Lecture Series Buenos Aires

18-3-2024 until 22-3-2024

Lecture M2 – Non-perturbative physics

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TABLE XL An abbreviated list of the CODATA recommended values of the fundamental constants of physics and chemistry based on the 2010 adjustment.

Quantity	Symbol	Numerical value	Unit	Relative std. uncert. u_r
speed of light in vacuum	c, c_0	299 792 458	m s^{-1}	exact
magnetic constant	μ_0	$4\pi \times 10^{-7}$	N A^{-2}	
		$= 12.566\,370\,614... \times 10^{-7}$	N A^{-2}	exact
electric constant $1/\mu_0 c^2$	ϵ_0	$8.854\,187\,817... \times 10^{-12}$	F m^{-1}	exact
Newtonian constant of gravitation	G	$6.673\,84(80) \times 10^{-11}$	m^3 kg^{-1} s^{-2}	1.2×10^{-4}
Planck constant	h	$6.626\,069\,57(29) \times 10^{-34}$	J s	4.4×10^{-8}
$h/2\pi$	$\hbar$	$1.054\,571\,726(47) \times 10^{-34}$	J s	4.4×10^{-8}
elementary charge	e	$1.602\,176\,565(35) \times 10^{-19}$	C	2.2×10^{-8}
magnetic flux quantum $h/2e$	Φ_0	$2.067\,833\,758(46) \times 10^{-15}$	Wb	2.2×10^{-8}
conductance quantum $2e^2/h$	G_0	$7.748\,091\,7346(25) \times 10^{-5}$	S	3.2×10^{-10}
electron mass	m_e	$9.109\,382\,91(40) \times 10^{-31}$	kg	4.4×10^{-8}
proton mass	m_p	$1.672\,621\,777(74) \times 10^{-27}$	kg	4.4×10^{-8}
proton-electron mass ratio	m_p/m_e	1836.152 672 45(75)		4.1×10^{-10}
fine-structure constant $e^2/4\pi\epsilon_0\hbar c$	α	$7.297\,352\,5698(24) \times 10^{-3}$		3.2×10^{-10}
inverse fine-structure constant	α^{-1}	137.035 999 074(44)		3.2×10^{-10}
Rydberg constant $\alpha^2 m_e c/2h$	R_∞	10 973 731.568 539(55)	m^{-1}	5.0×10^{-12}
Avogadro constant	N_A, L	$6.022\,141\,29(27) \times 10^{23}$	mol^{-1}	4.4×10^{-8}
Faraday constant $N_A e$	F	96 485.3365(21)	C mol^{-1}	2.2×10^{-8}
molar gas constant	R	8.314 4621(75)	J mol^{-1} K^{-1}	9.1×10^{-7}
Boltzmann constant R/N_A	k	$1.380\,6488(13) \times 10^{-23}$	J K^{-1}	9.1×10^{-7}
Stefan-Boltzmann constant $(\pi^2/60)k^4/\hbar^3c^2$	σ	$5.670\,373(21) \times 10^{-8}$	W m^{-2} K^{-4}	3.6×10^{-6}
Non-SI units accepted for use with the SI				
electron volt (e/C) J	eV	$1.602\,176\,565(35) \times 10^{-19}$	J	2.2×10^{-8}
(unified) atomic mass unit $\frac{1}{12}m(^{12}\text{C})$	u	$1.660\,538\,921(73) \times 10^{-27}$	kg	4.4×10^{-8}

Working with Atomic Units

Bohr radius: $a_0 = 4\pi\varepsilon_0 \dfrac{\hbar^2}{m_e e^2}$ = 5.2917721092(17)×10^{-11} m

Unit of energy: $E_H = \dfrac{m_e e^4}{4\pi\varepsilon_0 \hbar^2}$ = 4.359 744 34(19)×10^{-18} J = 27.2107 eV

Unit of velocity: $\dfrac{m_e v_0^2}{2} = \dfrac{E_H}{2} \Rightarrow v_0 = \dfrac{e^2}{4\pi\varepsilon_0 \hbar}$

= 2.187 691 263 79(71)x 10^{6} m s^{-1}

Unit of time: $\tau_0 = \dfrac{a_0}{v_0} = \dfrac{(4\pi\varepsilon_0)^2 \hbar^3}{m_e e^4}$

= 2.418 884 326 502(12) x 10^{-17} s

Atomic units are those units where $\hbar$ = $4\pi\varepsilon_0$= m_e = e = 1

Nice consequences

Bohr radius: $a_0 = 4\pi\varepsilon_0 \frac{\hbar^2}{m_e e^2}$ = 1

Unit of energy: $E_H = \frac{m_e e^4}{4\pi\varepsilon_0 \hbar^2}$ = 1

Unit of velocity: $\frac{m_e v_0^2}{2} = \frac{E_H}{2} => v_0 = \frac{e^2}{4\pi\varepsilon_0 \hbar}$ = 1

Unit of time: $\tau_0 = \frac{a_0}{v_0} = \frac{(4\pi\varepsilon_0)^2 \hbar^3}{m_e e^4}$ = 1

Etc…

In other words: the moment that we choose the afore-mentioned units as our basic units, it follows that $\hbar$ = $4\pi\varepsilon_0$= m_e = e = 1

Useful conversions

The atomic unit of fieldstrength corresponds to the field in a hydrogen atom at first Bohr orbit

How strong is this field (V/cm)?

$$F(r) = -eE(r) = \frac{1}{4\pi\varepsilon_0}\frac{-e^2}{r^2} \qquad E(r) = \frac{1}{4\pi\varepsilon_0}\frac{e}{r^2}$$

Fill in: $E(a_0)$ = 5.14×10^{11} V/m = 5.14×10^{9} V/cm

Which laser intensity does this correspond to?

$$I = \frac{1}{2}\varepsilon_0 c E^2$$

c=2.998×10^{8} m/s

Fill in: $E(a_0)$ = 3.51×10^{20} W/m^2 = 3.51×10^{16} W/cm^2

Interactions of atoms with strong laser fields

1905: Simple photo-effect

132

6. ***Über einen die Erzeugung und Verwandlung des Lichtes betreffenden heuristischen Gesichtspunkt;***
von A. Einstein.

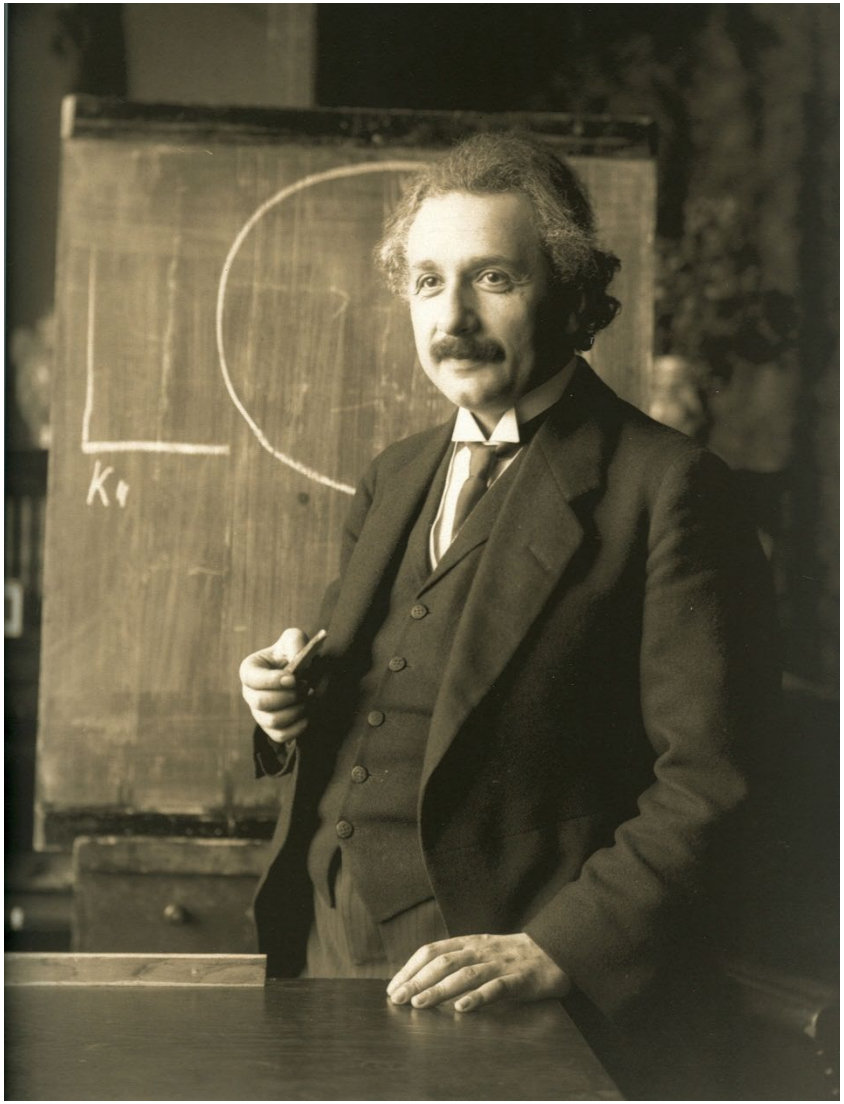
Zwischen den theoretischen Vorstellungen, welche sich die Physiker über die Gase und andere ponderable Körper gebildet haben, und der Maxwellschen Theorie der elektromagnetischen Prozesse im sogenannten leeren Raume besteht

146 *A. Einstein.*

versehenes Elektron wird, wenn es die Oberfläche erreicht hat, einen Teil seiner kinetischen Energie eingebüßt haben. Außerdem wird anzunehmen sein, daß jedes Elektron beim Verlassen des Körpers eine (für den Körper charakteristische) Arbeit P zu leisten hat, wenn es den Körper verläßt. Mit der größten Normalgeschwindigkeit werden die unmittelbar an der Oberfläche normal zu dieser erregten Elektronen den Körper verlassen. Die kinetische Energie solcher Elektronen ist

$$\frac{R}{N}\beta\nu - P.$$

Ist der Körper zum positiven Potential Π geladen und von Leitern vom Potential Null umgeben und ist Π eben imstande, einen Elektrizitätsverlust des Körpers zu verhindern



A. Einstein, Annalen der Physik 322, 132 (1905)

1930: Prediction of 2-photon absorption

Maria-Goeppert Mayer performed her PhD in Goettingen, with Max Born as her PhD supervisor, predicting the possibility that 2-photon absorption might be possible
However, light sources allowing to validate this prediction were unavailable at the time

In 1963 she became the 2nd female Nobel Laureate in physics, for her work on the nuclear shell model

Maria-Goeppert Mayer

Multi-Photon Ionization

SOVIET PHYSICS JETP VOLUME 23, NUMBER 1 JULY, 1966

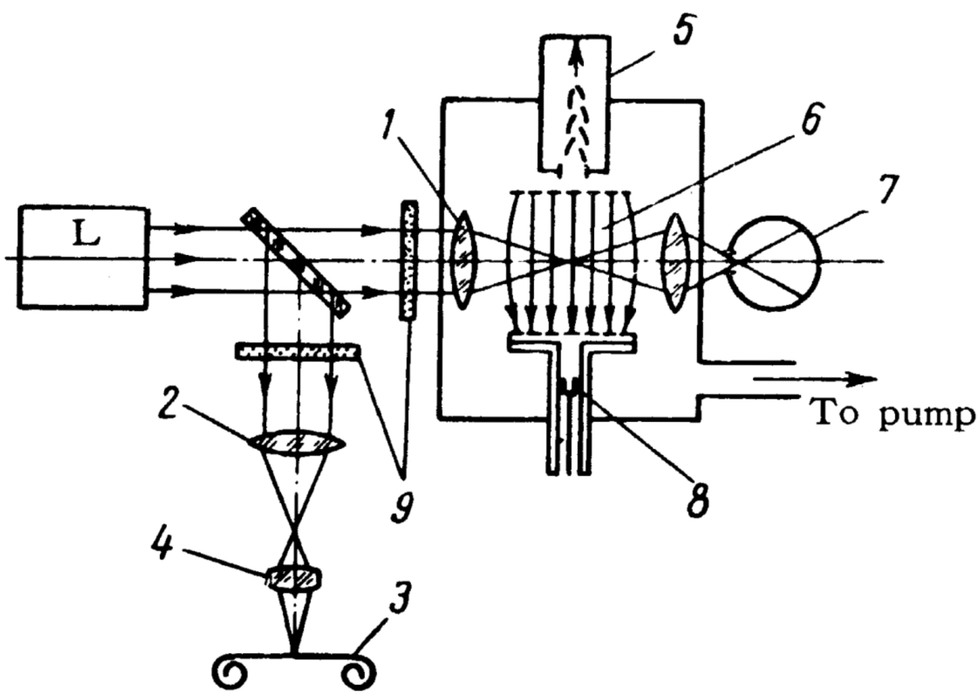
MANY-PHOTON IONIZATION OF THE XENON ATOM BY RUBY LASER RADIATION

G. S. VORONOV and N. B. DELONE

P. N. Lebedev Physics Institute, Academy of Sciences, U.S.S.R.

Submitted to JETP editor August 27, 1965

J. Exptl. Theoret. Phys. (U.S.S.R.) 50, 78-84 (January, 1966)



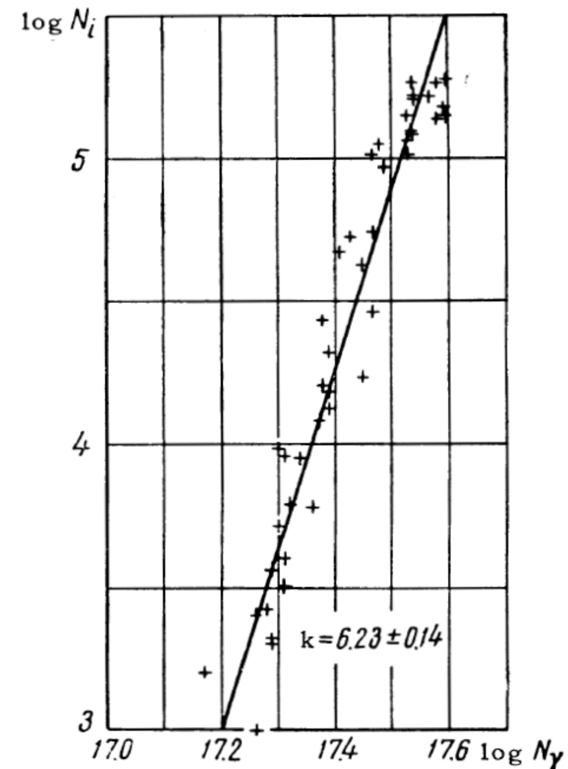


FIG. 2. Dependence of the number of ions formed N_i on the number of photons N_γ which have traversed the focusing region.

