Ultra high-resolution imaging of atoms and molecules by intense laser pulses

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Collaboration with:

Theory

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Experiment

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C. Lew Cocke, D. Ray (KSU, USA)
F. Sakai, S. Minemoto (Tokyo, Japan)

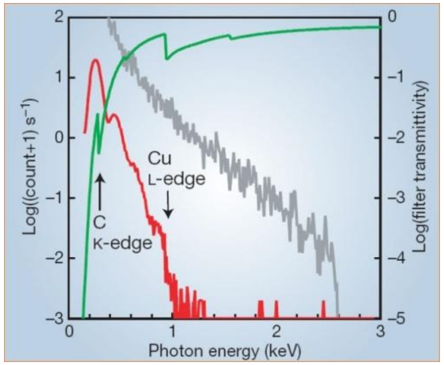
Outline

- Introduction
  - Ultrafast imaging of atoms & molecules
  - HHG, ATI with intense IR pulses
- Adiabatic theory
  - Basic idea
  - Examples
- Tunnel ionization
  - Atoms & Molecules in static fields
- Summary

HHG as a coherent X-ray source

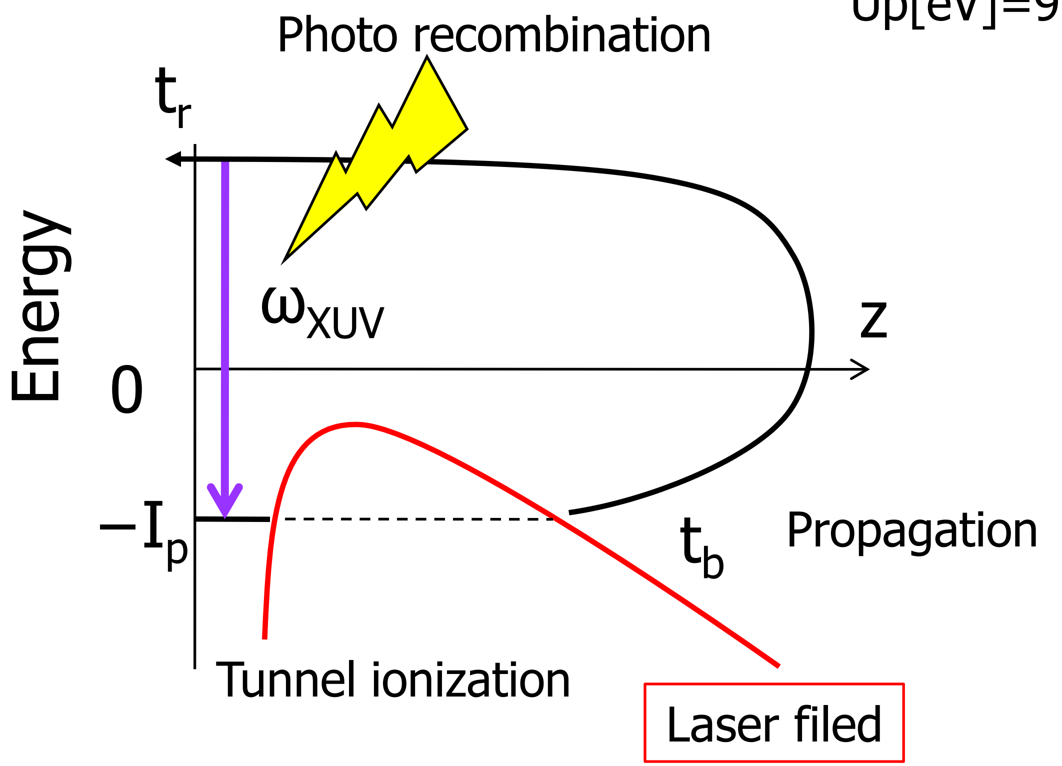
E cutoff= 3 Up + Ip
Up[eV]=93.8 x I [PW/cm] x $\lambda[\mu m]^2$

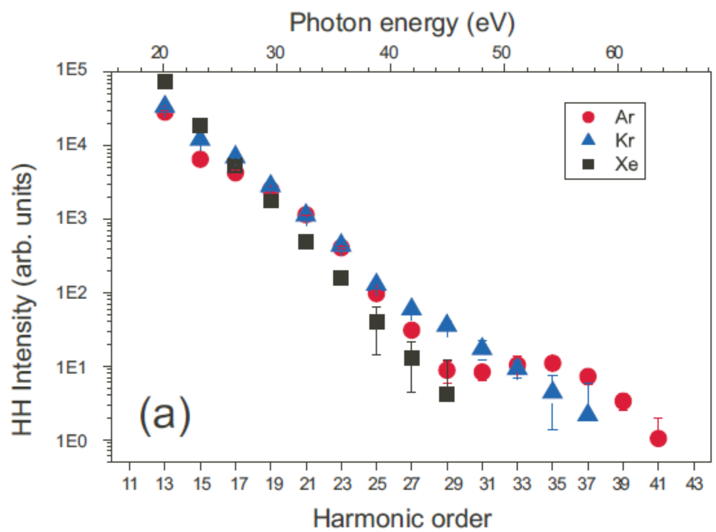
- High energy photon
  - $\hbar\omega$=1.4 keV, 14 PW, 720 nm
    - J Seres et al Nature 433 596 (2005)
- Ultrashort pulses
  - Single 80 asec pulses
    - G. Sansone et al., Science 314, 443 (2006).
- High intensity
  - Conversion Efficiency ~ 10^{-7}@ 300 eV, 1.6μm
    - Takahashi et al, PRL 101, 253901 (2008)



3 step model

$U_p[\mathrm{eV}] = 93.8 \times I\,[\mathrm{PW/cm}] \times \lambda[\mu\mathrm{m}]^2$





