Nonlinear processes in atoms

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Workshop on “Science with seeded FEL beams”
Hamburg, Germany
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What seeding might do for us ...

- Category A: smaller bandwidth → "long", spectrally narrow pulses
  - "Resonant by design"
    → x-ray driven Rabi oscillations
  - "Resonant by accident"
    → nonsequential (direct) two-photon absorption
    → enhanced multiple ionization
- Category B: shorter pulses → transform-limited sub-femtosecond pulses
  - Suppression of electronic damage in coherent diffractive imaging
  - Electronic coherence and correlation in the time domain
  - Imaging of electronic quantum motion

Acknowledgments I

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Acknowledgments II (xenon)

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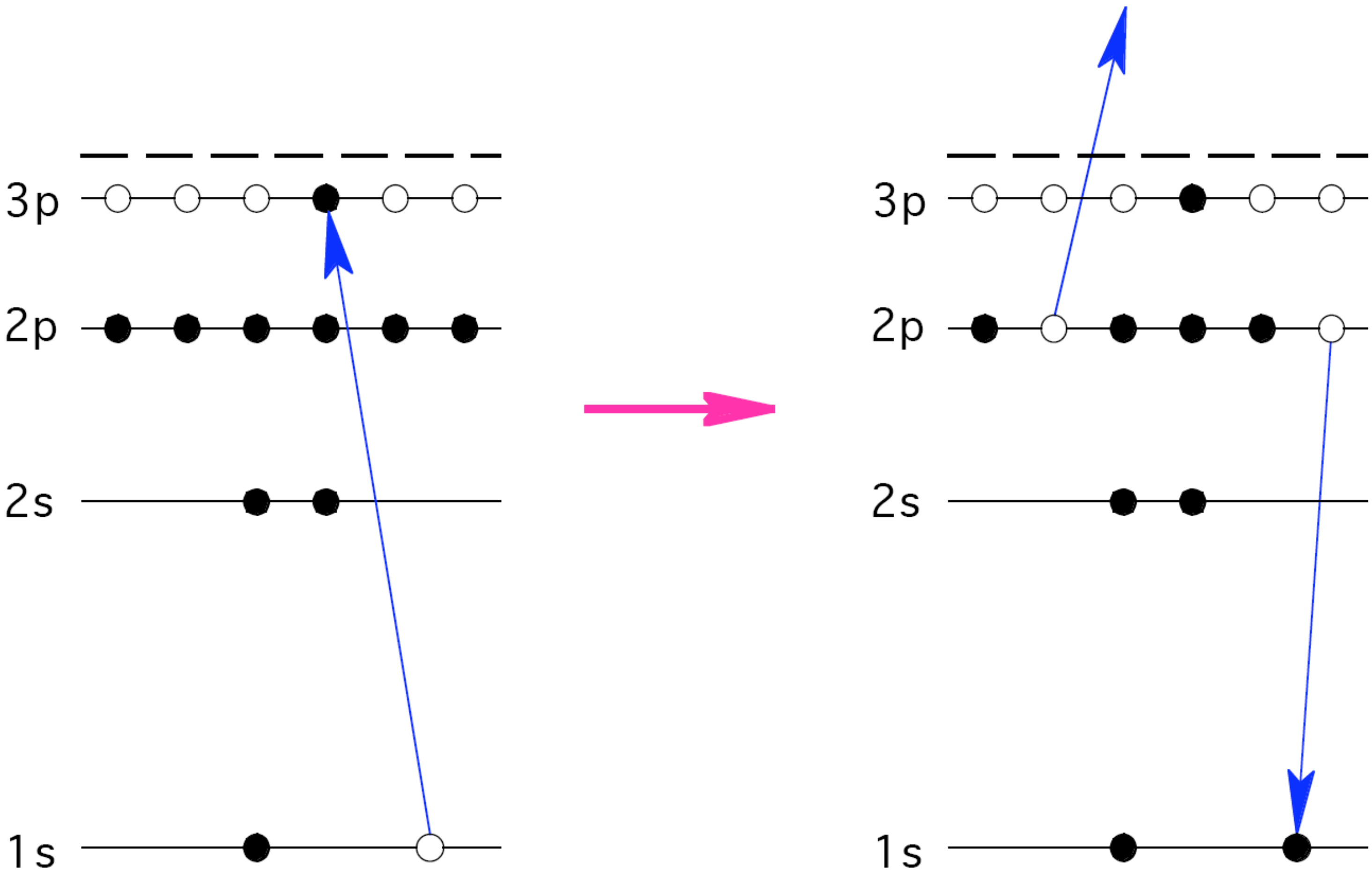
“Resonant by design”

- X-ray driven Rabi oscillations involving inner-shell electrons
- Modification of bound electronic dynamics leads to modification of Auger electron line profile
- Step towards the development of nonlinear spectroscopy in the x-ray domain

Resonant Auger effect



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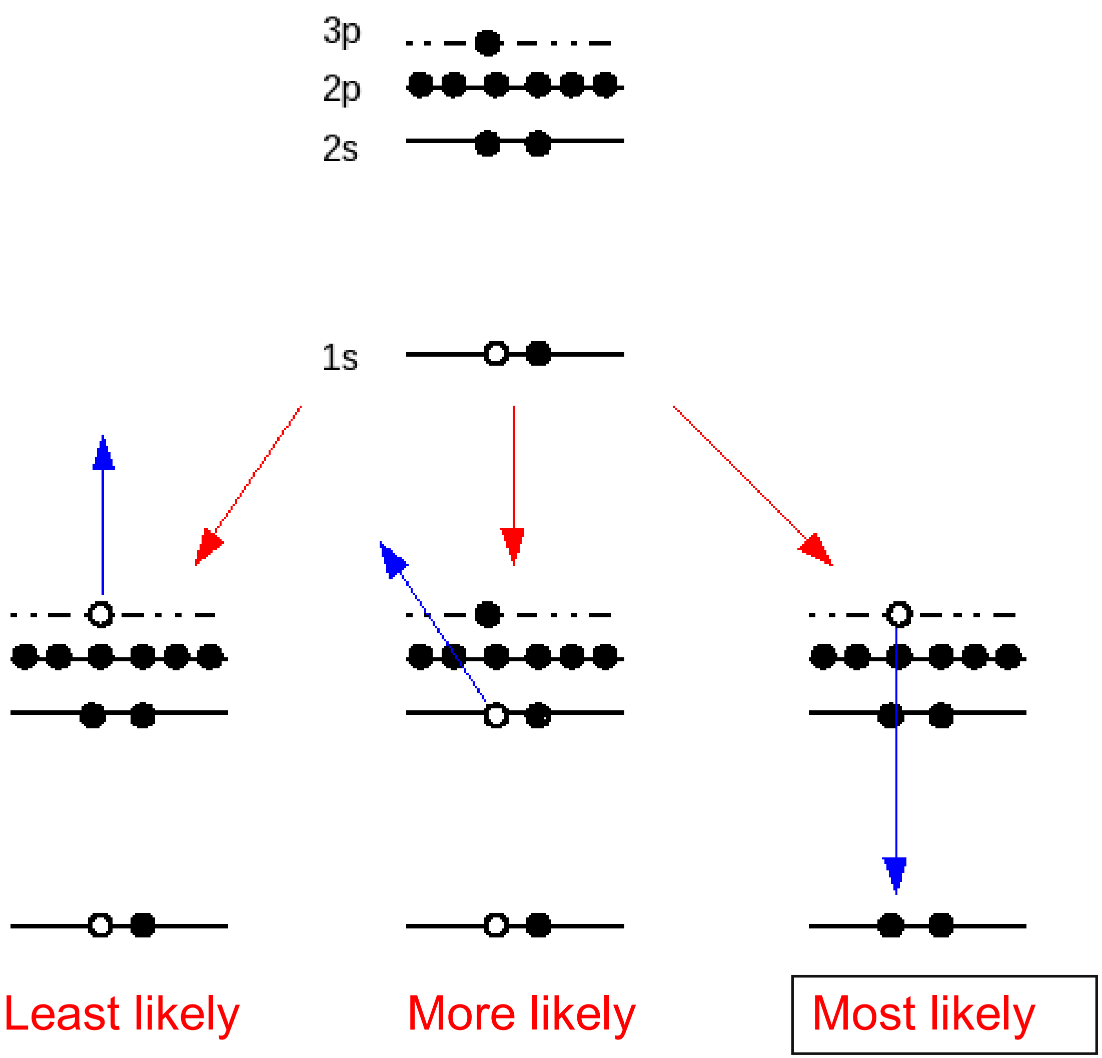
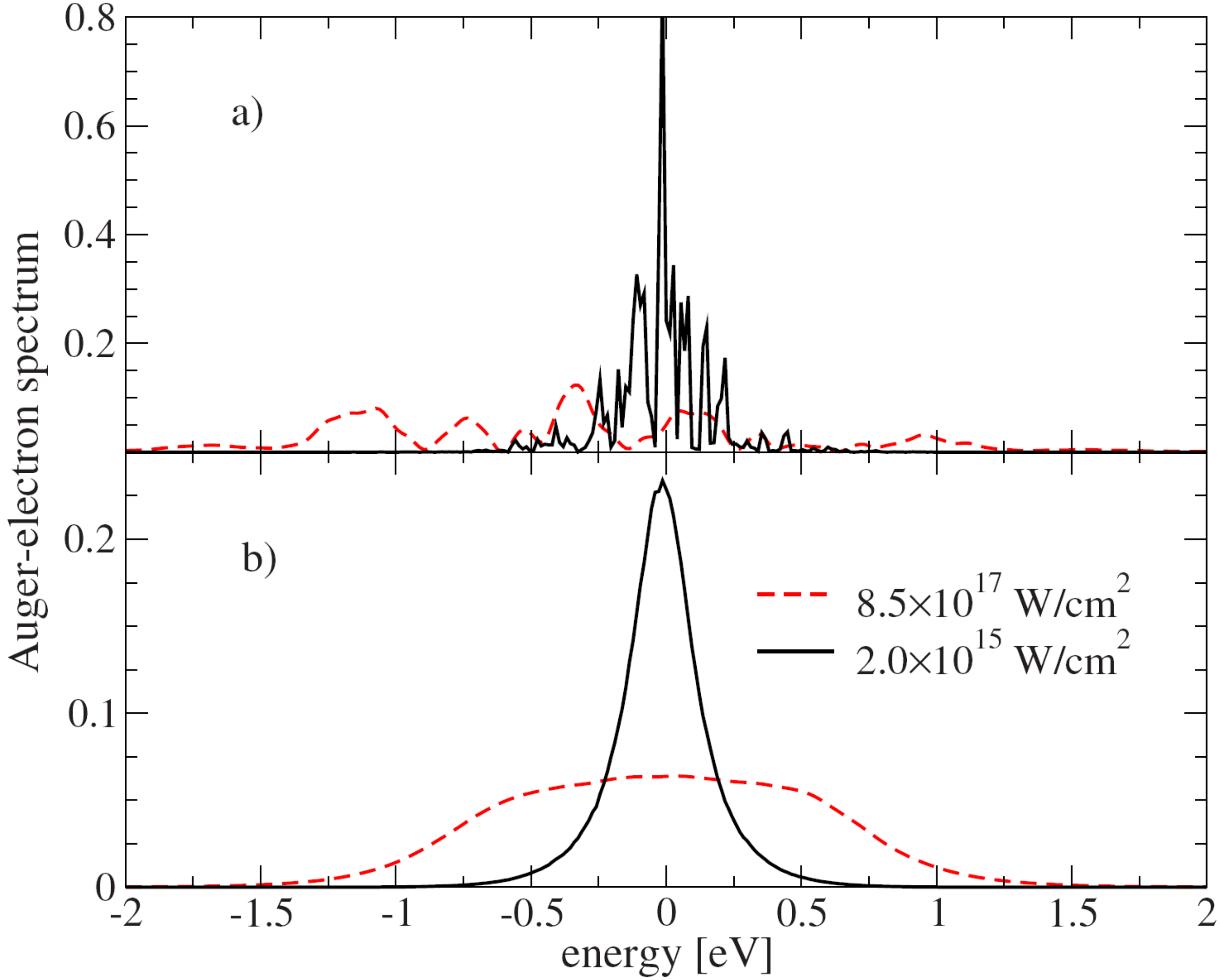
X-ray driven processes competing with resonant Auger decay



