Collective excitations in the strong-field regime

A closer look at the HHG spectrum of xenon

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KITP Workshop
Santa Barbara, 10th September 2014

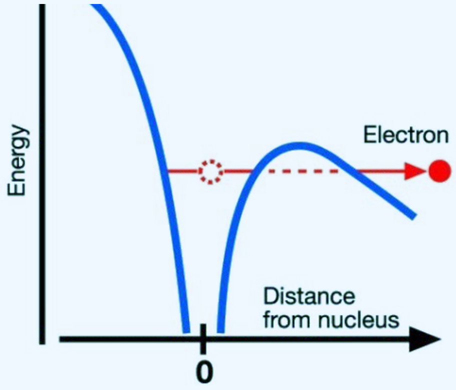
CFEL Theory Group

Lead by Robin Santra

Ultrafast Electron Motion

Using a Single-Electron Picture

Single-Active Electron (SAE) Approach



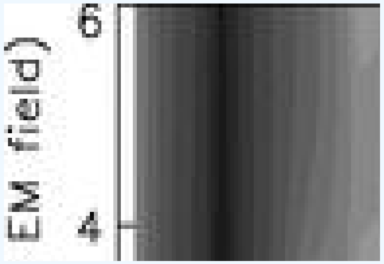
$$\hat{H} = \frac{\hat{p}^2}{2} + V(\hat{r}) + E(t)\,\hat{z}$$

- V(r) is local (and spherically symmetric)
- V(r) contains information about N-1 electrons
- Only a single orbital can be considered

- Strong-field ionization

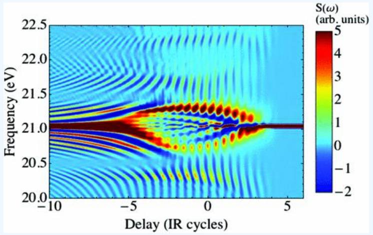
Nature Physics **6**, 428 (2010)

- HHG



PRA **79**, 053827 (2009)

- ATAS

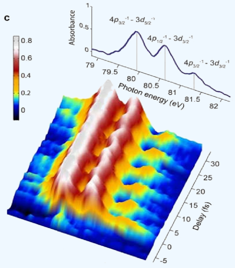


PRA **88**, 043416 (2013)

Multi-Electron Dynamics

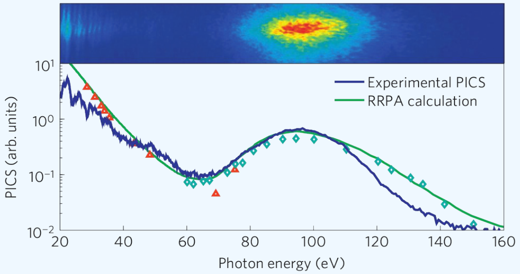
A Few Examples

- Multiorbital dynamics in HHG/tunnel ionization



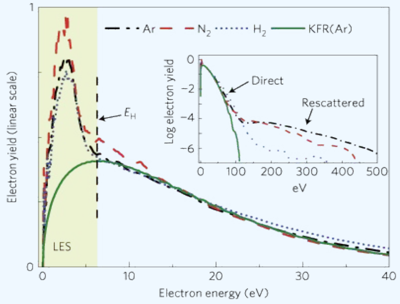
Nature **466**, 9212 (2010)

- Multielectron response in strong-fields



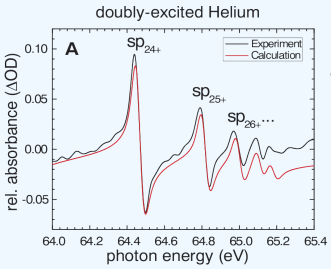
Nature Phys. **6**, 464 (2011)

- Low-energy ATI peak



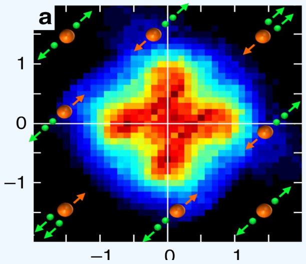
Nature Phys. **5**, 335 (2008)

- Manipulate electronic correlations



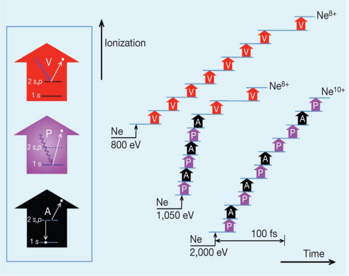
Science **340**, 716 (2013)

- Nonsequational double ionization



Nature Comm. **3**, 813 (2012)

- Multiple ionizations in the intense x-ray regime

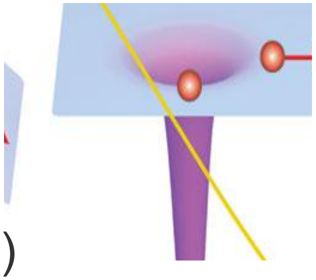


Nature **466**, 56 (2010)

Theoretical Approaches

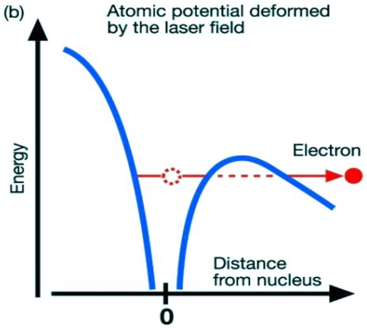
What Needs to be Described? Where are the Challenges?

