Atoms in strong laser fields

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Lecture I: General framework

- What is a strong laser field?
- How to obtain a strong laser field?
- Examples of strong-field phenomena
- Perspectives

References

- M. Gavrila, Atoms in Strong Laser Fields, Academic Press, 1992
- M. D. Perry and G. Mourou, Science 264, 917 (1994)
- L.F. DiMauro and P. Agostini, Adv.At. Mol. Opt. Phys. 35, 79 (1995)
- M. Protopapas, C.H. Keitel and P. L. Knight, Rep. Prog. Phys. 60, 389 (1997)
- C.F.M.F, Ph.D. thesis (TU-Berlin, 1999)
- T. Brabec and F. Krausz, Rev. Mod. Phys. 72, 545 (2000)
- C.J. Joachain, et al, Adv.At. Mol. Opt. Phys. 42, 225 (2000)

How strong is a "strong laser field"?

Central physical quantity: time-dependent Schrödinger equation

- One active electron:

$$i\partial_t \left|\psi(t)\right\rangle = \left[-\frac{\Delta}{2} + V + \mathbf{H}_{int}(t)\right] \left|\psi(t)\right\rangle$$

- Two active electrons:

$$i\partial_t \left|\psi_{1,2}(t)\right\rangle = \left[\sum_{n=1}^{2} -\frac{\Delta_n}{2} + V_n + \mathbf{H}_{int}^{(n)}(t) + V_{12}\right] \left|\psi_{1,2}(t)\right\rangle$$

- $H_{int}(t) \equiv$ interaction with the field
- $V \equiv$ binding potential
- Weak fields: laser fields$\ll$atomic binding forces
- Strong fields:
  - $\mathrm{I}_0 \sim 10^{13}\mathrm{W/cm}^2$: Stark shifts of the bound states $\sim$ photon energies→ 1^{st} discrepancies from perturbation theory
  - $\mathrm{I}_0 \sim 10^{16}\mathrm{W/cm}^2$: field can not be treated as a perturbation
  - $\mathrm{I}_0 \sim 10^{18}\mathrm{W/cm}^2$: relativistic treatment necessary

Strongest lasers available

- Lawrence Livermore Laboratory (USA)-10^{21}W/cm^2
- University of Rochester (USA)
- Lund Laser Center (Sweden)
- Laboratoire d'Optique Apliquee (France)
- Max-Born-Institut, Berlin (Germany)

Typical parameters

- length: $\tau \sim (1-100) fs$
- frequency: $\omega \sim 0.057 a.u. (\lambda \sim 800 nm)$

Gauge-equivalent Hamiltonians

$$H_i(t) = i\partial_t A_{j\leftarrow i}(t) A_{j\leftarrow i}(t)^{-1} + A_{j\leftarrow i}(t) H_j(t) A_{j\leftarrow i}(t)^{-1}$$