Resonant Auger line profile (SASE, 230 fs)



a) Single shot

b) Ensemble average

Rohringer, Santra, Phys. Rev. A **77**, 053404 (2008)

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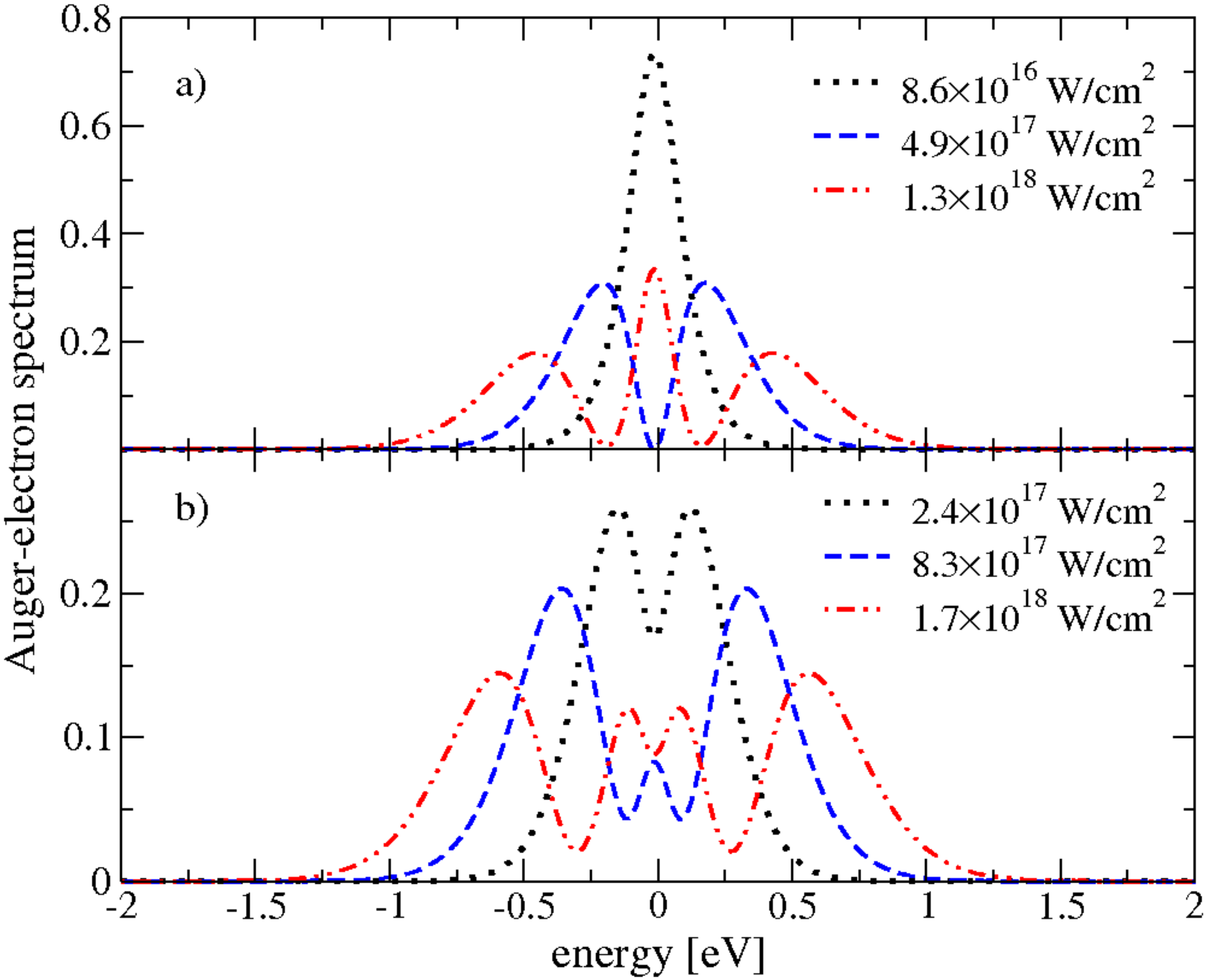
Resonant Auger line profile (Gaussian pulse, 2 fs)



Rohringer, Santra, Phys. Rev. A **77**, 053404 (2008)

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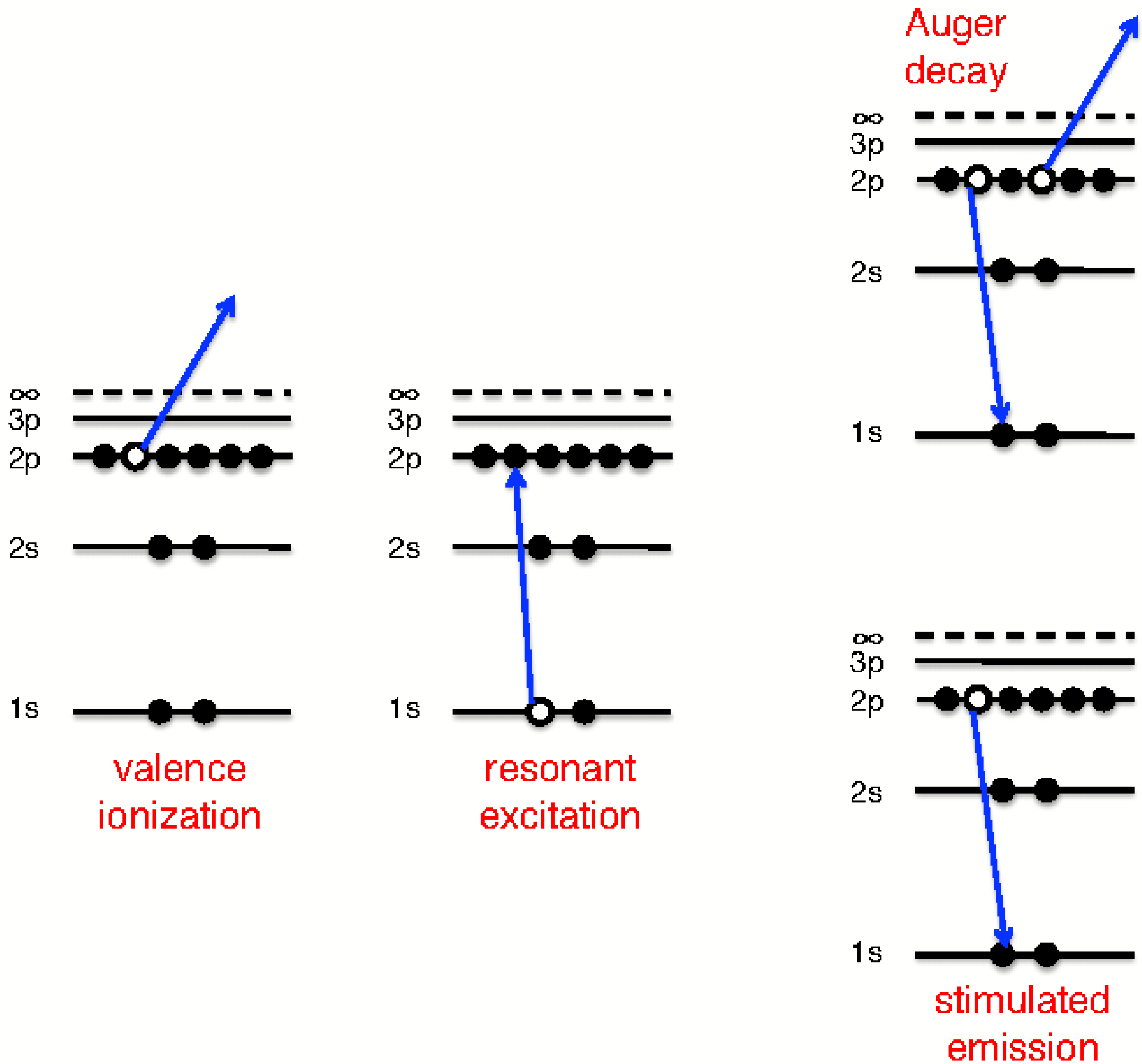
Neon interacting with a high-intensity x-ray pulse at 848 eV



E. P. Kanter *et al.*, Phys. Rev. Lett. **107**, 233001 (2011)

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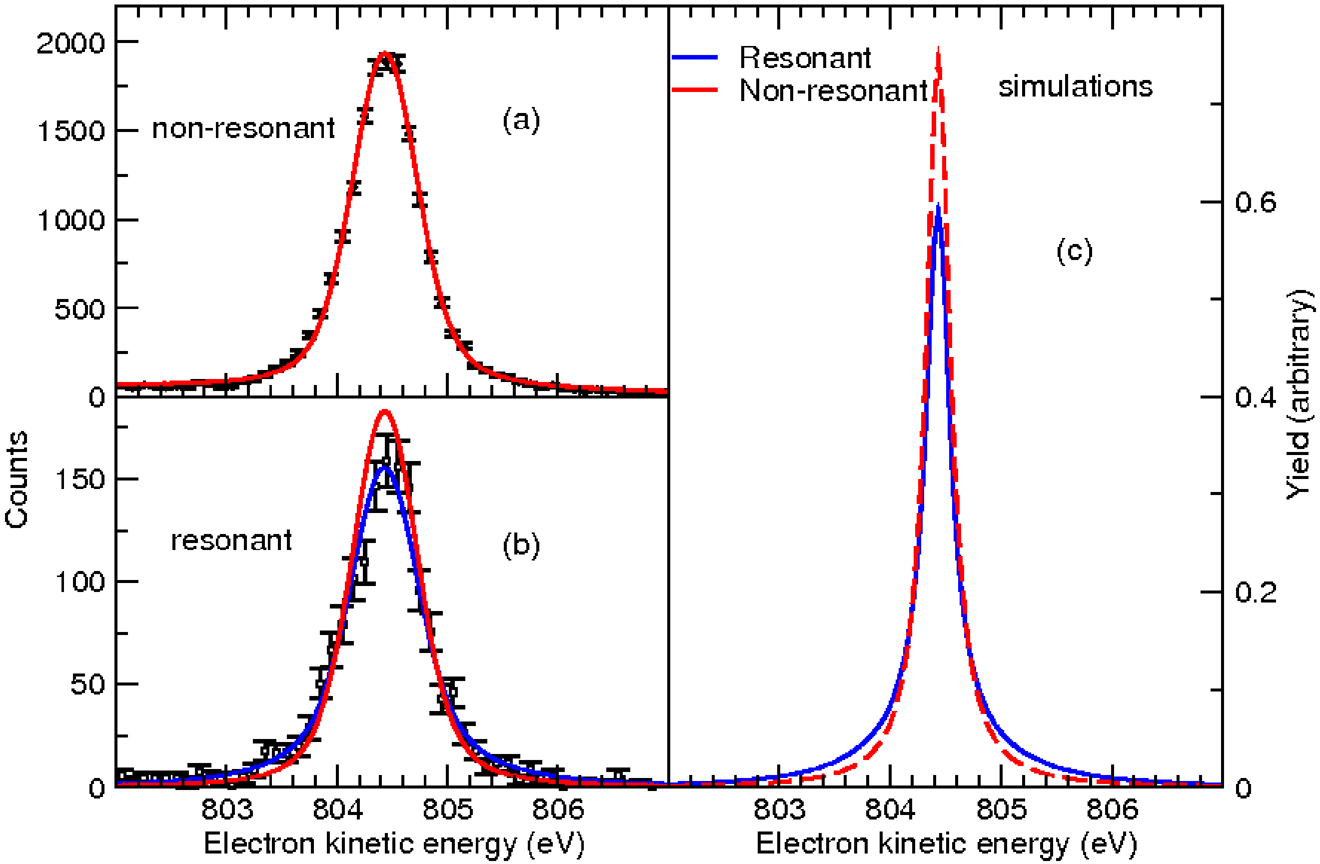
Modification of Auger line profile by high-intensity x rays



