IV- Multiphoton Processes/Strong-Field and Attosecond Physics

Outline

- **Overview**
  - **First systematic studies**
  - **How to obtain an intense laser field**
  - **How strong is „strong“?**
  - **Attosecond physics**
  - **Useful definitions**
  - **Atomic units**
- **Examples of strong-field phenomena**
  - **Above-threshold ionization**
  - **High-order harmonic generation**
  - **Nonsequential double ionization**
- **Main strong-field approaches**

1.(a) - First systematic studies

- Maria Goeppert-Mayer (PhD thesis)

Two-photon absorption, Ann. Der Physik 9, 273 (1931) (theoretical prediction)

Since this process is orders of magnitude weaker than one-photon absorption, it could only be observed experimentally in the 1960s, with the advent of lasers.

- Experimental verification
  - Second harmonic generation

    P. A. Franken, A. E. Hill, C. W. Peters and G. Weinreich, PRL 7, 118 (1961)
    W. Kaiser, C. G.B. Garret, PRL 7, 229 (1961)
  - $6S_{1/2} \longrightarrow 9D_{3/2}$ excitation in Cs by a ruby laser

    I.D. Abella, PRL 9, 453 (1962)

1.(b) – How to obtain an intense laser field

"Traditional" methods (Q- switching, mode locking)

Key idea: concentrate the pump energy in a very short time duration

➢ increase in peak power

- 1961- Q switching:

(proposed by Gordon Gould in 1958; independently discovered and demonstrated in 1961/1962 by R.W. Hellwarth and F.J. McClung)

The quality factor of the optical resonator in a laser is degraded during the pumping. The gain can build up to a very large value and does not exceed the laser-oscillation threshold
When the inversion reaches its peak, Q is restored (pulse laser)

- 1965 - Mode locking:

1.(b) – How to obtain an intense laser field

"Traditional" methods (Q- switching, mode locking)

- 1965 - Mode locking:

In general: the laser modes in a cavity will oscillate independently (the individual phases of the waves in each mode are not fixed)

Mode locking: each mode operates with a fixed phase between itself and the other modes

➢Constructive interference

➢Intense burst of light

	Peak power	Focused intensity
Free-running lasers	kW	10^9W/cm^2
Q-switching	MW	10^{12}W/cm^2
Mode-locking	GW	10^{13}W/cm^2

Details: H. Haken, "*Laser-light dynamics*", Siegman, "*Lasers*", Yariv, "*Quantum Electronics*"

1.(b) – How to obtain an intense laser field

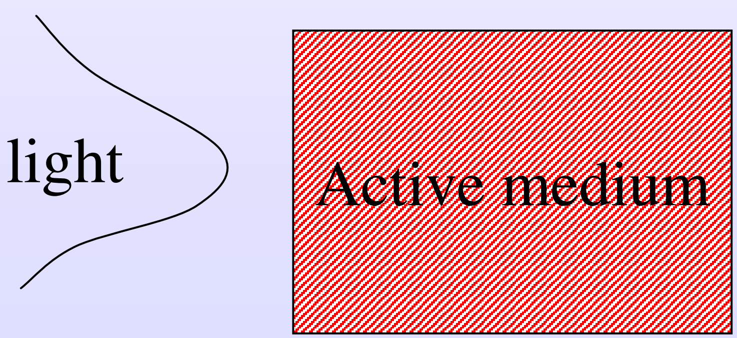
Chirped pulse amplification

D. Strickland and G. Mourou, Opt. Comm. 56, 219 (1985)

- Increase in intensities of 3 to 4 orders of magnitude
- Intensities up to the range of 10^{18}W/cm^2 were reached (nowadays over 10^{22} W/cm^2)

Main problem to achieve such intensities: self focusing

$$n = n_0 + n_1 I + n_2 I^2 + \ldots$$



The laser-beam profile varies with x,y,z

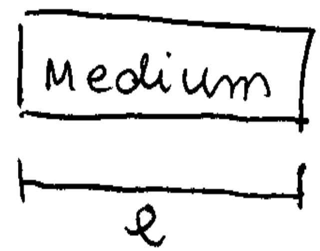
- ➢Center and edges have different refraction indices
- ➢Material behaves as a lense

This leads to distortions in the pulse and destroys the material

Overview

Nonlinear phase retardation ("B integral")

$$B = \frac{2\pi}{\lambda} \int_0^{\ell} n_2 I(z)\, dz$$



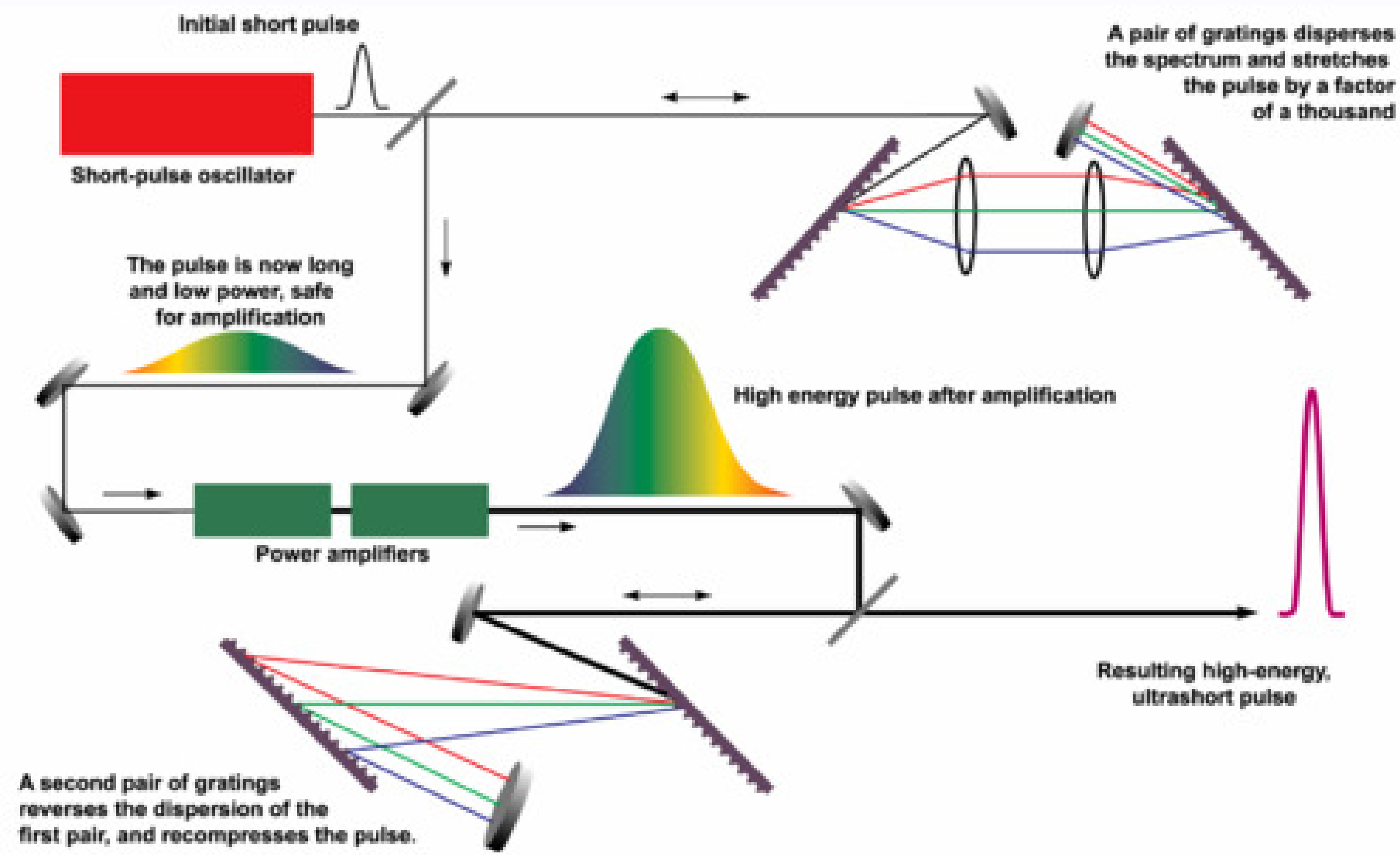
λ = light wavelength

- Related to the number of waves of nonlinear phase shifts accumulated when traversing the medium
- Spatial inhomogenities increase exponentially with B

➢ B has to be kept to a minimum

Solution: Chirped-Pulse Amplification

Chirped pulse amplification



Different optical ways for different frequencies due to the diffraction grating

- "Blue" and "red" components are separated
- Introduces a "chirp" in the pulse (hence the name) as some frequencies propagate more quickly than others

Important: At the end of the process the spectral distribution $\Delta\lambda/\lambda$ of the pulse should remain the same

Ideal pulse: close to the diffraction limit, as short as the initial pulse and temporally clean

Details:

- Input pulse: $I<10^{12}W/cm^2$ (pre-plasma formation intensity)
- Stretcher: stretching 10000 – 12000

  has a frequency dependent phase function $\beta(\omega)$

  Positively chirped: "red" earlier than "blue"

  Negatively chirped: "blue" earlier than "red"

- Compressor: chirp must be compensated

  Ideally: $\beta_{\text{stretcher}}(\omega) = -\beta_{\text{compressor}}(\omega)$

  In reality: residual chirp must be reduced to a minimum

1.(c) – How strong is “strong”?

