Probing Atomic Dynamics on the Attosecond Scale

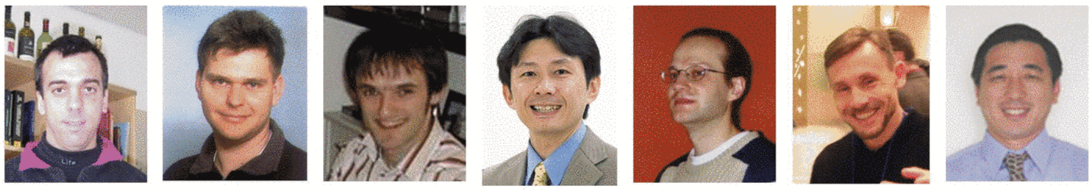
Joachim Burgdörfer

Institute for Theoretical Physics, Vienna University of Technology
Vienna, Austria, EU

Overview

- Attosecond electron wave-packets: time double slit, gratings, and holograms
- Electronic motion in atomic states: periodic orbit spectra
- Coherent excitation: wavepacket dynamics
  - time-domain images of Fano resonances
- "Artificial atoms": quantum dots and microwave cavities
- "Ultrashort" limit: half-cycle pulses

Collaborators:
Theory



D. Arbo (VUT) | I. Barna (Budapest) | A. Bärnthaler (VUT) | K. Ishikawa (U. Tokyo) | E. Persson (VUT) | C. Reinhold (ORNL) | X.-M. Tong (Tsukuba)

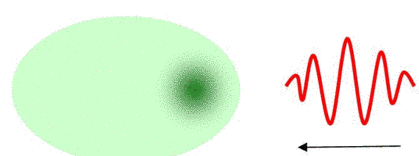
Experiment

J. Wang (UMass) | M. Wickenhauser (VUT) | S. Yoshida (VUT)

M. Drescher (Hamburg) | F. Krausz (MPI Garching)

Time resolved atomic dynamics

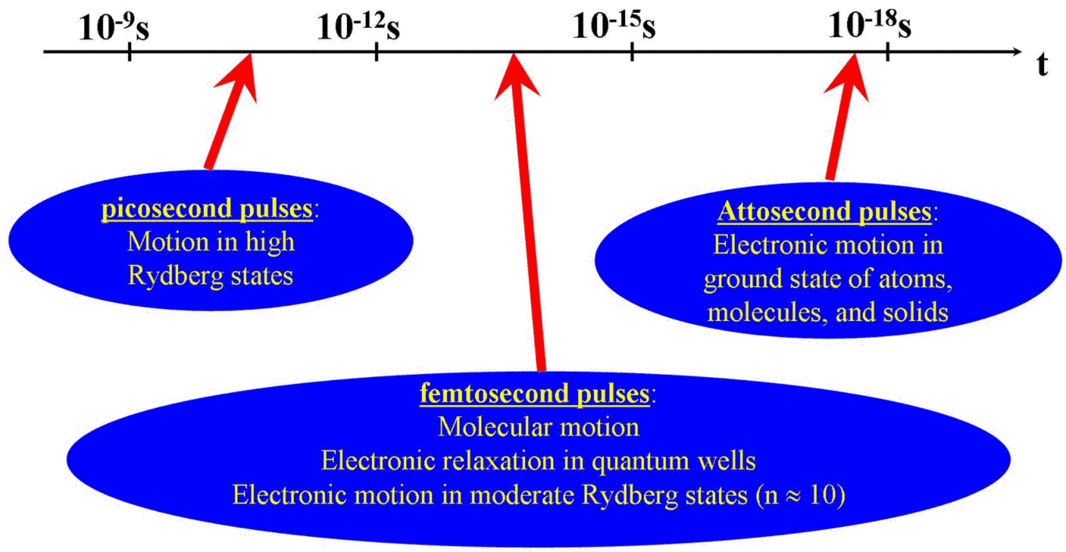
Goal: Time resolved electron wave packet



Time scale of electronic motion in atomic groundstate: ~100 as

How short is short?

"short": absolute time scale $\tau_p < \tau$



Electronic motion in atoms: periodic orbit "spectroscopy"

spectral density: $d(E) = \sum_n \delta(E - E_n)$

semiclassical limit (Gutzwiller trace formula)

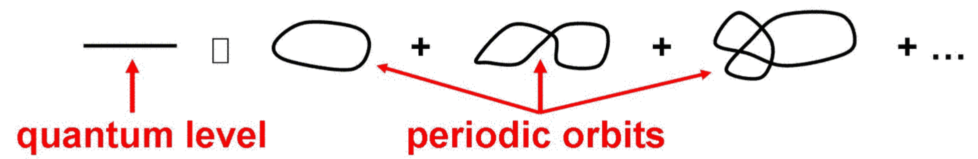
$$d(E) = d_{av}(E) + \frac{1}{2\pi} \sum_{j \in \{PPO's\}} T_j \sum_{k=-\infty}^{\infty} \frac{1}{\sqrt{\left|\det\left(M_j^k - I\right)\right|}} \cdot e^{i(kS_j - \sigma_{jk}\pi/2)}$$

$$S_j(E) = s_j \left|E\right|^{-1/2} = s_j z \, ; \qquad s_j = S(E=-1) = 2T_j$$

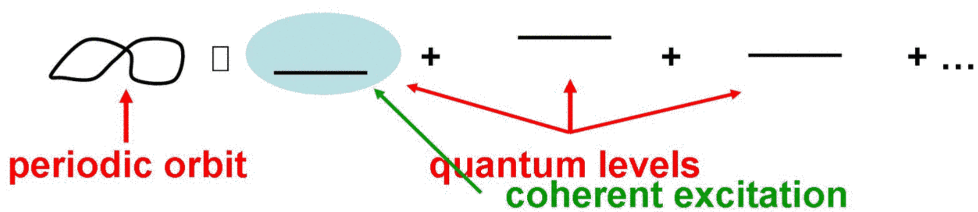
Coulomb-scaled action and periods

→ Fourier series of periodic orbits

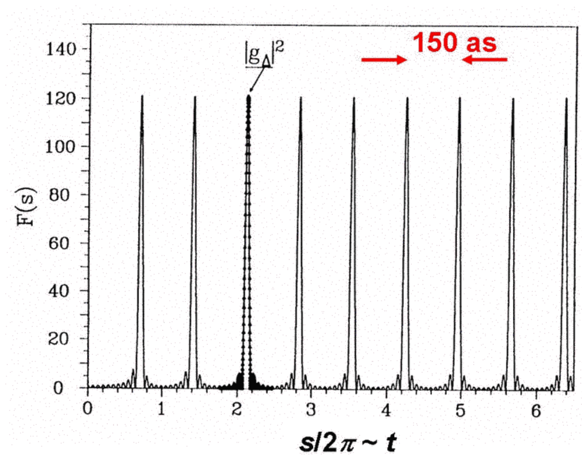
Fourier series of periodic orbits:



inverse Fourier transformation:

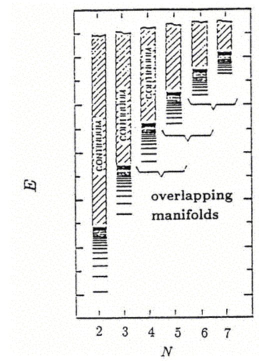


Periodic orbit spectrum of hydrogen

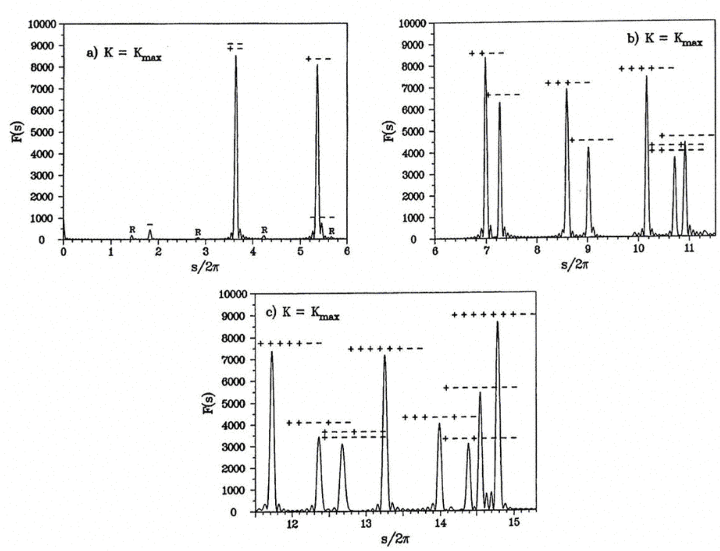


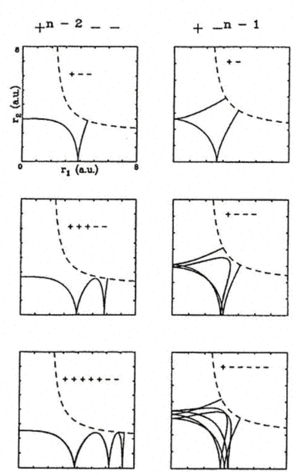
→ Bohr was right after all!

Periodic orbits of helium

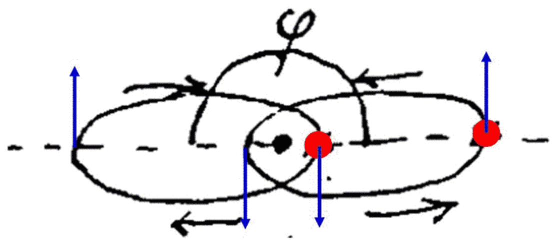


periodic orbits contributing near K_{max}





Correlated electron motion in He



(Heisenberg, 1922)

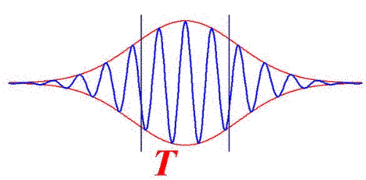
$$\vec{L} = 0$$

$$\left|\vec{\ell}_1\right| = \left|\vec{\ell}_2\right| \neq 0$$

QM: CI wavefunction

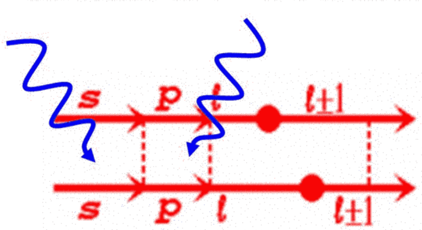
$$\left|\psi\right\rangle = \sum a_{ij}^{(s)} \left|s^{(i)}, s^{(j)}\right\rangle + \sum a_{ij}^{(p)} \left|p^{(i)}, p^{(j)}\right\rangle + \sum a_{ij}^{(d)} \left|d^{(i)}, d^{(j)}\right\rangle + \ldots$$

Electron correlation probed by attosecond pulses: two - photon double ionization (DI) of He



$+ He = He^{2+} + e_1^- + e_2^-$ **DI**

ω_{XUV}=91.6 eV
peak intensity: 10^{15} W/cm^2
duration: 150 - 450 attoseconds (10^{-18} s)



Laulan and Bachau, PRA 69, 033408 (2004)
Lambropoulos et al., PRA 72, 013410 (2005)
Parker et al., J. Phys B 34, L69 (2001)

Close-coupling method

Schrödinger Eqn. $$i\frac{\partial}{\partial t}\Psi(\mathbf{r}_1,\mathbf{r}_2,t) = \left(\hat{H}_{He} + \hat{V}(t)\right)\Psi(\mathbf{r}_1,\mathbf{r}_2,t),$$

Unperturb. ***H*** $$\hat{H}_{He} = \frac{\mathbf{p}_1^2}{2} + \frac{\mathbf{p}_2^2}{2} - \frac{2}{r_1} - \frac{2}{r_2} + \frac{1}{|\mathbf{r_1}-\mathbf{r_2}|},$$

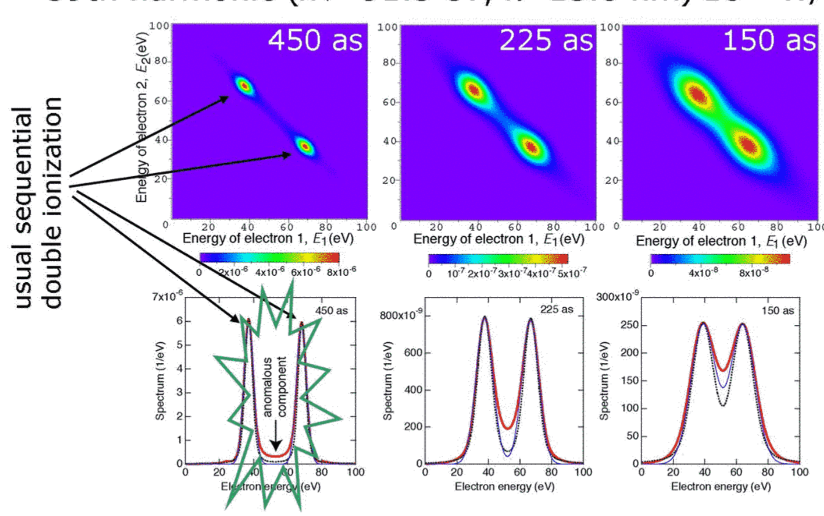
Perturbation $$\hat{V}(t) = -\sum_{i=1,2}\mathbf{E}(t)\cdot\mathbf{r}_i,$$

CC expansion $$\Psi(\mathbf{r}_1,\mathbf{r}_2,t) = \sum_{j=1}^{N} a_j(t)\Phi_j(\mathbf{r}_1,\mathbf{r}_2)e^{-iE_jt}.$$

