Role of Resonances and Re-collisions in ATI and Harmonic Generation

- Plateaux and enhancements in ATI spectra
- Resonances or «channel closings» ?
- Resonances + Multiple re-collisions
- About harmonic generation spectra

Alfred Maquet
Université Pierre et Marie Curie (Paris)
& KITP

ATI and HHG

General remark:
Around $I \approx 10^{14}$ Wcm^{-2} at 800 nm, the dynamics is neither pure multiphoton nor pure tunnelling (Keldysh parameter $\gamma \approx 1$).

$\Rightarrow$ no simple picture emerges for the details of the spectra ...

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Plateau in Above Threshold Ionization Spectra

G. G. Paulus,[1] W. Nicklich,[1] Huale Xu,[4] P. Lambropoulos,[1,3] and H. Walther[1,2]
[1]*Max-Planck-Institut für Quantenoptik, 85748 Garching, Germany*
[2]*Sektion Physik der Universität München, Germany*
[3]*Foundation for Research and Technology-Hellas, Institute of Electronic Structure and Laser, P.O. Box 1527, Heraklion 71110, Crete and Department of Physics, University of Crete, Greece*
[4]*Department of Physics, University of Southern California, Los Angeles, California 90089-0484*
(Received 28 October 1993)

Envelope of the spectra:
tunnelling + classical dynamics.

N.B. Fast electrons with E_{kin} > $2U_p$ have experienced at least one recollision, (Paulus *et al.* 1994).

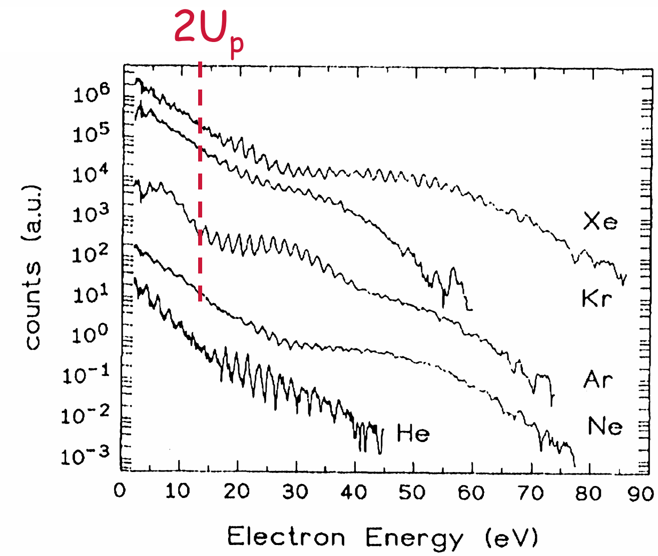

FIG. 1. ATI spectra of all rare gases. The intensity was 3×10^{14} W/cm^2 for He and 2×10^{14} W/cm^2 for all the others. (The curves are separated in the vertical direction.)

Conspicuous enhancements are seen in the plateaus(x) at selected intensities…

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LETTER TO THE EDITOR

Evidence for resonant effects in high-order ATI spectra

Marcus P Hertlein†, Philip H Bucksbaum†§ and H G Muller‡

† Physics Department, University of Michigan, Ann Arbor, MI 48109-1120, USA
‡ FOM-AMOLF, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands

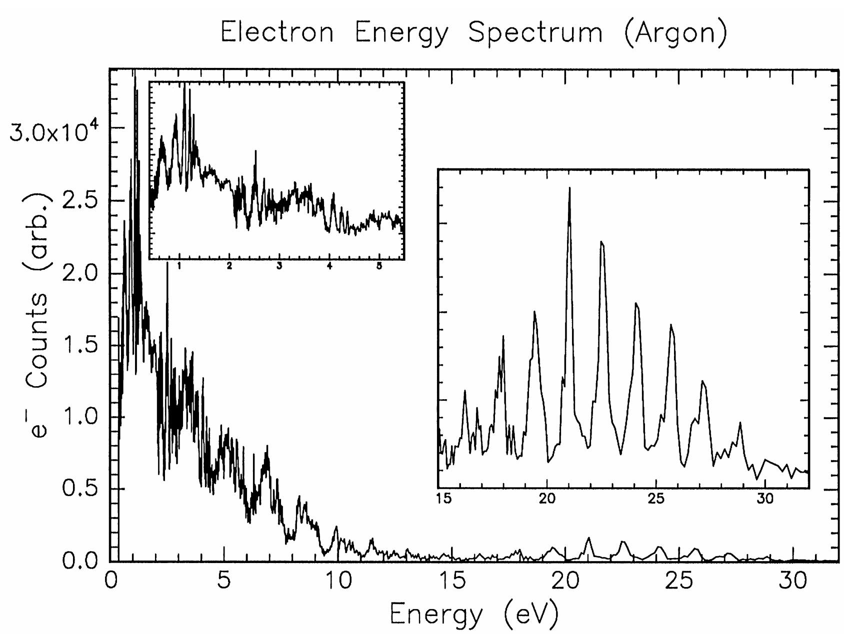


Figure 1. ATI spectrum for argon at an intensity of about 7×10^{13} W cm^{-2}. The left-hand inset shows the region of low-order Rydberg resonances with an expanded energy scale. The right-hand inset shows the region of high-energy electrons which are the focus of this paper.

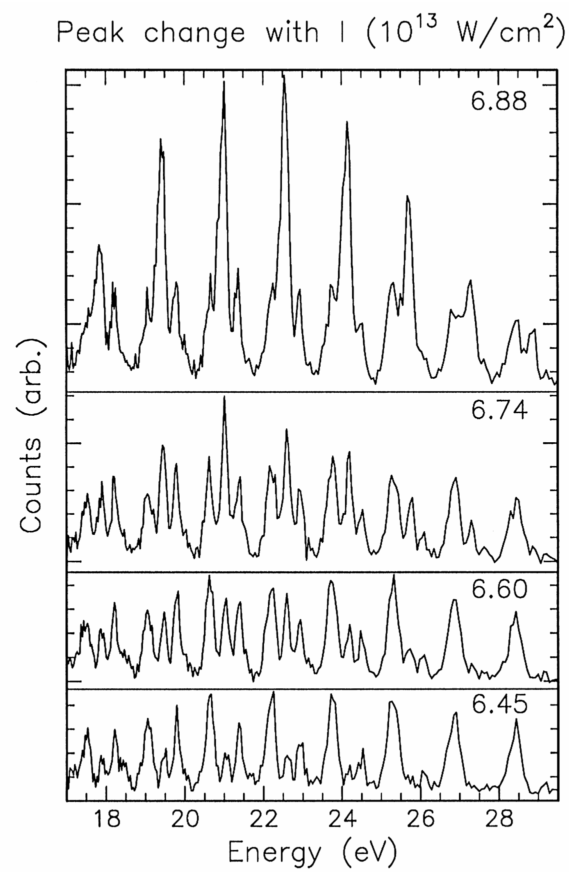
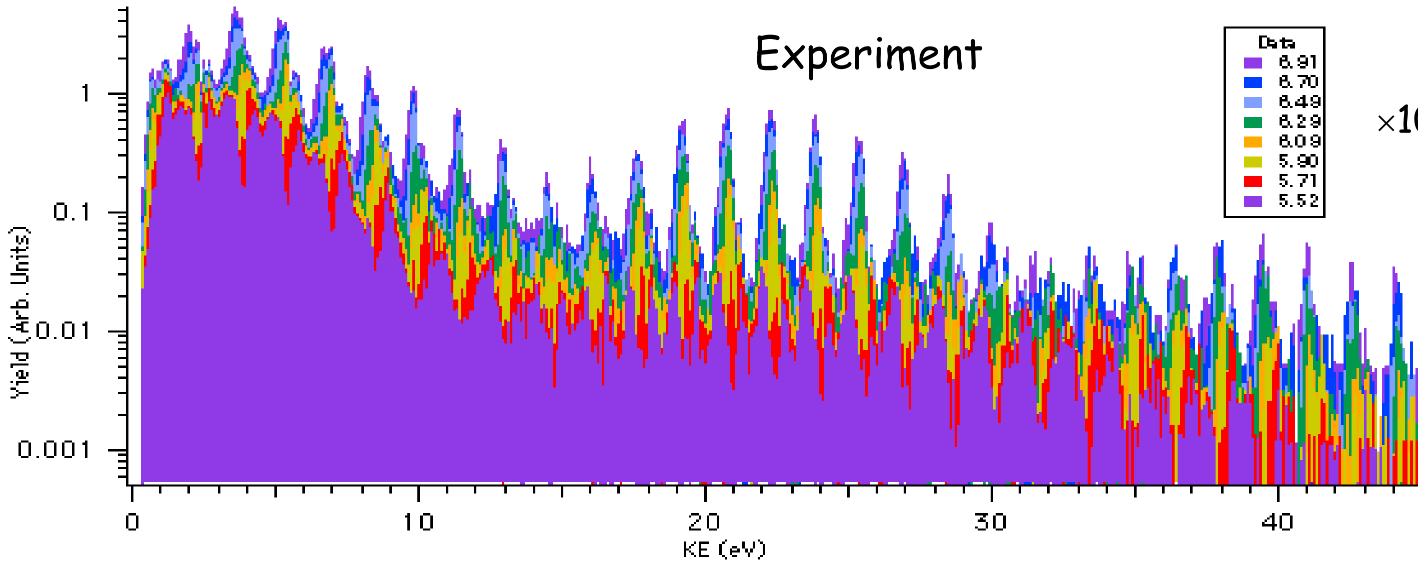


Figure 2. ATI electron spectrum in argon in the 15–35 eV range, over the intensity range 6.45×10^{13}–6.88×10^{13} W cm^{-2}.

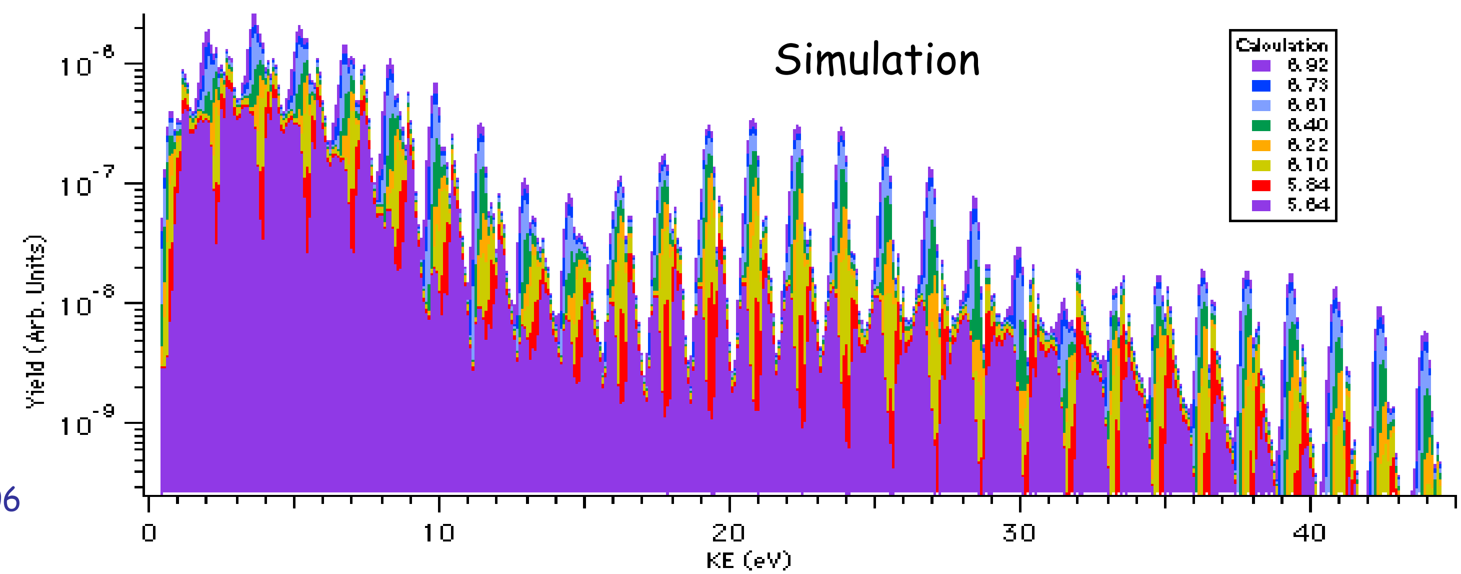
ATTO-06

Intensity dependence of plateau structures in Ar

Nandor, Van Woerkom, Muller *et al.* (1999)

