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Title: Prospects for strong field control beyond the single active electron model

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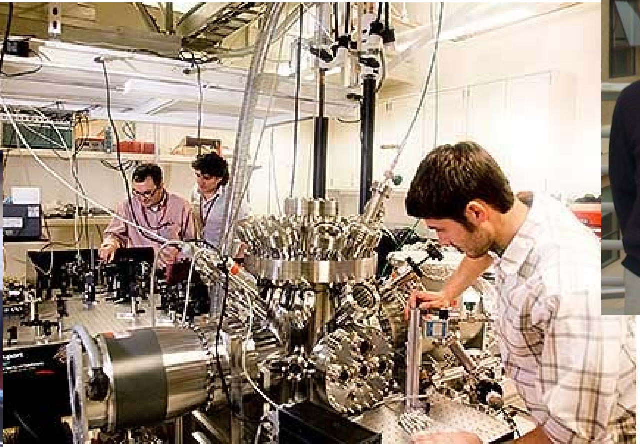
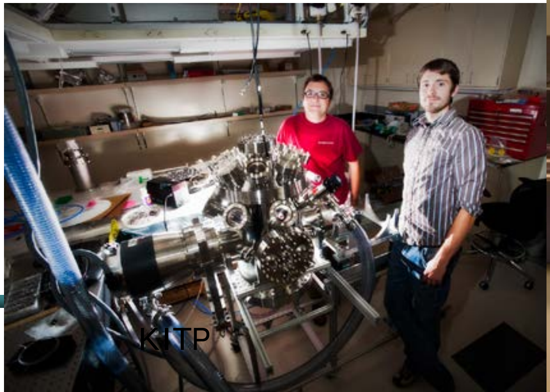
KITP, March 20, 2013, Quantum Control Program

Collaborators

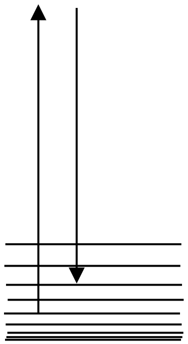




- Bucksbaum group (including alumni) : Adi Natan, James Cryan, Mike Glownia, Doug Broege, Andrei Kamalov, Julien Devin, James White, Song Wang, Jaehee Kim, Limor Spector, Joe Farrell, Brian McFarland, Vlad Petrovic, Lucas Zipp, Ian Tenney, Chelsea Liekhus-Schmaltz, Markus Guehr, Ryan Coffee

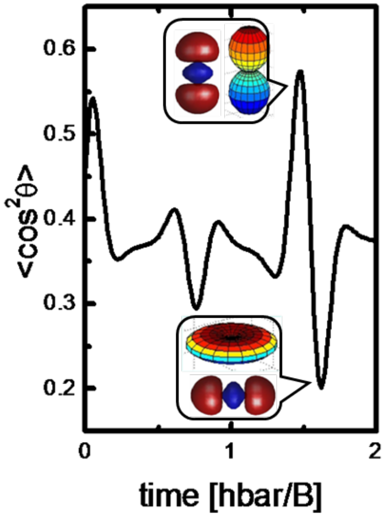
- Experimental groups contributing to our LCLS work: Nora Berrah et al, Kalamazoo; Lou Dimauro et al, OSU; Jon Marangos, Imperial College; Linda Young et al, ANL; Hamed Merdji, CEA Saclay; Raimund Feifel, Uppsala; Abbas Ourmazd et al., Milwaukee; AMO and Laser group, LCLS; PULSE Institute: Bucksbaum group, Guehr group; Reis group; Bogan group.
- Theory contributions: Tamar Seideman group, Northwestern; Alejandro Saenz group, Berlin; Martinez group, PULSE Institute




Coherent quantum control is already a practical tool in strong field physics.