> Spatial and kinetic aspects



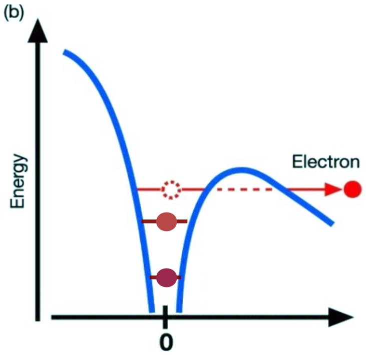
- Locally bound electrons (around $1a_0$)
- Large spatial motions ($100a_0$ and more)
- Small and large kinetic energies (1 eV – few 100eV)

> Light-matter interaction



- High photon energies, weak pulses (perturbative)
- Low photon energies, intense pulses (non-perturbative)

> Multielectron aspects



- Multiple orbital ionization
- Collective excitation

Theoretical Approaches

Many-Body Theories

Constructing the N-body wavefunction

- Ground state as reference state
- Construct excited states
    - Build from mean-field ground state
- Learn from time-independent theories
    - Converting into time-dependent theories

Configuration-Interaction

- Systematic inclusion of higher-order excitations

$$\left|\Phi_i^a\right\rangle,\ \left|\Phi_{ij}^{ab}\right\rangle,\ \left|\Phi_{ijk}^{abc}\right\rangle,\ \ldots$$

- Time-independent orbitals
    - Dynamics happens the expansion coefficients
- Many-electron systems
    - Atomic systems in 3D

TD-RASSCF Method

- Systematic inclusion of higher-order excitations

$$\left|\Phi_{i(t)}^{a(t)}\right\rangle,\ \left|\Phi_{i(t)j(t)}^{a(t)b(t)}\right\rangle,\ \ldots$$

- Time-dependent orbitals
    - Dynamics in the coefficients and in the orbitals
- Many-electrons systems
    - Atomic systems in 1D

Exact Wavefunction

- Everything is included

$$\left|\Psi(\mathbf{r}_1,\mathbf{r}_2,t)\right\rangle$$

- Spatial representation
    - Dynamics in the coefficients
- Two-electron systems
    - Helium and H_2
    - Lithium (no 1s electrons)

TDCIS
Wavefunction

Time-Dependent Configuration Interaction Singles (TDCIS)

- CI-Wavefunction

$$|\Psi(t)\rangle = \alpha_0(t)\,|\Phi_0\rangle + \sum_{ai} \alpha_i^a(t)\,|\Phi_i^a\rangle + \dots$$

Hatree-Fock GS

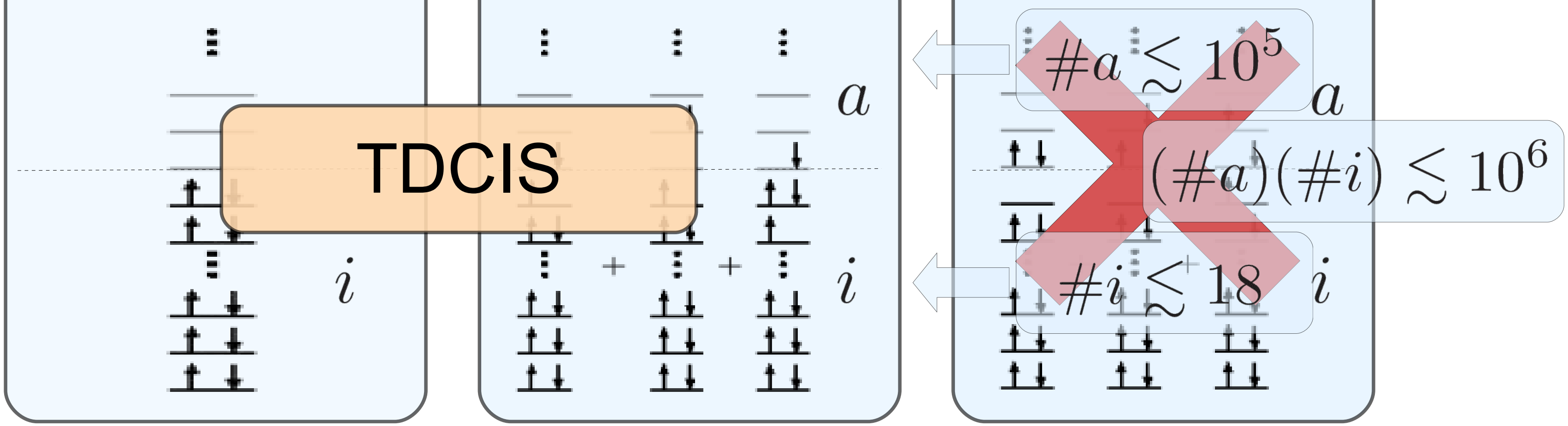
$$|\Phi_0\rangle = \prod_i \hat{c}_i^\dagger\,|\mathrm{vac}\rangle$$

Singles

$$|\Phi_i^a\rangle = \hat{c}_a^\dagger \hat{c}_i\,|\Phi_0\rangle$$

Doubles

$$|\Phi_{ij}^{ab}\rangle = \hat{c}_b^\dagger \hat{c}_i\,|\Phi_i^a\rangle$$



Greenman, *et al.*, PRA **82**, 023406 (2010)

TDCIS
Hamiltonian

Time-Dependent Configuration Interaction Singles (TDCIS)

> Hamiltonian

- No simplification of the Hamiltonian
- Exact two-electron Coulomb interaction is considered

$$\hat{H}(t) = \sum_i \left(\frac{\hat{\mathbf{p}}_i^2}{2} - \frac{Z}{\hat{\mathbf{r}}_\mathbf{i}} - \mathbf{E}(t) \cdot \hat{\mathbf{r}}_i \right) + \frac{1}{2} \sum_{i \neq j} \frac{1}{|\hat{\mathbf{r}}_\mathbf{i} - \hat{\mathbf{r}}_\mathbf{j}|}$$

$$= \underbrace{\hat{T} + \hat{V}_{\mathrm{MF}}}_{\hat{H}_0} + \underbrace{\frac{1}{|\hat{\mathbf{r}}_{12}|} - \hat{V}_{\mathrm{MF}}}_{\hat{H}_1} \underbrace{- \mathbf{E}(t) \cdot \hat{\mathbf{r}}}_{\hat{H}_{\mathrm{int}}(t)}$$

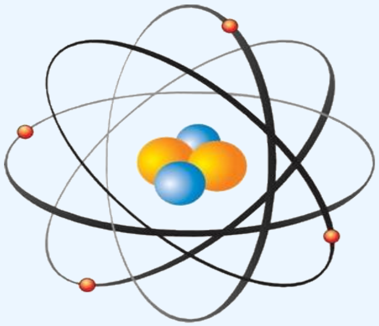
$$\hat{H}(t) = \hat{H}_0 + \hat{H}_1 + \hat{H}_{\mathrm{int}}(t)$$