Number of photons is so large that the field can be treated classically

Comparison with “normal” light sources

- He-Ne laser: Power ~ 1mW
- Lasers used in entertainment, etc: Intensity~1W/cm^2 -1KW/cm^2
- Sunny day (sunlight reaching the Earth): Intensity~0.135W/cm^2

Typical frequency: ω~0.057 a.u. (λ ~ 800nm)

Titanium –sapphire

Typical duration: 2.6*fs*

Matter in strong laser fields ($I > 10^{13}$W/cm^2):

- Starting point: time-dependent Schrödinger equation

$$\cancel{i\partial_t|\psi(t)\rangle = \left[-\frac{\Delta}{2} + V + \mathbf{z.E}(t)\right]|\psi(t)\rangle}$$

Atomic Hamiltonian

Coupling with field

- Weak fields ($I \ll 10^{13}$ W/cm^2): **laser fields << atomic binding forces**
  Field can be treated as a perturbation

- Strong fields:
  - ❖ $I \sim 10^{13}$ W/cm^2:
    Stark shifts of the atomic bound states ~ photon energies:
    First discrepancies from perturbation theory
  - ❖ $I \sim 10^{16}$ W/cm^2:
    laser fields ~ atomic binding forces
    Perturbation theory breaks down !
  - ❖ $I \sim 10^{18}$ W/cm^2: **ponderomotive energy (U_p=I/(4ω^2)) ~ e^- rest mass**
    Relativistic treatment is necessary

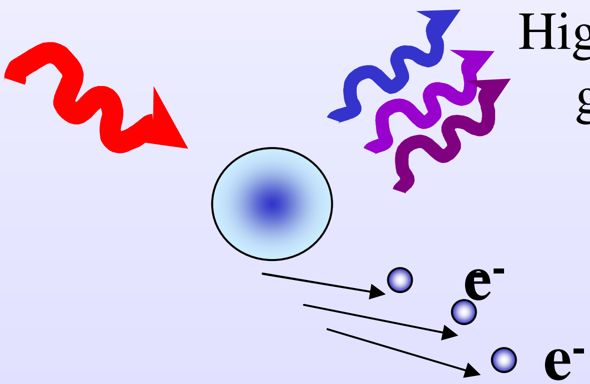
1.(d) – Perspectives: “Attosecond physics”

Intense-field laser physics deals also with ultrafast time scales

Typical cycle of an intense field: ~2.6fs

- Typical time scales of strong-field phenomena: a fraction of a field cycle (hundreds of atto (10^{-18}) seconds)

Attosecond processes

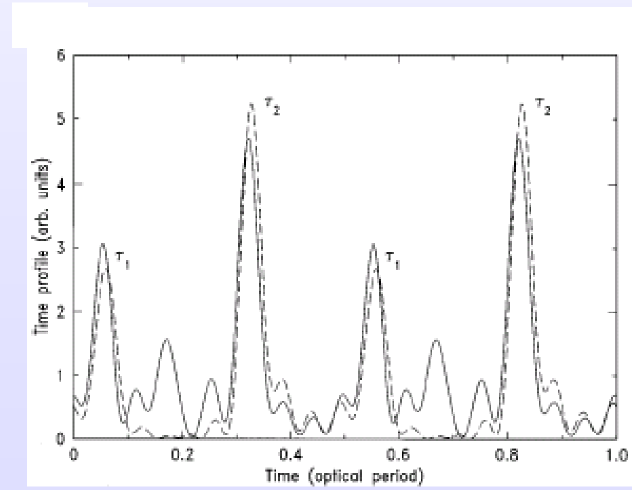


High-harmonic generation

Nonsequential double & multiple ionization

Attosecond pulses

(from groups of high-order harmonics)



P. Antoine, A. L‘Huillier, and M. Lewenstein, PRL **77**, 1234 (1996)

1- Overview

Powerful tools for *dynamic* measurements and control at atomic/molecular scales

1 *attosecond* ~ typical atomic dimensions (10^{-10}m)/speed of light (3×10^8 m/s)

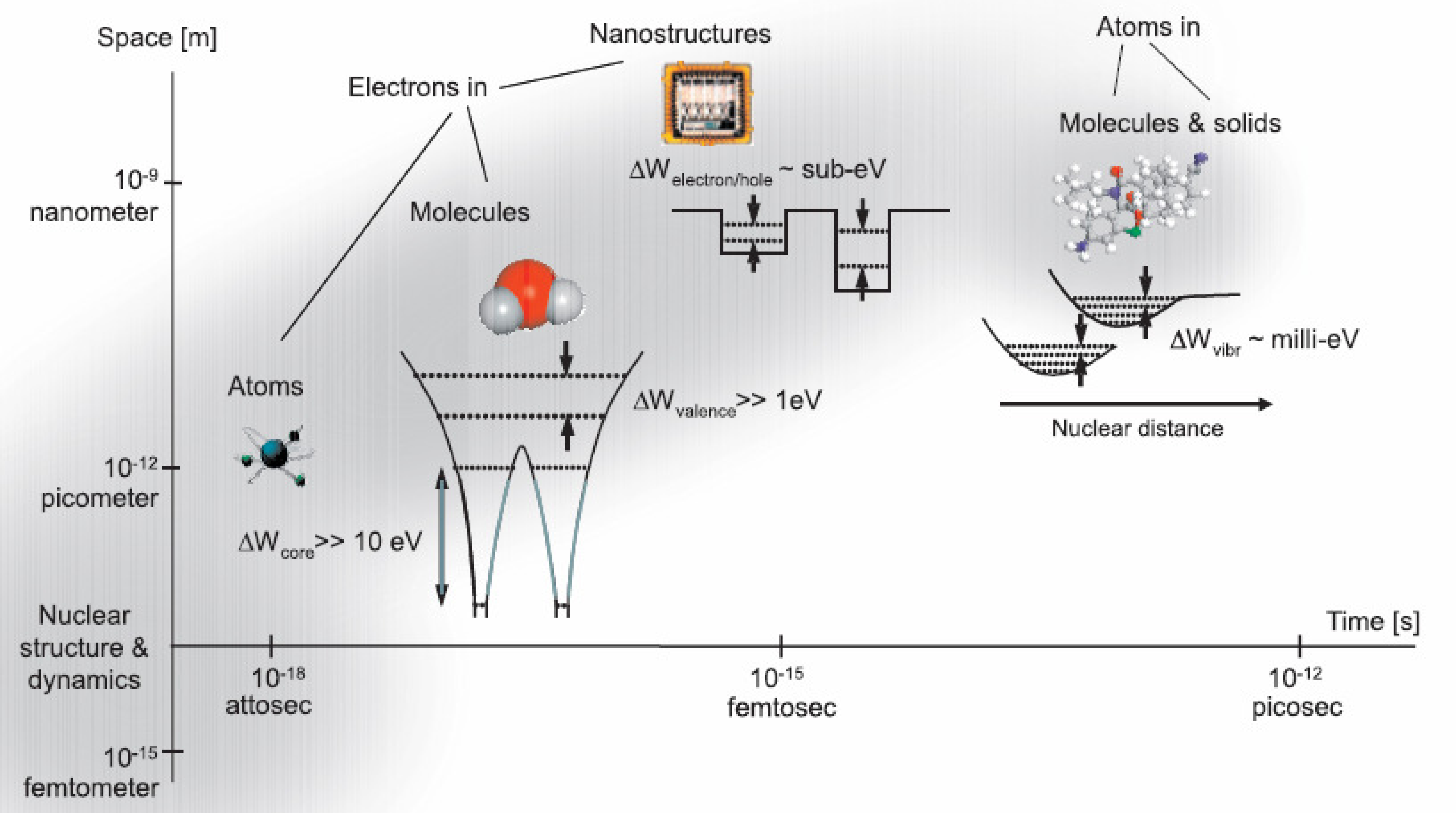
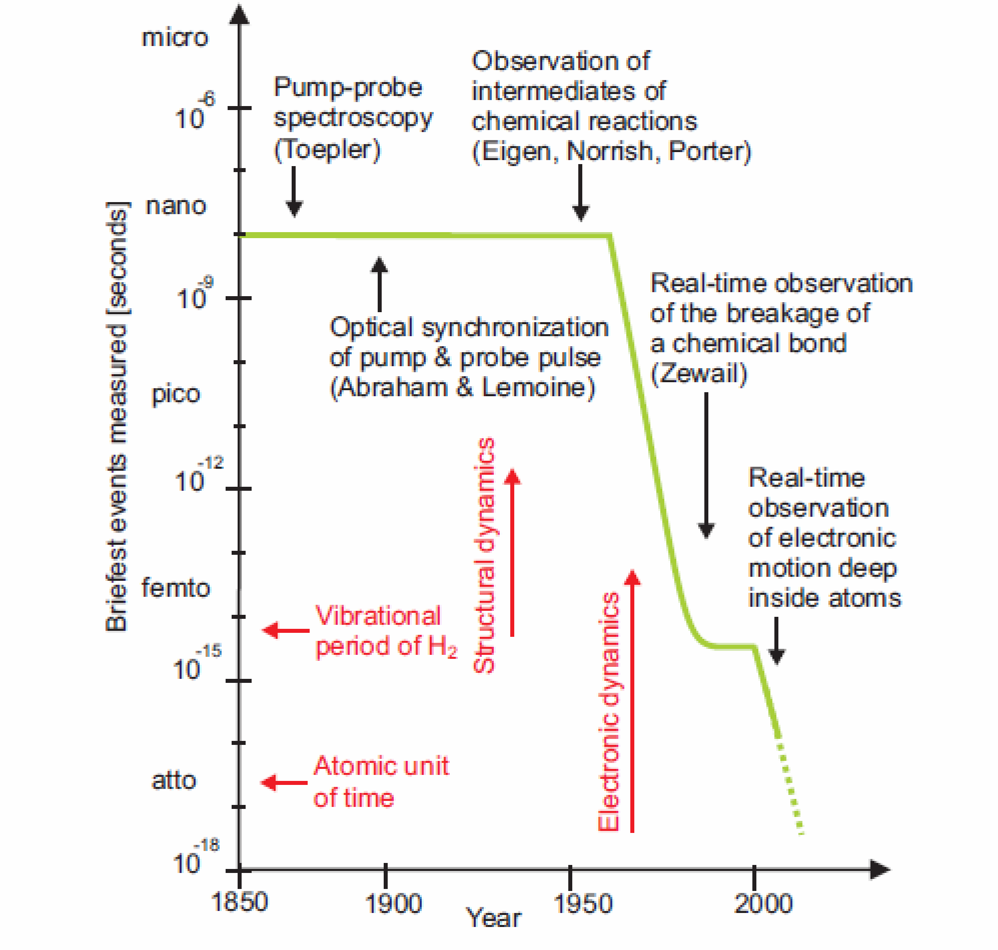


Fig. 1 in "Attosecond Physics", F. Krausz and M. Ivanov, Rev. Mod. Phys. 81, 163 (2009)

1- Overview



Example: Reconstruction of ultrafast changes in molecules, control of e^- motion in bound systems, etc.