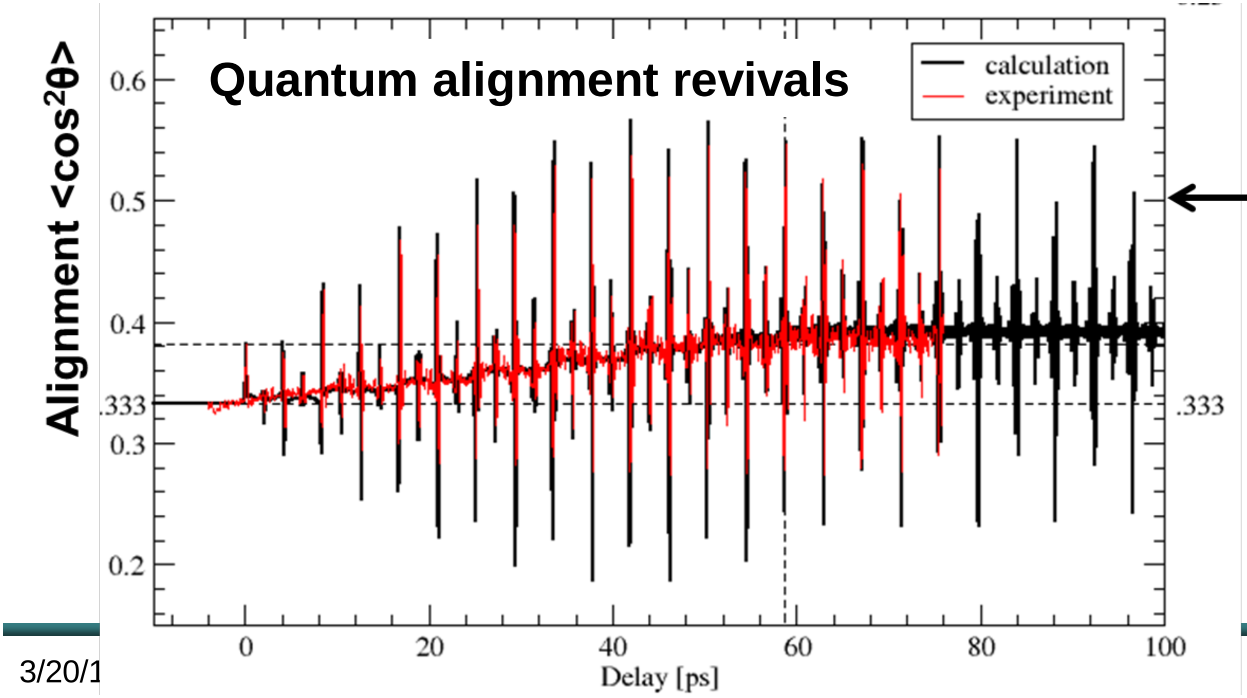
$\tau_{pump} < \tau_{rotation}$



Molecular Alignment

$U = -1/4\, \Delta\alpha\, E^2 \cos^2\theta$

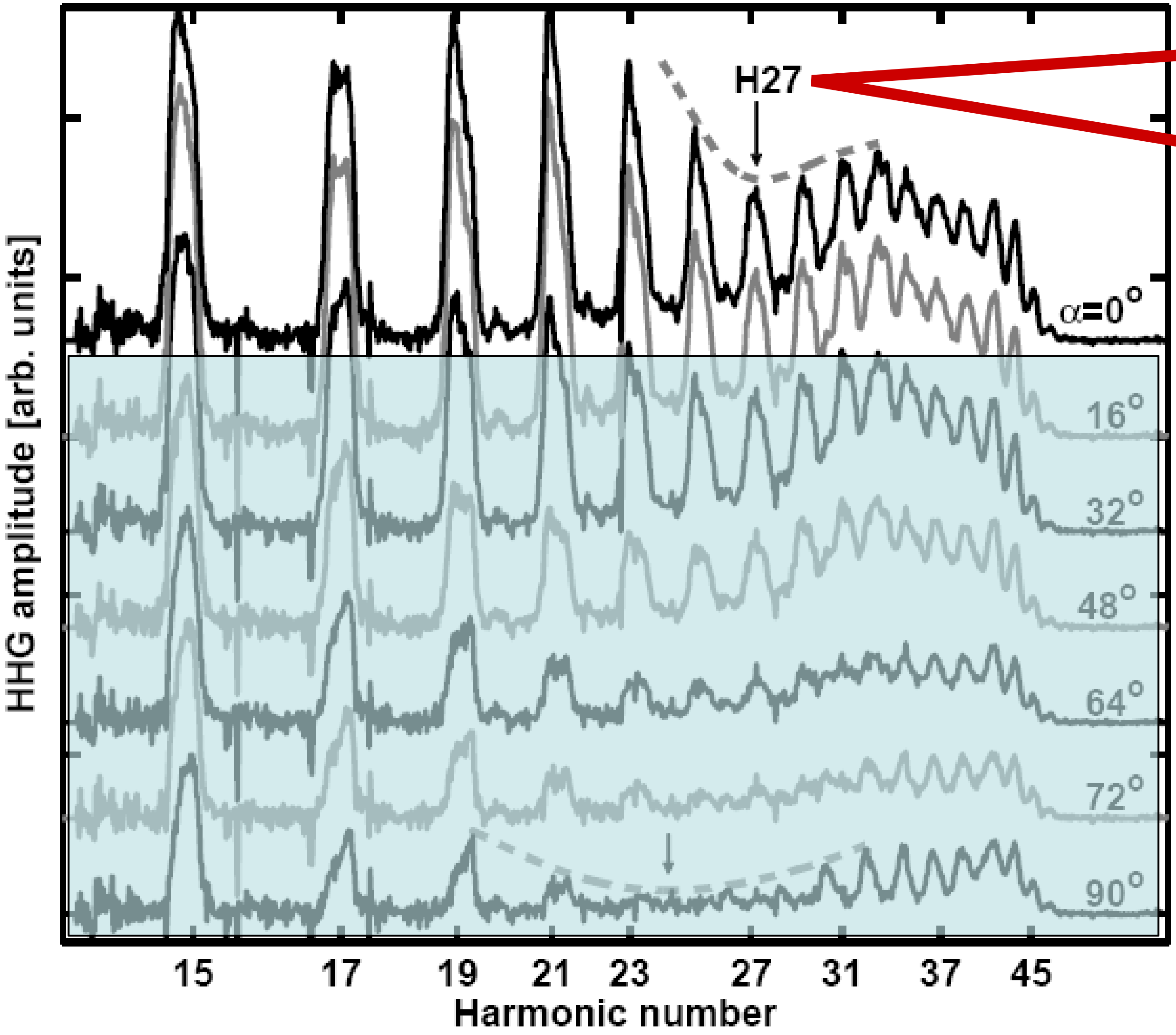
Stapelfeld, Seideman, Averbukh, others

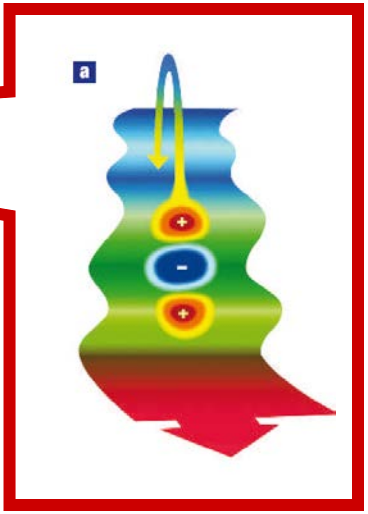