Minemoto et al, PRA08

Photo recombination

$A^{+} + e^{-} \rightarrow A + \hbar\omega_X$

Inverse process

Detailed balance

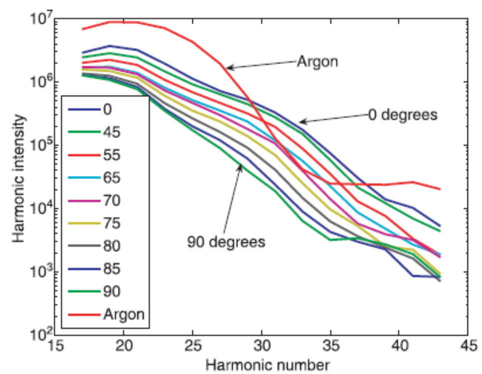
Photo ionization

$A + \hbar\omega_X \rightarrow A^{+} + e^{-}$

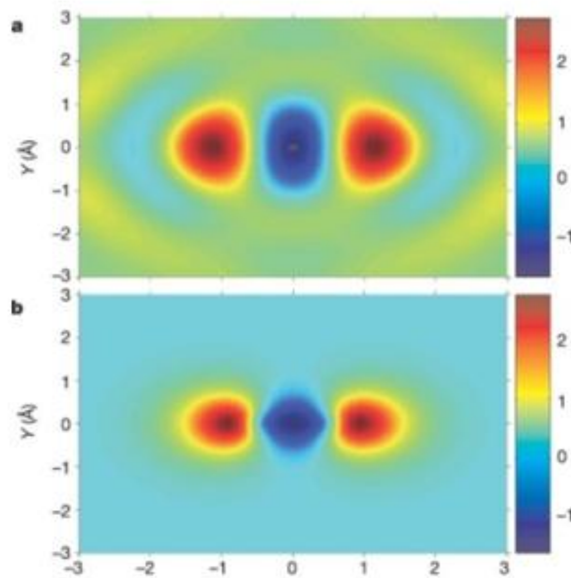
"Tomography" of molecules

- Re-construction of molecular orbitals from HHG spectra

HHG

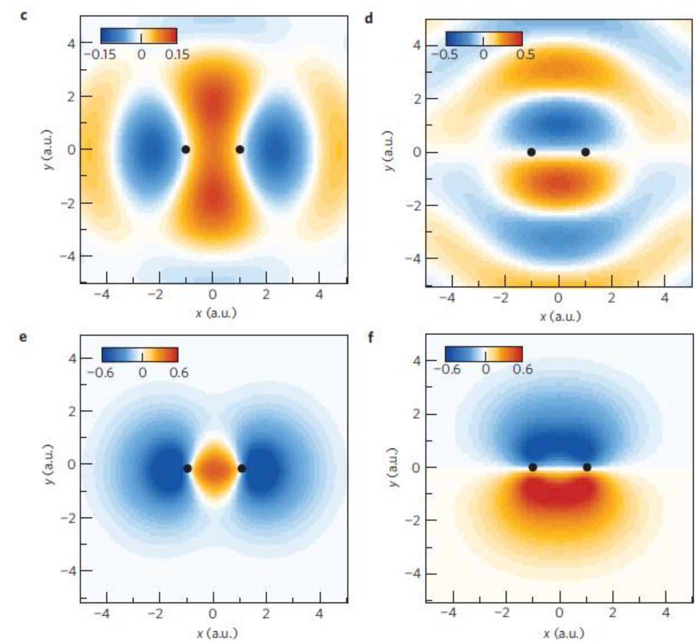


N_2 HOMO



FT

N_2 HOMO HOMO-1



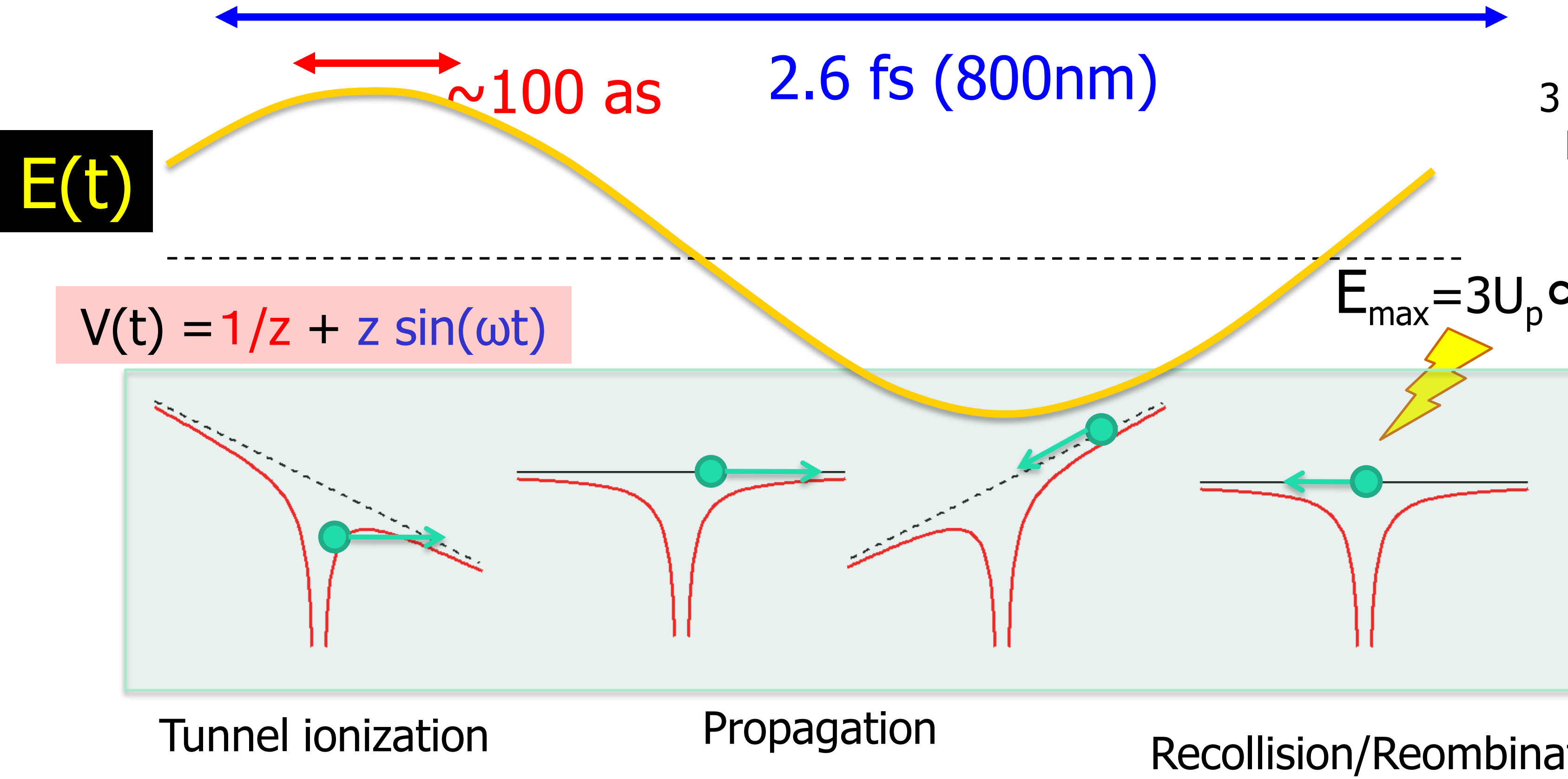
Itatani, J. et al. Nature 432, 867871 (2004)

Haessler et al, Nature Phys. 6 200 (2010)

Molecular imaging using rescattering electrons

- Laser induced phenomena
  - High Harmonic Generation (HHG)
  - High-energy Above Threshold Ionization (ATI)
  - Non seqtuential double ionization (NSDI)
  - etc

Self imaging



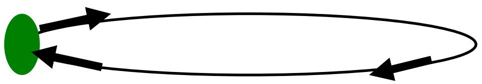
3 step model
P. Corkum

Re-examine: Factorization Formula

TM et al, PRL 100, 013903 (2008)

HHG & ATI --- Collision processes induced by laser

$$S = \sigma \; W$$



HHG spectrum
ATI spectrum
(Non linear process)

Momentum distribution of
rescattering electrons
(**laser info,Non linear**)

Photorecombination cross section
Elastic cross sections
(Target information, linear)

Dynamical imaging (Charge distribution etc.)

Beyond plane wave approx (FT or Born)
Scattering states (Coulomb + short range)
E ~ 50 eV @ 10^{14} W/cm^2

Target information from HHG

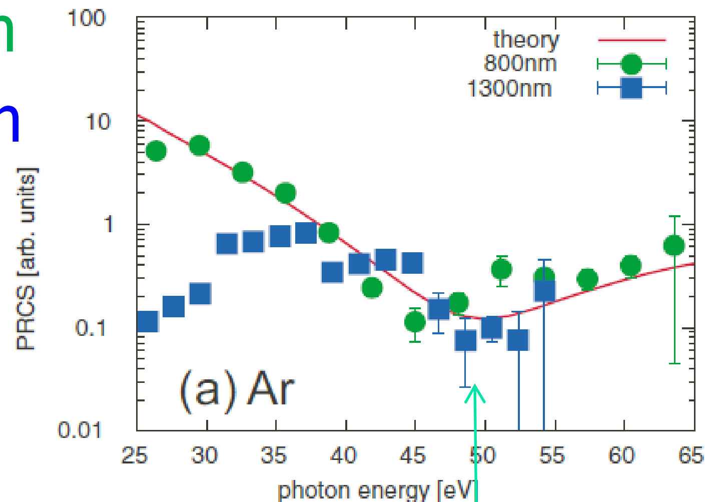
- Retrieval of photo recombination cross sections