Resonant: 848 eV
Non-resonant: 930 eV

Resonant Rabi period approaching 1 fs

E. P. Kanter *et al.*, Phys. Rev. Lett. **107**, 233001 (2011)

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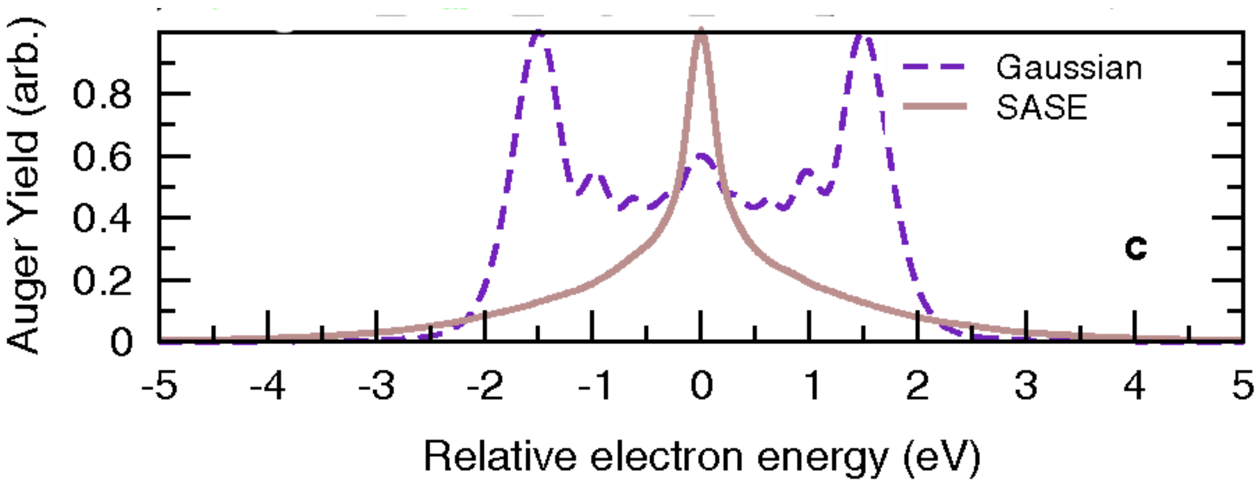
SASE vs. Gaussian (theory)

3.5×10^{17} W/cm^2



E. P. Kanter *et al.*, Phys. Rev. Lett. **107**, 233001 (2011)

The resonant Auger line shape generated by an ensemble of SASE pulses (averaged Gaussian temporal profile of 8.5 fs FWHM, 6 eV bandwidth) and a longitudinally coherent Gaussian pulse (8.5 fs FWHM, transform-limited).

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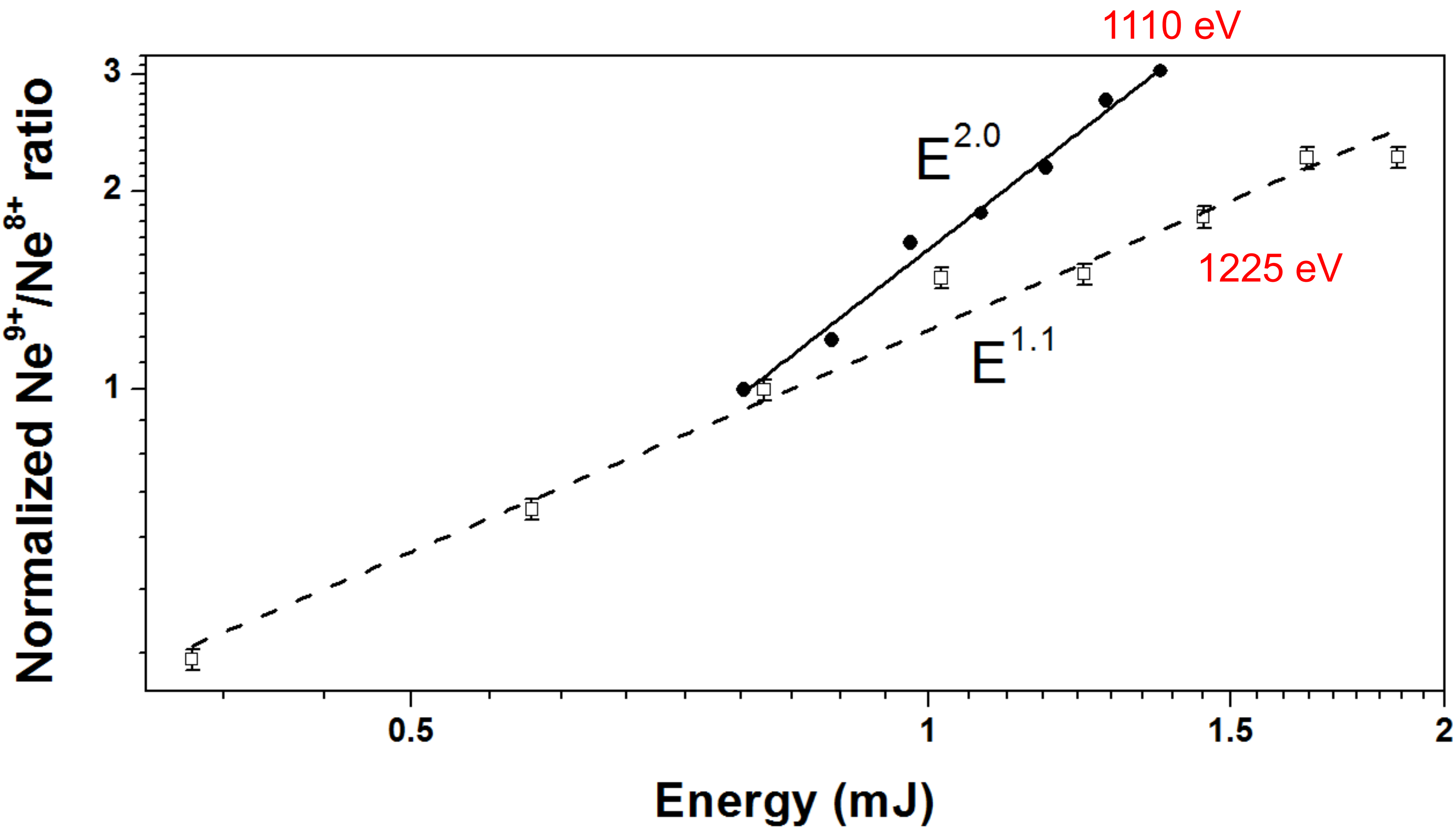
“Resonant by accident”

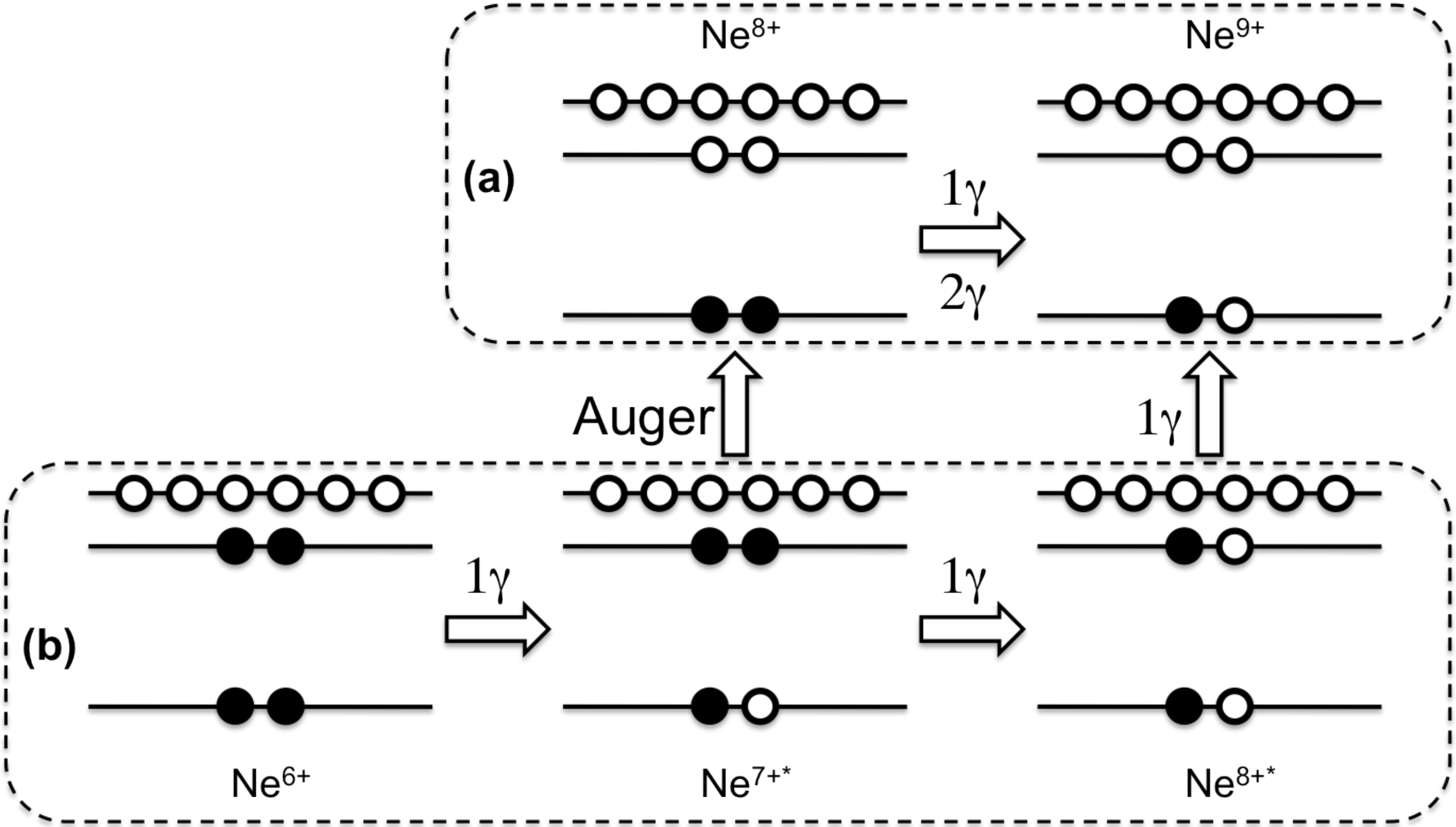
$\rightarrow$ nonsequential (direct) two-photon absorption

