Time *and* Quantum Mechanics

One is able to image electronic motion in atoms and molecules and to determine photoionization delays with temporal resolution at the natural attosecond scale.

What does that infer regarding measurement of time in quantum mechanics?

Status of the time variable : operator or parameter ?

Alfred Maquet, LCPMR, UPMC.
KITP Santa Barbara, Aug. 2014
alfred.maquet@upmc.fr

Outline

A) Heisenberg vs. Pauli.

B) Mathematical background: Weyl and Stone-von Neumann.

C) Derivations of Heisenberg inequalities.

D) Tunneling times.

E) Wigner *et al.* time delay.

F) Delays in photoionization.

Papers to appear on related topics :

- Burgdörfer et al. RMP (2014);
- Caillat et al. JPB (Sept 2014);
- Dahlström et al. Springer book (2014).

An Outstanding Problem since the Foundations of Quantum Mechanics

Heisenberg's position-momentum uncertainty inequality: $\Delta q_i \, \Delta p_i \geq \hbar / 2$

⇔ commutation relation (in matrix form) for operators associated to the ***canonical variables (e.g. position-momentum)*** of classical mechanics.

$$[Q,P] = QP - PQ = i\hbar 1 \quad (1)$$

Given the known time-frequency uncertainty for wave-like phenomena (e.g. in Fourier analysis) why not defining a time-operator T ? Its commutator with the hamiltonian would be:

$$\Delta E \, \Delta t \geq \hbar / 2$$

$$[T,H] = TH - HT = i\hbar 1 \quad (2)$$

In fact, that was Heisenberg's idea…(in 1927)

Heisenberg (1927)

und q_1 in der Beziehung

$$p_1 q_1 \sim h. \tag{1}$$

Daß diese Beziehung (1) in direkter mathematischer Verbindung mit der Vertauschungsrelation $pq - qp = \frac{h}{2\pi i}$ steht, wird später gezeigt werden. Hier sei darauf hingewiesen, daß Gleichung (1) der präzise

der Phase das Atom zerstört bzw. verändert. In einem bestimmten stationären „Zustand" des Atoms sind die Phasen prinzipiell unbestimmt, was man als direkte Erläuterung der bekannten Gleichungen

$$Et - tE = \frac{h}{2\pi i} \quad \text{oder} \quad Jw - wJ = \frac{h}{2\pi i}$$

ansehen kann. (J = Wirkungsvariable, w = Winkelvariable.)

$$E_1 t_1 \sim h. \tag{2}$$

Diese Gleichung entspricht der Gleichung (1) und zeigt, wie eine genaue Energiebestimmung nur durch eine entsprechende Ungenauigkeit in der Zeit erreicht werden kann.

From Classical to Quantum Mechanics

Classical		Quantum
Hamilton function + canonical variables		Hamiltonian operator in a Hilbert space
$H(q_i, p_i)$	→	$H(q_i, -i\hbar \frac{\partial}{\partial q_i})$
Poisson Bracket		Commutator
$\{A,B\} = \sum_i (\frac{\partial A}{\partial q_i}\frac{\partial B}{\partial p_i} - \frac{\partial A}{\partial p_i}\frac{\partial B}{\partial q_i})$	→	$\frac{1}{i\hbar}[A,B]$
Equation of motion		Heisenberg's equation
$\frac{dF(q_i,p_i,t)}{dt} = \frac{\partial F}{\partial t} + \{F,H\}$	→	$\frac{dF(q_i,p_i,t)}{dt} = \frac{\partial F}{\partial t} + \frac{1}{i\hbar}[F,H]$
Time variable is a ***parameter***	→	***Time variable is also a parameter ??***

Pauli (1930)

No such time operator does exist!

Pauli's argument: Relations of type (1) are only valid for unbounded operators
As H is semi-bounded, eq. (2) cannot hold:

[2] In the older literature on quantum mechanics, we often find the operator equation

$$H\,t - t\,H = \frac{h}{i} I,$$

which arises from (8.6) formally by substituting t for F. It is generally not possible, however, to construct a Hermitian operator (e.g. as function of p and q) which satisfies this equation. This is so because, from the C.R. written above, it follows that H possesses continuously all eigenvalues from $-\infty$ to $+\infty$ (cf. Dirac. Quantum Mechanics, First edition (1930), 34 and 56) whereas on the other hand, discrete eigenvalues of H can be present. *We, therefore, conclude that the introduction of an operator t is basically forbidden and the time t must necessarily be considered as an ordinary number ("c-number")* in Quantum Mechanics* (cf. for this E. Schrödinger, Berl. Ber. (1931) p. 238).**

W. Pauli (1933), in *General Principles of Quantum Mechanics*, (Springer, Berlin, 1980); footnote p. 63.

Time *and* Quantum Mechanics
(the parameter – operator conundrum)

Heisenberg 1927: three uncertainty relations

Position – Momentum:	$\Delta q_i \cdot \Delta p_i \geq \hbar$	Canonical variables
Angle – Action:	$\Delta w \cdot \Delta J \geq \hbar$	
Time – Energy:	$\Delta t \cdot \Delta E \geq \hbar$	

Time *and* Quantum Mechanics
(the parameter – operator conundrum)

Heisenberg 1927: three ~~uncertainty relations~~ ***inequalities***

Position – Momentum:	$\mathrm{D}q_i \cdot \mathrm{D}p_i \geq \hbar$	Canonical variables
Angle – Action:	$\mathrm{D}w \cdot \mathrm{D}J \geq \hbar$	
Time – Energy:	$\mathrm{D}t \cdot \mathrm{D}E \geq \hbar$	

Rationalized via Fourier-Transform analysis

Inequalities can be rationalized using algebraic properties of commutators or Fourier analysis.

A) Heisenberg vs. Pauli.

B) Mathematical background: Weyl and Stone-von Neumann.

C) Derivations of Heisenberg inequalities.

D) Tunneling times.

E) Wigner *et al.* time delay.

F) Delays in photoionization.

Heisenberg's inequalities for operators constructed from canonical variables (q_i, p_i)

Commutator of position and momentum operators (1-dimensional system)

$$[Q,P] = i\hbar 1 \qquad (1)$$

Q and P self-adjoint operators on an infinite Hilbert space H of complex-valued L^2 functions.

Pauli: If $\hbar \neq 0$, Eq. (1) has solutions only if Q and P are unbounded.
By contrast, it has no solution if:

- H is finite-dimensional (trace of commutator [Q,P] would be = 0);
- either Q or P is bounded (or semi-bounded);

⇔ ***Stone-von Neumann theorem***

W. Pauli (1933), in *General Principles of Quantum Mechanics*, (Springer, Berlin, 1980); footnote p. 63.
Weyl + Stone-von Neumann theorem: see: J. Rosenberg, in Contemporary Maths. **365**, 331-353 (2004)

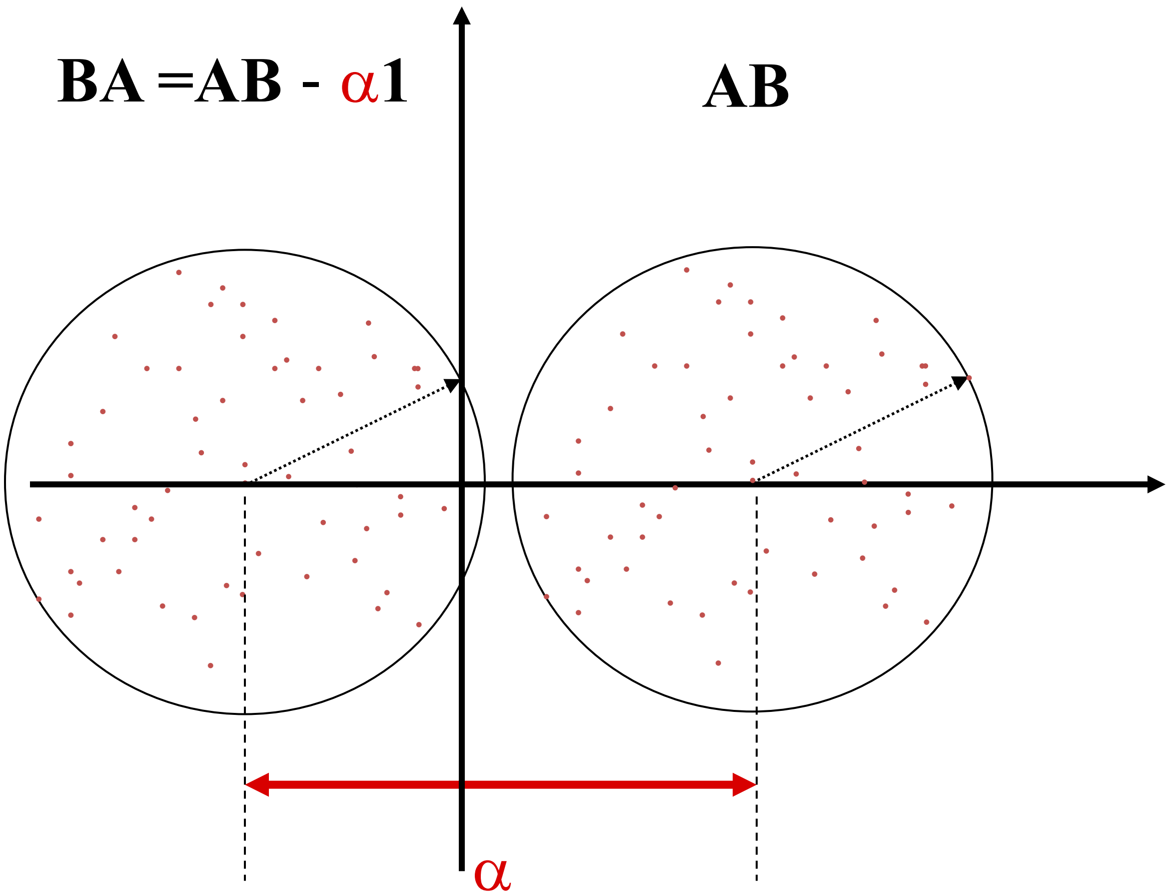
Spectra and commutators of bounded operators

1) Let: A and B bounded, then AB and BA are bounded. (i.e. finite spectral radius)

Suppose they have a non-zero commutator: $AB - BA = \alpha 1$ (1) AB and BA spectra would be translated by α.