- Length gauge

$$H_l(t) = -\frac{\Delta}{2} + V + z \cdot E(t)$$

$$A_{v\leftarrow l}(t) = e^{ib(t)z.}$$

- Velocity gauge

$$H_v(t) = \frac{1}{2}(-i\nabla - b(t)e_z)^2 + V$$

$$A_{v\leftarrow KH}(t) \; = e^{-ia(t)} e^{ic(t)p_z}$$

- Kramers-Henneberger frame

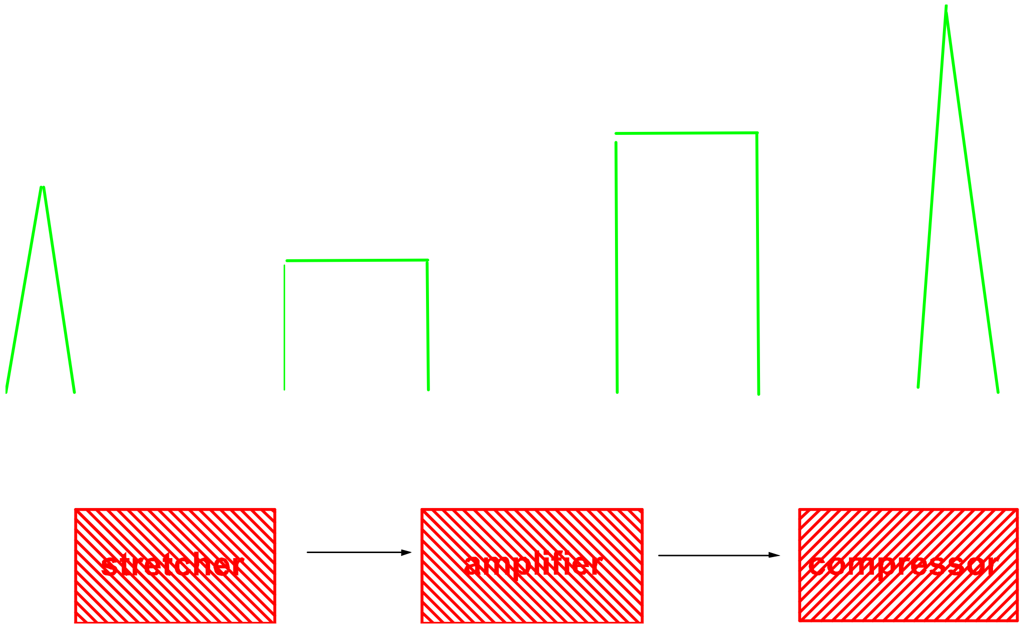
$$A_{l\leftarrow KH}(t) = e^{-ia(t)} e^{-ib(t)z} e^{ic(t)p_z}$$

$$H_{KH}(t) = -\frac{\Delta}{2} + V(\vec{x} - c(t)e_z)$$

- classical momentum transfer: $b\,(t) = \int\limits_0^t ds\, E\,(s)$
- classical displacement: $c\,(t) = \int\limits_0^t ds\, b\,(s)$
- classical energy transfer: $a\,(t) = \frac{1}{2} \int\limits_0^t ds\, b^2\,(s)$

How to obtain a strong laser field?

Chirped pulse amplification: Strickland and Mourou, Opt. Comm. 56, 219 (1985)

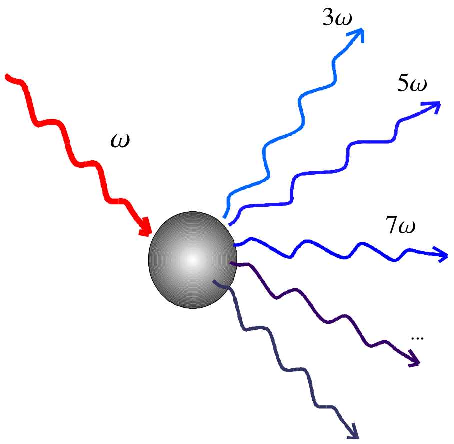


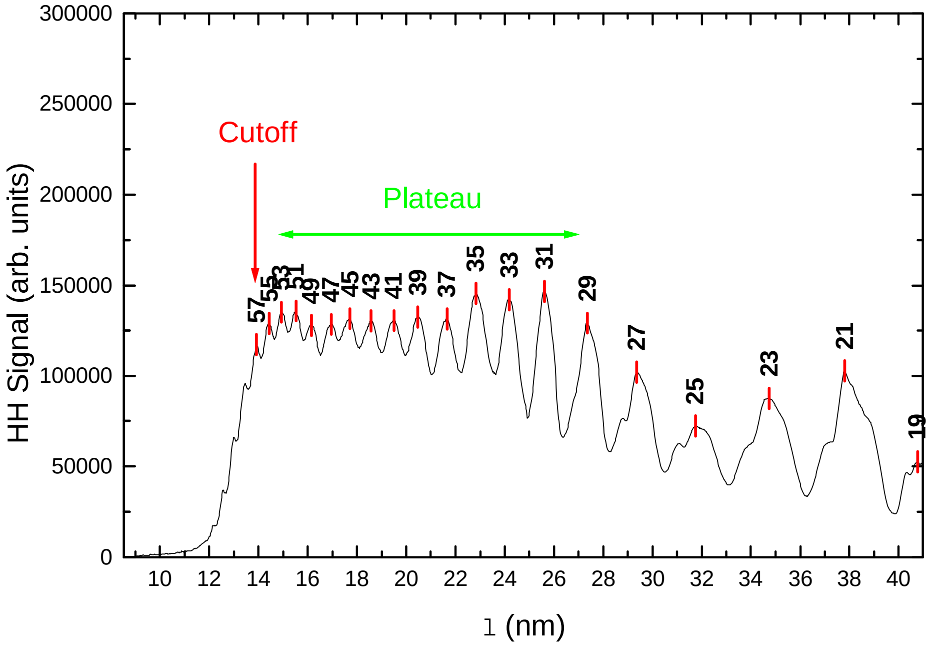
- Overcomes the problem of self-focusing
- Appropriate conditions:
  - Broadband amplifier
  - High-contrast pulses
  - Spectral distributions ($\Delta\lambda/\lambda$) remain the same