Multi-Photon Ionization

According to perturbation theory, the rate of n-photon absorption is given as

laser intensity

final state density

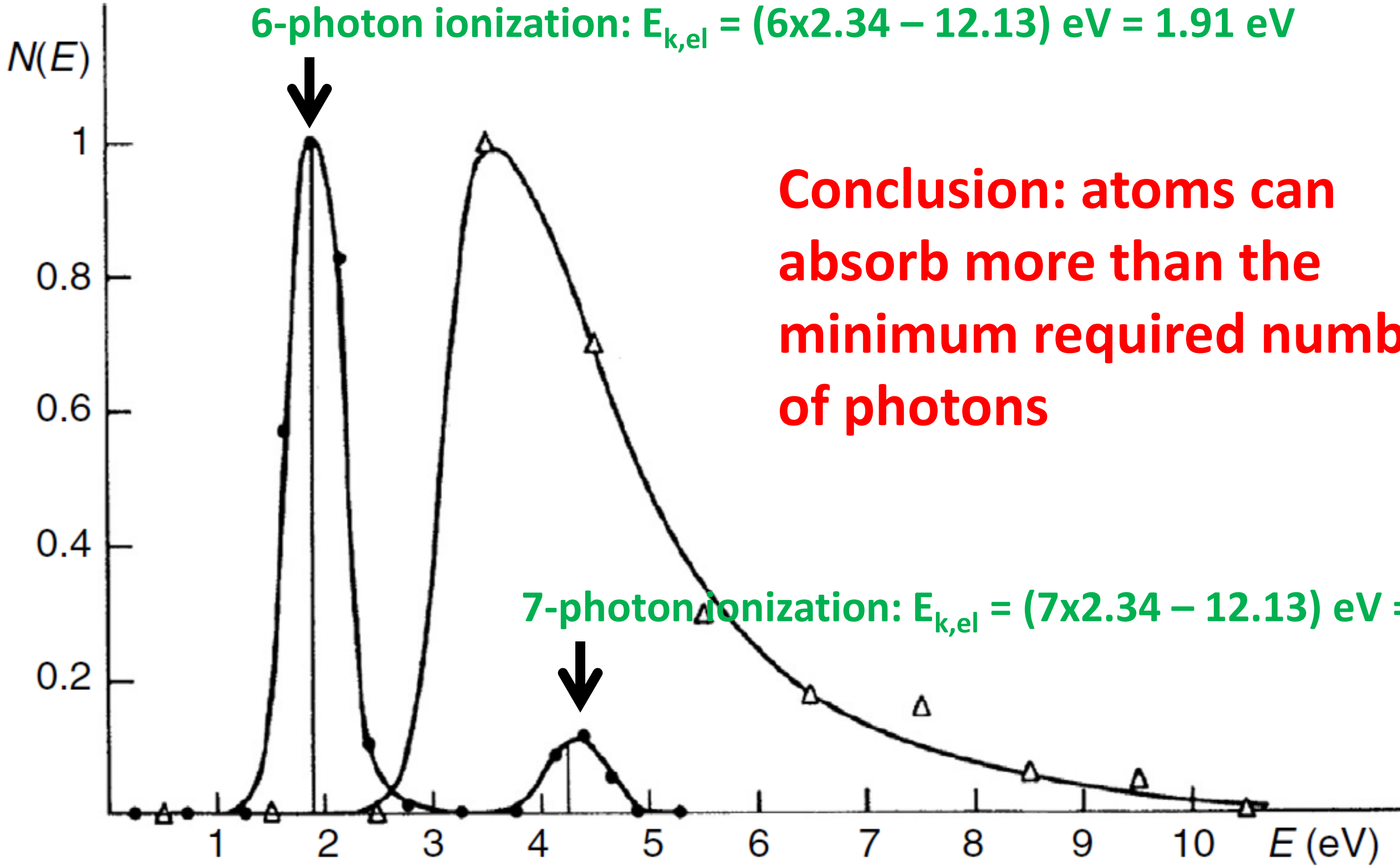
$$W_{fi}^{(n)} = \frac{2\pi}{\hbar}(2\pi\alpha\hbar)^n I_{laser}^n \left|\tilde{T}_{fi}^{(n)}\right|^2 \rho_f(E_f)$$

N-photon matrix element

Therefore, the lowest-order perturbation theory (LOPT) ionization rate is proportional to $I_{laser}^{n_0}$, where n_0 is the minimum number of photons needed

C.J. Joachain, N.J. Kylstra and R.M. Potvliege, Atoms in Intense Laser Fields, (Cambridge University Press, 2012)

Above-Threshold Ionization



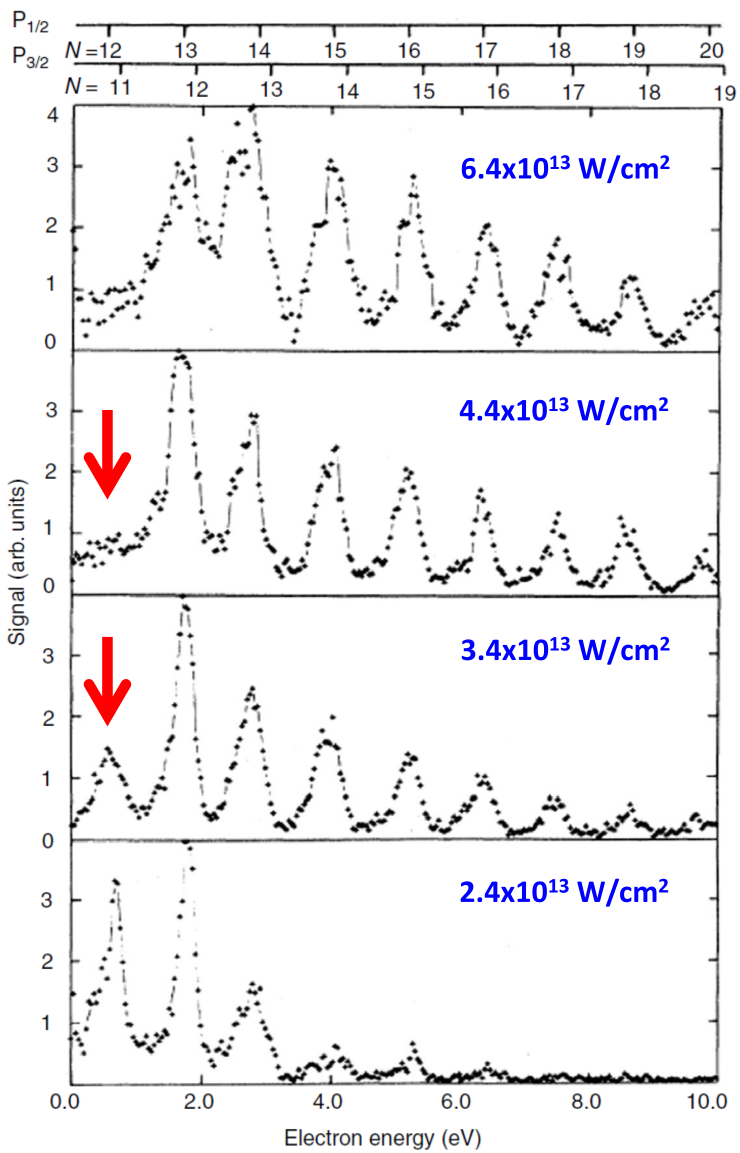
Conclusion: atoms can absorb more than the minimum required number of photons

Energy spectra of electrons produced by multiphoton ionization of xenon atoms, for two photon energies. Triangles: ω = 1.17 eV; Circles: ω =2.34 eV

P. Agostini et al., Phys. Rev. Lett. 42, 1127 (1979)

Pierre @ AMOLF (2004)

Above-Threshold Ionization



Ionization of Xe using 10 ns long, 1064 nm laser pulses

Measured ATI spectra as a function of intensity show a suppression of the lowest order ATI peak.

This shows that the magnitude of the laser electric field has a non-trivial influence on the ATI peak distribution (--> non-perturbative)

P. Kruit et al., Phys. Rev. A 28, 248 (1983)

Ponderomotive Energy

In an intense laser field, free electrons perform a wiggling motion

$$F = m_e a(t) = qE(t) = qE_0 \cos(\omega t)$$

$$v(t) = v_0 + \frac{qE_0}{m_e \omega} \sin(\omega t)$$

The kinetic energy of the free electron can be evaluated as

$$E_k(t) = \frac{1}{2} m_e v(t)^2 = \frac{1}{2} m_e v_0^2 + \frac{1}{2} \frac{q^2 E_0^2}{m_e \omega^2} sin^2(\omega t) + \frac{v_0 q E_0}{\omega} \sin(\omega t)$$

Cycle-averaged:

drift energy **ponderomotive energy U_p**

$$E_k(t) = \frac{1}{2} m_e v(t)^2 = \frac{1}{2} m_e v_0^2 + \frac{1}{4} \frac{q^2 E_0^2}{m_e \omega^2}$$

Remember Working Group 1: atomic units

In an intense laser field, free electrons perform a wiggling motion

$$F = \cancel{m_e} a(t) = \cancel{q} E(t) = \cancel{q} E_0 \cos(\omega t)$$

$$v(t) = v_0 + \frac{\cancel{q} E_0}{\cancel{m_e} \omega} \sin(\omega t)$$

The kinetic energy of the free electron can be evaluated as

$$E_k(t) = \frac{1}{2} \cancel{m_e} v(t)^2 = \frac{1}{2} \cancel{m_e} v_0^2 + \frac{1}{2} \frac{\cancel{q}^2 E_0^2}{\cancel{m_e} \omega^2} sin^2(\omega t) + \frac{v_0 \cancel{q} E_0}{\omega} \sin(\omega t)$$

Cycle-averaged:

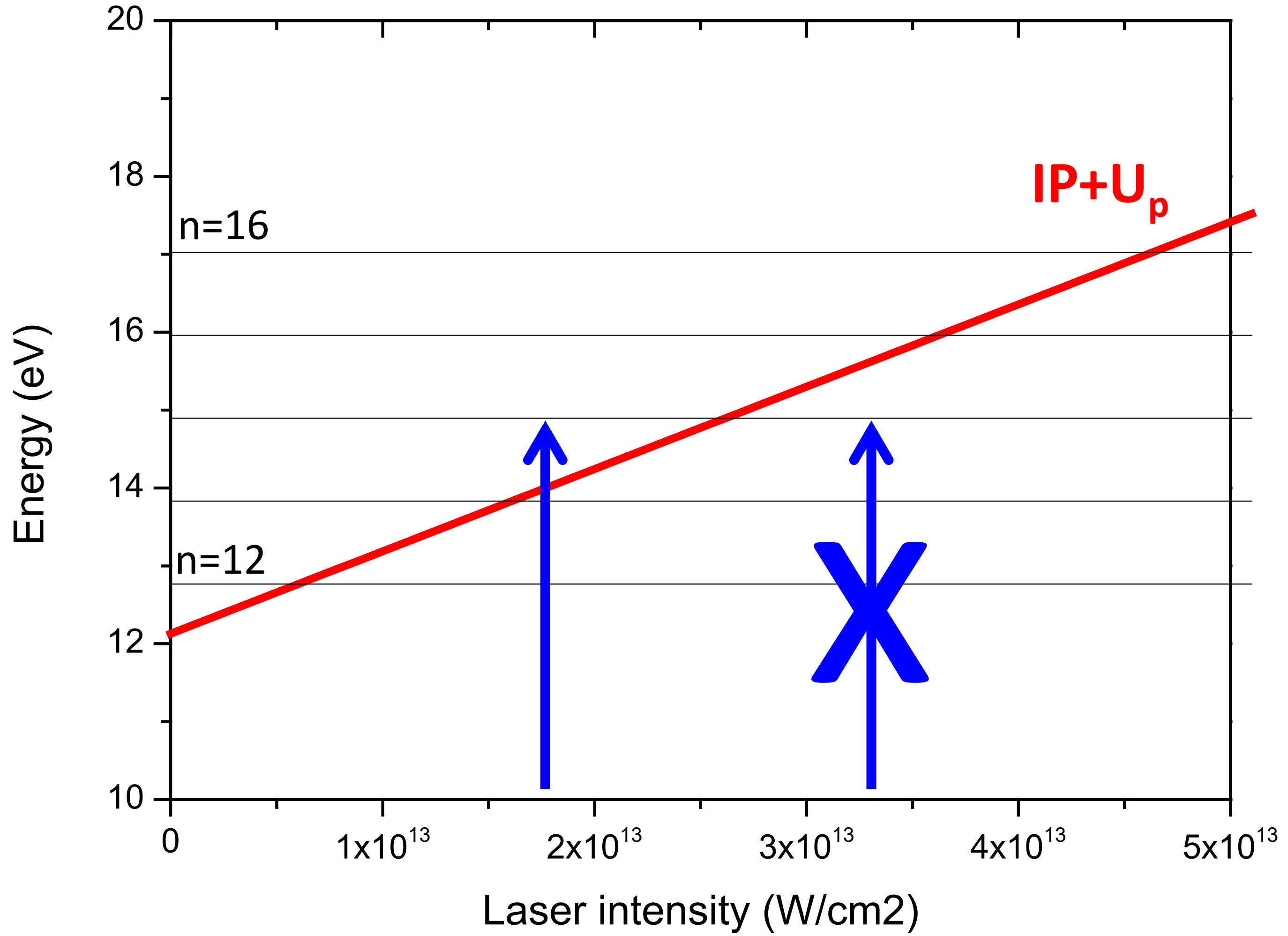
drift energy

ponderomotive energy U_p

$$E_k(t) = \frac{1}{2} \cancel{m_e} v(t)^2 = \frac{1}{2} \cancel{m_e} v_0^2 + \frac{1}{4} \frac{\cancel{q}^2 E_0^2}{\cancel{m_e} \omega^2}$$

