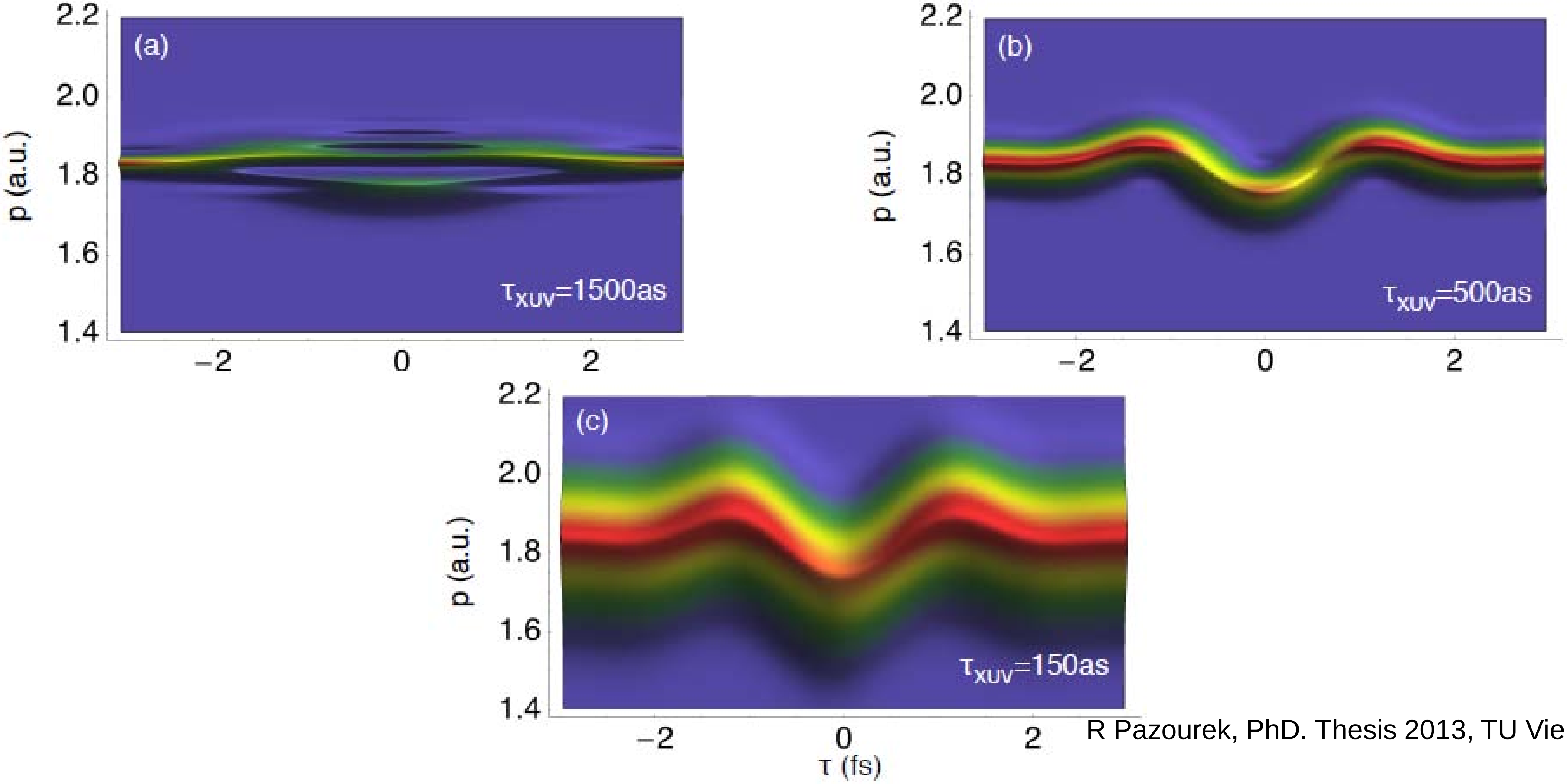
$$p_0 - A(\tau + t_{\mathrm{S}}^{(2\mathrm{s})})$$



$$t_{\mathrm{S}}^{(2\mathrm{p})} - t_{\mathrm{S}}^{(2\mathrm{s})} = 21\mathrm{as}$$