$S = \sigma W$ $\sigma = S/W$

Ar ~10^{14} W/cm^2

800nm

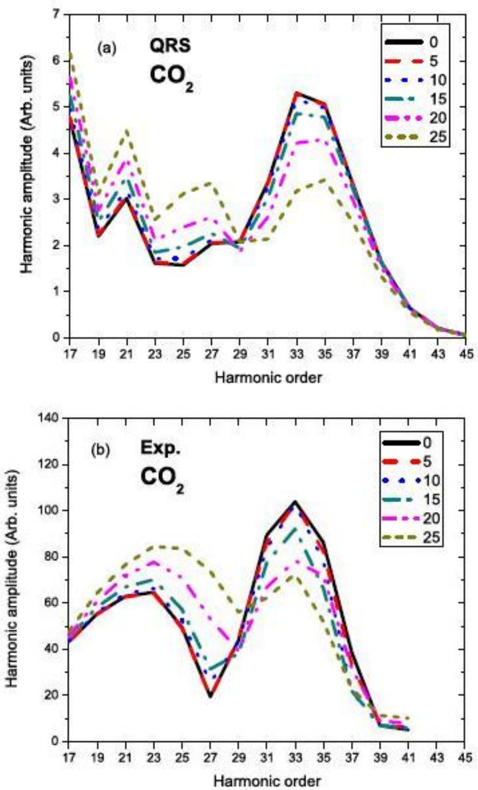
1300nm



Cooper minimum
From D to P states

S. Minemoto et al, PRA 061402 (2008)

CO_2



AT Le et al, PRA80, 013401 (2009)

Reconstruction of charge densities from PES

TM et al, JPB 09, J. Phys. Conf. Ser.09

Ueda, Okunishi $\sim 10^{14}$ W/cm^2

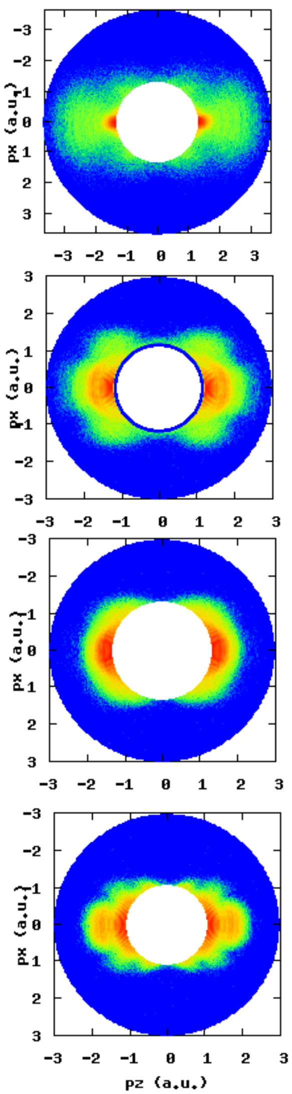
Exp. PES (ATI) **Elastic DCS, σ(p,θ)** **Charge density, Z(r)**

Ne

Ar

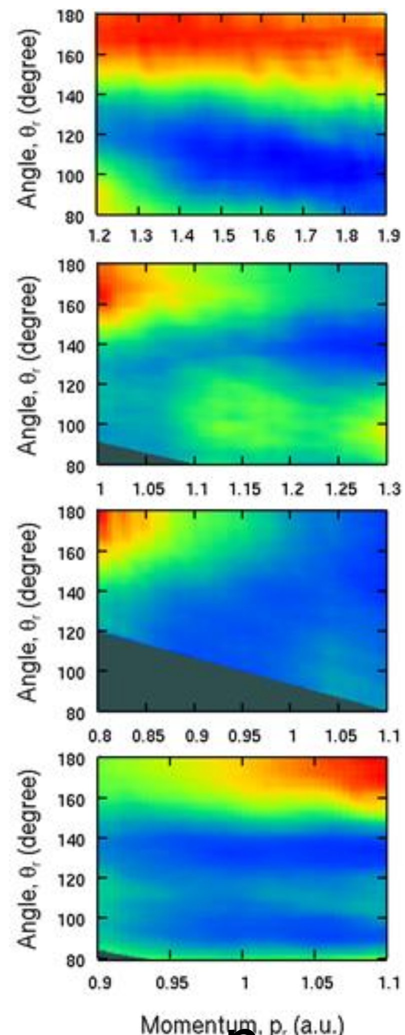
Kr

Xe



S=σW

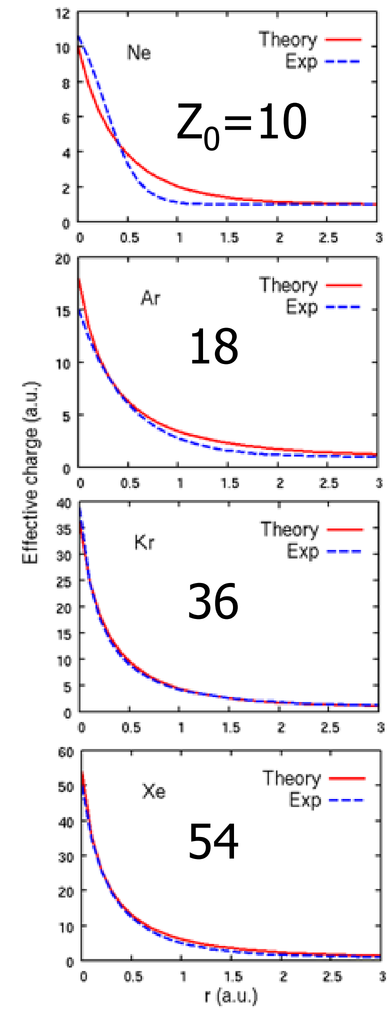
θ



p

Inv. scattering problem

(Fitting)