Fig.6 in "Attosecond Physics", F. Krausz and M. Ivanov, Rev. Mod. Phys. 81, 163 (2009)

1.(e) – Useful definitions

a) Ponderomotive energy: average kinetic energy transfer from the laser field to an electron

$$U_p = \frac{e\langle A^2(t)\rangle_t}{2m},$$ where $\langle \ \rangle_t$ = temporal average

- Occurs in several physical expressions in the context of matter + intense laser fields
- Can be associated with the A^2 term occurring in the interaction Hamiltonian in the velocity gauge

$$H_{int}(t) = \frac{[\mathbf{p} - e\mathbf{A}(t)]^2}{2m} = \frac{p^2}{2m} - \frac{e\mathbf{p}\cdot\mathbf{A}(t)}{m} + \frac{e^2A^2(t)}{2m}$$

For weak fields this term is neglected

1- Overview

- Monochromatic fields: $U_p = \frac{eE_0^2}{4m\omega^2}$

E_0^2 : driving-field intensity
ω :driving-field frequency

- Relativistic regime: $U_p \sim m_0c^2$

b) Electron excursion amplitude:

Maximal amplitude with which an electron oscillates in a laser field

$$\alpha = \frac{E_0}{\omega^2}$$ (atomic units, monochromatic field)

Please note: The above-stated expressions show that not only the intensity, but also the frequency of the laser field plays an important role. They delimit different physical regimes.

b) Multiphoton regime/tunneling regime

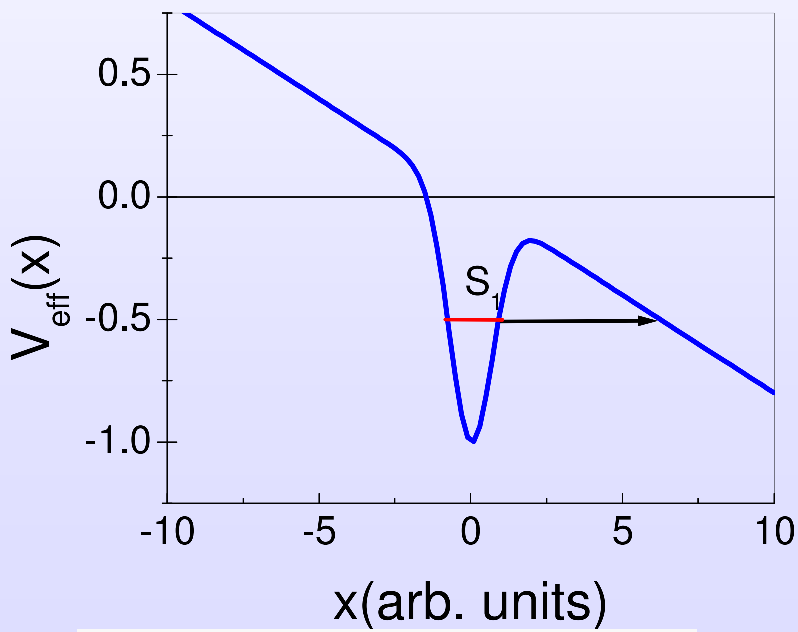
Determined by the Keldysh parameter

(L. V. Keldysh, Sov. Phys. JETP 20, 1307 (1965))

$$\gamma = \sqrt{\frac{I_p}{2U_p}}$$

I_p = ionization potential
U_p = ponderomotive energy

$\gamma < 1 \Rightarrow$ tunneling regime

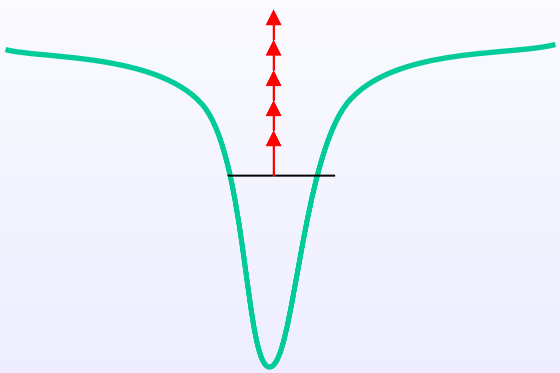


$$V_{eff}(r) = V(r) - \mathbf{r} \cdot \mathbf{E}(t)$$

- Atom is mainly a source term (internal structure is not very important)
- The e^- has enough time to tunnel through the potential barrier
- $\gamma << 1$ (very low frequencies): to a good approximation the field is taken to be static

1- Overview

$\gamma > 1 \Rightarrow$ multiphoton regime



- The electron reaches the continuum through a multiphoton transition
- Multiphoton transitions, and therefore atom-laser resonances, play a role
- $\gamma << 1$: the atom is being driven too quickly by the field for tunneling to occur

Please note: the Keldysh parameter may be defined with respect to any bound state of the atom in question. Normally one considers the ionization potential as it is related to the outer shell e^-s.

c) Atomic units: handy, since we are dealing with the ultra strong/ultrafast regime

- Length: 1 a.u = 0.53 x 10^{-10} m (Bohr radius)
- Charge: 1 a.u. = 1.6 x 10^{-19}C (electron charge)

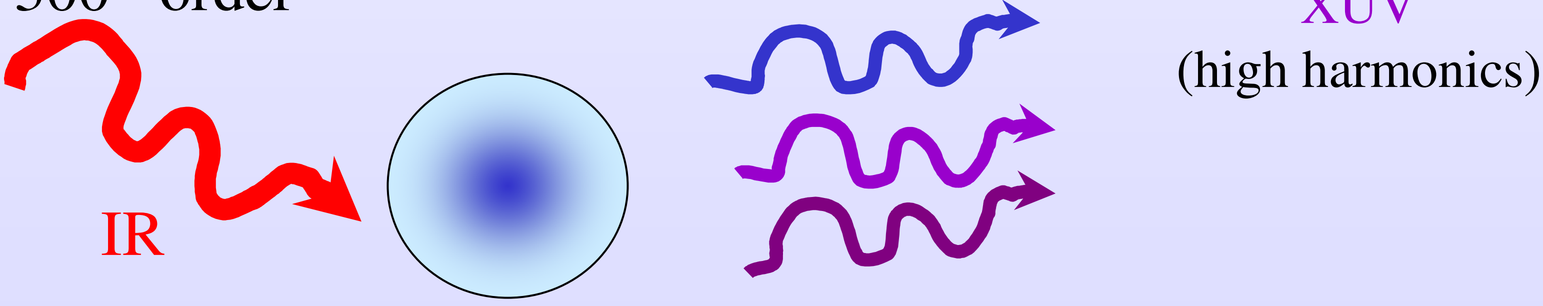
1- Overview

- Energy: 1 a.u.= $e^2/r_0 = 27.2$ eV
- Intensity: 1 a.u. = $\epsilon_0 c e^2/(2r_0^2) = 3.51 \times 10^{16}$W/cm^2
- Mass: 1 a.u. = 9.1×10^{-31} kg (electron mass)
- Frequency: 1 a.u. = 4.13×10^{16} s^{-1}
- The Planck constant is one in atomic units

2- Examples of strong-field phenomena

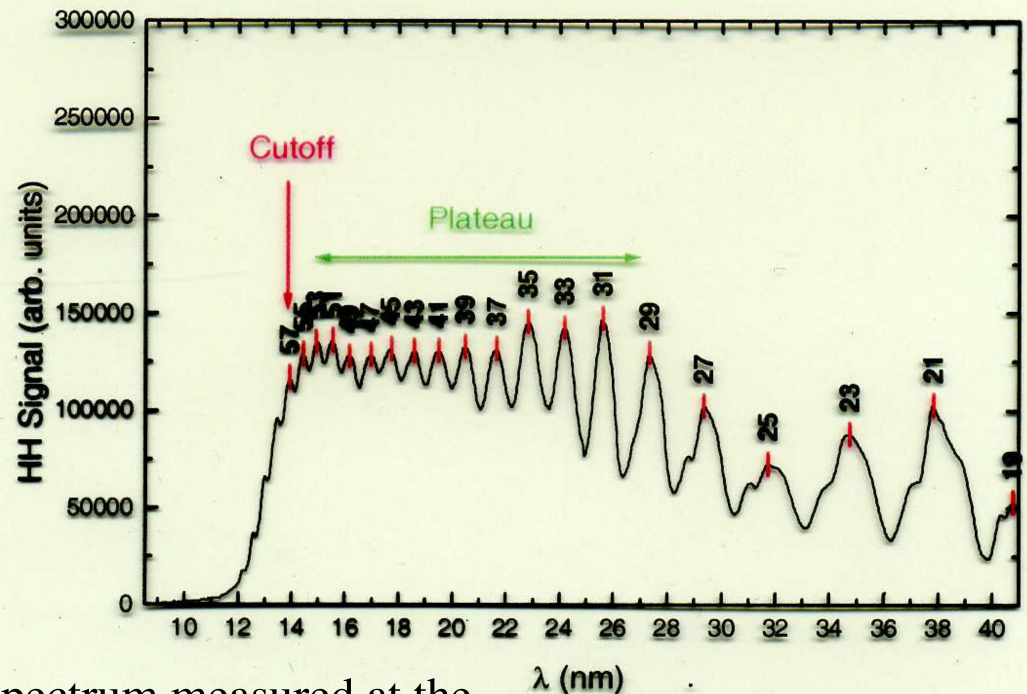
2.(a) – High-order harmonic generation

The highly nonlinear response of an atom to a strong laser field 10^{13}W/cm^2<I<10^{15}W/cm^2, emitting harmonics up to almost the 300th order