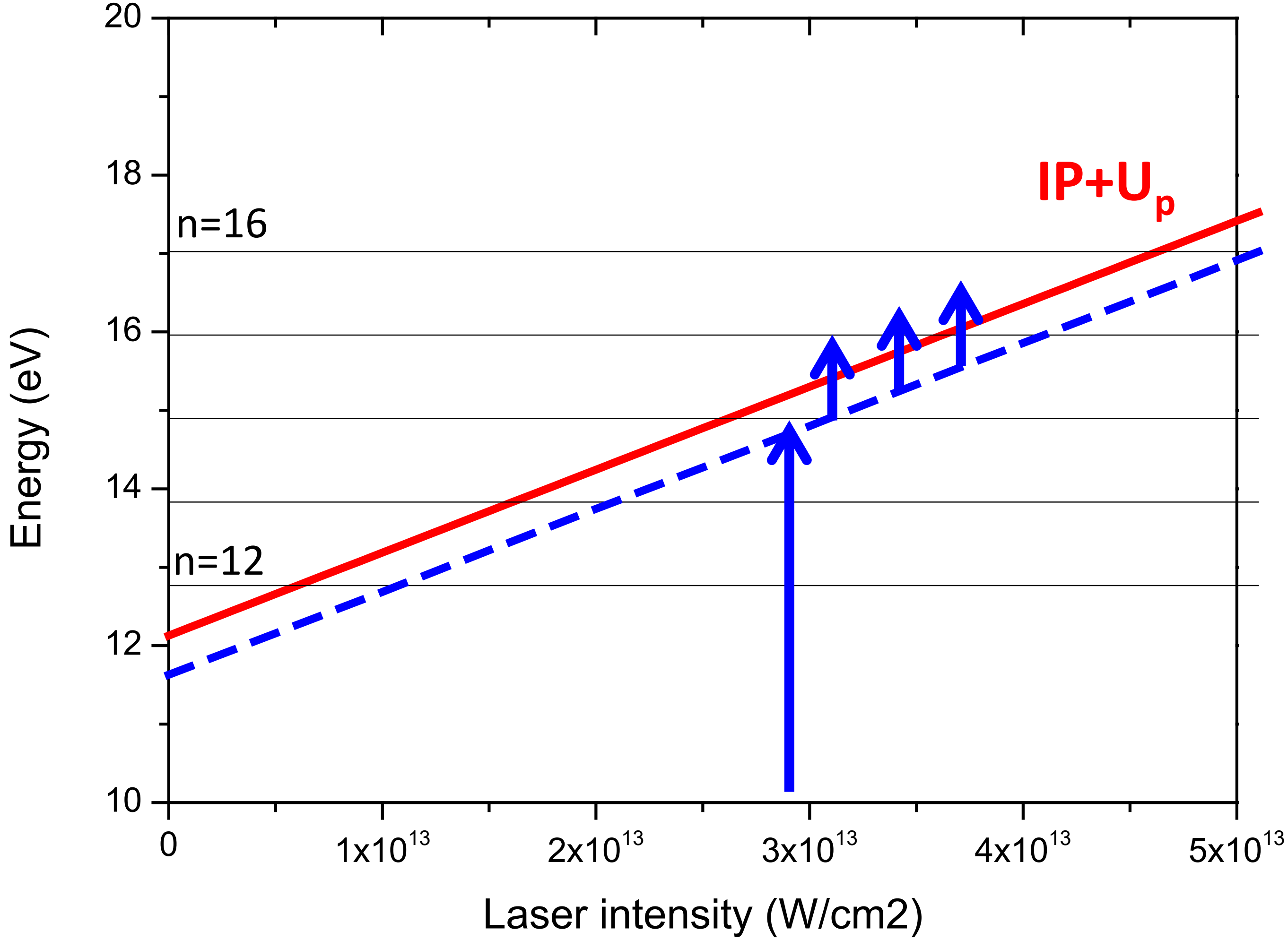
Channel closing

$$U_p(eV) = 9.337\, I_{laser}\left(10^{14}\frac{W}{cm^2}\right)\lambda^2\,(\mu m)$$



Freeman resonances

As the IP ponderomotively shifts, the laser can come into resonance with Rydberg states that also shift ponderomotively



Freeman resonances

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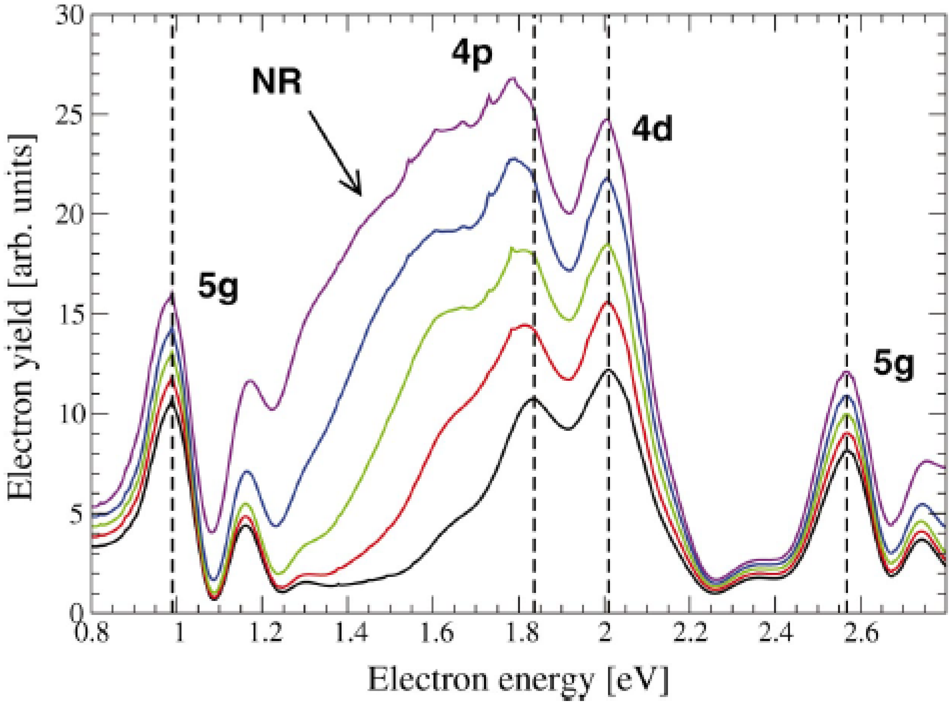
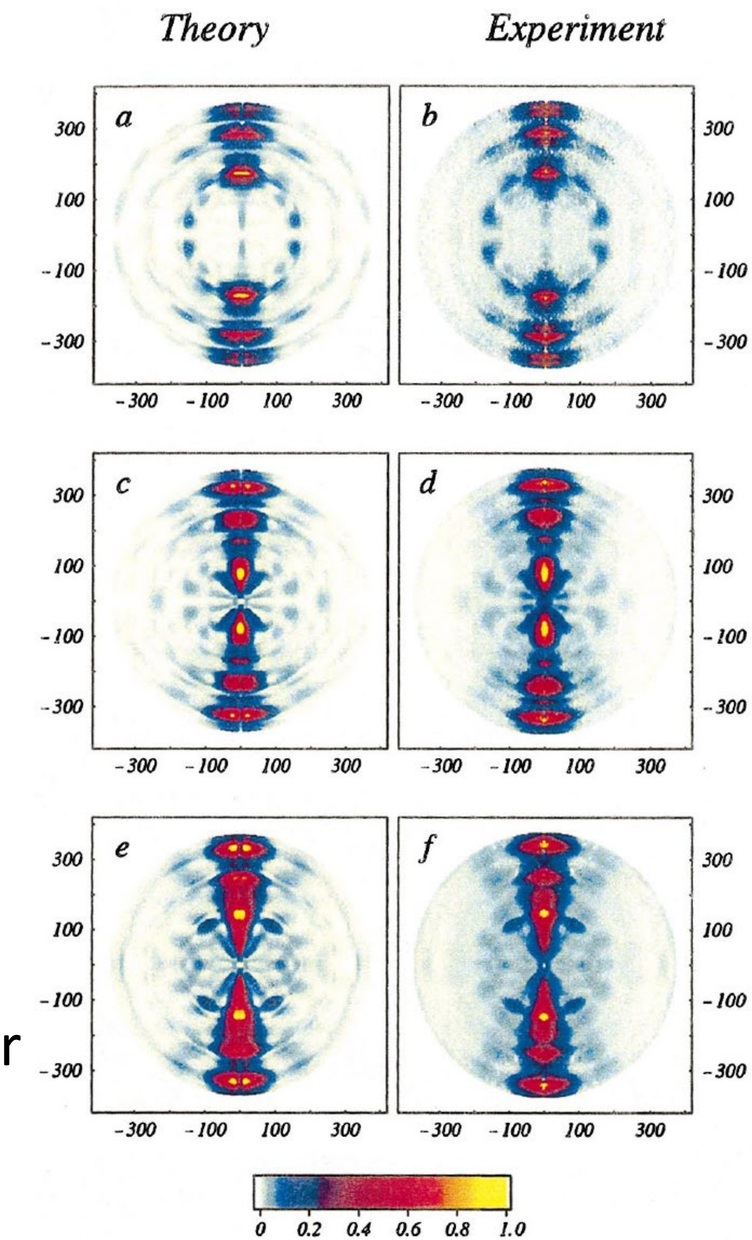
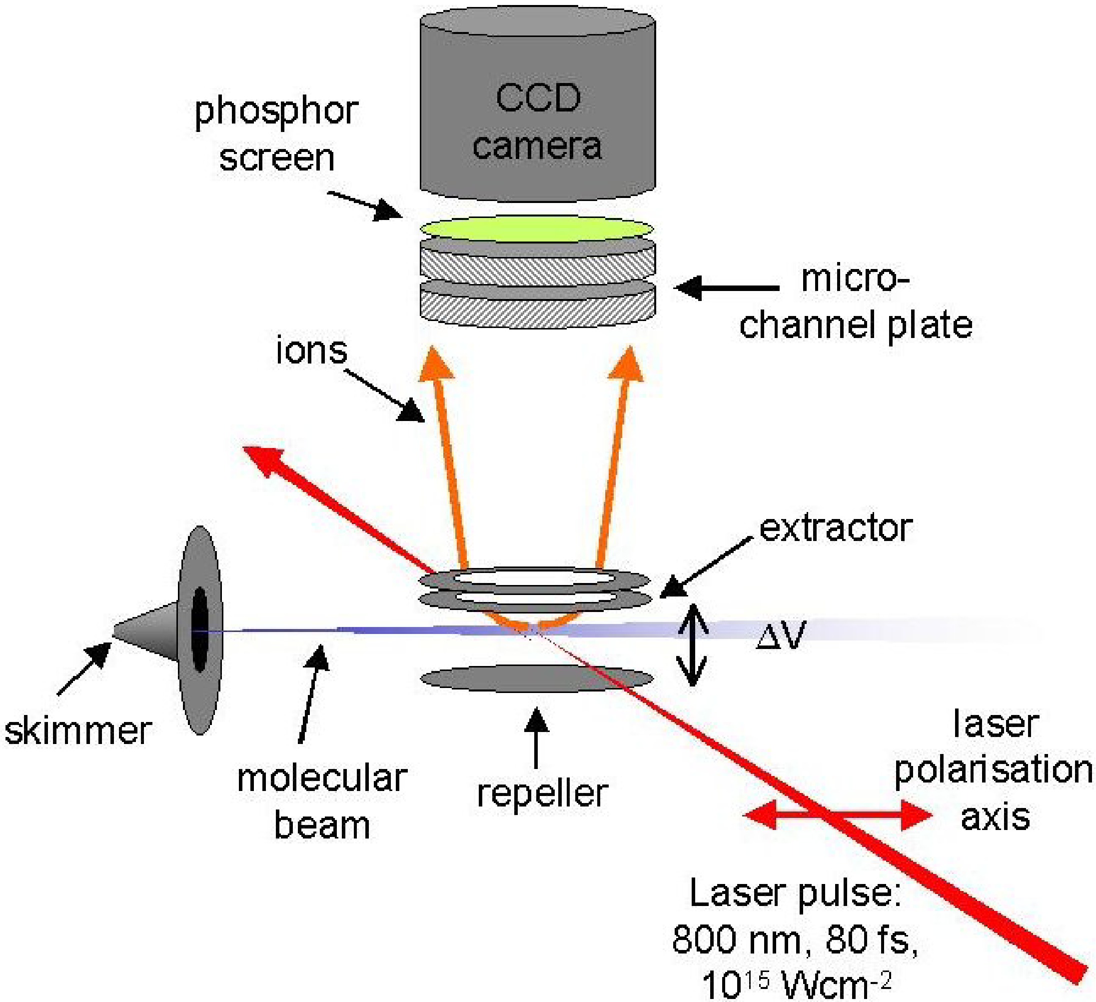


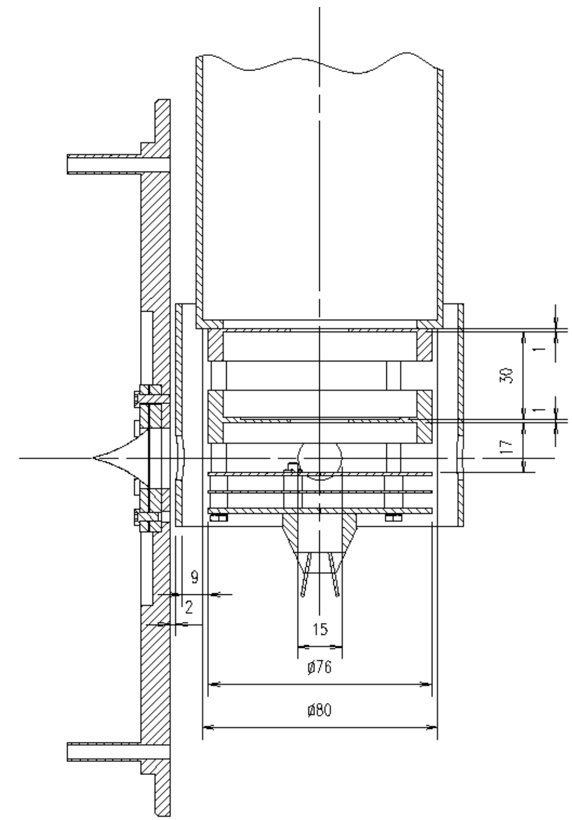
Calculation of Freeman resonances in Ar



R. Wiehle et al., Phys. Rev. A 67, 063405 (2003)

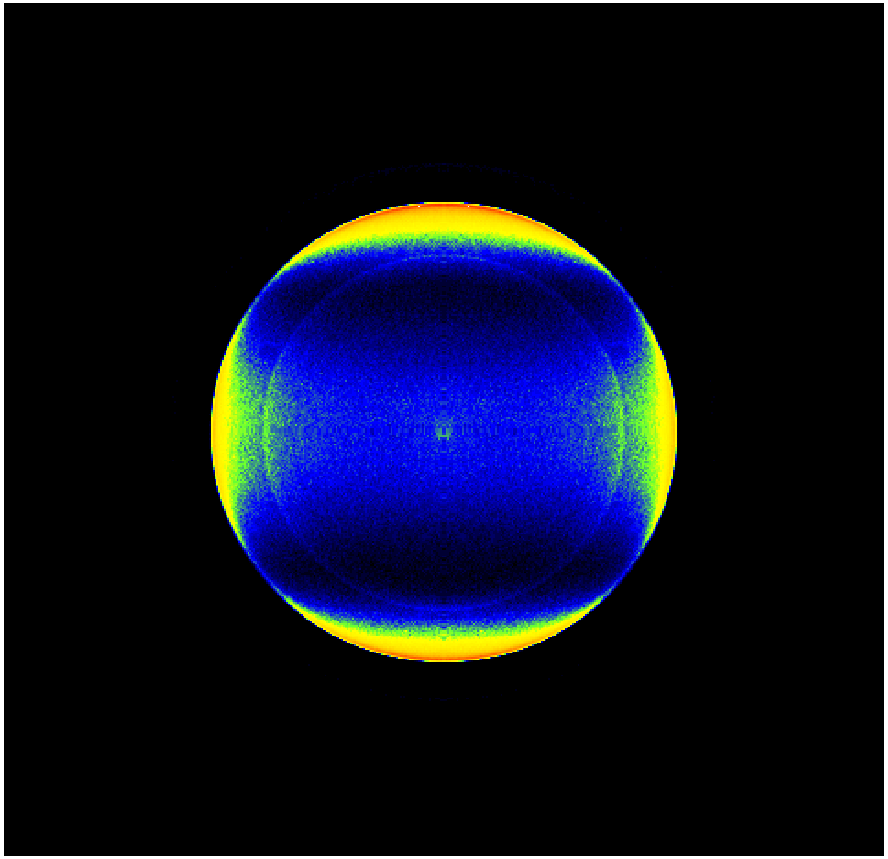
Velocity map ion & photoelectron imaging