TDCIS

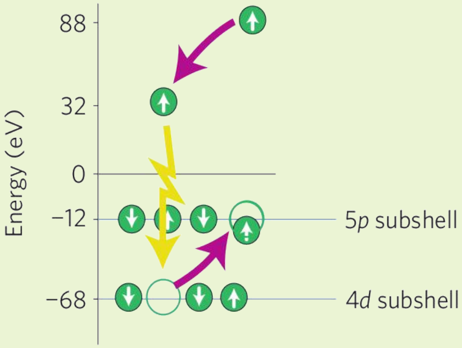
Hamiltonian

$$\hat{H}(t) = \hat{H}_0 + \hat{H}_1 + \hat{H}_{\mathrm{int}}(t)$$

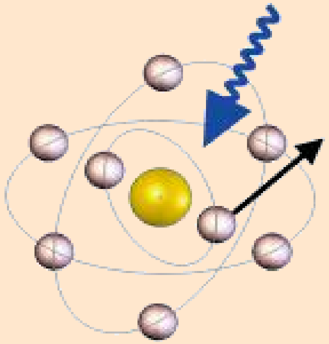
Fock Operator



Electron-Ion interaction



Laser Interaction



TDCIS

Residual Coulomb Interaction

> Residual Coulomb Interaction

- Captures everything beyond the HF mean-field level
- CIS: reduces to an electron-ion interaction

$$\left\langle \Phi_i^a \right| H_1 \left| \Phi_j^b \right\rangle = 2v_{ajib} - v_{ajbi}$$

Intrachannel interaction

- Ionic state does not change

$$\left\langle \Phi_i^a \right| H_1 \left| \Phi_j^b \right\rangle \qquad i = j$$

Interchannel interaction

- Ionic state changes due to presence of the electron

$$\left\langle \Phi_i^a \right| H_1 \left| \Phi_j^b \right\rangle \qquad i \neq j$$

TDCIS

Equation of Motion

$$\boxed{\hat{H}(t) = \hat{H}_0 + \hat{H}_1 + \hat{H}_{\text{int}}(t)}$$

$$+$$

$$\boxed{|\Psi_{\text{CIS}}(t)\rangle = \alpha_0(t)\,|\Phi_0\rangle + \textstyle\sum_{ai} \alpha_i^a(t)\,|\Phi_i^a\rangle}$$

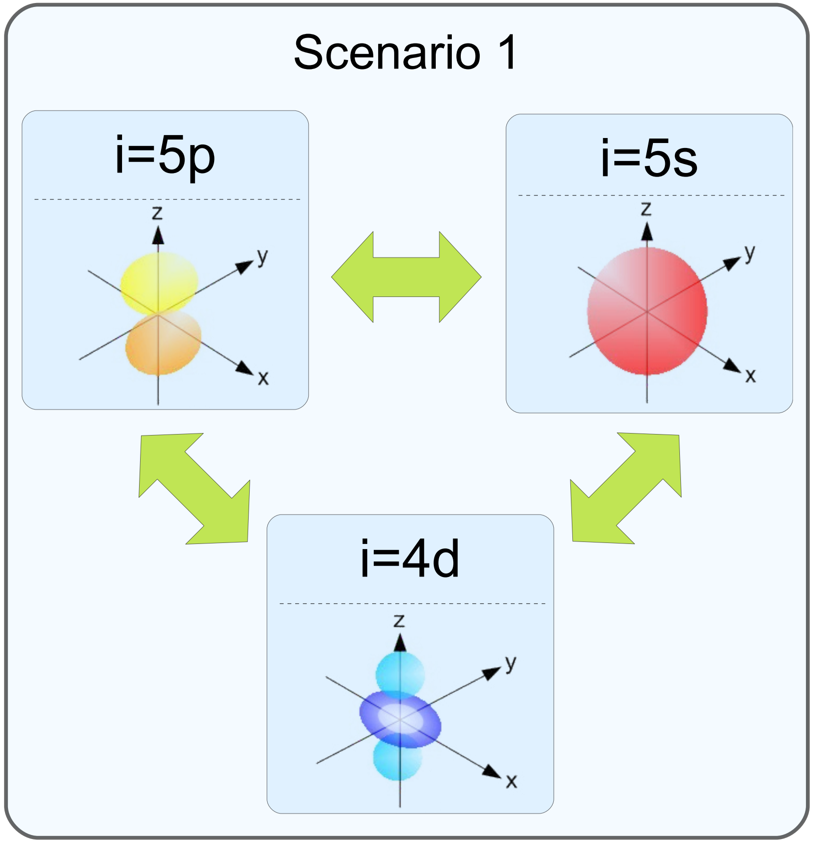
$$=$$

$$\boxed{i\partial_t\,|\Psi_{\text{CIS}}(t)\rangle = \hat{H}(t)\,|\Psi_{\text{CIS}}(t)\rangle}$$

TDCIS

Multiple Orbital Physics

$$|\Psi_{\mathrm{CIS}}(t)\rangle = \alpha_0(t)\,|\Phi_0\rangle + \sum_{a,i} \alpha_i^a(t)\,|\Phi_i^a\rangle$$