2- Strong-field phenomena

2.(a) – High-order harmonic generation



Spectrum measured at the
Max Born Institut, Berlin

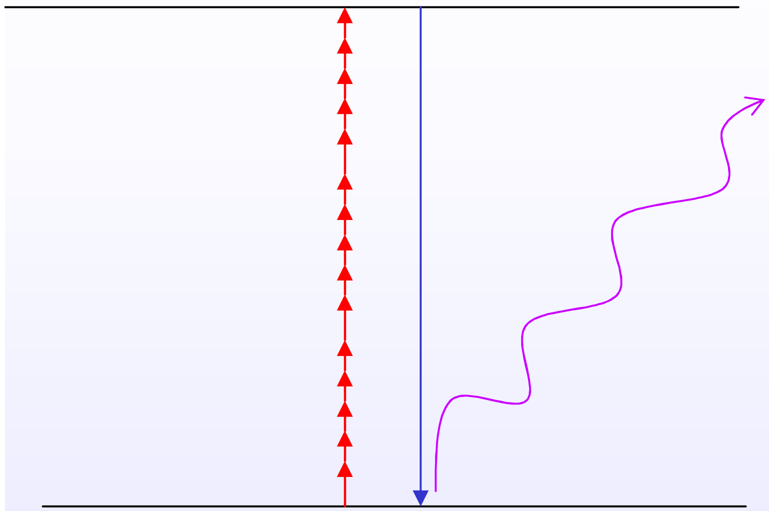
Features: plateau + cutoff

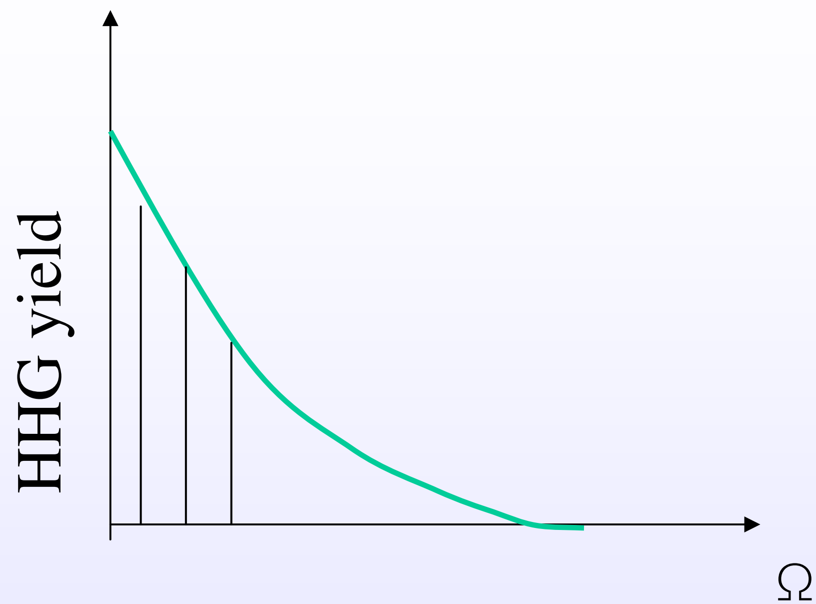
Cutoff law at $\Omega_{max} = I_p + 3.17\ U_p$

Several features in contradiction with high-order perturbation theory

2- Strong-field phenomena

"Perturbative" expectations





In perturbation theory, the field excites or de-excites one atom but does not change its structure

Transition probability proportional to I^N (N= number of absorbed photons)

➢No plateau+ cutoff

First measurements:

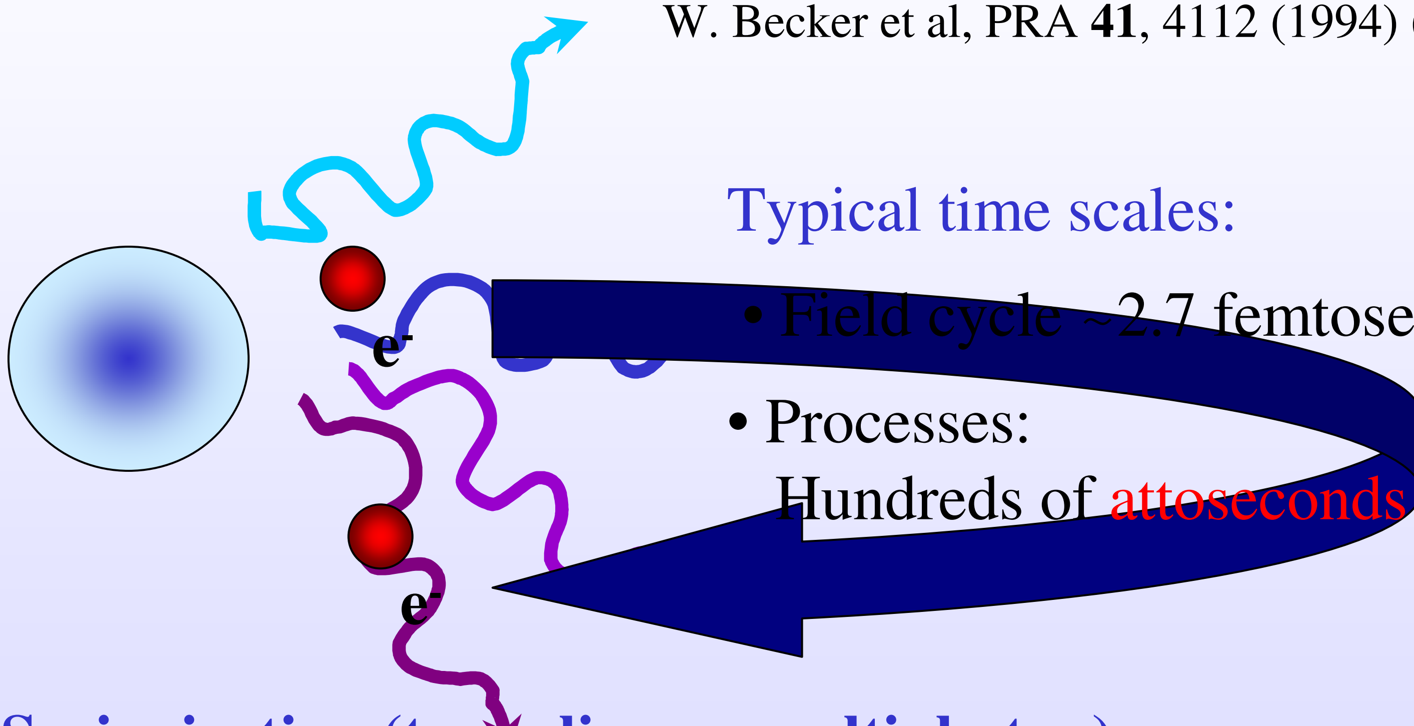
University of Illinois, Chicago: A. McPherson et al JOSA B 4, 595 (1987)

Saclay, France: M Ferray et al, J. Phys. B, 21, L31 (1988); X. F. Li et al, PRA 39, 5751 (1988)

2- Strong-Field Phenomena

Paradigm: laser-induced recombination

- Classical formulation: P.B. Corkum, PRL **71**, 1994 (1993) (1163 citations).
- Quantum mechanical formulation: M. Lewenstein et al, PRA **49**, 2117 (1994) (598 citations);
  W. Becker et al, PRA **41**, 4112 (1994) (130 citations).



Typical time scales:

- Field cycle ~2.7 femtoseconds (10^{-15}s)
- Processes:
  Hundreds of attoseconds (10^{-18}s)

S_1: ionization (tunneling or multiphoton)

S_2: propagation

S_3: High-order harmonic generation (HHG): recombination of e^-

2- Strong-Field Phenomena

Predictions:

- Plateau
- Harmonic energy: $\Omega = I_p + E_{kin}(t_1,t_0)$
- Cutoff: Maximal $E_{kin}(t_1,t_0)$
- Monochromatic fields: $\Omega_{max} = I_p + 3.17U_p$

