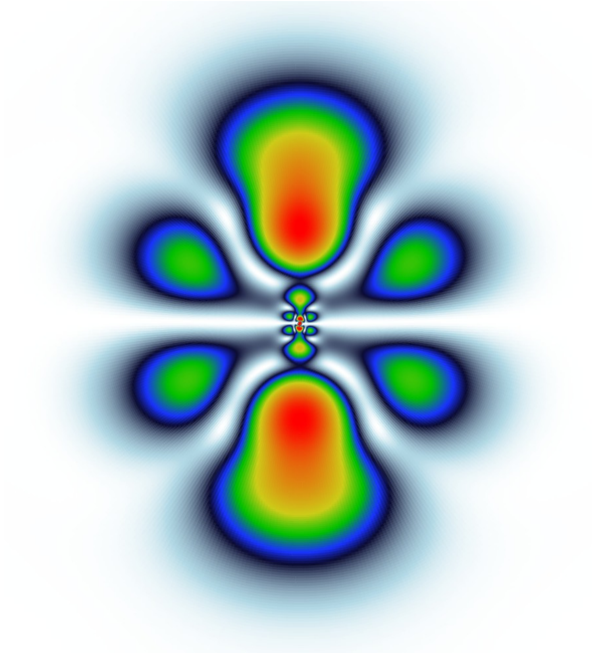
Controlling ionic hole populations and coherences with ultrashort pulses



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May 28th, 2015

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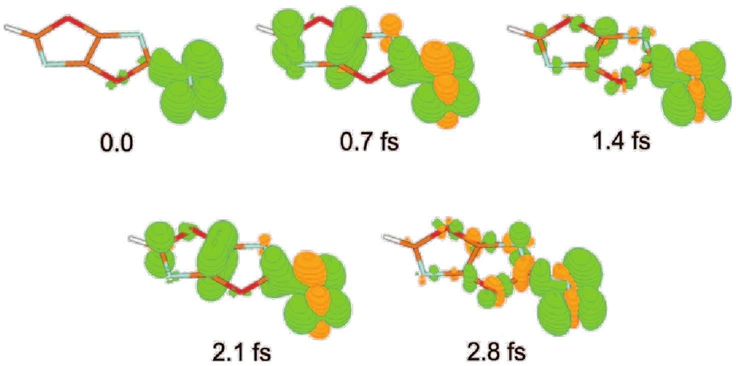
Alexander von Humboldt Stiftung/Foundation

ITAMP Institute for Theoretical Atomic Molecular and Optical Physics

Charge Transfer Dynamics

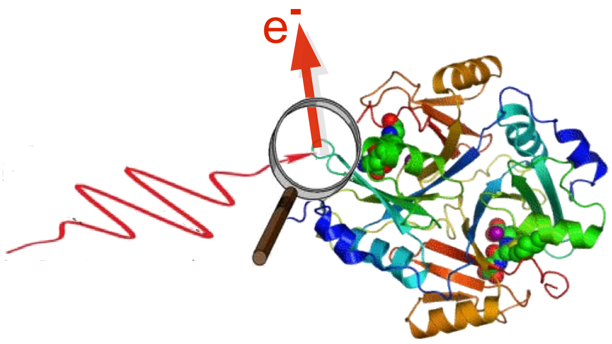
Hole Creation via Photoionization

- Ultrafast hole migration



Lünnemann *et al.*, JCP **130**, 154305 (2009)

- Photoionization prepares the hole
- Understanding the preparation process is crucial



Is the hole coherent ?

Control hole distribution ?

Outline

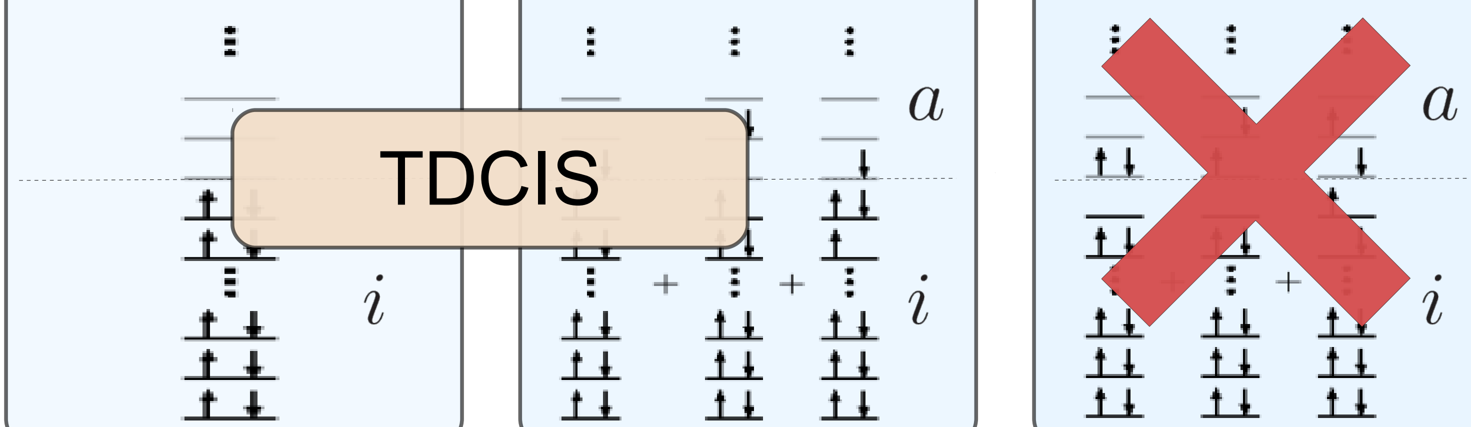
- TDCIS
  - Multi-orbital processes
  - Interchannel coupling (e.g. Fano)
- How to create coherent hole motion?
  - 1-photon ionization vs. tunnel ionization
  - Creating attosecond motion with long IR pulses?
- Controlling hole populations using Fano resonances?
  - Detuning dependent hole alignment
  - Stable 2s holes (2-photon process > 1-photon process)

Time-Dependent Configuration Interaction Singles (TDCIS)

- CI-Wavefunction

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

Hatree-Fock GS	Singles	Doubles
$\|\Phi_0\rangle = \prod_i \hat{c}_i^\dagger \|\mathrm{vac}\rangle$	$\|\Phi_i^a\rangle = \hat{c}_a^\dagger \hat{c}_i \|\Phi_0\rangle$	$\|\Phi_{ij}^{ab}\rangle = \hat{c}_b^\dagger \hat{c}_i \|\Phi_i^a\rangle$



Time-Dependent Configuration Interaction Singles (TDCIS)

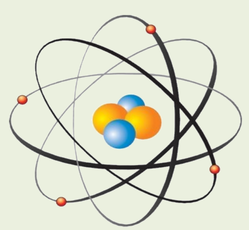
Hamiltonian

- Exact Hamiltonian (no approximations)