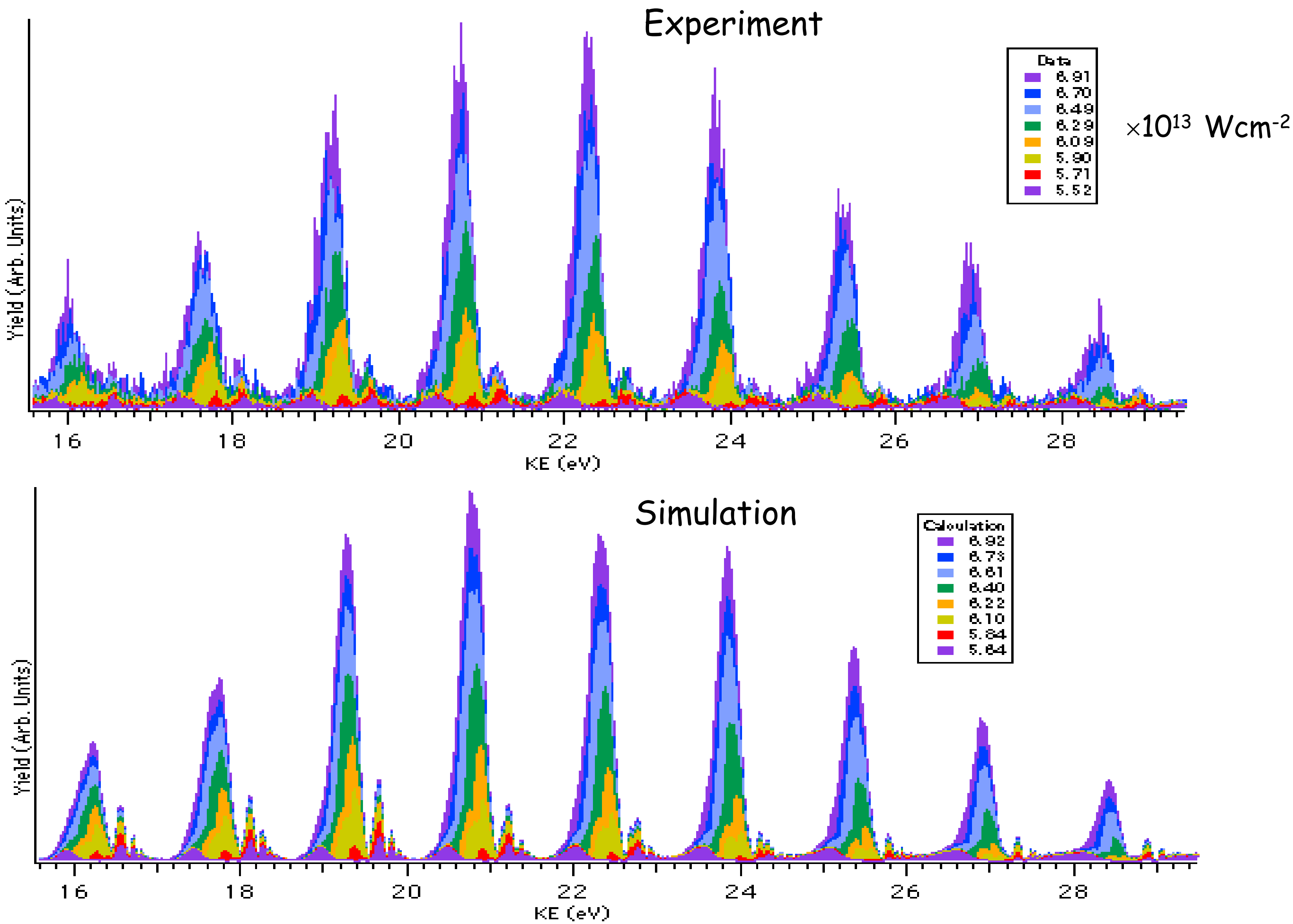


$\times 10^{13}$ Wcm^{-2}

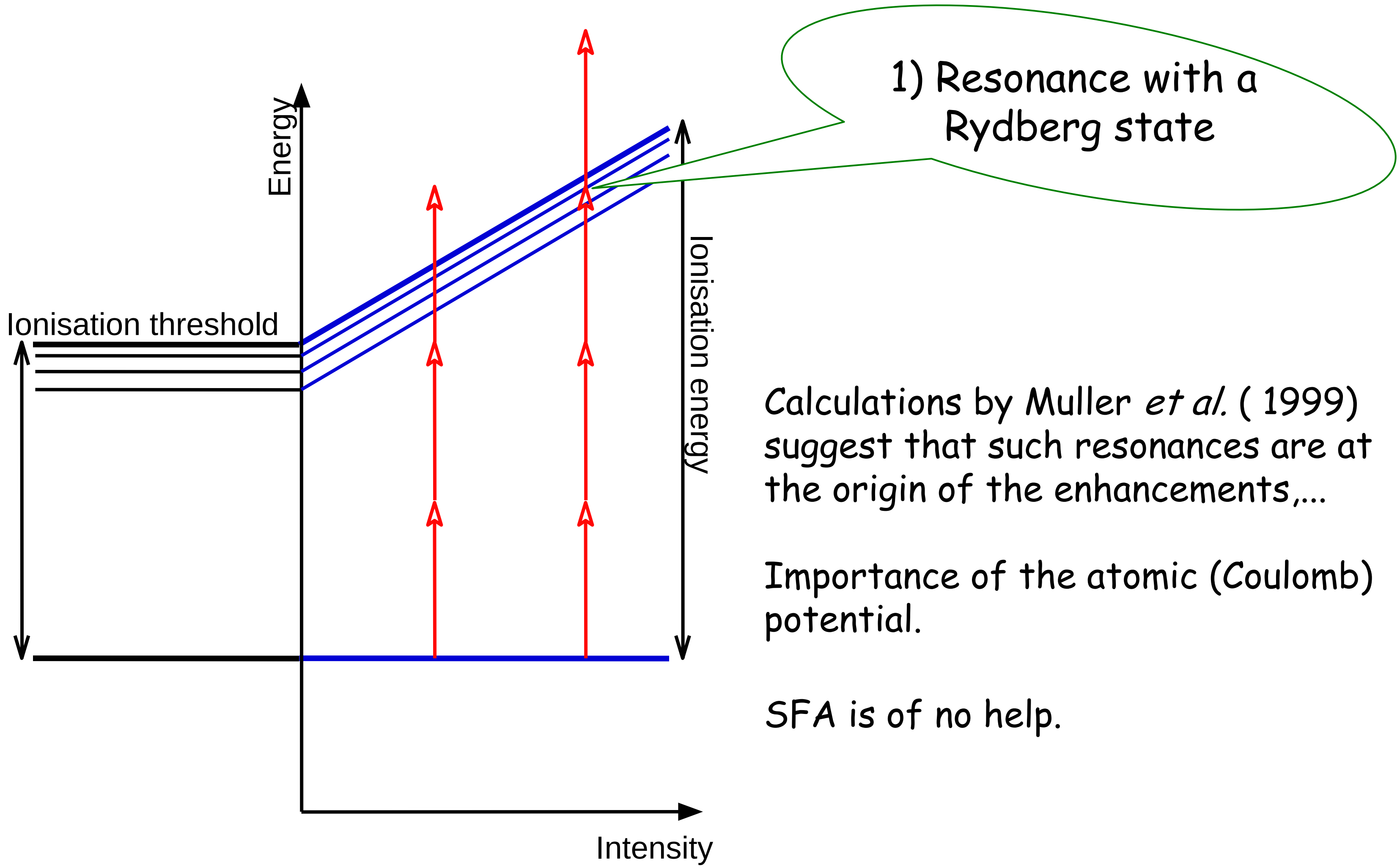


Non linear dependence of the magnitude of the peaks on the intensity

Nandor, Van Woerkom, Muller *et al.* (1999)



Question: what is the origin of these enhancements?
Two mechanisms have been proposed in the literature



Calculations by Muller *et al.* (1999) suggest that such resonances are at the origin of the enhancements,...

Importance of the atomic (Coulomb) potential.

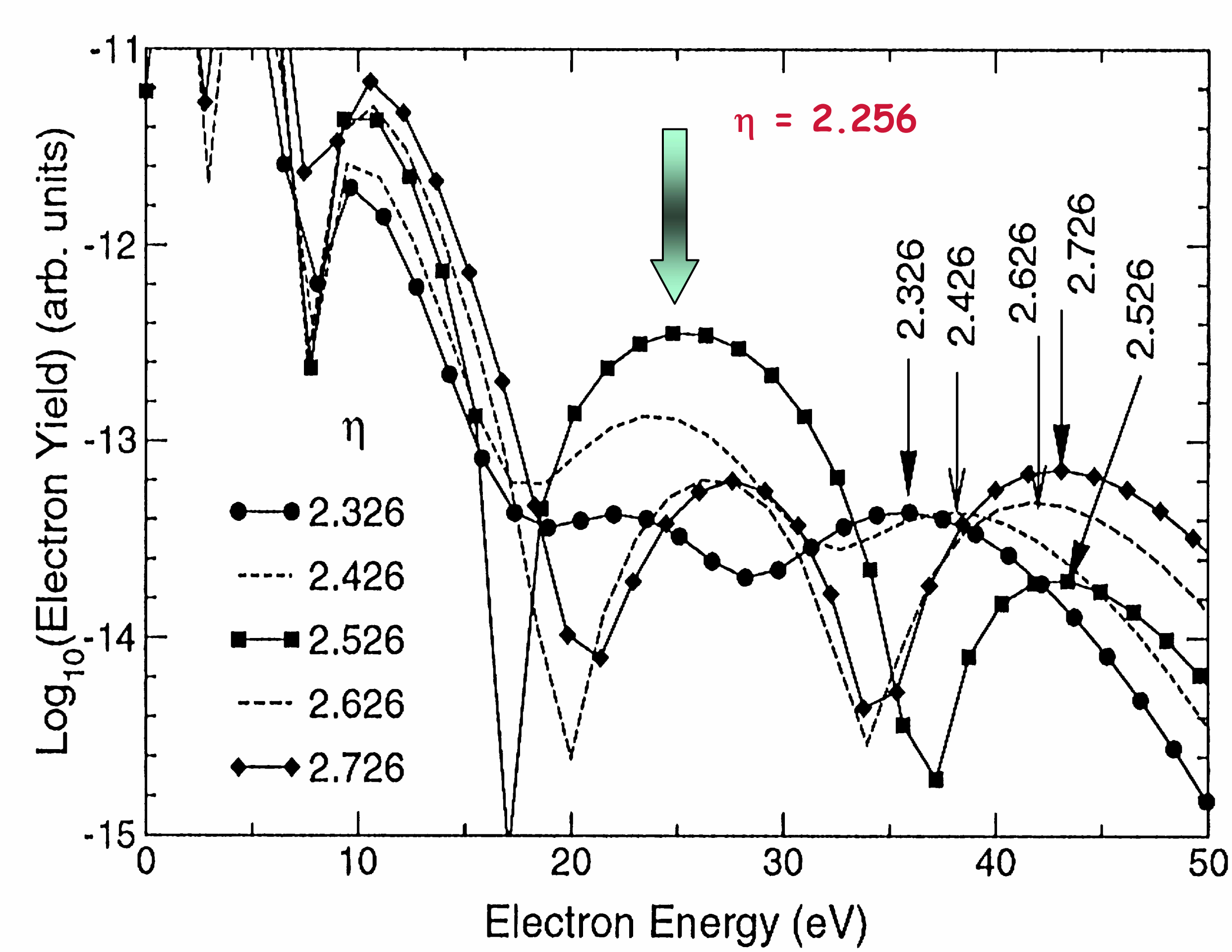
SFA is of no help.

However: One can reproduce the enhancements with the help of S-matrix calculationsfor a zero-range (3-D δ–function) model potential !

[R. Kopold, W. Becker, M. Kleber and G. G. Paulus 2002]

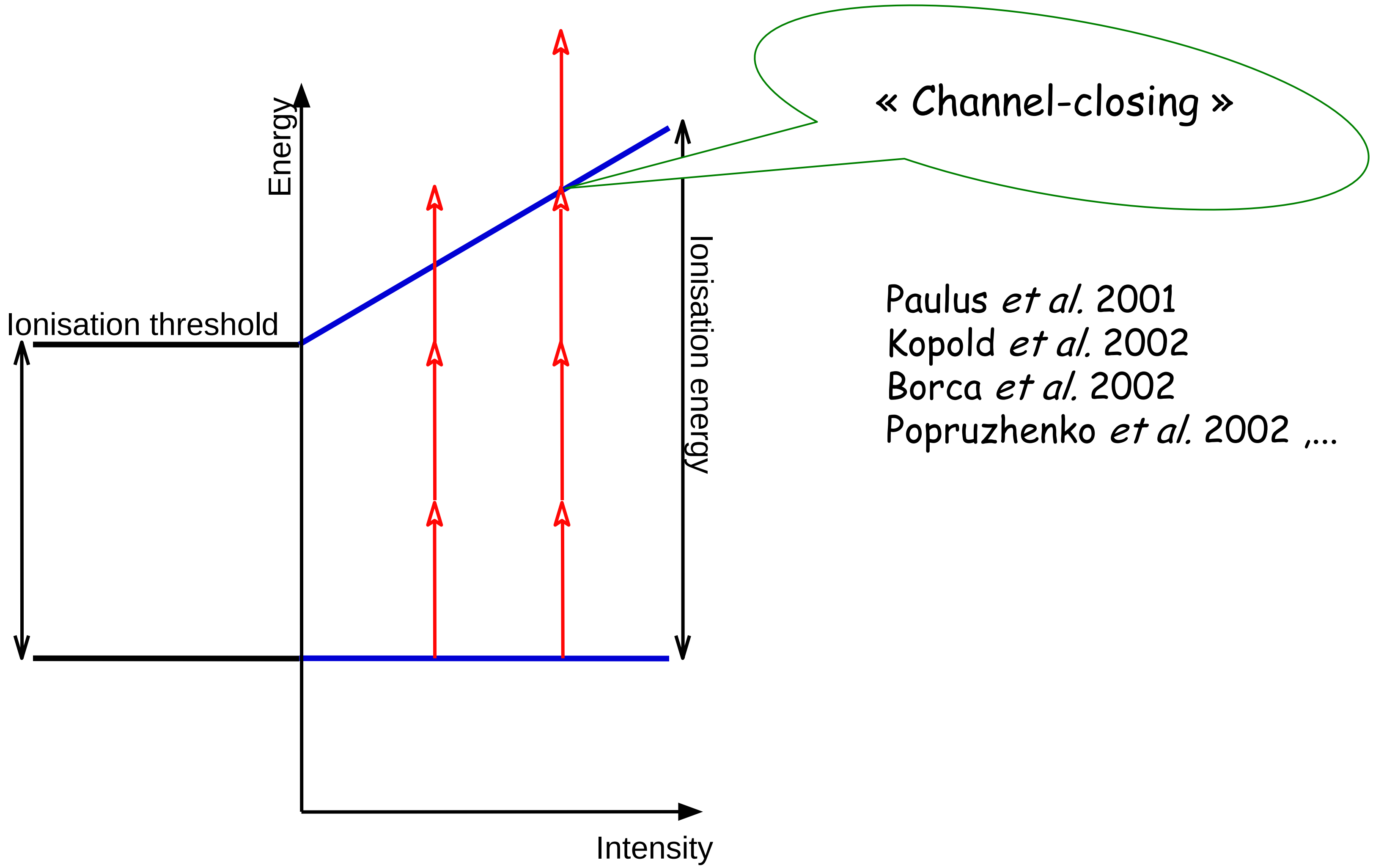
zero-range (δ–function) potential $\Longrightarrow$ Only one bound state

$\eta = U_p/\omega$



12-photon « Channel-Closing » at :
$\eta = 2.256$

2) « Channel-closing » in a zero-range potential
(no Rydberg states!)



Paulus *et al.* 2001
Kopold *et al.* 2002
Borca *et al.* 2002
Popruzhenko *et al.* 2002 ,...

Two questions and two approaches:

1) Quantum. If it is a resonant process, how can we identify the resonant state(s)?
(Difficult from numerical resolution of TDSE, see however Muller *et al.*)

2) Classical. As the enhancements involve fast electrons, (E_{kin} > $2U_p$) what is the role of the recollisions?