Shaped pulse solution (Eight 100 fs kicks over 64 ps) to optimize alignment

Cryan, et al. PRA, *80, 063412 (2009).*

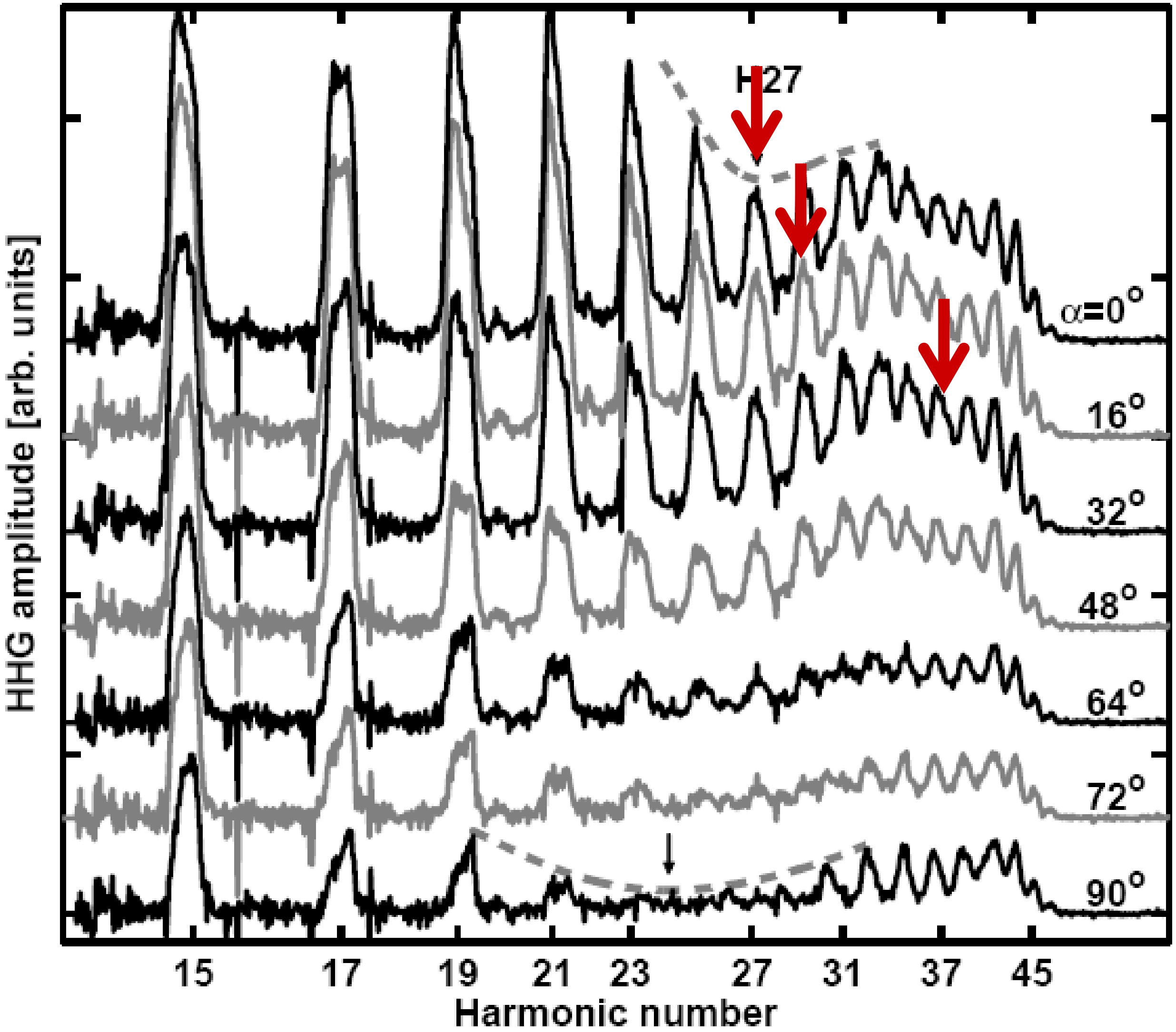
HHG example: Does structural interference cause the dip in the N_2 HHG spectrum?