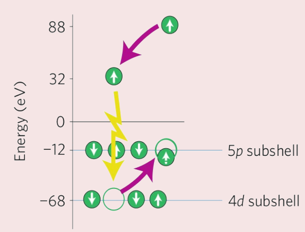
$$\hat{H}(t) = \sum_i \left(\frac{\hat{\mathbf{p}}_i^2}{2} - \frac{Z}{\hat{\mathbf{r}}_\mathbf{i}} - \mathbf{E}(t) \cdot \hat{\mathbf{r}}_i \right) + \frac{1}{2} \sum_{i \neq j} \frac{1}{|\hat{\mathbf{r}}_\mathbf{i} - \hat{\mathbf{r}}_\mathbf{j}|}$$

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

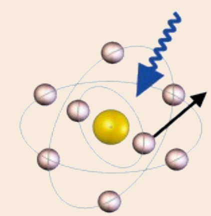
Mean-Field System



Residual Coulomb Interaction



Laser-Matter Interaction



TDCIS

Time-Dependent Schrödinger Equation

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

$$+$$

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

$$=$$

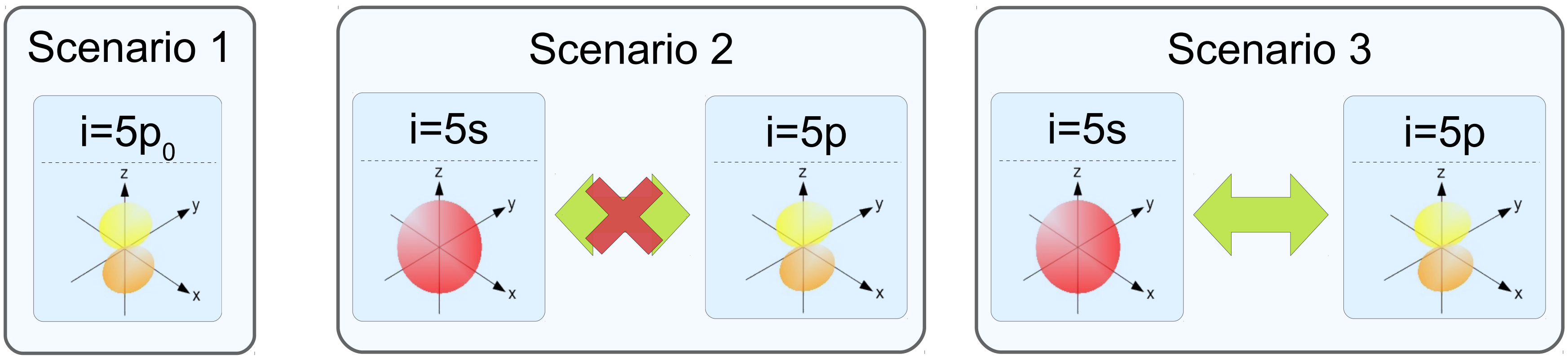
$$i\partial_t\,|\Psi_{\text{CIS}}(t)\rangle = \hat{H}(t)\,|\Psi_{\text{CIS}}(t)\rangle$$

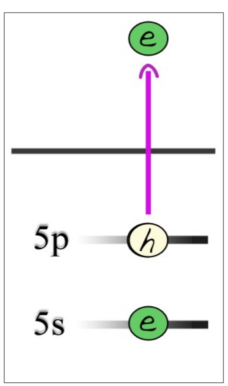
Greenman *et al.,* PRA **82**, 023406 (2010)

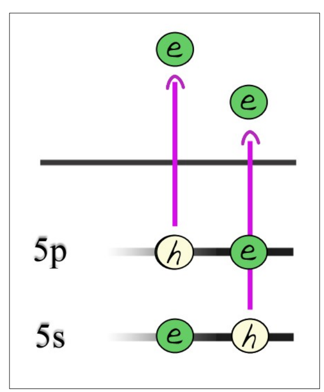
TDCIS

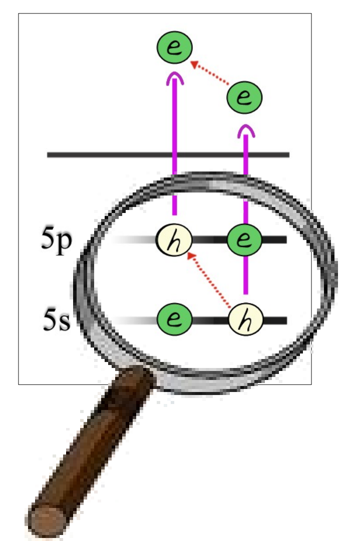
Multiple Orbital Physics

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





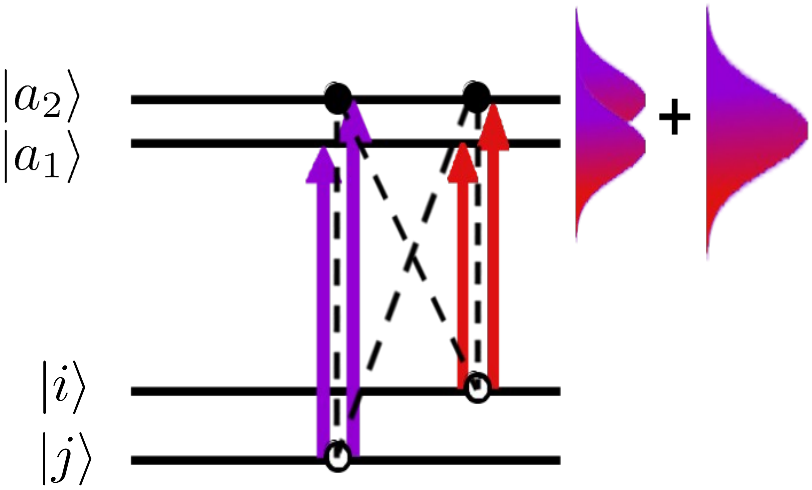




Ionic subsystem

Degree of Coherence – Ion Density Matrix

$$|\Psi(t)\rangle = \alpha_0(t)\,|\Phi_0\rangle + \sum_a \alpha_i^a(t)\,|\Phi_i^a\rangle + \sum_a \alpha_j^a(t)\,|\Phi_j^a\rangle$$



Subsystem: Ion

Ion Density Matrix

$$\hat{\rho}(t) = |\Psi(t)\rangle\langle\Psi(t)|$$

$$\hat{\rho}^{\mathrm{IDM}}(t) = \mathrm{Tr}_a\left[\hat{\rho}(t)\right]$$

Degree of Coherence

$$g_{ij}(t) = \frac{|\rho_{ij}^{\mathrm{IDM}}(t)|}{\sqrt{\rho_{ii}^{\mathrm{IDM}}(t)\rho_{jj}^{\mathrm{IDM}}(t)}} \leq 1$$

Outline

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- Interchannel coupling (e.g. Fano)