However, as $AB = A(BA)A^{-1}$ (similarity transform) AB and BA have the same spectrum.

$\Leftrightarrow$ Eq. (1) verified only if $\alpha = 0$.



2) If H is finite-dimensional, with dimension N, the trace of the commutator $\mathrm{Tr}[A,B] = 0$, while $\mathrm{Tr}\, 1 = N$

The Stone-von Neumann Theorem (1930-31)

Original idea from Weyl (1928):
Q and P can be exponentiated to one-parameter unitary groups:

$$V_t = e^{itQ} \text{ and } U_t = e^{itP},\ t \in \mathbf{R}$$

One constructs the operator: $U_\tau Q U_{-\tau}$, (same spectum as Q),
and expanding the exponentials, one finds:

$$U_t Q U_{-t} = e^{itP} Q e^{-itP} = Q + it\,[P,Q] + \frac{(it)^2}{2!}[P,[P,Q]] + \frac{(it)^3}{3!}[P,[P,[P,Q]]]\ldots$$

(Baker-Campbell-Hausdorff formula + Hadamard)

One has,
for instance:

$$e^{itP} Q e^{-itP} = (1 + itP + \frac{(it)^2}{2!}PP + \cdots)Q(1 - itP + \frac{(-it)^2}{2!}PP + \cdots)$$

$$= Q + it(PQ - QP) + \frac{(it)^2}{2!}\underbrace{(PPQ - 2PQP + QPP)}_{=\,[P,[P,Q]]} + \cdots$$

The Stone-von Neumann Theorem (II)

$$U_t Q U_{-t} = e^{itP} Q e^{-itP} = Q + it\,[P,Q] + \frac{(it)^2}{2!}[P,[P,Q]] + \frac{(it)^3}{3!}[P,[P,[P,Q]]]\ldots$$

- a) As: $[P,Q] = -i\hbar$, $\Rightarrow [P,[P,Q]] = 0$, idem for higher-order terms, then:

$$\Rightarrow \boxed{U_\tau Q U_{-\tau} = Q + \hbar\tau}$$

shows that the spectrum of Q is invariant under a translation $\hbar\tau$

$\Leftrightarrow$ spectrum of Q is the real line and is unbounded.

- b) Generalization: Applies to any power Q^n and, thus to any function $f(Q)$ expressible as a power series. Thus:

$$U_\tau f(Q) U_{-\tau} = f(Q + \hbar\tau)$$

In particular: $U_\tau V_\sigma U_{-\tau} = U_\tau e^{i\sigma Q} U_{-\tau} = e^{i\sigma(Q + \hbar\tau)} = e^{i\hbar\sigma\tau} V_\sigma$

One has also: $V_\sigma U_\tau V_{-\sigma} = e^{i\hbar\sigma\tau} U_\tau$

$\Leftrightarrow$ multiplicative form of the commutation relation (Weyl)

The Stone-von Neumann Theorem (III)

This is the mathematical background Pauli used to formulate his famous statement.

Theorem states the unicity (i.e. irreducibility) of the representations of the Heisenberg commutation relations in Weyl form.

Generalizations exploiting the Lie algebra of commutators, constitute the main outgrow of the theorem.

Further extensions related to the so-called Heisenberg group, have given rise also to an abundant literature (Mackey, Weil, Lions, etc.)

No hermitian « time operator » in quantum mechanics !

From the preceding analysis, (+Stone-von Neumann theorem):
Since, based on physical grounds,
• time is unbounded ($t \in]-\infty, +\infty[$)
• Hamiltonian H is semi-bounded (or with discrete eigenvalues),
⇒ there is no Hermitian time operator in quantum mechanics **(*)**
⇒ time is not an "observable" quantity.
In other words: a time delay cannot be associated
to eigenvalues of an hermitian operator.

See also the papers by Briggs & Rost: Time enters Quantum Mechanics as the result of the coupling of the system with the (classical) environment, which encompasses the time measurement device itself.

Euro. Phys. J. D**10**, 311 (2000) and Found. Phys. **31**, 693 (2001).

(*) As Joachim Burgdörfer said last week, this assertion is still discussed in the mathematical physics literature. See for instance:
Time in Quantum Mechanics, Muga *et al.* editors, Lecture Notes in Physics **789** (2009)

A) Heisenberg vs. Pauli.

B) Mathematical background: Weyl and Stone-von Neumann.

C) Derivations of Heisenberg inequalities.

D) Tunneling times.

E) Wigner *et al.* time delay.

F) Delays in photoionization.

From Commutators of Canonical Variables Operators to Heisenberg's Inequalities

(an operational approach, via expectation values of operators)

$$\langle Q \rangle = \langle Y, QY \rangle$$

Variance: $(DQ)^2 = \left\langle (Q - \langle Q \rangle)^2 \right\rangle; \quad (DP)^2 = \left\langle (P - \langle P \rangle)^2 \right\rangle$

Using commutator + Cauchy-Schwarz: $(DQ)^2 \cdot (DP)^2 \geq \frac{1}{4}\left(\langle [Q, P] \rangle\right)^2$

Heisenberg: $DQ \cdot DP \geq \frac{1}{2}\left|\langle [Q, P] \rangle\right| = \frac{\hbar}{2}$

Robertson-Schrödinger Inequality

Let A and B a pair of hermitian operators. For a system in state |Ψ>, define:

$$|f\rangle = \left(A - \langle A\rangle\right)|\Psi\rangle \quad ; \quad |g\rangle = \left(B - \langle B\rangle\right)|\Psi\rangle$$

$$\text{Variances:} \quad \sigma_A^2 = \langle f|f\rangle \quad ; \quad \sigma_B^2 = \langle g|g\rangle$$

$$\sigma_A^2 \cdot \sigma_B^2 = \langle f|f\rangle\langle g|g\rangle \geq \left|\langle f|g\rangle\right|^2 \qquad \text{(Cauchy - Schwarz)}$$

$$\text{as: } \langle f|g\rangle^* = \langle g|f\rangle, \text{ one has: } \left|\langle f|g\rangle\right|^2 = \left(\frac{\langle f|g\rangle + \langle g|f\rangle}{2}\right)^2 + \left(\frac{\langle f|g\rangle - \langle g|f\rangle}{2i}\right)^2$$

$$\langle f|g\rangle = \langle\Psi|AB - A\langle B\rangle - \langle A\rangle B + \langle A\rangle\langle B\rangle|\Psi\rangle = \langle AB\rangle - \langle A\rangle\langle B\rangle$$

$$\text{and: } \langle g|f\rangle = \langle BA\rangle - \langle A\rangle\langle B\rangle$$

$$\text{Thus:} \quad \langle f|g\rangle + \langle g|f\rangle = \langle AB + BA\rangle - 2\langle A\rangle\langle B\rangle, \quad \textbf{\textit{note the anti - commutator}}!$$

$$\text{and: } \langle f|g\rangle - \langle g|f\rangle = \langle [A, B]\rangle$$

$$\Delta A^2 \cdot \Delta B^2 = \sigma_A^2 \cdot \sigma_B^2 \geq \left(\frac{\langle AB + BA\rangle - 2\langle A\rangle\langle B\rangle}{2}\right)^2 + \left(\frac{\langle [A, B]\rangle}{2i}\right)^2$$

Remark: Harmonic Oscillator and Other Systems

$$DA^2 \cdot DB^2 = s_A^2 \cdot s_B^2 \quad \geq \quad \left(\frac{\langle AB + BA \rangle - 2\langle A \rangle \langle B \rangle}{2}\right)^2 + \left(\frac{\langle [A, B] \rangle}{2i}\right)^2$$

When specializing to the case of non-diagonal operators (such as x and p_x), the Heisenberg inequality is often written: $\Delta x \, . \, \Delta p_x \geq \hbar/2$, with $[x, p_x] = i\hbar$. This expression does not make apparent the first term of the inequality written above for the product of the variances.

For most systems, this first term is ≠ 0.

The equality $\Delta x \, . \, \Delta p_x = \hbar/2$ is verified for the ground state of the harmonic oscillator sytem.

Remark: One checks that the minimum uncertainty $\Delta x \, . \, \Delta p_x = \hbar/2$ is verified for the normal (i.e. Gaussian) momentum and spatial distributions in the ground state.

One can make a parallel with the Fourier Transform, where the minimum of the time-bandwidth product is reached for Gaussian distributions.

Time-Energy Inequality

As there is no commutation relation for time and energy, one has to rely on the properties of Fourier transforms.