Nonlinear production of Ne^{9+}: observation



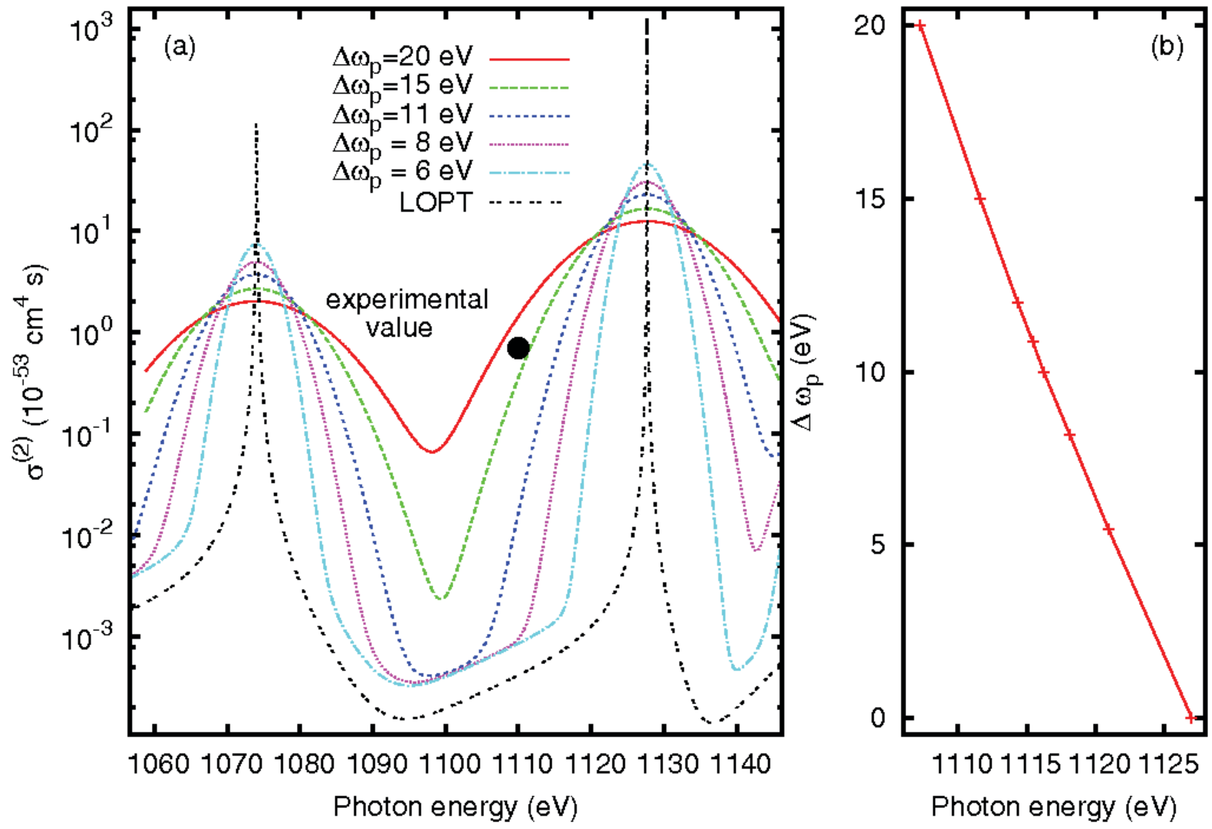
G. Doumy *et al.*, Phys. Rev. Lett. **106**, 083002 (2011)

Nonlinear production of Ne^{9+}: mechanisms



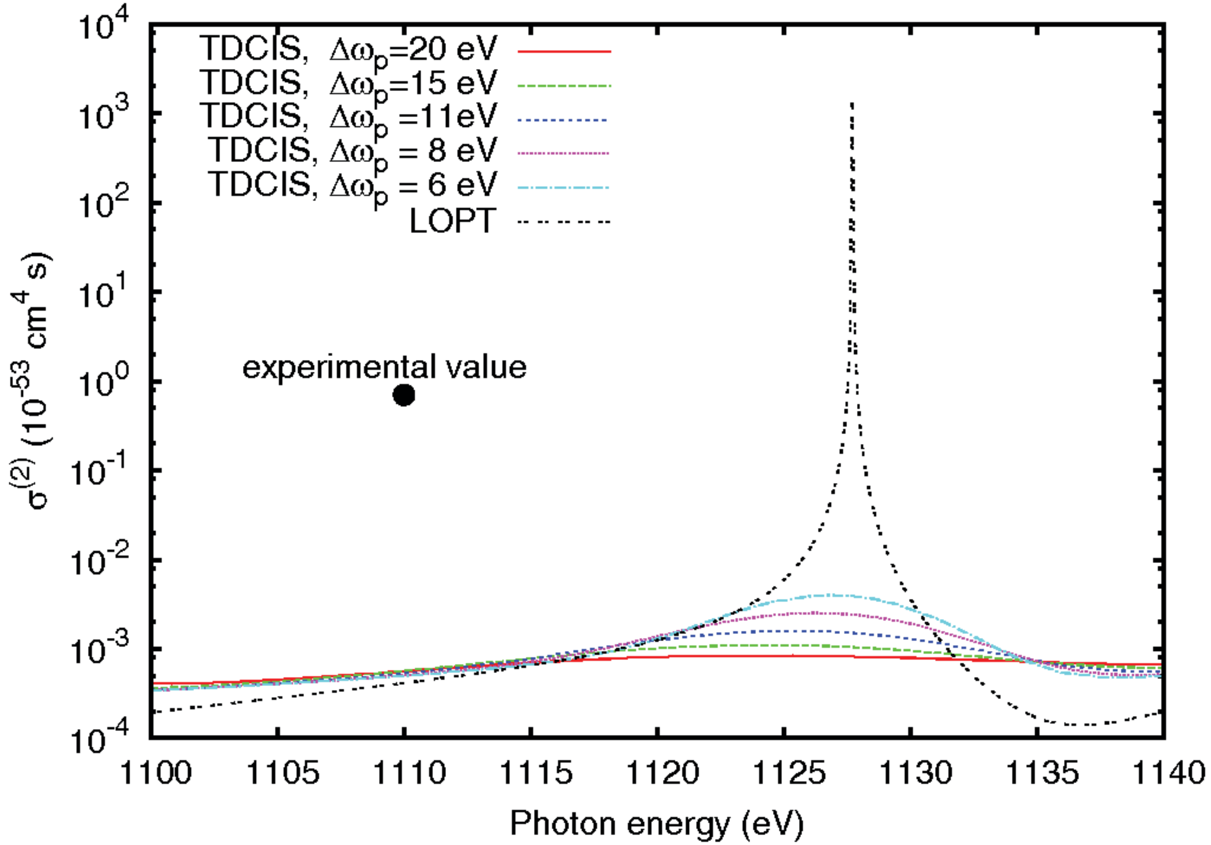
G. Doumy *et al.*, Phys. Rev. Lett. **106**, 083002 (2011)

Effective two-photon cross section of Ne^{8+}: SASE



A. Sytcheva *et al.*, Phys. Rev. A **85**, 023414 (2012)

Effective two-photon cross section of Ne^{8+}: coherent case



A. Sytcheva *et al.*, Phys. Rev. A **85**, 023414 (2012)

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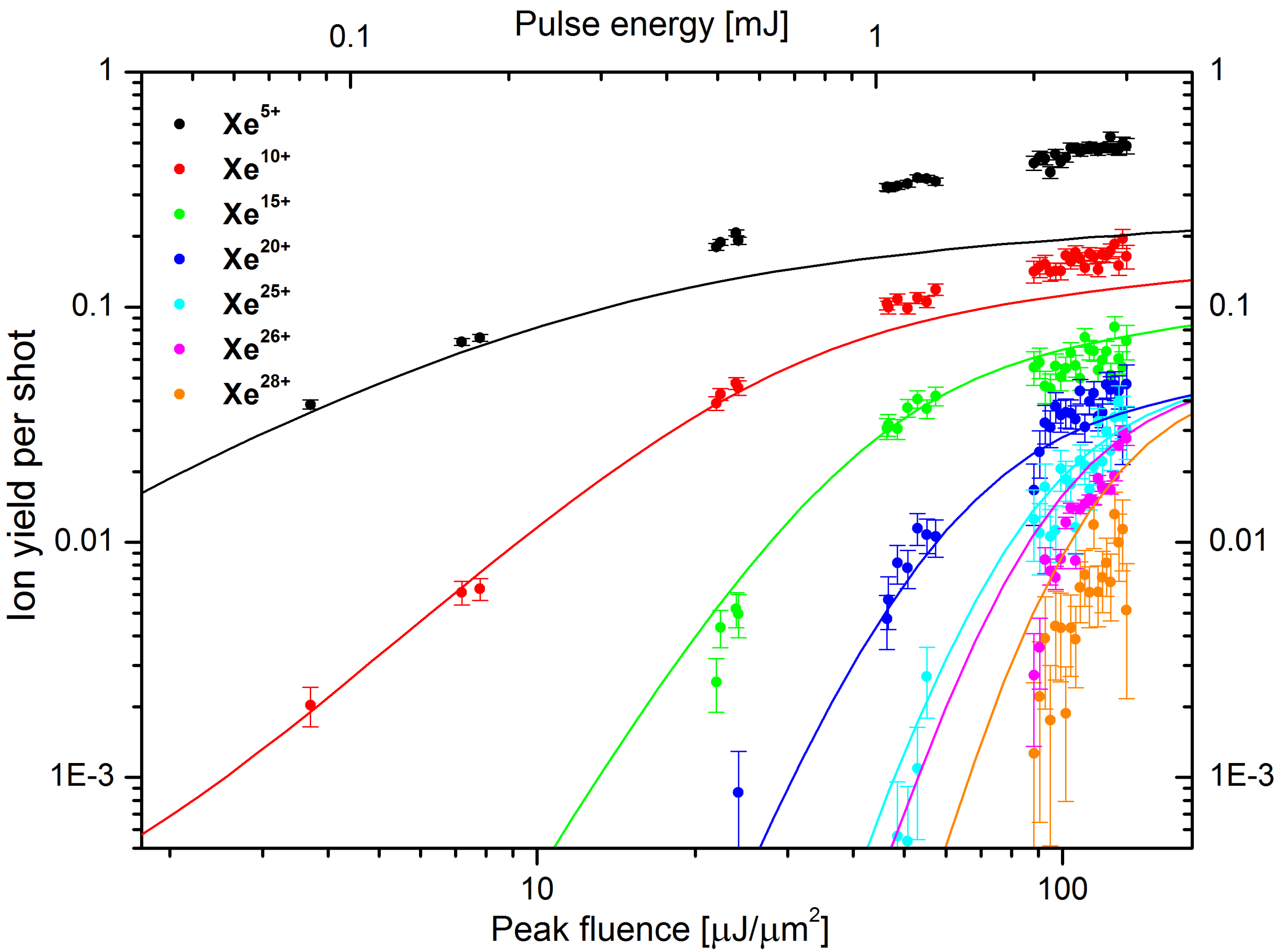
“Resonant by accident”