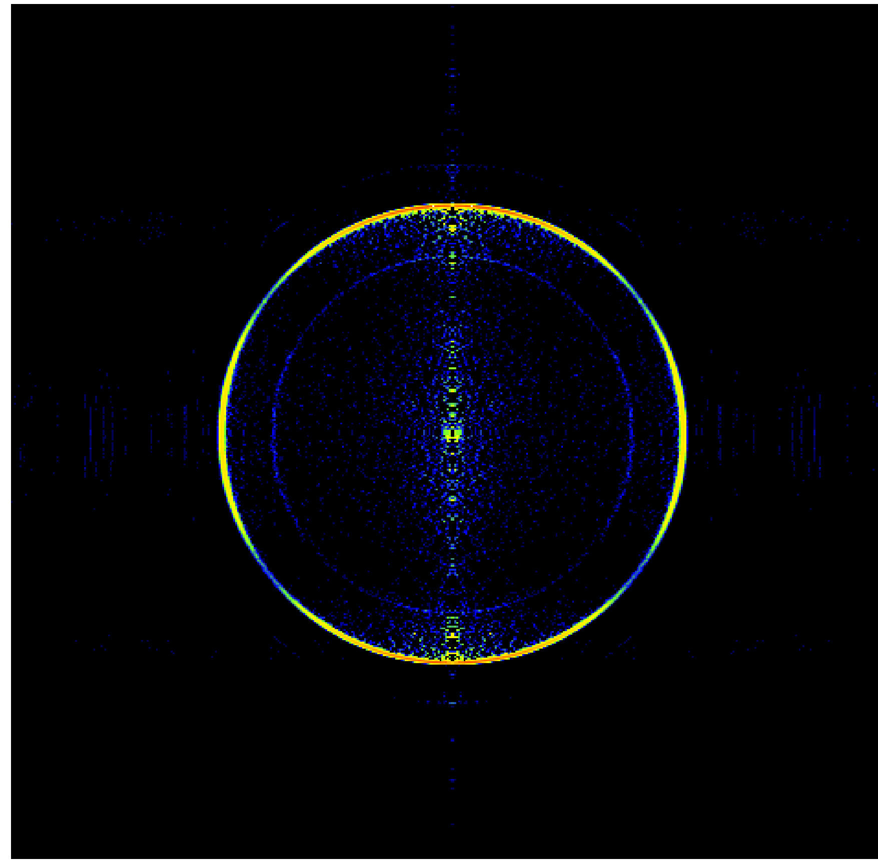


A. T. J. B. Eppink, D. H. Parker, *Review of Scientific Instruments* 68, 3477 (1997).

Extraction of the energy and angular distribution using an iterative procedure



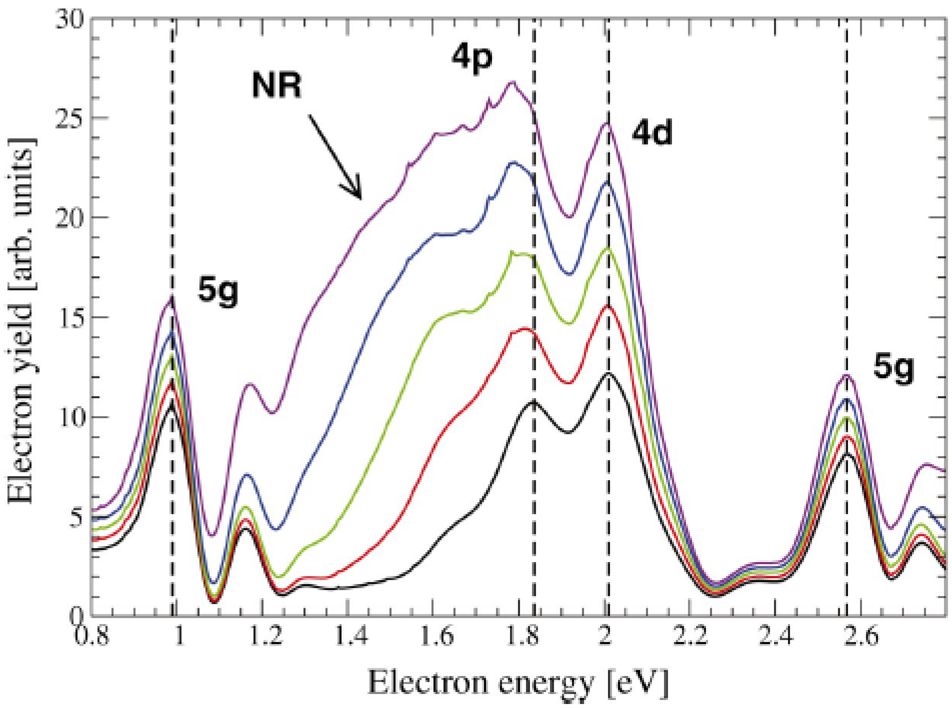
Raw image for 2-photon ionisation of Ar by 532 nm light

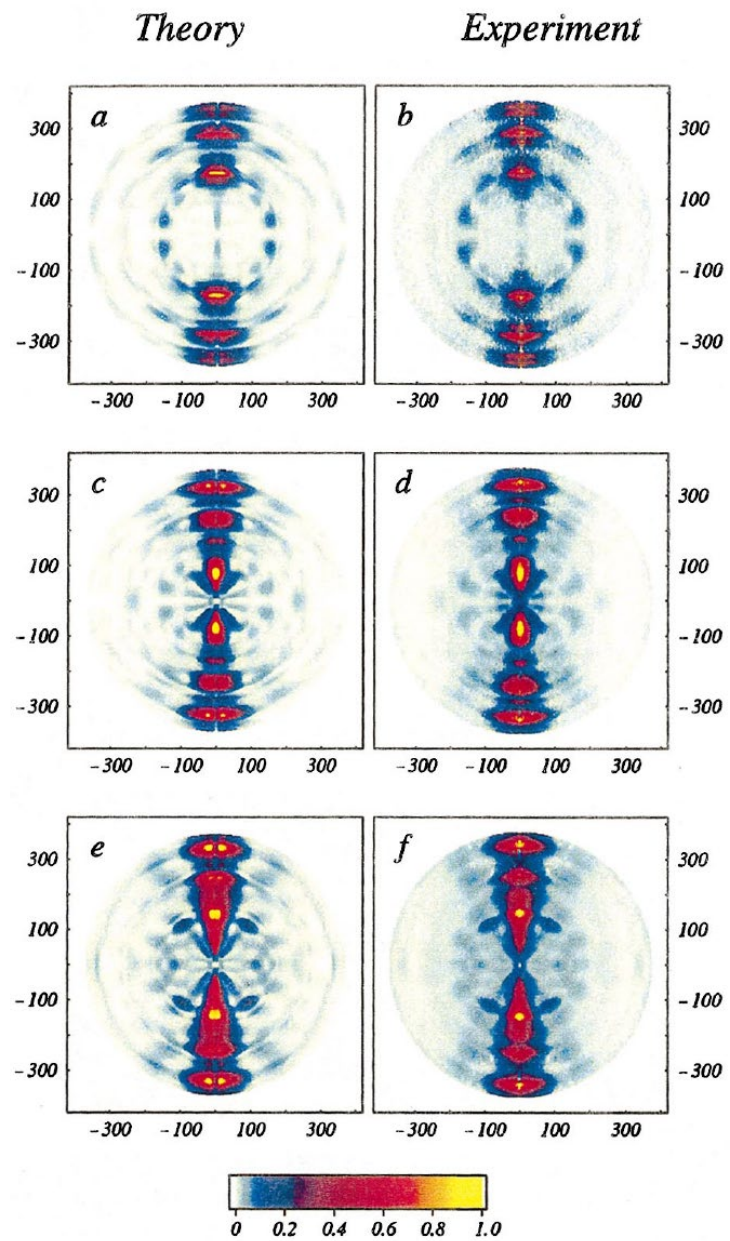


Slice through the 3D velocity distribution, obtained by Abel inversion of the image $\Delta v/v = 1\%$

Freeman resonances

As the IP ponderomotively shifts, the laser can come into resonance with Rydberg states that also shift



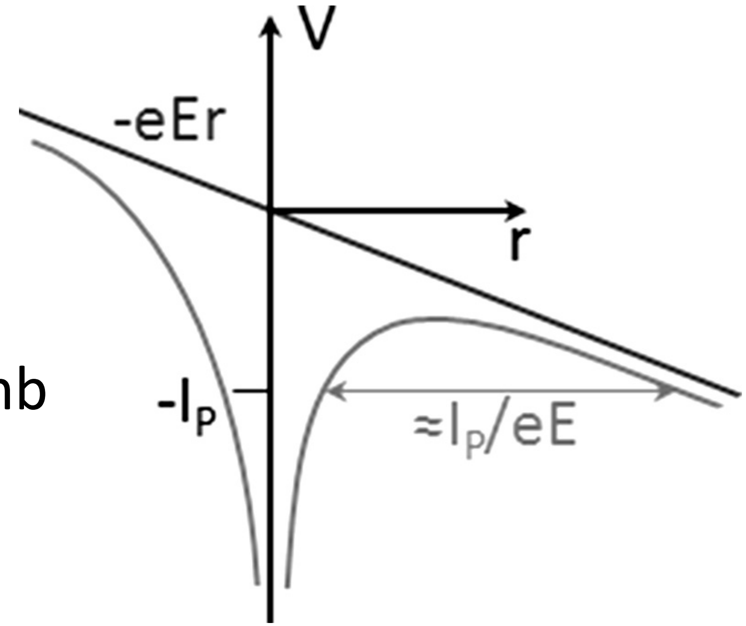


Calculation of Freeman resonances in Ar

R. Wiehle et al., Phys. Rev. A 67, 063405 (2003)

Distortion of the Coulomb potential

$V(z) = -\frac{1}{z} - E_{laser} z$ (a.u.)



For sufficiently strong laser fields the electron can tunnel through the Coulomb + laser field potential, or pass over the saddlepoint in this potential

Saddlepoint: $$\frac{\partial V(z)}{\partial z} = \frac{1}{z^2} - E_{laser} = 0$$

$$z_{Saddle} = \frac{1}{\sqrt{E_{laser}}}; V(z_{saddle}) = -2\sqrt{E_{laser}}$$

Over-the-barrier ionization: $$IP = -2\sqrt{E_{laser}}; E_{laser} = \frac{IP^2}{4}; \; I_{laser,OTB} = \frac{IP^4}{16}$$

Distortion of the Coulomb potential

Over-the-barrier ionization: $I_{laser,OTB} = \frac{IP^4}{16}$

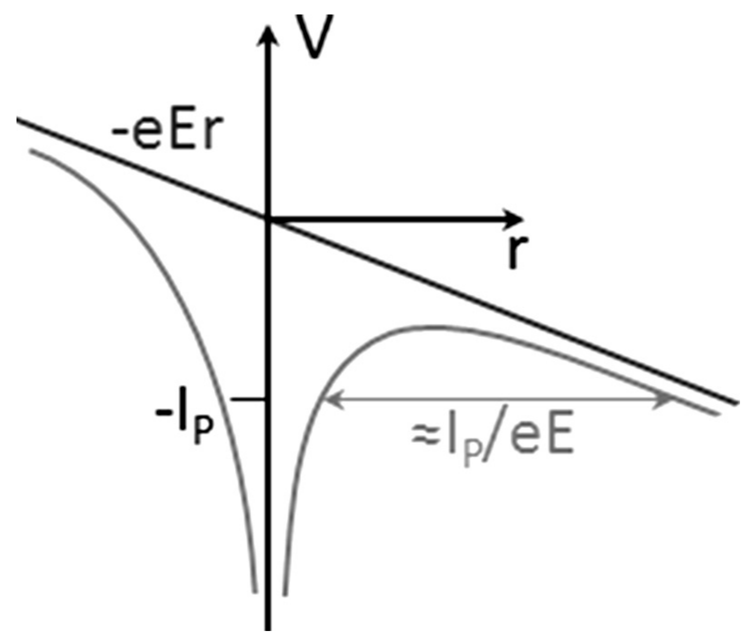
Remember:
1 a.u. = 3.51x10^{16} W/cm^2

Hydrogen atom (IP=0.5 a.u.) : $I_{laser} = 0.0039$ a.u. = 1.4x10^{14} W/cm^2

Below $I_{laser,OTB}$ the electron can escape the atom by tunneling through the Coulomb + laser electric field potential, provided that the potential is sufficiently quasi-static