Solve a coupled set of ***N*** ODEs

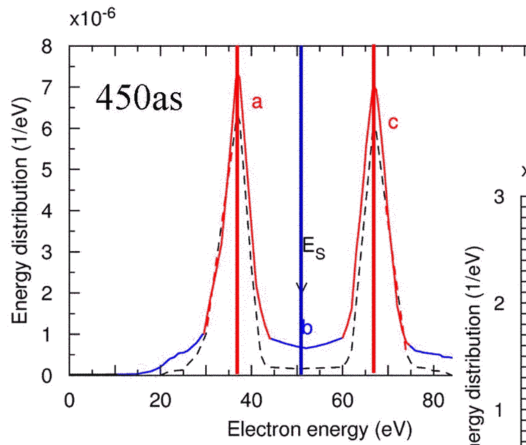
Electron energy distribution

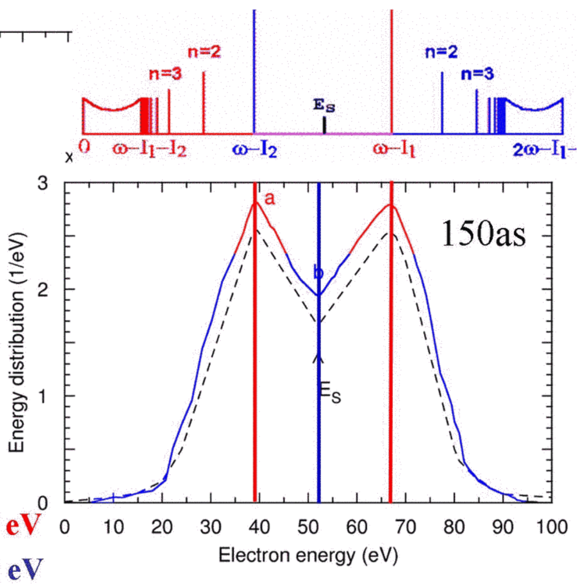
59th harmonic ($h\nu$=91.5 eV, λ=13.6 nm) 10^{15} W/cm^2



K.L. Ishikawa and K. Midorikawa, Phys. Rev. A 72, 013407 (2005)