1) Finding the resonances (if any!) associated to the enhancements. Compare long-range and short-range potentials

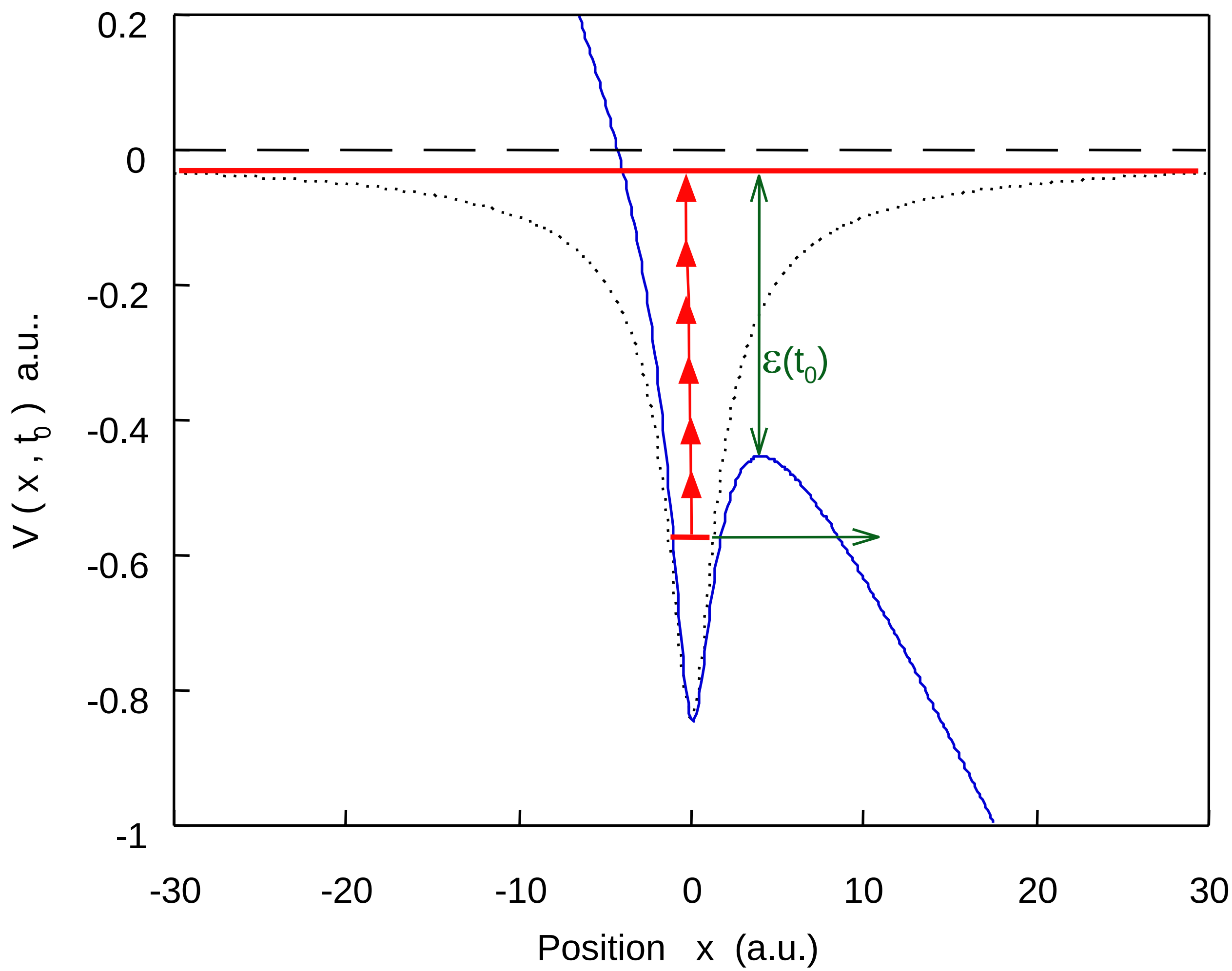
Idea: look at the Floquet spectrum (at these intensities excited atomic states are significantly shifted and broadened, bare-state basis is not well adapted)

Model: - 1-D soft Coulomb potential
- Pöschl-Teller
- «soft» Yukawa } not discussed here

Calculations: - TDSE for photoelectron spectra
- Floquet analysis

NB. Calculations are under way for more realistic 3-D potentials (H and Ar)

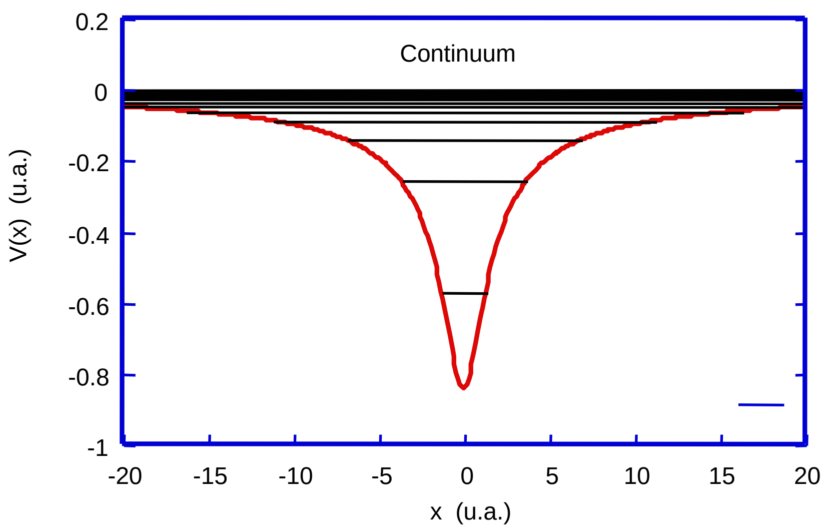
Questions: Rydberg states are located so high above the barrier. How they can play a role in the dynamics for fast photoelectons that have experienced at least one recollision?? The lifetimes of these states should be very short (e.g. shorter than a half cycle...)?

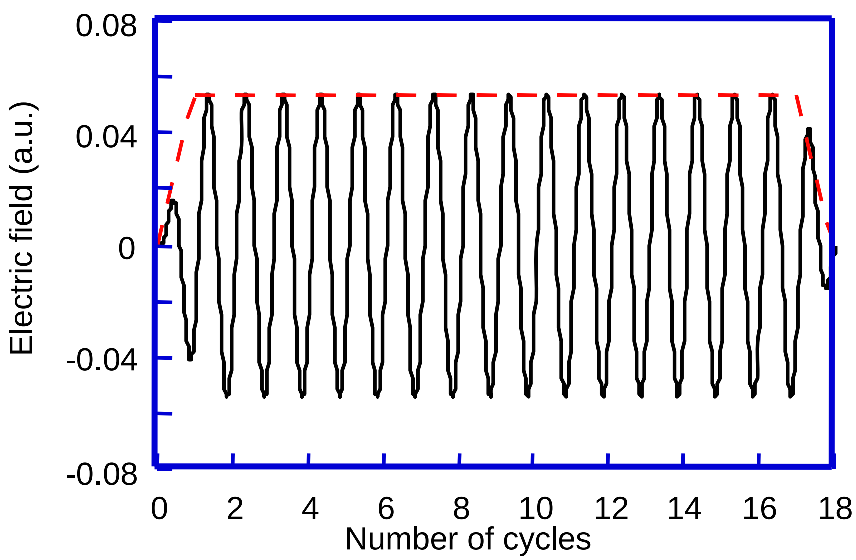


Model calculation for investigating the origin of the enhancements in ATI spectra: long-range «soft-Coulomb» potential

$$H_0 = -\frac{1}{2}\frac{\partial^2}{\partial x^2} - \frac{1}{\sqrt{a + x^2}}$$

with a = 1. 41 ; $E_0 \approx$ - 0.58 a.u. (Ar atom)

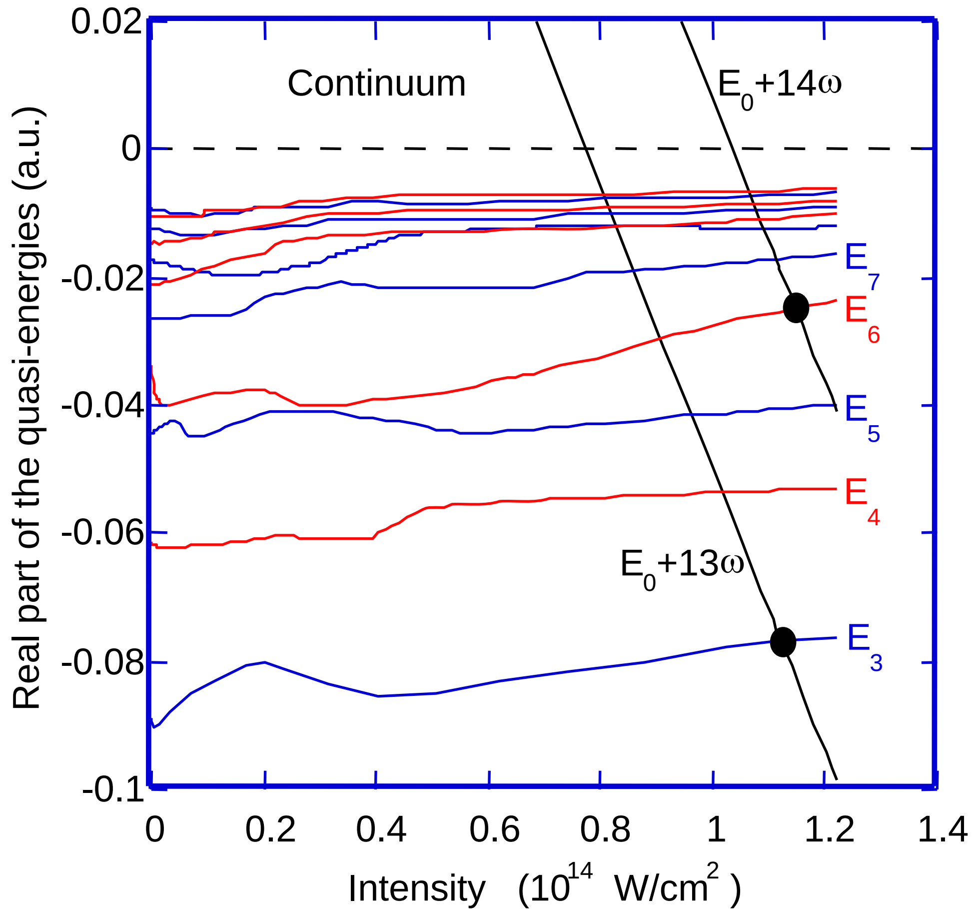




Time-dependent Schrödinger equation

$$i\frac{\partial \psi(x,t)}{\partial t} = \left[H_0 + i\frac{A(t)}{c}\frac{\partial}{\partial x} \right]\psi(x,t)$$

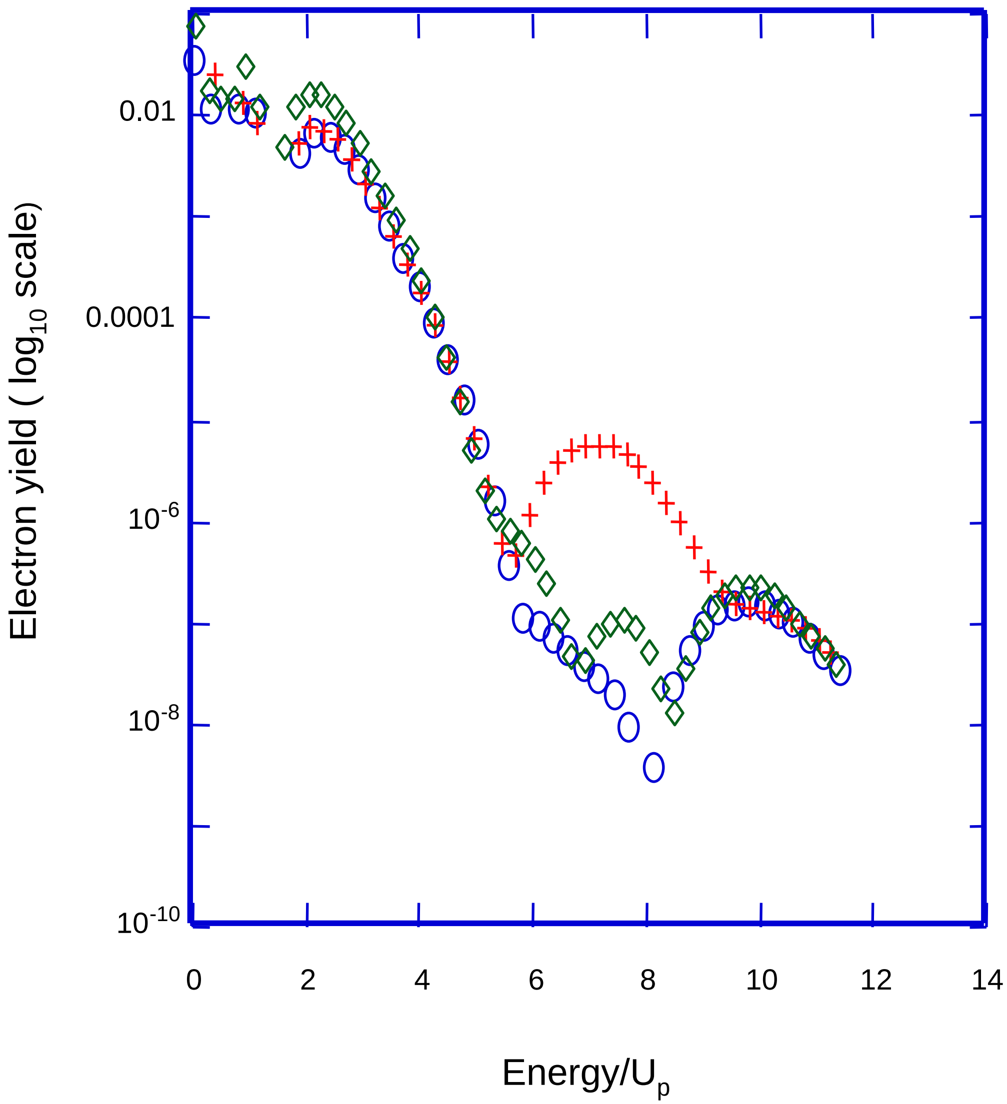
Quantum (1-D «soft-Coulomb»): Floquet analysis: locations of resonances



* 13-photon resonance with n = 3 at $I = 1.12 \times 10^{14}$ W/cm^2.

* 14-photon resonance with n = 6 at $I = 1.145 \times 10^{14}$ W/cm^2.