This condition is expressed by the Keldysh parameter γ

$$\gamma = \sqrt{\frac{IP}{2U_p}} \ll 1$$



Keldysh parameter

The Keldysh parameter can be interpreted as the ratio of the time it takes the electron to tunnel out and the laser period

$$\gamma = \sqrt{\frac{IP}{2U_p}} = \frac{\tau_{tunneling}}{\tau_{laser}}$$

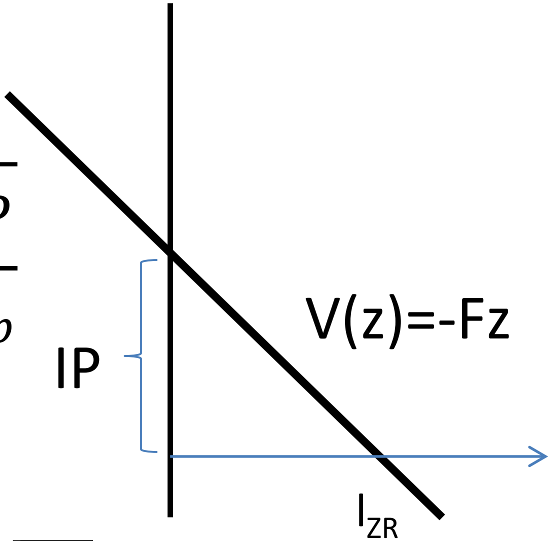
Derivation for zero-range potential:

$$l_{ZR} = \frac{IP}{F}$$

$$< v >= \sqrt{2IP}$$

$$\Rightarrow \frac{\tau_{tunneling}}{\tau_{laser}} \sim \frac{\omega\sqrt{IP}}{F} \sim \sqrt{\frac{IP}{U_p}}$$

Keldysh



$$v(t) = v_{max} - Ft \rightarrow 0$$

$$v_{max} = \sqrt{2IP}$$

$$\Rightarrow \frac{\tau_{tunneling}}{\tau_{laser}} \sim \frac{\omega\sqrt{IP}}{F} \sim \sqrt{\frac{IP}{U_p}}$$

Ivanov

Tunneling formulas

Provided suitable approximations are made, the rate of tunnel ionization can be described by simple formulas

Strong field approximation:
Assume that after the ionization process the interaction of the electron with the core is negligible, and that the electron only interacts with the laser electric field

Adiabatic approximation:
Assume that in the presence of the laser field the atom remains in the lowest available state, and that no population is transferred to excited states

Single active electron approximation:
Assume only the most weakly bound electron is ionized

Tunneling formulas

In the adiabatic approximation, the ionization rate at time t, when the electric field equals E(t), is given by $\Gamma_{DC}(E(t))$

Ground state hydrogenic atom:

$$\Gamma_{DC}(E) = \frac{4Z^5}{E} exp\left(-\frac{2Z^3}{3E}\right)$$

Non-hydrogenic atoms in arbitrary states:

$$\kappa = (2IP)^{1/2}$$

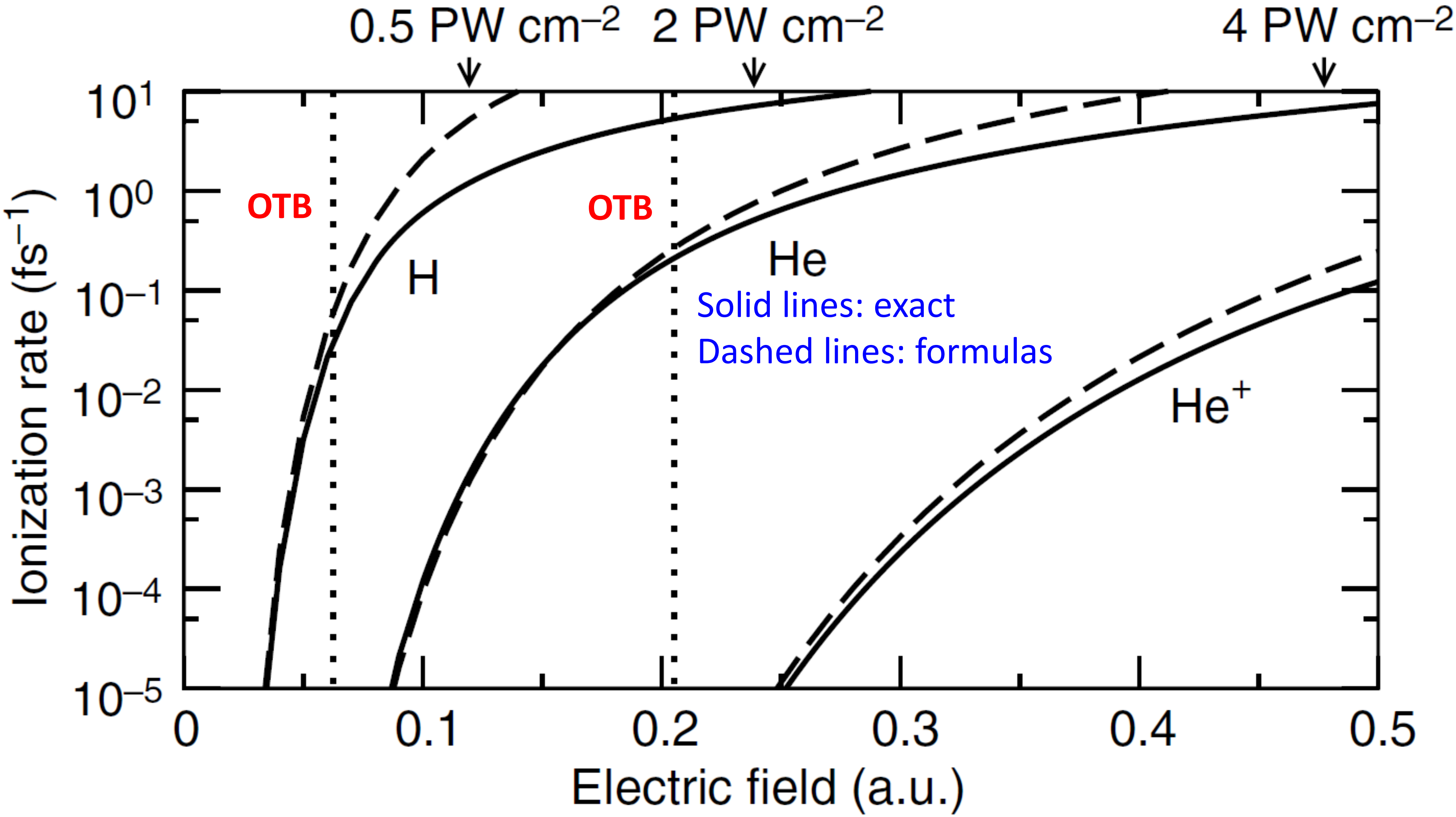
$$\Gamma_{DC}(E) = C_{as}^2 A(l,m) \frac{\kappa^2}{2} \left(\frac{2\kappa^3}{E}\right)^{\frac{2Z_c}{\kappa}-|m|-1} exp\left(-\frac{2\kappa^3}{3E}\right)$$

$$A(l,m) = \frac{2l+1}{2^{|m|}|m|!} \frac{(l+|m|)!}{(l-|m|)!} \qquad C_{as} = \left[\frac{2^{2n^*}}{n^*\Gamma(n^*+l+1)\square(n^*-l)}\right]^{1/2}$$

C.J. Joachain, N.J. Kylstra and R.M. Potvliege, Atoms in Intense Laser Fields, (Cambridge University Press, 2012)

NB Simplified to ADK by Ammosov-Delone-Krainov

Tunneling formulas



C.J. Joachain, N.J. Kylstra and R.M. Potvliege, Atoms in Intense Laser Fields, (Cambridge University Press, 2012)

After ionization: Propagation assuming the strong-field approximation (SFA)

Assume that the electron does not feel the ion anymore as soon as it has tunneled out
Assume, moreover, that the Coulomb-free motion starts with v=0 at r=0, and that the laser amplitude is constant

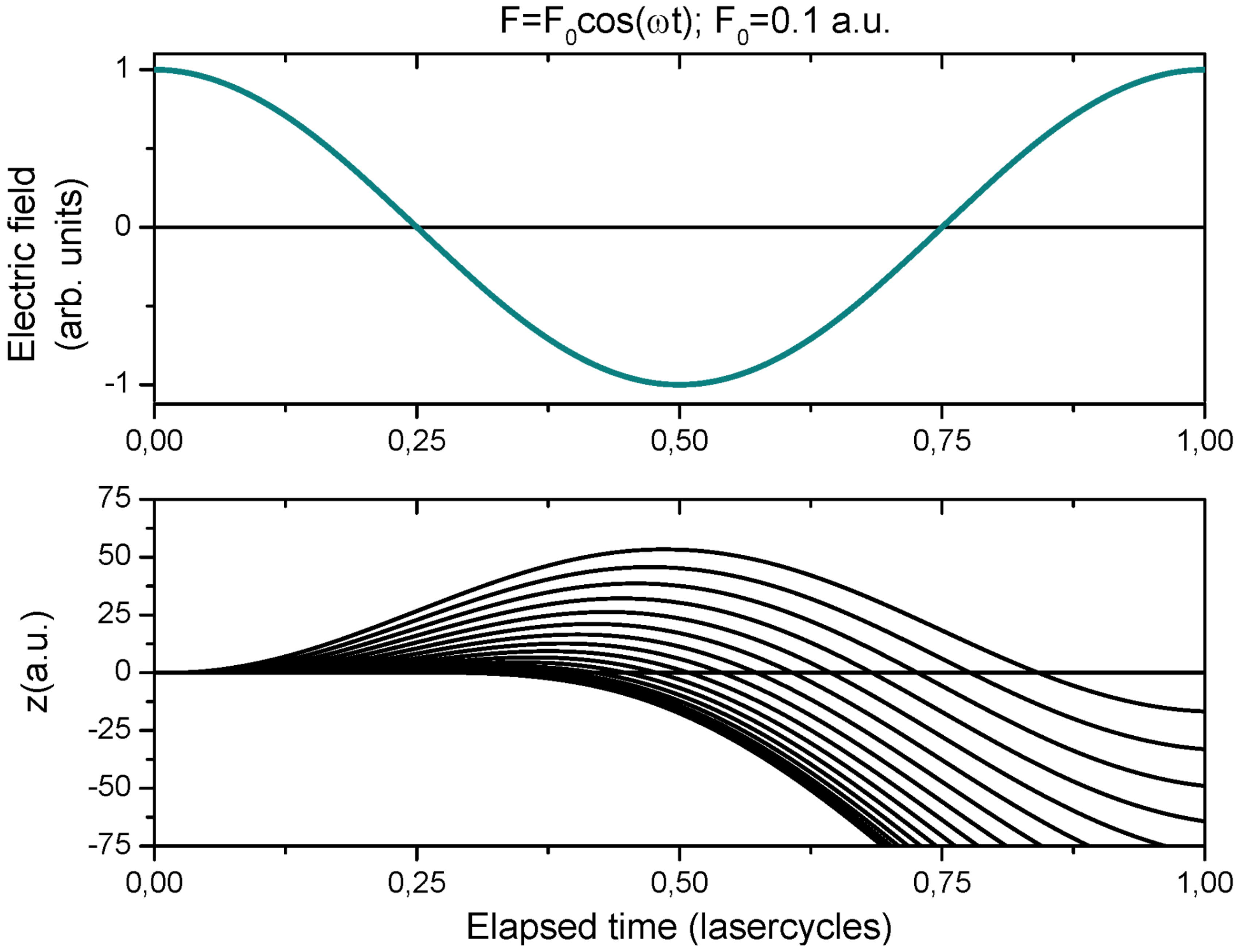
$a(t) = E_0 cos\omega t$ (a.u.)

$v(t) = v_0 sin\omega t + \ v_{0z}$ $\qquad v_0 = E_0/\omega$ (a.u.)

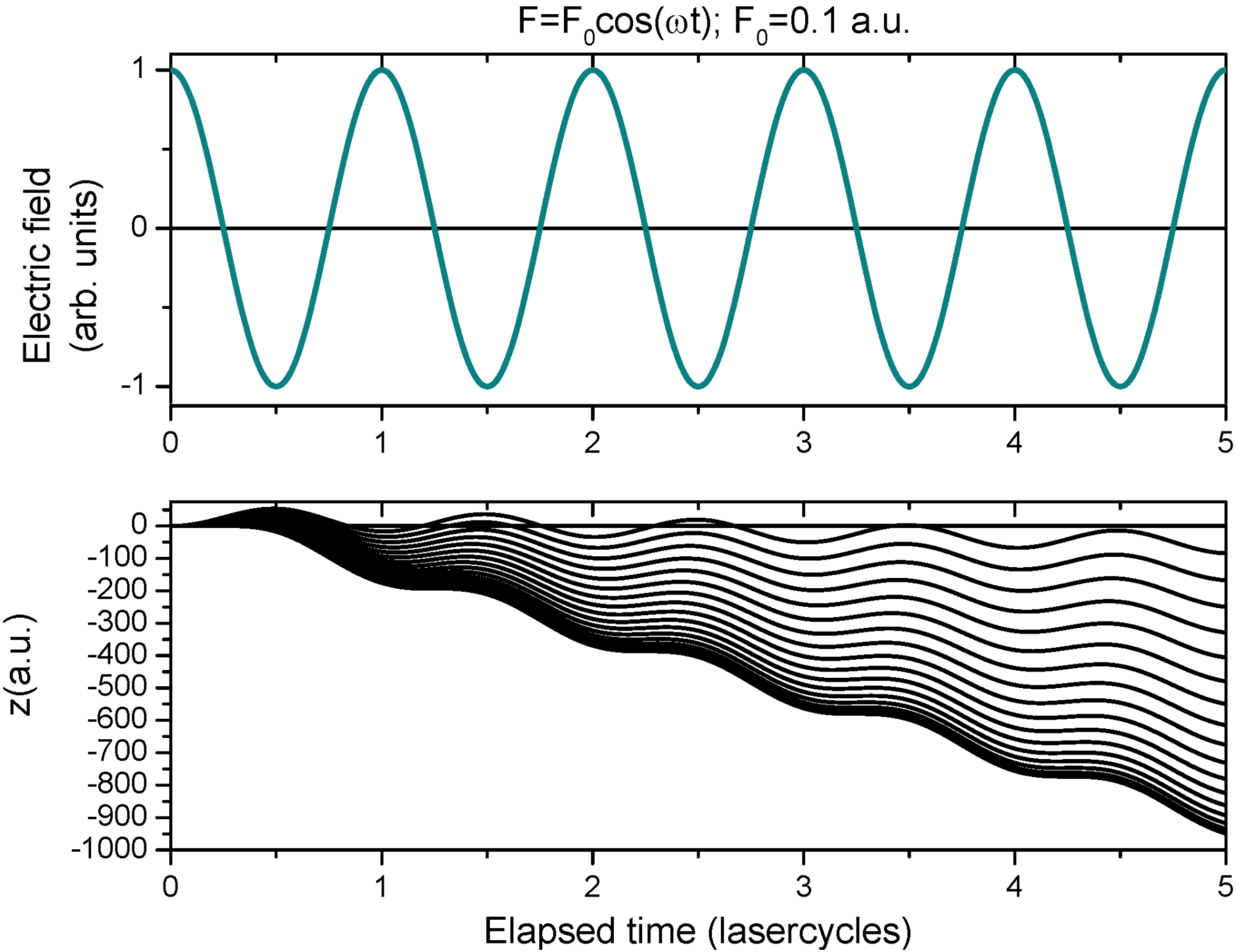
$z(t) = z_0(-cos\omega t) + \ v_{0z}t + \ z_{0z}$ $\qquad z_0 = E_0/\omega^2$(a.u.)