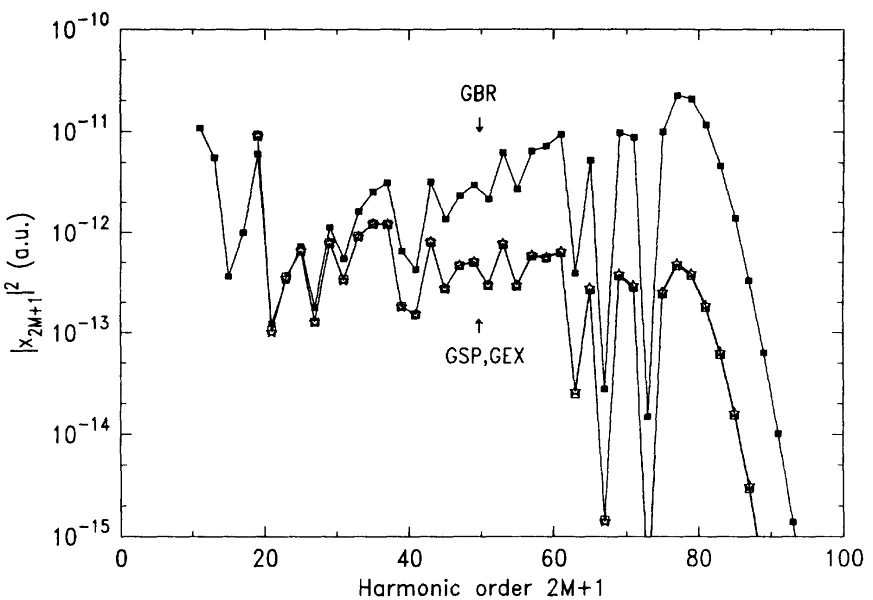


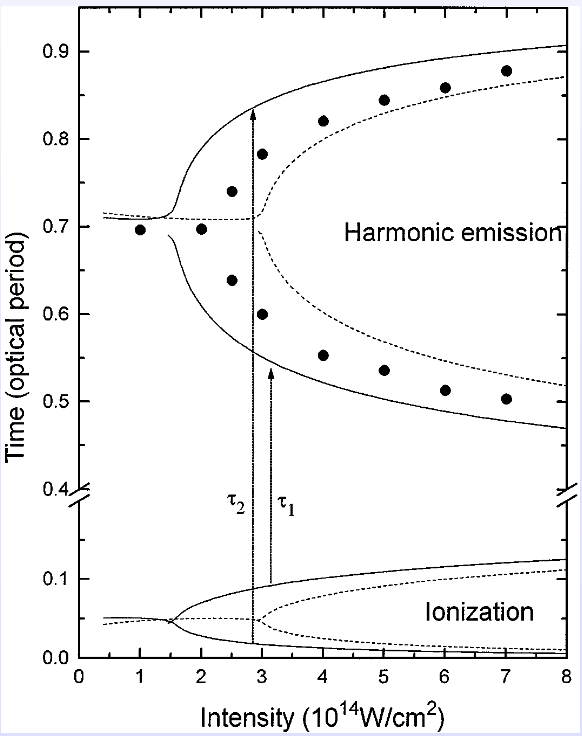
FIG. 2. Comparison of harmonic spectra obtained with GEX (open squares), GSP (stars), and GBR (black squares) methods; $I_p = 13.6$, $U_p = 20$, $\alpha = 2I_p$.

From M. Lewenstein et al, PRA **49**, 2117 (1994)

2- Strong-Field Phenomena

Consequences:

- Cutoff energies can be predicted by classical computations
- For more complex field there may be more than one cutoff, related to local maxima of $E_{kin}(t_0,t_1)$.
- Most contributions to high-order harmonic generation within a field cycle occur at well-defined times, i.e., when the e^- comes back:



➢ This is in contrast with the predictions of perturbation theory, for which there are no preferential times for HHG

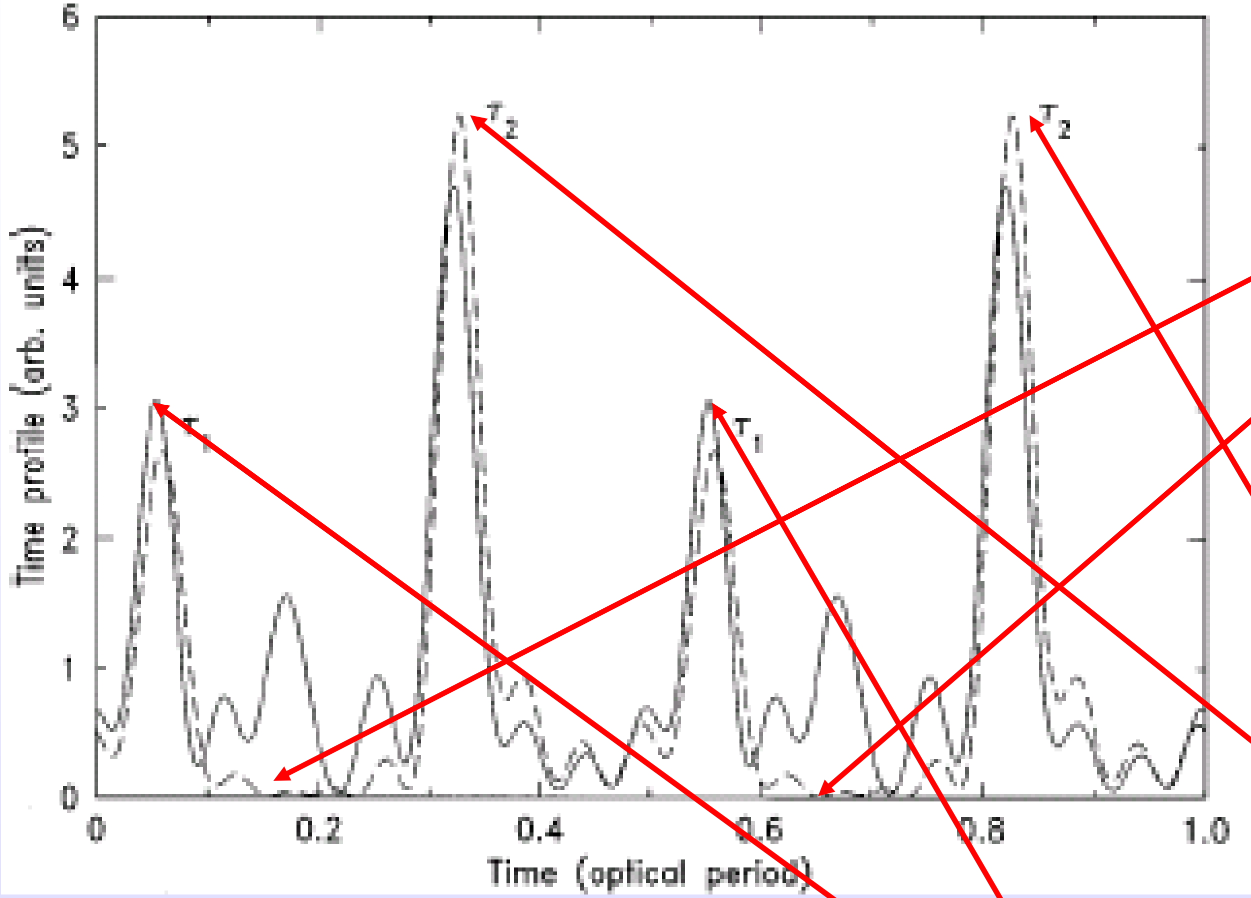
➢ One may use this fact to produce attosecond pulses.

- Quantum mechanically, there may be more than one possible path for the e^- to return, which will then interfere.

From P. Antoine, A. L'Huillier, and M. Lewenstein, PRL **77**, 1234 (1996)

2- Strong-field phenomena

- Electron return times



Short orbit suppressed through propagation

Longer orbit

Shorter orbit

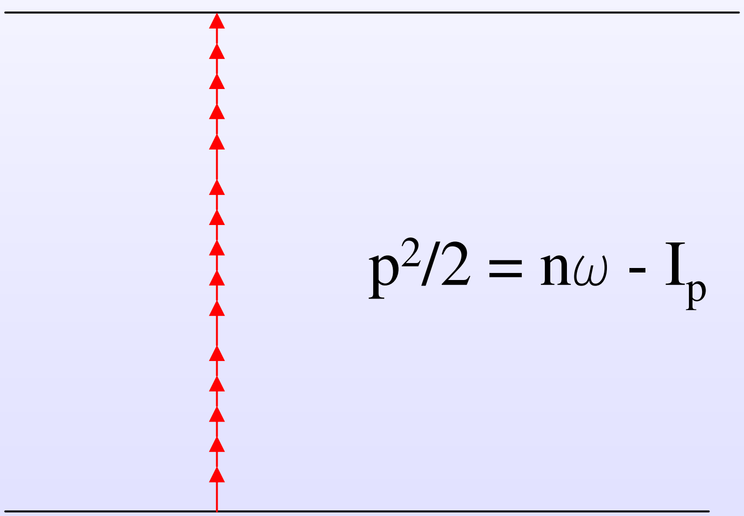
Attosecond pulses are generated by grouping high harmonics and are very important for metrology

2- Strong-Field Phenomena

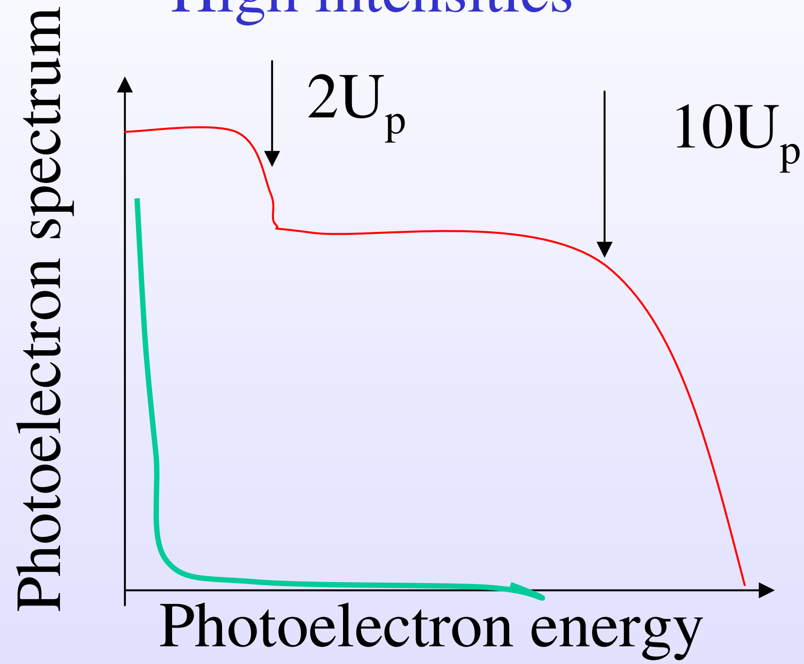
2.(b) – Above-threshold ionization

The atom absorbs photons in excess, i.e., more than the necessary amount for it to ionize.