r

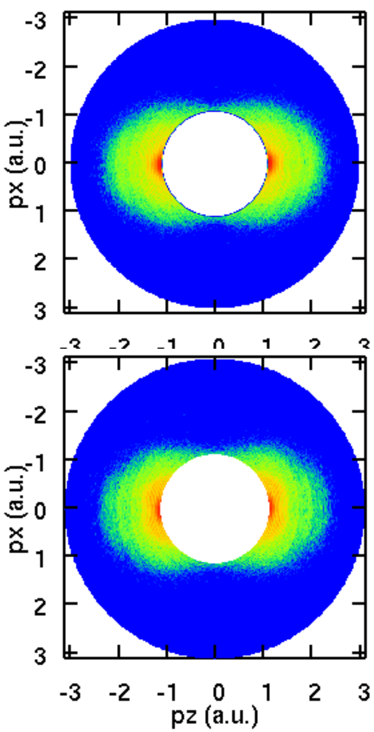
DCS of molecular targets

R. Luccese, M. Okunishi, TM, K. Ueda in preparation

2 center interference
p cosθ = const

PES

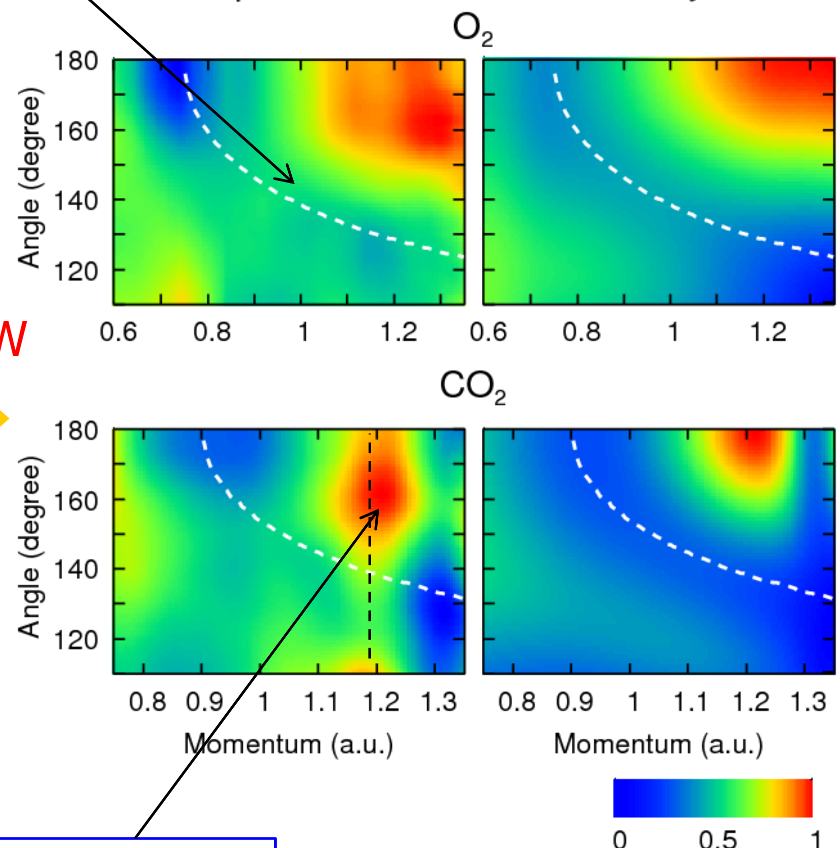
O_2



CO_2

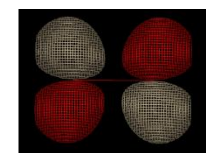
S=σW

DCS



Shape
resonance

Inv.



Underway

Factorization formula

- **S=σ W** [QRS: C. D. Lin et al, JPB 43 122001 (2010)]
  - HHG&ATI with TDSE
    - for Atoms, PRL100,013903(08) etc
  - Experiments
    - PE spectra:PRL100,143001&143002(08) etc
    - HHG spectra: PRA78,061402 (08) etc
  - Applications:
    - Molecular dipole transion matrix (CO_2 etc): AT Le, et al, PRA (09)
    - CEP determination: J. Phys. B 42 065402 (09) etc
    - NSD of He: Z. Chan et al, PRL (10)
  - Analytical derivation
    - SFA (Born): A. Čerkić, E. Hasović, D. B. Milošević & Becker
    - Effective range theory: Frolov, Manakov & Starace
    - Adiabatic theory: Tolstkhin &TM

Re-Re-examine: Adiabatic theory

With O.I. Tolstikhin

- Adiabatic parameter

$$\varepsilon = \frac{\text{Atomic time scale } (1/E_0)}{\text{Laser period } (T_0/2\pi)} \sim \frac{1}{T_0\alpha^2}$$

$E_0 = -\alpha^2/2$

F_0 : Electric field strength

T_0 : Laser period

- Perturbation parameter

$$\xi = \frac{F_0}{|E_0| \times |2E_0|^{1/2}} \sim \frac{F_0}{\alpha^3}$$

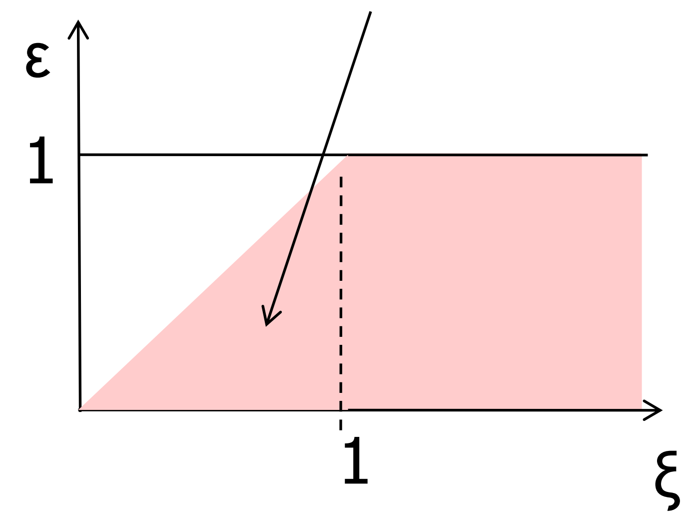
Keldysh $\gamma = \frac{\varepsilon}{\xi}$

$\varepsilon < \xi < 1$

- Adiabatic theory
  - Asymptotics:

$$\varepsilon \to 0, \;\; \xi(\varepsilon^0) = const$$

- Region of validity
  - $\varepsilon < \min(1, \xi)$



Basic idea of adiabatic theory

- Shrödinger Equation

$$i\frac{\partial}{\partial t}\psi(r,t)=\left[-\frac{1}{2}\nabla^2+V(r)+F(\varepsilon t)z\right]\psi(r,t)$$

- Asymptotics $\varepsilon \to 0$

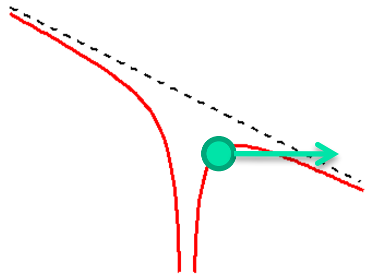
$$\psi(\nu,\mu)=A(\varepsilon^{\nu})\exp[iS(\varepsilon^{\mu})]$$

$$\psi=\psi_a(0,-1)+\psi_r(\nu>0,-3)$$

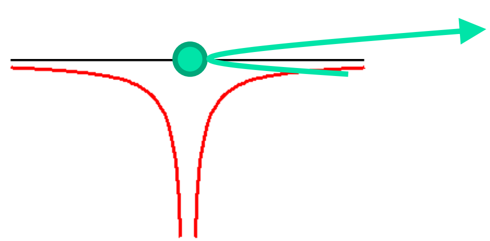
$\nu = 1/2$ (1D)

$3/2$ (3D)

Adiabatic part



Rescattering part



1D-Zero range potential model

Tolstikhin, TM, SW,
PRA 81, 033415 (2010)
[27 pages, 165 Eqs]

Schrodinger eq

$$i\frac{\partial}{\partial t}\psi(x,t)=\left[-\frac{1}{2}\frac{\partial^2}{\partial x^2}-\alpha\delta(x)+F(\varepsilon x)x\right]\psi(x,t)$$

Wave function

$$c(t)=\alpha^{-1/2}\psi(0,t)$$

$$\psi(x,t)=\frac{e^{i\pi/4}\alpha^{3/2}}{\sqrt{2\pi}}\int_{-\infty}^{t}\exp\left[iS(x,t;0,t')\right]\frac{c(t')dt'}{\sqrt{t-t'}}$$

Integral equation

$$c(t)=\frac{e^{i\pi/4}\alpha}{\sqrt{2\pi}}\int_{-\infty}^{t}\exp\left[iS(0,t;0,t')\right]\frac{c(t')dt'}{\sqrt{t-t'}}$$

Action

$$S(y_f,t_f;y_i,t_i)=\int_{t_i}^{t_f}\exp\left[\frac{1}{2}u^2(t)-F(t)\right]dt$$

$$u(t)=-\int_{-\infty}^{t}F(t)dt+u(t\to-\infty)$$

Ionization spectrum

$$P(k)=\alpha^3\left|\int_{-\infty}^{\infty}\exp[-iS(k,t)]c(t)dt\right|^2$$

Asymptotics

Schrodinger eq

$$c(t)=\frac{e^{i\pi/4}\alpha}{\sqrt{2\pi}}\int_{-\infty}^{t}\exp[iS(0,t;0,t')]\frac{c(t')dt'}{\sqrt{t-t'}}$$