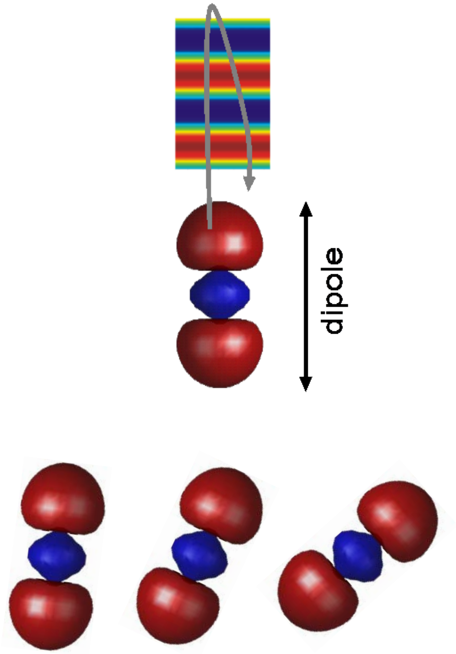




e.g. Itatani, J. *et al. Nature 432, 867–871 (2004).*

Test: The structural interference should move to higher energy with angle

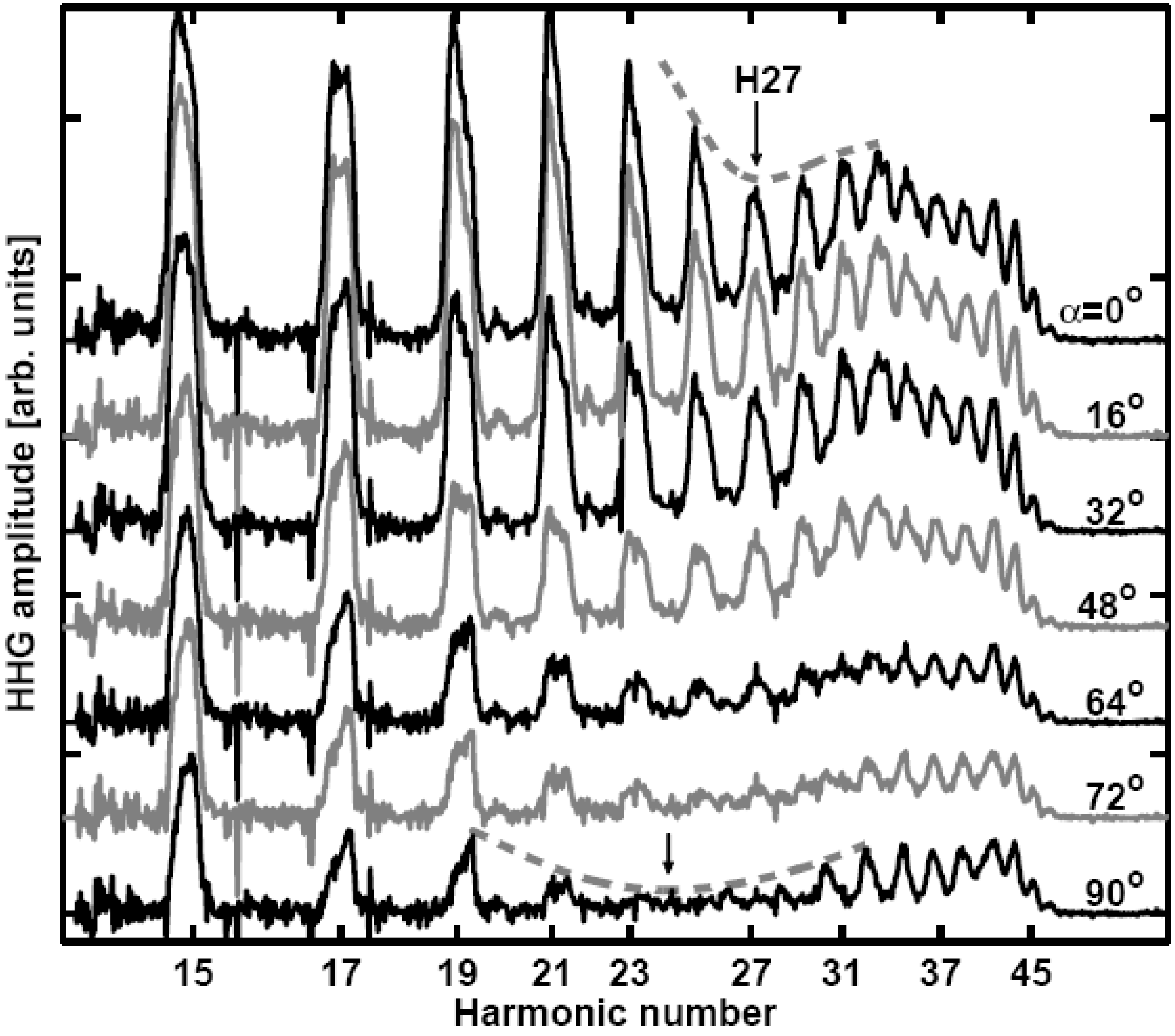


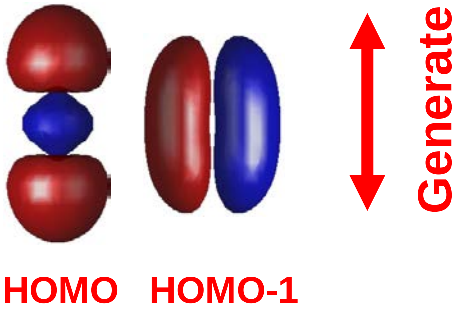


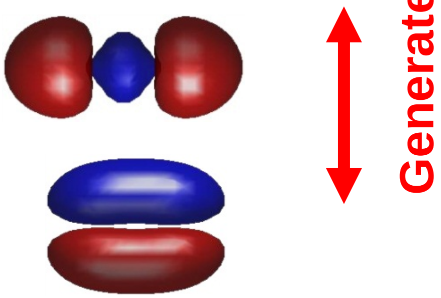
$H_{min} \sim 1/(2d\cos\theta)^2$

Farrell, J. P. et al., Chemical Physics 366(1-3): 15 (2009).