N.B. E$(t) = -dA/dt, i.e. v_0 = A_0$

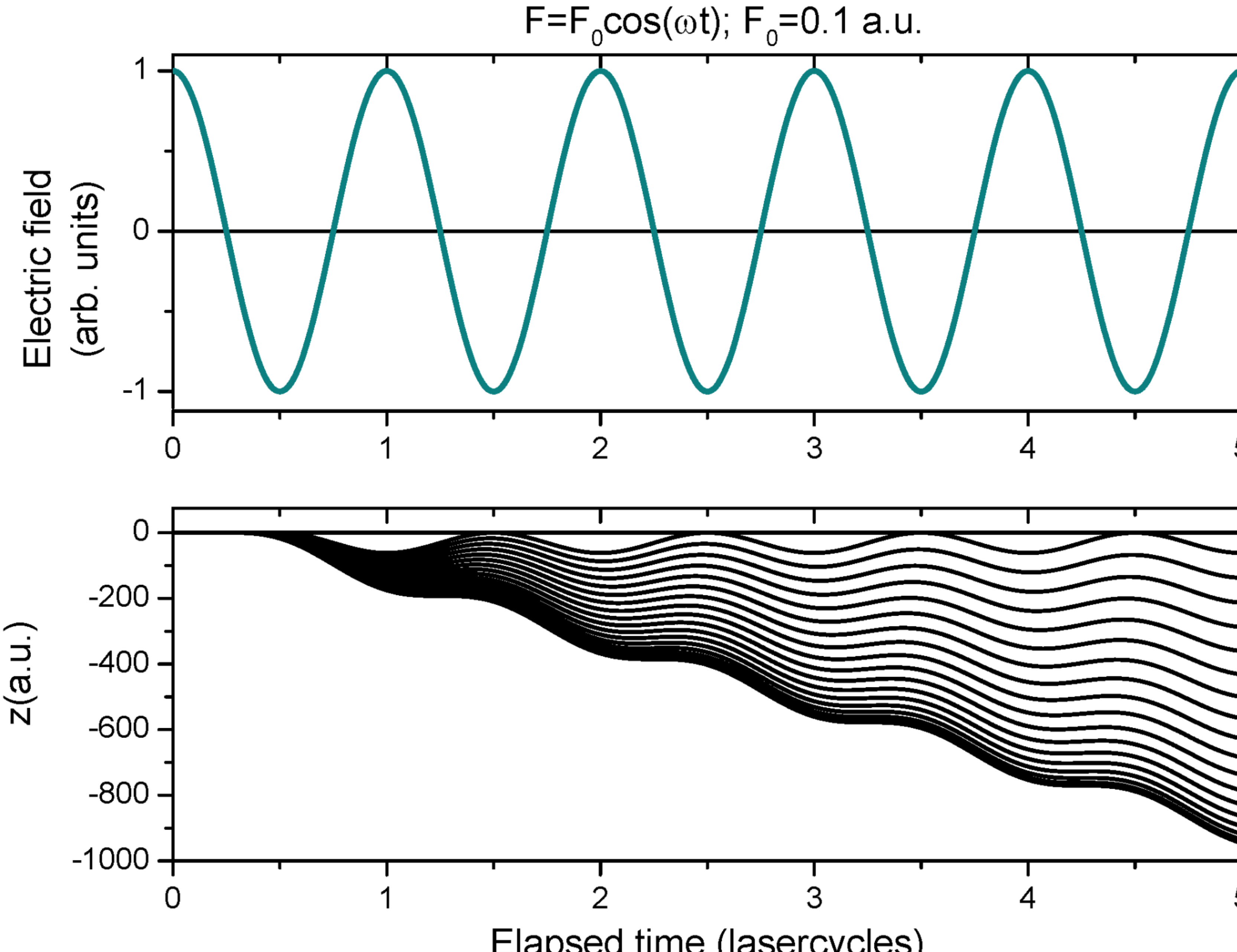
P. Corkum. Phys. Rev. Lett. 71, 1994 (1993)



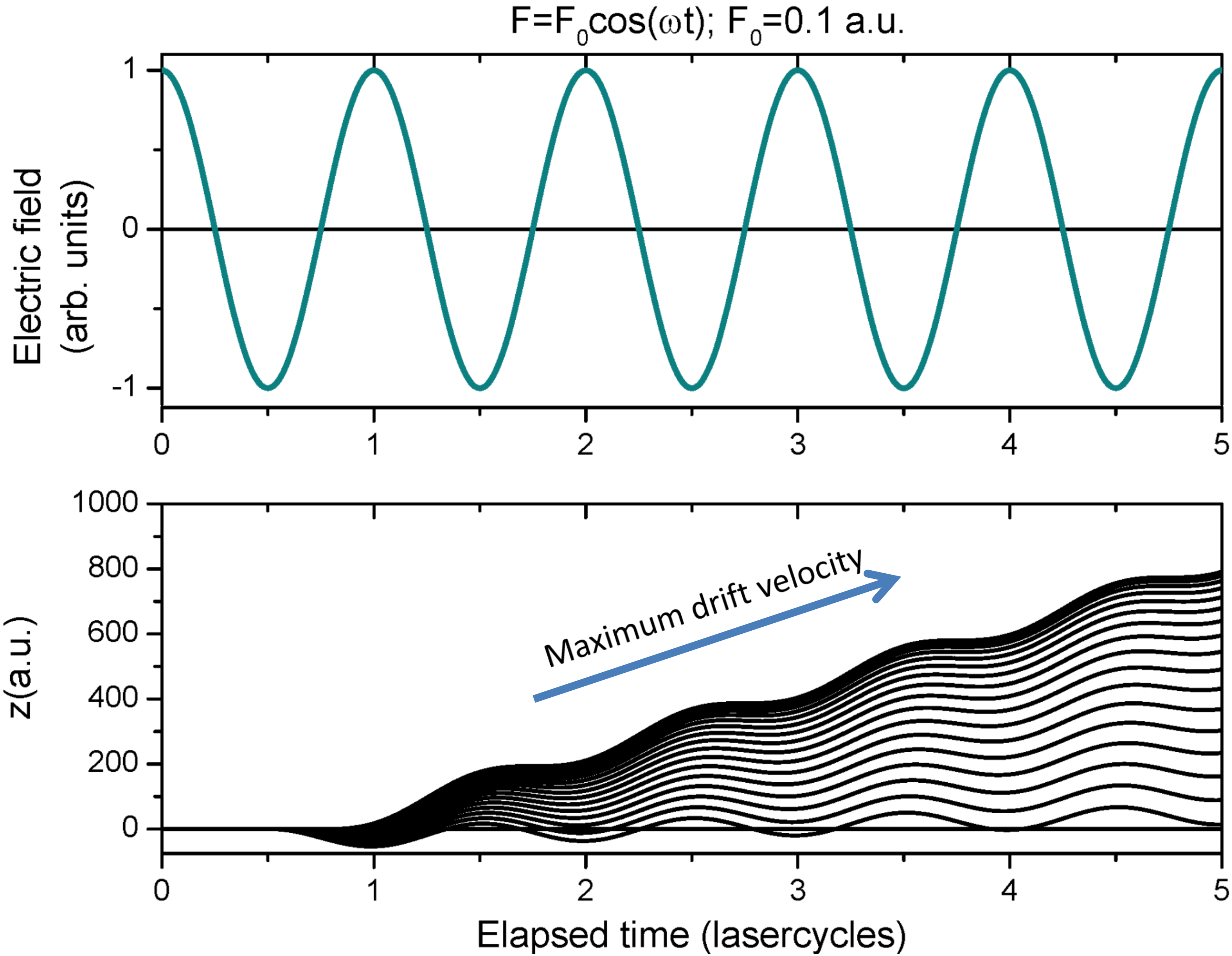
F=F$_0$cos(ωt); F$_0$=0.1 a.u.
Electric field (arb. units)
1
0
-1
0,00
0,25
0,50
0,75
1,00
z(a.u.)
75
50
25
0
-25
-50
-75
Elapsed time (lasercycles)



F=F0cos(ωt); F0=0.1 a.u.
Electric field (arb. units)
1
0
-1
0 1 2 3 4 5
z(a.u.)
0
-100
-200
-300
-400
-500
-600
-700
-800
-900
-1000
0 1 2 3 4 5
Elapsed time (lasercycles)



F=F0cos(ωt); F0=0.1 a.u.
Electric field (arb. units)
1
0
-1
0
1
2
3
4
5
0
-200
-400
-600
-800
-1000
z(a.u.)
0
1
2
3
4
5
Elapsed time (lasercycles)



F=F0cos(ωt); F0=0.1 a.u.
Electric field (arb. units)
1
0
-1
0
1
2
3
4
5
1000
800
600
400
200
0
z(a.u.)
Maximum drift velocity
Elapsed time (lasercycles)

Maximum drift velocity

$$v_\infty(t_{ionization}) = \int_{t_{ionization}}^{\infty} a(t)dt =$$

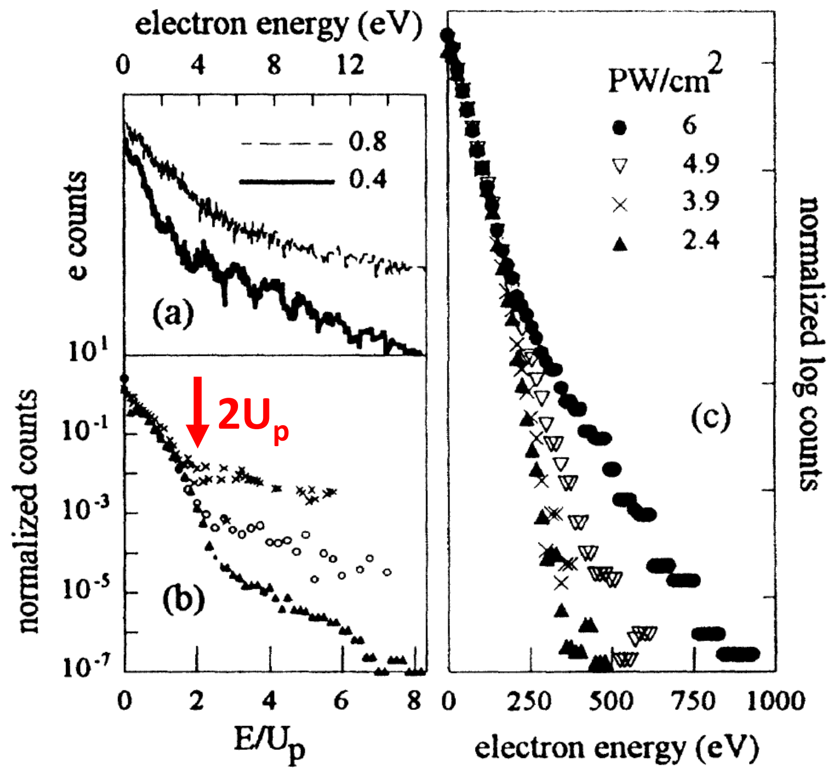
$$\int_{t_{ionization}}^{\infty} dt\,[-E_0 cos\omega t] \sim \frac{E_0}{\omega} sin\omega t\Big|_{t_{ionization}}^{\infty} -$$

$$= \frac{-E_0}{\omega} sin\omega t_{ionization} = -A(t_{ionization})$$

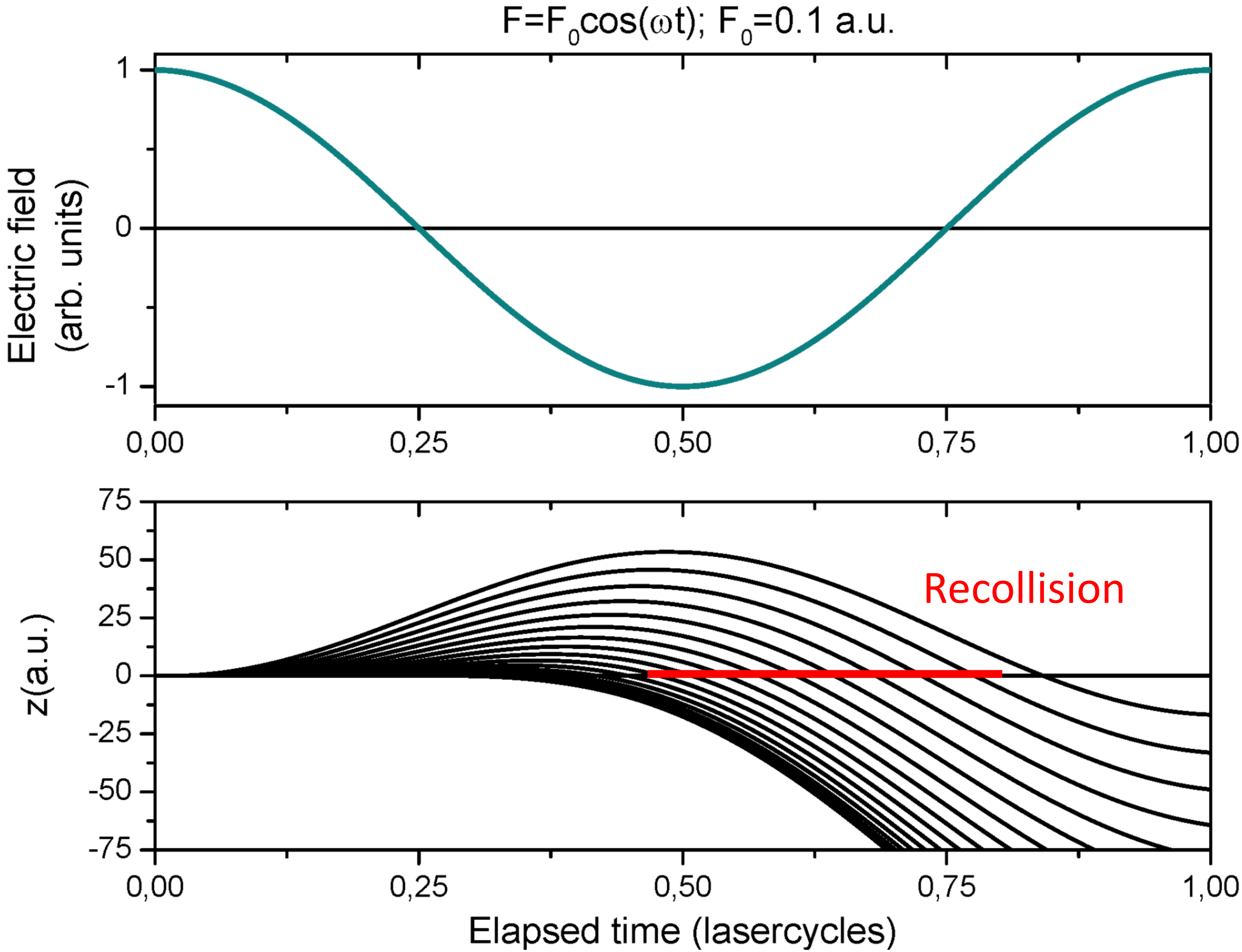
$$E_{k,max} = \frac{1}{2} v_{max}^2 = \frac{E_0^2}{2\omega^2} = \frac{A_0^2}{2}$$