→ enhanced multiple ionization

X-ray multiphoton ionization of xenon at 2 keV



Comparison between experiment and theory

B. Rudek, S.-K. Son *et al.*, submitted

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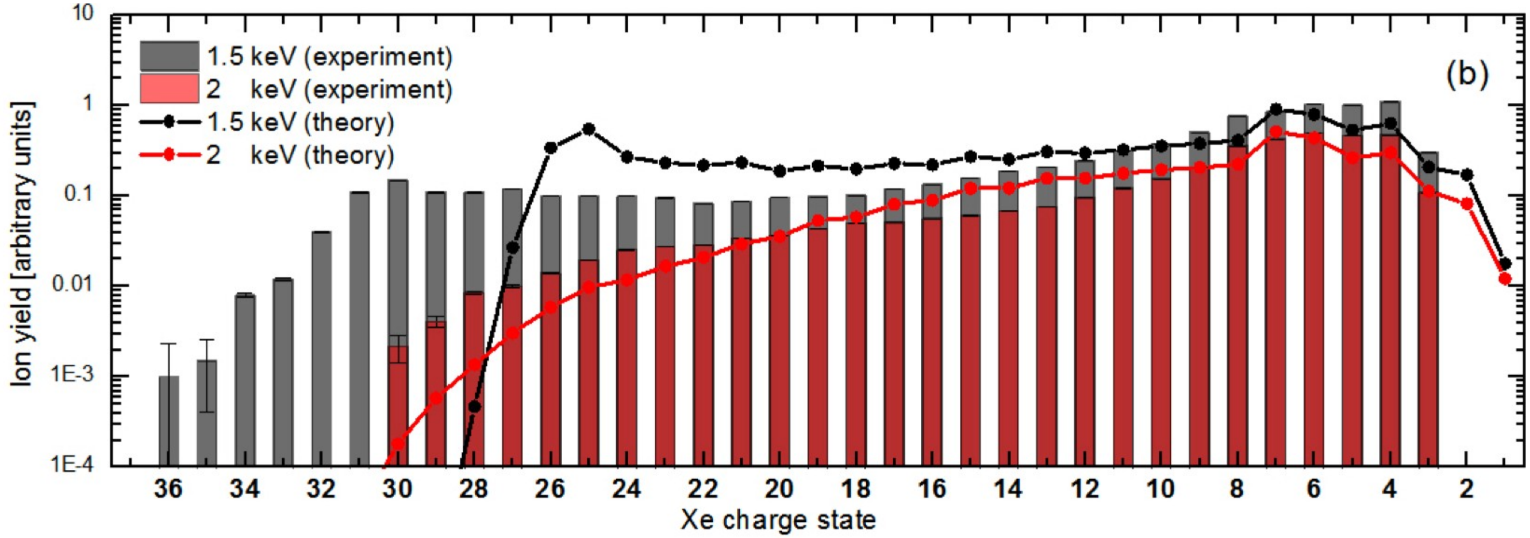
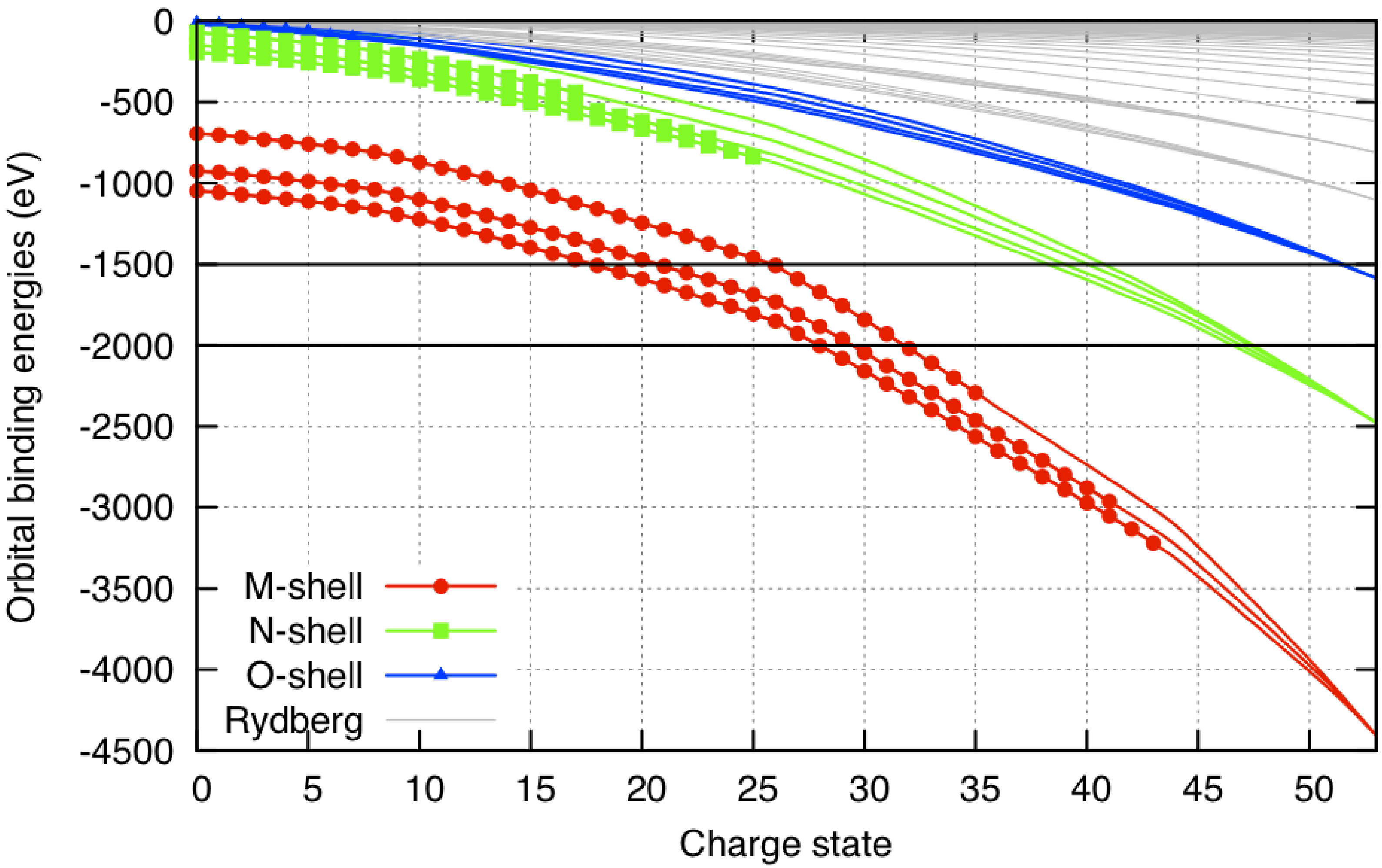
Enhanced multiple ionization at 1.5 keV



B. Rudek, S.-K. Son *et al.*, submitted

Orbital binding energies of the ground configuration of Xe^{q+}



S.-K. Son and R. Santra, Phys. Rev. A **85**, 063415 (2012)

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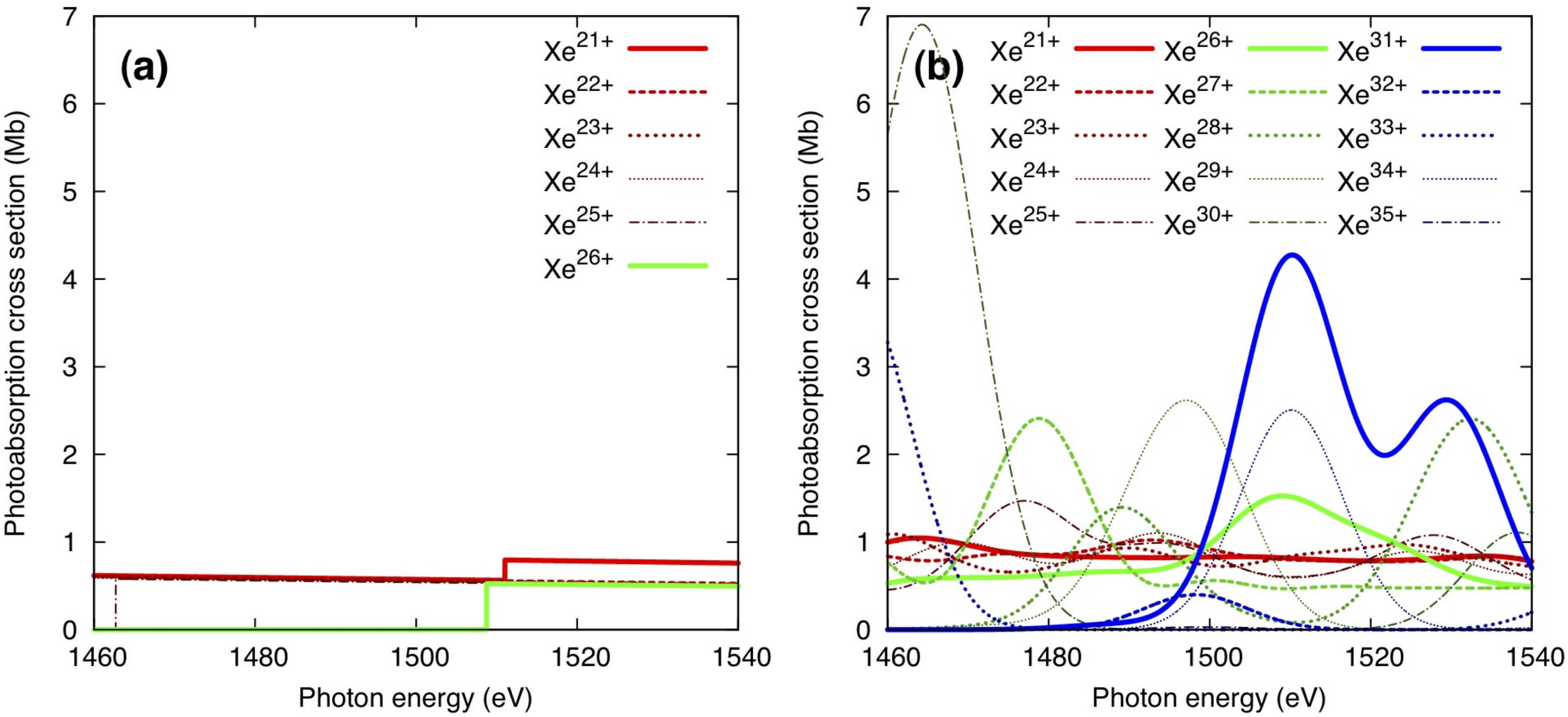
Resonantly enhanced multiple ionization caused by large SASE bandwidth



B. Rudek, S.-K. Son *et al.*, submitted

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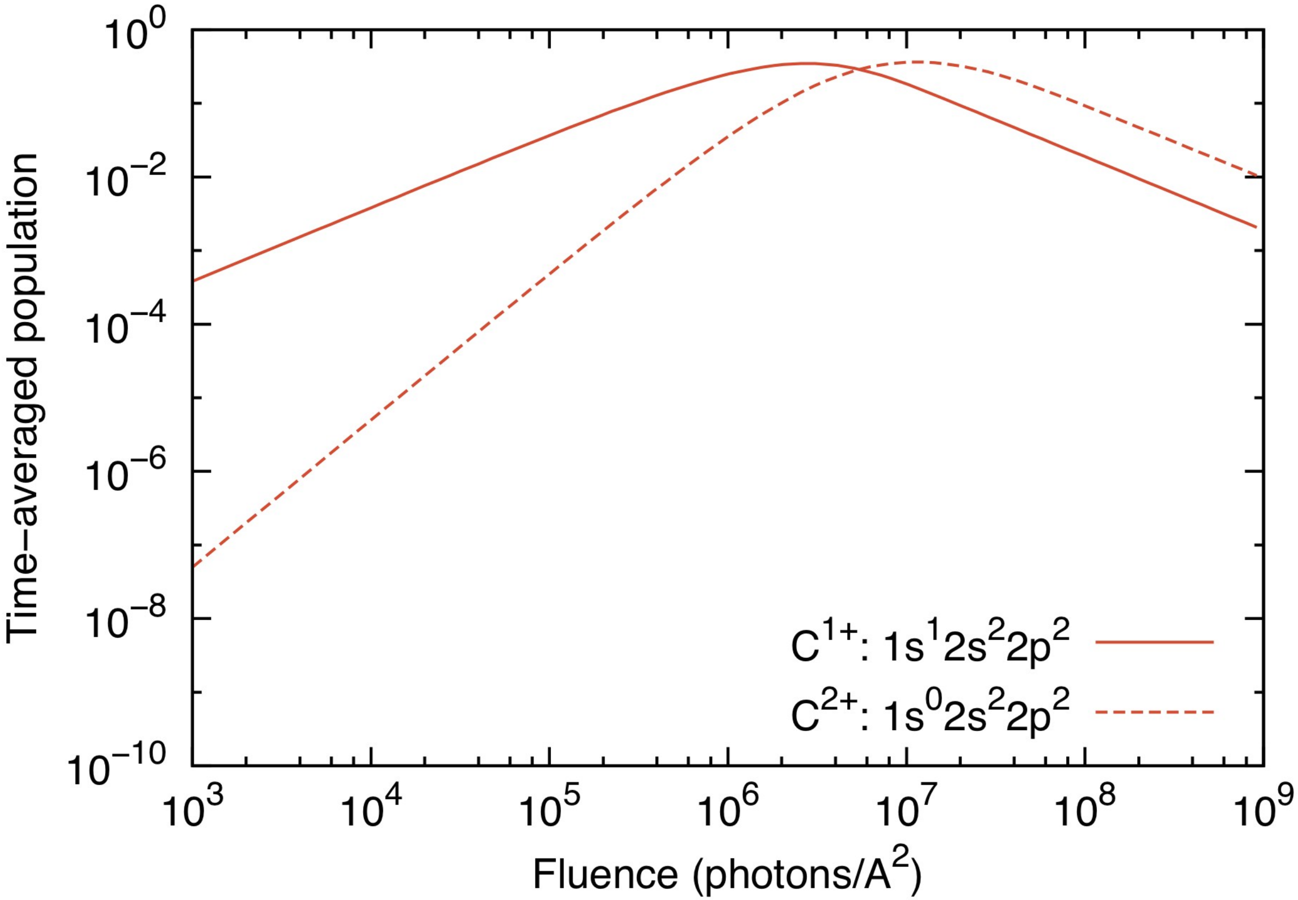
Suppression of electronic damage in coherent diffractive imaging