TDSE: Numerical simulations of ATI spectra at intensities close to the n = 3 resonance (1-D «soft-Coulomb»)



○ $I = 1.02\ 10^{14}$ W/cm^2

+ $I = 1.12\ 10^{14}$ W/cm^2

◊ $I = 1.22\ 10^{14}$ W/cm^2

$\omega = 0.0577$

see: Wassaf *et al.*
PRL **90**, 013001 (2003);
PRA **67**, 053405 (2003)

Identifying the resonances in the ATI peaks

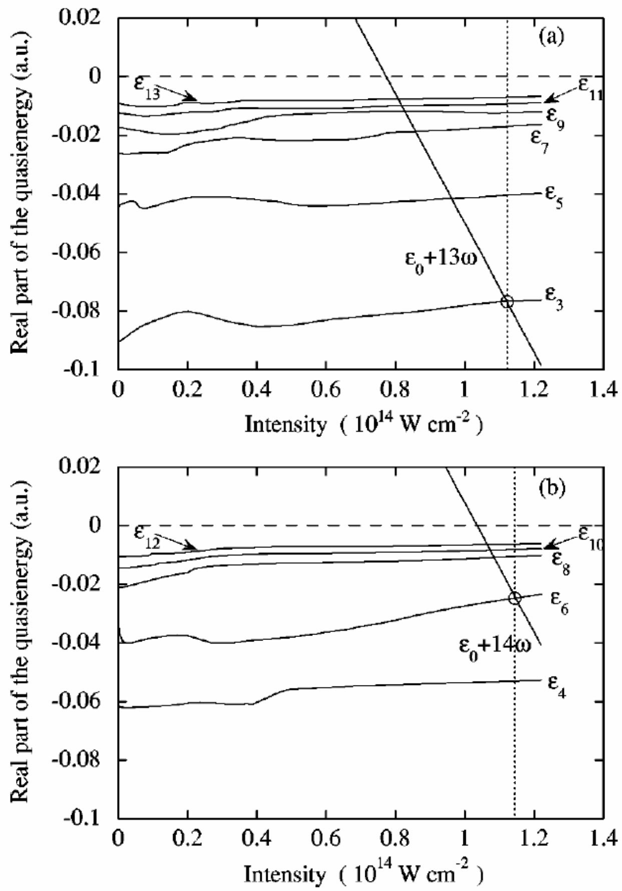


FIG. 4. (a) Odd-parity states quasienergies; (b) even-parity states quasienergies. The crossings between the ground state and the dressed states $n=3$ and $n=6$ occur at intensities 1.12 $\times 10^{14}$ W cm^{-2} and 1.145$\times 10^{14}$ W cm^{-2}, respectively.

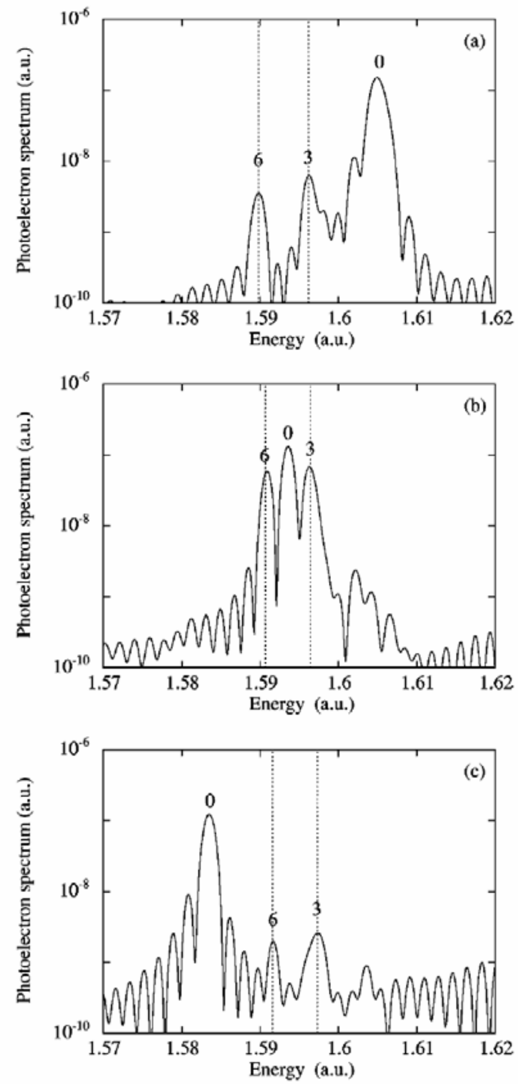
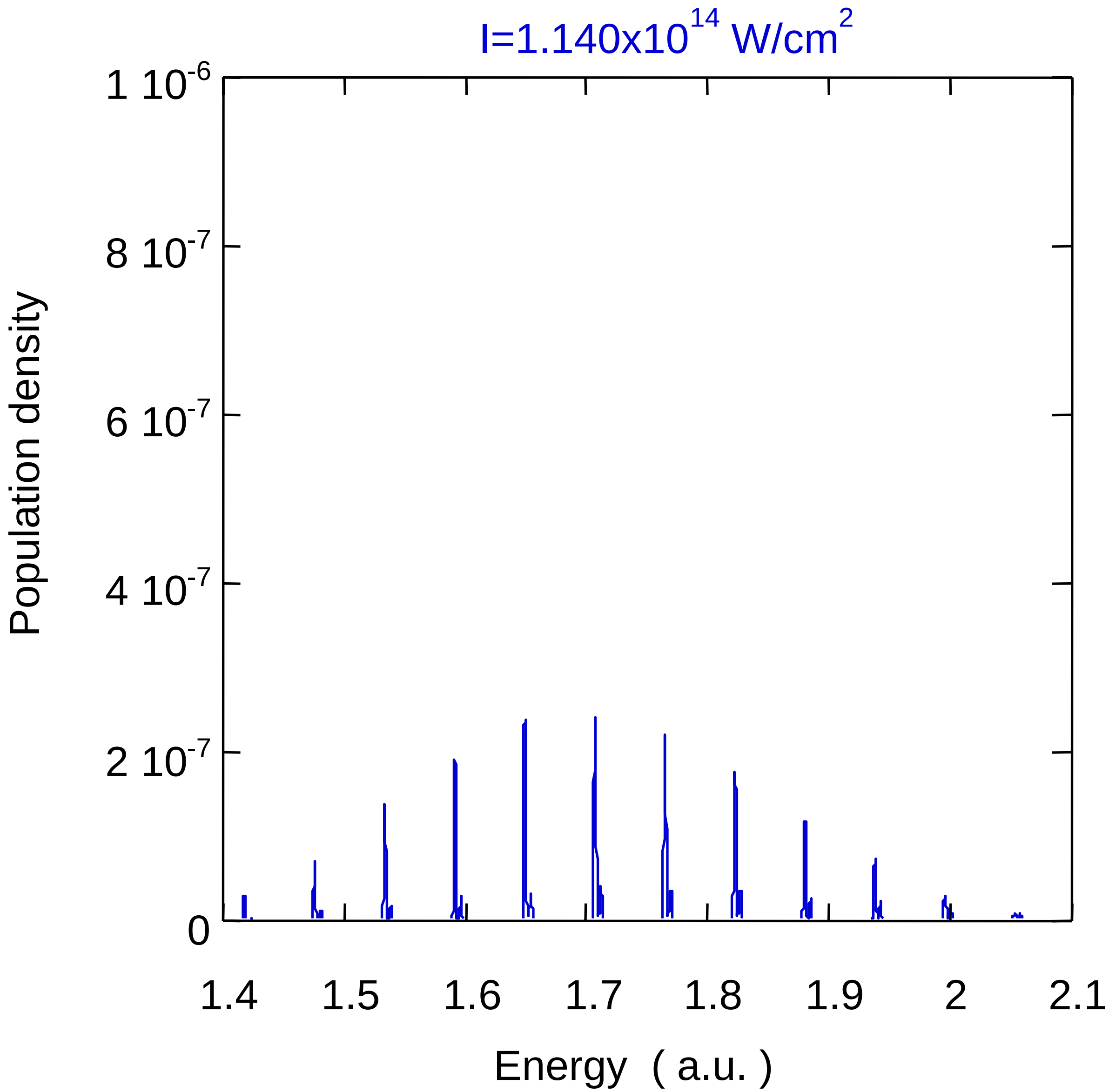
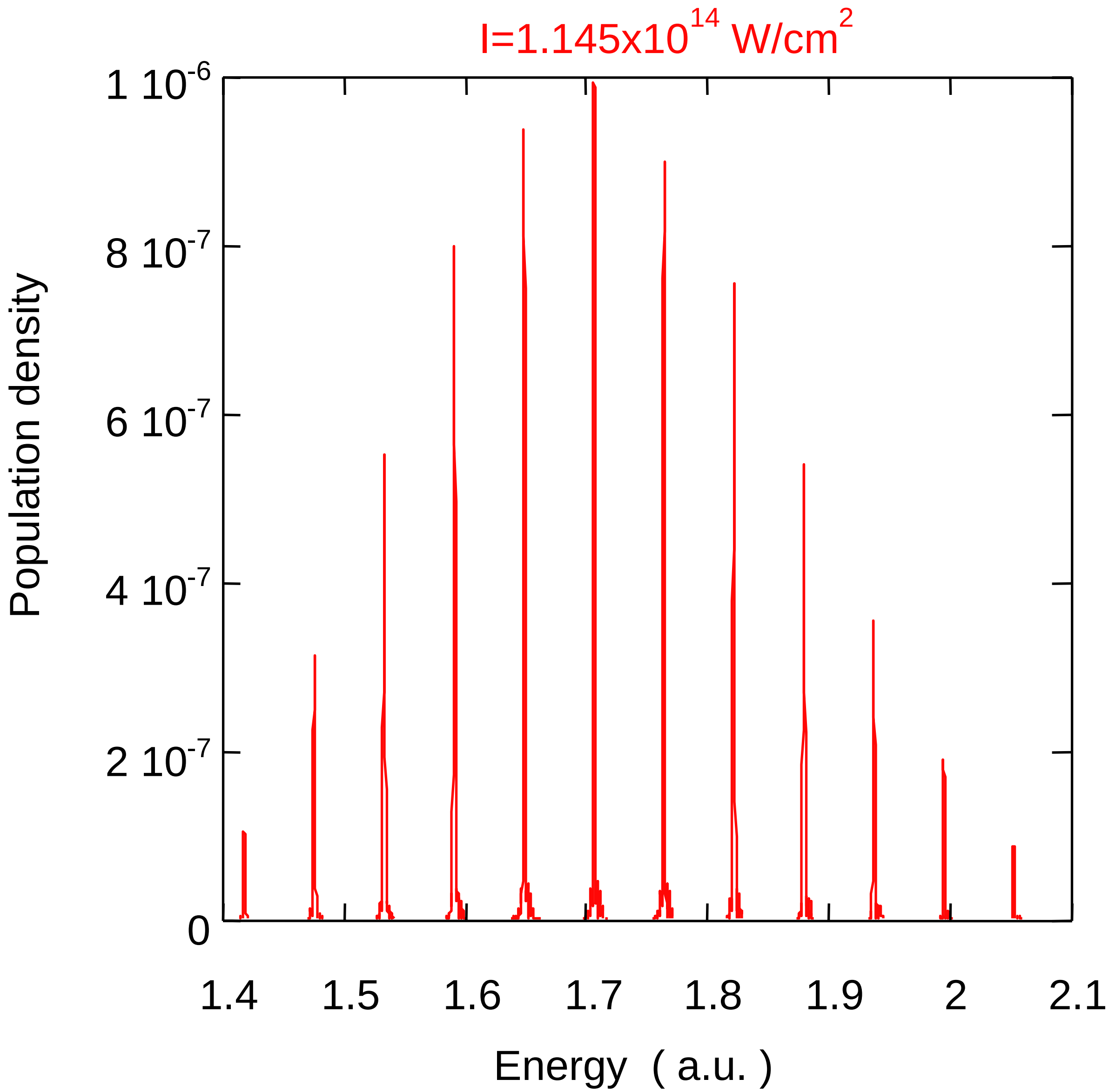


FIG. 5. Details of one photoelectron peak showing subcomponents. (a) corresponds to an intensity of 1.081×10^{14} W cm^{-2}, (b) to 1.135×10^{14} W cm^{-2}, and (c) to 1.180×10^{14} W cm^{-2}. The peak labeled 0 corresponds to the direct ionization process, while the peaks labeled 3 and 6 are associated to Freeman resonances via the dressed states $n=3$ and $n=6$.

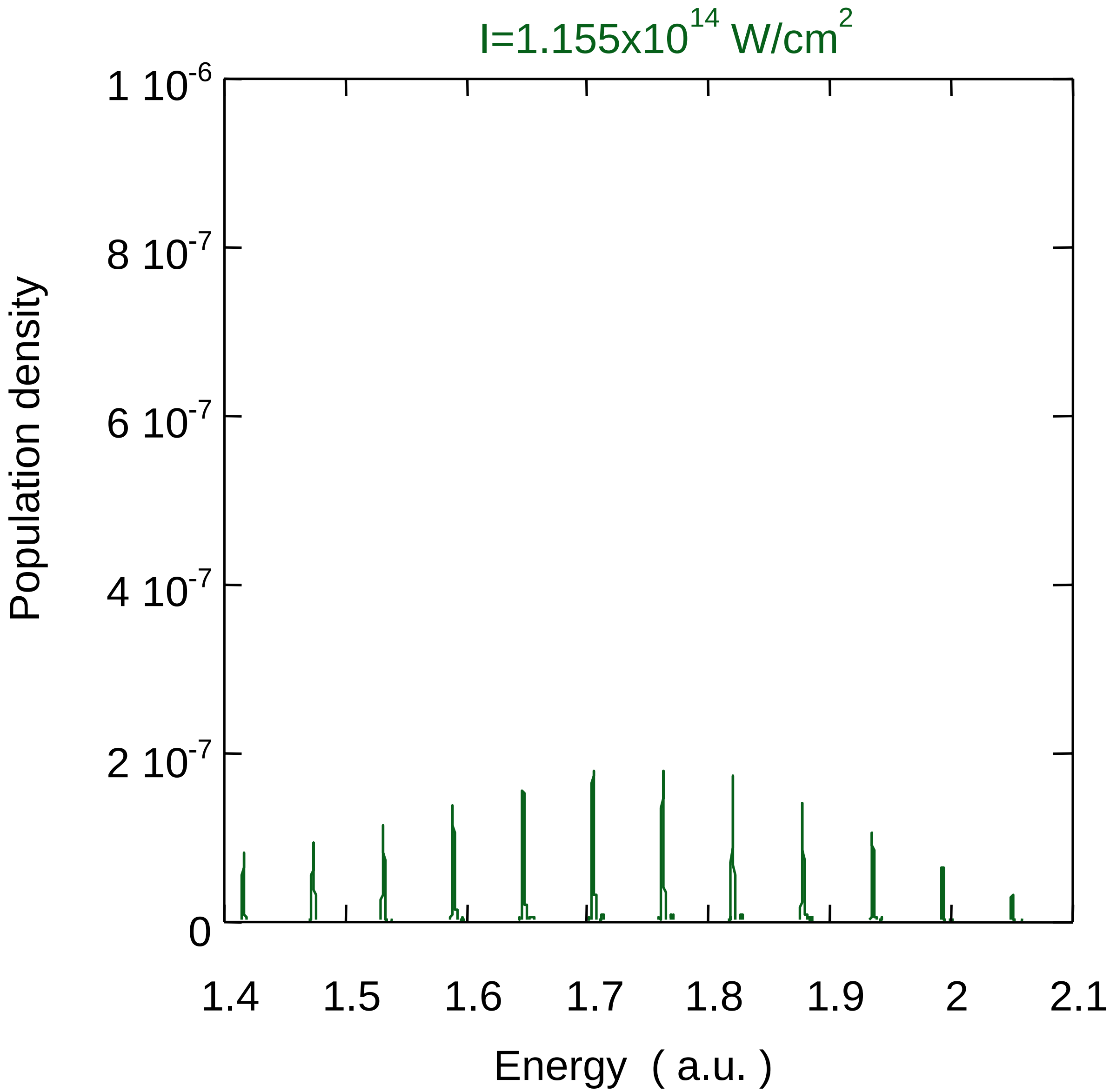
Resonance on n = 6



Resonance on n = 6



Resonance on n = 6



Floquet calculations for the soft-Coulomb potential (a « Sturmian-on-a-grid » analysis)

