Lecture 1 17.9.13 Ionization of Atomic Systems

Motivation: Present day laser experiments

Goal: to probe valence & inner shell electron dynamics inside molecules & atoms

"Dream": to observe electron dynamics in a chemical reaction in "real time"

What does "real time" mean?

Time-scale of electron dynamics inside atoms & molecules ⇒ atomic units of time - au

1 au ≈ 24 as, where 1 as = 10^{-18} s

Atomic units: $m_e = 1$; $\hbar = 1$; $|e| = 1$

Hydrogen (Bohr model): orbital period = $2\pi \approx 150$ au

To observe electron dynamics in real time, need resolution on the attosecond time scale.

⇒ Need attosecond pulses

⇒ First pulse train produced and characterized in 2001 ⇒ attosecond science was born

Science, 292, 1689, (2001)

(2) Going back in time...

Previously: Ahmed Zewail: Nobel Prize in Chemistry in 1999 for observing a chemical reaction on a femtosecond (10^{-15} sec) time-scale

⇒ motion of nuclei is on a fs time scale

⇒ Has been called "father of femtochemistry"

⇒ To view chemical reaction on a fs time-scale, needed "ultra-short" laser pulses or pulses on duration of fs

Going even further back in time: The birth of high-speed photography - 1878

- used to investigate whether horse's feet were off the ground all @ the same time

⇒ resolution milli-seconds!

⇒ The time-scale of the phenomena being probed determines the duration of the light-pulse needed to resolve the phenomena

⇒ The next frontier & the goal of attosecond science is to observe electron motion using "snapshots" of attosecond pulses

(3) Jumping ahead: the next frontier ⇒

as pump ⇒ as probe experiments

⇒ as pump excites the atom/molecule on an as fine scale

⇒ as probe probes it

This is how ~~I~~ we nail observed chemical reaction on a fs time-scale (using 2 ultrashort (fs) pulses separated in time)

Pump: $\psi^0(0) \rightarrow \psi'(0)$; then evolves field free

Probe: $\psi'(\tau) \rightarrow \psi^2(\tau)$ ⇒ the final state

⇒ temporal information (e.g. molecular vibration) can be extracted by repeating the experiment for different τ

⇒ intermediate steps in a chemical reaction have been investigated

(4)

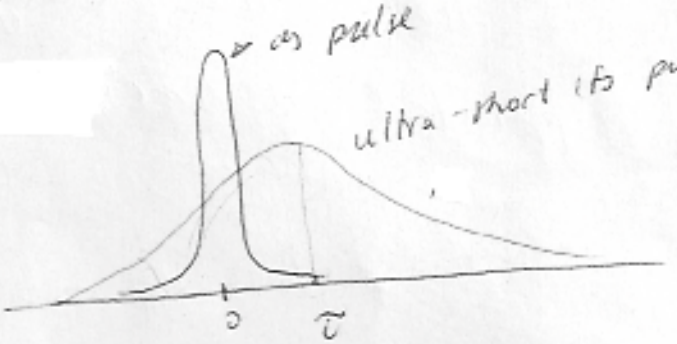
NO as pump as probe currently exists!

⇒ As pulses are produced by HHG & currently energy flux is too low

⇒ Alternative sources (plasma mirrors, FEL) promise such sources ⇒ much work currently in progress

However, there is a way to time processes with attosecond precision, even with fs (ultrafast) pulses

⇒ streaking



Atom absorbs a XUV photon from an as pulse (10 nm – 100 nm) ⇒ pump

⇒ it is probed by a fs pulse, which acts as a streak camera providing timing information

Timing information extracted by observing final electron momenta @ the detector. Attosecond streak camera:

$$\vec{p}_f = \vec{p}_0 - e\vec{A}(\tau)$$

$\vec{p}_0$: probe-free; $\vec{A}(\tau)$: timing information

where $\vec{A}(t)$ is the vector potential of the

Ultra-short pulse: $A(t) = A_0 f(t) \sin(\omega t + \phi)$

"Attosecond streak camera" is an approximation

Assumptions ① dipole approximation (neglects spatial dependence in the E-field: $E = \frac{-\partial A(t)}{\partial t}$

② SFA - Strong Field Approximation - neglects Coulomb field after ionization

SFA ⇒ subject of next lecture

Recap ⇒ Experimental "toolbox" in as science

① Ultrashort laser pulse ~ FWHM 5 fs

② Attosecond pulse ~ FWHM 100 - 200 as

Need Ultrashort laser pulse to create an attosecond pulse via the process KNOWN as High Harmonic Generation (HHG)

2 Types of pulses have different physics when they interact w. an atom / molecule due to different frequencies / intensities.

① Ultra-short (fs) pulse ⇒ produced by Ti-sapphire lasers ~ typical $\lambda \sim 800\,nm$

ω (in a.u) $= \frac{45.56}{800} = 0.057$ au $= \hbar\omega$

much slower than the frequency of electron motion inside atoms; $\omega << \omega_e$, where $\omega_e \sim O(1)$ au

⇒ From the view-point of bound-electron E-field changes slowly

I_p (Helium) $= 0.9$ au $>> \hbar\omega$ ⇒ cannot have single photon ionization!

electron has to absorb $\frac{I_p}{\hbar\omega} = 16$ photons!

Two regimes believed to exist

1) Tunnel ionization
2) Multi-photon ionization

} separated by Keldysh parameter, $\gamma = \omega\frac{\sqrt{2I_p}}{F}$

⇒ historically perturbation theory was used → SFA introduced by Keldysh in '60s resolved the problem

Lecture 1 17.9.13

$\gamma \ll 1$ ⇒ Tunnel ionization ⇒ strong field, low frequency limit

$\gamma \gg 1$ ⇒ multi-photon ionization ⇒ weaker field, higher frequency limit

The theory needed to understand ionization with ultra-short pulses: strong Field Approx. (SFA)

Tunnel ionization is, by far, the most important concept ⇒ holds even in the $\gamma \sim 1$ regime where most experiment operate

$\gamma \ll 1$ adiabatic tunneling

$\gamma \sim 1$ non-adiabatic tunneling

⇒ Tunnel ionization is the first step in production of as pulses via the process known as HHG

As pulse – much weaker; much higher photon energy (Harmonic multiples of the generating ultrafast pulse – hence HHG)