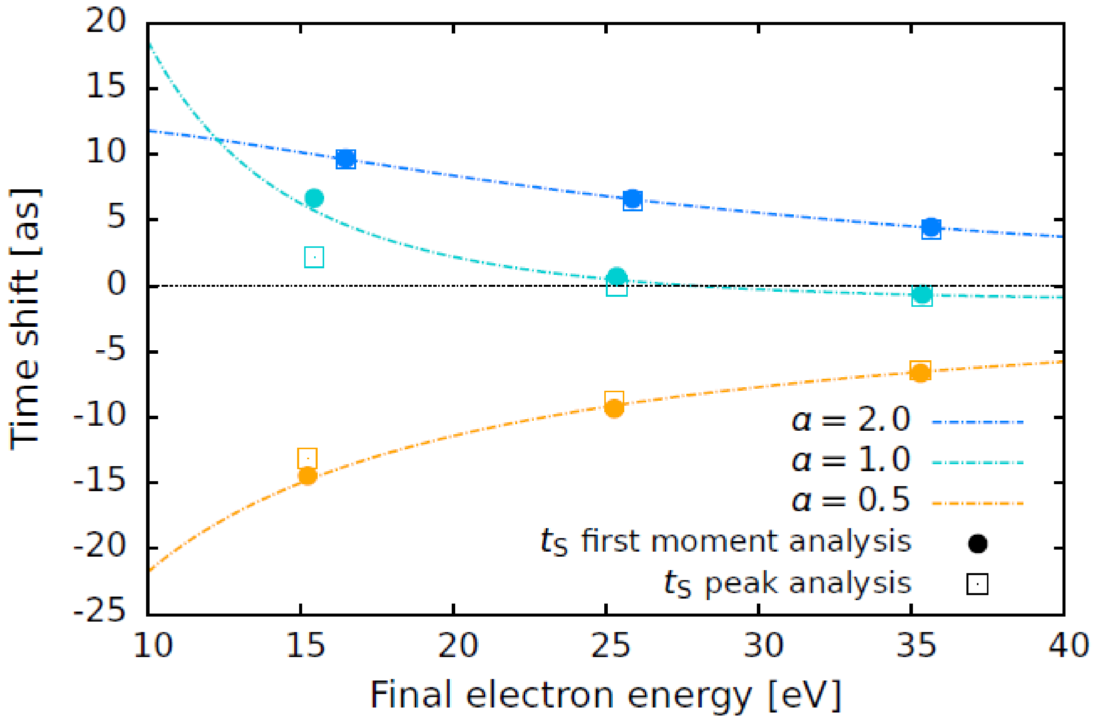
M. Schultze et al., Science **328**, 1658 (2010)

Streaking for different pulse durations: from quantum interferences to the classical clock



R Pazourek, PhD. Thesis 2013, TU Vienna

Streaking for photodetachment

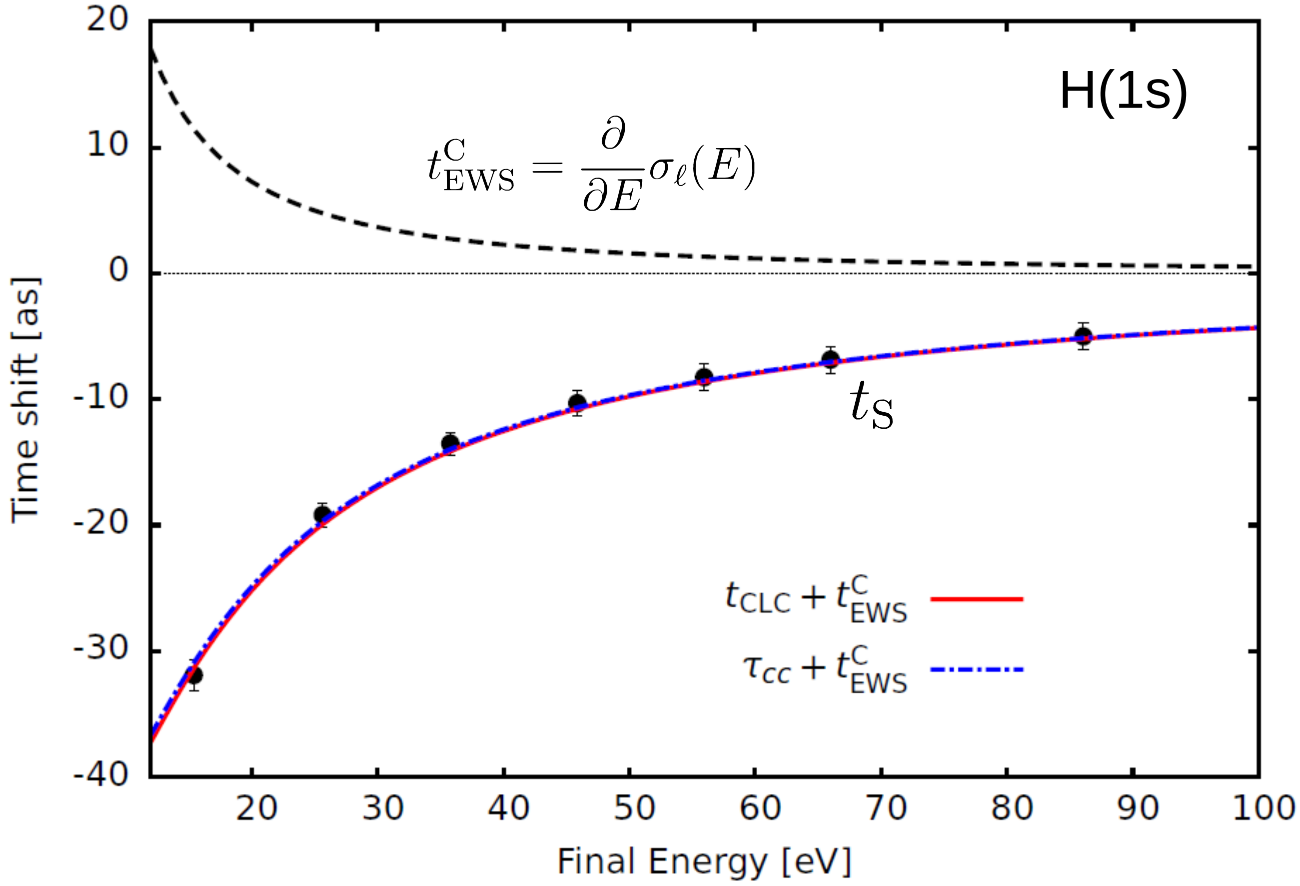


Yukawa
potential

$$\boxed{t_S = t_{EWS}}$$

Nagele et al., J. Phys. B **44 FT**, 081001 (2011)