Floquet:

$$\Psi(x,t) = e^{-i\mathcal{E}t} \sum_{n=-\infty}^{n=+\infty} e^{in\omega t} \phi_n(x).$$

Floquet-Fourier components:

$$[H_0 + n\omega]\phi_n(x) + i\frac{A_0}{c}\frac{\partial}{\partial x}\phi_{n+1}(x) + i\frac{A_0}{c}\frac{\partial}{\partial x}\phi_{n-1}(x) = \mathcal{E}\phi_n(x). \quad (7)$$

Floquet calculations for the soft-Coulomb potential (a « Sturmian-on-a-grid » analysis)

Schrödinger:

$$\left(-\frac{1}{2}\frac{\partial^2}{\partial x^2}-\frac{\alpha}{\sqrt{c+x^2}}\right)\psi_n(\alpha,x)=\lambda_n\psi_n(\alpha,x),$$

Associated « Sturmian » equation:

$$\sqrt{c+x^2}\left(\frac{1}{2}\frac{\partial^2}{\partial x^2}+\lambda\right)S_\nu(\lambda,x)=\alpha_\nu S_\nu(\lambda,x),$$

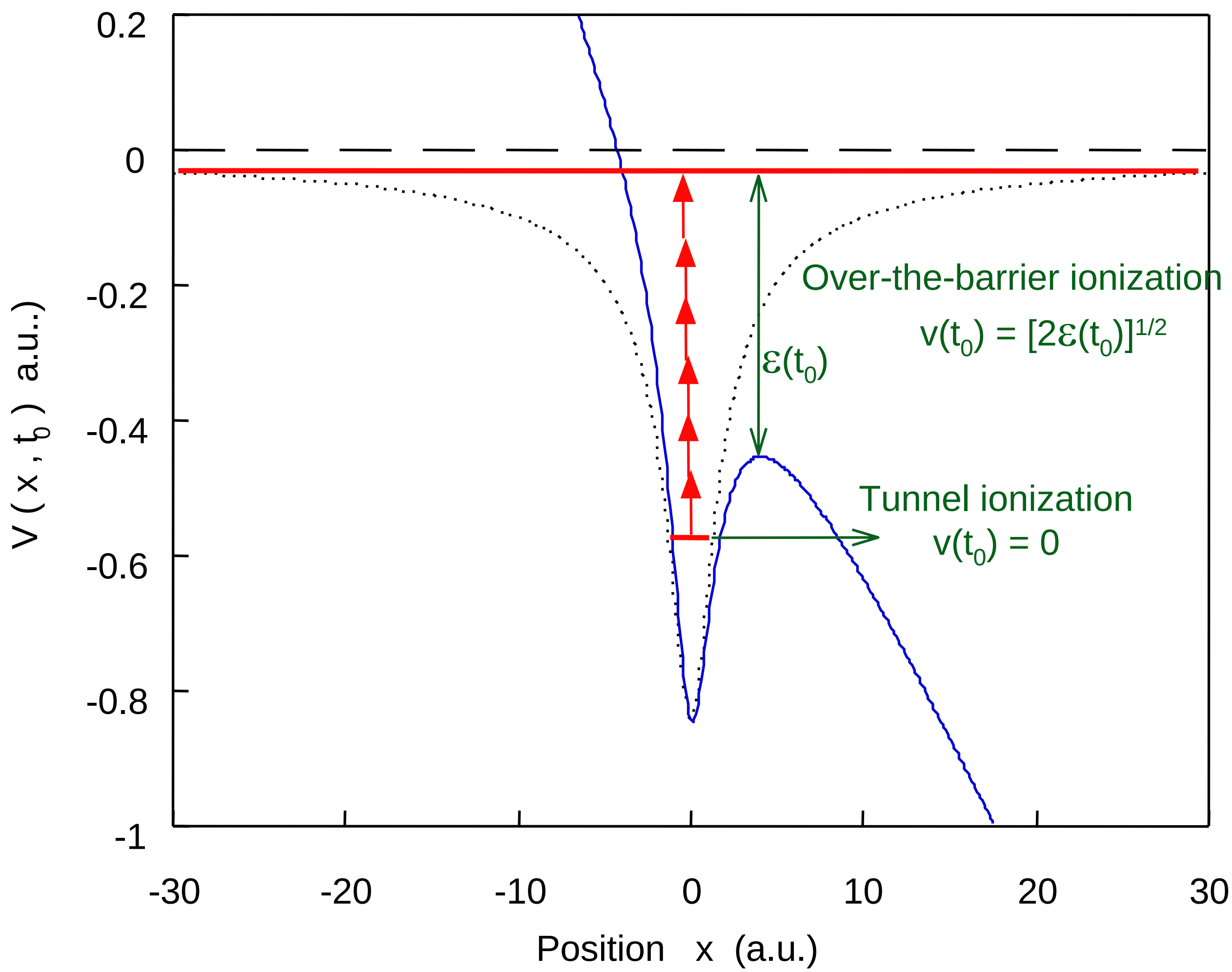
In matrix form: $\mathbf{M}(\lambda)\mathbf{S}(\lambda) = a_\lambda \mathbf{S}(\lambda)$; $\{S_\nu(\lambda)\}$: discrete basis ;
$\lambda \in C$: free parameter

With elements:

$$\begin{cases} M_{i,i}=\sqrt{c+x_i^2}\left(\dfrac{1}{\delta x^2}+\lambda\right), \\ M_{i,i+1}=-\sqrt{c+x_i^2}\left(\dfrac{1}{2\,\delta x^2}\right), \\ M_{i+1,i}=-\sqrt{c+x_{i+1}^2}\left(\dfrac{1}{2\,\delta x^2}\right), \end{cases}$$

Diagonalizing **M**, one gets the S_ν and α_ν

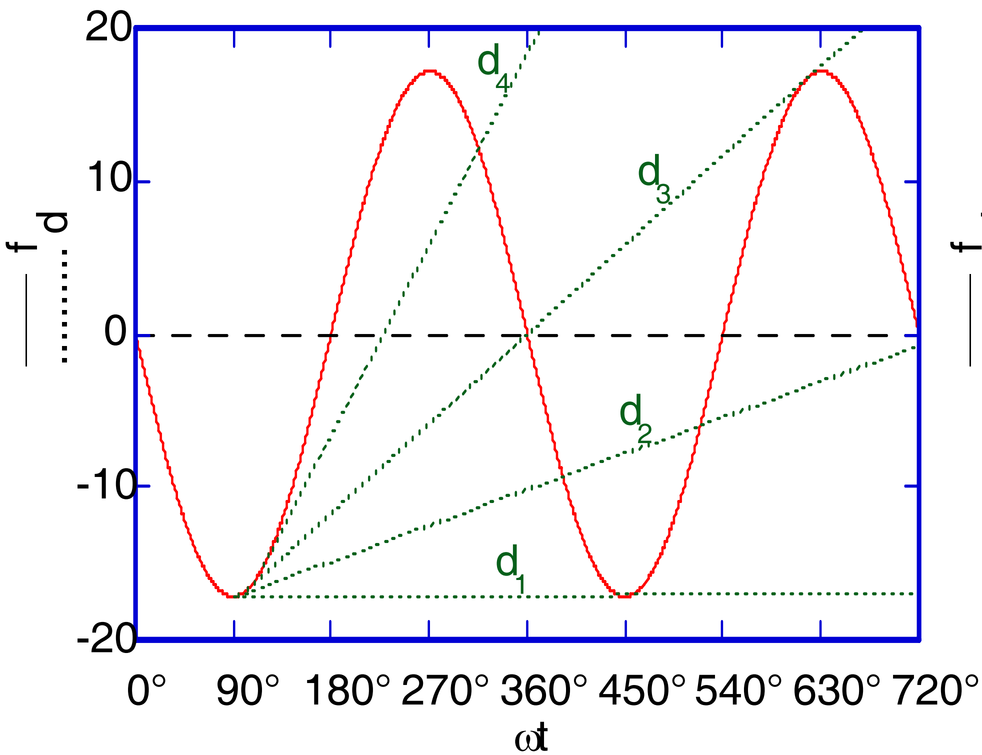
Classical : Electron resonantly excited in Rydberg state is well over the barrier.
When released in the continuum, its initial velocity is non-zero



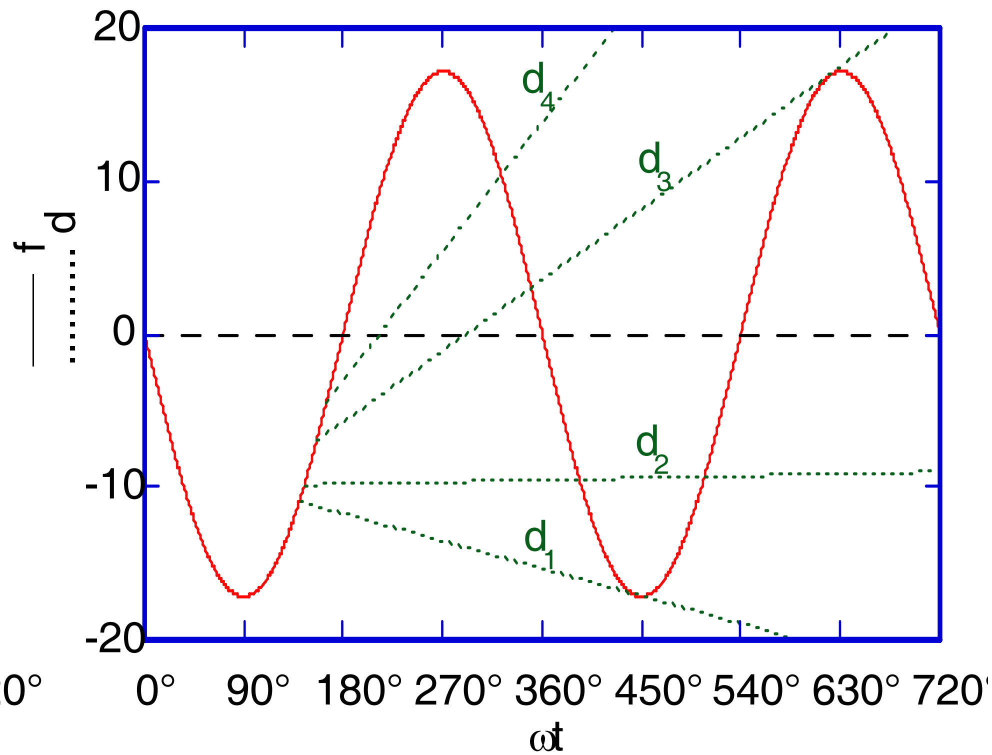
It can nevertheless come back to the origin and experience multiple recollisions

$$x(t)=\frac{E_0}{\omega^2}\left[\sin(\omega t)-\sin(\omega t_0)\right]+(t-t_0)\left[-\frac{E_0}{\omega}\cos(\omega t_0)\pm\sqrt{2\varepsilon(t_0)}\right]$$

$V(t_0) = 0$



$V(t_0) = [2E_{kin}(t_0)]^{1/2}$



N.B. Periodic trajectory (d_1):

E_{kin} = 0 when returning at x = 0

N.B. Periodic trajectory (d_2):

$E_{kin} \neq 0$ when returning at x = 0

An example of (almost) periodic trajectory (d_2)

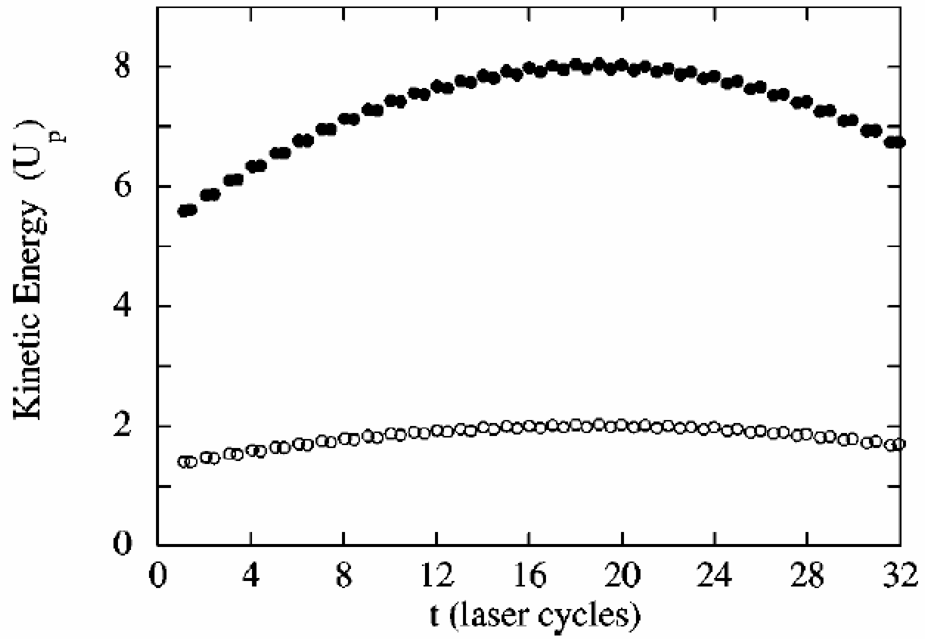


FIG. 7. Kinetic energies as a function of return times t_1 of an electron following the trajectory labeled A in Fig. 6(b). Open circles, return kinetic energy at $x=0$, see Eq. (17); filled circles, final kinetic energy after recollision, see Eq. (18).