DI electron spectra





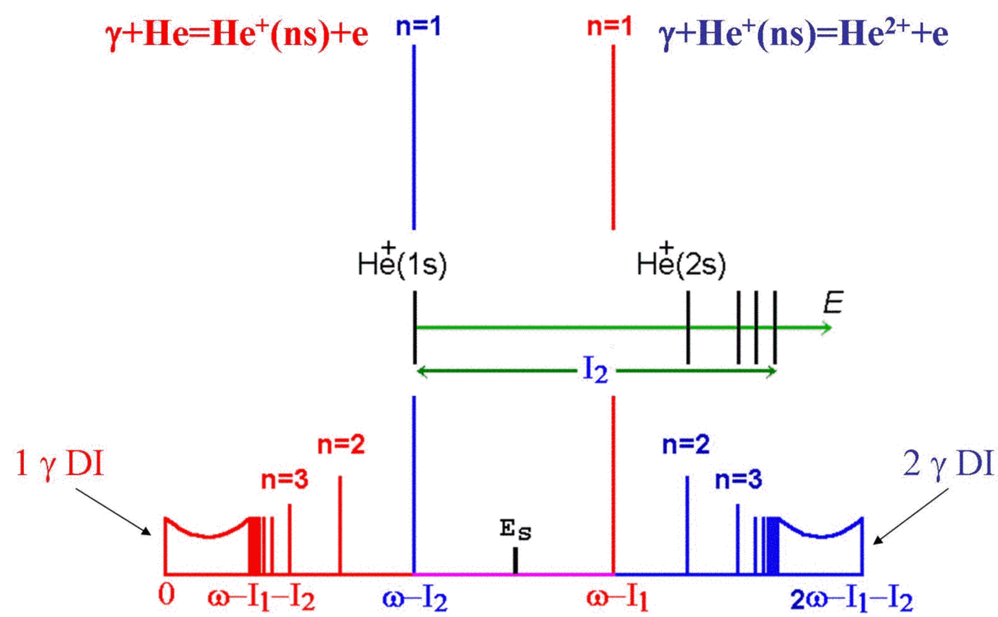
independent particle (IP): red, 37 eV

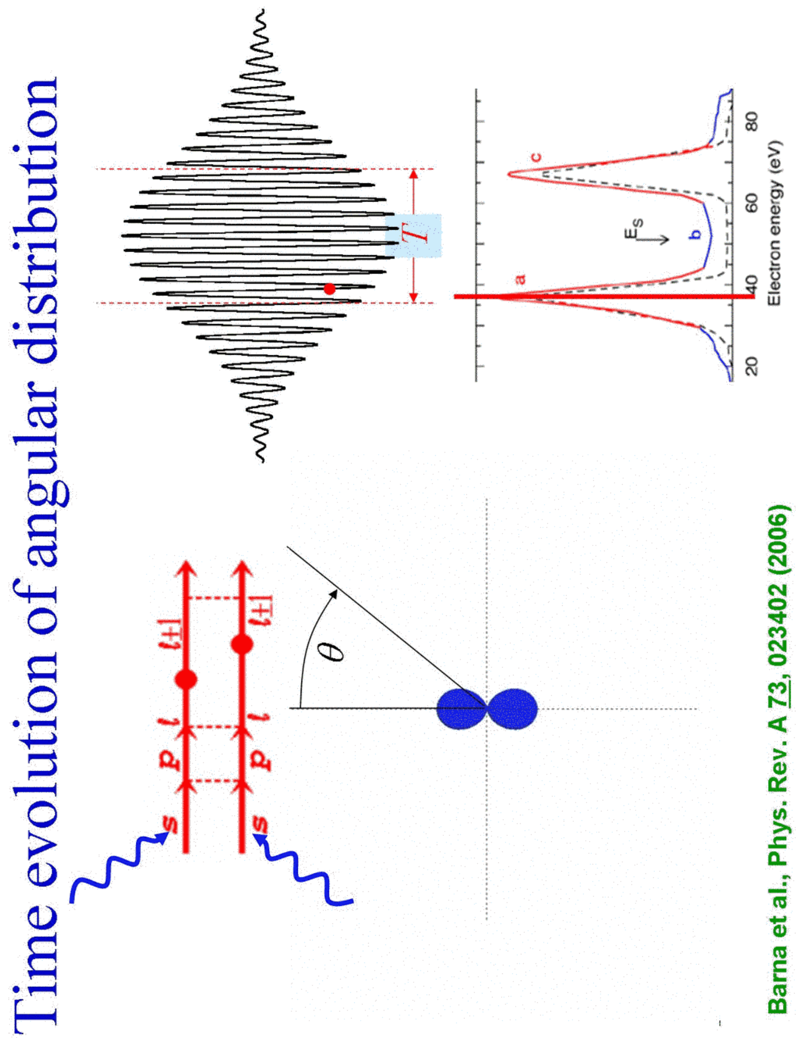
correlation induced (CI): blue, 52 eV

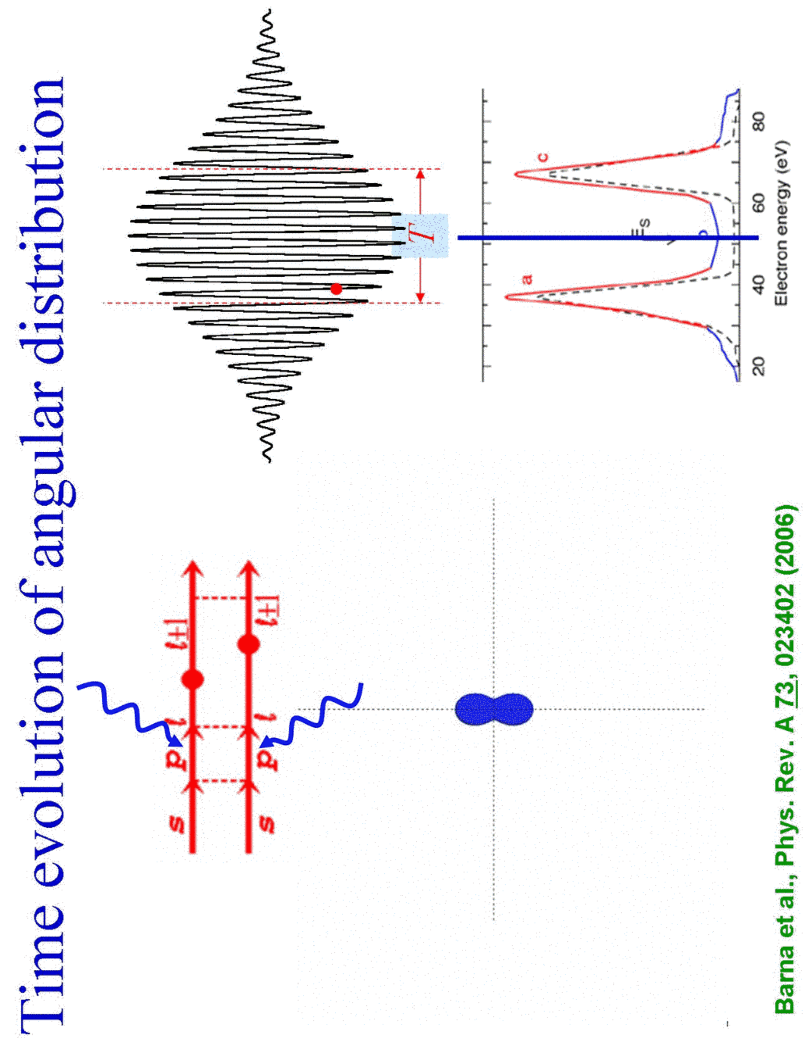
Ishikawa et al., PRA 72, 013407 (2005)

Barna et al., PRA 73,023402(2006)

Emission spectrum, schematically




Time evolution of angular distribution
T
Electron energy (eV)
20
40
60
80
a
b
c
E_S
s
p
l
l±1
θ
Barna et al., Phys. Rev. A 73, 023402 (2006)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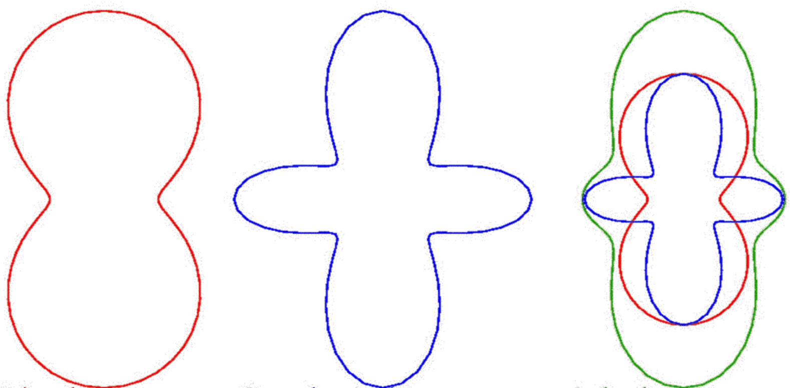
Barna et al., Phys. Rev. A 73, 023402 (2006)

Multipoles

$$\frac{d\sigma}{d\Omega} = \frac{\sigma_0}{4\pi}\left[1 + \beta P_2(\cos\theta) + \gamma P_4(\cos\theta)\right]$$

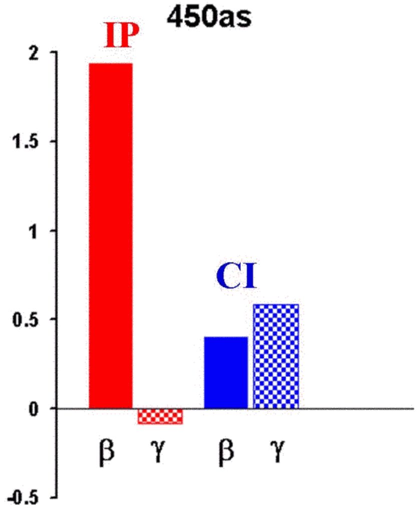


Dipole + Quadrupole = Admixture

β=*p*-wave

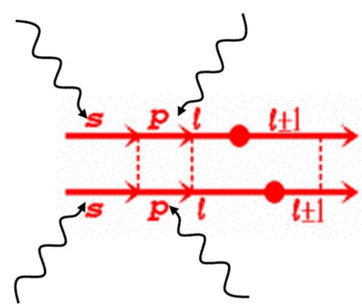
γ=*d*-wave

β & γ



initial state:

$$|i\rangle = a|ss\rangle + b|pp\rangle + \ldots$$



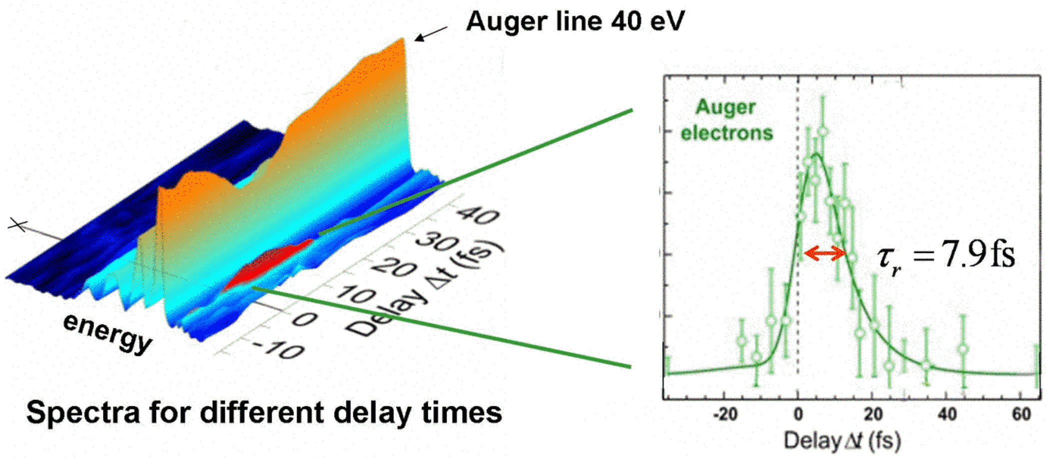
$$|f\rangle = b'|sd\rangle + c'|dd\rangle + \ldots$$