The spectrum shows interference of higher orbitals

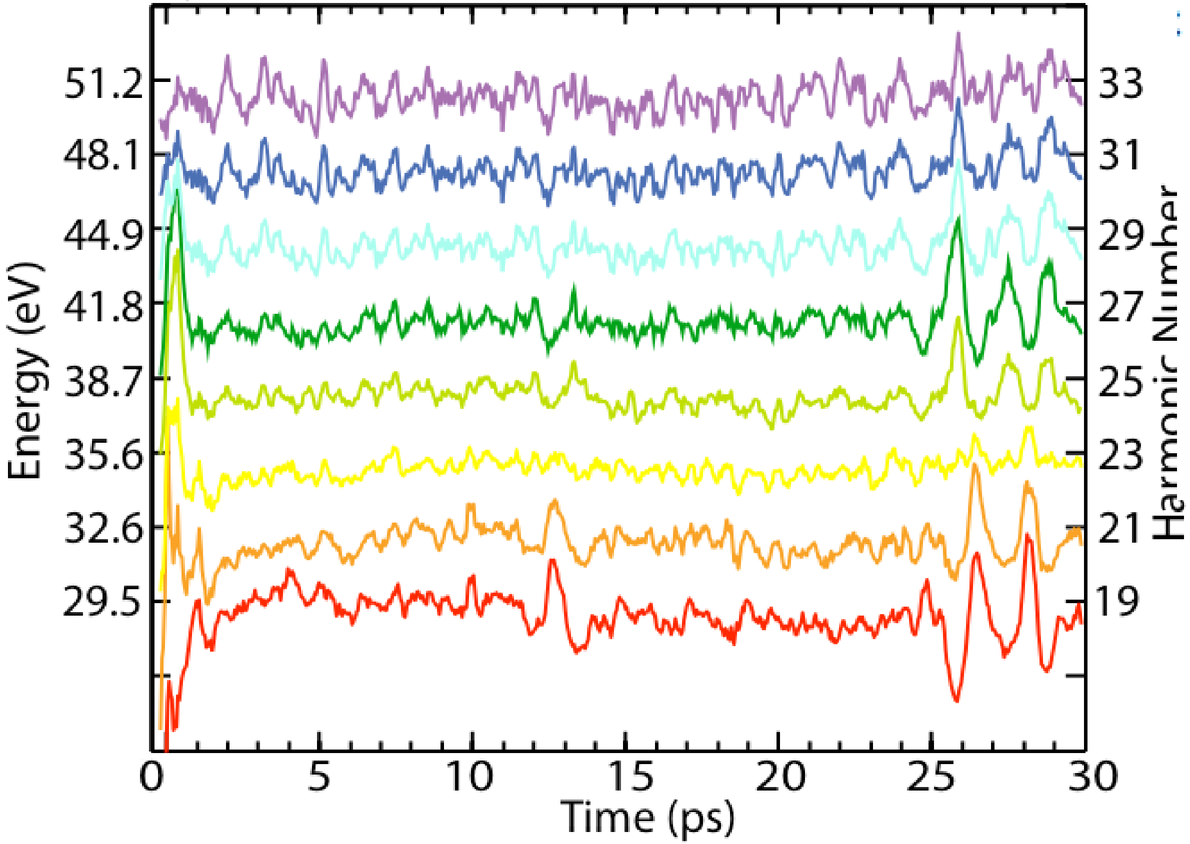


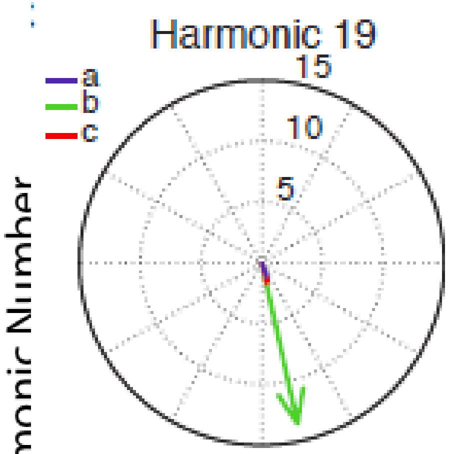


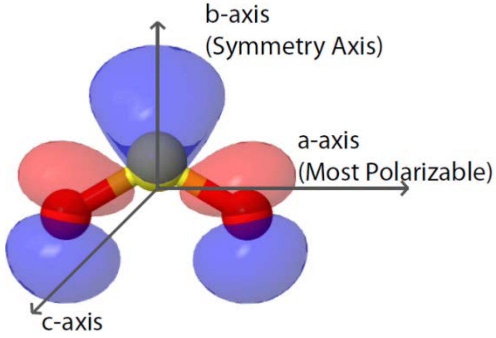


McFarland et al,
Science 322, 1232 (2008)