Time-averaged population of core-hole configurations



8 keV and 1 fs FWHM

10^6 photons/Å^2 = 10^{12} photons/(100 nm)^2

S.-K. Son, L. Young, and R. Santra, Phys. Rev. A **83**, 033402 (2011)

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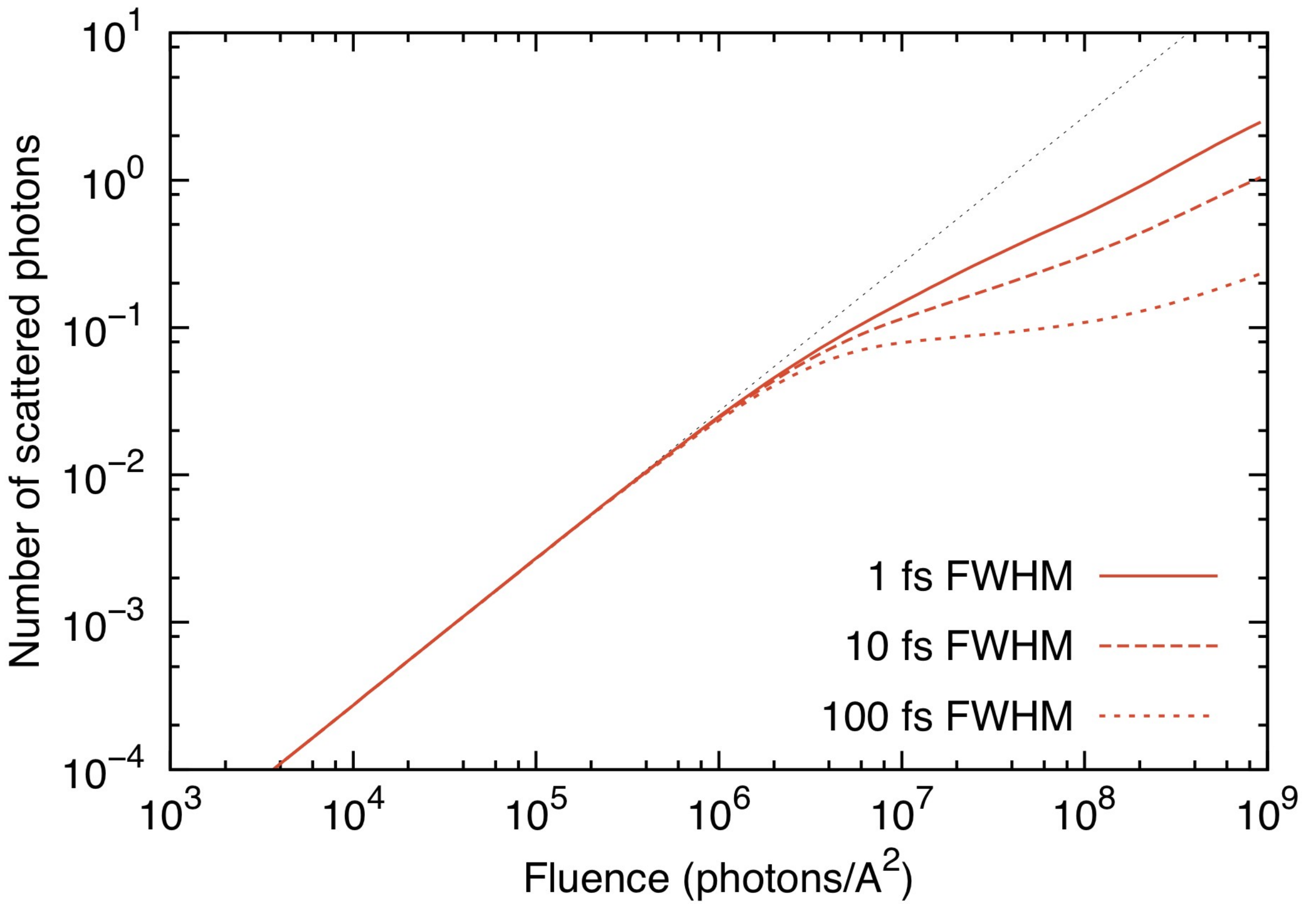
Number of photons scattered by a single carbon atom



8 keV with spatial resolution of 1.7 Å

S.-K. Son, L. Young, and R. Santra, Phys. Rev. A **83**, 033402 (2011)

“x-ray transparency” [Nature **466**, 56 (2010)]
“frustrated absorption” [Phys. Rev. Lett. **104**, 253002 (2010)]

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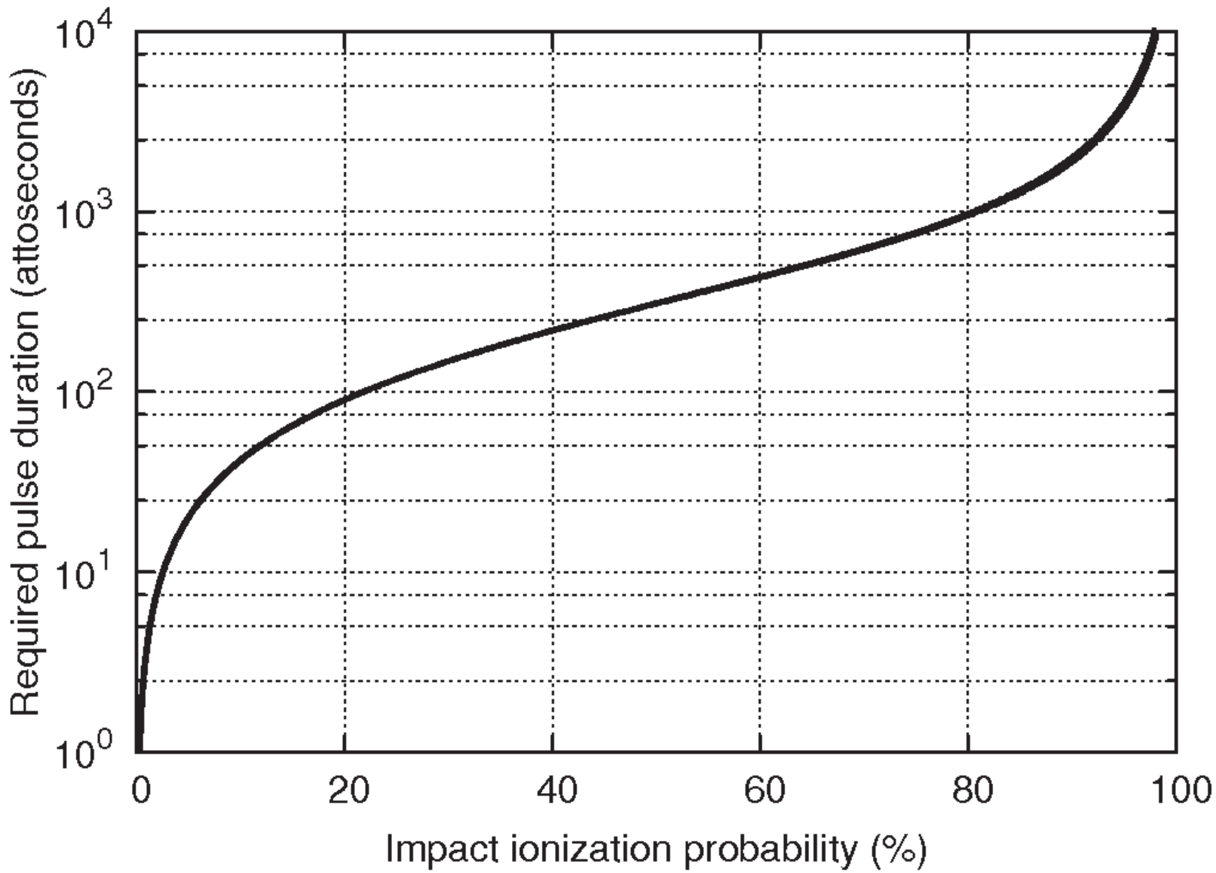
Suppression of electron impact ionization



S.-K. Son, L. Young, and R. Santra, Phys. Rev. A **83**, 033402 (2011)

Pulse duration required for a given impact ionization probability, for a photoelectron with a kinetic energy of 12 keV in a carbon-based medium with a mean free path of 13 nm

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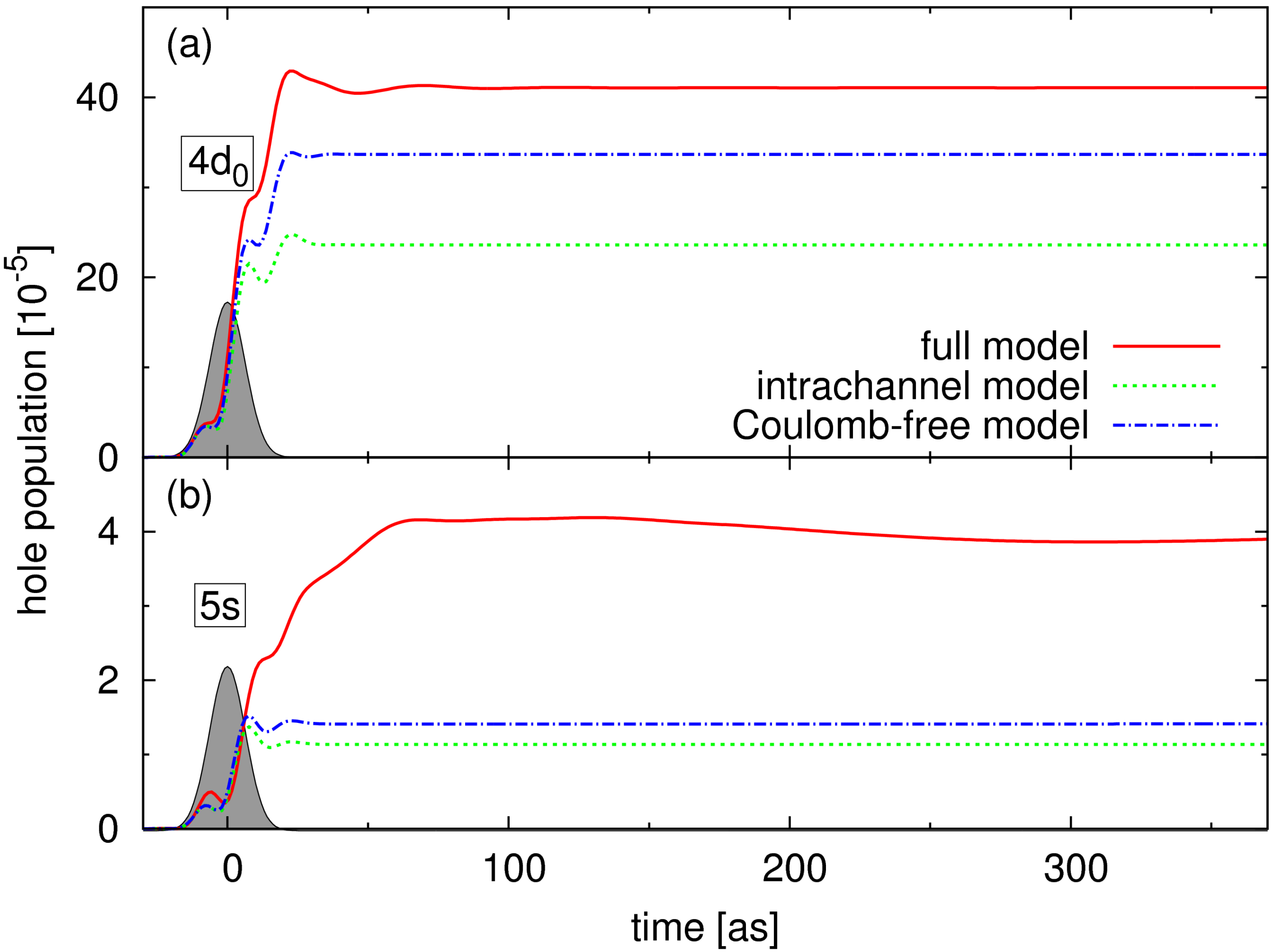
Electronic coherence and correlation in the time domain