Mapping of initial angular correlation

Proof-of-principle experiment:

Auger decay $\tau_r = 8\ \text{fs}$

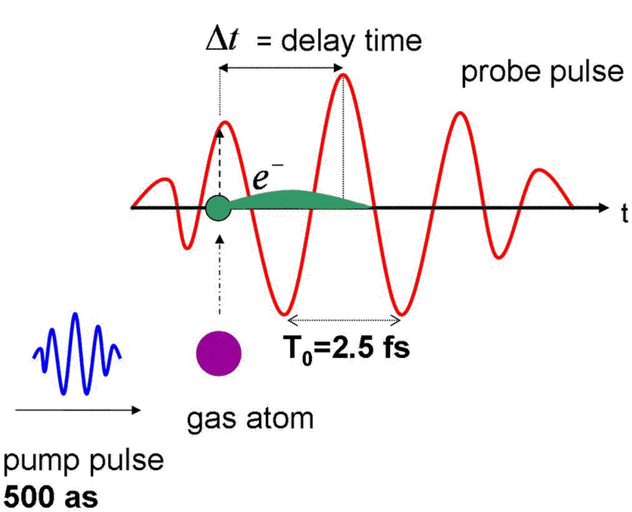
M. Drescher F. Krausz

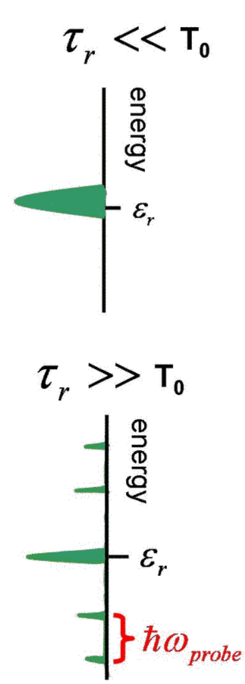


Spectra for different delay times

⇨ **extract lifetime of Auger decay**

XUV-IR pump-probe: attosecond streaking

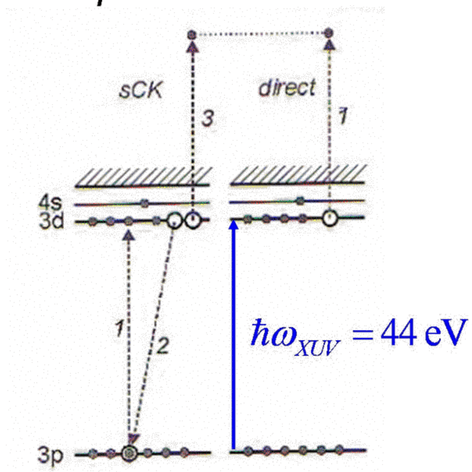




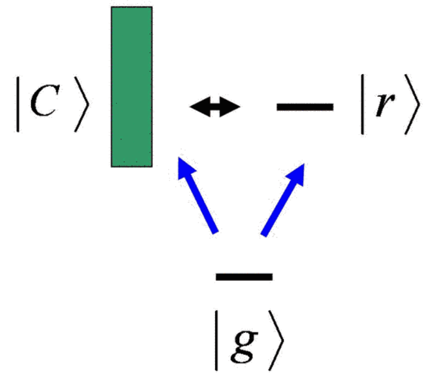
Coherent excitation

super Coster Kronig

Cr: τ_r = 800 as



schematic:



Goal: Time resolved evolution of coherent superposition

Model Hamiltonian:

$$H = H_0 + \underbrace{V_{bc}}_{\text{bound-continuum coupling}} + \underbrace{H_L}_{\text{Laser}}$$

$$H_a = H_0 + V_{bc}$$

Approximations:

- **XUV-pump pulse:** $(\hbar\omega_{XUV} \sim 50-100eV)$
  - First order perturbation theory

$$|\psi(t)\rangle = -i\int_{-\infty}^{t} dt' U(t,t') H_{XUV}(t') |g\rangle_{t'}$$

- **Probe pulse:**
  - Strong field approximation $(\hbar\omega_L \sim 1.6eV)$

$$U(t,t') = U_{probe}(t,t') - i\int_{t'}^{t} dt'' U_{probe}(t,t'') H_{CI} U_{CI}(t'',t')$$

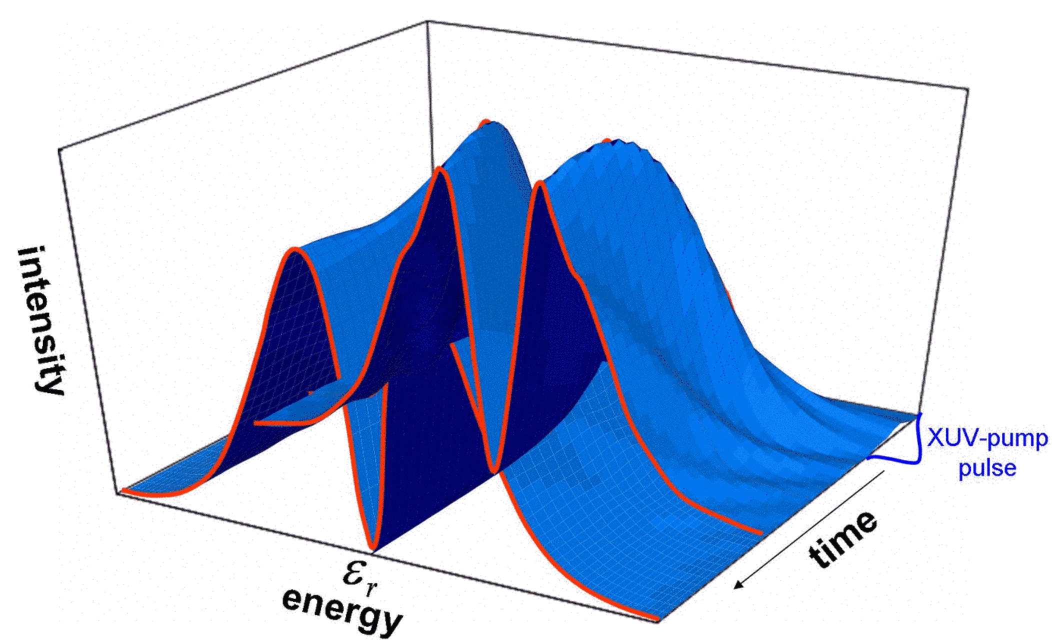
Ionization amplitude:

$a = a_{\text{resonant}}$ =

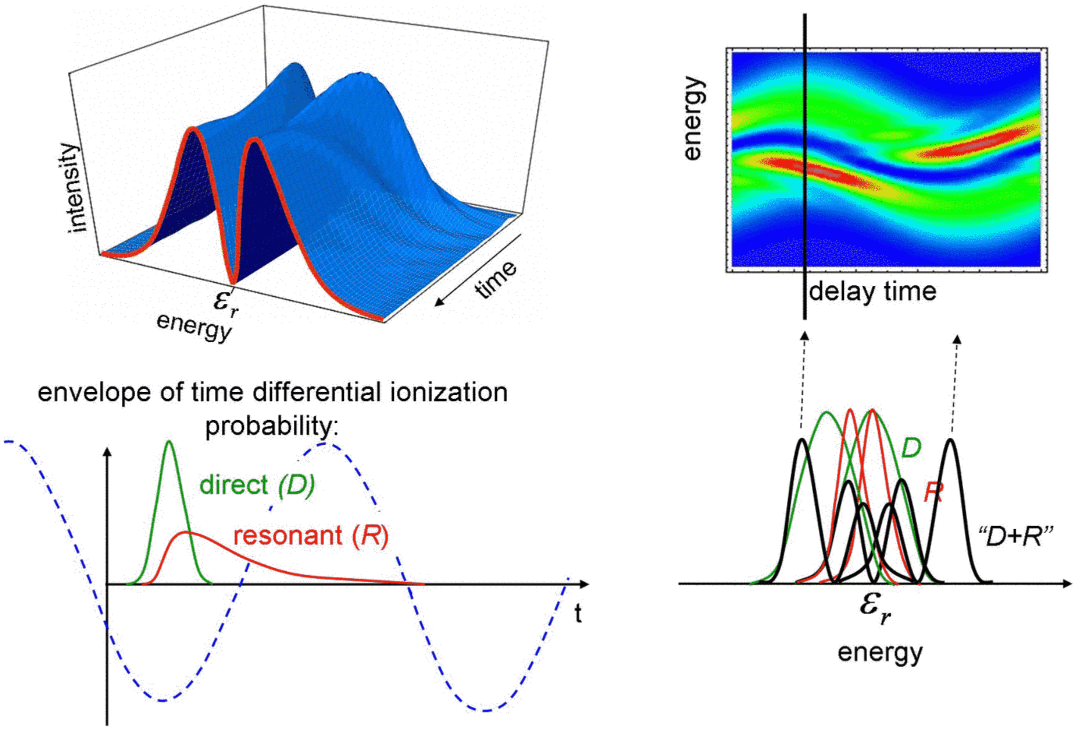
$$= -\int_{-\infty}^{t} dt' e^{i\Phi_E(t',t)} V_{E(t',t)} e^{-iE_r t'} \int_{-\infty}^{t'} dt'' e^{-\frac{\Gamma}{2}(t'-t'')} \; {}_{t''}\langle r | D(t'') | g \rangle_{t''}$$

Time dependent ionization probability

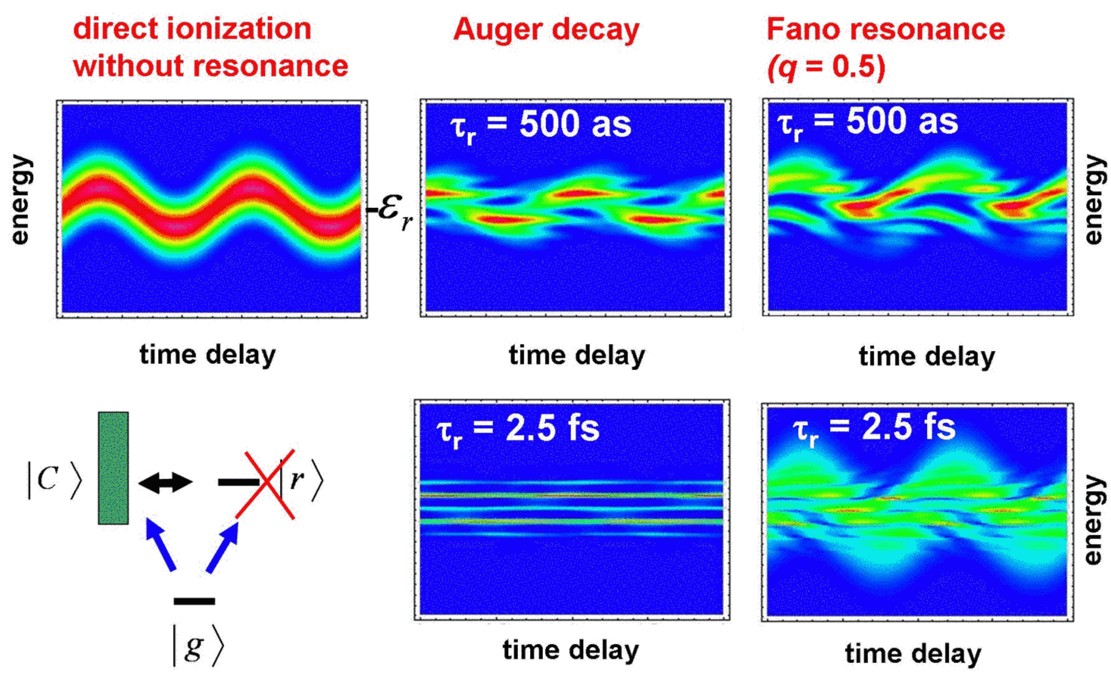
for window resonance ($q = 0$)



$q = 0$: window resonance



Streak images

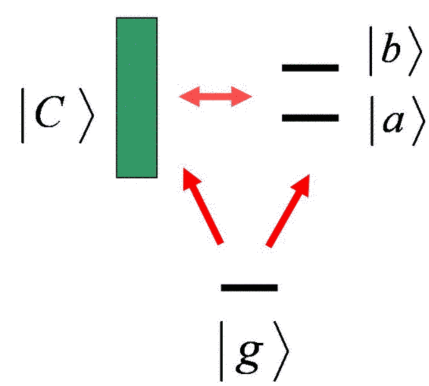


Wickenhauser et al., Phys. Rev. Lett. 94, 023002 (2005)

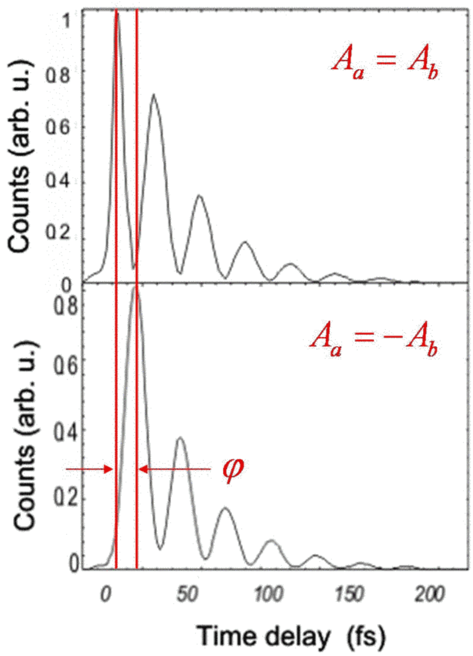
Novel information accessible ?