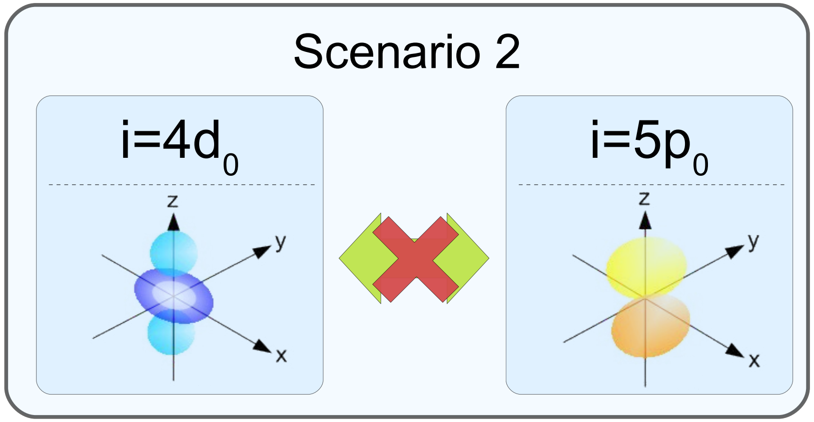


Scenario 1

$i=5p$

$i=5s$

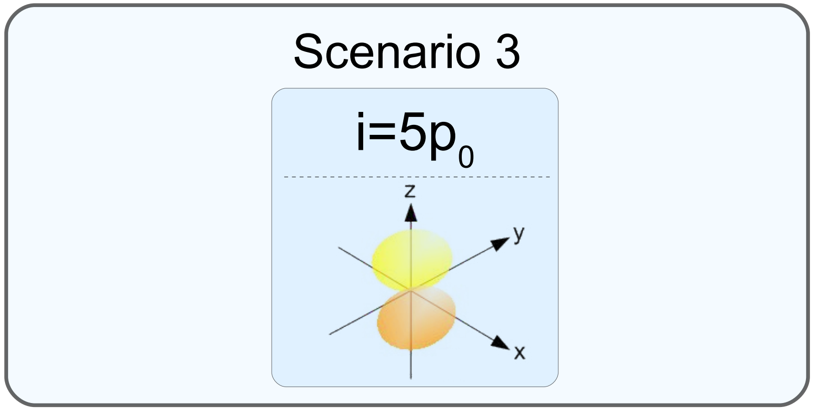
$i=4d$



Scenario 2

$i=4d_0$

$i=5p_0$



Scenario 3

$i=5p_0$

TDCIS

Wide Spectrum of Applications

Theory / Education

- Technical description
  - Greenman *et al.*, PRA **82**, 023406 (2010)
- Review
  - Pabst, EPJ ST **221**, 1 (2013)
- Educational description
  - Krebs *et al.*, AJP **82**, 113 (2014)

Strong-Field Ionization

- Adiabaticity of tunnel ionization
  - Kamaratskou *et al.*, PRA **87**, 043422 (2013)
- Sub-cycle ionization dynamics
  - Wirth *et al.*, Science **334**, 195 (2011)
  - Pabst *et al.*, PRA **86**, 063411 (2012)

HHG

- Multichannel effects in argon
  - Pabst *et al.*, PRA **85**, 0234111 (2012)
- Collective excitations in xenon
  - Pabst *et al.*, PRL **111**, 0233005 (2013)
- Spin-orbit effects in krypton
  - Pabst *et al.*, JPB **47**, 124026 (2014)

X-ray/XUV-Triggered Processes

- Decoherence in photoionization
  - Pabst *et al.*, PRL **106**, 0053003 (2011)
- Nonlinear x-ray/xuv ionization
  - Sytcheva *et al.*, PRA **85**, 023414 (2012)
  - Kamaratskou *et al.*, PRA **89**, 033415 (2014)
- Exploiting Fano resonances
  - Heinrich-J. *et al.*, PRA **89**, 043415 (2014)

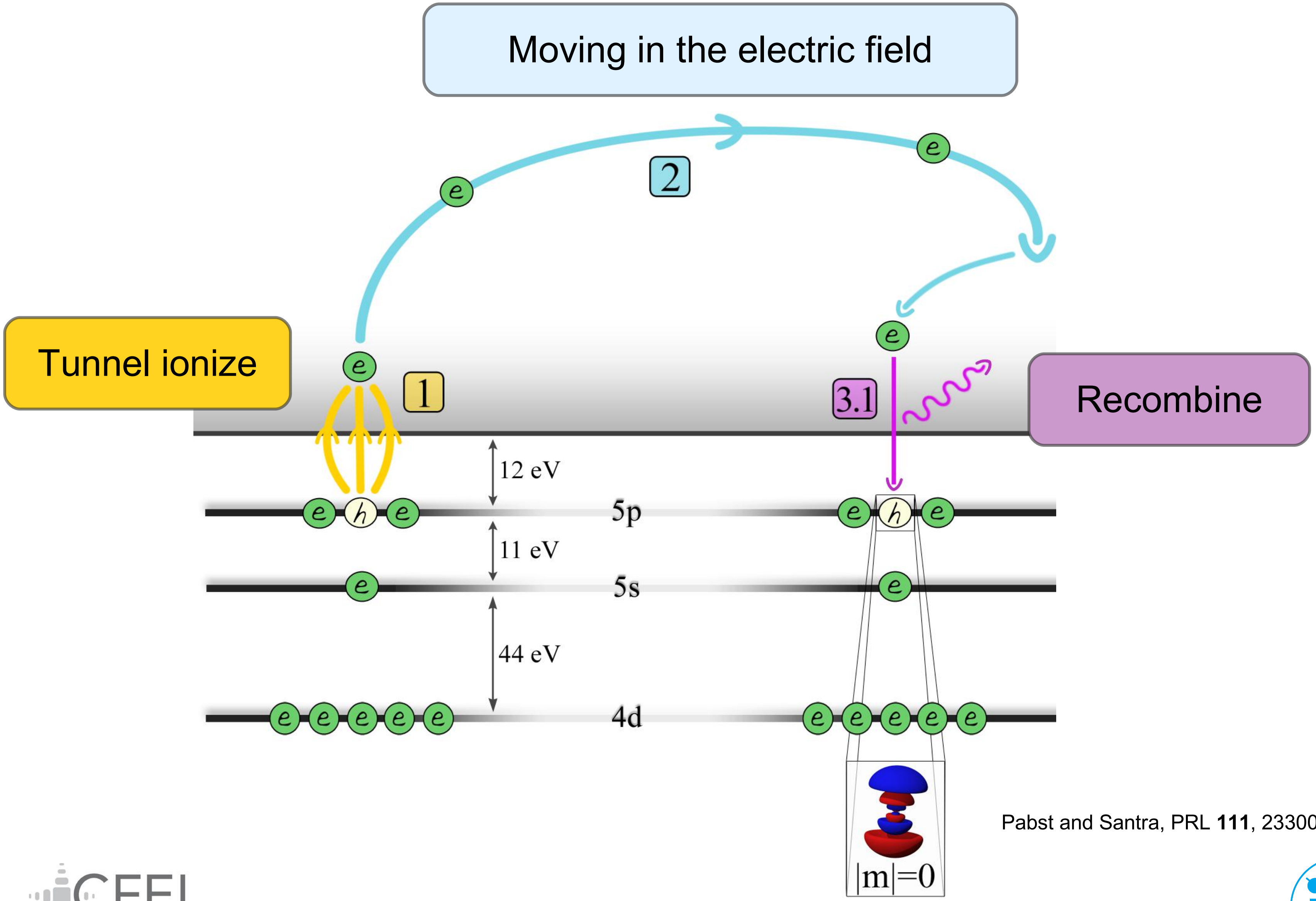
Many-body Physics on the Attosecond Scale