Asymptotics

$$c(t)=c_a(t)+c_r(t)$$

Adiabatic

$$c_a(t)=a(\varepsilon^0)\exp[is(\varepsilon^{-1})]$$

Rescattering

$$c_r(t)=a(\varepsilon^{\nu>0})\exp[is(\varepsilon^{-3})]$$

Solution

$$c_a(t)=\alpha^{-1/2}\phi(0;F(t))\exp\left(-iE_0t-i\int_{-\infty}^{t}[E_0(F(t'))-E_0]dt'\right)$$

Siegert state in a static E-field

Regularity & Outgoing

$$c_{r(1loop)}(t)=T(\tilde{u}_f^+)c_r^{(a)}(t) \qquad c_r^{(a)}(t)=\frac{e^{i\pi/4}\alpha}{\sqrt{2\pi}}\int_{-\infty}^{t}\exp[iS(0,t;0,t')]\frac{c_a(t')dt'}{\sqrt{t-t'}}$$

Scattering（Transmission index）

Factorization formula: Saddle Point approximation

$$P(k)=\alpha^3\left|\int_{-\infty}^{\infty}\exp[-iS(k,t)]c(t)dt\right|^2$$

Spectrum $$P(k)=2\pi\left|I_a(k)+I_r(k)\right|$$

$$\zeta(k)=\left[\frac{3i}{4}\left[S_0(k,t_i^-)-S_0(k,t_i^+)\right]\right]^{2/3}$$

Adiabatic $$I_a(k)=\alpha^{3/2}U(\zeta(k))\left[\frac{dt_i^+/dk}{s_i\alpha(t_i^+)}\right]^{1/2}e^{iS(k,t_i^+)}c_a(t_i^+)$$

$$U(\zeta)=2\sqrt{\pi}\exp\left(\frac{2}{3}\zeta^{3/2}\right)\zeta^{1/4}Ai(\zeta)$$

Rescattering $$I_{r(1loop)}(k,\sigma_1)=A(\tilde{u}_f^+)\alpha^{3/2}U(\zeta(t_f^+))\left[\frac{dt_i^+/dk}{s_i\alpha(t_i^+)}\right]^{1/2}e^{iS(k,t_f^+)+iS(t_f^+,t_i^+)}c_a(t_i^+)$$

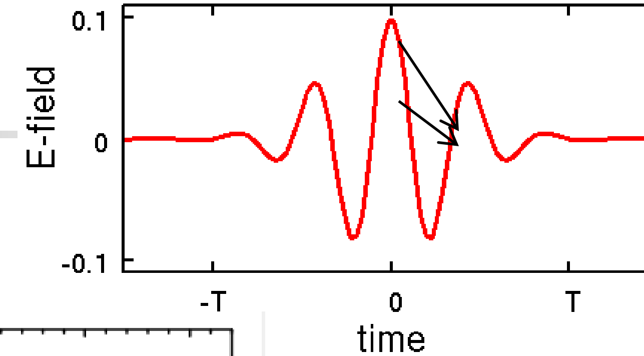
$$P_{1loop}(k)=\underbrace{\left|c_a(t_i(t))\right|^2\times\Gamma(t_i(t))\times\left|\frac{dt_i(t)}{dk}\right|}_{W}\times\underbrace{\left|R(k+v(t_r(k))\right|^2}_{\sigma}$$

Survival Prob | Ionizaion rate | propagation | Cross section

$$P_{sl}(k)=P_{1loop}(k)\times Ai(S_{short}+S_{long})$$

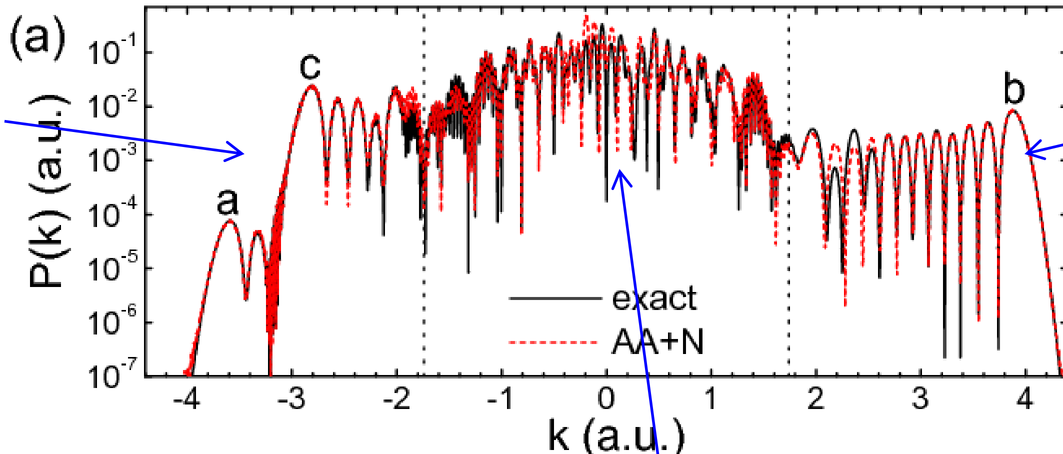
For HHG, Okajima, TM, OTI

Ex) 5 cycle pulse



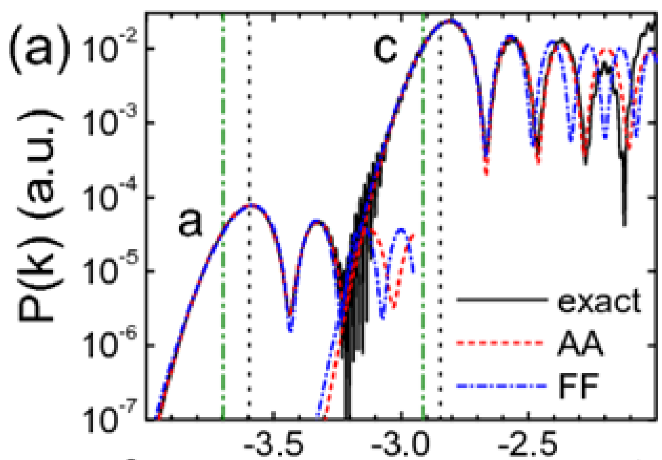
- 5 fs, 800 nm, 3.5×10^{14} W/cm^2

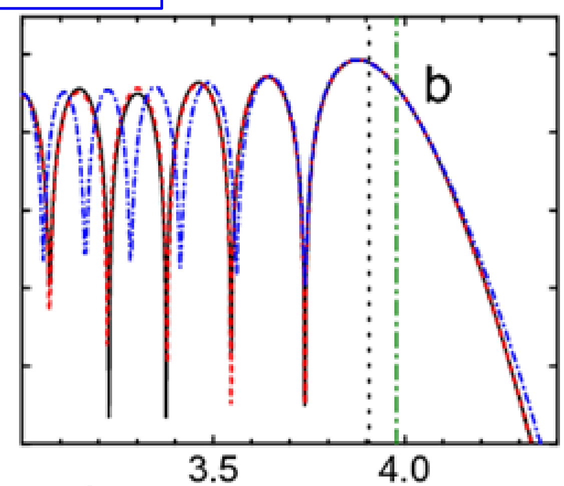
Rescattering Long+short



Rescattering Long+short

Adaiabtic＋Rescattering



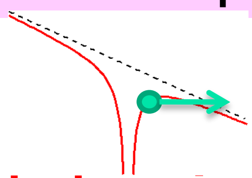


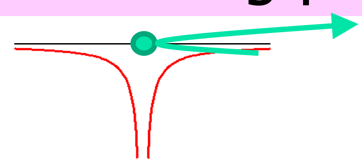
Re-examination of atomic states in a electric field