Typical strong-field phenomena

High-harmonic generation

- HHG: the highly nonlinear response of an atom to a strong laser field ($10^{13}/cm^2$ <I<$10^{15}/cm^2$)





- Spectra $\sim \left| \int\limits_0^\infty \exp[i\omega t]\ddot{x}(t)dt \right|^2$ $(\ddot{x}(t) \equiv \text{dipole acceleration})$
- Media: gases (atoms or small molecules)

First measurements:

- University of Illinois, Chicago: A. McPherson et al, JOSA B 4, 595 (1987); R. Rosman et al, JOSA B 5, 1237 (1988)
- Saclay, France: M. Ferray et al, J. Phys. B 21, L31 (1988); X.F. Li et al, Phys. Rev. A 39, 5751 (1988)

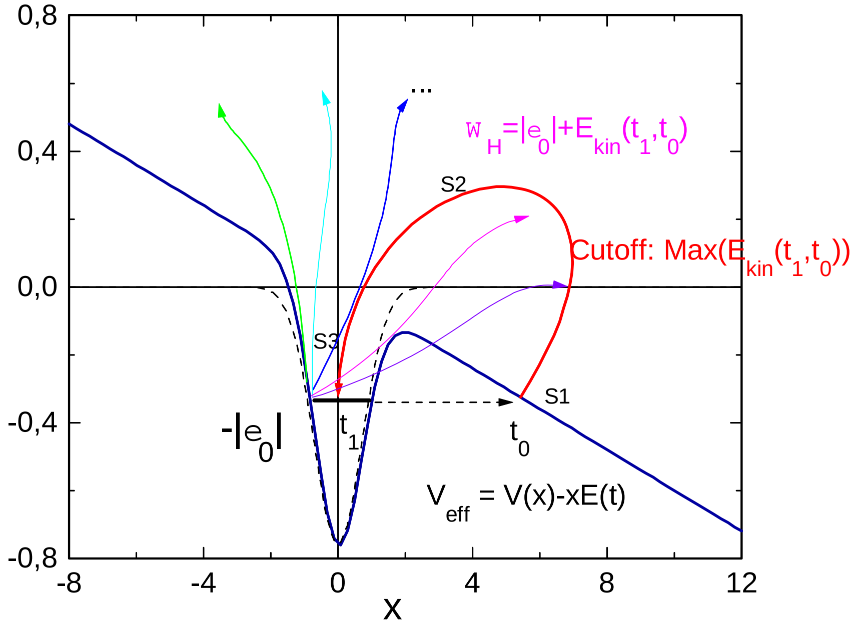
World record: $n \sim 300$ (Technical University Vienna, Austria)

Applications:

- high-frequency sources
- attosecond pulses: important for attosecond metrology
  - Short, e.g., few-cycle pulses (TU Vienna)
  - Counter-rotating polarizations in the input fields (Corkum,et al, Opt. Lett. 19, 1870 (1994))
  - Influence HH propagation through appropriate geometrical conditions (Antoine et al, PRL 77, 1234 (1996); PRA 54, R1761; Salières et al, Adv. At. Opt. Mol. Pys. 41, 83 (1999))