Collective Excitations
in the HHG spectrum of Xe

Pabst and Santra, PRL **111**, 233005 (2013)

High-Harmonic Generation

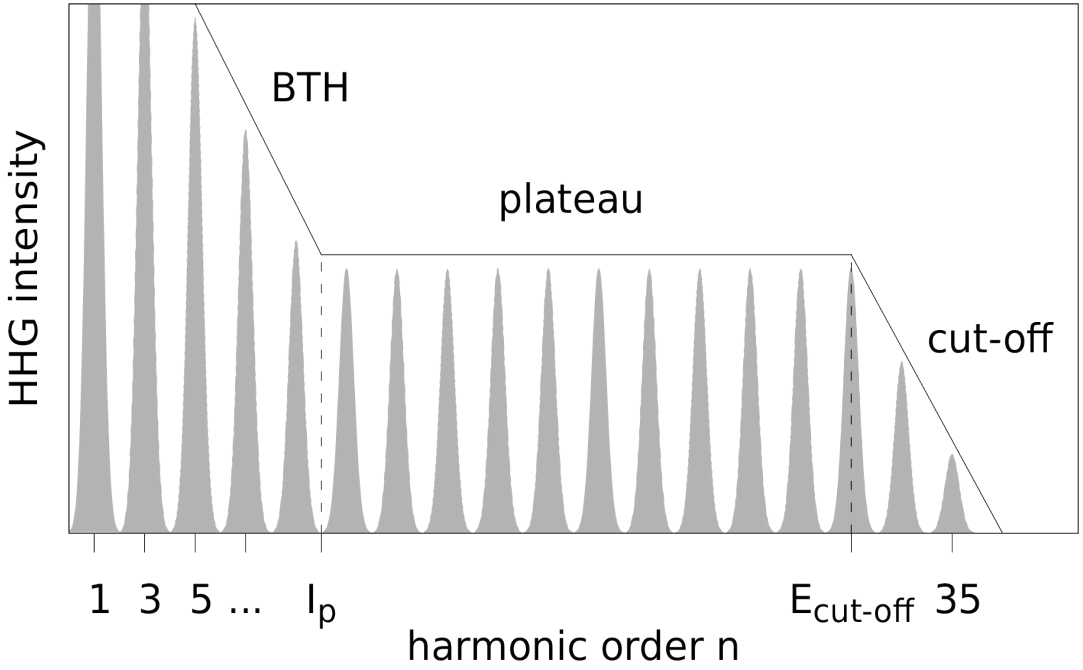
Basic Picture



Pabst and Santra, PRL **111**, 233005 (2013)

High Harmonic Generation

The Spectrum



Pabst, EPJ ST **221**, 1 (2013)

Collective Excitations in the Strong-Field Regime

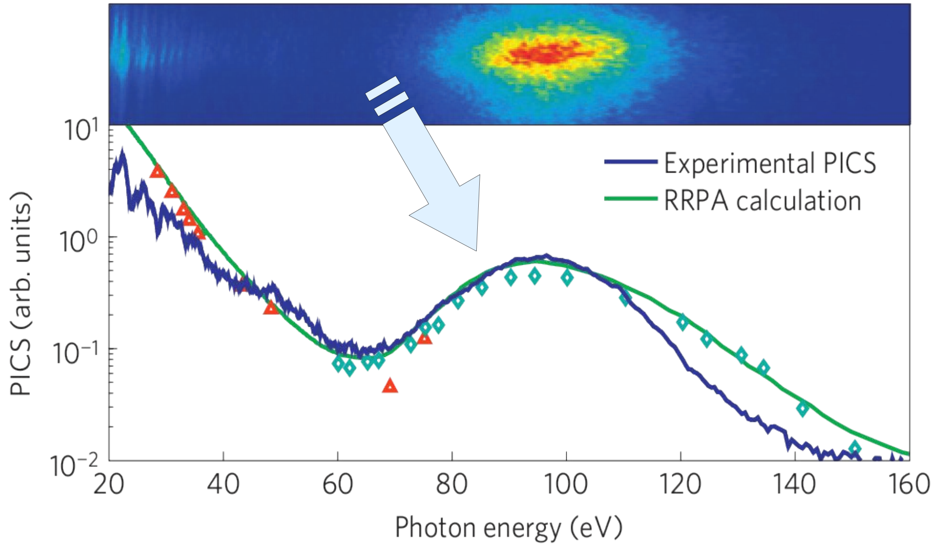
First Observation and Various Interpretations

What is the origin?

- Single-particle phenomenon
  - Is it a Cooper minimum?
    PRL **110**, 063002 (2013)
- Propagation effects
  - Phase matching (Gouy phase) ?
    New J. Phys. **13**, 073003 (2011)
- Many-body mechanism
  - Collective excitation ?
    Shiner et al., Nat. Phys. 6, 464 (2011)

- Why is so hard to do first-principle calculations?
  - Long driving wavelength: >1500 nm
  - Large ponderomotiv energies: >120 eV
  - Large spatial motion up to 150 a_0
  - Many bound electrons are involved

