ATTOSECOND PHYSICS

UNIT XII RABBIT

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RABBIT

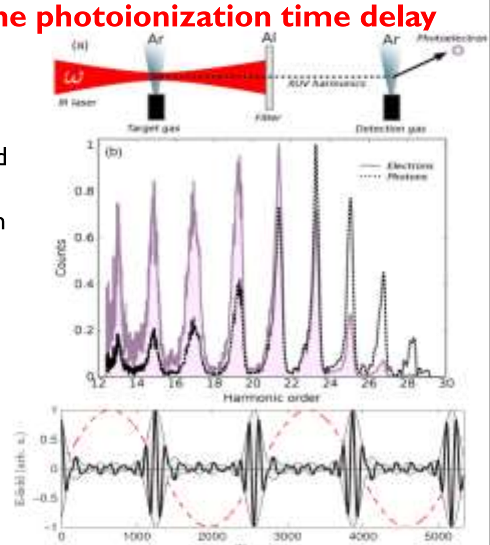
Reconstruction of Attosecond harmonic Beating By Interference of Two-photon transitions

Goal: To determine the photoionization time delay

A typical HHG experiment.

(a) A fraction of the IR laser field is converted to XUV through HHG. The XUV field is then filtered out and used to photoionize the detection gas.

(b) A representative photoelectron spectra (full) and XUV photon spectra (dotted) from an HHG experiment using Ar atoms and Al filter.



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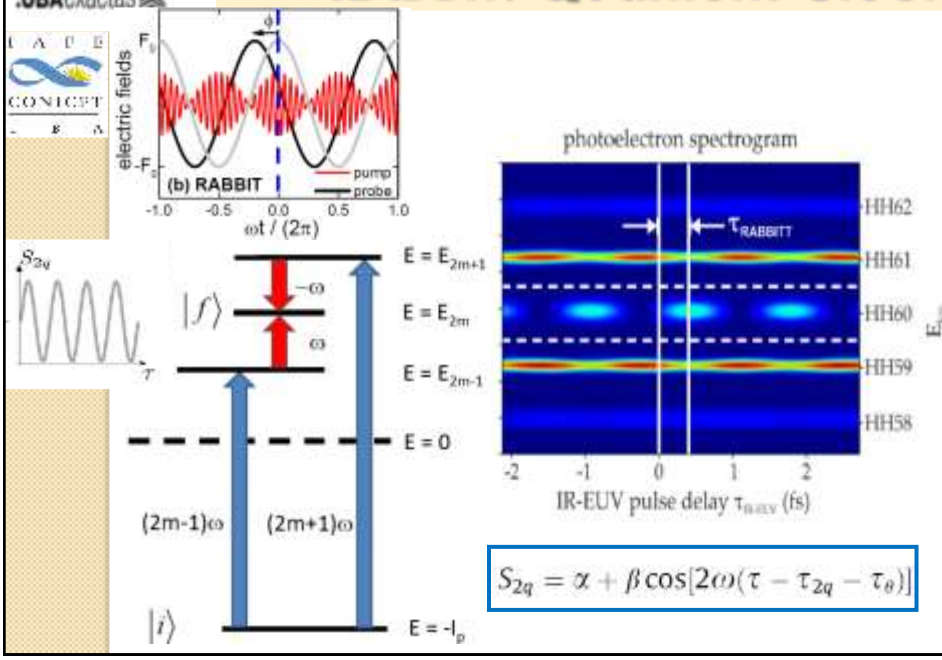
RABBIT: Quantum Clock



$$S_{2q} = \alpha + \beta \cos[2\omega(\tau - \tau_{2q} - \tau_\theta)]$$

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ABSORPTION OF A HH PHOTON

$$H = \frac{p^2}{2} + V(r) + zF(t) \qquad \mathbf{F}(t) = \mathbf{F}_X(t) + \mathbf{F}_L(t)$$