HHG revivals in an asymmetric top molecule, SO_2

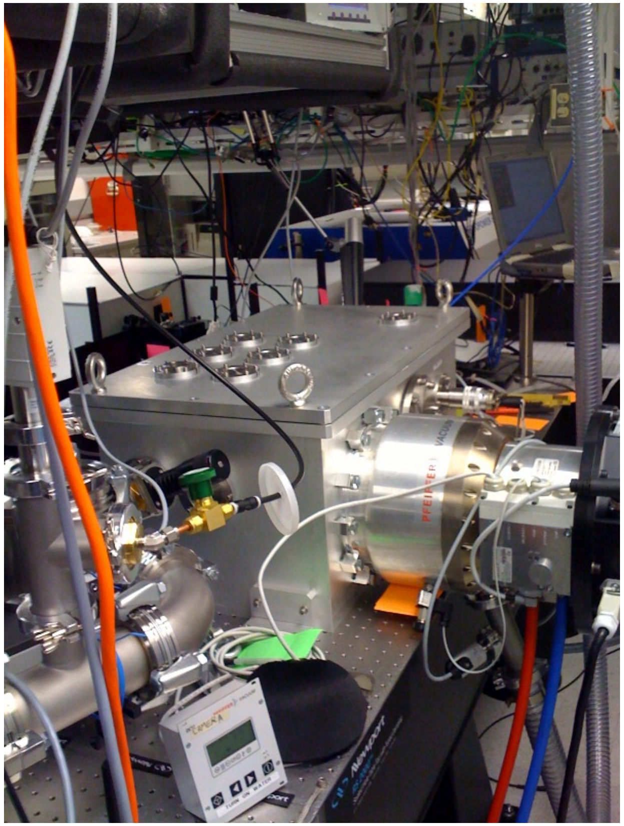


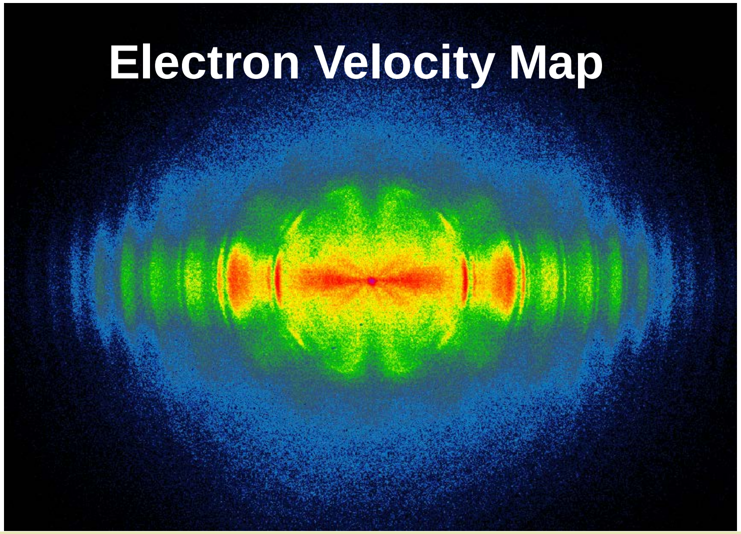




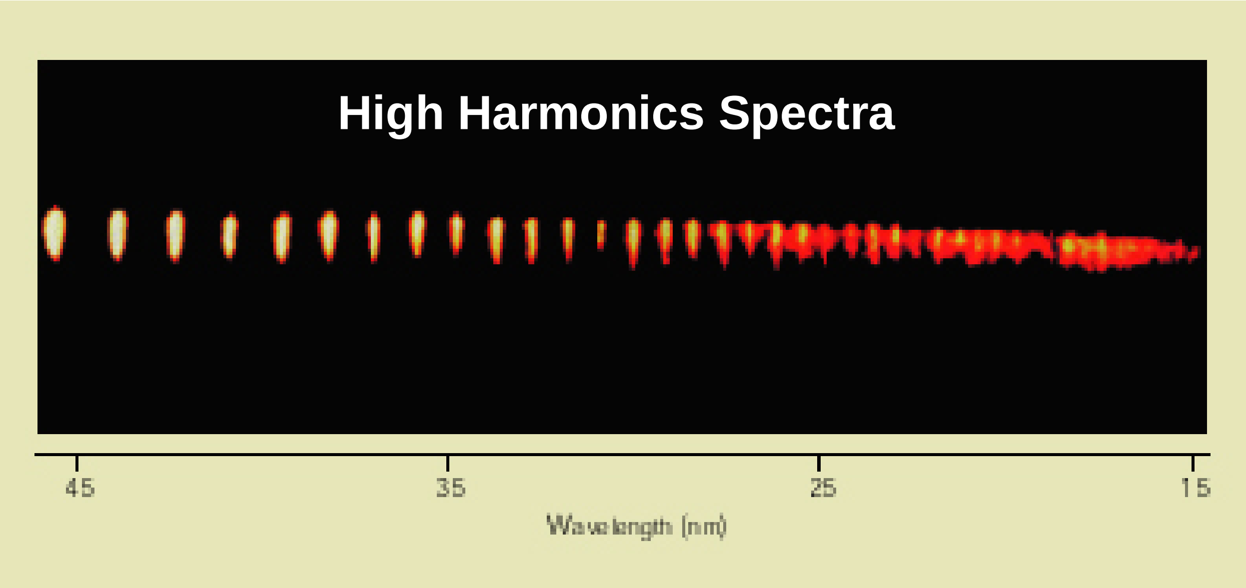
L. Spector, et al., arXiv:1207.2517v2 [physics.atom-ph]; also
L. Spector et al., work in progress with A. Saenz, et al.

Strong laser fields and bound electrons:



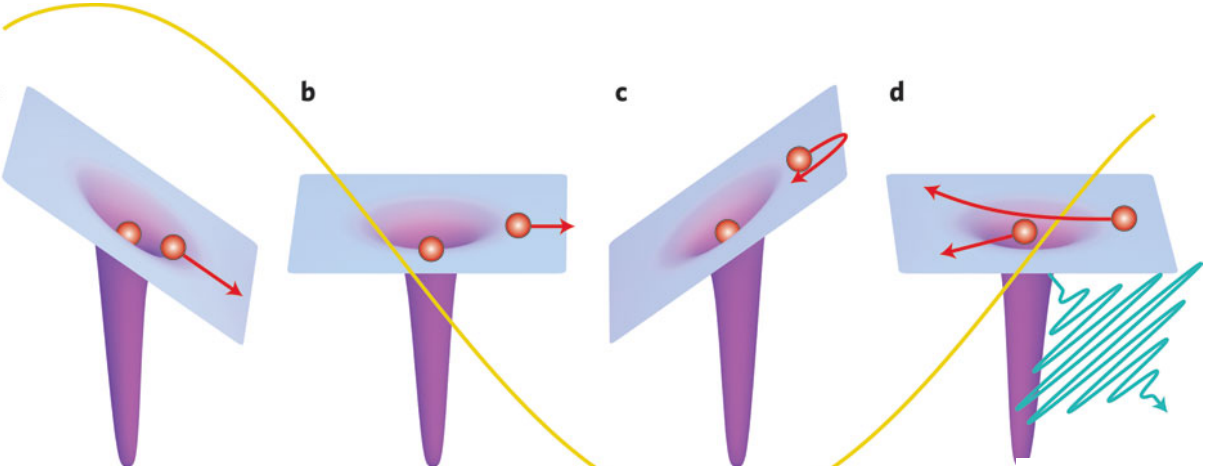


Even the raw data look beautiful.