Giant enhancement in the HHG spectrum

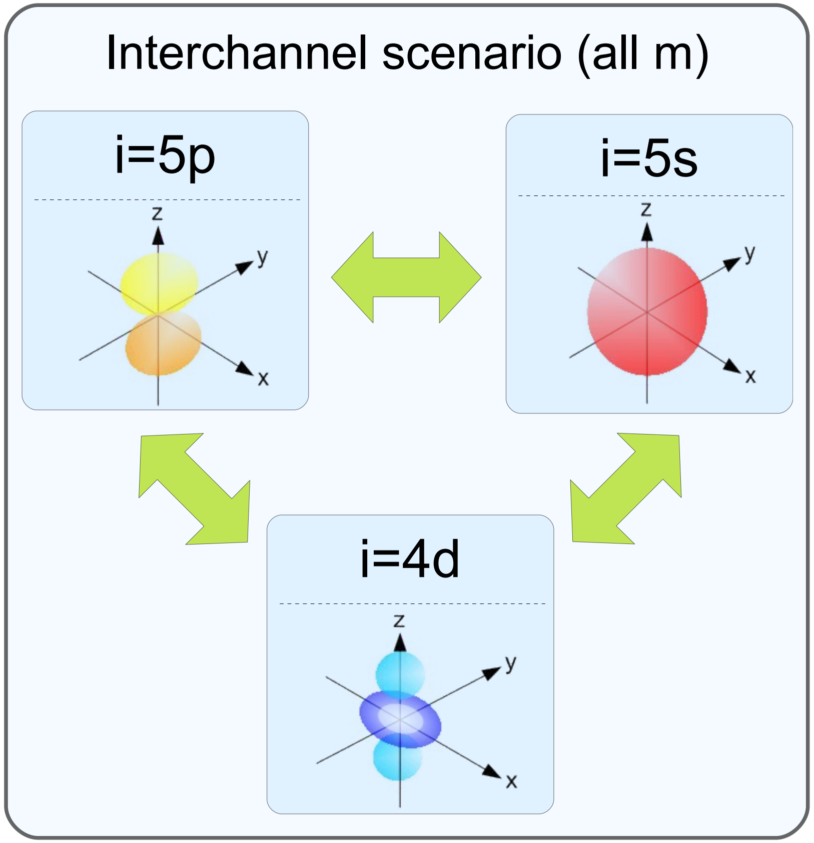


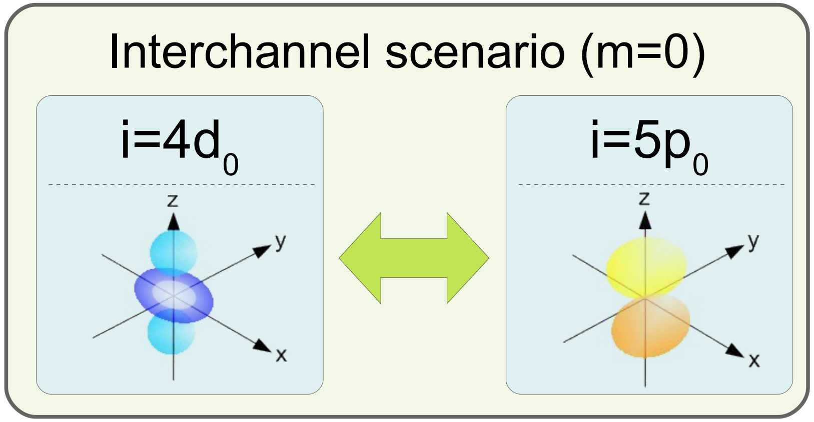
Nature Phys. **6**, 464 (2011)

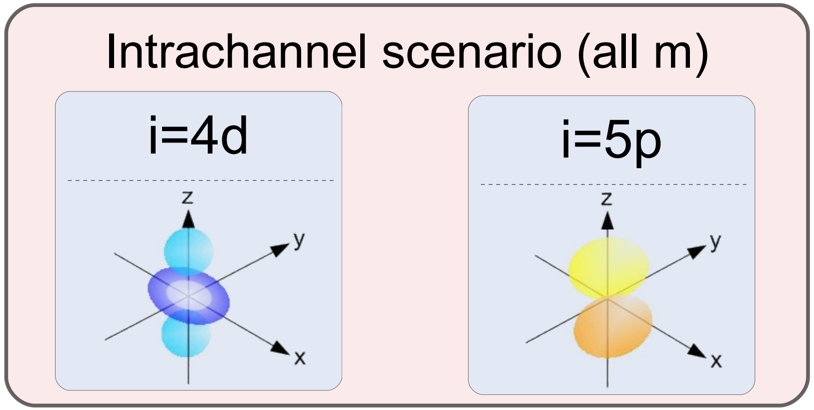
Collective Excitations in the Strong-Field Regime

Testing Multiorbital Nature

$$|\Psi_{\mathrm{CIS}}(t)\rangle = \alpha_0(t)\,|\Phi_0\rangle + \sum_{a,i} \alpha_i^a(t)\,|\Phi_i^a\rangle$$