1st order perturbation theory: Absorption of one harmonic photon

$$\left|\Psi(t)\right\rangle = \sum_\alpha a_\alpha(t)\left|\alpha(t)\right\rangle + \int d\vec{k}\, b(\vec{k},t)\left|\psi_{\vec{k}}(t)\right\rangle$$

$$a_\alpha(t) = -i\int_0^t dt' F_X(t')\left\langle \alpha \right| z \left| \varphi_i \right\rangle e^{i(E_\alpha - E_i)t'}$$

$$a_\alpha(t) = -i\left\langle \alpha \right| z \left| \varphi_i \right\rangle \int_0^t dt' F_X(t') e^{i(E_\alpha - E_i)t'}$$

$$a_\alpha(t) = -iz_{\alpha i}\tilde{F}_X(t, E_\alpha - E_i)$$

In the same way:

$$b(\vec{k},t) = -iz_{\vec{k}i}\tilde{F}_X(t, E_{\vec{k}} - E_i)$$

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ABSORPTION OF A HH PHOTON

$$F_X(t) = F_{X0}\cos\omega_X t = \frac{F_X}{2}\left(e^{i\omega_X t} + e^{-i\omega_X t}\right)$$

$$\Rightarrow \tilde{F}_X(t, E_{\vec{k}} - E_i) = \frac{F_X}{2}\left(\frac{1-e^{i\left(E_{\vec{k}}-E_i+\omega_X\right)t}}{E_{\vec{k}} - E_i + \omega_X} + \frac{1-e^{i\left(E_{\vec{k}}-E_i-\omega_X\right)t}}{E_{\vec{k}} - E_i - \omega_X}\right)$$

We neglect the first term: emission of an ω_X photon

$$\tilde{F}_X(t, E_{\vec{k}} - E_i) \simeq \frac{F_X}{2}\left(\frac{1-e^{i\left(E_{\vec{k}}-E_i-\omega_X\right)t}}{E_{\vec{k}} - E_i - \omega_X}\right)$$

$$\tilde{F}_X(t, E_{\vec{k}} - E_i) \simeq \frac{F_X}{2}e^{-i\left(E_{\vec{k}}-E_i+\omega_X\right)t/2}\left(\frac{e^{-i\left(E_{\vec{k}}-E_i+\omega_X\right)t/2} - e^{i\left(E_{\vec{k}}-E_i+\omega_X\right)t/2}}{E_{\vec{k}} - E_i + \omega_X}\right)$$

$$= -iF_X e^{-i\left(E_{\vec{k}}-E_i+\omega_X\right)t/2}\left(\frac{\sin\left(E_{\vec{k}} - E_i + \omega_X\right)t/2}{\left(E_{\vec{k}} - E_i + \omega_X\right)/2}\right)$$

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ABSORPTION OF A HH PHOTON

$$\left|\Psi(t)\right\rangle = \sum_\alpha a_\alpha(t)\left|\alpha(t)\right\rangle + \int d\vec{k}\,b(\vec{k},t)\left|\psi_{\vec{k}}(t)\right\rangle$$

$$b(\vec{k},t) = -iz_{\vec{k}i}\tilde{F}_X(t, E_{\vec{k}} - E_i)$$

$$= -z_{\vec{k}i}F_X e^{-i\left(E_{\vec{k}}-E_i+\omega_X\right)t/2}\left(\frac{\sin\left(E_{\vec{k}} - E_i + \omega_X\right)t/2}{\left(E_{\vec{k}} - E_i + \omega_X\right)/2}\right)$$

$$\left|b(\vec{k},t)\right|^2 = F_X^2\left|z_{\vec{k}i}\right|^2\left|\tilde{F}_X(t, E_{\vec{k}} - E_i)\right|^2$$

$$= F_X^2\left|z_{\vec{k}i}\right|^2\left(\frac{\sin\left(E_{\vec{k}} - E_i + \omega_X\right)t/2}{\left(E_{\vec{k}} - E_i + \omega_X\right)/2}\right)^2$$

$$\Rightarrow \lim_{t\to\infty}\left|b(\vec{k},t)\right|^2 = \pi t F_X^2\left|z_{\vec{k}i}\right|^2\delta\left(E_{\vec{k}} - E_i + \omega_X\right)$$ Fermi's Golden Rule