- Asymptotics in the adiabatic theory

$$\psi = \psi_a(\varepsilon^{-1}) + \psi_r(\varepsilon^{-3})$$

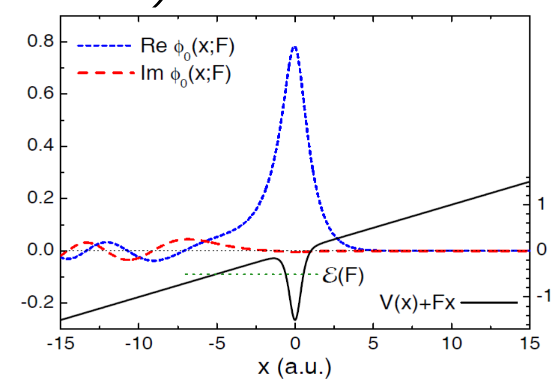
Adiabatic part　　Rescattering part





- Siegert states in a static electric field
  - Regularity & Outgoing

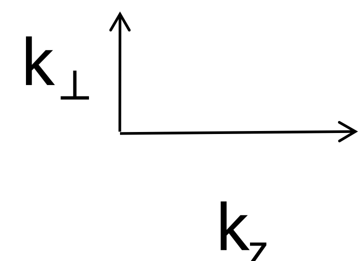
$$\psi_a(\mathbf{r},t) = \varphi_0(\mathbf{r};F(t))\exp\left(-iE_0 t - i\int_{-\infty}^{t}\left[E_0(F(t)) - E_0\right]dt'\right)$$

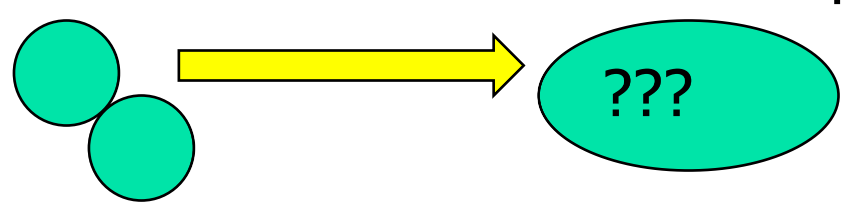


Atoms & molecules in a static field

J. Stark

- Stark effect
  - J. Stark (1913, Nobel Prize 1916)
  - Splitting of the atomic bound state energies
  - Perturbation among bound states
- Tunnel ionization
  - Keldysh (1964), ADK(1986)
  - Ionization rate, momentum distributions in week field limit
  - $P(k_\perp) \propto \exp(-k_\perp^2/3E)$ for H(1s)
- Quantum mechanical treatment
  - Strong field ? Over the barrier ?
  - Target dependence ?





Atomic Siegert states in an E-field

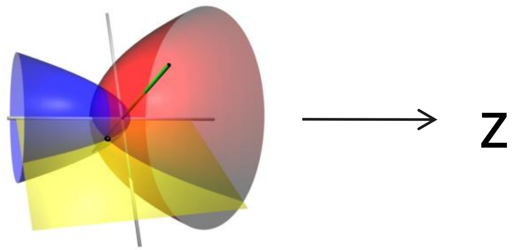
Batishchev, OIT, TM, PRA accepted

- Schrödinger equation + Regularity & Outgoing

$$\left[-\frac{1}{2}\Delta + \mathrm{V}(\mathbf{r}) + \mathrm{Fz}\right]\varphi_0(\mathbf{r};\mathrm{F}) = \mathrm{E}_0(\mathrm{F})\,\varphi_0(\mathbf{r};\mathrm{F})$$

  - Parabolic coordinates

$$\xi = r + z, \quad \eta = r - z, \ \phi$$

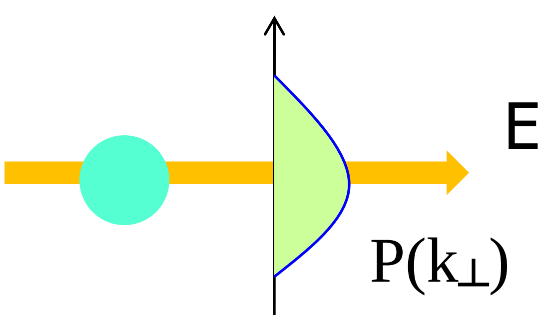


  - Asymptotic form (η→∞)

$$\phi(\xi,\eta;F)\big|_{\eta\to\infty} = \phi(\xi)\times\frac{2^{1/2}}{F^{1/2}\eta^{3/4}}\exp\left[\frac{iF^{1/2}\eta^{3/2}}{3}+\frac{iE\eta^{1/2}}{F^{1/2}}\right]$$

  - Transverse momentum distribution

$$P(k_\perp) = \frac{8\pi^2}{F}\left|\phi\left(\frac{k_\perp{}^2}{F}\right)\right|^2$$