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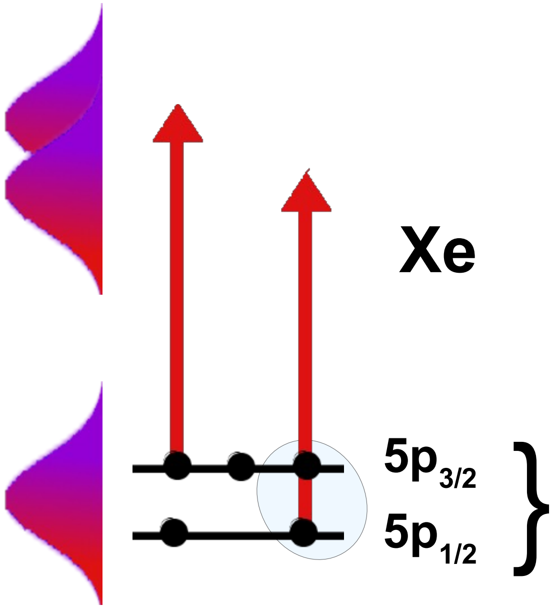
- Detuning dependent hole alignment
- Stable 2s holes (2-photon process > 1-photon process)

How to create coherent hole motion?

1-photon ionization



Target State:

$$|\text{hole}\rangle = c_1 \left|5p_{3/2}^{-1}\right\rangle + c_2 \left|5p_{1/2}^{-1}\right\rangle$$

$$\Delta E = \varepsilon_{5p_{1.5}} - \varepsilon_{5p_{0.5}} \approx 1.3\,\text{eV}$$

$$\Rightarrow \tau_{\text{FWHM}} \lesssim 1.4\,\text{fs}$$

attosecond pulses are required

Coherence window

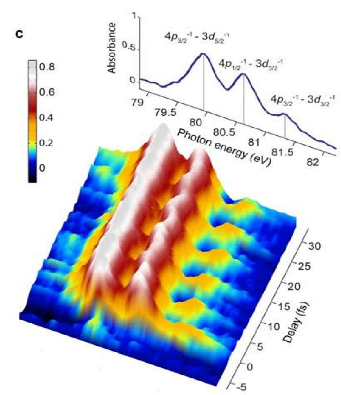
all ΔE for which a coherent superposition is possible

$$\Delta E \lesssim \delta\omega = \frac{4\,\ln(2)}{\tau_{\text{FWHM}}}$$

How to create coherent hole motion?
Tunnel ionization

- What is the coherence window for tunnel ionization?

- Previous experiments in krypton
  - 1-photon argument: $\Delta E=0.67$ eV → 2.7 fs or shorter
  - Get coherence via transient absorption
  - Pulse durations: 2 fs and 4 fs
  - Degree of coherences: 0.85 and 0.6



no clear contradiction to
1-photon coherence window

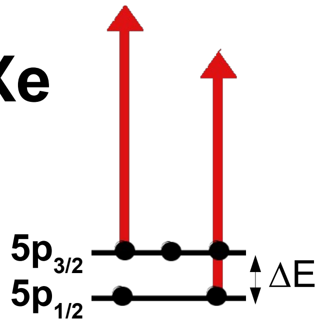
Goulielmakis, *et al.*, Nature **466**, 739 (2010)
Wirth, et al., Science **334**, 194 (2011)

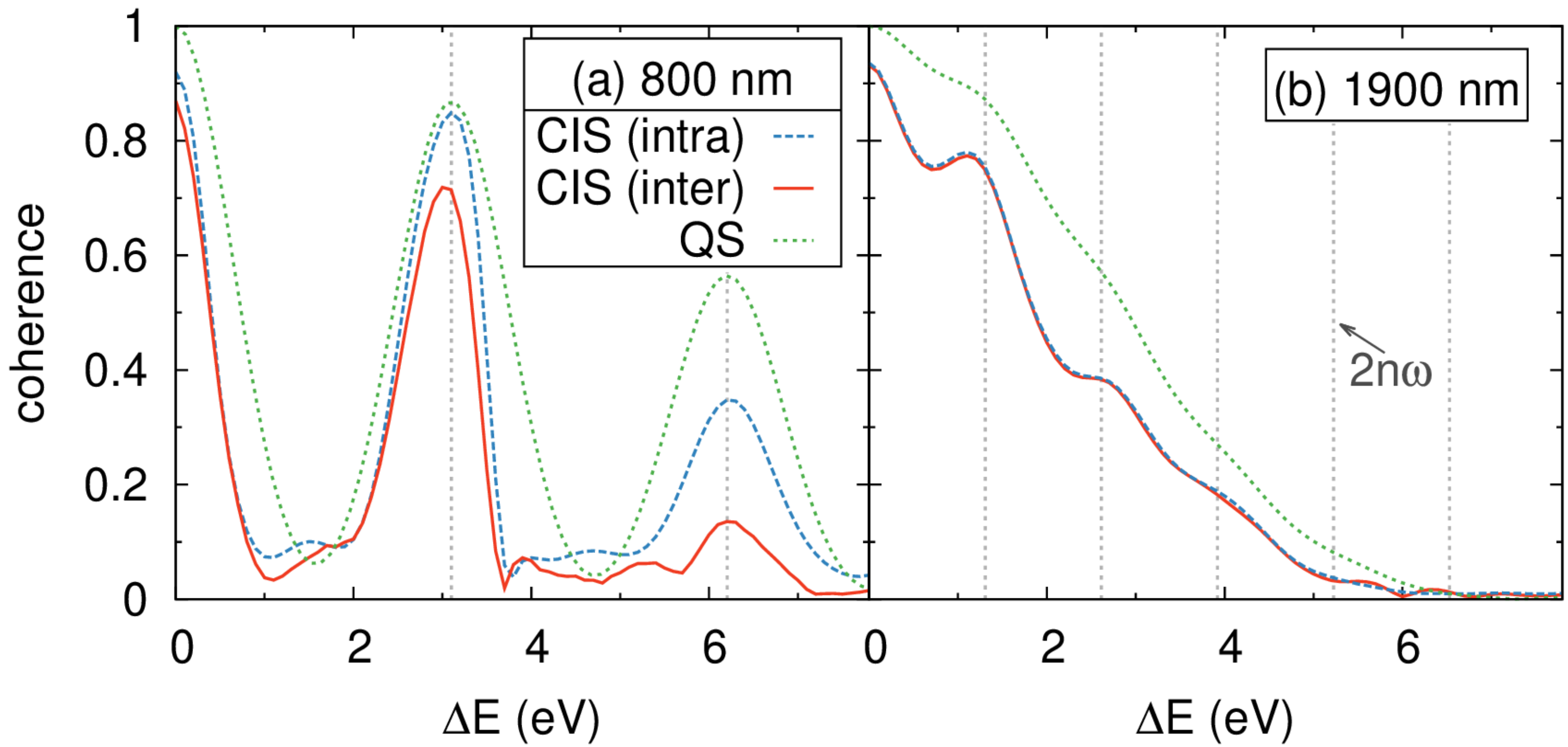
- Xenon:
  - Duration: T < 1.2 fs → no IR pulse could create $5p^{-1}_{1/2}$–$5p^{-1}_{3/2}$ superposition

How to create coherent hole motion?