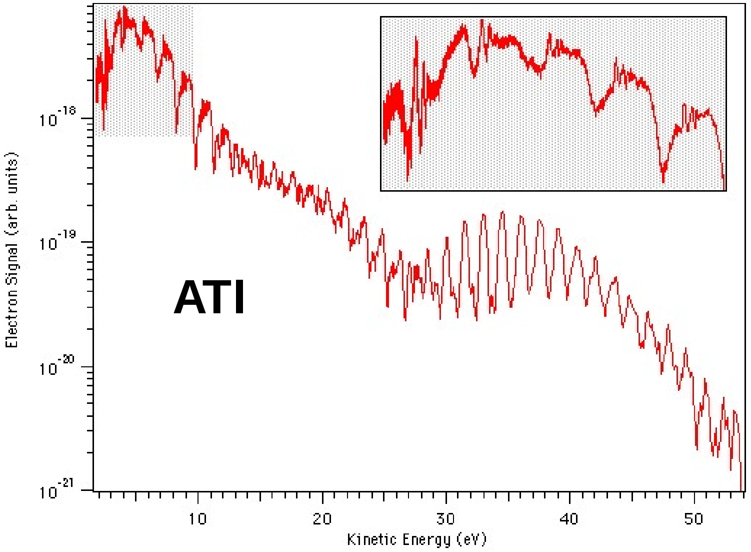


800 nm
10-1000 TW/cm^2

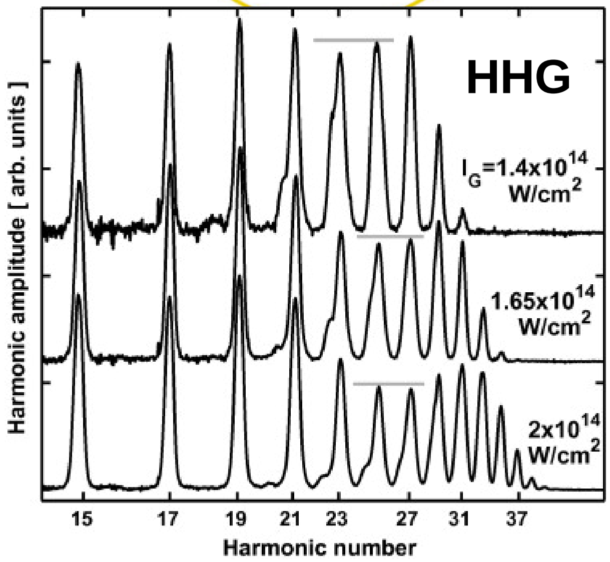
Strong field control of electrons: A single active electron theory is successful



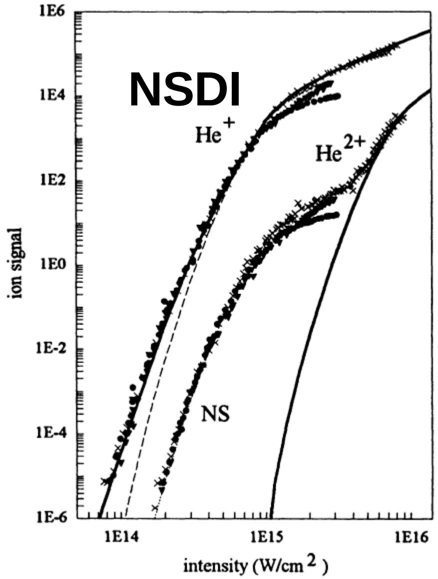
Midorikawa, Nature Photonics 5, 640–641 (2011)



(M. J. Nandor, et al. PRA 60, 1771 (99).)



J.P. Farrell, et al., Chem Phys 366 15 (09).

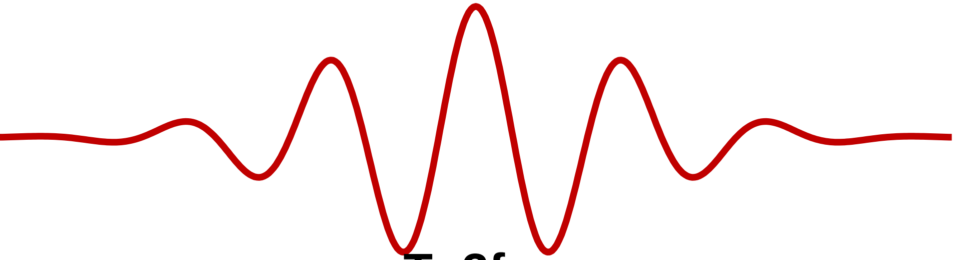


B Walker, et al. PRL 73 1227 (94)

Hypothesis: Transient state strong field physics may break the single active electron paradigm