Coulomb EWS time delay and Coulomb-laser-coupling (CLC)



$$\phi_C = \frac{Z}{k}\ln(2kr) + \sigma_\ell$$

$$t^{\mathrm{C}}_{\mathrm{EWS}}$$

$$t_{\mathrm{CLC}}(E, Z, \omega_{\mathrm{IR}}) \approx \frac{Z}{k^3}\left[2 - \ln(2E\pi/\omega_{\mathrm{IR}})\right]$$

$$\tau_{\mathrm{cc}} = t_{\mathrm{CLC}}$$

continuum-**c**ontinuum delay in **RABBIT**

$$t_{\mathrm{S}} = t_{\mathrm{EWS}} + t_{\mathrm{CLC}}$$

Time (phase) shifts due to interplay between streaking field and long-range interaction (classical)

$$\vec{p}(\tau) = \vec{p}_0 - \vec{A}(\tau + t_{EWS}) \; + \int_{\tau}^{\infty} dt \left[\vec{a}_{C,L}(\vec{F}, t) - \vec{a}_C(t) \right]$$

acceleration by laser and Coulomb field

acceleration by Coulomb field

$$\approx \vec{p}_0 - \vec{A}(\tau + t_{EWS}) + c(Z, \omega)\vec{F}(\tau)$$

$$\approx \vec{p}_0 - \vec{A}(\tau + t_{EWS} + t_{CLC})$$ → **IR field induced "time" (phase) shift**

→ Classical Coulomb-laser coupling (CLC) time shift

Nagele et al., J. Phys. B **44 FT**, 081001 (2011)
Pazourek et al., Faraday Discussions **163**, 353 (2013)

Additivity of long-range interaction corrections

$$\boxed{t_{\mathrm{S}} = t_{\mathrm{EWS}} + t_{\mathrm{CLC}} + t_{\mathrm{dLC}}}$$

Coulomb-laser coupling of outgoing electron

$$V_C \sim \frac{1}{r}$$

dipole-laser coupling

$$V_d \sim \frac{1}{r^2}$$

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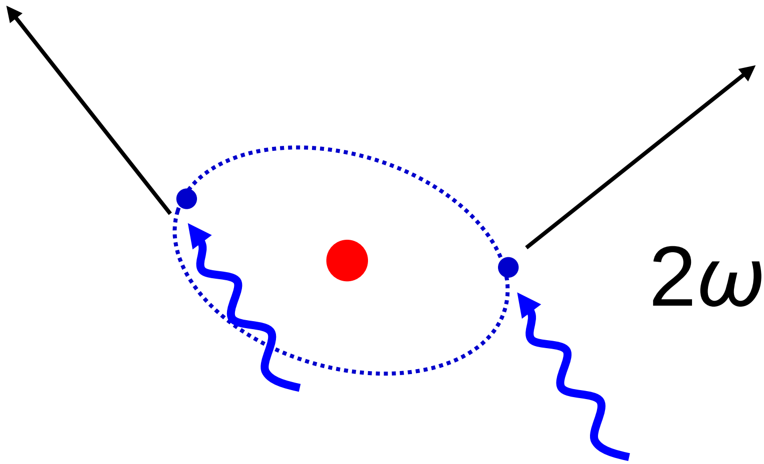
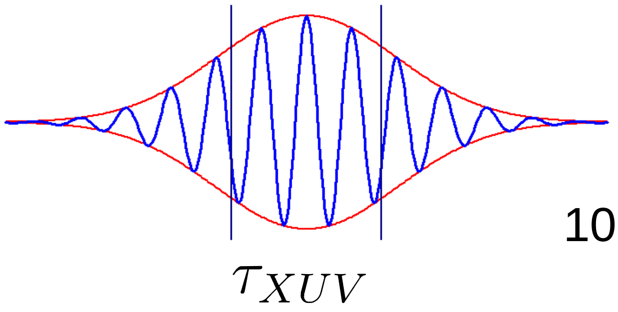
Two-photon double ionization (TPDI) of He: (non) sequential emission in time domain



$$2\omega_{XUV} + He = He^{2+} + e_1^- + e_2^-$$

$\omega_{XUV} \geq 40$ eV: “non-sequential”

$\omega_{XUV} > 54$ eV: “sequential”