Fourier-Transform pair:

$$g(E) = \frac{1}{\sqrt{2\pi\hbar}} \int_{\mathbf{R}} e^{-iEt/\hbar} f(t)\, dt; \qquad f(t) \in L^2(\mathbf{R});$$

$$f(t) = \frac{1}{\sqrt{2\pi\hbar}} \int_{\mathbf{R}} e^{+iEt/\hbar} g(E)\, dE; \qquad g(E) \in L^2(\mathbf{R});$$

Parseval:

$$\int_{\mathbf{R}} |g(E)|^2\, dE = \int_{\mathbf{R}} |f(t)|^2\, dt \;;$$

Variances:

$$\Delta t^2 = \sigma_t^2 = \int_{\mathrm{R}} (t - t_0)^2 |f(t)|^2\, dt = \int_{\mathrm{R}} (t^2 - t_0^2) |f(t)|^2\, dt;$$

$$\Delta E^2 = \sigma_E^2 = \int_{\mathrm{R}} (E - E_0)^2 |g(E)|^2\, dE = \int_{\mathrm{R}} (E^2 - E_0^2) |g(E)|^2\, dE$$

where t_0 and E_0 are the average (expectation) values:

$$t_0 = \int_R t |f(t)|^2\, dt \quad \text{and} \quad E_0 = \int_R E |g(E)|^2\, dE\,;$$

t_0 and E_0 can be removed via variable changes like $t \to t - t_0$, etc.

$\Rightarrow$ one can consider: $\Delta t^2 = \int_{\mathrm{R}} t^2 |f(t)|^2\, dt \;;\; \Delta E^2 = \int_{\mathrm{R}} E^2 |g(E)|^2\, dE$

Time-Energy Inequality (follow)

Derivation of Heisenberg inequality (Weyl):

Start from: $\frac{d}{dt}(t\,f(t)) = t\frac{df(t)}{dt} + f(t) \Rightarrow |f(t)|^2 = f(t)\bar{f}(t) = \left[\frac{d}{dt}(t\,f(t))\,\bar{f}(t)\right] - \left(t\frac{df(t)}{dt}\bar{f}(t)\right)$

Then : $\int_R |f(t)|^2 dt = \int_{\mathbf{R}}\left[\frac{d}{dt}(t\,f(t))\,\bar{f}(t)\right]dt - \int_{\mathbf{R}} t\frac{df(t)}{dt}\bar{f}(t)\,dt$

Integrating first term by part : $\int_R |f(t)|^2 dt \quad = \left[t|f(t)|^2\right]_{-\infty}^{+\infty} - \int_{\mathbf{R}} t\,f(t)\frac{d\bar{f}(t)}{dt}\,dt - \int_{\mathbf{R}} t\frac{df(t)}{dt}\bar{f}(t)\,dt$

First term is zero and : $\int_R |f(t)|^2 dt \quad = -2\,\mathbf{Re}\left[\int_{\mathbf{R}} t\,f(t)\frac{d\bar{f}(t)}{dt}\,dt\right] \;\leq\; 2\left|\int_{\mathbf{R}} t\,f(t)\frac{d\bar{f}(t)}{dt}\,dt\right|;$

Cauchy-Schwarz : $\left|\int_{\mathbf{R}} t\,f(t)\frac{d\bar{f}(t)}{dt}\,dt\right|^2 \quad \leq \int_{\mathbf{R}} t^2|f(t)|^2 dt \cdot \int_{\mathbf{R}}\left|\frac{d\bar{f}(t)}{dt}\right|^2 dt$

Derivative: $\frac{d\bar{f}(t)}{dt} = \frac{-i}{\hbar}\frac{1}{\sqrt{2p\hbar}}\int_{\mathbf{R}} e^{-iEt/\hbar} E\,g(E)\,dE$; + **Parseval:** $\int_{\mathbf{R}}\left|\frac{d\bar{f}(t)}{dt}\right|^2 dt = \frac{1}{\hbar^2}\int_{\mathbf{R}} E^2|g(E)|^2 dt$

Thus : $\int_R |f(t)|^2 dt \leq \frac{4}{\hbar^2}\int_{\mathbf{R}} t^2|f(t)|^2 dt \cdot \int_{\mathbf{R}} E^2|g(E)|^2 dt = \frac{4}{\hbar^2}\Delta t^2 \cdot \Delta E^2$

If *f(t)* is normed: Inequality for the product of the square roots of the variances :

$$\Rightarrow \Delta t \cdot \Delta E \geq \frac{\hbar}{2}$$

Time-Energy Inequality (follow)

To summarize: Given the absence of time operator, one has to use a Fourier Transform technique to establish the time-energy Heisenberg inequality.

NB. There exists other derivations, notably the one by Mandelstam and Tamm: Δt is the time interval associated to the energy change ΔE in the expectation value of H.
J. of Physics (USSR) **9** (4) 249 (1945). See also the book by Griffiths: *Introduction to QM* (Pearson) p. 114.

How to Measure "Characteristic Times" or "Time Delays" in Quantum Systems?

With the recent technological advances, the question has arisen of how to "clock" quantum electronic processes on their natural time scale (in the attosecond range).

Two topics have emerged in this context:
- Tunneling times;
- Time delays in photoionization.

NB. I will not discuss the closely related "attoclock" concept as Joachim Burgdörfer already dicussed it and Sasha Landsman (ETH Zürich) can answer all your questions!

A) Heisenberg vs. Pauli.

B) Mathematical background: Weyl and Stone-von Neumann.

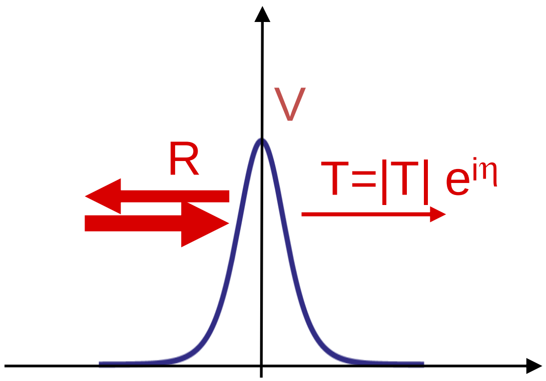
C) Derivations of Heisenberg inequalities.

D) Tunneling times.

E) Wigner *et al.* time delay.

F) Delays in photoionization.

Tunneling Time (I)



The question of the status of time (parameter or dynamical variable?) remained academic, until the advent of STM (Scanning Tunneling Microscopy), of semi-conductor devices and of Josephson junctions measurements (in the 1980s).

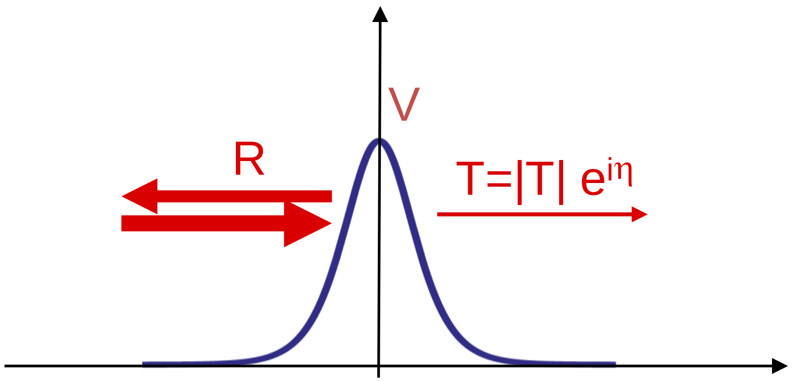
Then arose the question of tunneling times *i.e.*:

"How much time does a tunneling particle spend in the barrier region?"; Steinberg, PRL **74**, 2405 (1995).

An abundant literature has been (and continues to be) devoted to this topic. A non-exhaustive (and subjective) list includes:

- Büttiker and Landauer, "*Traversal Time for Tunneling*", PRL **49**, 1739 (1982).
- Landauer and Martin, "*Barrier interaction time in tunneling*", RMP **66**, 217 (1994).
- Carvalho and Nussenzveig, "*Time Delays*", Phys. Reports **364**, 83 (2002).
- Yamada, "*Unified Derivation of Tunneling Times from Decoherence Functionals*", PRL **93**, 170401 (2004).
- Winful, "*Tunneling time, the Hartman effect and superluminality…*", Phys. Reports **436**, 1 (2006).
- Book: *Time in Quantum Mechanics*, Muga *et al.* editors, Lecture Notes in Physics **789** (2009)
- Galapon, "*Only Above Barrier Energy Components Contribute to Barrier Traversal Time*", PRL **108**, 170402 (2012), etc.

Tunneling Time (II)



Four (4 !) different definitions have been proposed for ***tunneling times*** for a particle with energy E < V (height of barrier).

For a transmission amplitude $T = |T|\, e^{i\eta}$, one distinguishes:

- Baz or Larmor time = $-\hbar\partial\eta/\partial V$ (spin precession in B field) ;
- Büttiker-Landauer time = $-\hbar\partial \ln |T|/\partial V$;
- **Bohm-Wigner time= $\hbar\partial\eta/\partial E$ (or group delay, see below);**
- Pollack-Miller time = $\hbar\partial \ln |T|/\partial E$.

⇒**There is no consensus:** This results essentially from the fact that the high-energy components of the incoming w-p:
i) arrive first;
ii) cross the barrier with a higher probability;
⇒ deformation of w-p…

Tunneling Time (III)

The notion remains extremely controversial !!

Many other "times" have been proposed to deal with the question: Let us mention a few: "dwell time", "time-of-arrival", "traversal time", "group delay", etc..

Joachim Burgdörfer has pointed out that the concept of a "time delay operator" is also used in different areas of physics: Solid state, Optics, Acoustics, etc. All deal with wave phenomena (see Winful's review).

The possibility of defining an hermitian operator associated to time delays has been introduced by Smith (Phys. Rev. **118**, 349, 1960) in an R-matrix context: The eigenvalues of the "lifetime matrix" can be associated to "time delays" in each reaction channel. The basis used describes the system in the "inner region" $r \leq R$.