Physical picture: three-step model

- Classical version (or "simpleman's model"):M. Yu. Kuchiev, JETP Lett. 45, 404 (1987); K. C. Kulander et al, SILAP proceedings (Plenum, New York, 1993); P.B. Corkum, PRL71, 1994 (1993).
- Quantum-mechanical version: M. Lewenstein et al, Phys. Rev. A 49, 2117 (1994); W. Becker et al, Phys. Rev. A 41, 4112 (1990) and 50, 1540 (1994)



- Model:

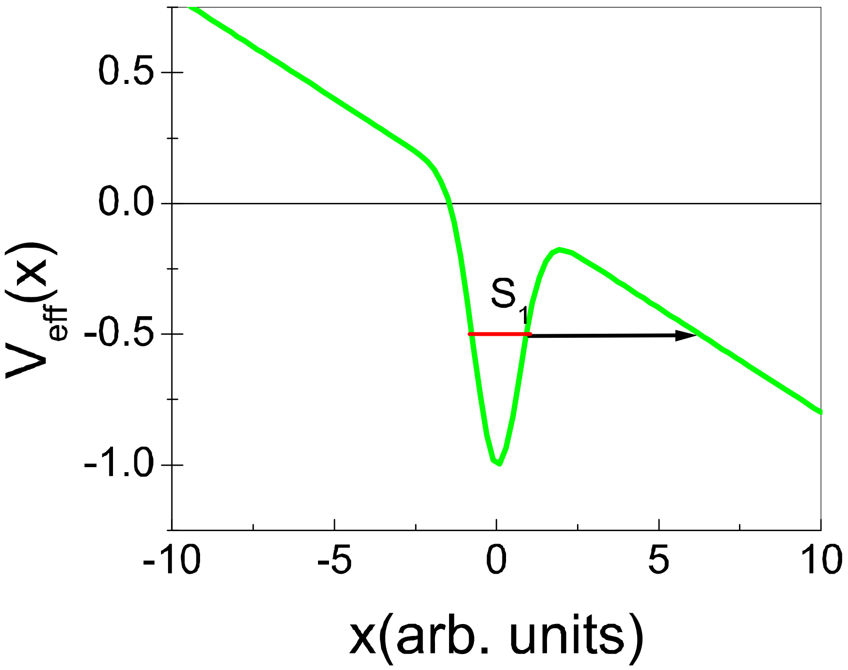
S_1: Electron leaves the atom at t_0
S_2: Electron propagates in the continuum
S_3: Electron recombines with the ground state at t_1

- Predictions:
  - Plateau
  - Harmonic energy: $\Omega_h = E_{kin}(t_0, t_1) + |\varepsilon_0|$
  - Cut-off: Maximum $E_{kin}(t_0, t_1)$.
  - Monochromatic fields: $\varepsilon_{coff} = |\varepsilon_0| + 3.17U_p$

Has become the paradigm for describing HHG in atoms

Consequences

- Most contributions to HHG within a field cycle occur at definite times, i.e., when the electron comes back
- Cutoff energies can be predicted using classical computations
- By changing the motion of the electron in the continuum, one may, for instance, influence groups of high harmonics
- For more complex driving fields, there may be several local maxima for $E_{kin}(t_0, t_1)$
- Relevance of a cutoff trajectory depends on
  - Field at the emission time (ionization rate $\sim \exp[-2^{5/2}|\varepsilon_0|^{3/2}/3|E(t_0)|]$)



  - Excursion time $t_1 - t_0$ of the electron in the continuum
  - Interference between different trajectories

Example: Bichromatic driving fields

C.F.M.F., M. Dörr, W. Becker and W. Sandner, PRA 60, 1377 (1999); C.F.M.F., W. Becker, M. Dörr and W. Sandner, Laser Phys. 9, 388 (1999); C.F.M.F, D.B. Milošević and G.G. Paulus, PRA 61, 063415 (2000)

$$E(t) = E_{01}\sin(\omega t) + E_{02}\sin(n\omega t + \phi)$$

The plateau structure depends on E_{01}, E_{02}, n, ϕ:

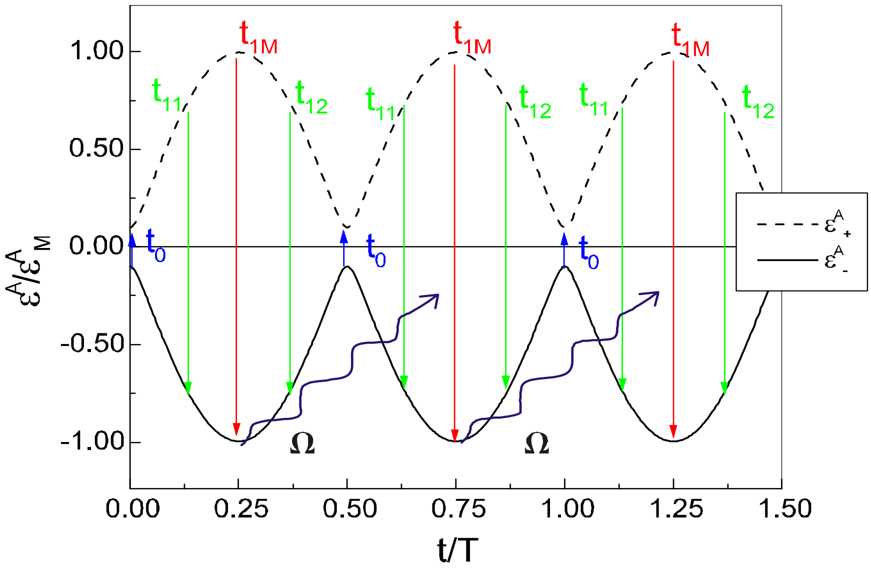
Example

-
  - $E_{01} = 0.1\ a.u.\ (I_\omega \sim 3 \times 10^{14}\ W/cm^2)$
  - $\omega = 0.057\ a.u.\ (\lambda \sim 800\ nm)$
  - $n = 2$ (i.e., a $\omega - 2\omega$ field)
  - E_{02}, ϕ varied

Two-level atom

B. Sundaram and P. Milonni, Phys. Rev. A 41, 6571 (1990) ; L. Plaja and L. Roso-Franco, J. Opt. Soc. Am. B 9(1992) and many other groups.

- HHG results from bound-bound transitions
- Plateau and cut-off at much lower frequencies
- Harmonic generation occurs due to transitions between the dressed states.



(C.F.M.F and I Rotter, Physical Review A 66, 013402 (2002)

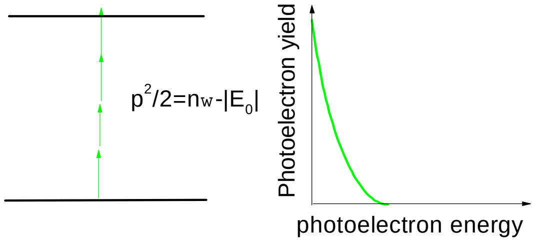
- S₁: Transition $\left|\phi_0^A(t)\right\rangle \longrightarrow \left|\phi_1^A(t)\right\rangle$ at t_0
  S₂: Adiabatic following
  S₃: Recombination with the ground state at t_1
- Inadequate for describing HHG in atoms
  - Cutoff law does not agree with experiments
  - Atom would immediately ionize in the required intensity range
  - time profiles do not agree with experiments
- Useful for alternative (e.g., solid-state) systems

Above-threshold ionization (ATI):

An atom absorbs more photons than the necessary amount for it to ionize

Low/moderate intensities

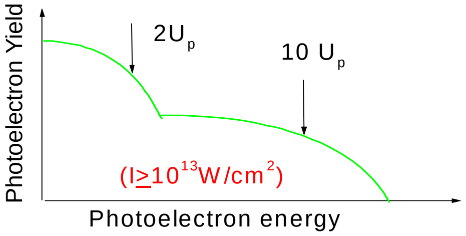
Multiphoton ionization



Clear physical picture
Approach: perturbation theory

High intensities

Above-threshold ionization (ATI)