Tunnel ionization

- Change ΔE
- Keep IR pulse fixed (T=6.3fs , 10^{14} W/cm^2)



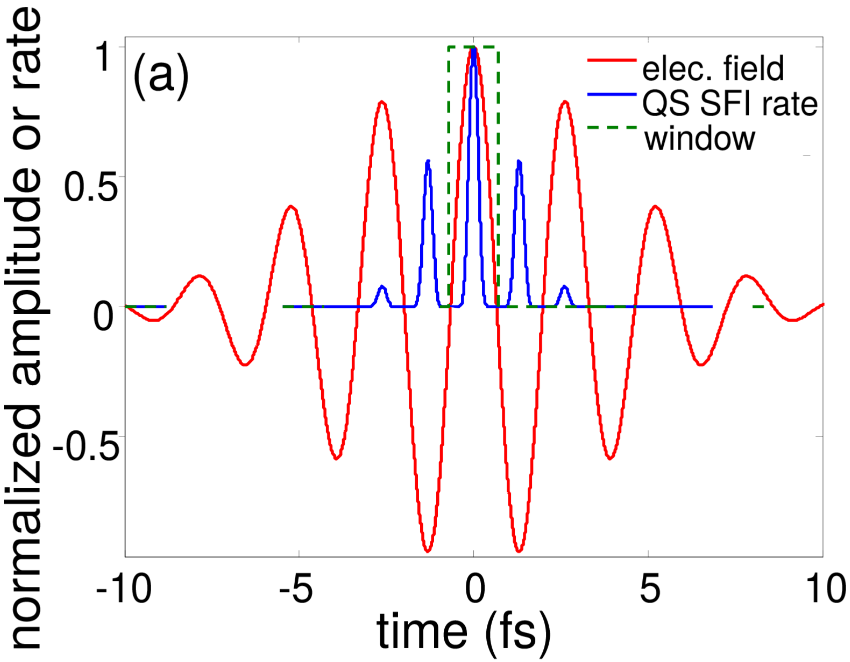


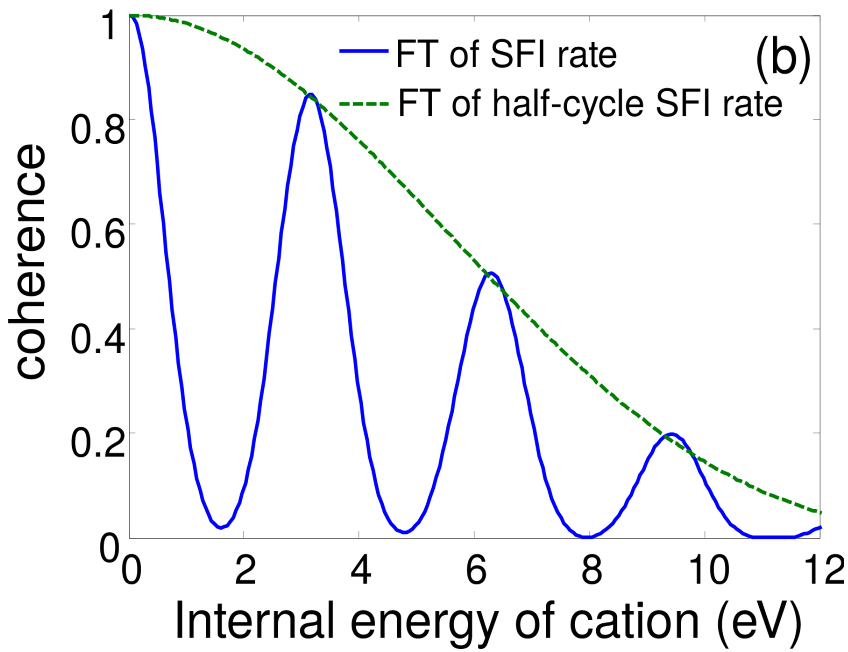
How to create coherent hole motion?

Tunnel ionization – Coherence window

$$\Gamma(t) := \Gamma\big(E(t)\big)$$

$$\Gamma(\omega) = \mathrm{FFT}\big[\Gamma(t)\big]$$





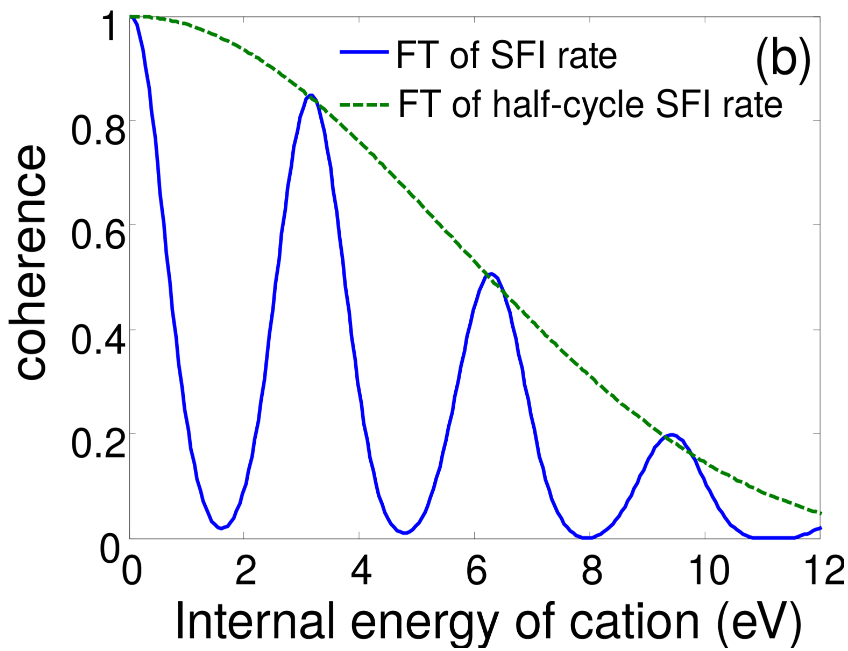
$$i\partial_t \hat{\rho}(t) = e^{i\hat{H}_0\, t}\hat{\Gamma}(t)e^{-i\hat{H}_0\, t}$$

$$\rho_{IJ}(\infty) = \Gamma_{IJ}(\Delta E_{JI})$$

$$g_{IJ} = \frac{|\Gamma_{IJ}(\Delta E_{JI})|}{\sqrt{\Gamma_{II}(0)\Gamma_{JJ}(0)}}$$

How to create coherent hole motion?