Role of multiple recollisions

At each recollision, the electron can either:

- be elastically scattered, staying available for new recollisions...
- or recombine with the nucleus, giving rise to harmonic generation...
- or experience an inelastic collision, with final ejection energy:

$$5.5\ U_p < E_{kin} < 9.2\ U_p$$

$\Rightarrow$the relative probability for a given electron of being ejected within this energy range grows with the number of recollisions, i.e. with the pulse duration...

NB: if they are ejected from the ground state via tunnelling, the electrons experiencing large numbers of recollisions revisit the origin with a low velocity. They end mostly with kinetic energies below 2 U_p .

Dependence of the magnitudes of the enhancements on the duration of the pulse at resonance

Doubling the pulse duration: (16 → 32 cycles) ≈ double the yields

Ratios of areas under peaks:

$R_{(0\rightarrow 12)} = A_{32}/A_{16} \approx 1.95$

$R_{(0\rightarrow 5.5)} = A_{32}/A_{16} \approx 1.84$

$R_{(5.5\rightarrow 9.2)} = A_{32}/A_{16} \approx 3.26$

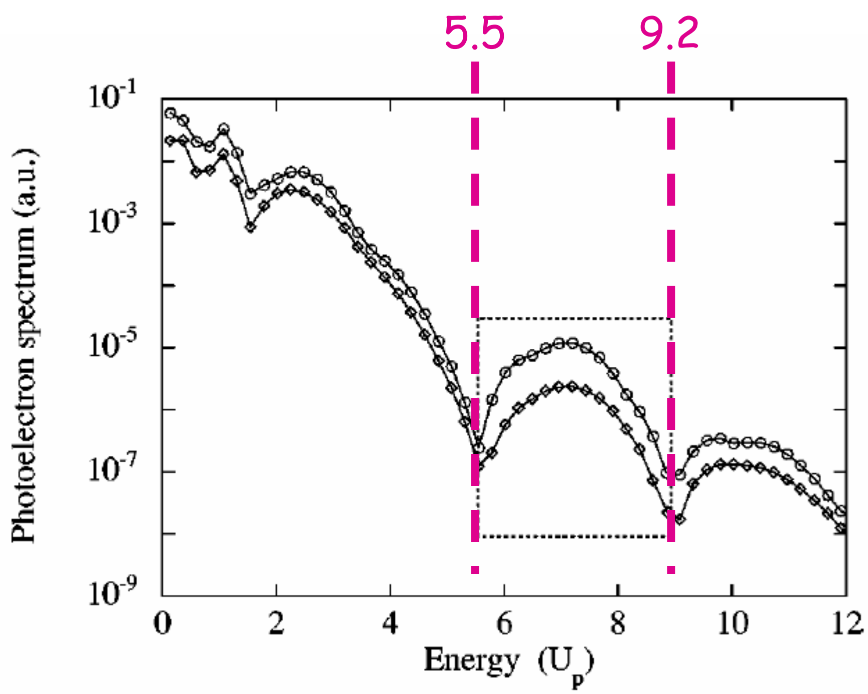


FIG. 8. Photoelectron spectra for the long-range potential Eq. (1), for $I = 1.145\times 10^{14}$ W cm^{-2} and for two different pulse durations. Open circles, 32 cycles; diamonds, 16 cycles.

Role of the pulse duration at resonance

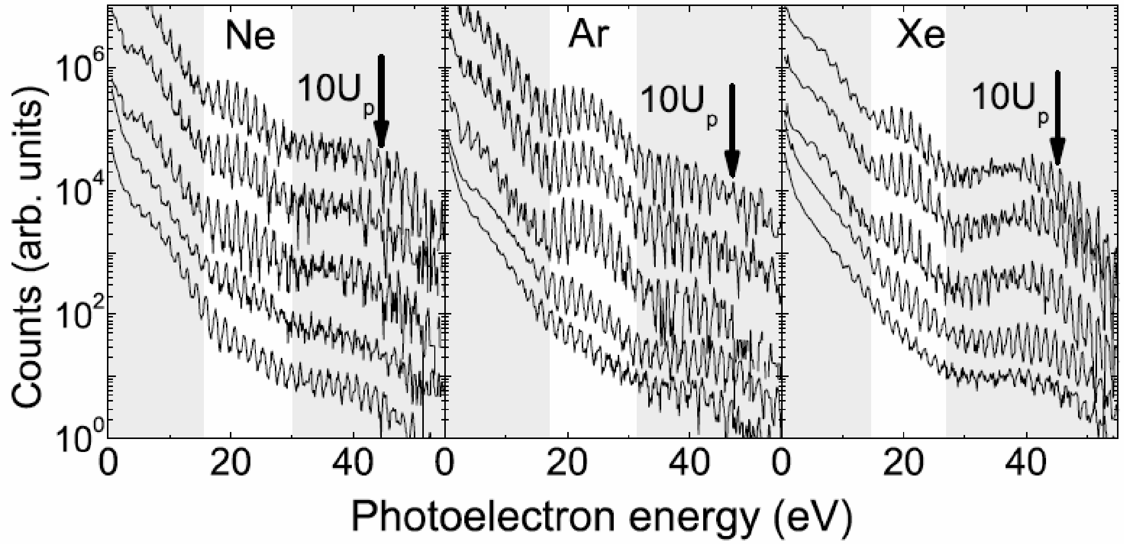


FIG. 1: ATI spectra of Ne, Ar, Xe for different pulse durations. For the curves from bottom to top, the pulse durations are 14, 24, 45, 60 and 100 fs, respectively. Notice that a clear enhancement of ATI peaks develops as the pulse duration is increased, which is highlighted in the figure. For visual convenience, the curves at different pulse durations are separated in vertical direction, i.e. the relative ratio of curves has no meaning. The laser intensities are chosen such that the enhancements are optimized, i.e. the LIS are right on resonance. This is the case at 7.5×10^{13}W/cm^2 for Ne, 8.0×10^{13}W/cm^2 for Ar, and 7.5×10^{13}W/cm^2 for Xe.

Courtesy of G. Paulus *et al.* 2006, to appear

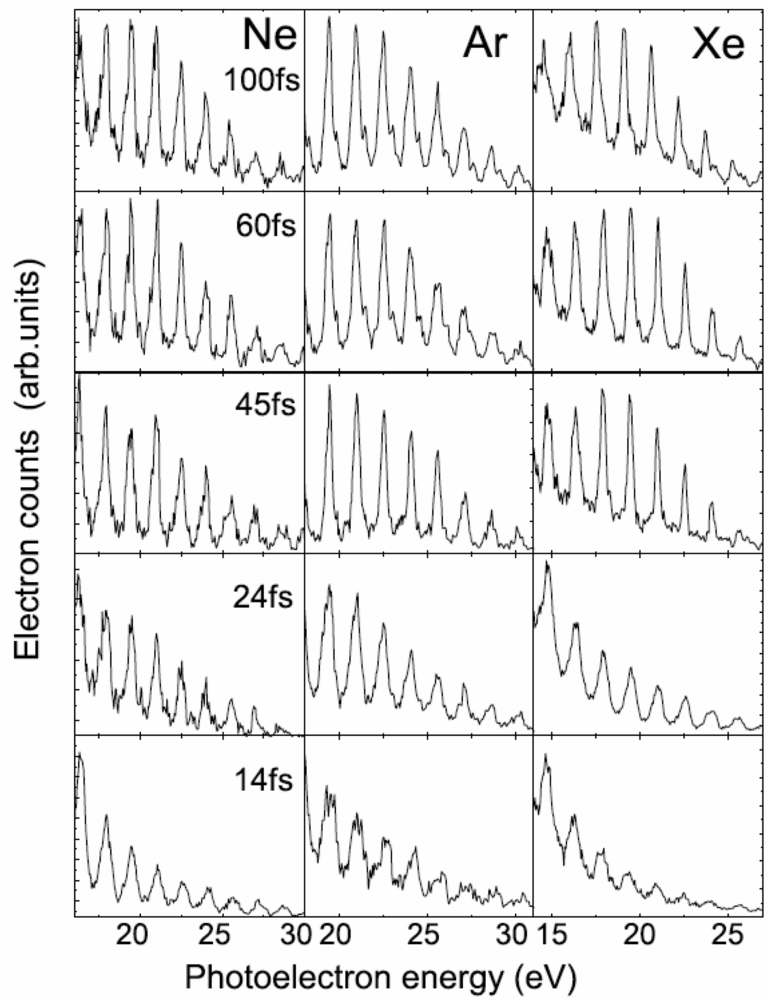
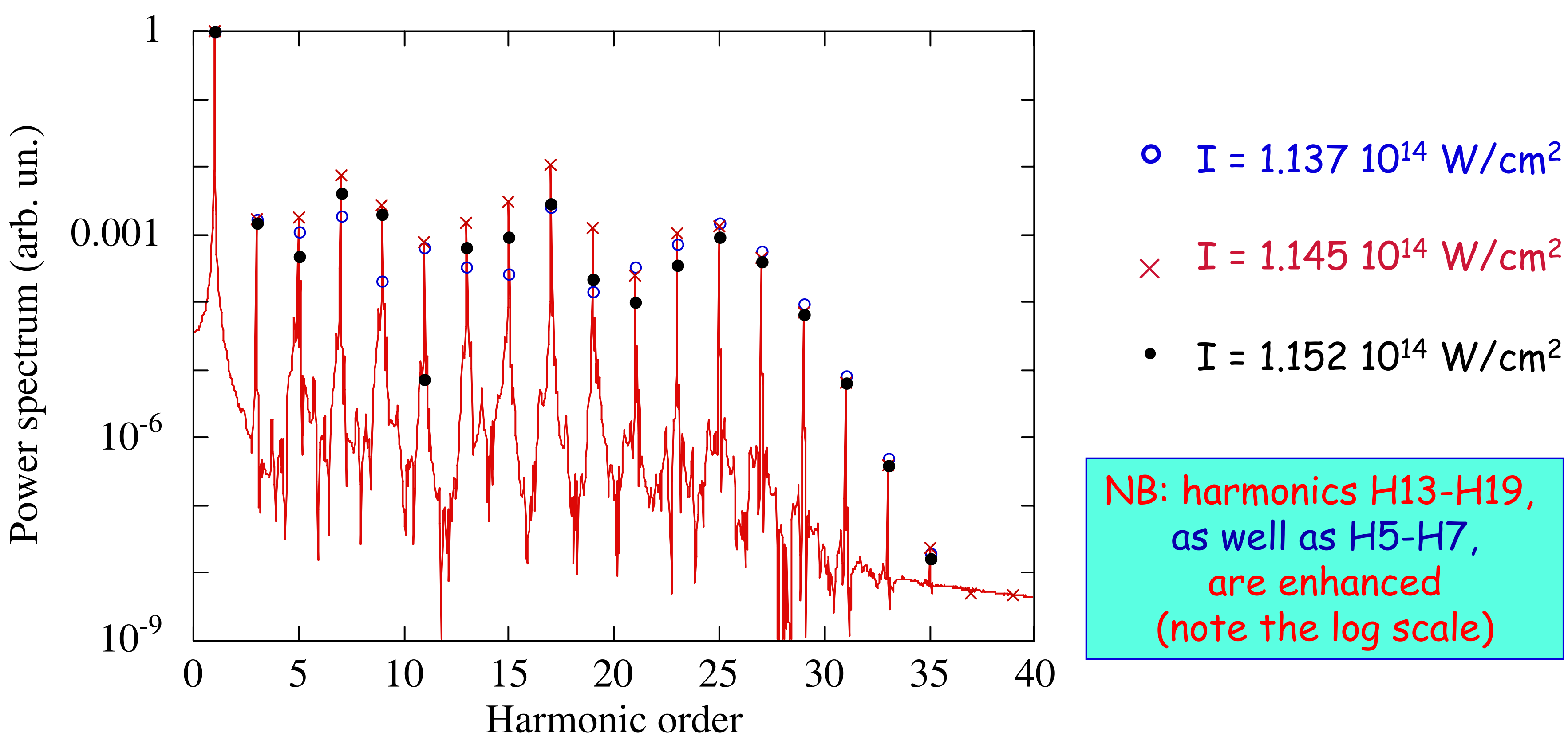


FIG. 2: Photoelectron spectra of inert gases in the energy region where the resonance-like enhancement occurs. The experimental parameters are the same as in Fig.1.

ATTO-06 Courtesy of G. Paulus *et al.* 2006, to appear

Role of resonances in harmonic spectra ?