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ABSORPTION OF A HH PHOTON

Instead, we turn on the ionizing field adiabatically:

$$F_X(t) = (F_X/2)\,e^{-i\omega_X t+\varepsilon t}$$

$$\tilde{F}_X(t, E_{\vec{k}} - E_i) = \lim_{\varepsilon\to 0^+} \frac{F_X}{2} \frac{e^{i(E_{\vec{k}}-E_i-\omega_X)t+\varepsilon t}}{E_{\vec{k}} - E_i - \omega_X - i\varepsilon}$$

$$= \frac{F_X}{2}\left(\wp \frac{1}{E_{\vec{k}} - E_i - \omega_X} + i\pi\delta\left(E_{\vec{k}} - E_i - \omega_X\right)\right) e^{i(E_{\vec{k}}-E_i-\omega_X)t}$$

$$\left|\Psi^{(1)}(t)\right\rangle = i\sum_\alpha z_{\alpha i}\tilde{F}_X(t, E_\alpha - E_i)\left|\alpha(t)\right\rangle + i\int d\vec{k} z_{\vec{k}i}\tilde{F}_X(t, E_k - E_i)\left|\psi_{\vec{k}}(t)\right\rangle$$

$$= i\sum_\alpha z_{\alpha i}\tilde{F}_X(t, E_\alpha - E_i)\left|\alpha(t)\right\rangle + i\int d\vec{k} z_{\vec{k}i}\tilde{F}_X(t, E_k - E_i)\left|\psi_{\vec{k}}(t)\right\rangle$$

$$= i\frac{F_X}{2}\left[\pi z_{\vec{k}i}\left|\psi_{\vec{k}}\right\rangle + \int d\vec{k} z_{\vec{k}i}\wp\frac{1}{E_{\vec{k}} - E_i - \omega_X}\left|\psi_{\vec{k}}\right\rangle + \sum_\alpha z_{\alpha i}\wp\frac{1}{E_\alpha - E_i - \omega_X}\left|\alpha\right\rangle\right] e^{-i(E_i+\omega_X)t}$$

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Long-range potential: quantum

For the case of a hydrogenic atom, the quantum Coulomb scattering wave is:

$$\begin{cases} \Phi_f(t) = \Phi_{\vec{k}}(\vec{r}, t) = e^{-i\varepsilon t}\dfrac{e^{i\vec{k}.\vec{r}}}{(2\pi)^{3/2}} D_c(Z, \vec{k}, t) \\ D_c(Z, \vec{k}, t) = N_T(k)\,{}_1F_1\left(-i\frac{Z}{k}, 1, -ikr - i\vec{k}.\vec{r}\right) \\ N_T(k) = e^{\frac{\pi Z}{2k}}\Gamma(1 + i\frac{Z}{k}); \quad Z \text{ is the ion charge} \end{cases}$$

The asymptotic behavior of the final state is

$$F_\ell(Z,k) \to \sin\left(kr + \frac{Z}{k}\ln(2kr) - \frac{\ell\pi}{2} + \sigma_\ell(k)\right)$$

The asymptotic behavior of the outgoing Coulomb wave is given by

$$\left|\Psi^{(1)}\right\rangle \sim e^{i\left(kr + \frac{Z}{k}\ln(2kr) - \frac{l\pi}{2} + \sigma_l(k)\right)}$$

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Long-range potential: quantum

$$\left|\Psi^{(1)}(t)\right\rangle \sim e^{i\phi_{\omega_X}+if(\omega_X,r,t)-i(\omega_X-E_i)t}$$

$$f(\omega_X,r,t)=\left(kr+\frac{Z}{k}\ln(2kr)-\frac{l\pi}{2}+\sigma_l(\kappa)\right)$$

We can use the SPA

$$\frac{\partial\phi_{\omega_X}}{\partial\omega_X}+\frac{\partial f(\omega_X,r,t)}{\partial\omega_X}-t=\frac{\partial\phi_{\omega_X}}{\partial\omega_X}+r\frac{\partial k}{\partial\omega_X}+\frac{Z}{k}\frac{\partial\ln(2kr)}{\partial\omega_X}+\frac{\partial\sigma_l(\kappa)}{\partial\omega_X}-t=0$$

$$\Rightarrow t=\tau_{GD}+\tau_{\text{free}}+\tau_{LR}+\tau_{EWS}$$ Arrival time at r

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EXCHANGE OF A PROBE PHOTON

$$H=\frac{p^2}{2}+V(r)+zF(t) \qquad \mathbf{F}(t)=\mathbf{F}_X(t)+\mathbf{F}_L(t)$$