- Density matrix of coherently excited manifolds
- Decoherence and dephasing times

Extension: two (or more) resonances



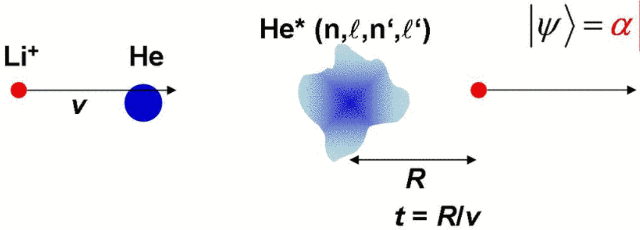
sideband intensity



➔ sensitivity to excitation amplitude

Wickenhauser et al., J. Mod. Opt. 53, 247 (2006)

"Attosecond" streaking of a coherently excited autoionizing resonance



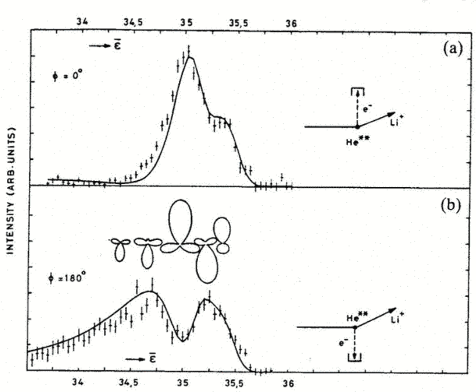
$$|\psi\rangle = \alpha |2s2p\,^1P\rangle + \beta |2p\,^2D\rangle$$

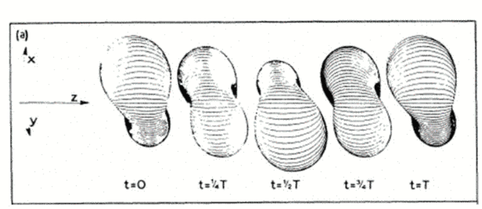
"PCI shift"

$$E(t) = E_0 + \frac{1}{R(t)}$$

quantum beats

τ=36 fs



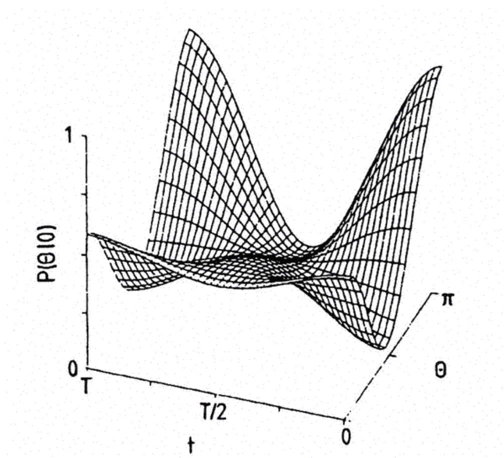


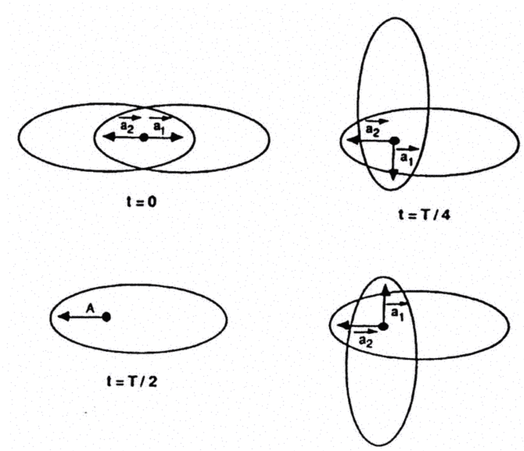
→ collective dipole oscillation

P. Van der Straten and R. Morgenstern, Comm. At. Mol. Phys. 19, 243 (1986)
J. Burgdörfer and R. Morgenstern, Phys. Rev. A 38, 5520 (1988)

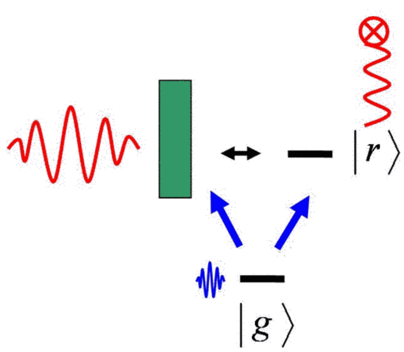
Quasi- classical vibrations:

$$|\psi\rangle = \alpha|2s^{2}\,{}^{1}S\rangle + \beta|2p^{2}\,{}^{1}S\rangle$$





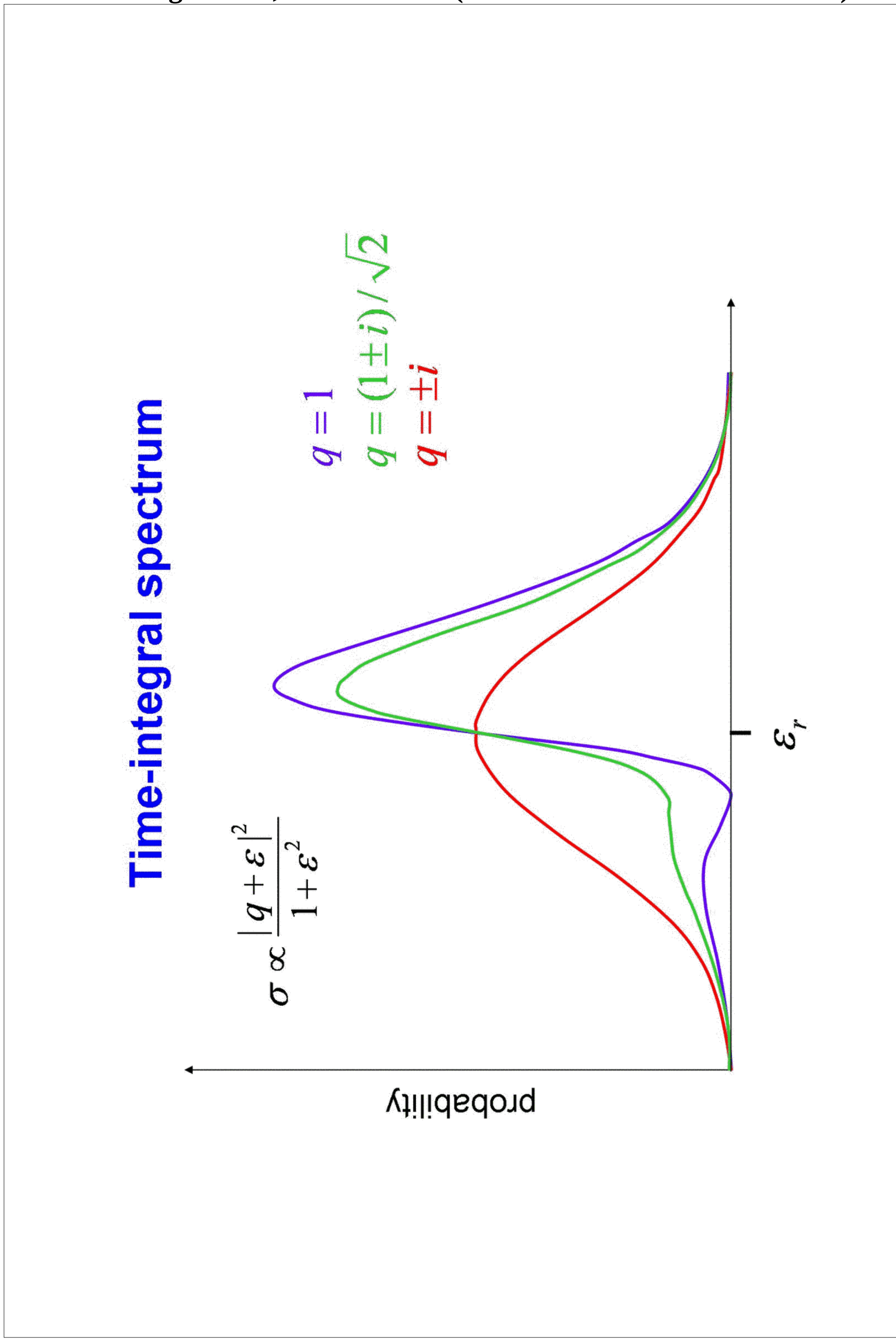
Can attosecond streak images measure complex *q* ?

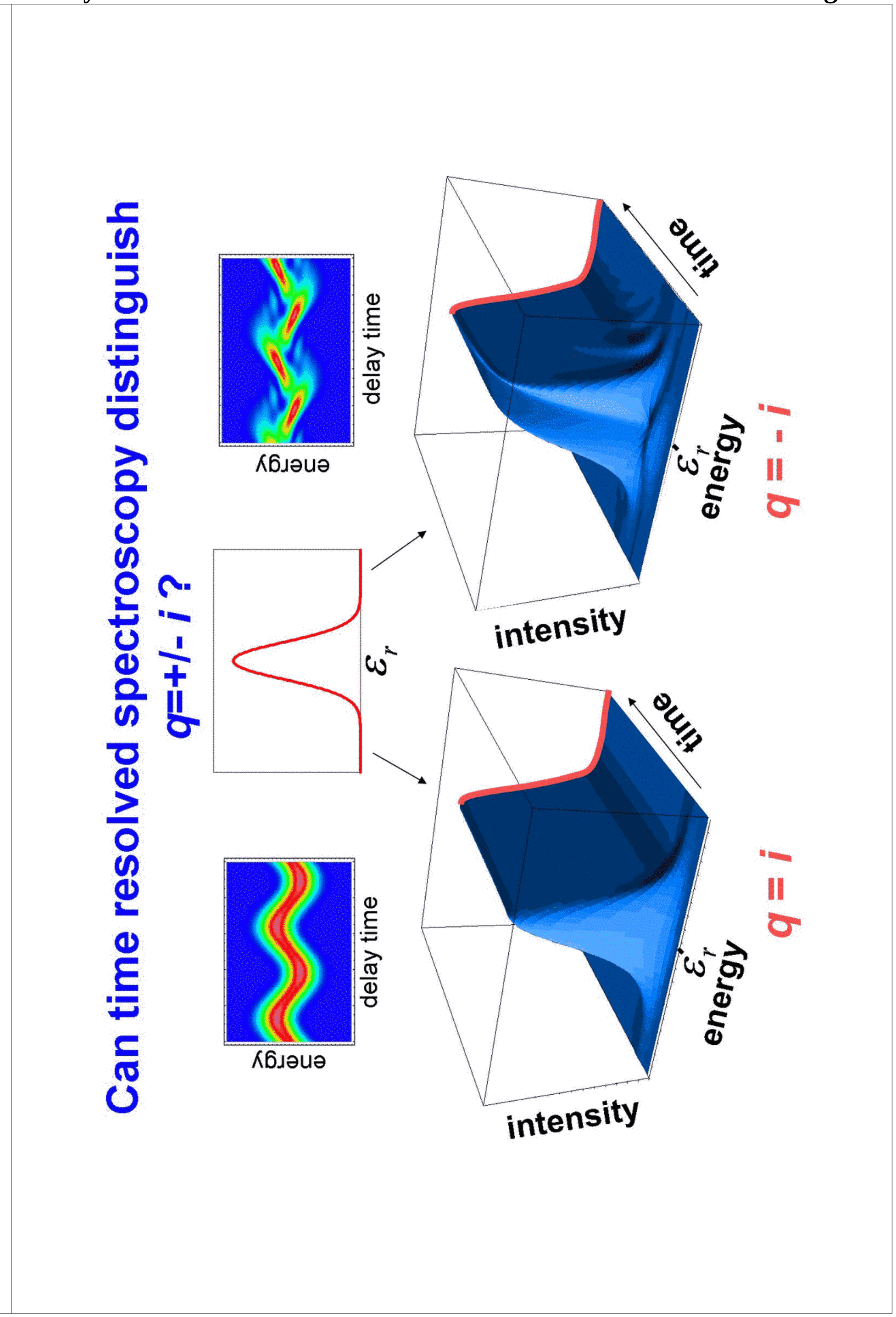


System with broken time reversal symmetry

- Resonances in magnetic field
- Decoherence in open quantum systems

⇒ complex *q*


Time-integral spectrum
$\sigma \propto \frac{|q+\varepsilon|^2}{1+\varepsilon^2}$
$q = 1$
$q = (1 \pm i)/\sqrt{2}$
$q = \pm i$
probability
ε_r


Can time resolved spectroscopy distinguish
q=+/- i ?
energy
delay time
ε_r
intensity
time
energy
ε_r
q = i
q = - i

Summary and outlook

- Attosecond dynamics in atoms can be probed by ultrashort lasers
- Interferences between attosecond electron wavepackets
- Following time evolution of Fano resonance in real time possible
- Extracting novel information
- Attosecond XUV-XUV interaction probe correlation dynamics
- Application to atomic systems
  - super Coster Kronig resonance
  - helium
- Quantum dots
- Decoherence in open systems