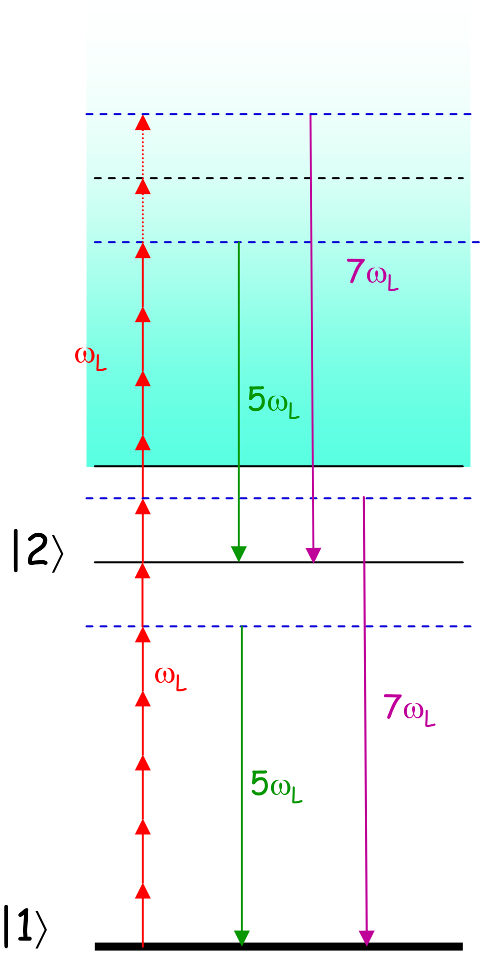
Model: (« soft-Coulomb », Ar-like) at intensities close to the 14-photon resonance on the state n = 6 (I_{res} = 1.145 10^{14} W/cm^2)



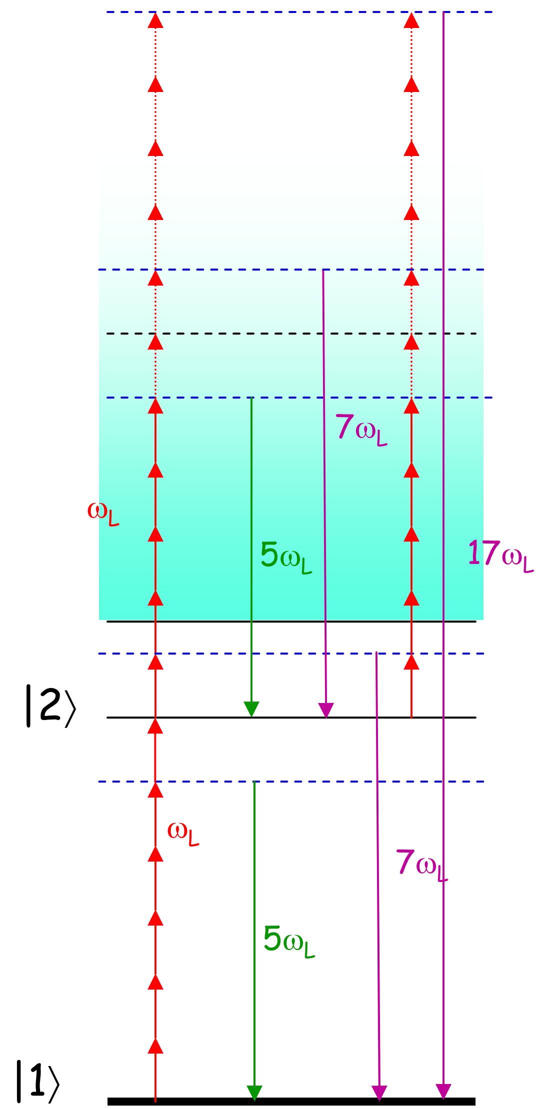
NB: harmonics H13-H19,
as well as H5-H7,
are enhanced
(note the log scale)

Taïeb *et al.* PRA **68**, 033403 (2003)

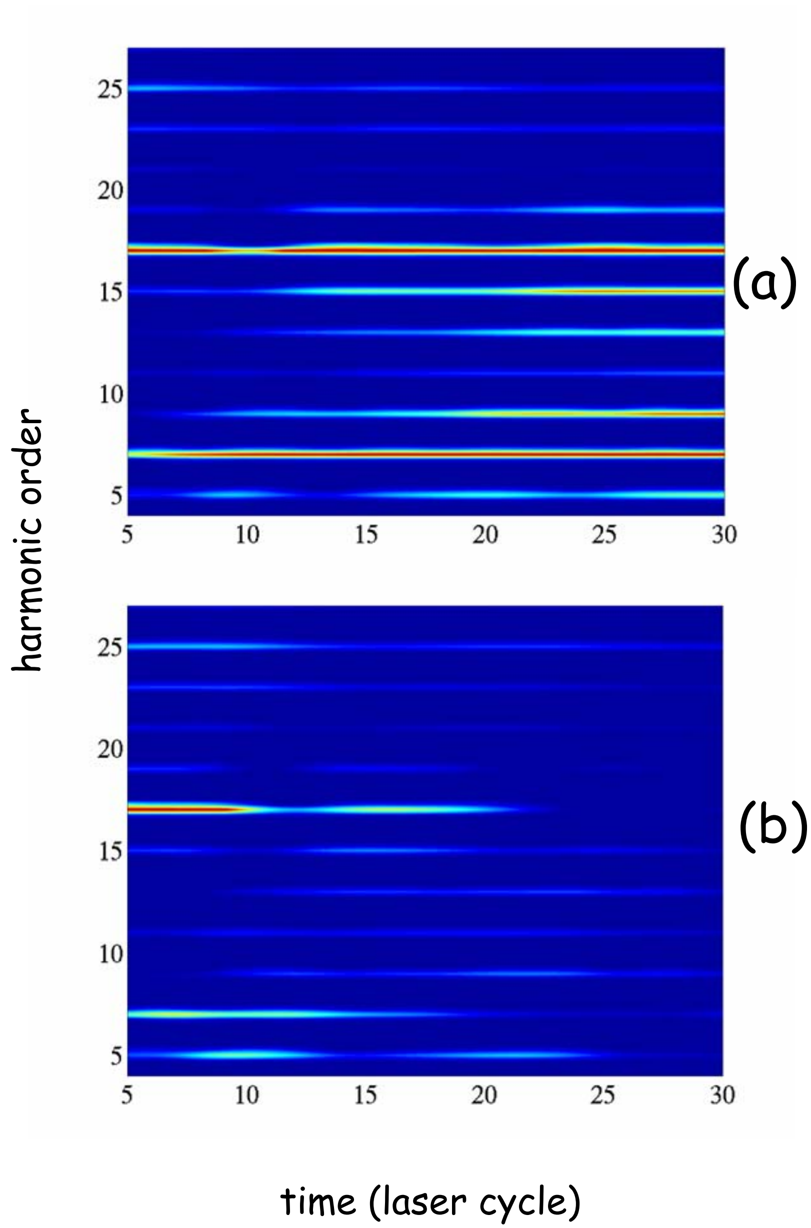
An example: a 6-photon resonance can be responsible for the enhancements of H5 and H7 (and higher…)
Taïeb *et al.* (PRA **68** 033403, 2003)



The larger polarizability of the excited state can enhance H5 and H7



Wavelet-like (Gabor) analysis of the time dependence of harmonic emission rates



Constant amplitude field after one-cycle turn-on

Gabor transform coefficients vs.time:

(a) resonant intensity (n=6)

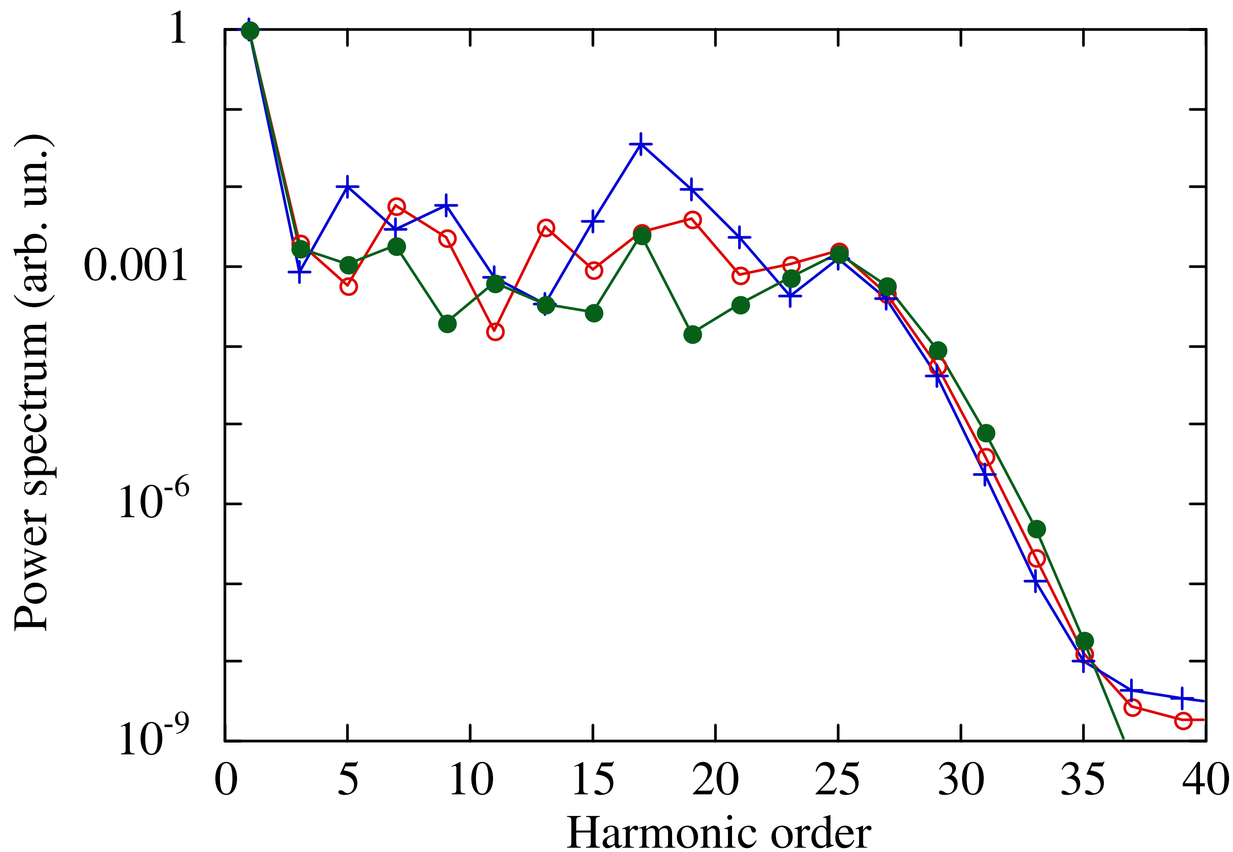
$I = 1.145 \times 10^{14}$ W/cm^2

(b) non-resonant intensity

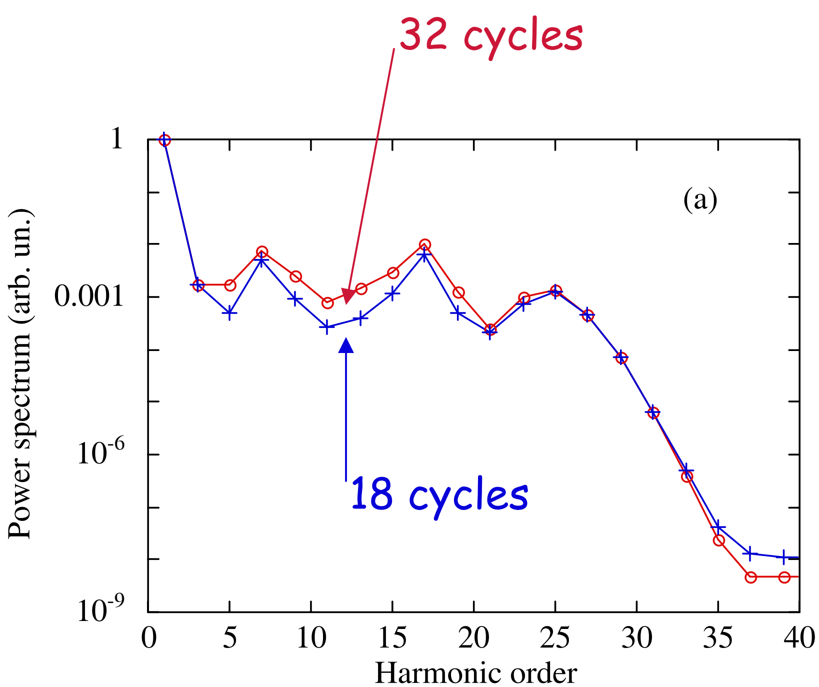
$I = 1.137 \times 10^{14}$ W/cm^2

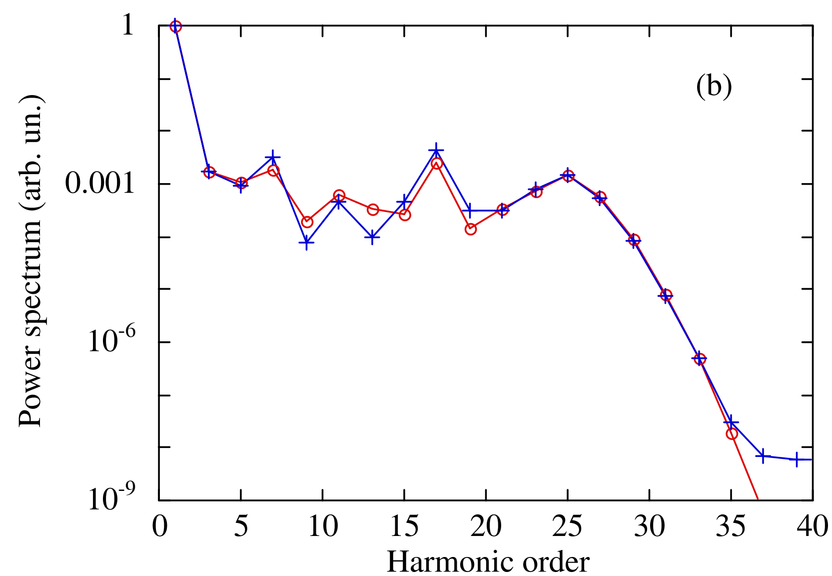
- H7 and H17 are dominant. Part of them are emitted when, after being ejected from the state n=6, electrons recombine either in n=6 (H7) or n=0 (H17) states.
- In non-resonant conditions (b), harmonic emission declines in time because of the depletion of the ground state.
- At resonance, (a), emission rates keep growing because of *multiple* recollisions.