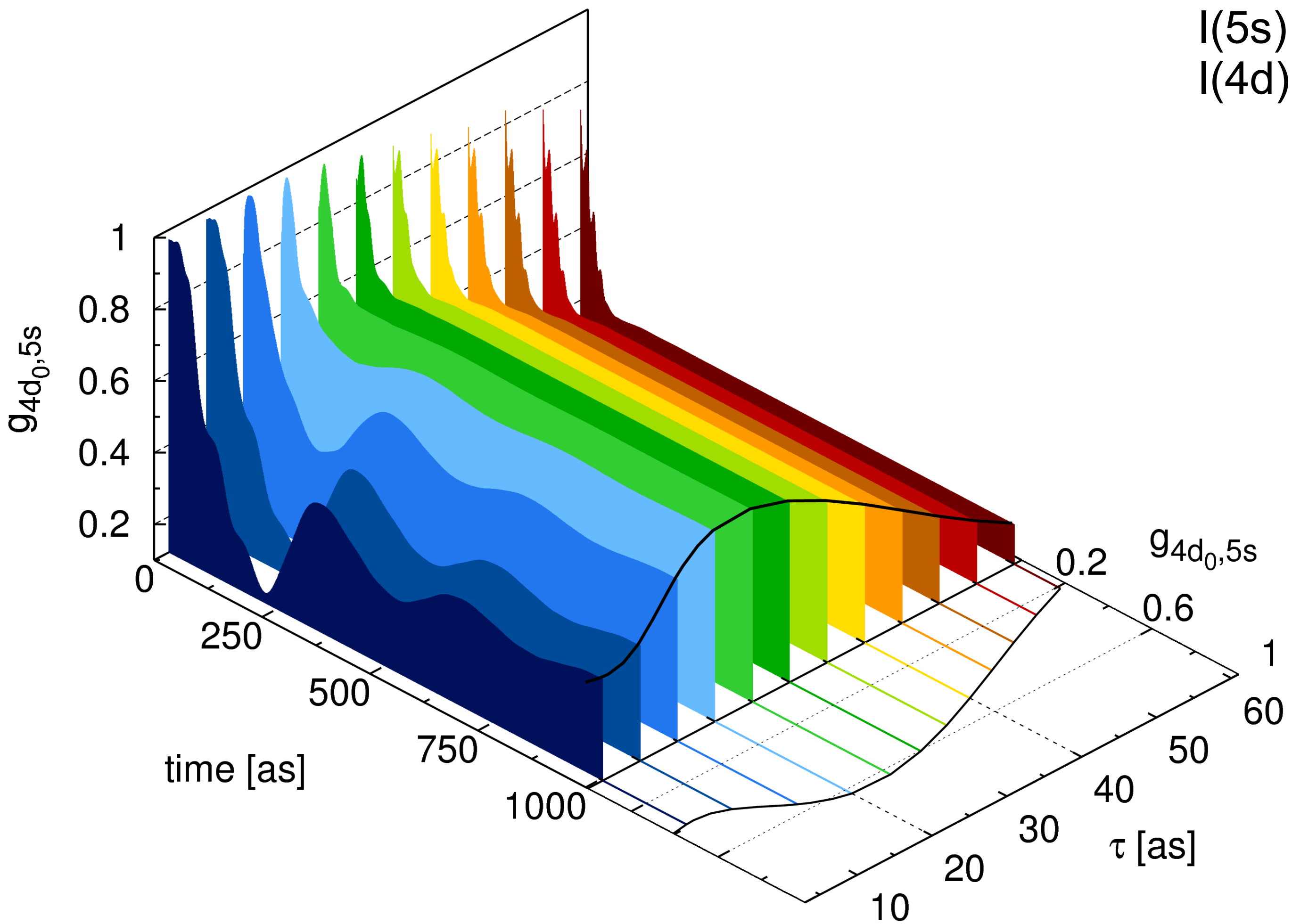
Attosecond photoionization of xenon: hole populations



S. Pabst *et al.*, Phys. Rev. Lett. **106**, 053003 (2011)

Pulse duration 20 as; photon energy 136 eV

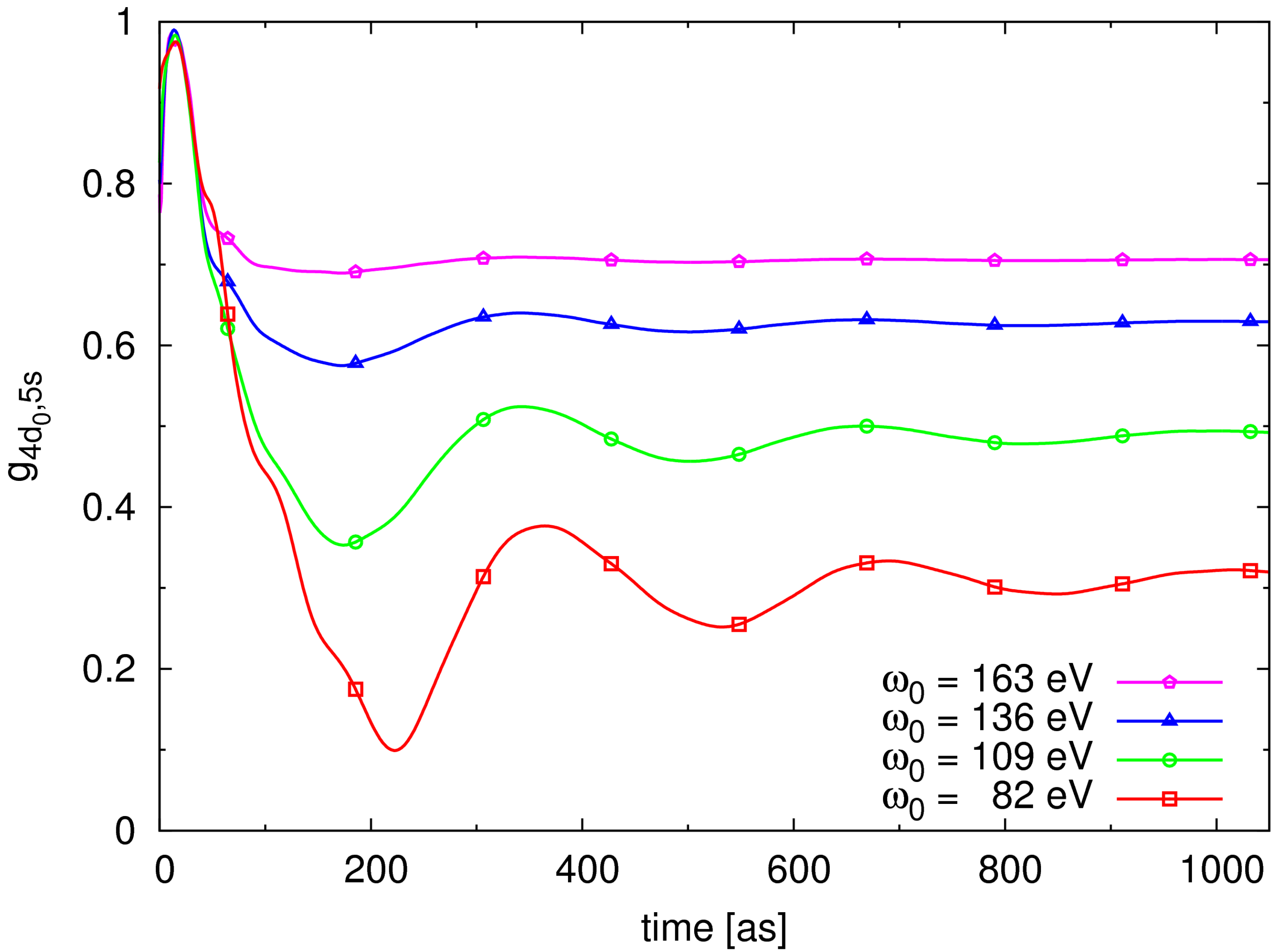
Time evolution of the degree of coherence (full theory)



Photon energy 136 eV
I(5s) ≈ 20 eV
I(4d) ≈ 70 eV

S. Pabst *et al.*, Phys. Rev. Lett. **106**, 053003 (2011)

Time evolution of the degree of coherence (full theory)



S. Pabst *et al.*, Phys. Rev. Lett. **106**, 053003 (2011)

Pulse duration 20 as

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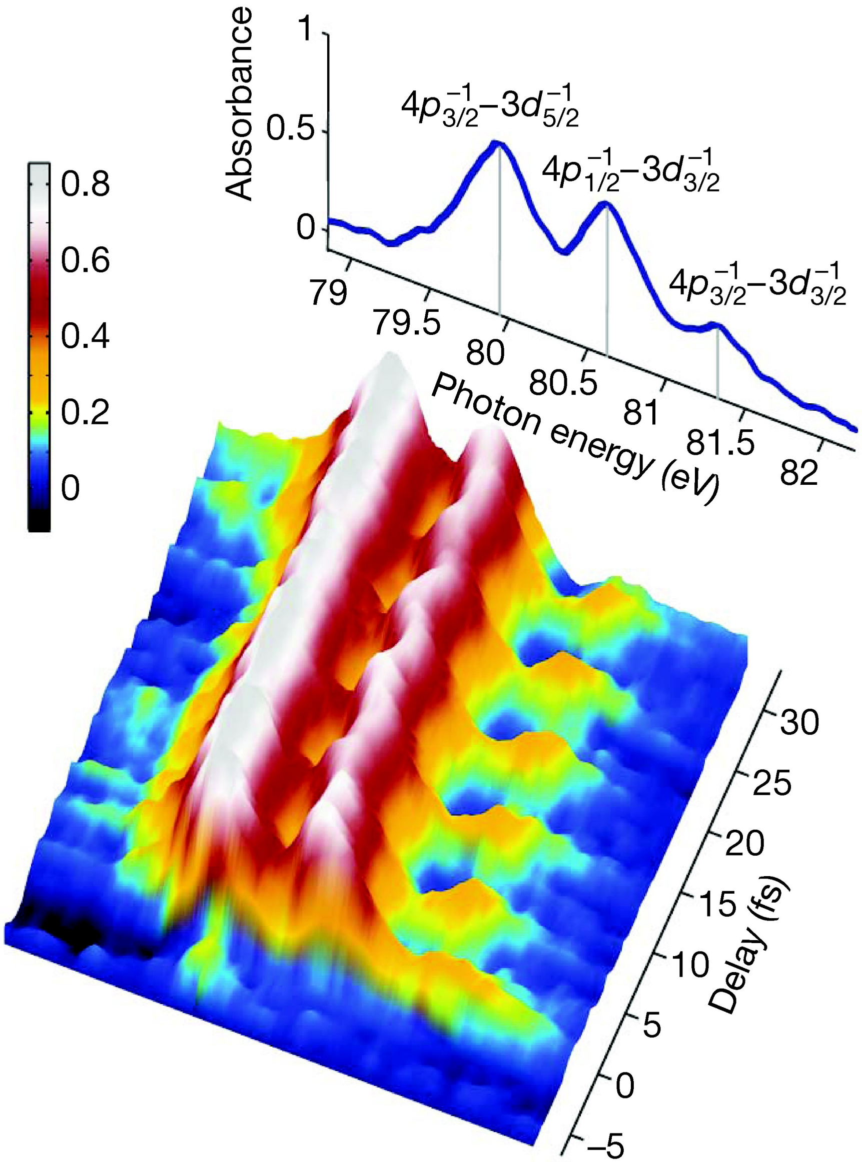
Attosecond transient absorption spectroscopy