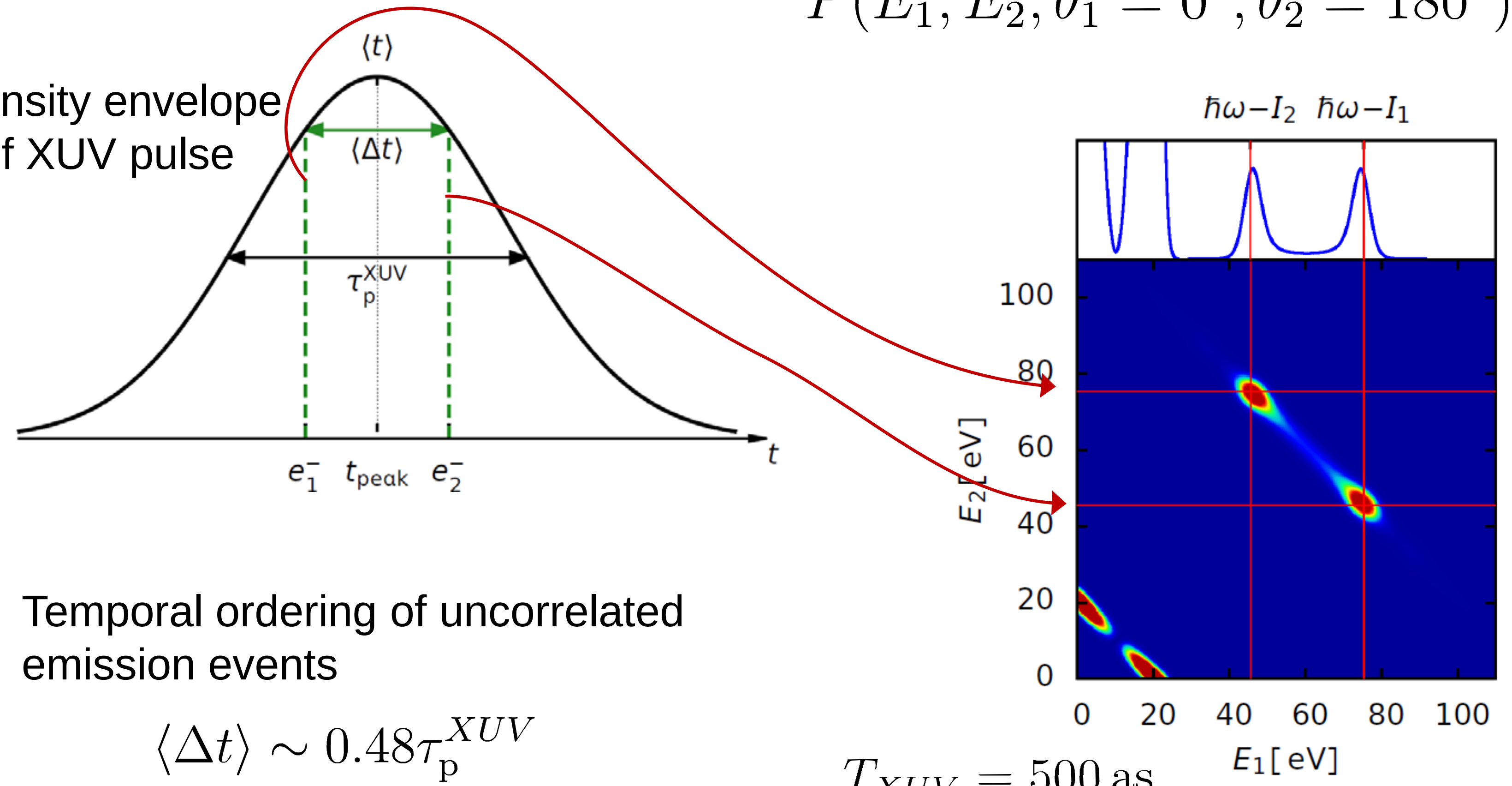
100as ≤ τ_{XUV} ≤ 1fs

R. Pazourek et al., arXiv **1405.1779** (2014)

TPDI: Timing information

$$P(E_1, E_2, \theta_1 = 0°, \theta_2 = 180°)$$

intensity envelope
of XUV pulse



Temporal ordering of uncorrelated
emission events

$$\langle \Delta t \rangle \sim 0.48 \tau_\mathrm{p}^{XUV}$$
$$\sim 240\, as$$

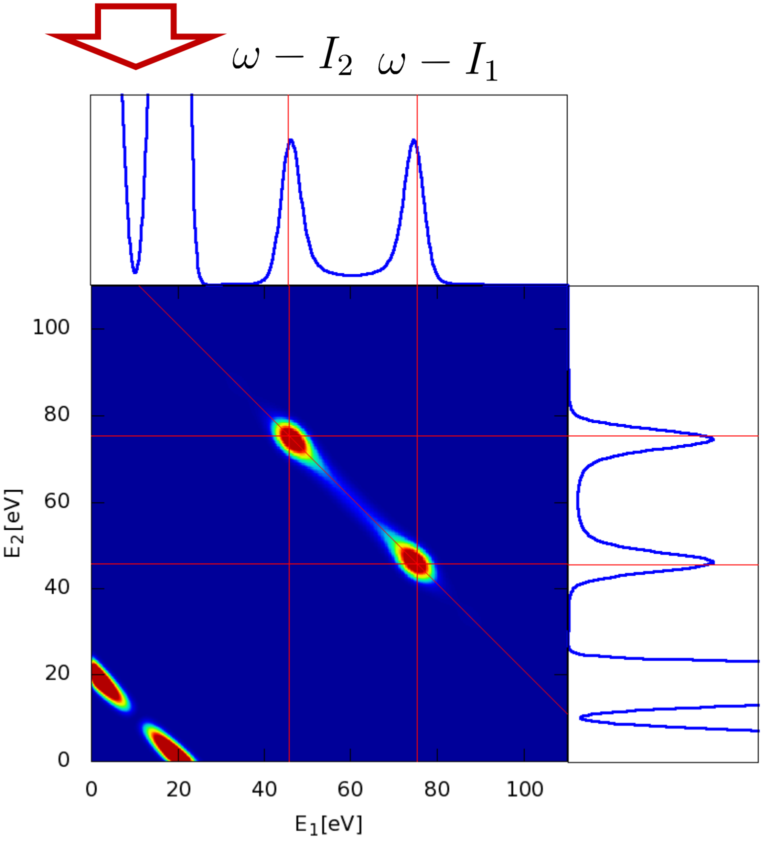
$$T_{XUV} = 500\,\mathrm{as}$$
$$\hbar\omega = 100\,\mathrm{eV}$$