2nd order perturbation theory:
Absorption/emission of one NIR photon

$$\left|\Psi^{(1)}(t)\right\rangle=\sum_\alpha a_\alpha^{(1)}(t)\left|\alpha(t)\right\rangle+\int d\vec{\kappa}\,b^{(1)}(\vec{\kappa},t)\left|\psi_{\vec{\kappa}}(t)\right\rangle$$

$$\left|\Psi^{(2)}(t)\right\rangle=\sum_\alpha a_\alpha^{(2)}(t)\left|\alpha(t)\right\rangle+\int d\vec{k}\,b^{(2)}(\vec{k},t)\left|\psi_{\vec{k}}(t)\right\rangle$$

$$b_f^{(2)}(t)=-i\int_{-\infty}^{t}dt'F_L(t')\left\langle f(t')\right|z\left|\Psi^{(1)}(t')\right\rangle$$

$$=-iF_L z_{f\Psi}\int_{-\infty}^{t}dt'e^{i\left(E_f\mp\omega_L-\omega_X-E_i\right)t'}$$

$$=-iz_{f\Psi}\tilde{F}_L(t,E_f\mp\omega_L-\omega_X-E_i)$$

$$\lim_{t\to\infty}b_f^{(2)}(t)=-iz_{f\Psi}F_L\,2\pi\delta\left(E_f\mp\omega_L-\omega_X-E_i\right)$$

EXCHANGE OF A PROBE PHOTON

We use (i) the random phase approximation and (ii) adiabatic switch on of the probe pulse

$$F_L(t) = (F_L / 2) e^{\mp i\omega_L t + \varepsilon t}$$

$$\tilde{F}_L(t, E_{\vec{k}} - E_i) = \lim_{\varepsilon \to 0^+} \frac{F_L}{2} \frac{e^{i\left(E_{\vec{k}} - E_i - \omega_X \mp \omega_L\right)t + \varepsilon t}}{E_{\vec{k}} - E_i - \omega_X \mp \omega_L - i\varepsilon}$$

$$= \frac{F_L}{2}\left(\wp \frac{1}{E_{\vec{k}} - E_i - \omega_X \mp \omega_L} + i\pi\delta\left(E_{\vec{k}} - E_i - \omega_X \mp \omega_L\right)\right) e^{i\left(E_{\vec{k}} - E_i - \omega_X \mp \omega_L\right)t}$$

$$z_{f\Psi} = \left\langle f \middle| z \middle| \Psi^{(1)} \right\rangle$$

$$= \sum_\alpha a_\alpha^{(1)}(t) \left\langle f | z | \alpha \right\rangle + \int d\vec{\kappa}\, b^{(1)}(\vec{\kappa}, t) \left\langle f | z | \psi_{\vec{\kappa}} \right\rangle$$

$$= i \frac{F_X}{2}\left[i\pi z_{\vec{k}\vec{\kappa}} z_{\vec{\kappa} i} + \int d\vec{\kappa}\, z_{\vec{k}\vec{\kappa}} z_{\vec{\kappa} i} \wp \frac{1}{E_{\vec{\kappa}} - E_i - \omega_X} + \sum_\alpha z_{\vec{k}\alpha} z_{\alpha i} \wp \frac{1}{E_\alpha - E_i - \omega_X} \right]$$

EXCHANGE OF A PROBE PHOTON

Asymptotic approximation

$$z_{f\Psi} = \left\langle f \middle| \Psi^{(1)} \right\rangle$$

$$= \int_0^\infty dr \sin\left(kr + \frac{Z}{k}\ln(2kr) - \frac{l\pi}{2} + \sigma(k) \right) z e^{i\left(\kappa r + \frac{Z}{\kappa}\ln(2\kappa r) - \frac{l\pi}{2} + \sigma(\kappa)\right)}$$

$$\simeq \frac{1}{2i}\int_0^\infty drz e^{i\left((\kappa - k).r + \frac{Z}{\kappa}\ln(2\kappa r) - \frac{Z}{k}\ln(2kr) + \sigma(\kappa) - \sigma(\kappa)\right)}$$

$$= -\frac{e^{i(\sigma(\kappa) - \sigma(\kappa))}}{2i} \frac{(2\kappa)^{i/\kappa}}{(2k)^{i/k}} \left(\frac{i}{\kappa - k} \right)^{2 + i\xi} \Gamma(2 + i\xi)$$

$$\xi = \frac{1}{\kappa} - \frac{1}{k}$$

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EXCHANGE OF A PROBE PHOTON

$$T_{if}^{(2)} = -iz_{f\Psi} F_L 2\pi\delta\left(E_f \mp \omega_L - \omega_X - E_i\right)$$

$$\propto \underset{(A)}{F_L F_X} \underbrace{e^{i(\sigma(\kappa)-\sigma(k))}}_{(B)} \underbrace{\frac{(2\kappa)^{i/\kappa}}{(2k)^{i/k}} \left(\frac{i}{\kappa - k}\right)^{2+i\xi} \Gamma(2+i\xi)}_{(C)}$$