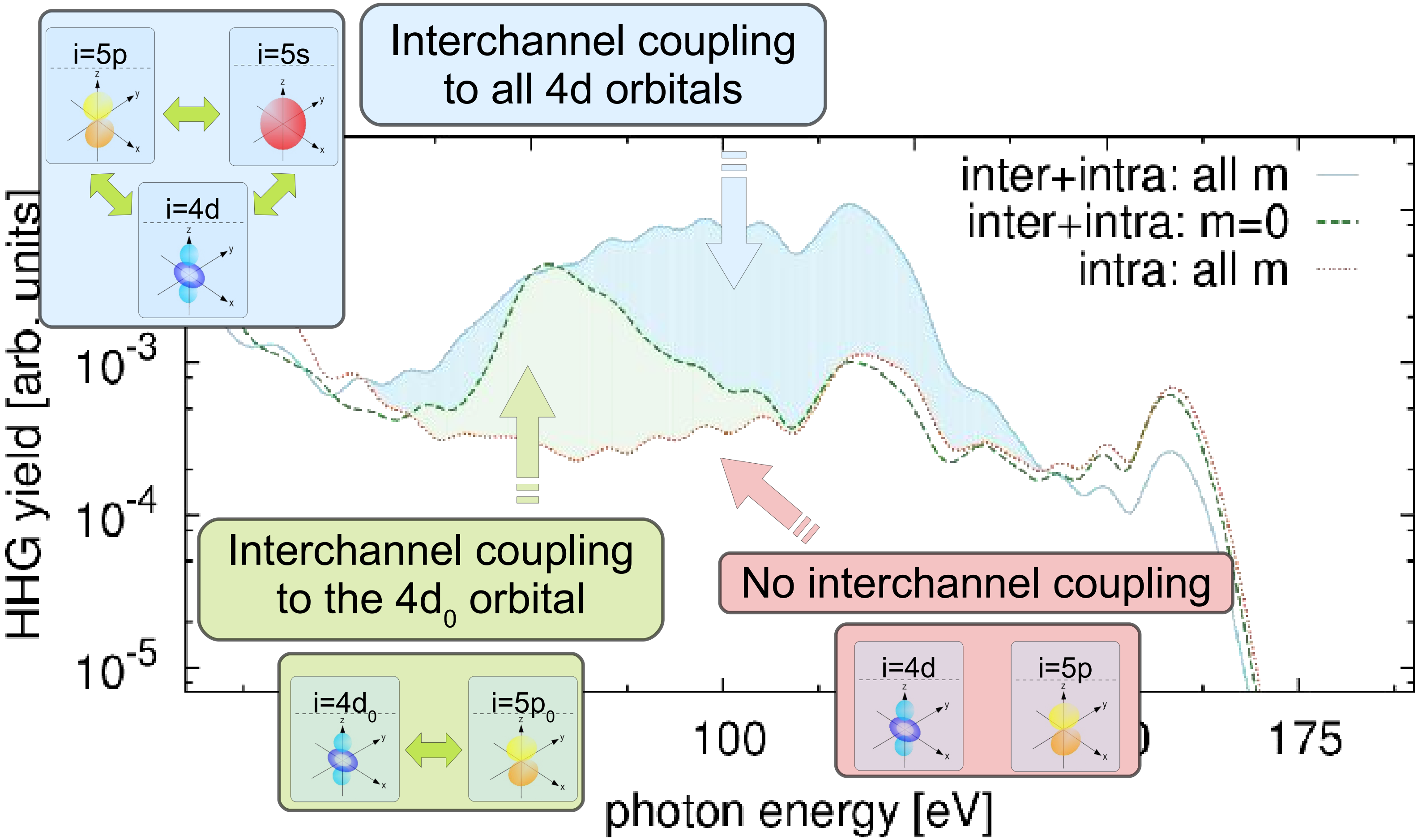

Interchannel scenario (all m)


Interchannel scenario (m=0)


Intrachannel scenario (all m)

Collective Excitations in the Strong-Field Regime

TDCIS Results


Interchannel coupling to all 4d orbitals

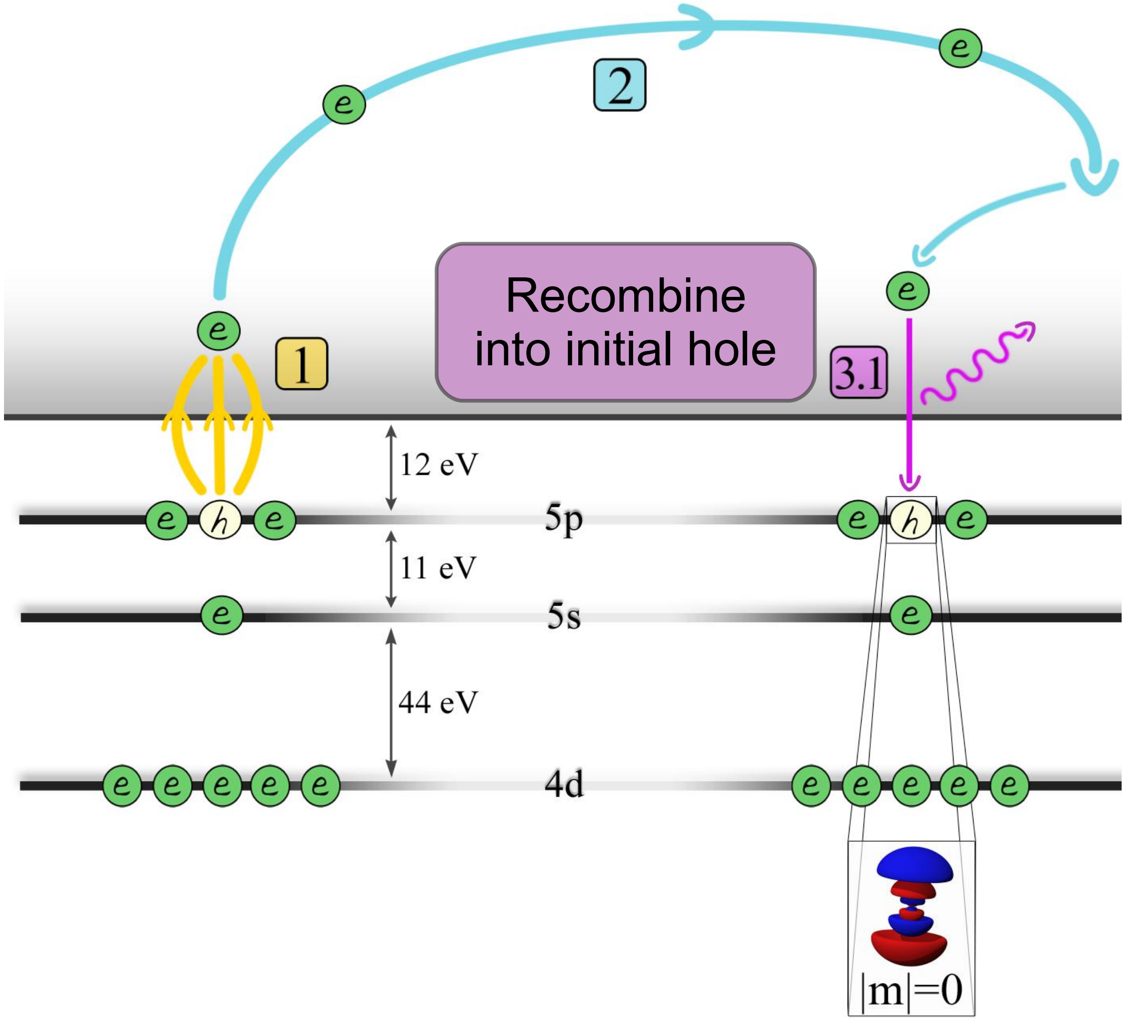
Interchannel coupling to the $4d_0$ orbital

No interchannel coupling

Pabst and Santra, PRL **111**, 233005 (2013)

High-Harmonic Generation

Complete the Picture – Include Collective Excitations



Pabst and Santra, PRL **111**, 233005 (2013)