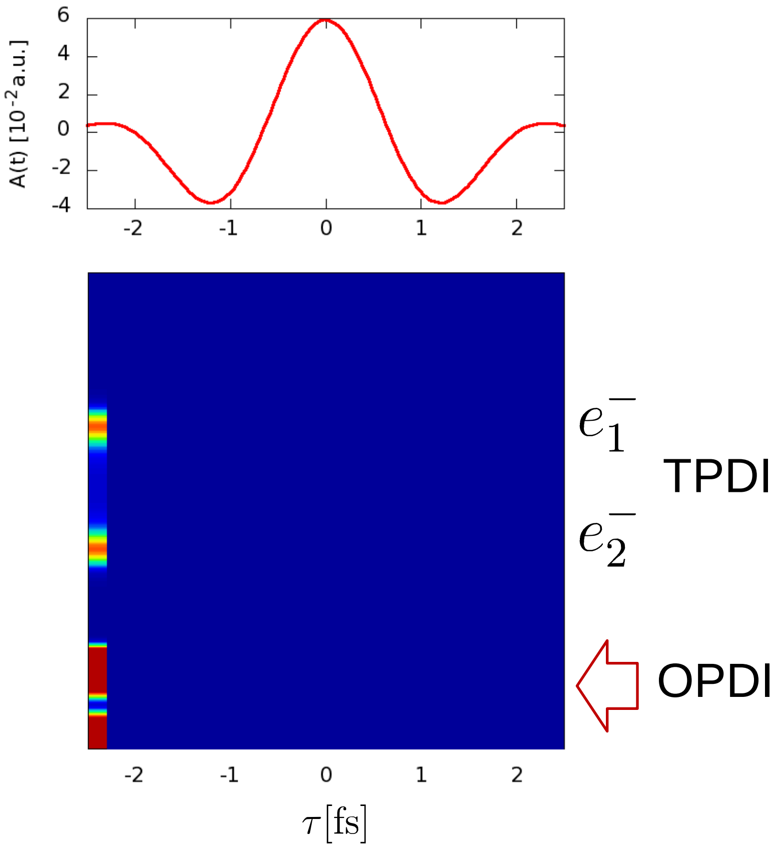
Streaking of two electrons in the continuum

One-photon double ionization **(OPDI)**

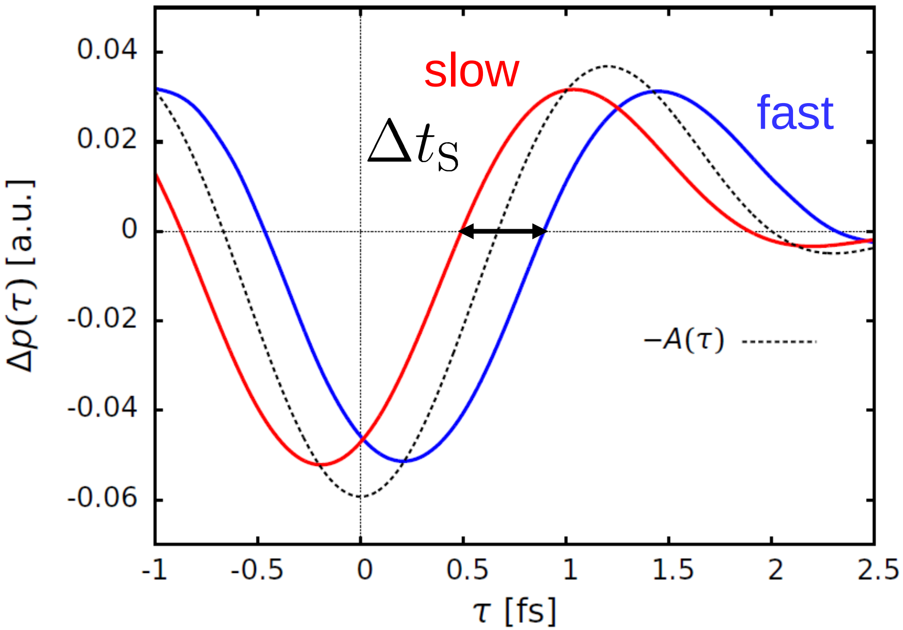
two-photon double ionization **(TPDI)**

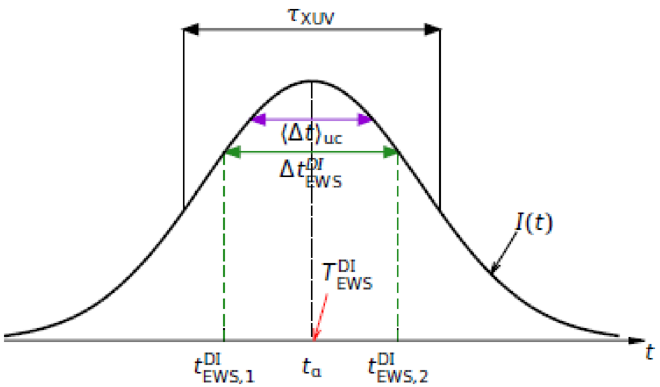
R. Pazourek et al., arXiv **1405.1779** (2014)





Streaking time shift for TPDI





$$\theta_1 = 0^\circ,\ \theta_2 = 180^\circ$$

“first“ electron: t_S= -211as

“second“ electron: t_S= 177as

$$\Delta t_S = 388\text{as} > \langle \Delta t \rangle \sim 240\text{as}$$

Time-ordering of the two photoabsorption events is measurable!