Recent references include:

- Rotter *et al.* *"Generating particle scattering states in wave transport"* PRL **106**, 120602, 2011 (transmission of electrons through mesoscopic systems)
- Barr & Reichl, "*Quasi-bound states in 2- and 3-dimensional open quantum systems*" PRA **81**, 022707 2010 (non-hermitian hamiltonian)

A) Heisenberg vs. Pauli.

B) Mathematical background: Weyl and Stone-von Neumann.

C) Derivations of Heisenberg inequalities.

D) Tunneling times.

E) Wigner *et al.* time delay.

F) Delays in photoionization.

Wigner's Time-Delay

Wigner had defined "time-delays" in collision processes (*)

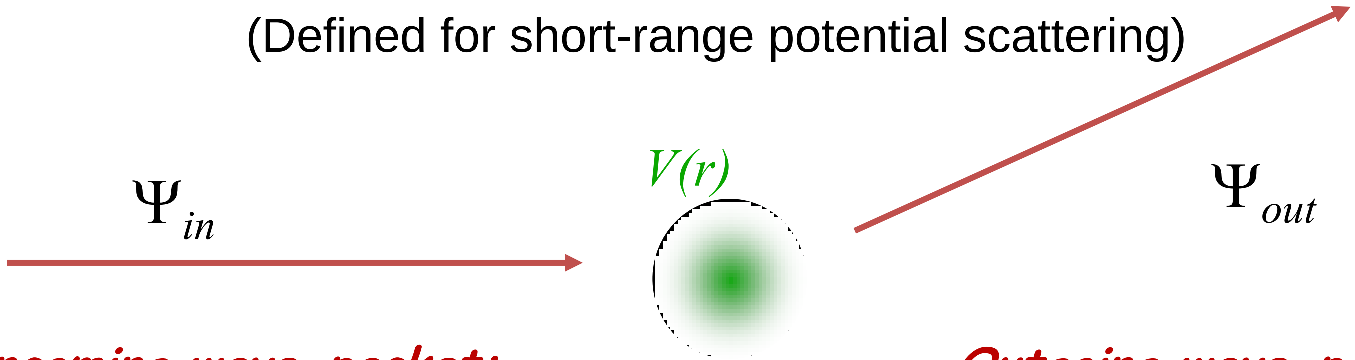
• Idea: In the course of a scattering process by a potential, a particle experiences a "time-delay" ***as compared to free motion***. NB. Reference to classical motion is implicit.

• Main result: The delay is intimately related to the scattering phase-shift induced by the potential (see below).

(*) Wigner, Phys. Rev. **98**, 145 (1955)

"Wigner time-delay" ⇔ scattering phase-shift

(Defined for short-range potential scattering)



Incoming wave-packet:

$$\Psi_{in}(r,t) \propto \int_0^{+\infty} dE\,|A(E)|\,e^{i(-kr-Et)}$$

$S = -kr - Et$ ($r = 0$ at $t = 0$)
Time-position relation for the maximum of w-p, given by ***stationary phase*** : $\partial S/\partial E = 0$,
with $k = (2E)^{1/2}$ and $dk/dE = 1/k$
$\Rightarrow$ $t = -r/k$; ($t < 0$)

Outgoing wave-packet:

$$\Psi_{out}(r,t) \propto \int_0^{+\infty} dE\,|A(E)|\,e^{i(kr-Et+\eta(E))}$$

Here, $\eta(E)$ is the scattering phase-shift induced by $V(r)$.
With $S = kr - Et + \eta(E)$ and $\partial S/\partial E = 0$,
the time-position relation becomes:
$t = r/k + t_W$; ($t > 0$)

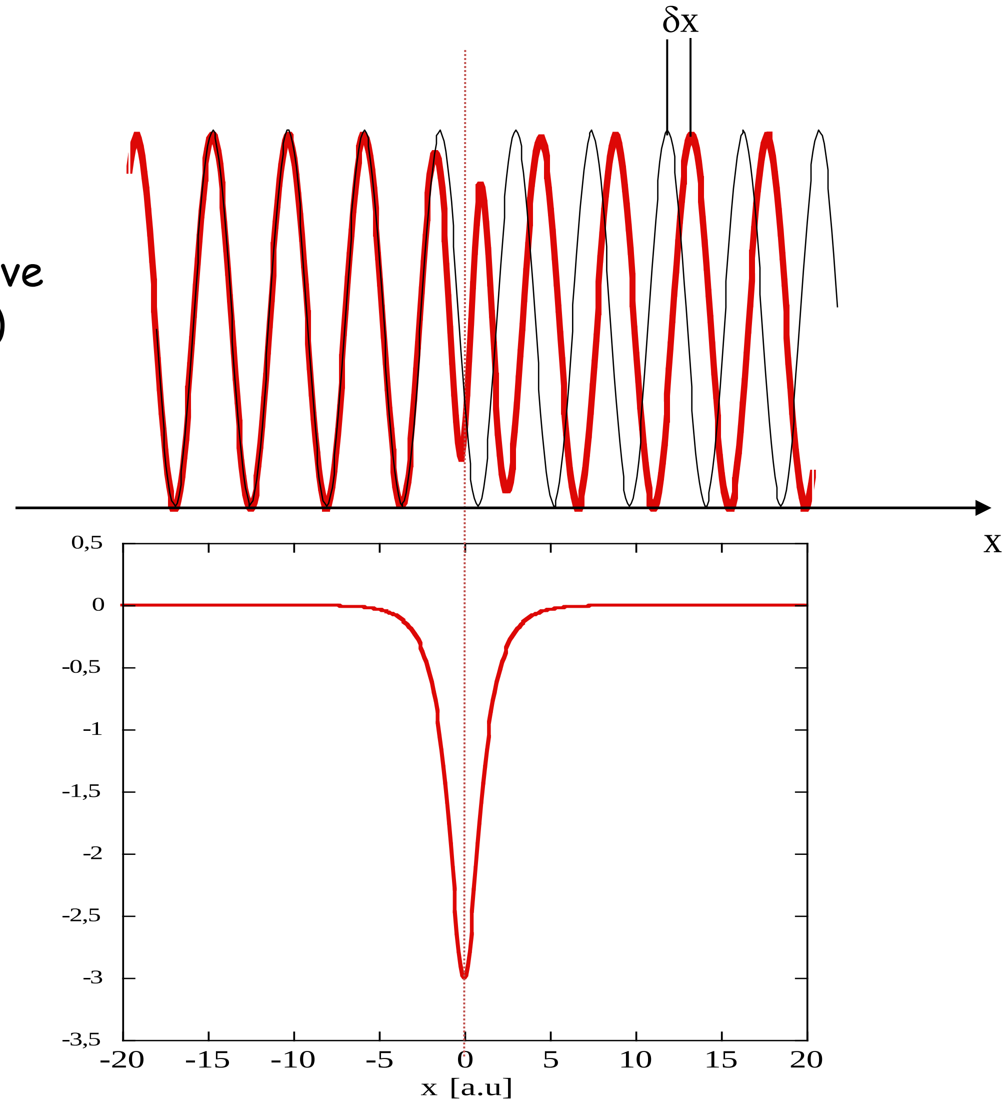
Where: $$t_W = \frac{d\eta(E)}{dE}$$ = "Wigner time-delay"

Wigner, Phys. Rev. **98**, 145 (1955)

Wigner: 1-D scattering and phase-shifts

Free particle = Plane wave
(wave number k=1 a.u.)

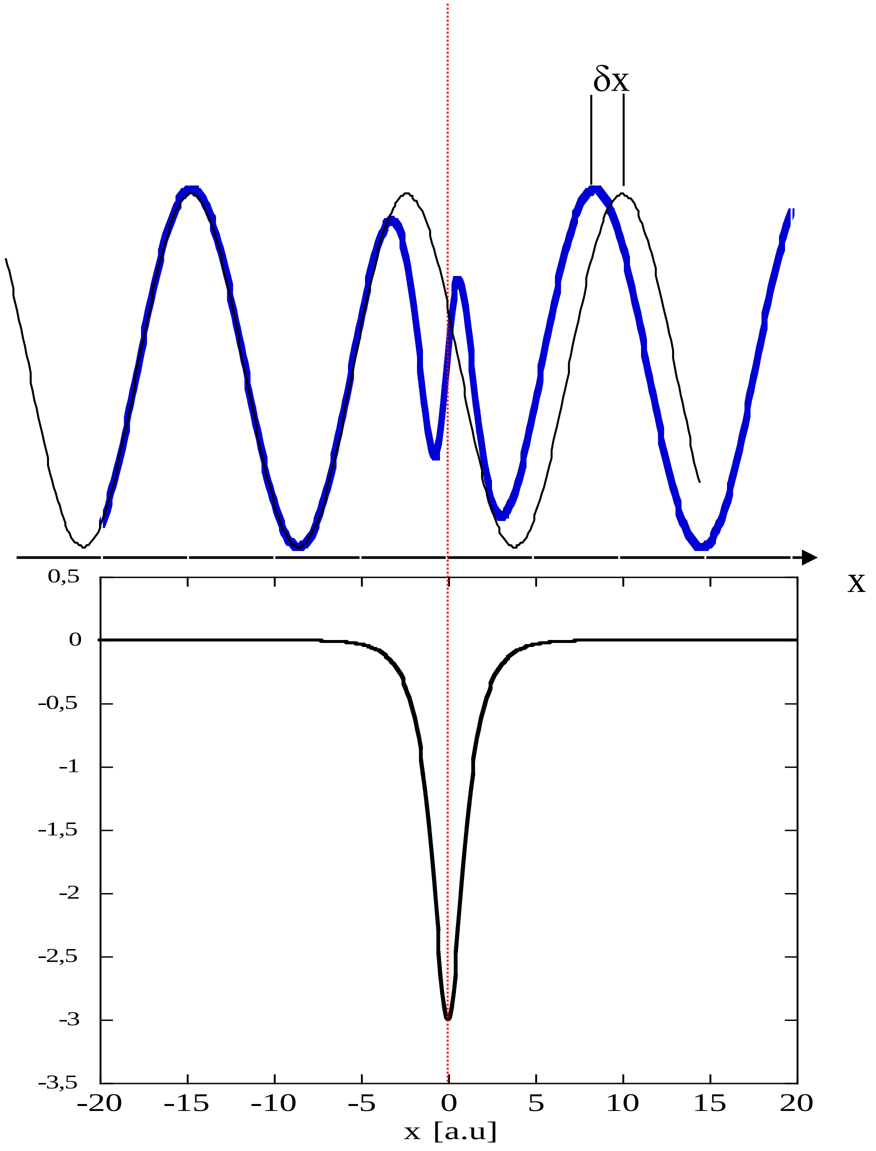
Scattering by a
short-range potential:



The potential induces a phase shift

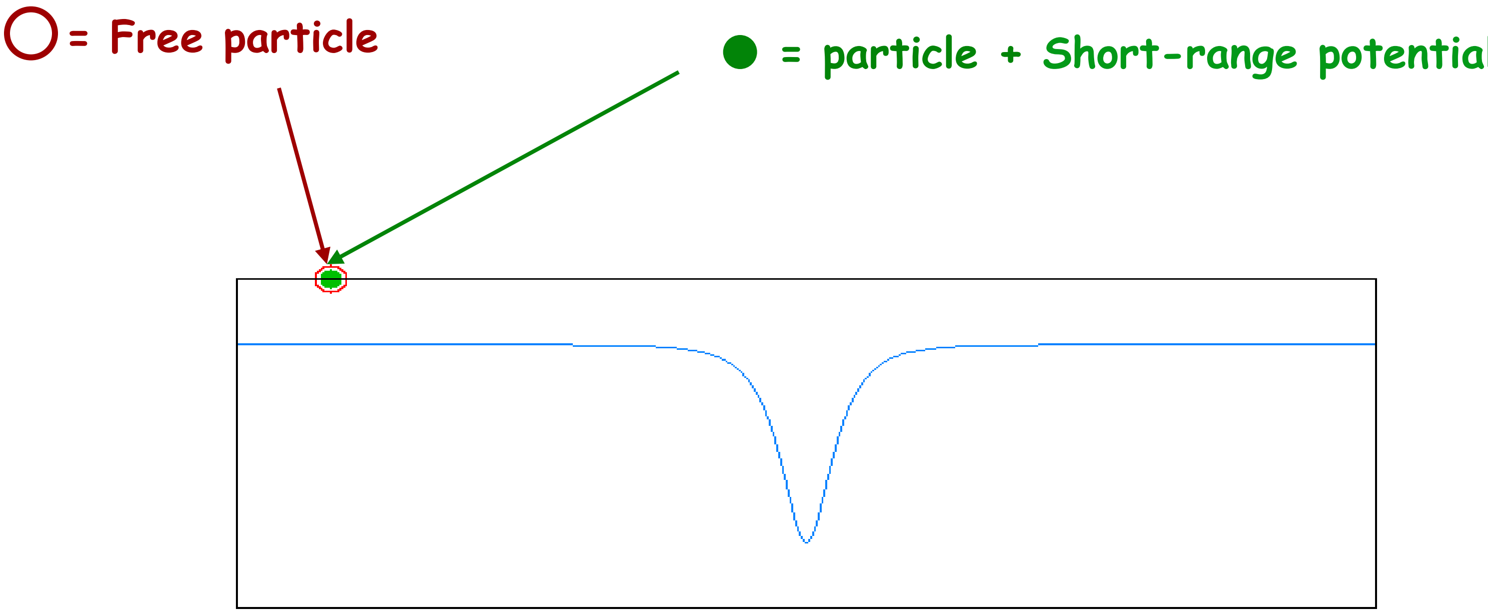
1-D scattering and phase-shifts

A slower free particle
(wave number k=0.5 a.u.)



The phase shift depends on the particle's energy.

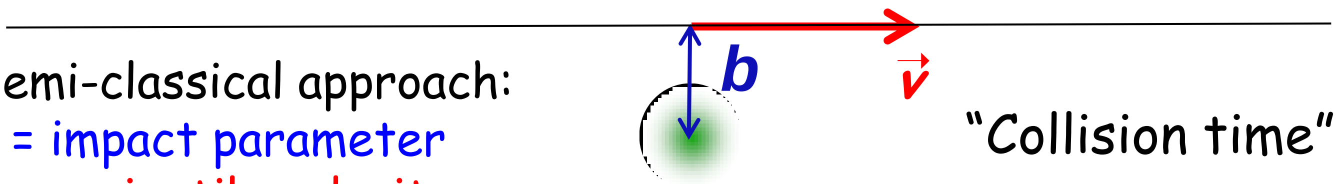
1-D scattering and time-delay: a classical perspective



Measurement of Time Delays

The above delays are associated to "gedanken" experiments.
In experiments with particle beams it is not feasible to determine a delay between particles in free motion and the ones experiencing a collision or tunneling.

Morever, "Collision times" were expected to range in the attosecond range or shorter, i.e. inaccessible to experiments



Semi-classical approach:
b = impact parameter
v = projectile velocity

"Collision time" = b/v

Things have changed with the advent of XUV sources delivering "attosecond" pulses with the possibilty to synchronize them to an IR pulse which will play the role of a "clock".

Remarks on “Wigner time-delay”

• Wigner time delay is defined via a stationary phase analysis:
It is based on the motion of the maximum of the w-p spatial distribution.
This implies that the w-p keeps a decent spatio-temporal shape.

• It is a group delay, defined with reference to free classical motion.
• Other definitions have been proposed e.g. motion of the average value of the position operator, etc.

• When applied to tunneling this group delay is different from a traversal time (see Landauer and Winful’s papers).
Another problem is that if you have an incoming gaussian w-p, the transmitted one is never a pure gaussian. Not to mention the reflected component which interferes with the incoming wave.

A) Heisenberg vs. Pauli.

B) Mathematical background: Weyl and Stone-von Neumann.

C) Derivations of Heisenberg inequalities.

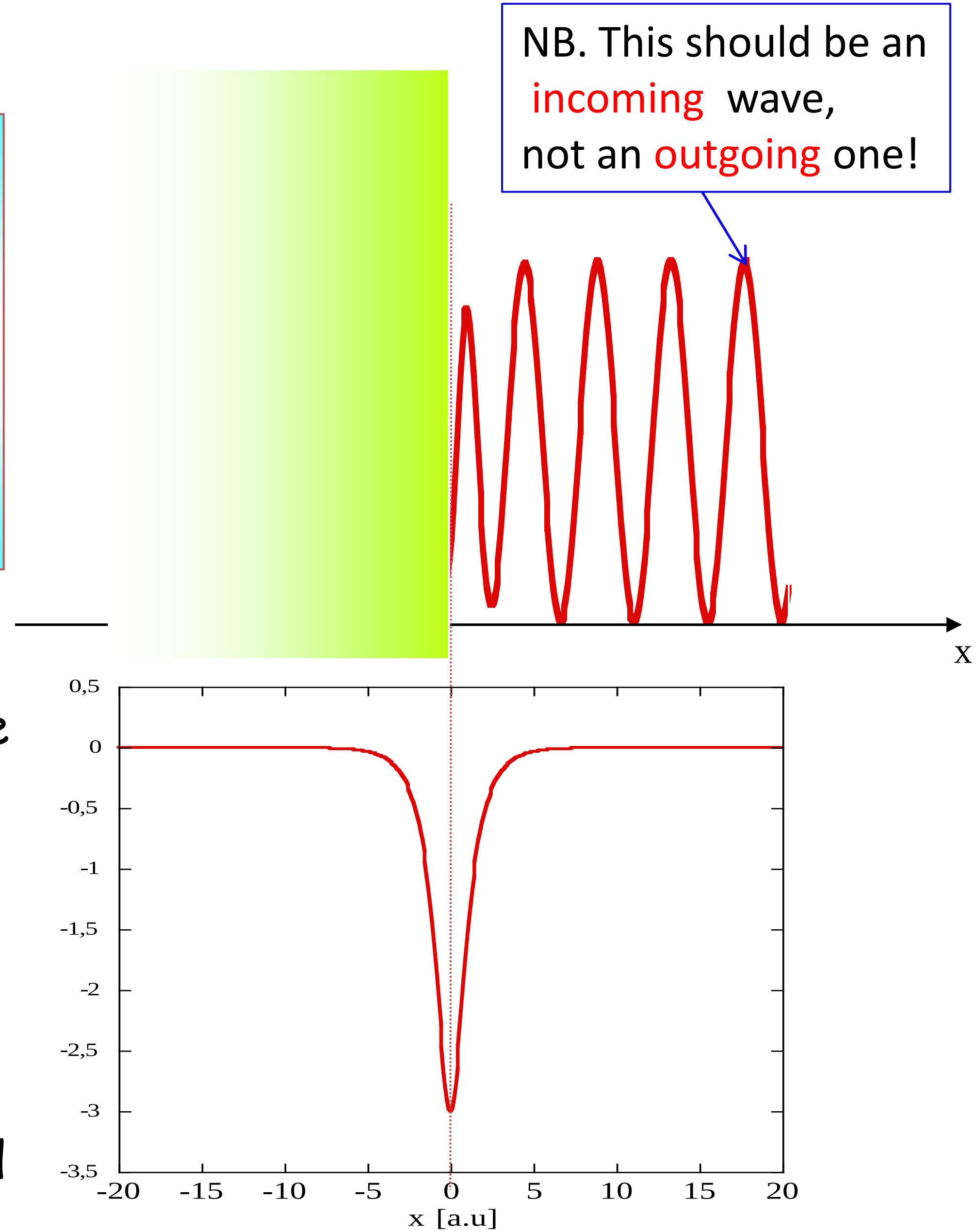
D) Tunneling times.