I dropped the delta function in the last expression for simplicity

(A) $$F_L \equiv |F_L| e^{i\varphi_1} \quad ; \quad F_X \equiv |F_q| e^{\pm i\varphi_q}$$

(B) Difference in scattering phase between the intermidiate state and the final state. Related to the Wigner delay of the electron.

(C) Contains an additional phase: continuum-continuum phase.

$$\phi_{cc} = \arg\left[\frac{(2\kappa)^{i/\kappa}}{(2k)^{i/k}} \left(\frac{i}{\kappa - k}\right)^{2+i\xi} \Gamma(2+i\xi)\right]; \quad \xi = \frac{1}{\kappa} - \frac{1}{k}$$

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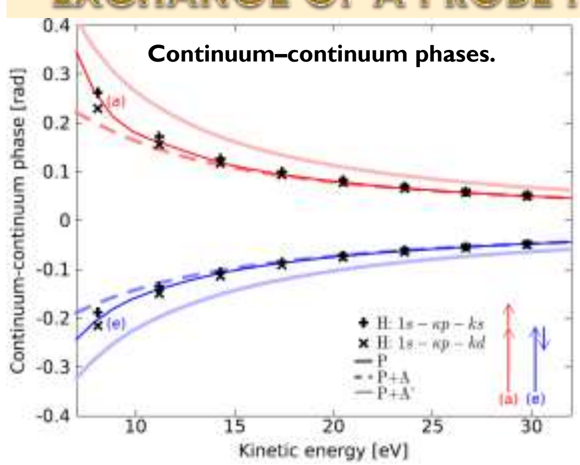
EXCHANGE OF A PROBE PHOTON



ATI phaseshifts for absorption (a) of one 800 nm laser photon and for stimulated emission (e) of one laser photon, in a Coulomb potential with Z = 1. The asymptotic approximation (P) provides the correct qualitative behavior, while the long-range amplitude corrections (P + A and P + A') lead to quantitative agreement, at high enough energy, with the exact calculations in hydrogen (+ and ×).

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EXCHANGE OF A PROBE PHOTON

$$\frac{\kappa^2}{2} \pm \omega = \frac{k^2}{2}$$

+ corresponds to the absorption
– to the emission of a probe photon

$$\left|T_{if}^{(2)}\right|^2 = \left|T_{if}^{(2,+)} + T_{if}^{(2,-)}\right|^2$$

$$= \left|T_{if}^{(2,+)}\right|^2 + \left|T_{if}^{(2,-)}\right|^2 +$$

$$+2\left|T_{if}^{(2,+)}\right|\left|T_{if}^{(2,-)}\right| \cos\left[2\varphi_1 - (\varphi_{q+2} - \varphi_q) - (\sigma(\kappa^+) - \sigma(\kappa^-)) - (\phi_{cc}^- - \phi_{cc}^+)\right]$$

$\varphi_1 = \omega\tau$ — IR delay

$\varphi_{q+2} - \varphi_q \approx 2\omega\tau_{GD}$ — GD of the attosecond pulses

$\sigma(\kappa^+) - \sigma(\kappa^-) \approx 2\omega\tau_W$ — EWS delay

$\phi_{cc}^- - \phi_{cc}^+ \approx 2\omega\tau_{cc}$ — CC delay

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EXCHANGE OF A PROBE PHOTON

$$\left|T_{if}^{(2)}\right|^2 = \left|T_{if}^{(2,+)}\right|^2 + \left|T_{if}^{(2,-)}\right|^2 +$$

$$+2\left|T_{if}^{(2,+)}\right|\left|T_{if}^{(2,-)}\right| \cos\left[2\omega\left(\tau - \tau_{GD} - \tau_W - \tau_{cc}\right)\right]$$

Therefore, the maximal probability for photoemission occurs when the amplitudes associated with paths (a) and (e) are in phase, arg[T_+] = arg[T]:

$$\tau = \tau_{GD} + \tau_W + \tau_{cc}$$

- τ_{GD}: the group delay of the XUV field is *when* the attosecond pulse arrived at the target, i.e. when the XUV and NIR fields added constructively at the atom.