Experiment
on krypton



E. Goulielmakis *et al.*,
Nature **466**, 739 (2010)

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Imaging of electronic quantum motion

Time-resolved x-ray scattering theory

If x rays could be treated as a classical field, then one would expect

$$\frac{dP}{d\Omega} = \frac{dP_e}{d\Omega}\left|\int d^3x\ \rho(\mathbf{x},t)\ e^{i\mathbf{Q}\cdot\mathbf{x}}\right|^2$$

$$= \frac{dP_e}{d\Omega}\int d^3x \int d^3x' \left\langle \Psi,t \middle| \hat{n}\left(\mathbf{x}'\right) \middle| \Psi,t \right\rangle \left\langle \Psi,t \middle| \hat{n}\left(\mathbf{x}\right) \middle| \Psi,t \right\rangle e^{i\mathbf{Q}\cdot(\mathbf{x}-\mathbf{x}')}$$

Quantum electrodynamics, on the other hand, predicts

$$\frac{dP}{d\Omega} = \frac{dP_e}{d\Omega}\int d^3x \int d^3x' \left\langle \Psi,t \middle| \ \hat{n}\left(\mathbf{x}'\right)\ C(\hat{H})\ \hat{n}\left(\mathbf{x}\right)\ \middle| \Psi,t \right\rangle e^{i\mathbf{Q}\cdot(\mathbf{x}-\mathbf{x}')}$$

$$C(\hat{H}) = \frac{\tau\ \Delta E}{\hbar\sqrt{\pi\ 8\ln 2}}\exp\left(-\frac{\tau^2}{8\ln 2\ \hbar^2}(\hat{H}-\langle\hat{H}\rangle_t)^2\right)$$

G. Dixit, O. Vendrell, and R. Santra, PNAS, in press

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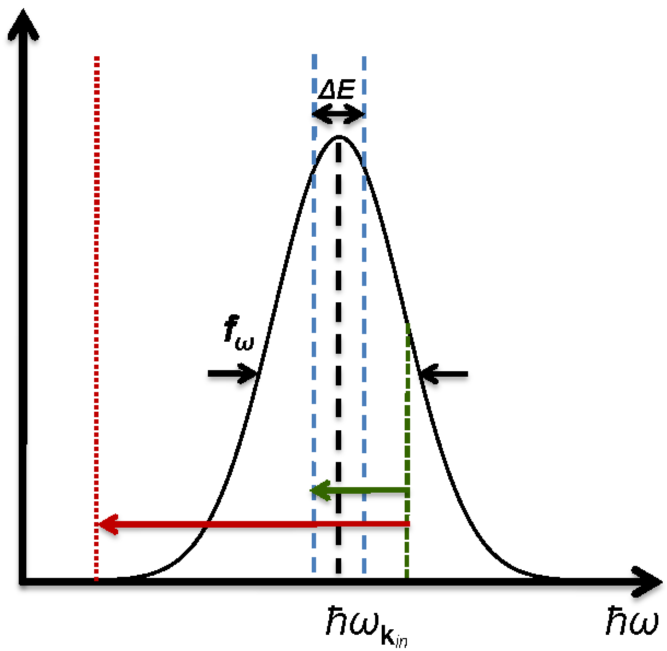
Key idea



- Probing dynamics requires a finite spectral bandwidth.
- As a consequence of this bandwidth, it becomes impossible to decide whether the detected photon was scattered elastically or inelastically.
- This problem persists even if the scattering detector has perfect energy resolution.

G. Dixit, O. Vendrell, and R. Santra, PNAS, in press

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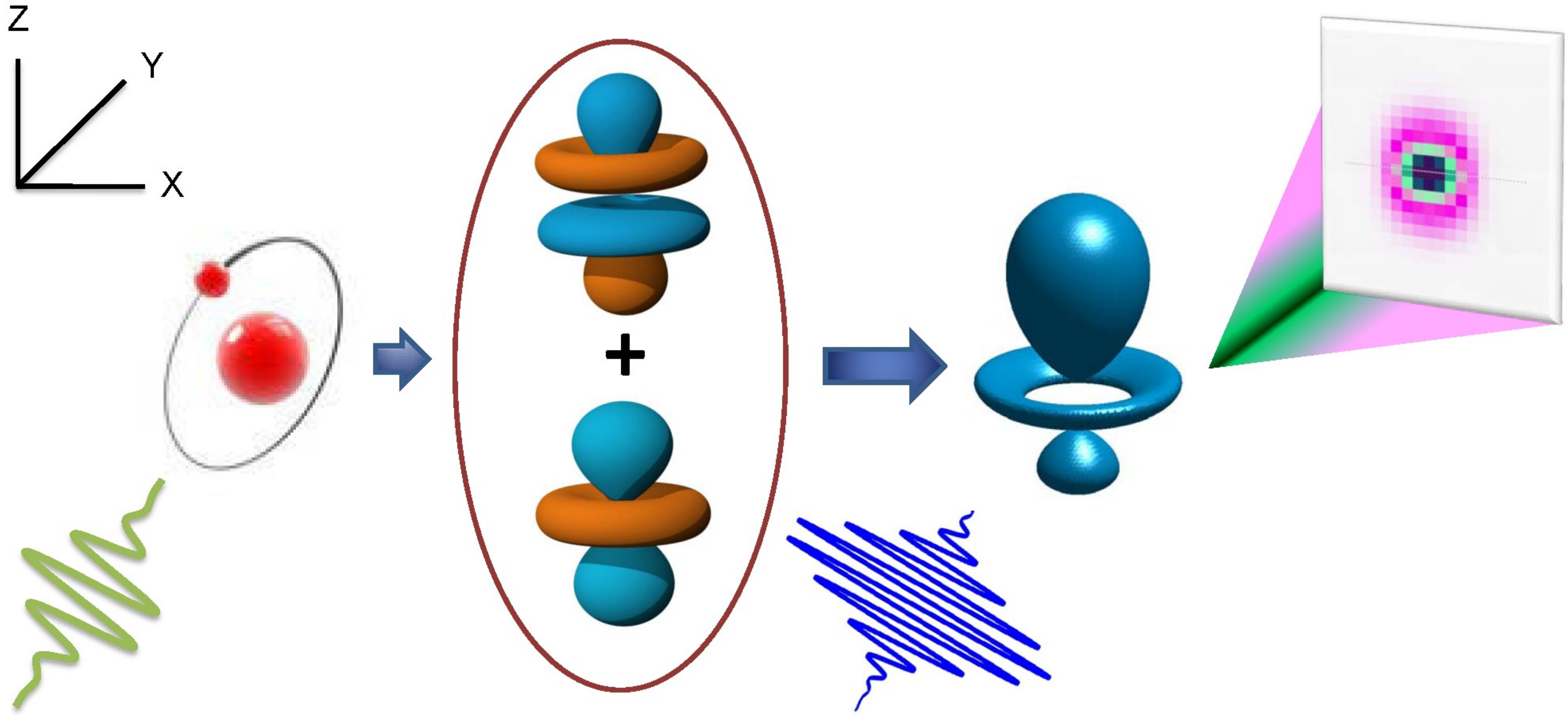
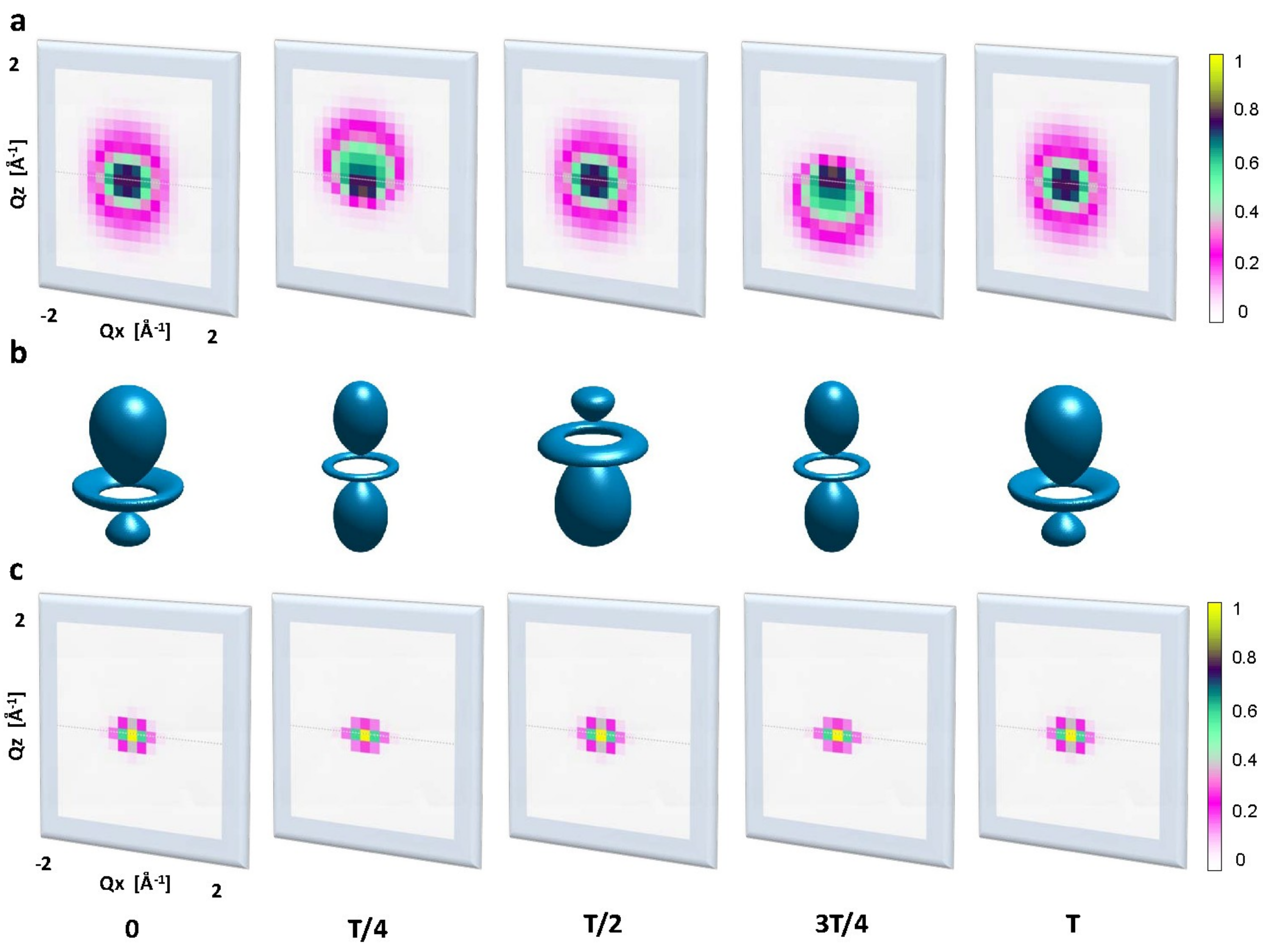
Example



G. Dixit, O. Vendrell, and R. Santra, PNAS, in press

QED vs. semiclassical theory



T = 6.25 fs

G. Dixit, O. Vendrell, and R. Santra, PNAS, in press

Conclusions

X-ray free-electron lasers are great.

If they produced shorter, more coherent pulses, they would be even greater!