E) Wigner *et al.* time delay.

F) Delays in photoionization.

First idea: Photoionization = "half-collision"

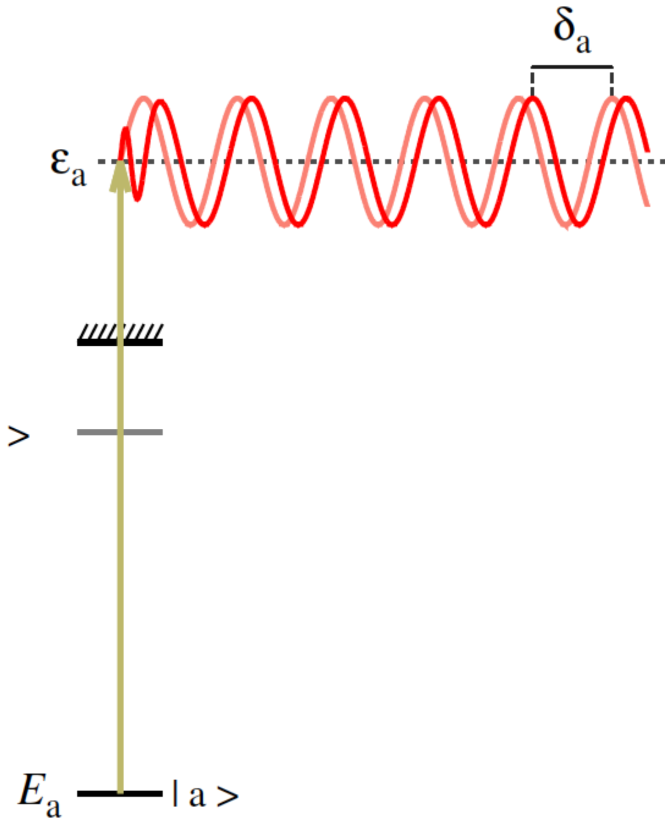
⇒ Delay linked to scattering phase-shifts of the photoelectron wave function ? (Wigner picture)



• Remark: Again, such delays are defined from "gedanken" experiments. In photoionization of neutrals, one has to determine a delay between electrons in a pure Coulomb potential and the ones in the potential of a structured ionic core.

Attosecond XUV Photoionization

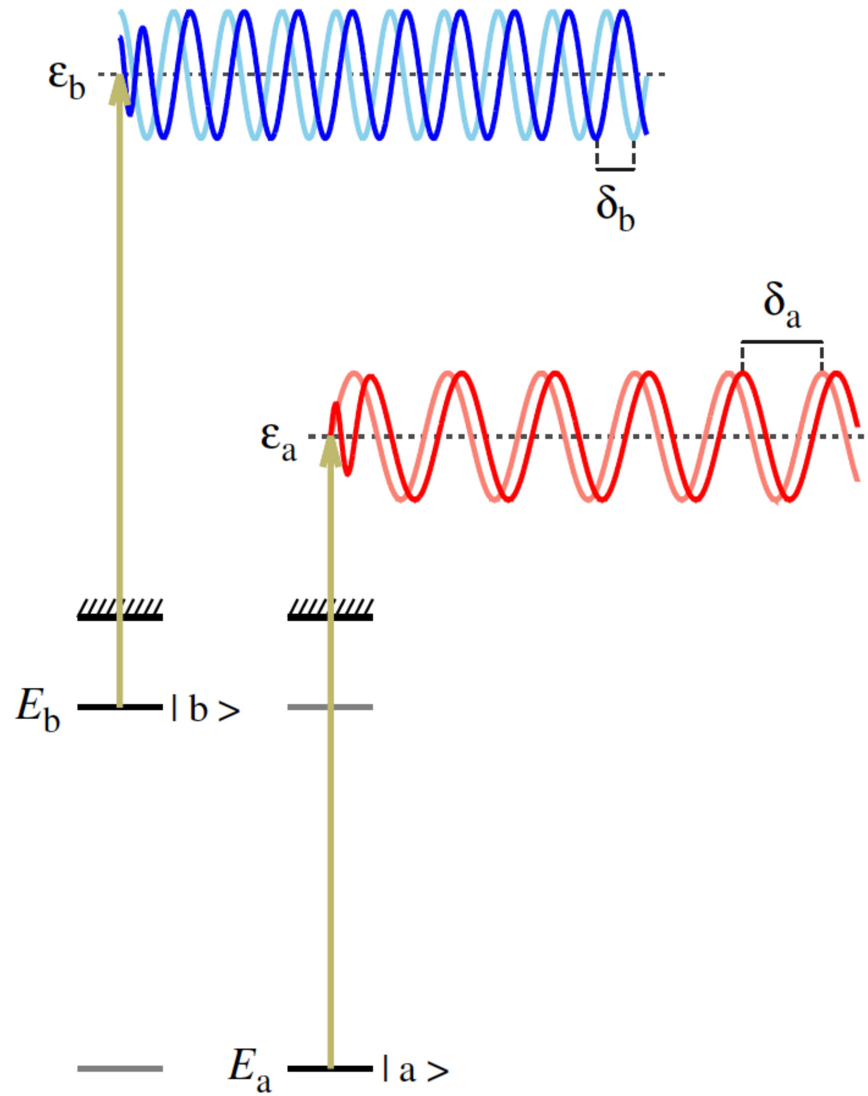
However,
the time delay,
as compared to free motion,
is not measurable.
Ones needs a “clock”.



Attosecond XUV Photoionization

When ionizing atoms from either one of two distinct states with the same attosecond pulse, one can compare the time evolution of the photoelectron wave-packets
(with different phase-shifts).
Measured delays in atoms:

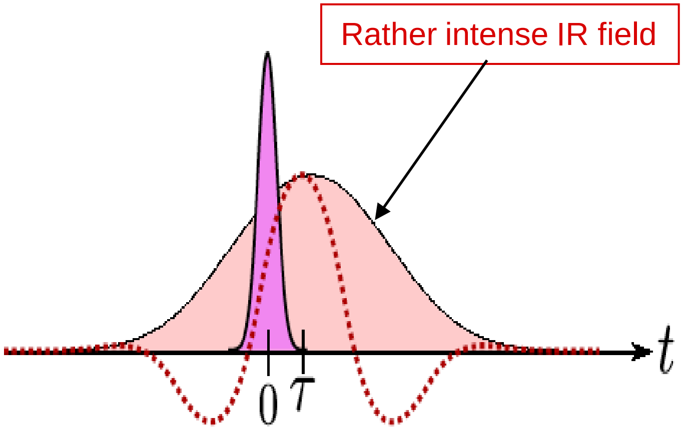
• $\Delta\tau_{2s-2p}$ (Ne) ≈ 20 as (Garching, Streaking)
□ $\Delta\tau_{3s-3p}$ (Ar) ≈ 100 as (Lund, RABBIT)



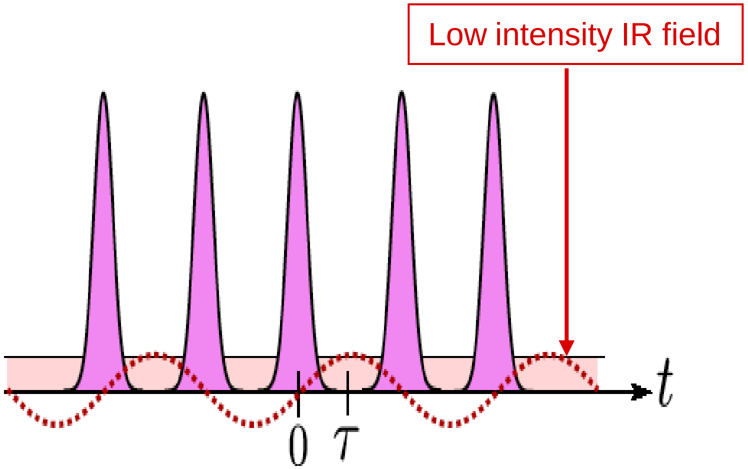
NB. One needs the presence of an additional (IR) field to "clock" the process.