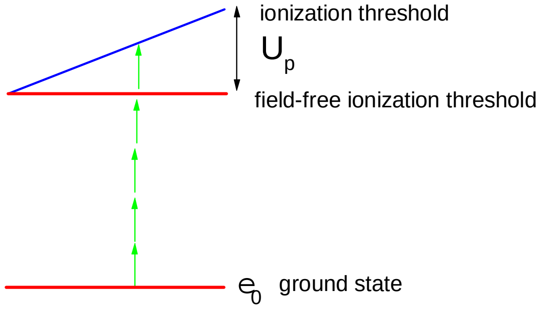


Physical picture?
Approaches?

- First measurements: Saclay: Agostini et al, PRL 42, 1127 (1979)
- This was the first clear evidence that perturbation theory breaks down:
  - Intensity dependence contradicts perturbation theory
  - Low-energy peaks are reduced in magnitude

Explanation: The ionization threshold is effectively shifted by the field:

Channel-closing condition

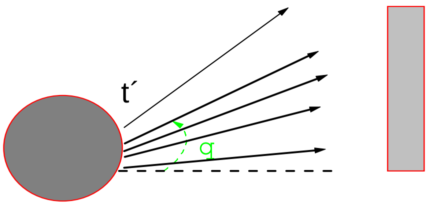


Plateau

(first measurements: G.G. Paulus et al, PRL 72, 2581 (1994))

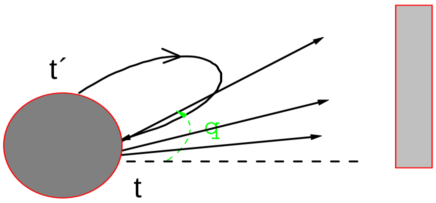
Physical picture

Direct electrons



Cutoff: $2U_p$

Rescattered electrons



Cutoff: $10U_p$

- Cutoff: maximal electron kinetic energy
- $(t, t') =$ (start time, return time)
- Classical description: G.G. Paulus et al, PRA 52,4043 (1995).
- Quantum-mechanical description: See, e.g., A. Lohr et al, PRA 55, R4003 (1997); W. Becker et al, PRA 56, 645 (1997).

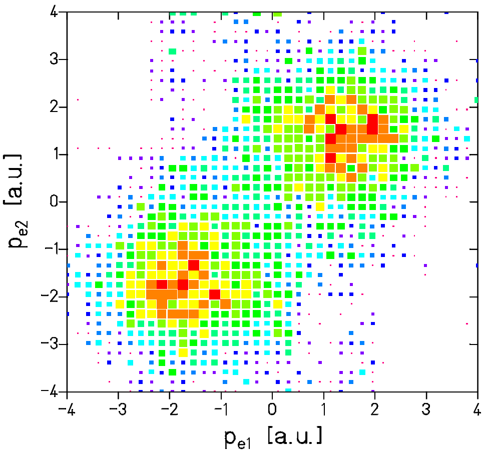
- In particular the method of “quantum orbits” has been successfully applied in this context: (R. Kopold et al, Opt. Comm. 179, 39 (2000); P. Salières et al, Science 292, 902 (2001); S. P. Popruzhenko et al, PRL 89, 023001 (2002), CFMF et al, PRA 66, 043413 (2002))

Nonsequential double ionization
First evidences:

- A. L'Huillier et al, PRA 27, 2503 (1983)
- B. Walker et al, PRL 73, 1227 (1994)
- Electron-electron correlation effects are huge
- Treatment beyond the Single Active Electron Approximation is required