Coherence window



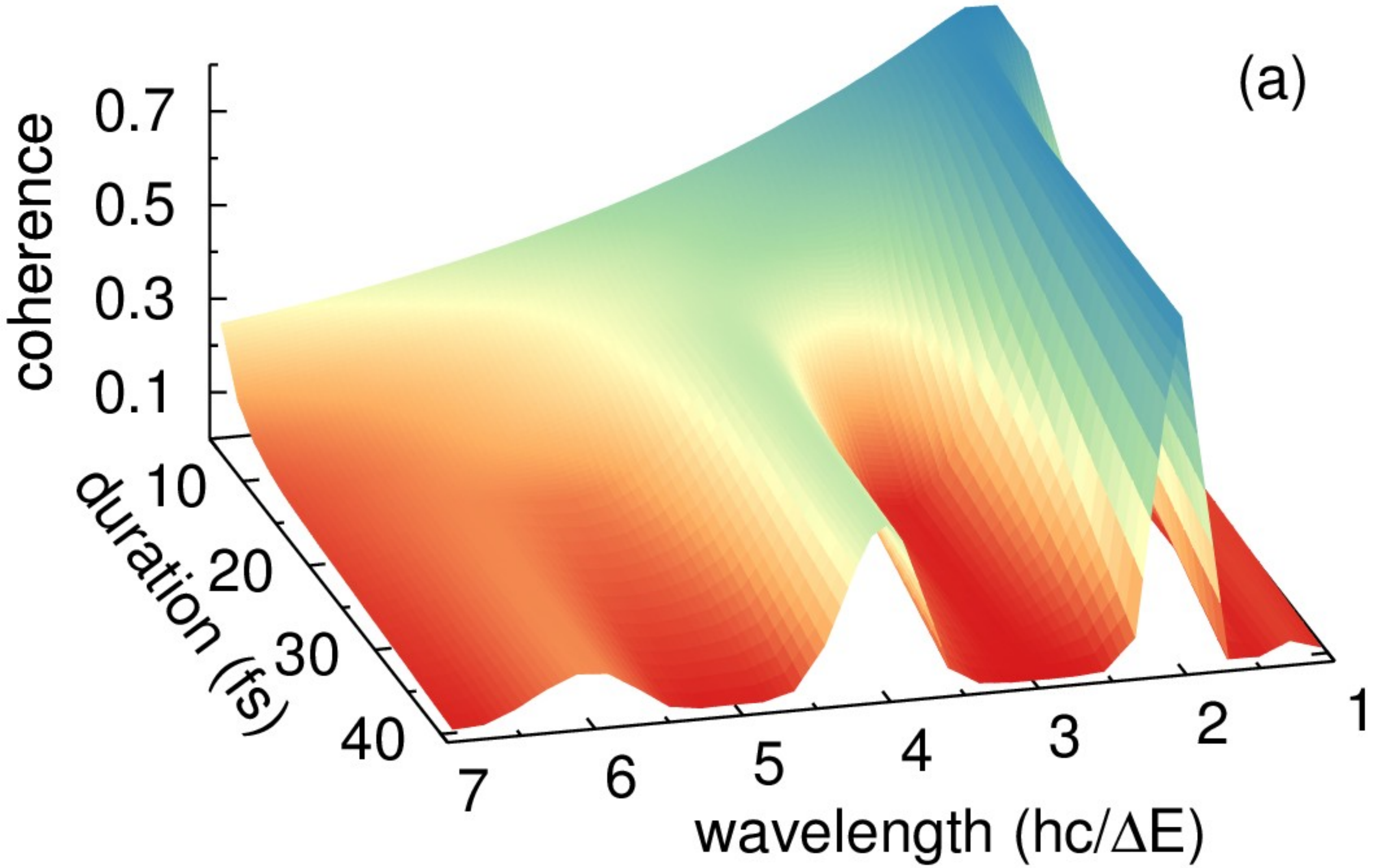
Coherence window:

$$|\Gamma_{IJ}(\Delta E_{JI})|$$

- Strong wavelength dependence (2ω or 2ΔE)
- Envelope is defined by the sub-cycle
- Pulse envelope affects sub-structure

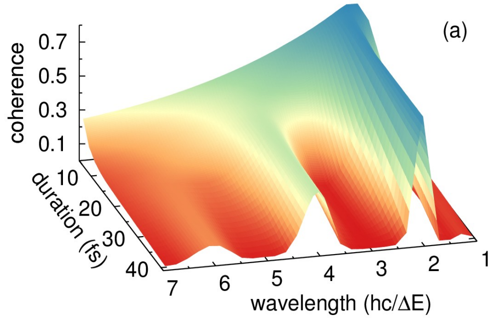
How to create coherent hole motion?

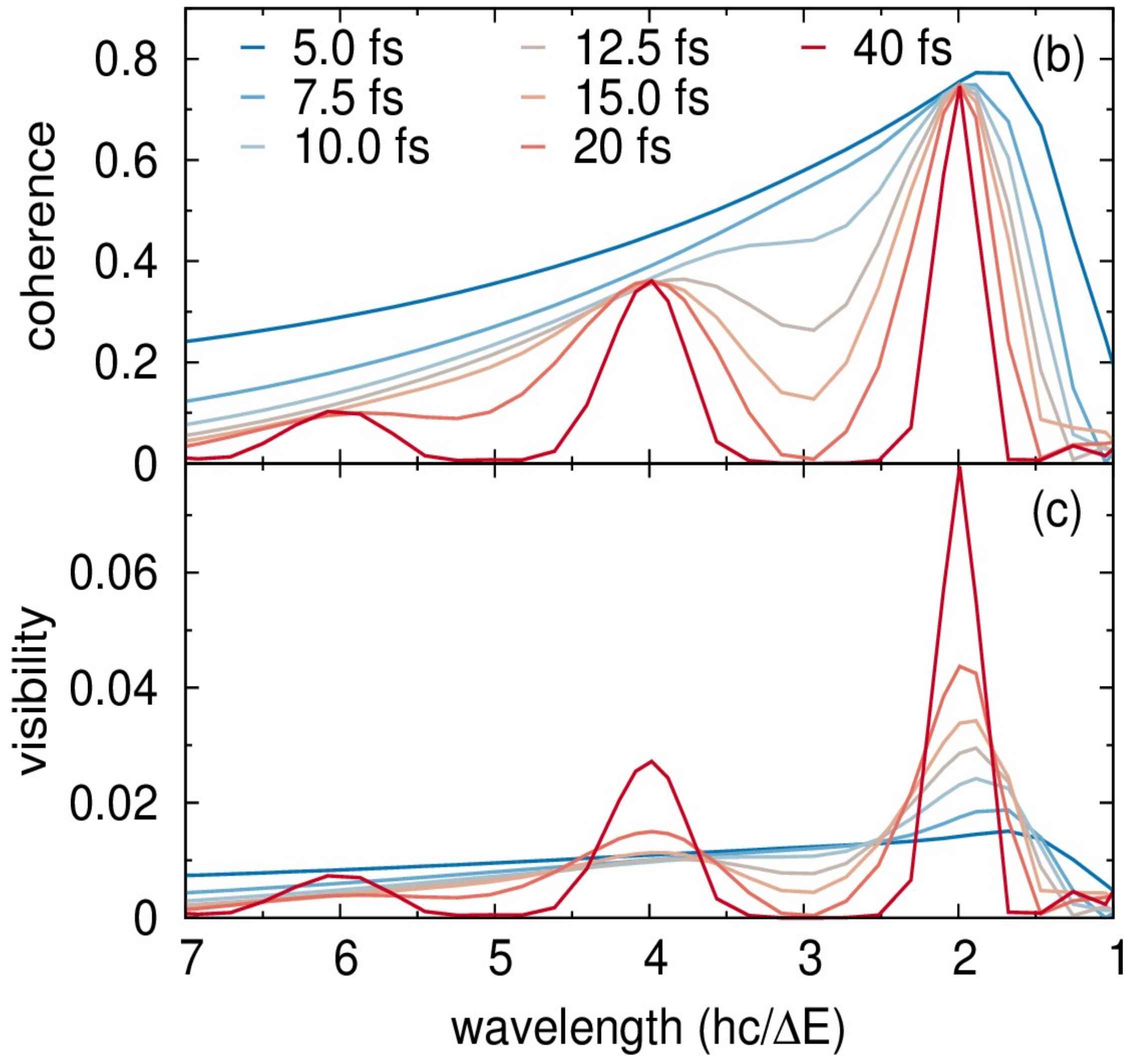
Coherence window – wavelength/duration dependencies