Two-Color IR-XUV Attosecond Pump-Probe Experiments

Streaking
Single attosecond XUV pulse
+ few-cycle IR pulse with controlled Carrier-Envelope Phase;
(Garching)

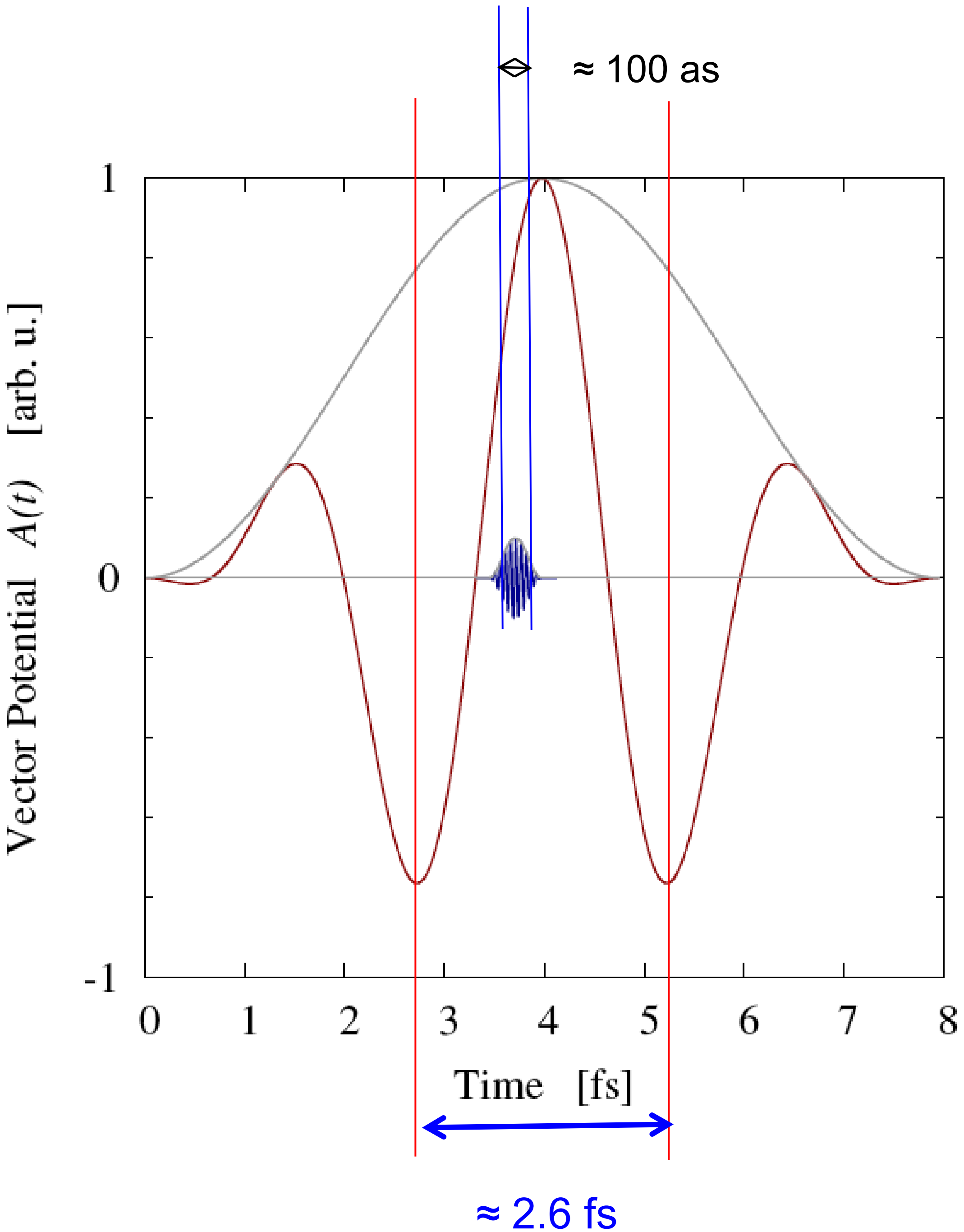


RABBIT
Attosecond XUV pulse train + (long-)IR pulse;
(Lund)

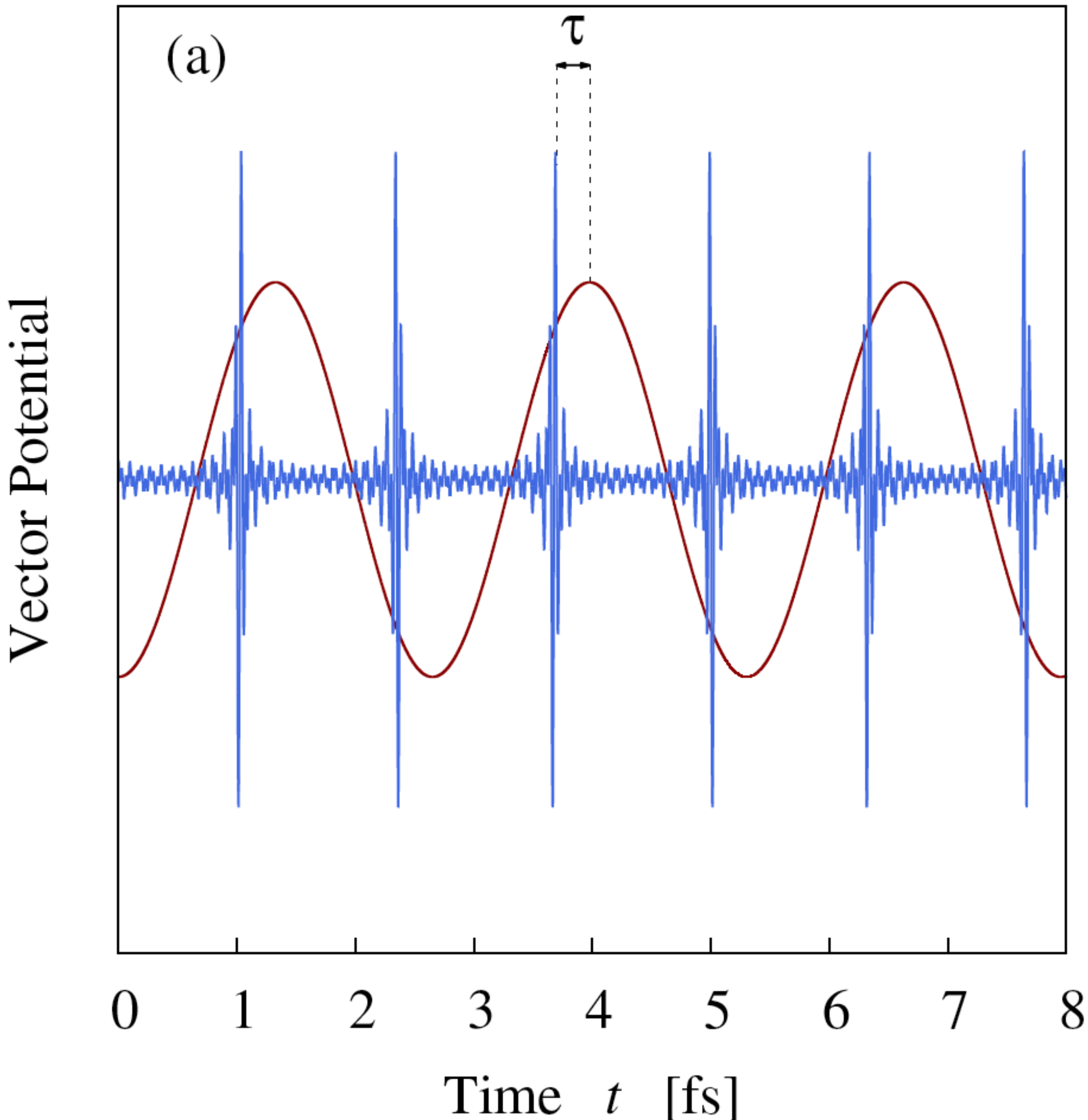


See: Dahlström et al. JPB **45**, 183001 (2012) (tutorial).

Two-color
IR-XUV pulses:
Attosecond
range;
Streaking



RABBIT



Phase-Shifts

Idea: One needs to determine the energy dependence of the phase-shifts in order to compute "Wigner-like time-delays".

However: Computation of phase-shifts in wave functions of photoelectrons remains a challenge for theory.

Experimental determination is difficult.
one needs Photoelectron Angular Distributions (PADs).

We have experts here, who can tell you more than myself on the topic: Marcus Dahlström, Anatoli Kheifets & Eva Lindroth

However, Wigner time-delays computed either from phase shifts or deduced from TDSE calculations for Ne 2s and 2p amount to:

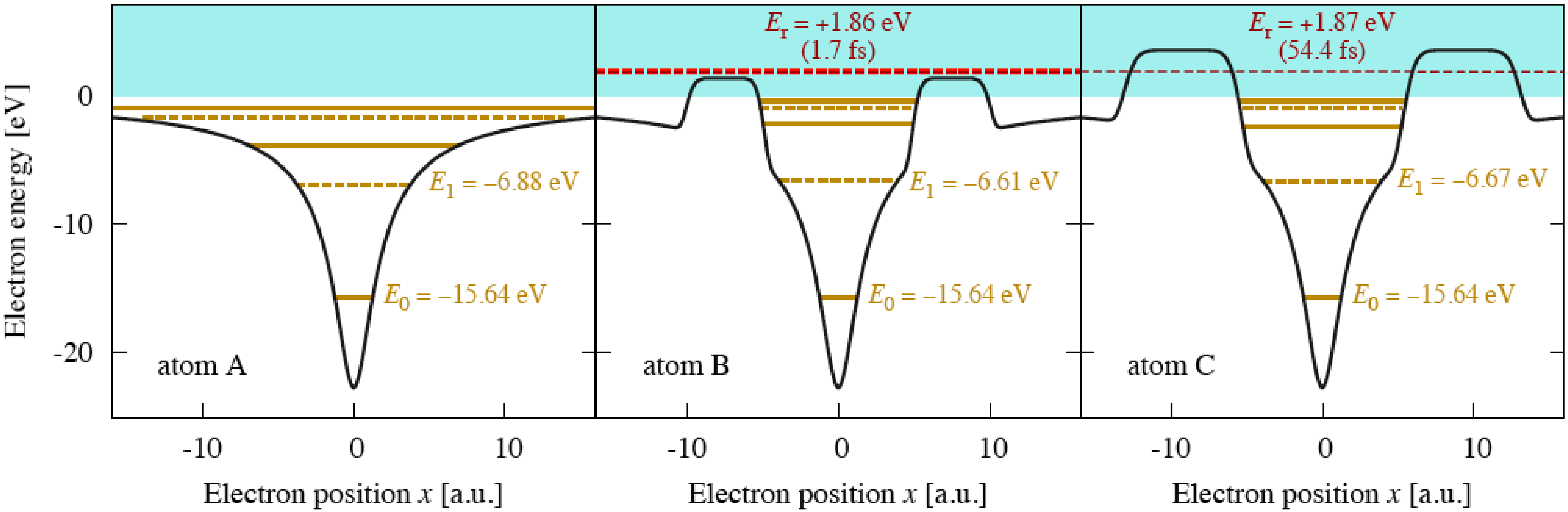
$$\Delta\tau_W \approx 5 - 12 \text{ as} \;<\; \Delta t \approx 21 \text{ as}.$$

Physical origin of the discrepancy ?
Electron correlations ?