Low/moderate intensities

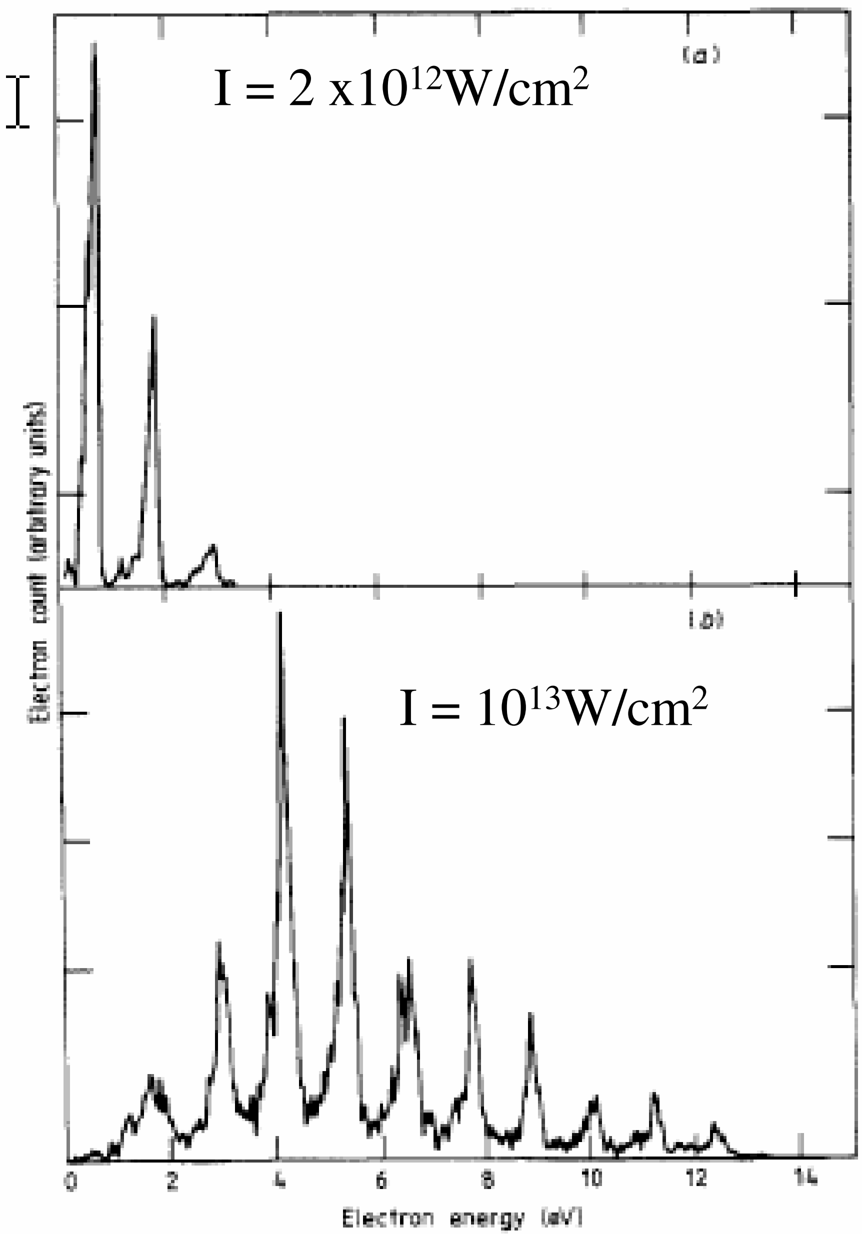


High intensities



- First measurements: CEA Saclay: Agostini et al, PRL 42, 1127 (1979)
- This was the first clear evidence that perturbation theory breaks down:
  - Peak intensities do not follow the predictions of perturbation theory
  - Low-energy peaks are reduced in magnitude.
- Explanation: the ionization threshold is effectively shifted by the field

2- Strong-field phenomena



From G. Petite, P. Agostini and H. G. Muller, J. Phys. B 21,4097 (1988)

2- Strong-field phenomena

ATI Plateau

First measurements: G.G. Paulus, Phys. Rev. Lett. 72, 2851 (1994) (150 citations)

We present photoelectron energy spectra for the rare gas atoms in strong 40 fs, 630 nm laser pulses. A new property in the above threshold ionization distribution is described, namely, a plateau. Numerical calculations using one- and three-dimensional models suggest that at least in part this is a one-electron effect. All rare gas atoms investigated show similar behavior, indicating that the plateau in above threshold ionization is a universal phenomenon. We discuss a simple mechanism possibly responsible for the plateau.

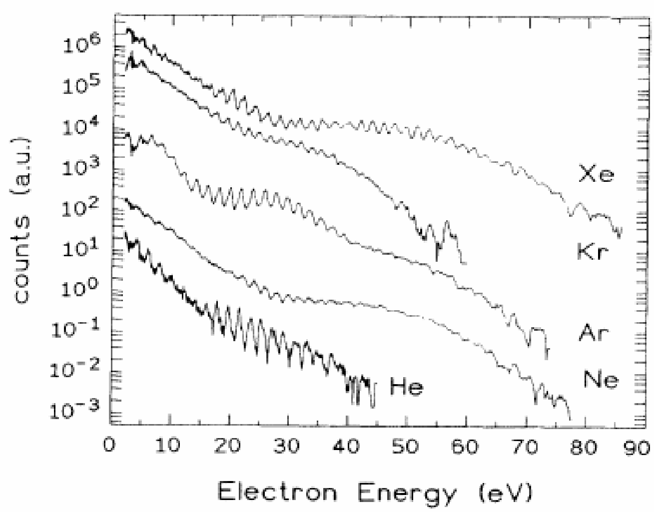


FIG. 1. ATI spectra of all rare gases. The intensity was 3×10^{14} W/cm^2 for He and 2×10^{14} W/cm^2 for all the others. (The curves are separated in the vertical direction.)

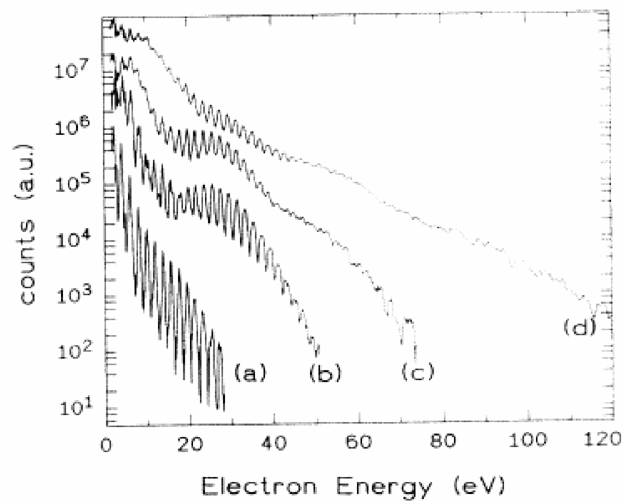
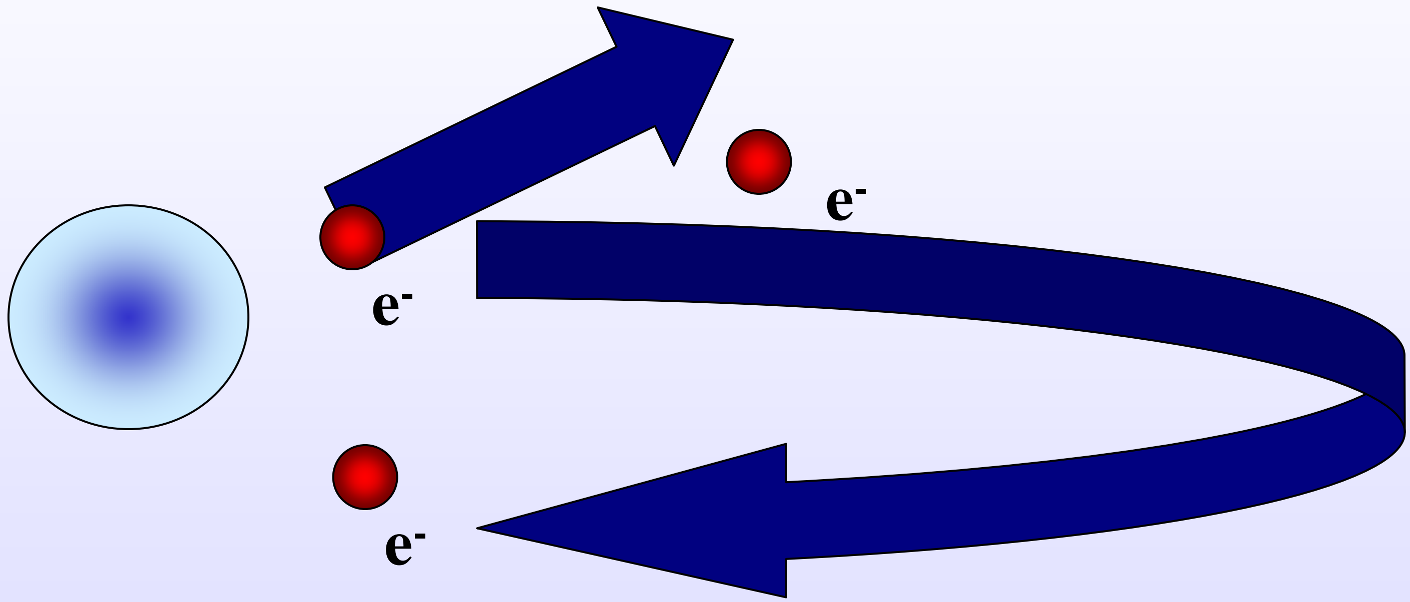


FIG. 2. ATI spectra from Ar with 40 fs, 630 nm pulses at intensities of 6×10^{13} W/cm^2 (*a*), 1.2×10^{14} W/cm^2 (*b*), 2.4×10^{14} W/cm^2 (*c*), and 4.4×10^{14} W/cm^2 (*d*) (the curves are separated slightly in the vertical direction for visual convenience).