How to create coherent hole motion?

Coherence window – wavelength/duration dependencies





> **Pulse duration**

- 2ΔE peaks become more narrow
- Peak height unaffected
- Off-diagonal matrix element increases

Outline

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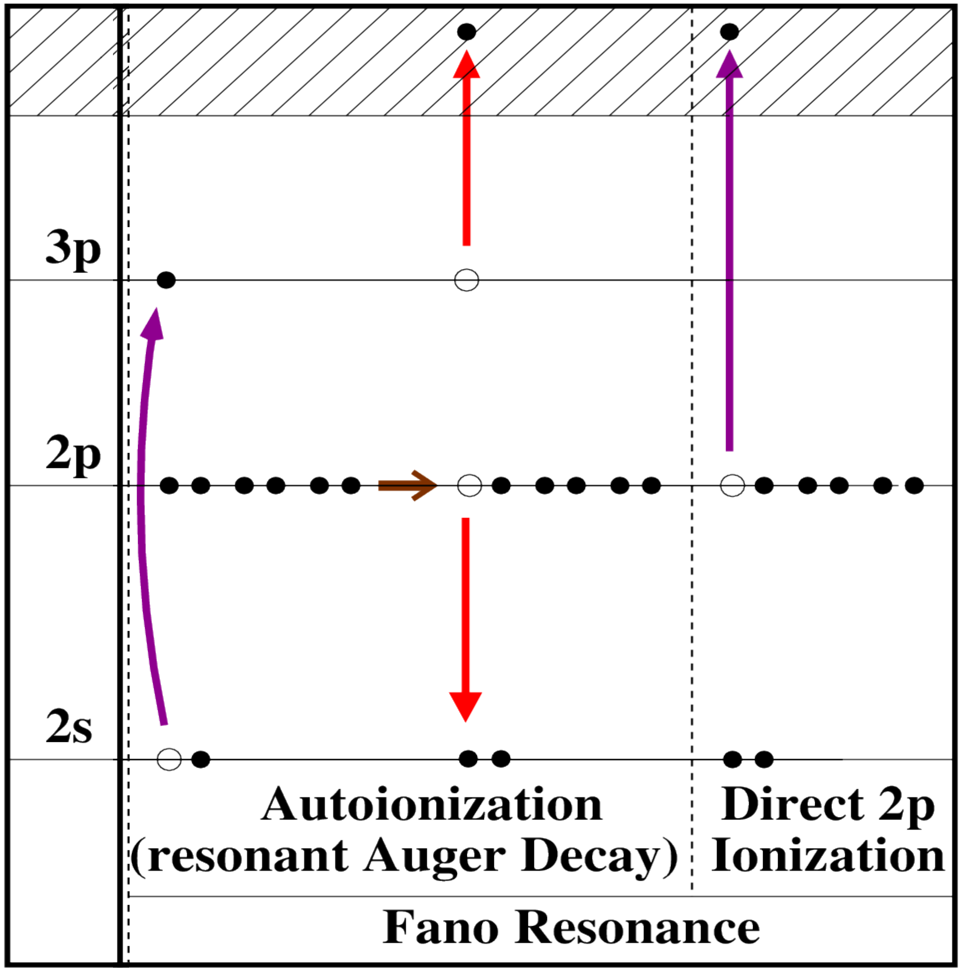
Hole alignment using Fano resonances

Test System: Neon



Final hole in 2p shell

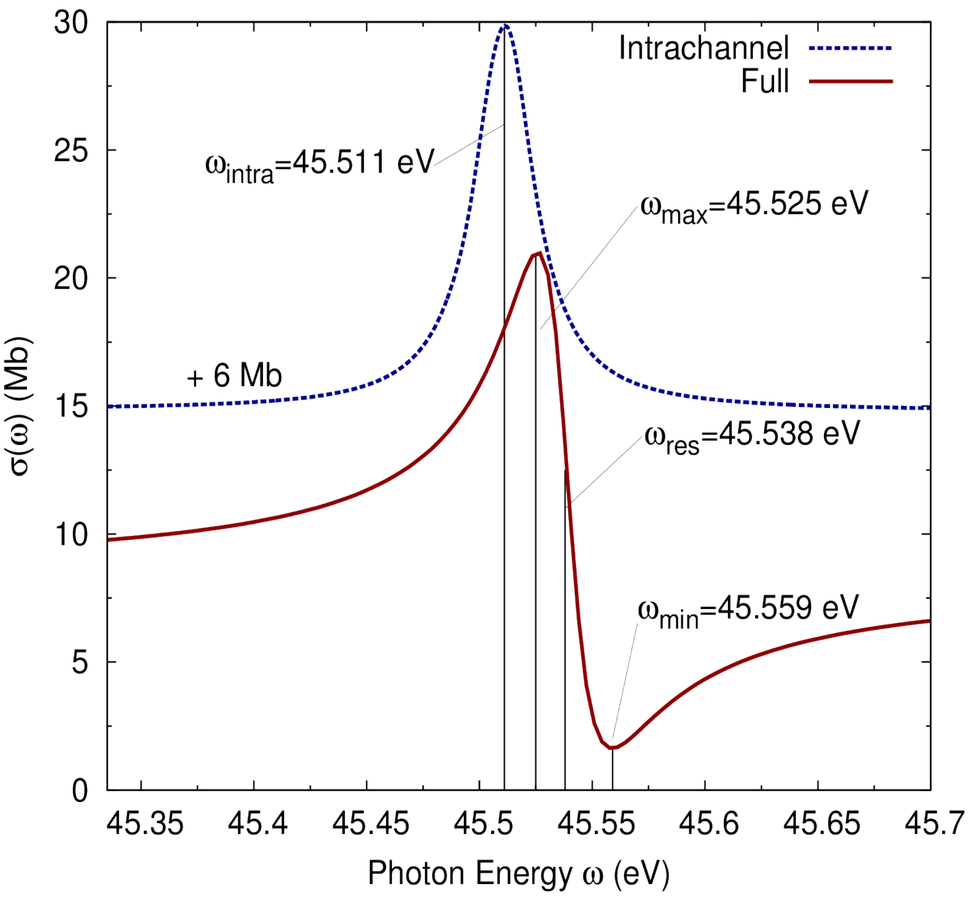
What is the hole alignment (m distribution)