- τ_W: the Wigner delay is the 'delay' in single-photon ionization, i.e. an asymptotic temporal shift of the photoelectron wave packet.

- τ_{cc}: the continuum–continuum delay, i.e. a *measurement induced delay* due the electron being probed by an IR laser field in a long-range potential with a Coulomb tail of charge *Z*. This delay can be traced back to the phaseshifts of the ATI matrix elements.

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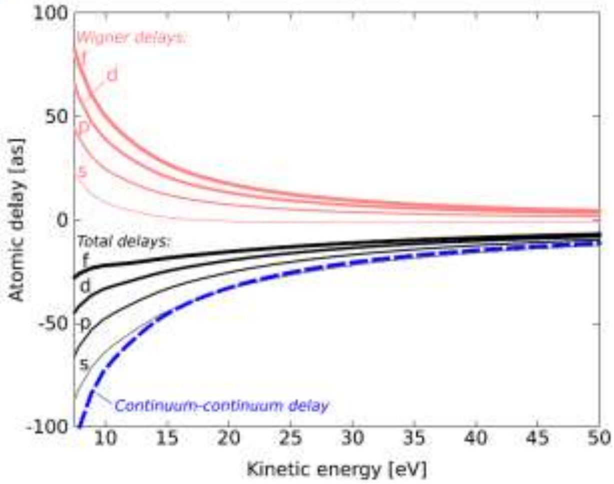
EXCHANGE OF A PROBE PHOTON



Atomic delays in laser-assisted ionization. The Wigner delays for hydrogen with a photoelectron of s, p, d or f-character (red, upper curves) plus the universal continuum–continuum delay for the laser-probing process with $Z = 1$ and $\omega = 1.55$ eV (blue, dashed curve) yields the total delay in laser-assisted photoionization (black, lower curves).

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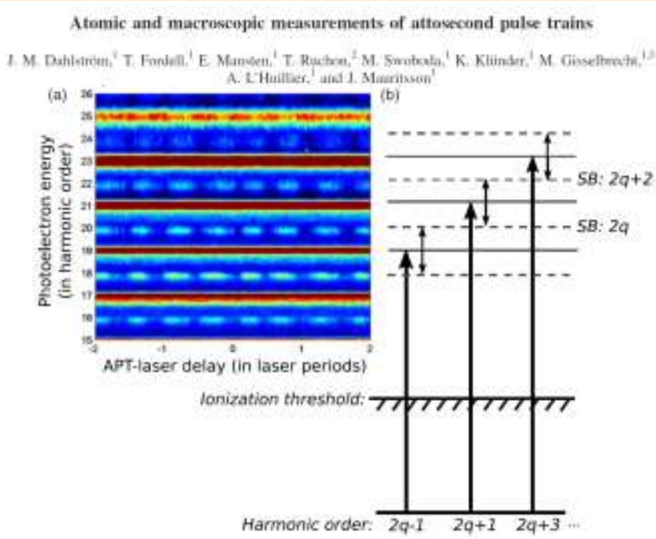
EXPERIMENTS OF RABBIT



Photoelectron spectrogram of Ar over photon energy and delay between the APT and the IR field. (b) Schematic energy diagram over the quantum paths leading to the same final energy in SB 2q.

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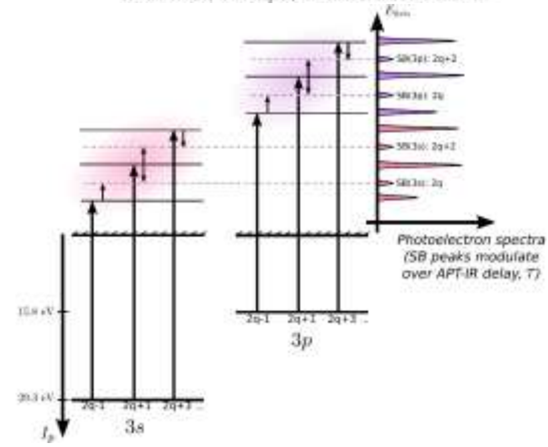
EXPERIMENTS OF RABBIT