2- Strong-field phenomena

Explanation: three-step model

- Classical formulation: G. G. Paulus et al, Phys Rev A 52, 4043 (1995)
- Quantum mechanical formulation: See, e.g., A. Lohr et al, Phys Rev A 55, R4003 (1997); W. Becker et al, Phys Rev A 56, 645 (1997)



S_1: ionization (tunneling or multiphoton)
S_2: propagation (if it reaches the detector: direct ATI)
S_3: elastic collision (rescattered ATI)

Cutoff: Maximal kinetic energy

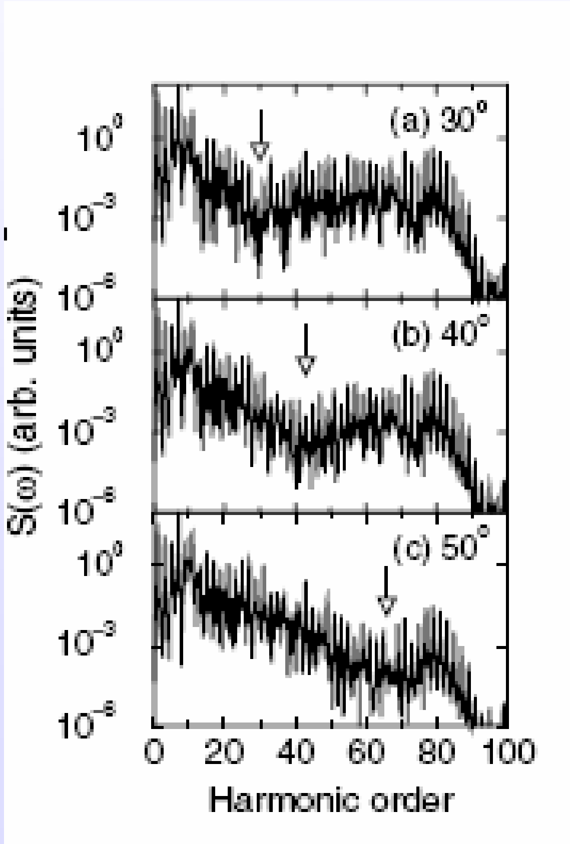
2- Strong-Field Phenomena

Recent applications: ultrafast imaging of molecules

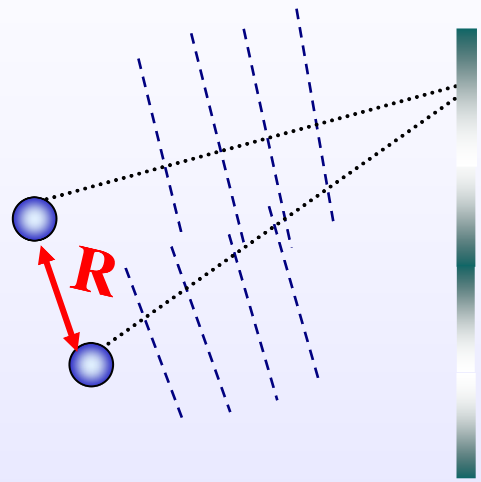
M. Lein, N. Hay, R. Velotta, J.P. Marangos and P.L. Knight, PRL **88**, 183902 (2002);

PRA **66**, 023805 (2002)

(H_2^+, TDSE computation)



Analogy: double-slit experiment

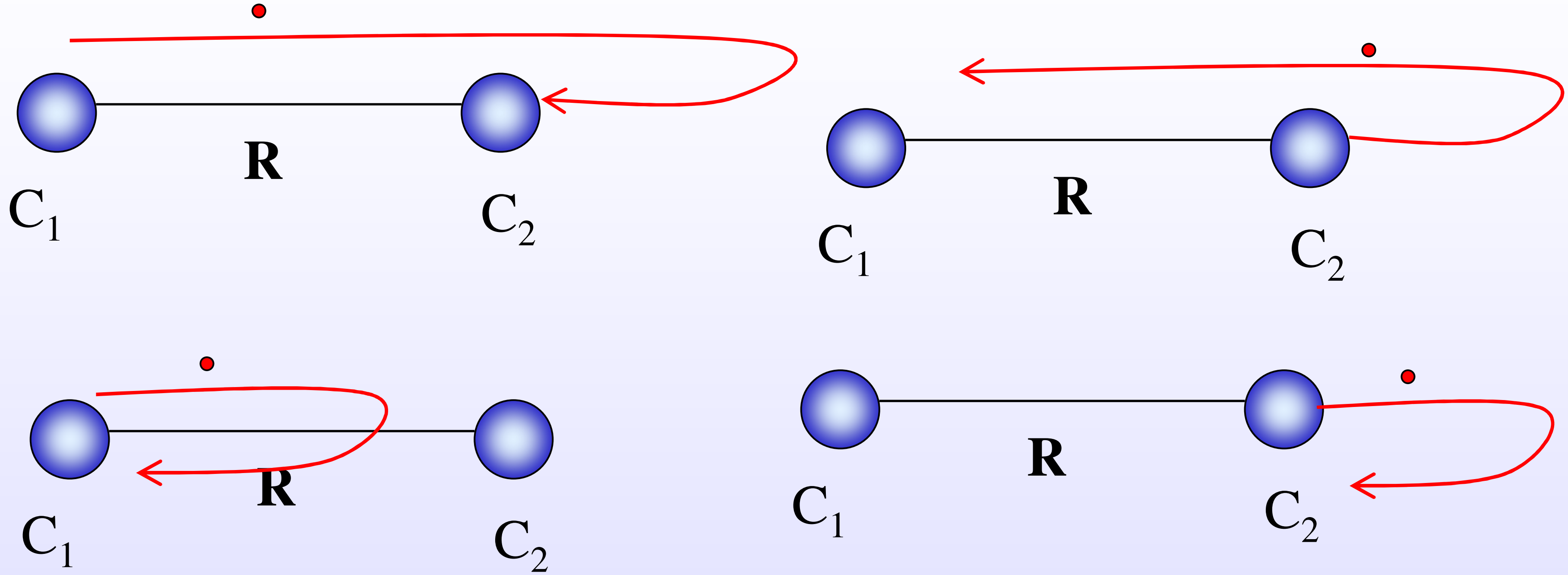


Maxima/Minima due to interference of HHG/ATI at different centers

2- Strong-Field Phenomena

Quantum Interference

Simplest case: diatomic molecules



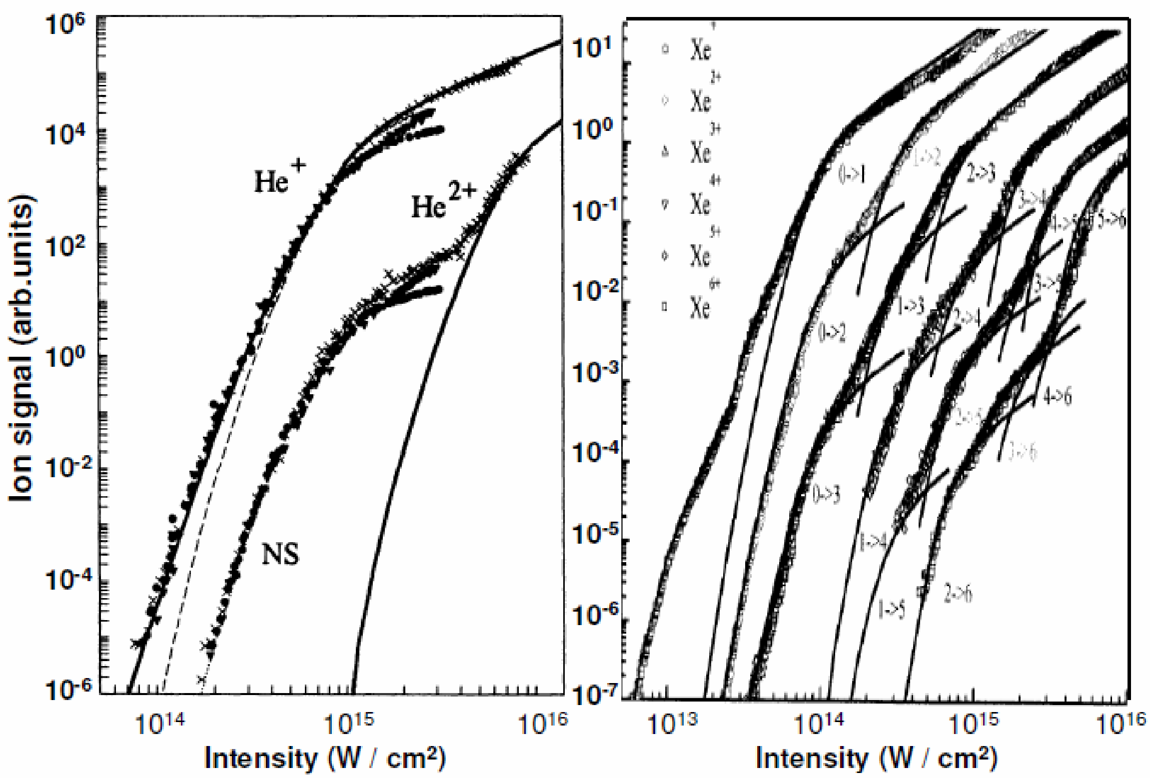
Apart from that: shape of the molecular wavefunctions also influence the spectra

2c- Nonsequential double ionization

- Electron-electron correlation effects are huge

(Review: C.F.M.F. and X. Liu, in press; a link will be put from the course website)