$2U_p$

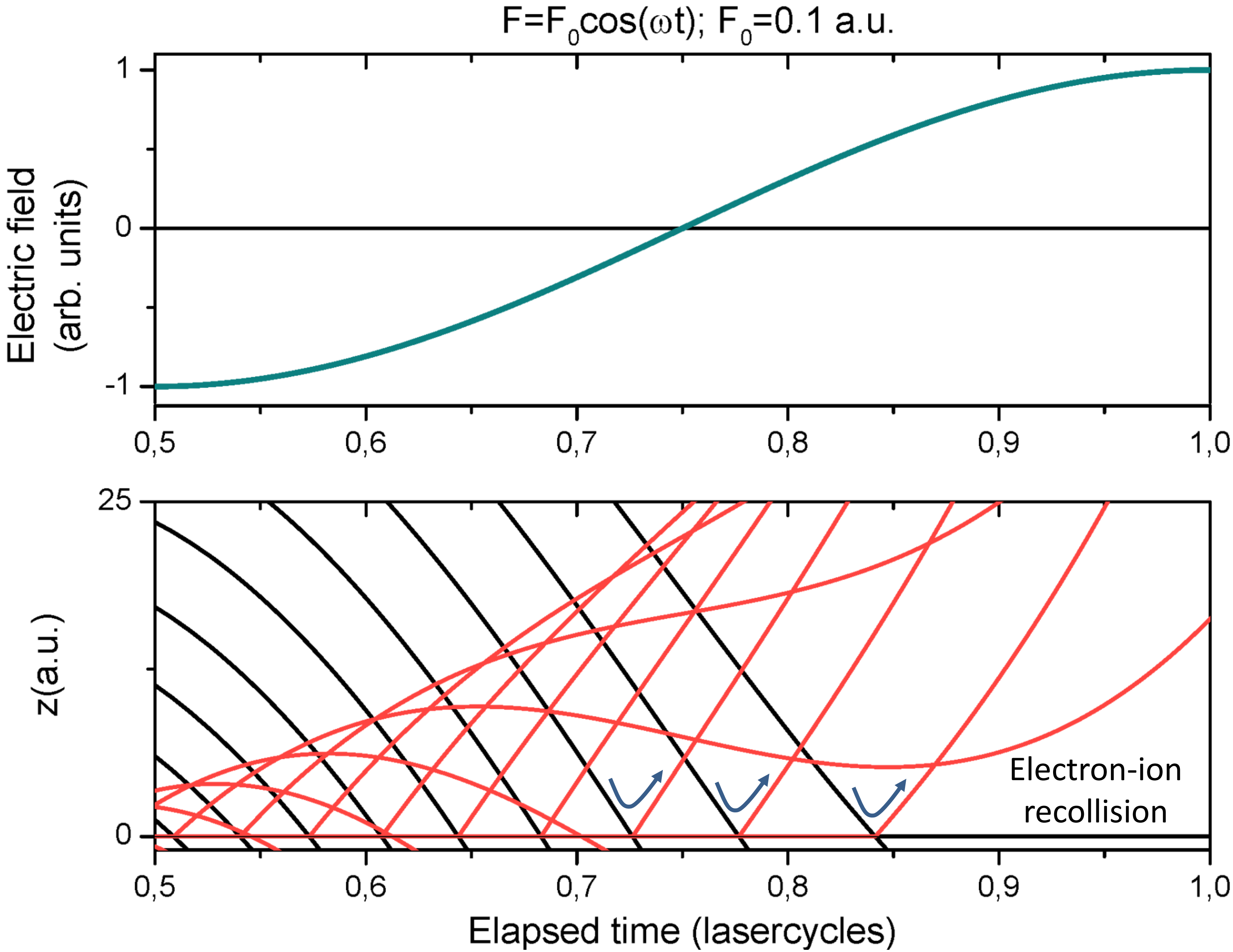
The photoelectron spectrum also contains energies > $2U_p$



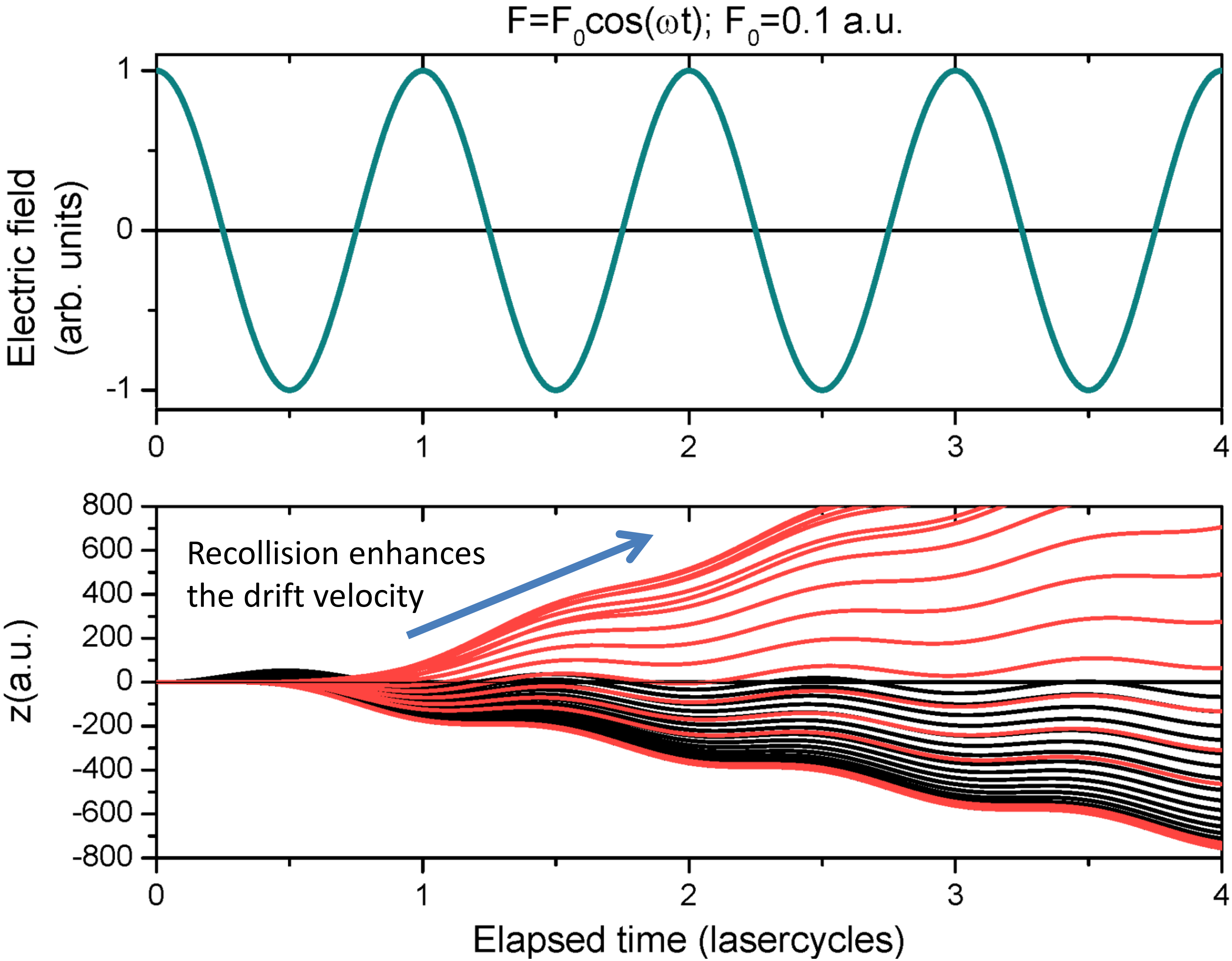
B. Walker et al., Phys. Rev. Lett. 73, 1227 (1994)



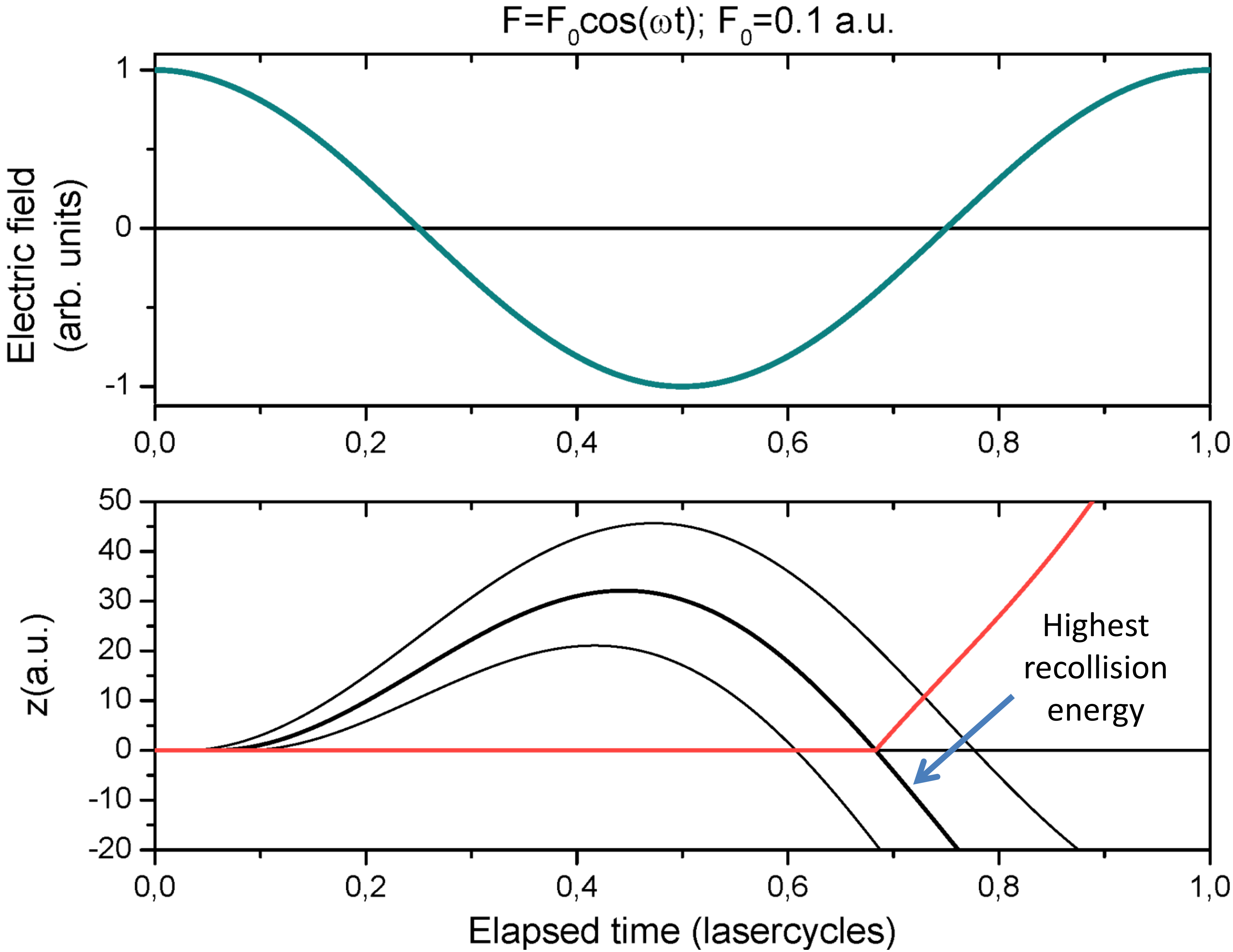
F=F0cos(ωt); F0=0.1 a.u.
Electric field (arb. units)
1
0
-1
0,00
0,25
0,50
0,75
1,00
z(a.u.)
75
50
25
0
-25
-50
-75
Recollision
Elapsed time (lasercycles)



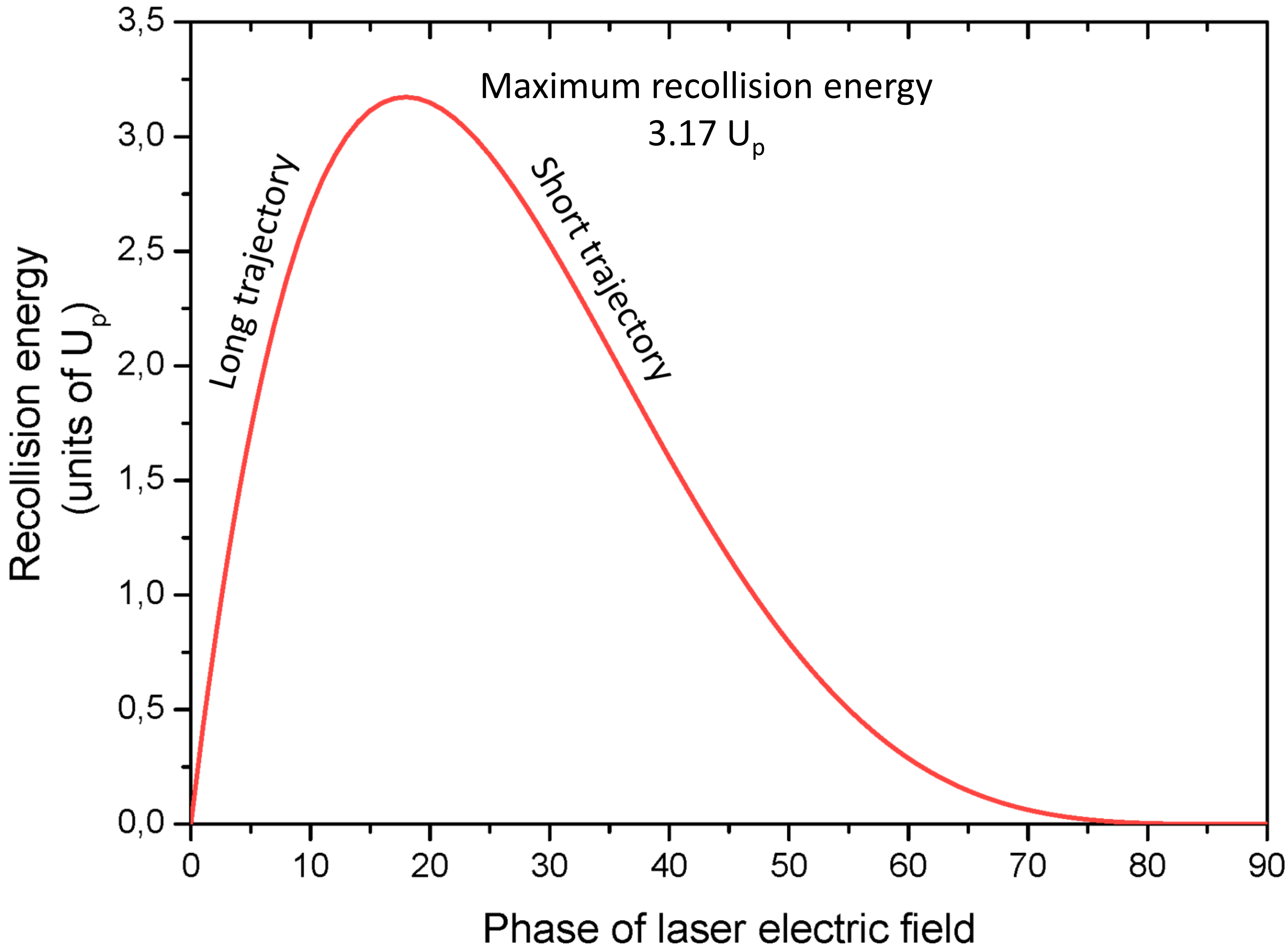
F=F0cos(ωt); F0=0.1 a.u.
Electric field (arb. units)
1
0
-1
0,5
0,6
0,7
0,8
0,9
1,0
25
z(a.u.)
0
Electron-ion recollision
Elapsed time (lasercycles)