?

Heinrich-Josties *et al.*, PRA **89**, 0434015 (2014)

Hole alignment using Fano resonances

Total cross Section



$$\rho(t;\omega) = \sigma(\omega) \int_{-\infty}^{t} dt' \; I_\omega(t')$$

Modulation of total 2p population

Do each m component behave similarly ?

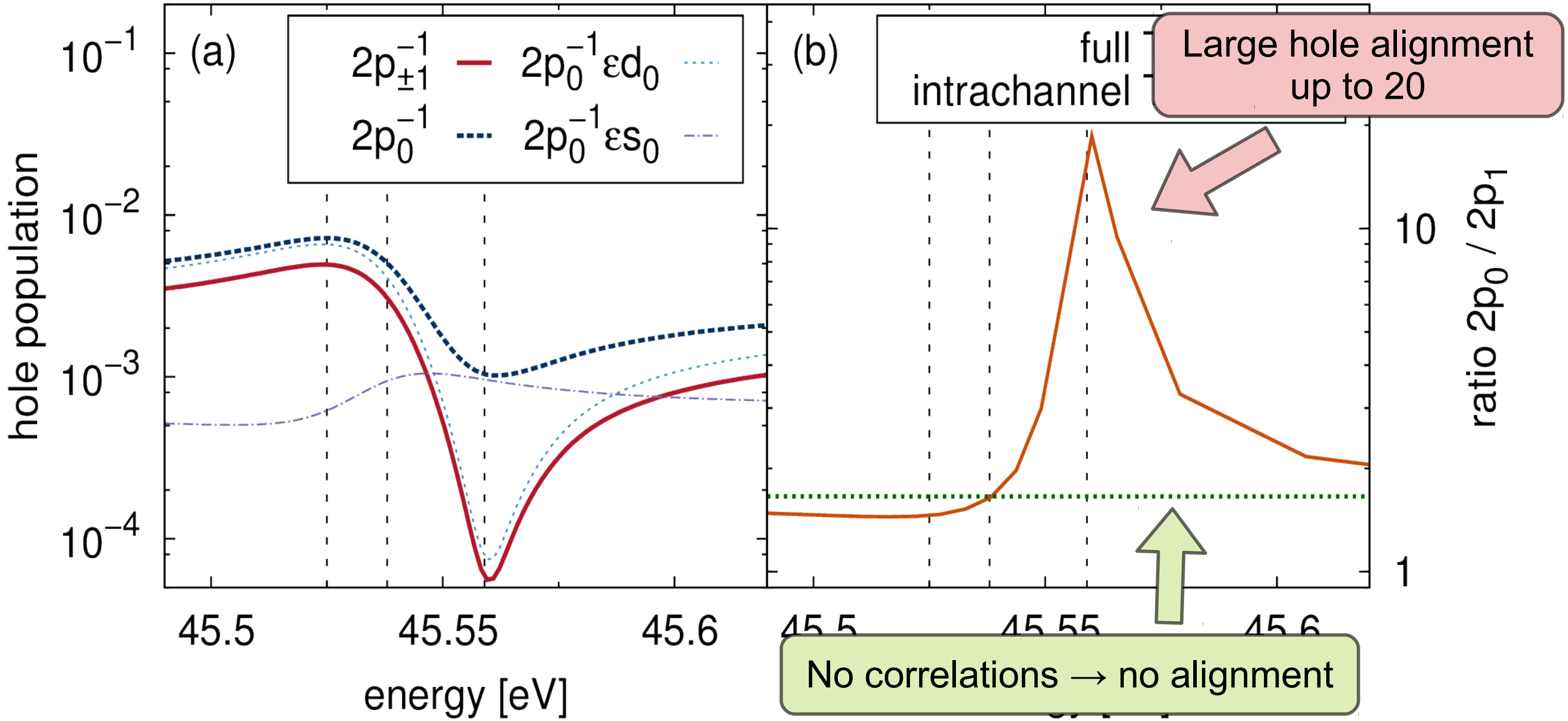
Heinrich-Josties *et al.*, PRA **89**, 0434015 (2014)