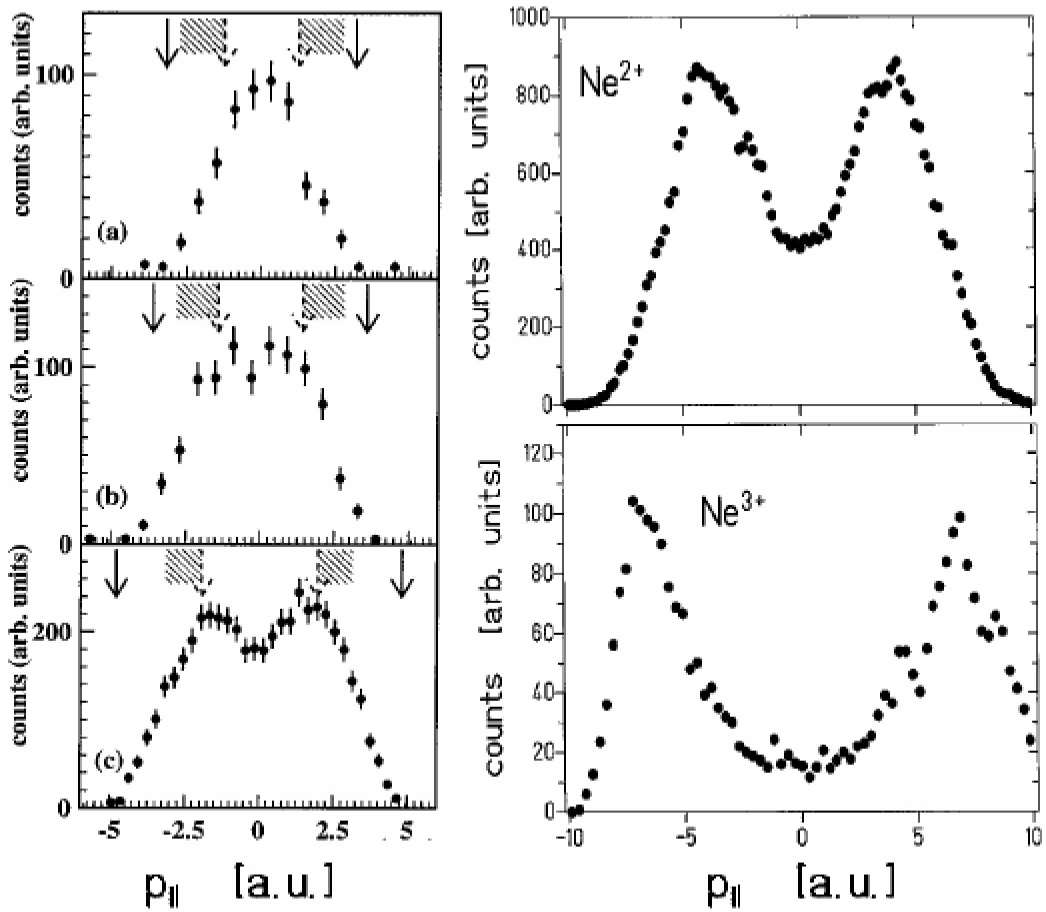
  - First evidence: A. l'Huillier, et al, . *Phys. Rev. A* 27, 2503-2512 (**1983**): Multiphoton ionization of Xenon atoms
  - „The knee“: Double ionization yield deviated in orders of magnitude from the predictions of sequential models, in which one electron is ripped after each other.



From Walker, B.; Sheehy, B.; DiMauro, et al *PRL* 73, 1227-1230 (**1994**); Larochelle, S.; Talebpour, A.; Chin, S. L., *J. Phys. B* 31, 1201-1214 (**1998**).

2c- Nonsequential double ionization

Ion momentum distributions: possible with the advent of the COLTRIMS (COLd Target Recoil Ion Momentum spectrometer) technique

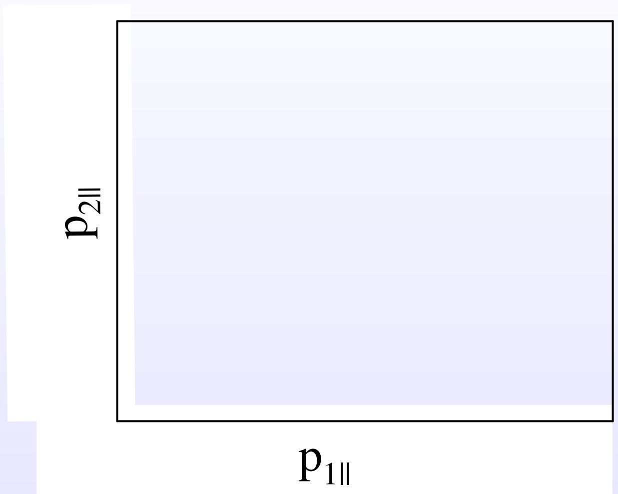


First measurements: double ionization of helium (Frankfurt: Weber, Th.;et al *Phys. Rev. Lett.* **2000**, 84, 443-446.) and neon (Heidelberg/Berlin: Moshammer, R.; et al, *Phys. Rev. Lett.* **2000**, 84, 447-450.).

2c- Nonsequential double ionization

Electron-momentum distributions

- Freiburg/Berlin group: PRL **84**(4), 447 (2000), PRL **87**(4), 043003(2001)
- Frankfurt/Marburg group: PRL **84** (4), 443 (2000); Nature **404**, 608 (2000)



System: Neon

Field: Linearly polarized light

- Frequency: $\omega = 0.057$ a.u.
- Intensity: $I = 10^{14} - 10^{15}\,\mathrm{W/cm^2}$

Doubly-humped differential e^- momentum distributions:

Maxima at $p_{\|} = \pm\, 2[U_p]^{1/2}$,

with $p_{\|}$ = momentum components parallel to the laser-field polarization

$U_p = I/(4\omega^2)$ = ponderomotive energy

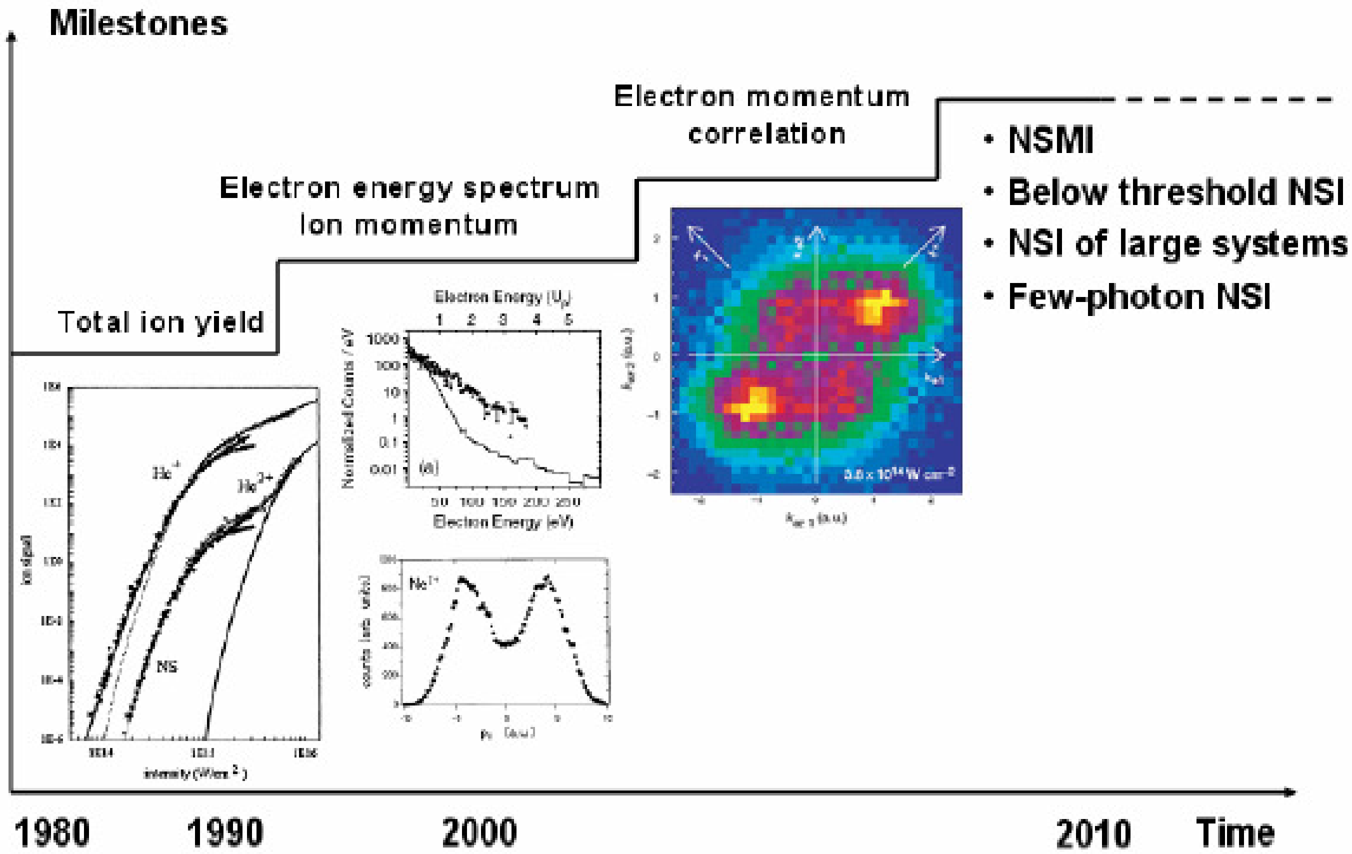
Can only be explained by a non-sequential physical mechanism:

S_1: tunneling ionization of first e^-

S_2: propagation of first e-

S_3: inelastic rescattering (NSDI)

2c- Nonsequential double ionization



Please note:

There may be more than one rescattering mechanism, depending on the species

Signature of the electron-electron interaction can be inferred from the distributions

2c- Nonsequential double ionization