F=F$_0$cos(ωt); F$_0$=0.1 a.u.
Electric field (arb. units)
1
0
-1
0
1
2
3
4
800
600
400
200
0
-200
-400
-600
-800
z(a.u.)
Recollision enhances the drift velocity
Elapsed time (lasercycles)



F=F$_0$cos(ωt); F$_0$=0.1 a.u.
Electric field (arb. units)
1
0
-1
0,0
0,2
0,4
0,6
0,8
1,0
z(a.u.)
50
40
30
20
10
0
-10
-20
Highest recollision energy
Elapsed time (lasercycles)



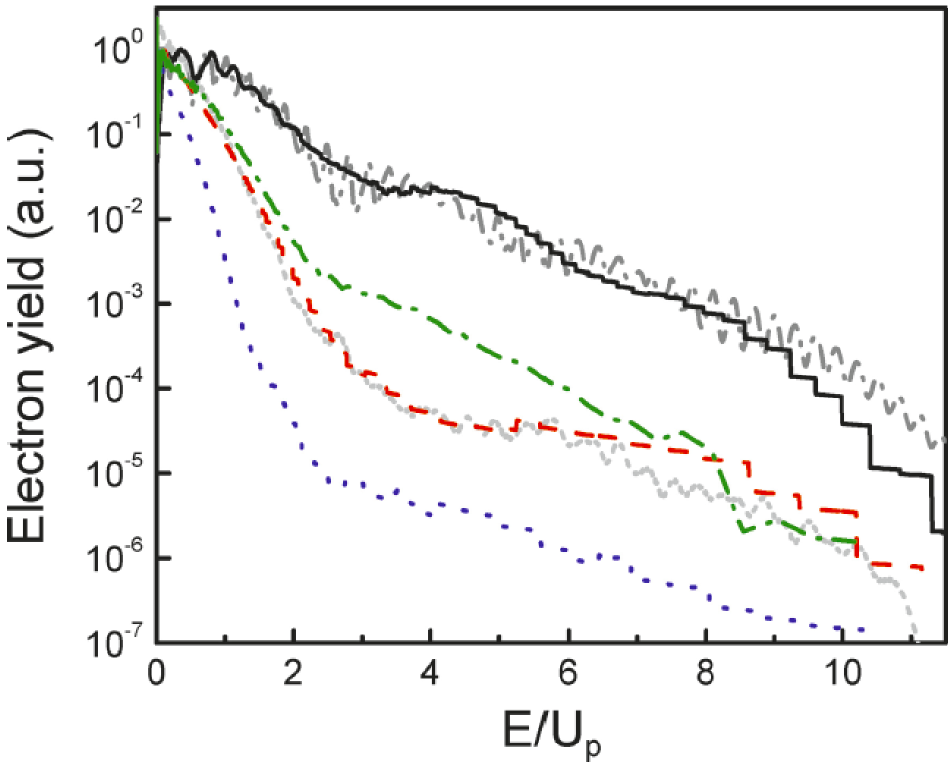
Maximum recollision energy
3.17 U_p
Long trajectory
Short trajectory
Recollision energy
(units of U_p)
3,5
3,0
2,5
2,0
1,5
1,0
0,5
0,0
0
10
20
30
40
50
60
70
80
90
Phase of laser electric field

Recollision trajectories

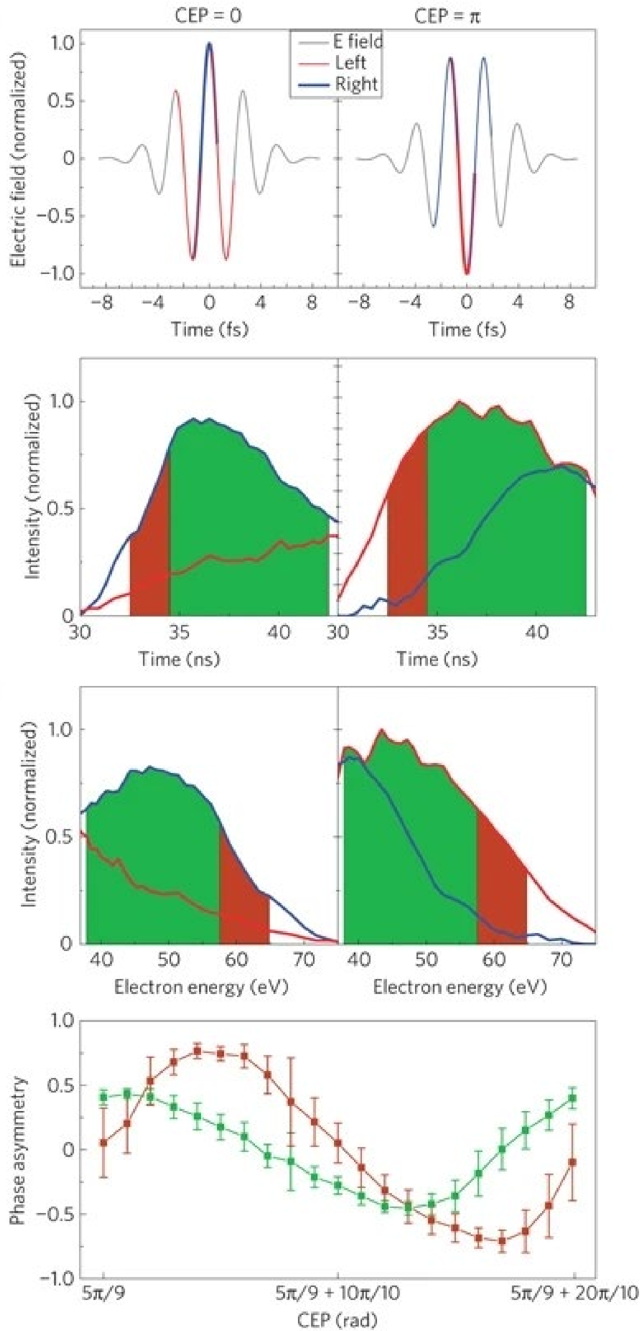
The highest possible return energy is 3.17 U_p = 0.7925 A_0^2, corresponding to a velocity of 1.259 A_0. This recollision occurs near a zero crossing of the field.

When the recollision flips the sign of the velocity, the field increases the velocity to appr. -2.259 $A_{0,}$ corresponding to an energy of 10.2 U_p

ATI of Argon at 0.8 μm (black line), 1.3μm (green line), 2 μm (red line), and 3.6μm (blue line) at an intensity of 0.08 PW/cm2.

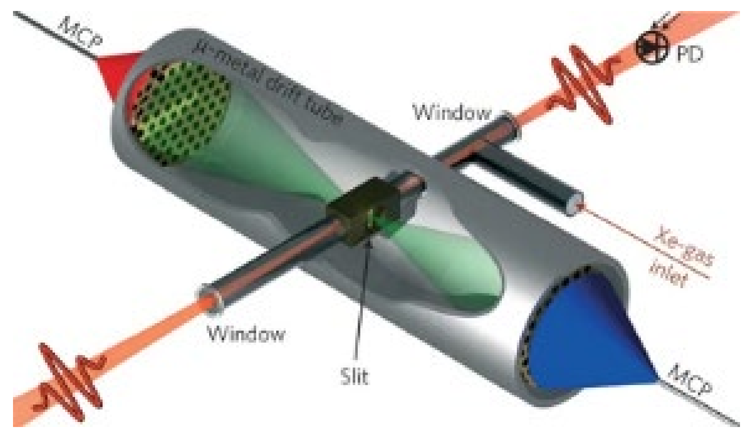


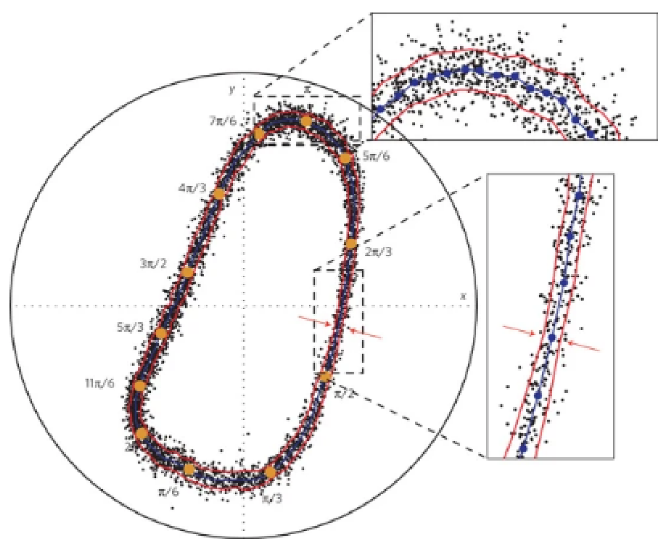
Colosimo, P. et al., Nat. Phys., 4, 386 (2008)



Stereo-ATI

The sensitivity of the recollision electrons to the carrier envelope phase (CEP) of the laser can be exploited to develop a single-shot diagnostic of this parameter





The „phase potato“

Wittmann et al., Nat. Phys. 5, 357 (2009)

Canonical momentum

$$E(t) = E_0 \cos(\omega t) = -\frac{dA(t)}{dt}$$

$$A(t) = -A_0 \sin(\omega t) + constant$$

Vector potential

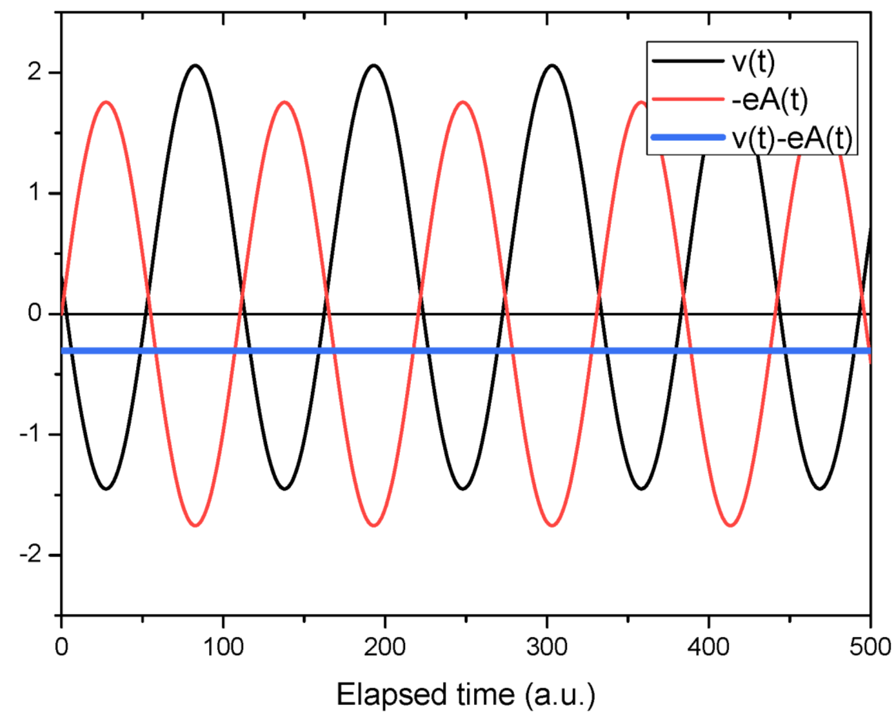
$$A_0 = \frac{E_0}{\omega}$$

$$v(t) = v_0 + \int_{t_0}^{t} a(t)dt = v_0 + \int_{t_0}^{t} \frac{-eE(t)}{m} dt = v_0 + \frac{e}{m}(A(t) - A(t_0))$$

$$v(t) - \frac{e}{m}A(t) = v_0 - \frac{e}{m}A(t_0)$$

In a strong laser field the conserved quantity is the canonical momentum:

p = m**v**(t) – eA(t)



Canonical momentum

$\mathbf{p} = m\mathbf{v}(t) - eA(t)$

At the end of the laser pulse, A(t) is equal to zero

$m\mathbf{v}_{\infty}(t) - e\cancel{A}(t) = m\mathbf{v}_{\mathbf{initial}}(t) - eA_{initial}(t)$

This is the basis for attosecond streaking methods that we now encounter

- If we know $v_{initial}(t)$ and $A_{initial}(t)$ then we can find $t_{initial}$
- If we know $v_{initial}(t)$ and $t_{initial}$ then we can find A(t), and thus E(t)
- (to be shown later) characterize attosecond pulses by streaking

Useful materials for further reading (strong field ionization):

C.J. Joachain, N.J. Kylstra and R.M. Potvliege, *Atoms in Intense Laser Fields,* (Cambridge University Press, 2012)

M. Ivanov et al., *Anatomy of strong field ionization,* J. Mod. Optics 52, 165 (2005)

L. DiMauro and P. Agostini, Adv. At. Mol. And Opt. Physics 35, 79 (1995)

+ several chapters (DiMauro, Ivanov, Smirnova, L´Huillier) in upcoming book „Attosecond and XUV Physics“ (ed. by M.J.J. Vrakking and Th. Schultz, Wiley, december 2013)