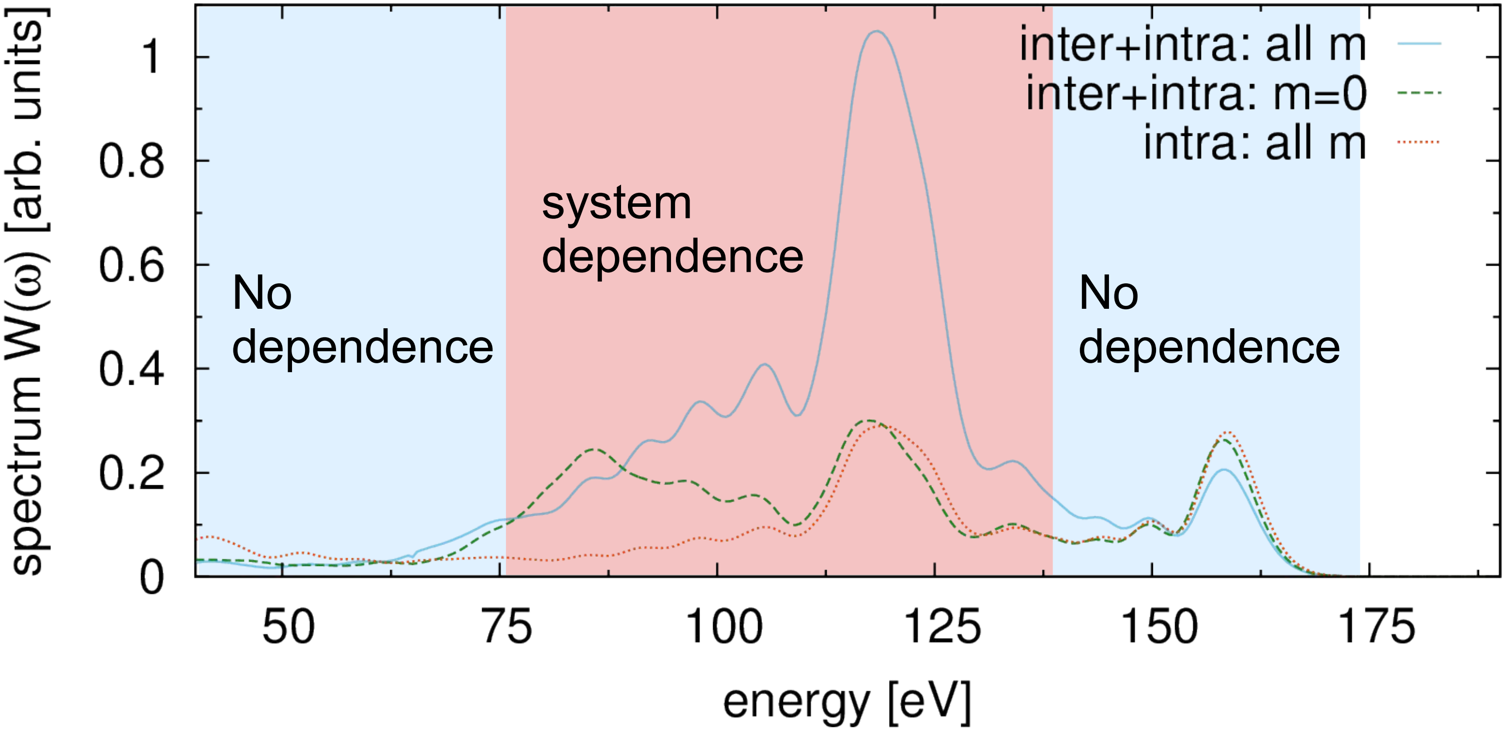
Collective Excitations in the Strong-Field Regime

Spectrum of the Returning Electron

Is the electron spectrum system-independent ?

⇒

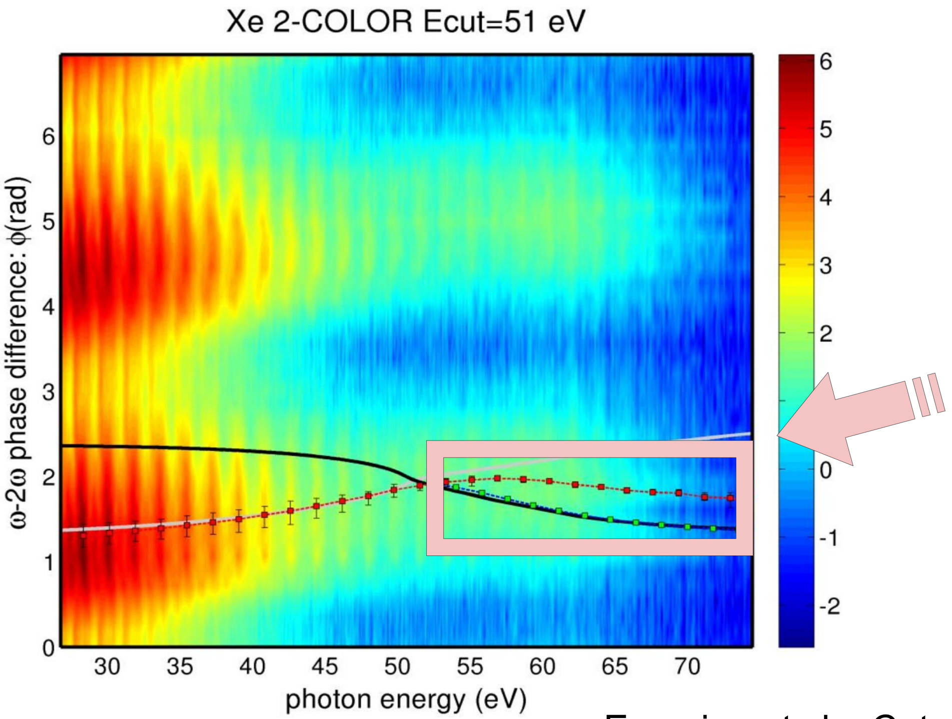
$$W(\omega) = \frac{S_{\mathrm{HHG}}(\omega)}{\sigma_{5p_0}(\omega)}$$



Time Delay in Recombination?

> 2color HHG spectrum

- Driving wavelength is 1400nm
- Second color: 700nm (perpendicular polarized)



- Discrepancy to the Lewenstein model
- Indication that recombination is delayed

Experiments by Caterina Vozzi