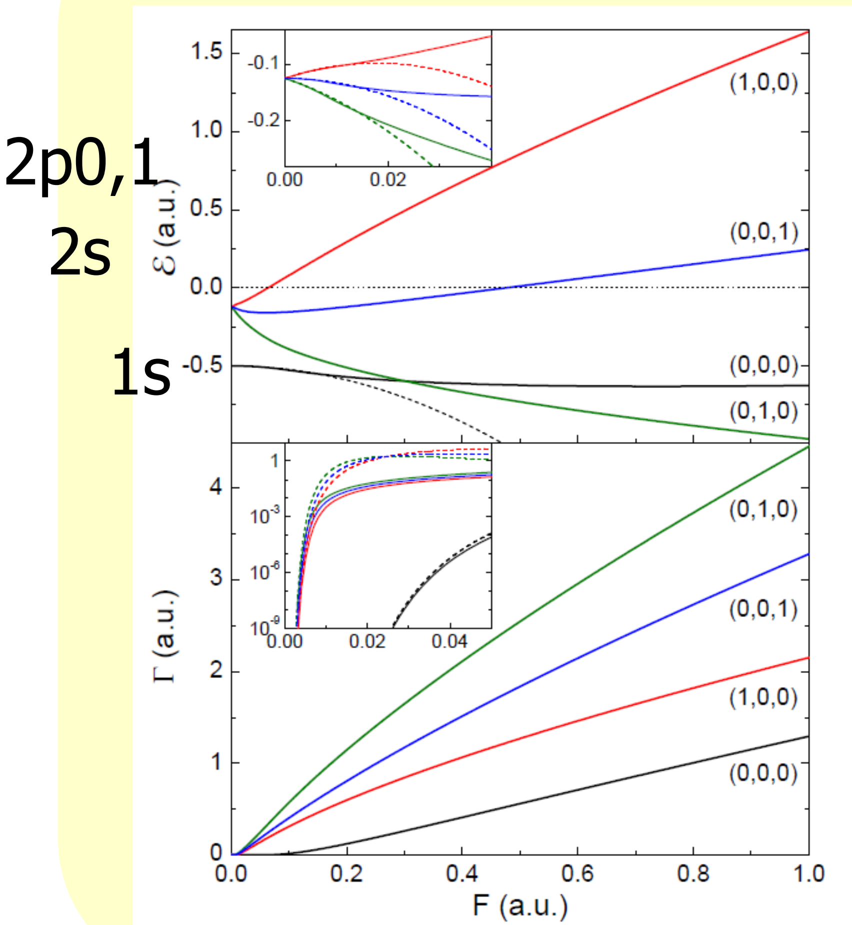


    - Complex R-matrix

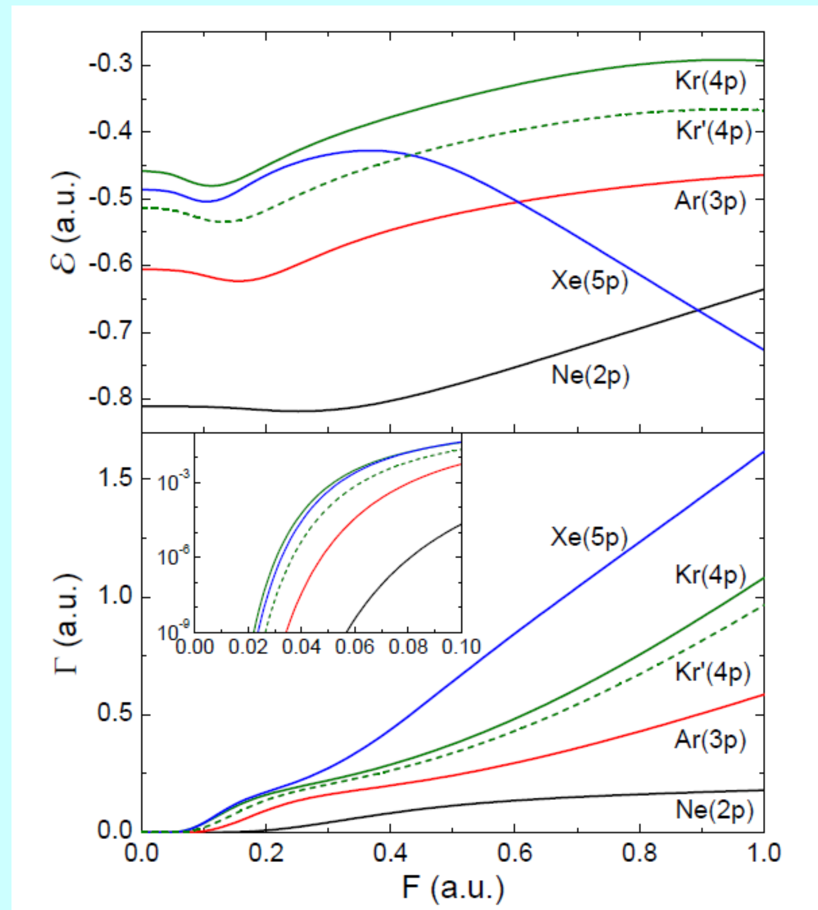
Energies & Width

$E = \varepsilon - i\Gamma/2$

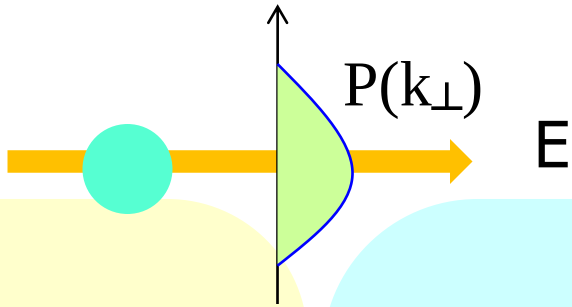
H



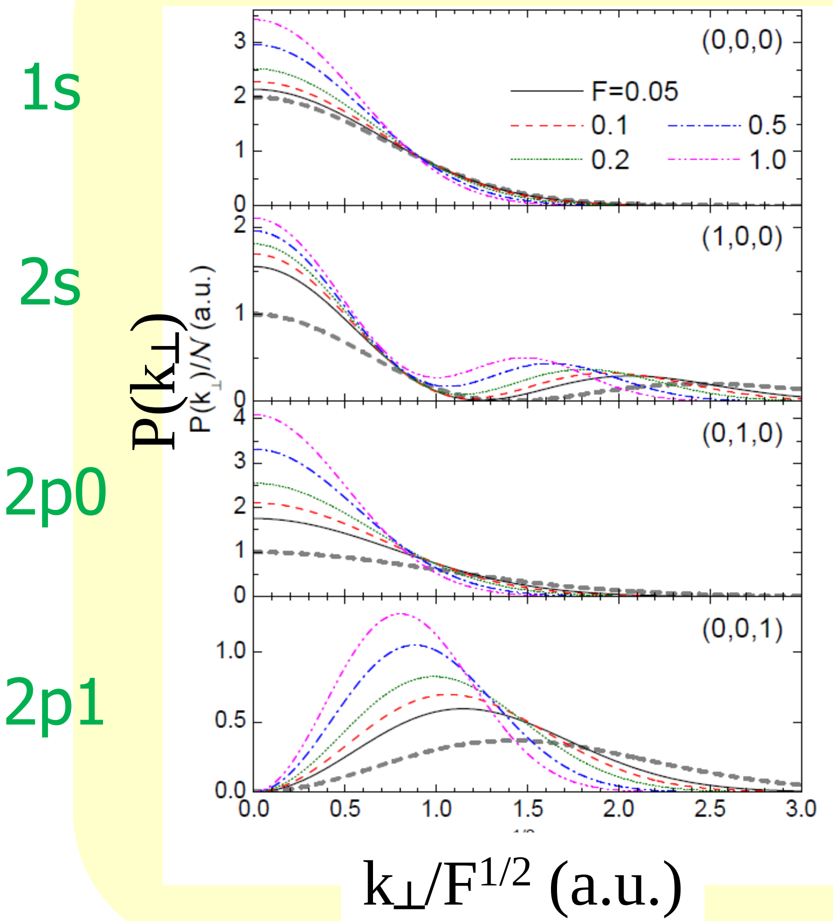
Rare gas atoms



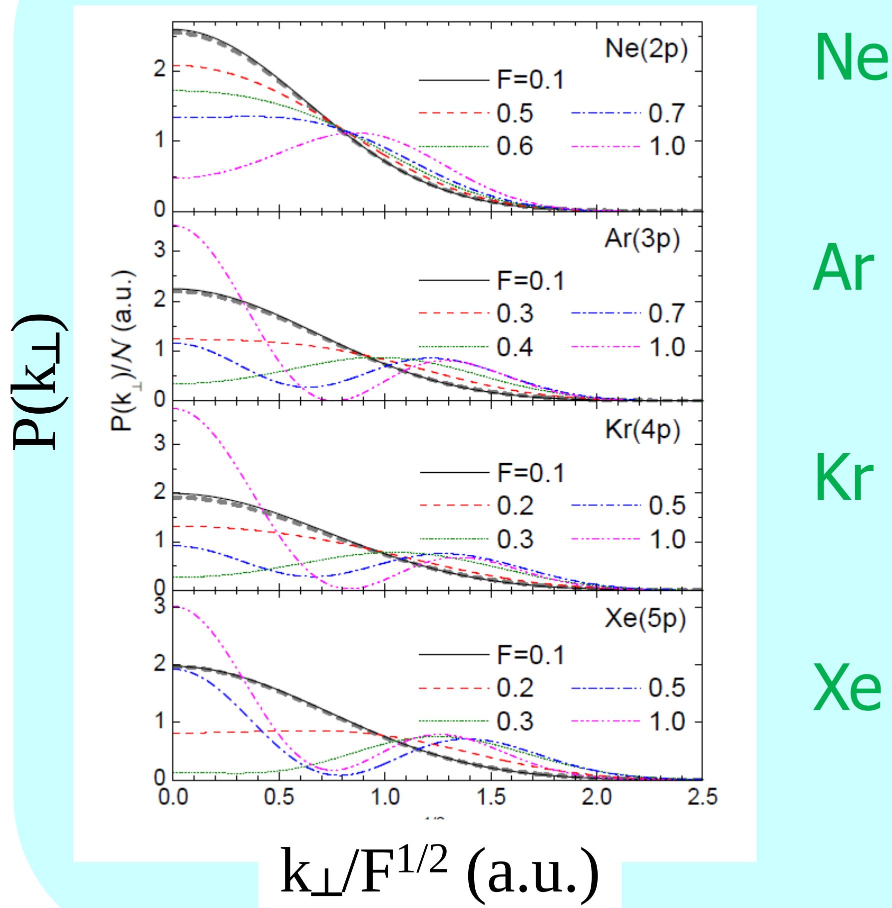
Transverse momentum distribution



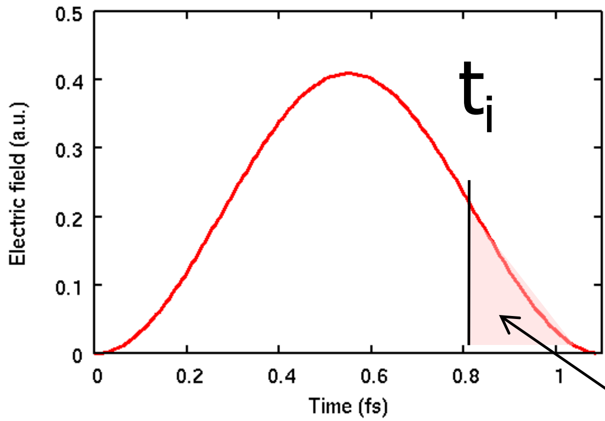
H atom



Rare gas atoms



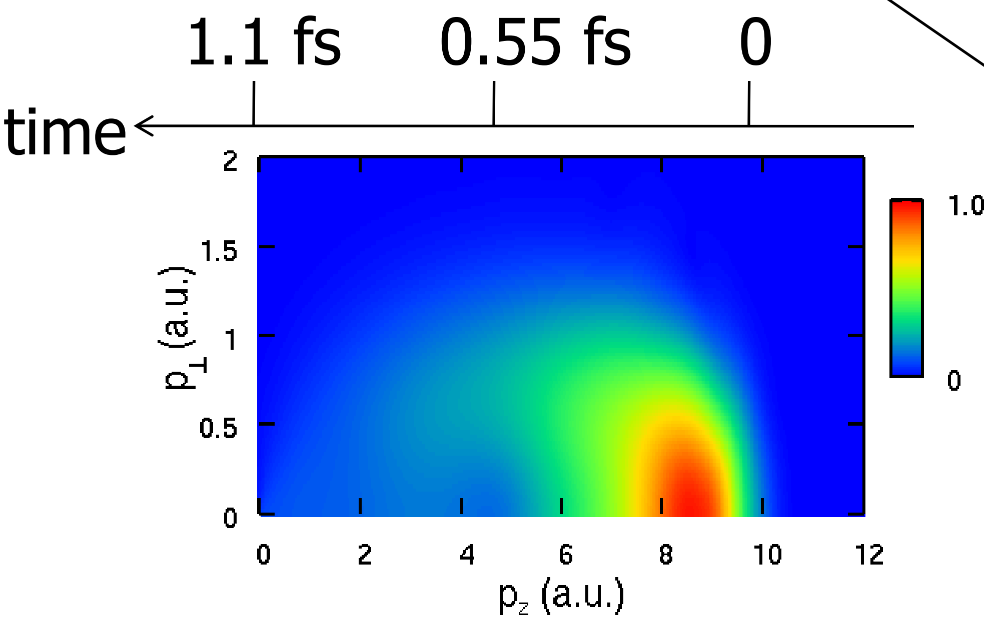
Ionization by a half cycle pulse



$I = 6 \times 10^{15}\ W/cm^2$

$T = 1.1\ fs$

Ar (3p)



$$p_z = \int_{t_i}^{\infty} E(t)\, dt$$