PRL 106, 143002 (2011) PHYSICAL REVIEW LETTERS 8 APRIL 2011

Probing Single-Photon Ionization on the Attosecond Time Scale

K. Klünder,[1] J. M. Dahlström,[1] M. Gisselbrecht,[1] T. Fordell,[1] M. Swoboda,[1] D. Guénot,[1] P. Johnsson,[1] J. Caillat,[2] J. Mauritsson,[1] A. Maquet,[2] R. Taïeb,[2] and A. L'Huillier[1,*]



A delay of 110 as between the 3p and 3s states in argon has been measured experimentally by Klünder *et al* using an APT with a photon energy of 35 eV

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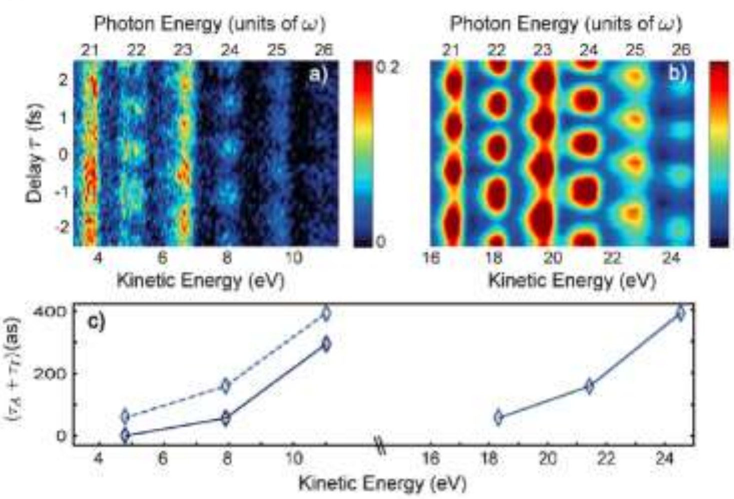

Energy spectra as a function of delay from electrons liberated from the 3s shell (a) and the 3p shell (b), respectively. (c) Retrieved delays corrected for the Cr group delay. Also shown are the 3p delays shifted down in energy for comparison with the 3s delays (dashed line).

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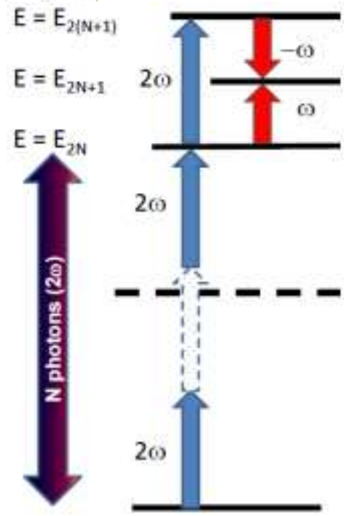
RABBIT-LIKE EXPERIMENTS