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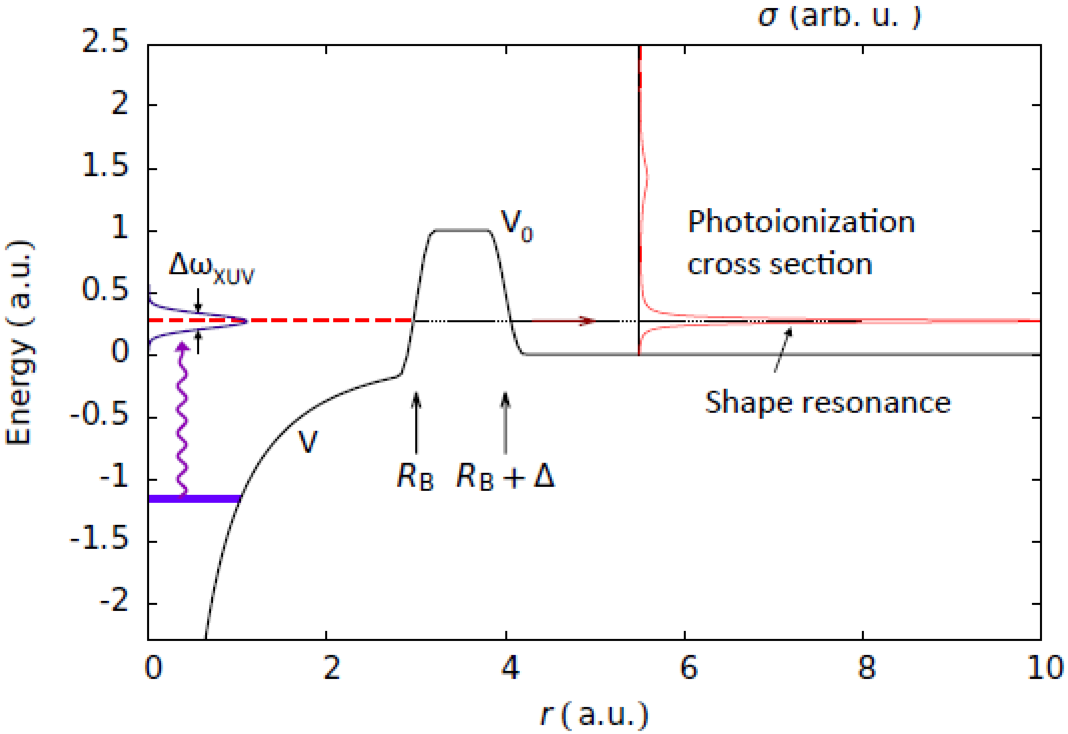
Attosecond streaking of tunneling time?

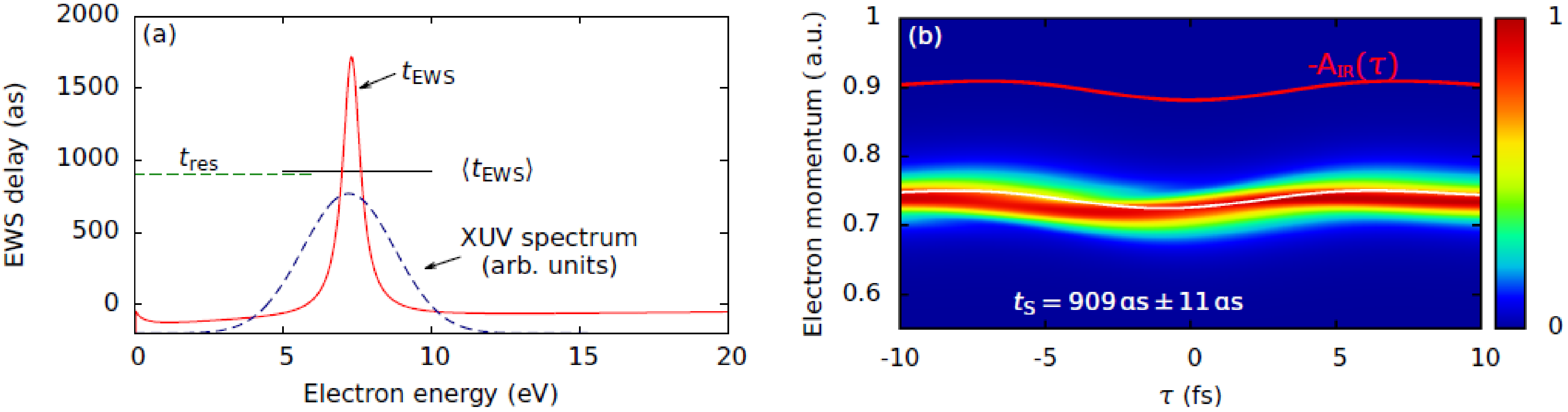


Excitation of shape-resonance:
thin-barrier regime

$$\tau_{\mathrm{XUV}} \ll t_{\mathrm{res}} \ll \tau_{\mathrm{IR}}$$

$$\omega_{\mathrm{IR}} \ll \Gamma \ll \Delta\omega_{\mathrm{XUV}}$$

Attosecond streaking of tunneling time?