Scaled harmonic spectra in the vicinity of the n = 3 (13- photon) resonance

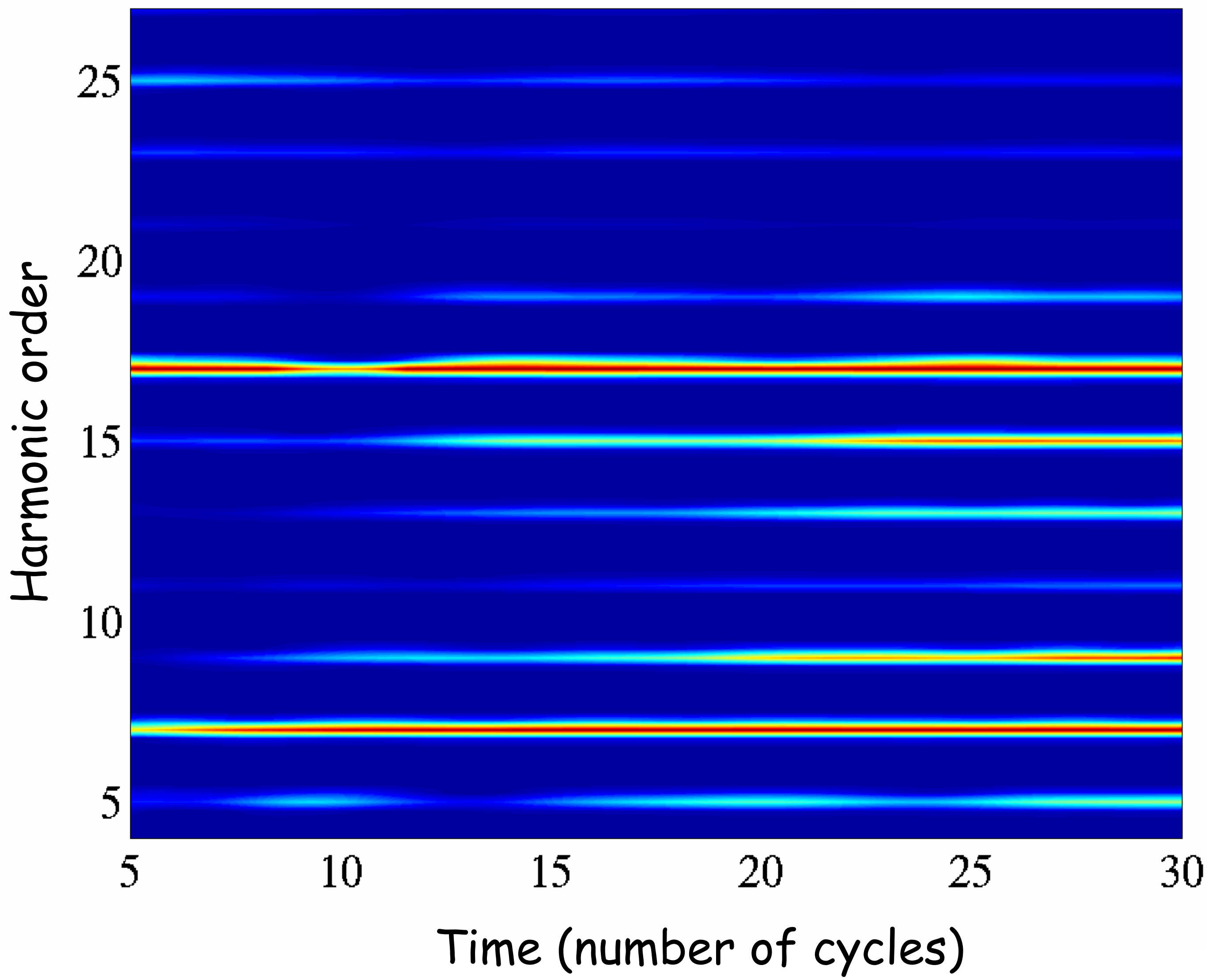


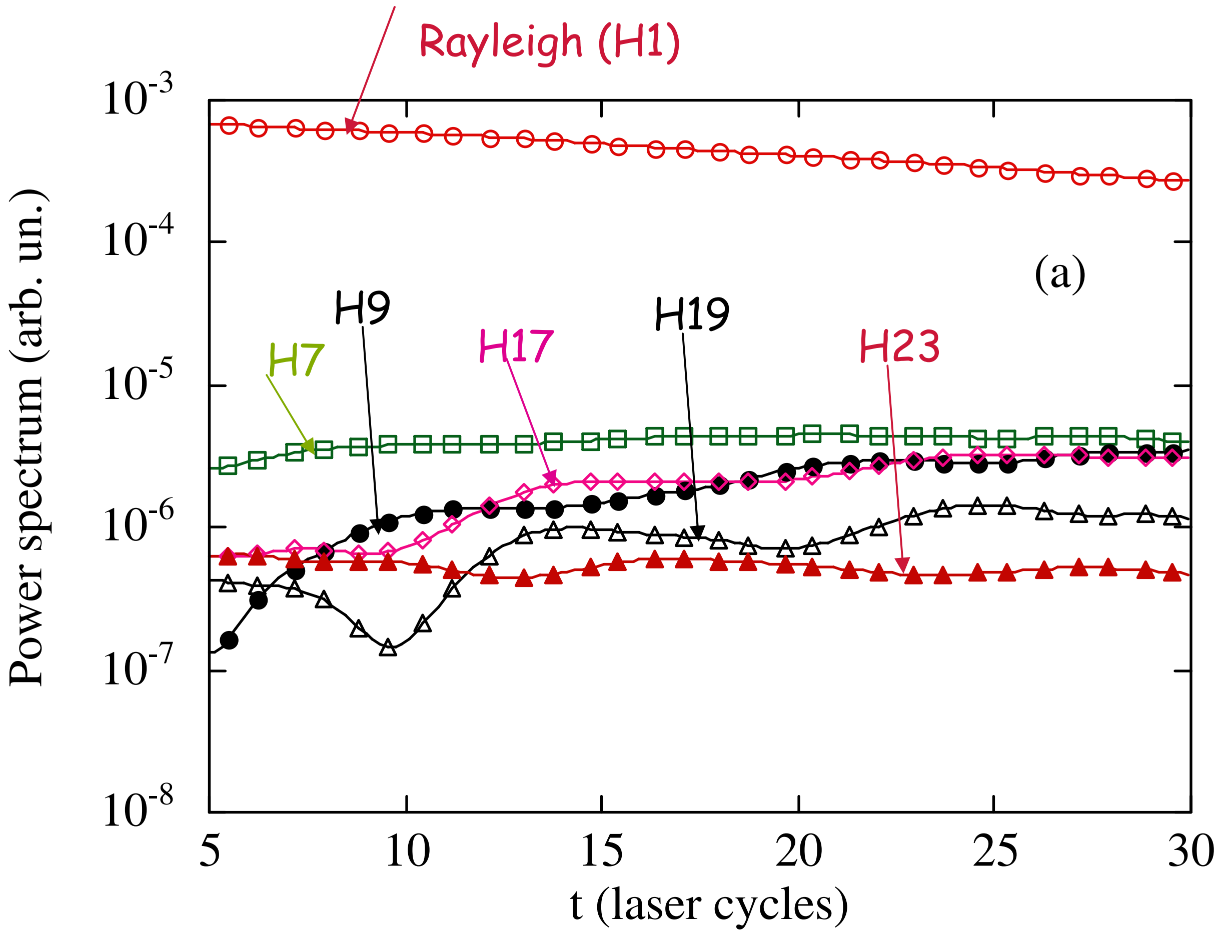
Role of the pulse duration
Scaled harmonic spectra in the vicinity of the n = 6 (14- photon) resonance
(a) At resonance
(b) At a slightly different intensity





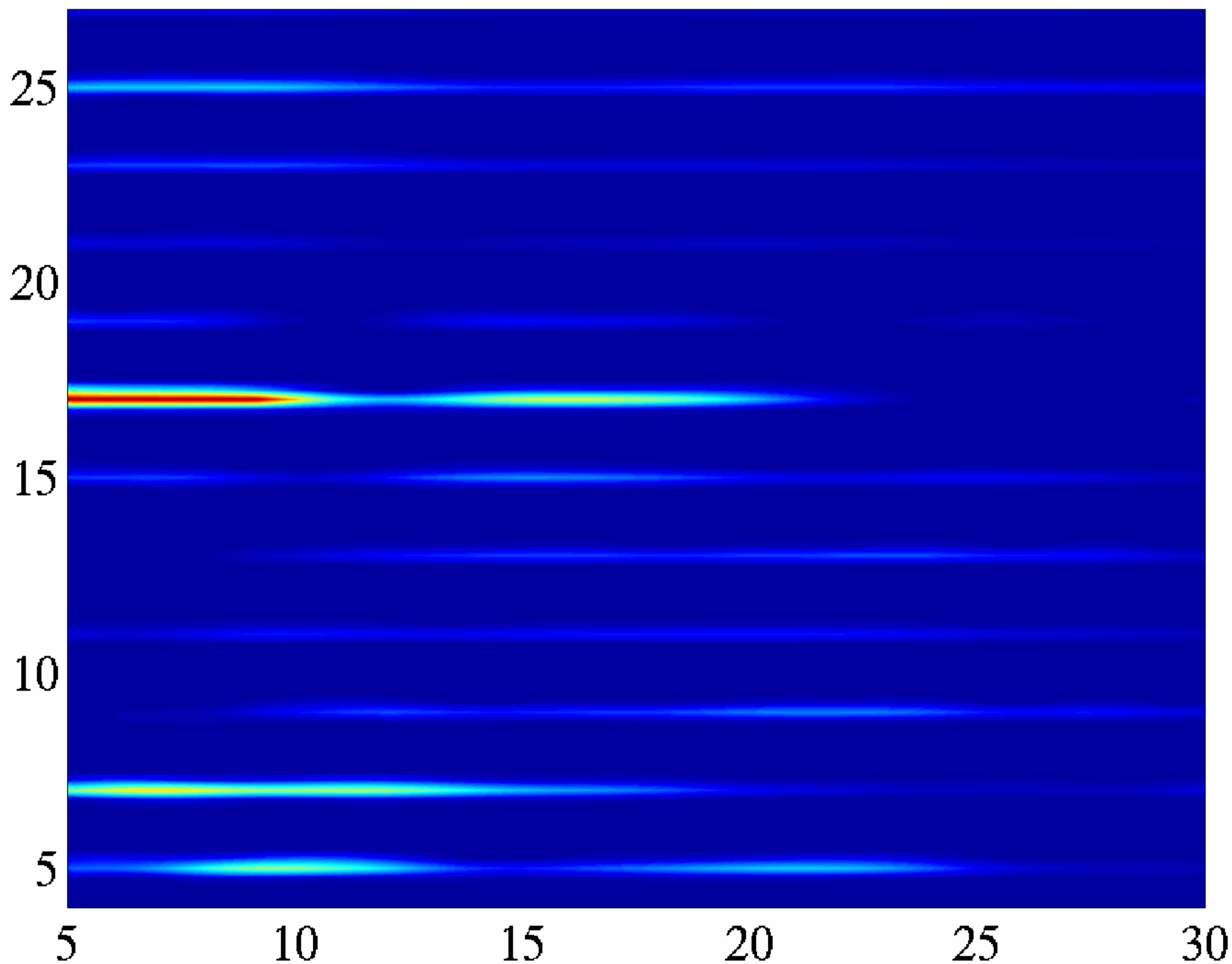
Constant intensity pulse. Time dependence of the harmonic emission
(a) at resonance on n = 6 state

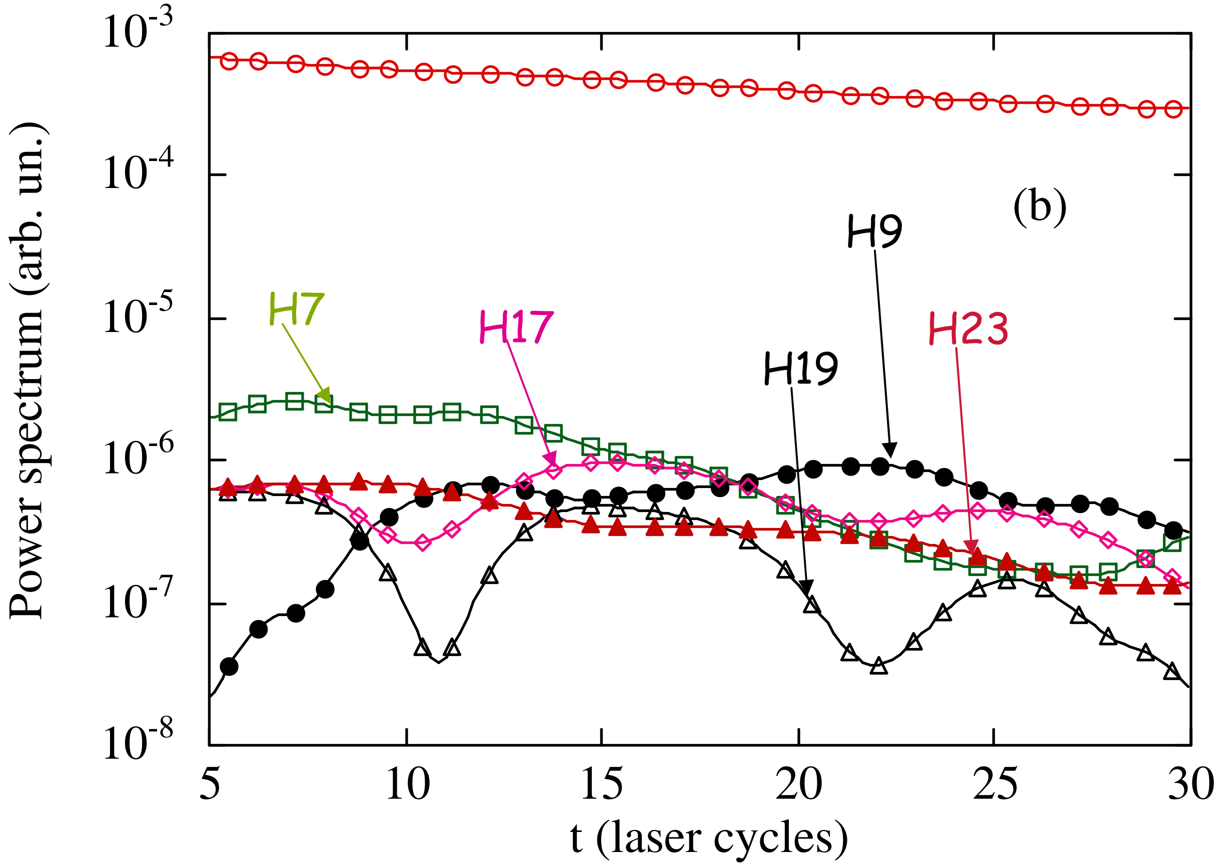




Rayleigh (H1)
(a)
H9
H17
H19
H23
H7
Power spectrum (arb. un.)
t (laser cycles)
10^{-3}
10^{-4}
10^{-5}
10^{-6}
10^{-7}
10^{-8}
5
10
15
20
25
30

Constant intensity pulse. Time dependence of the harmonic emission
(b) Slightly off resonance





Power spectrum (arb. un.)
t (laser cycles)
(b)
H7
H17
H19
H9
H23
10^{-3}
10^{-4}
10^{-5}
10^{-6}
10^{-7}
10^{-8}
5
10
15
20
25
30

Conclusions

Multiphoton resonances involving excited states and/or laser-induced states can induce enhancements observed in the high-energy part of ATI spectra and in the plateau of harmonic spectra

Long-range potential:
enhancements related to resonances on Rydberg states located above the barrier.
Confirmed by quantum Floquet and classical analysis.
The magnitude of the enhancement grows non-linearly with the pulse duration (see Paulus experiments)
Laser-Induced States?
Channel closing?

Short-range potentials:
Enhancements related to resonances on Laser-Induced States. The question of the role of «channel-closings» is not settled yet

An open question: Will these results be confirmed in 3-D calculations?.
Preliminary results in H and Paulus' data are encouraging.