Arises the question of the role of the measurement process (based on the use of an auxiliary IR field which "dresses" the system's states)

Role of resonances: Numerical experiments

Idea: Use one-electron, 1-D model potentials designed to mimic the presence of a shape resonance in the continuous spectrum



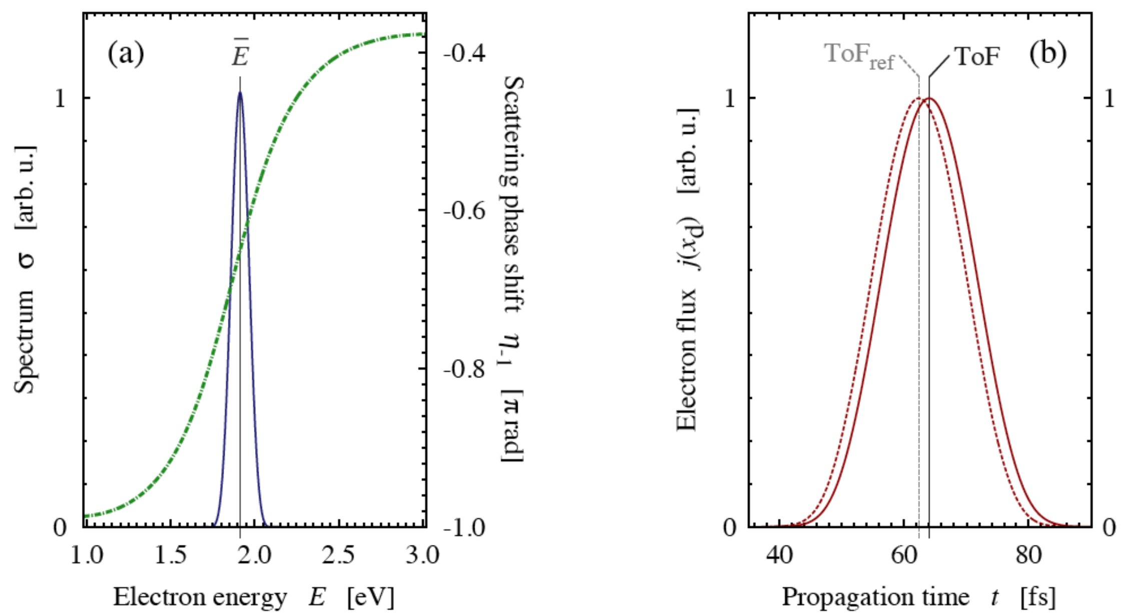
The energy and width (or lifetime) of the resonance can be changed by tuning the position, height and width of the barriers.

Caiilat *et al*. PRL **106**, 093002 (2011);
Dahlström *et al*. to appear (2014)

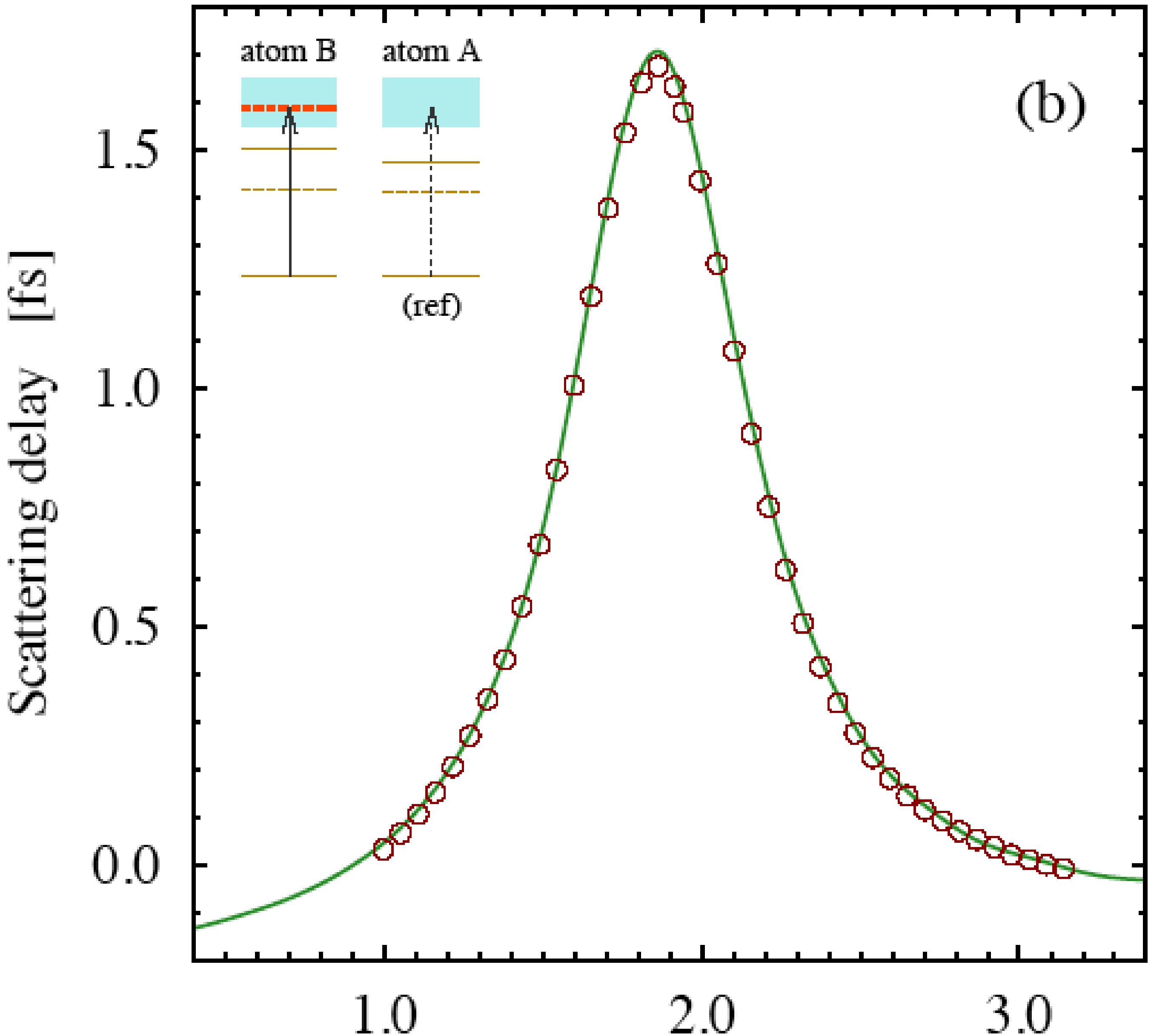
Validation of the concept of Wigner time delay

Idea: Compute the Wigner time delay from the phase shifts and their energy derivatives and compare to numerical "times of flight".
The system A is used as a reference. Case (B) of a broad resonance.

Lifetime of the resonance B shorter than pulse duration.
(a): Photoelectron spectrum and phase-shift of w-f.
(b): Time dependence of photoelectron fluxes for potentials A and B. Vertical lines: Average time under the peaks = ToF. Fluxes are computed at x_d =1000 a.u.



The difference between the averaged "times of flight":
$\Delta T = ToF_{(C)} - ToF_{(A)} \approx 1.65$ fs, agrees with the Wigner-like delay



atom B
atom A
(ref)
(b)
Scattering delay [fs]
1.5
1.0
0.5
0.0
1.0
2.0
3.0

Eugene P. Wigner

COMMUNICATIONS ON PURE AND APPLIED MATHEMATICS, VOL. XIII, 001–14 (1960)

The Unreasonable Effectiveness of Mathematics in the Natural Sciences

Richard Courant Lecture in Mathematical Sciences delivered at New York University, May 11, 1959

EUGENE P. WIGNER

Princeton University

"and it is probable that there is some secret here which remains to be discovered." (*C. S. Peirce*)

" Physics is becoming so unbelievably complex that it is taking longer and longer to train a physicist. It is taking so long, in fact, to train a physicist to the place where he understands the nature of physical problems that he is already too old to solve them. ”

E. P. Wigner

Collaborations

LCP-MR:
Alfred Maquet
Richard Taïeb
+ Jérémie Caillat

Atomic Physics, Lund
J.M. Dahlström
T. Fordell
M. Gisselbrecht
D. Guénot
P. Johnsson
K. Klünder
A. L'Huillier
J. Mauritsson
M. Swoboda

Collaborations

LCP-MR:
Jérémie Caillat
Roland Guichard
Alfred Maquet
Richard Taïeb

CEA Saclay:
Thierry Auguste
Pierre Breger
Bertrand Carré
Zsolt Diveki
Stefan Haessler
Thierry Ruchon
Pascal Salières

CELIA:
Eric Constant
Baptiste Fabre
Julien Higuet
Yann Mairesse
Eric Mével