Hole alignment using Fano resonances

Partial cross section

$$\rho_i(t;\omega) = \sigma_i(\omega) \int_{-\infty}^{t} dt' \; I_\omega(t')$$

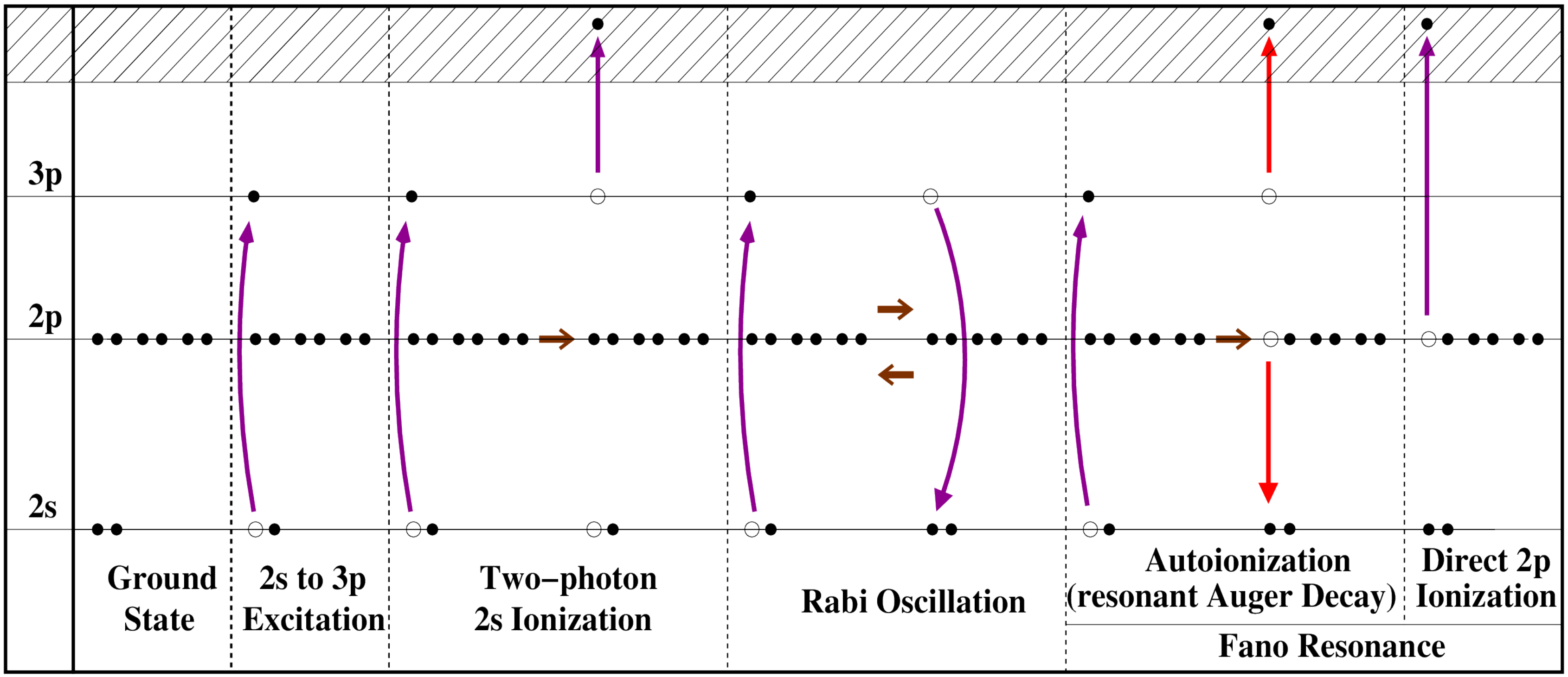
$$a = \frac{\rho_{2p_0}}{\rho_{2p_{\pm 1}}}$$



Heinrich-Josties *et al.*, PRA **89**, 0434015 (2014)

Hole alignment using Fano resonances
Driving with an intense FEL pulse

> Increasing XUV intensity

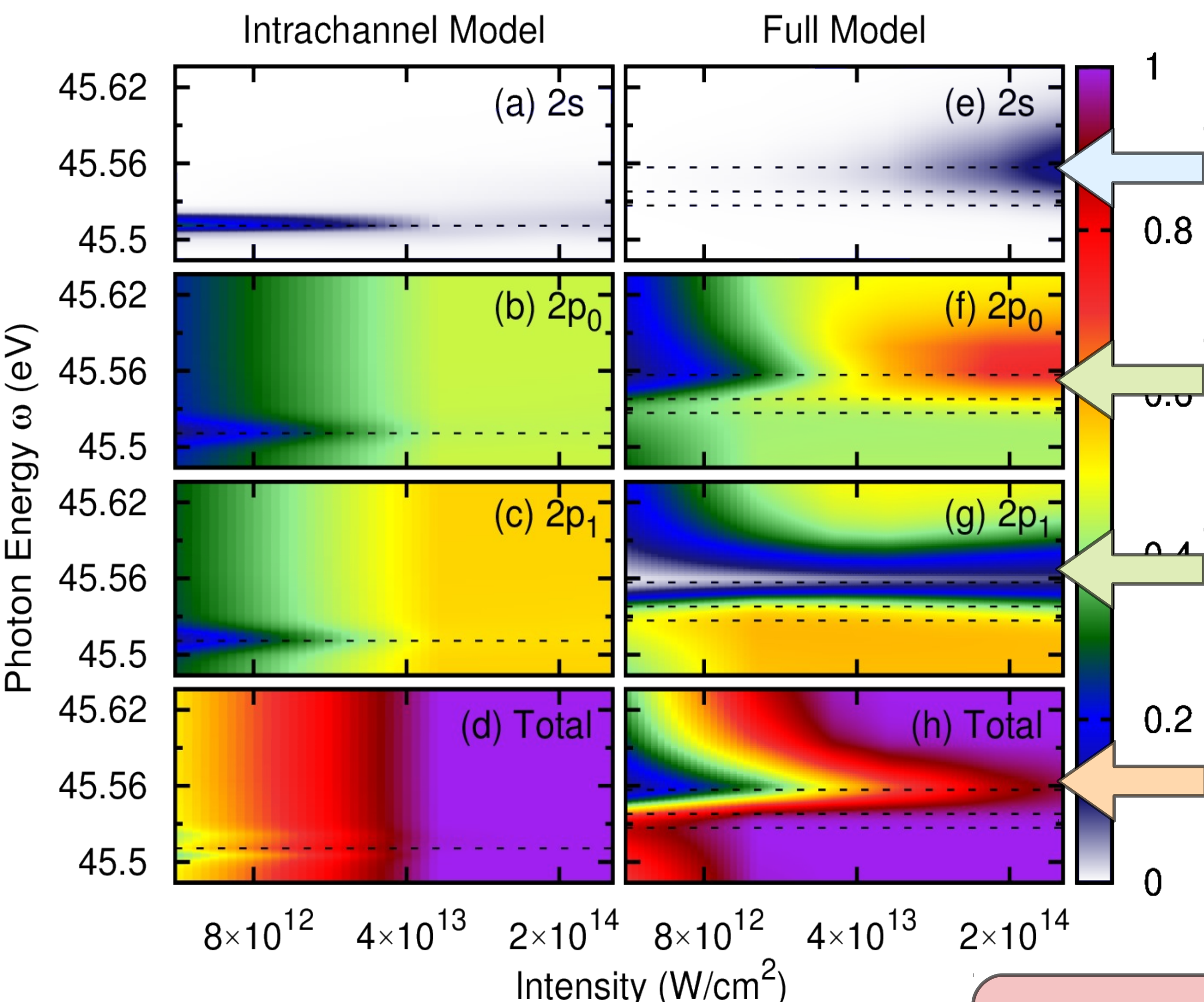


> More processes

- Multi-photons processes
- Multi-electron ionization (ignored here)

Hole alignment using Fano resonances

Driving with an intense FEL pulse



2s hole stability
2-photon ionization

Non-trivial $2p_0$ ionization behavior

Reduced $2p_{\pm 1}$ ionization

Modified saturation behavior

FERMI experiment in Nov. 2014

Summary

- Creating novel ionic states with ultrashort pulses

- Create coherent motion via strong-field ionization
  - Mechanism/conditions quite different than in few-photon ionization
  - Long pulses can create fast hole motion

- Exploiting electron correlations can significantly change the hole distributions
  - Mimicking strong-field ionization with one photon
  - 2-photon ionization can be larger than 1-photon ionization

Acknowledgment

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