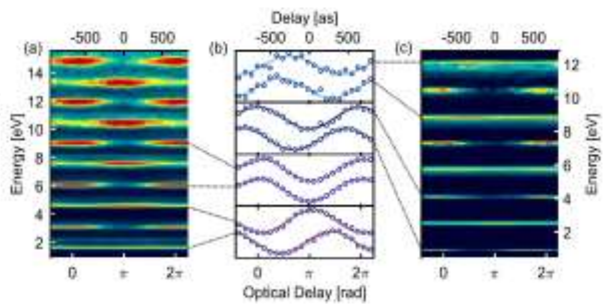
ω–2ω ionization in Ar



optica

Probing electron delays in above-threshold ionization

Lucas J. Zipp, Adi Natan, and Philip H. Bucksbaum



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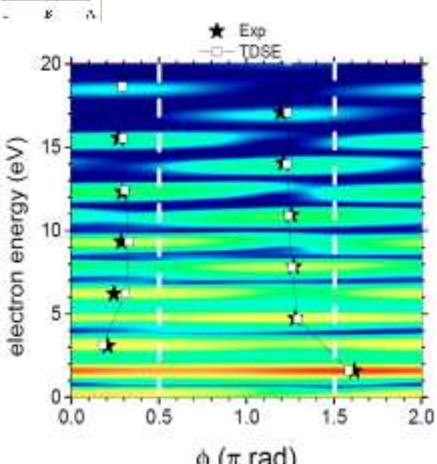
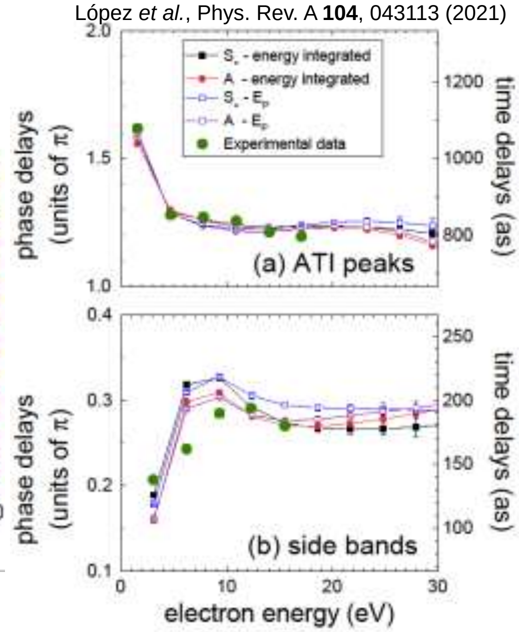
RABBIT-LIKE EXPERIMENTS

López *et al.*, Phys. Rev. A **104**, 043113 (2021)





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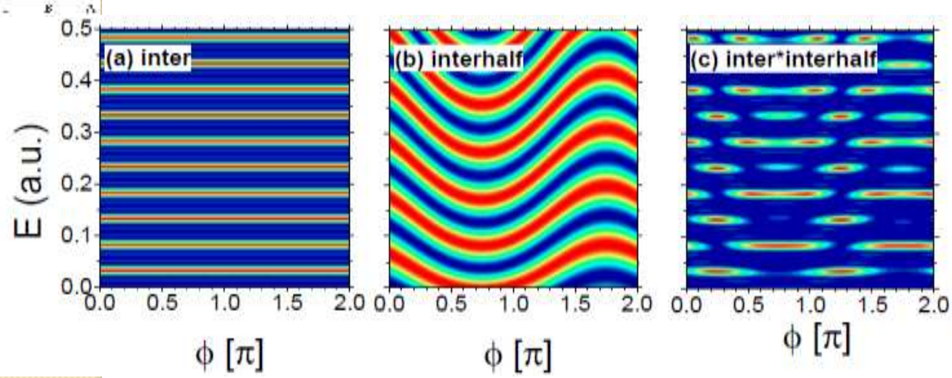
RABBIT-LIKE EXPERIMENTS

Origin of phase delays in ω–2ω



$$P(E, \cos\theta = 1) = c_0(E) + \sum_{i\geq 1} c_i(E)\cos\left[i\phi - \delta_i(E)\right]$$

D. G. Arbó, S. D. López, and J. Burgdörfer, Phys. Rev. A 106, 053101 (2022)

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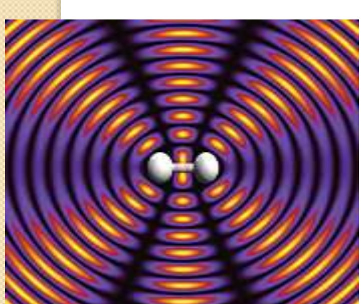
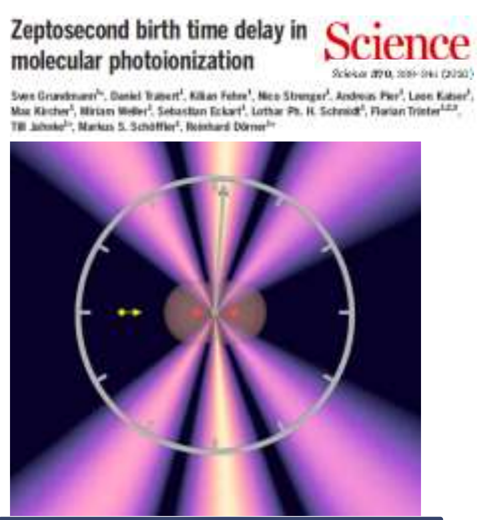
THE TINIEST TIME EVER MEASURED

Ionización de H_2: F. Martín (Madrid) Science **315**, 629 (2007).



Double photoionization of H_2 by circularly polarized light



$$\tau_b = 247 \text{ zs} = 2.47\times 10^{-19} \text{ s}$$

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