$$t_{\rm S} \approx \langle t_{\rm EWS} \rangle \approx t_{\rm res}$$

→ dwell time, not tunelling time

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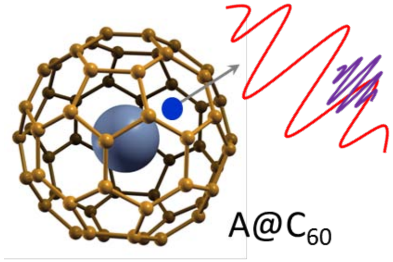
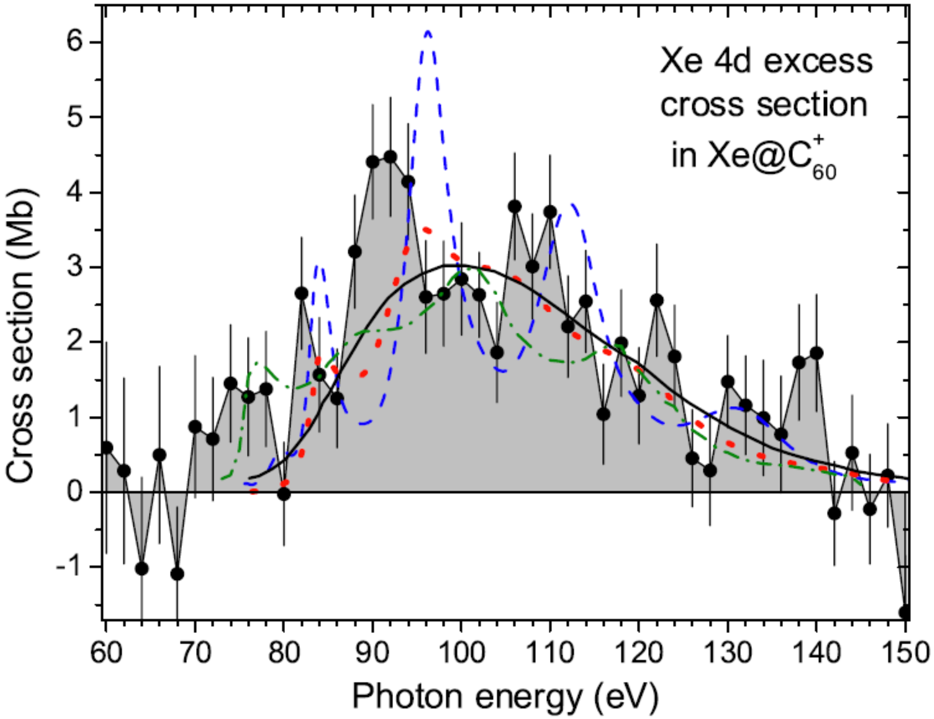
Ionization delays in endohedral fullerenes

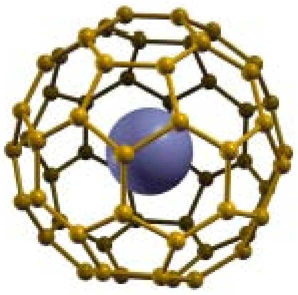


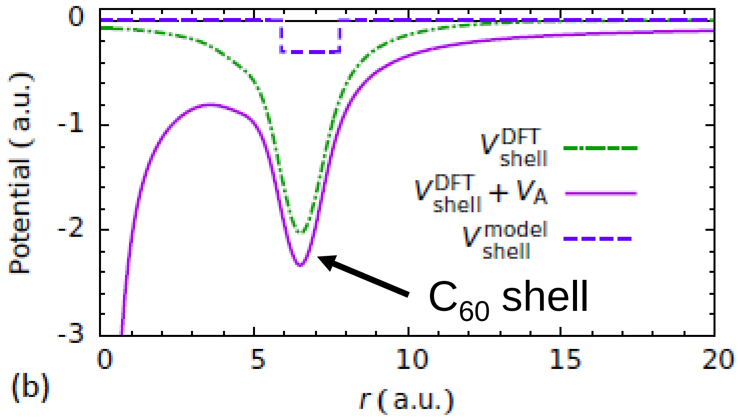
Photoionization of central atom
“confinement resonances“