Ionization time

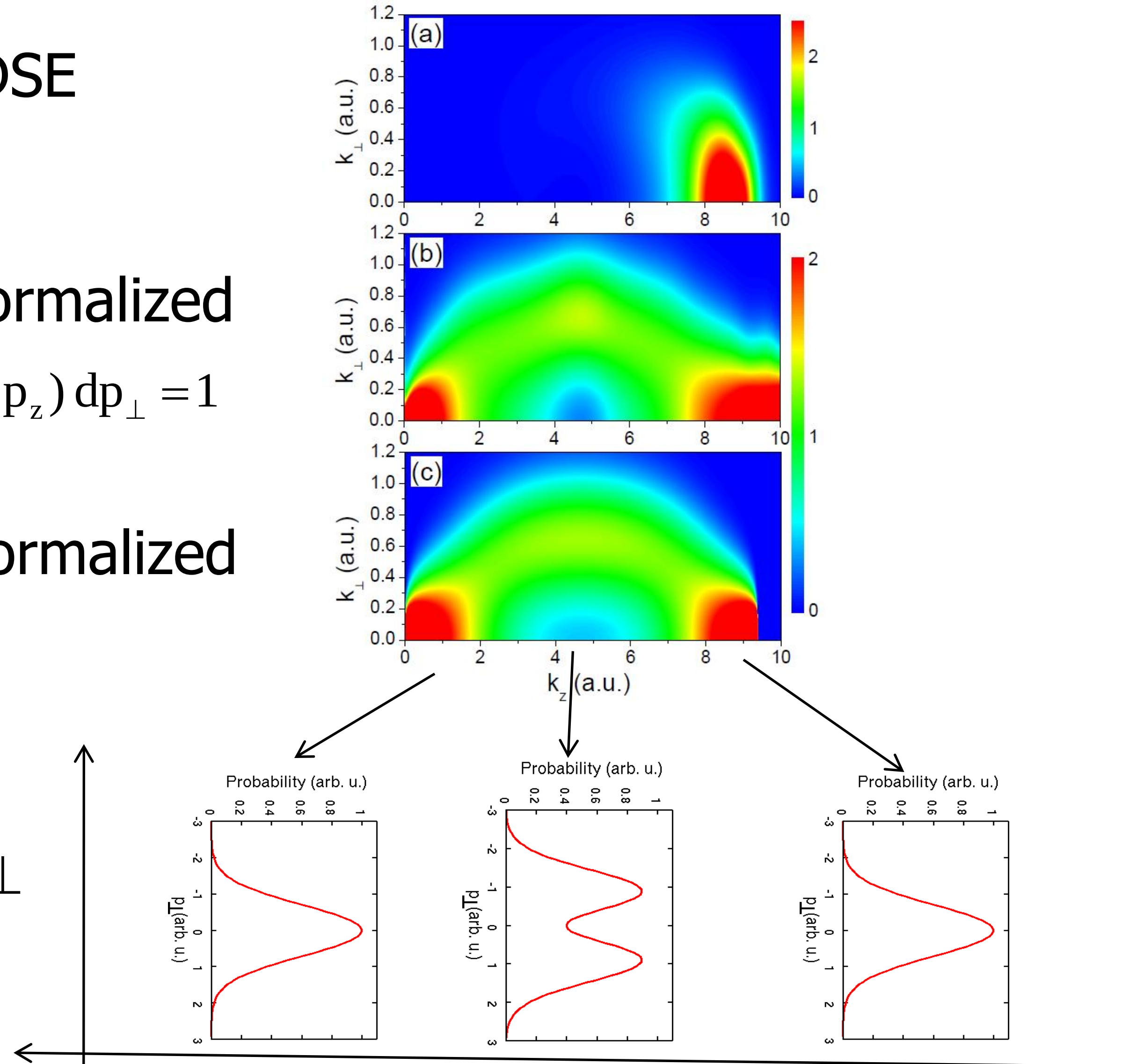
Time dependence of the tunneling wave packets

TDSE

TDSE, normalized

$$\int_0^\infty P(p_\perp, p_z)\, dp_\perp = 1$$

Static, normalized



$p_\perp$

time

1.1 fs

0.55 fs

0

Summary

- Ultra high resokution imaging
  - with HHG, ATI, etc
- Factorization formula
  - S=σ W
- Adiabatic theory
  - Adiabatic + Rescattring
- Tunnel ionization
  - Siegert states in a static field
- Adiabatic theory for Super intense X-ray ($\sim 10^{16}$ W/cm^2)
  - Stabilization against ionization
    - Toyota et al, PRA08, PRL09