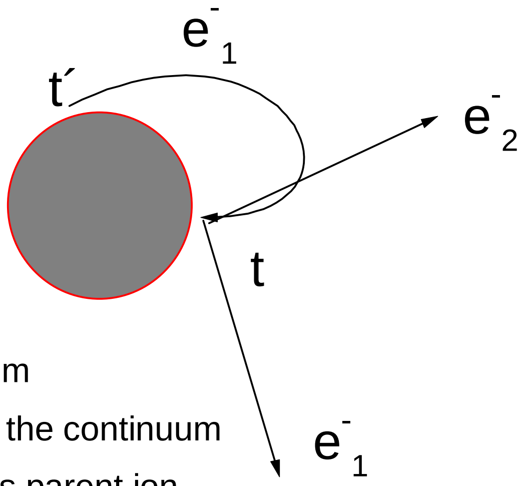
Recent experiments (Neon):

- 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)
- Distributions peaked around $p = \pm 2\sqrt{U_p}$
  ($U_p = I_0/4\omega^2$=ponderomotive energy)



- Linearly polarized light (Approximated by $E(t) = E_0 \sin(\omega t)$)
  - Intensity: $5{\times}10^{14} - 10^{15} W/cm^2$
  - Wavelength: $\lambda \sim 800nm$ (Ti:sapphire)
- Momentum distributions peaked around $p = \pm 2\sqrt{U_p}$

Physical picture



S_1: e^-_1 leaves the atom
S_2: e^-_1 propagates in the continuum
S_3: e^-_1 collides with its parent ion
and frees e^-_2

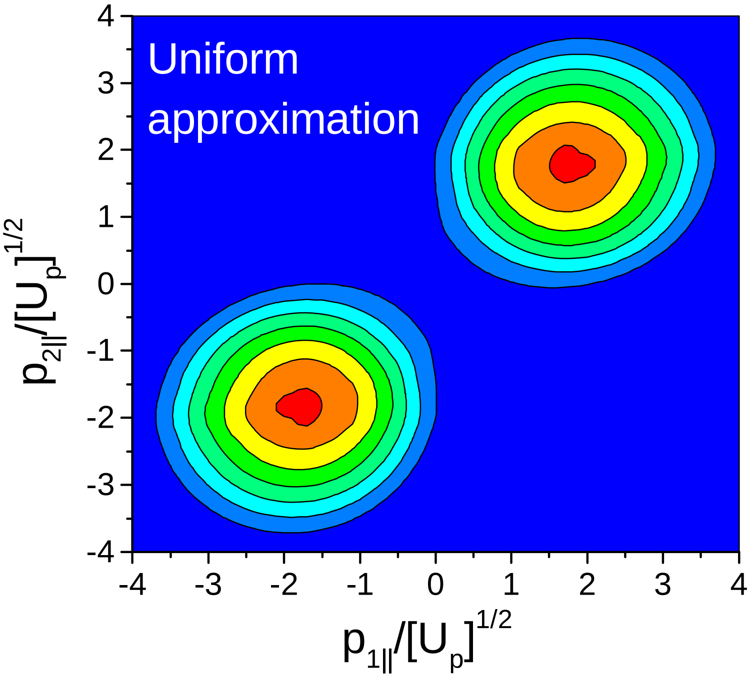
- High-order ATI: elastic rescattering
- Non-sequential double ionization: inelastic rescattering

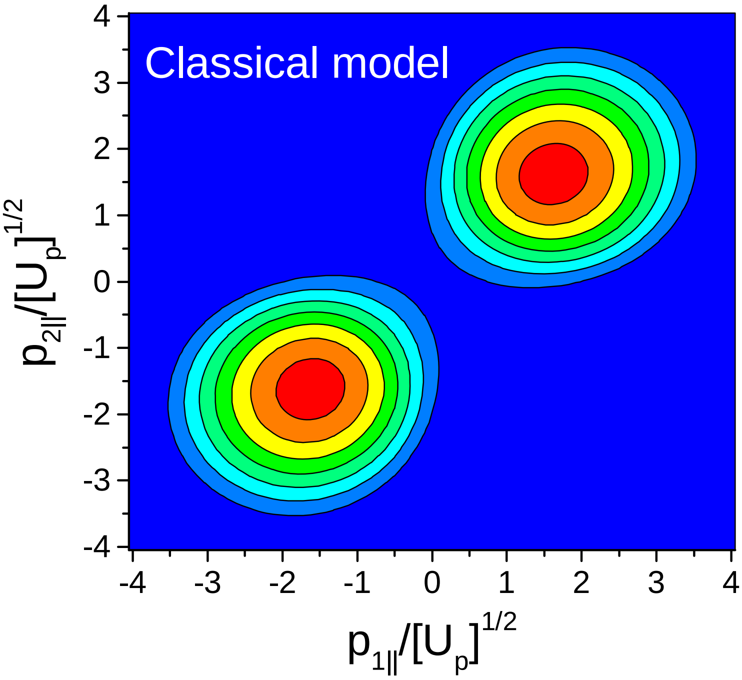
Conservation of energy at the rescattering time:

$$(p_{1x} - A(t))^2 + (p_{2x} - A(t))^2 = E_{kin}(t) - I_p - p_{1\perp}^2 - p_{2\perp}^2$$

- Circle centered at $p_{1x} = p_{2x} = A(t)$
- Maximum probability near $A(t) \simeq 2\sqrt{U_p}$
- Radius depend on :
  - the ionization potential I_p for the ion
  - the transverse momenta $p_{i\perp}$ for both e^-s
  - the kinetic energy $E_{kin}(t)$ of the 1^{st} e^- upon return

Theory (examples)





($U_p = I_0/4\omega^2$=ponderomotive energy)Very good agreement!
How can one compute such yields?

Stabilization

"Stabilization is the decrease of the ionization probability with the strength of the external field"

- Controversial issue:

"Stabilization exists": J. Eberly, Q. Su et. al (Illinois/Rochester group); K. Kulander et al.; P. Knight, K. Burnett et al (London and Oxford groups); M. Gavrila et. al (Amsterdam group); H. Reiss; F. Faisal; R. Potvliege, R. Shakeshaft et. al

"Stabilization does not exist": S. Geltman, Chen and Bernstein Potvliege, R. Shakeshaft et. al

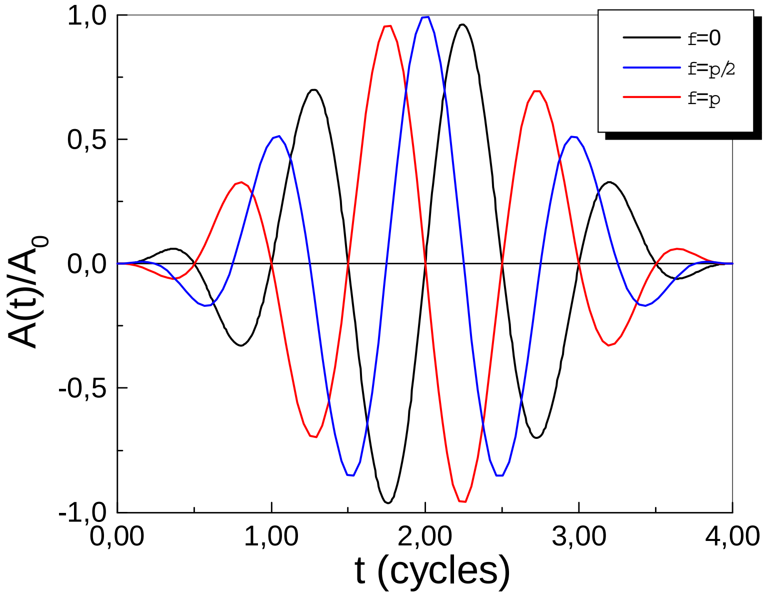
For reviews on the subject: K. Burnett et al, J. Phys.B 26 (1993) 561; J.H. Eberly and K.C. Kulander, Science 262 (1993) 1229; S. Geltman, Chem. Phys. Lett. 237 (1995) 286

- Picture: electron gets trapped in a field-distorted potential
- Occurrence: high frequencies and intensities (both 1 a.u.)
- Strongest evidence (still far below the required intensity range): N.J. Van Druten et al, PRA 55, 622 (1997)

Matter not settled yet! (no experiments exist)

Perspectives

- Few-cycle pulses



- More complex (many-body) systems in strong laser fields:
  - Clusters
  - Dielectrics
  - Surfaces