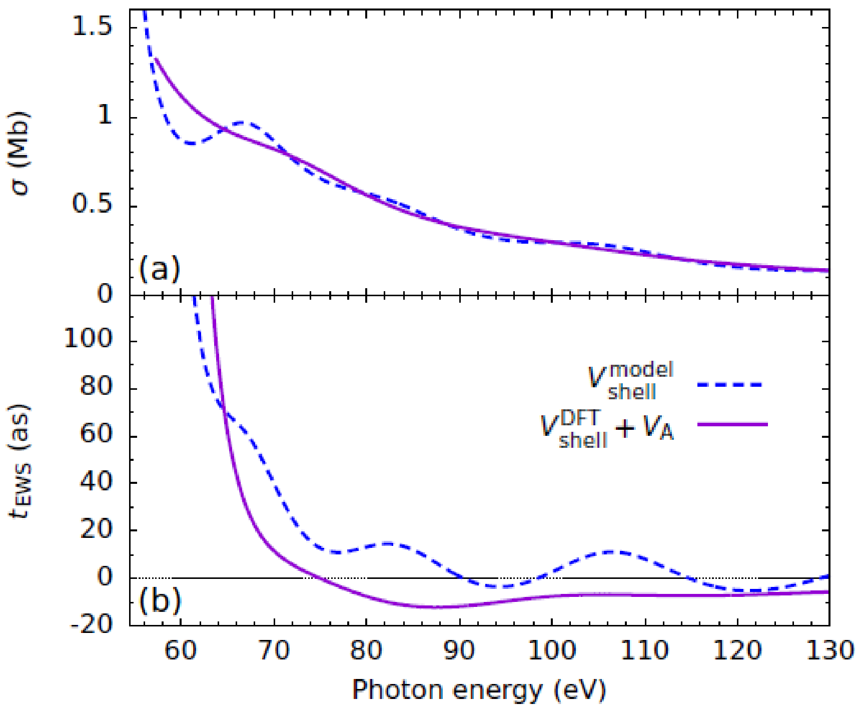


A. L. D. Kilcoyne et al, PRL **105**, 213001 (2010)

EWS delay in endohedral fullerenes





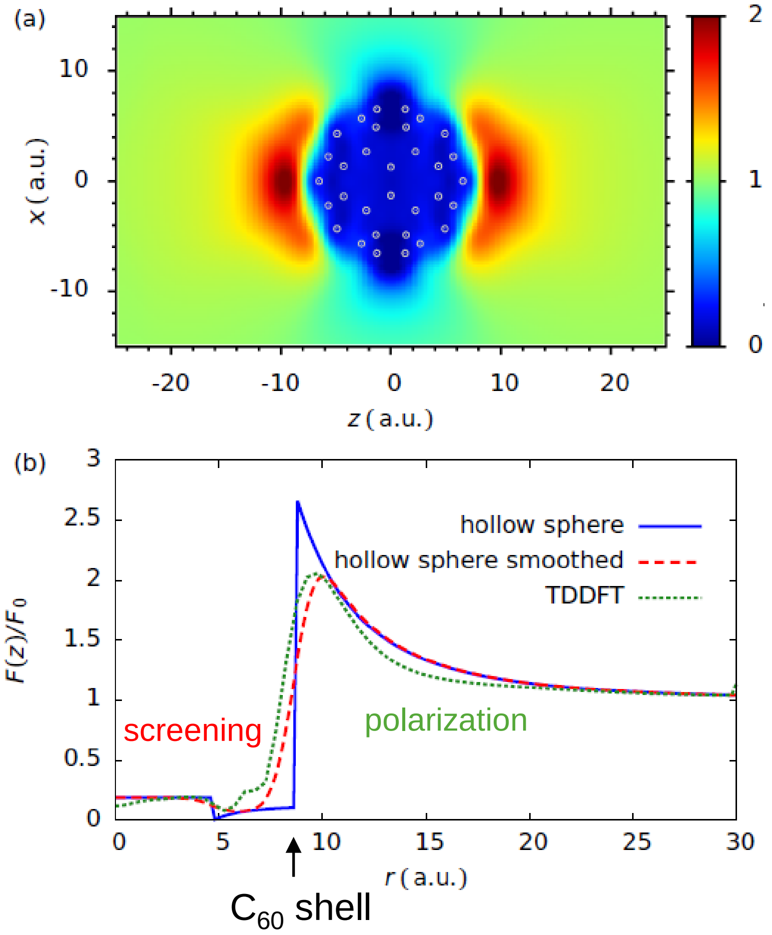


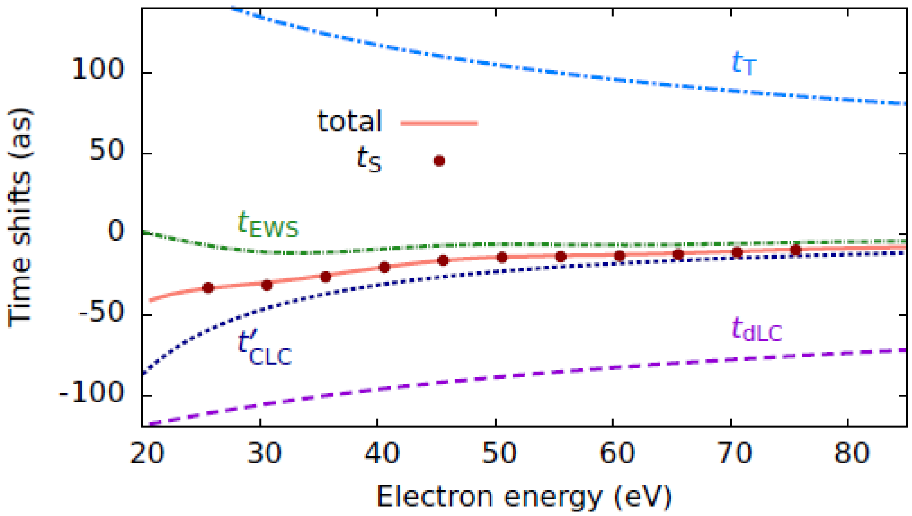
C_{60} potential
calculated by DFT

EWS delay

R Pazourek, S Nagele, J Burgdörfer., Faraday Discussions **163**, 353 (2013)
R Pazourek, S Nagele, J Burgdörfer, Rev.Mod.Phys. (submitted 2014)

Response to streaking field: screening and field enhancement





Streaking delay

S Nagele et al. (2014)

Summary

Attosecond chronoscopy:

- Quantum dynamics in the time domain
- Time delay of photoionization accessible
- Observation of coherent and incoherent processes on the attosecond scale
- Control and manipulation of dynamics in the time domain

Collaborators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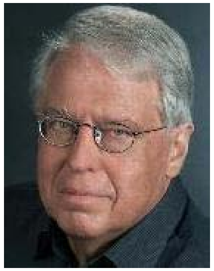
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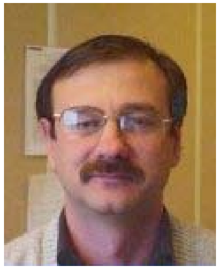
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