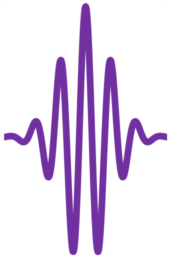
- **Excited molecules are highly dynamic multi-particle systems.**
- **Strong fields can couple to the multi-electron dynamics in molecules in ways that are easily observed.**
- **Example 1: Strong field interactions with molecules in transition**
- **Example 2: Strong field interactions with core-excited molecules at an x-ray free electron laser**

Laser Lab: hv=1.5 eV



T=3fs

LCLS: hv=1500 eV



T=3as

UV-induced ring opening, accompanied by strong field fragmentation

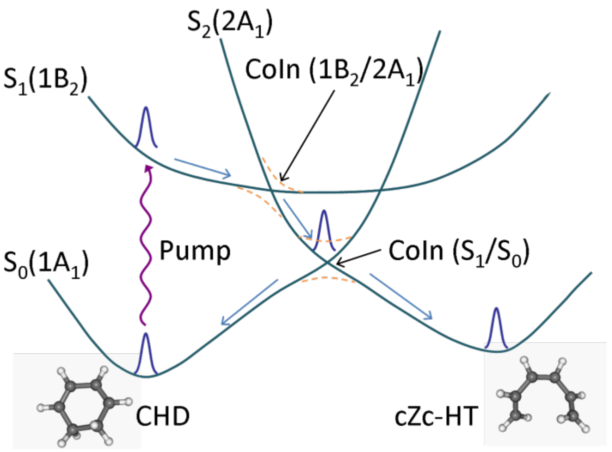
Experimental setup:

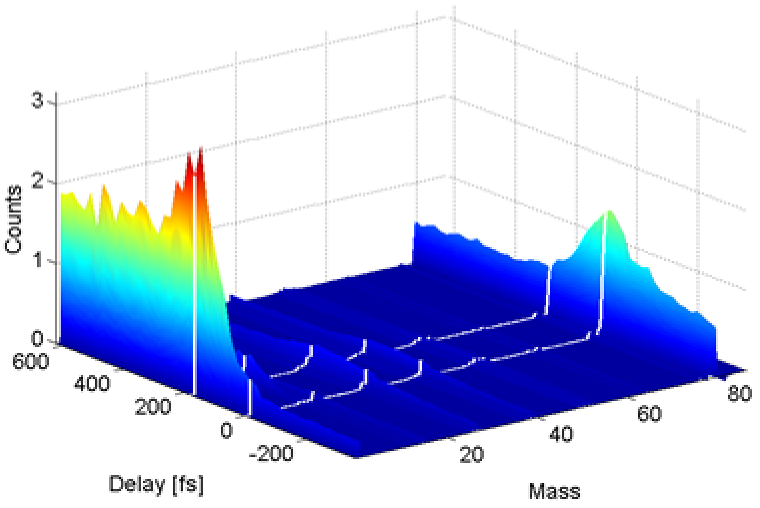
- S0→S1 induced by 266 nm pulses
- 800 nm laser focused to 10^{14} W/cm^2 induces ionization, photofragmentation

Analysis:

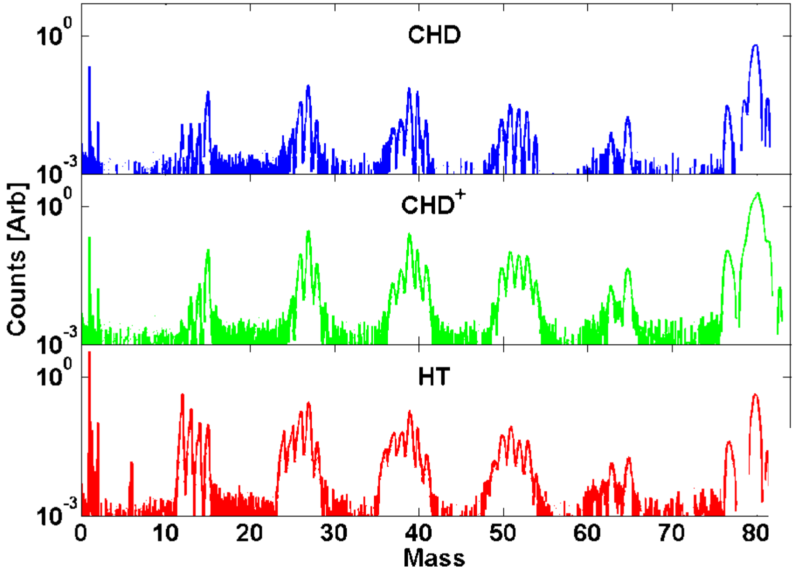
- Photofragment patterns reveal the state of the ring-opening process
- Coulomb explosion energies are sensitive to multiple ionization

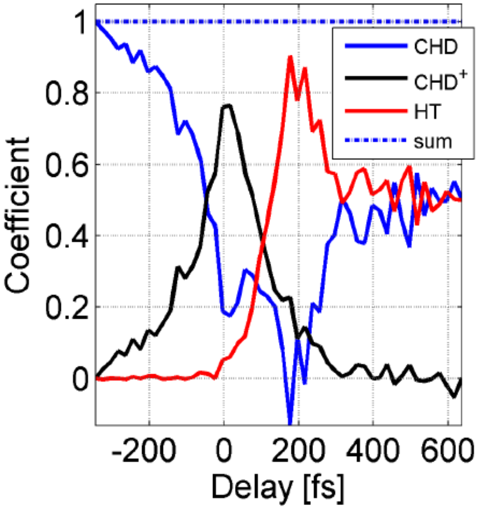
J. L. White, et al., JCP 136, 054303 (2012).





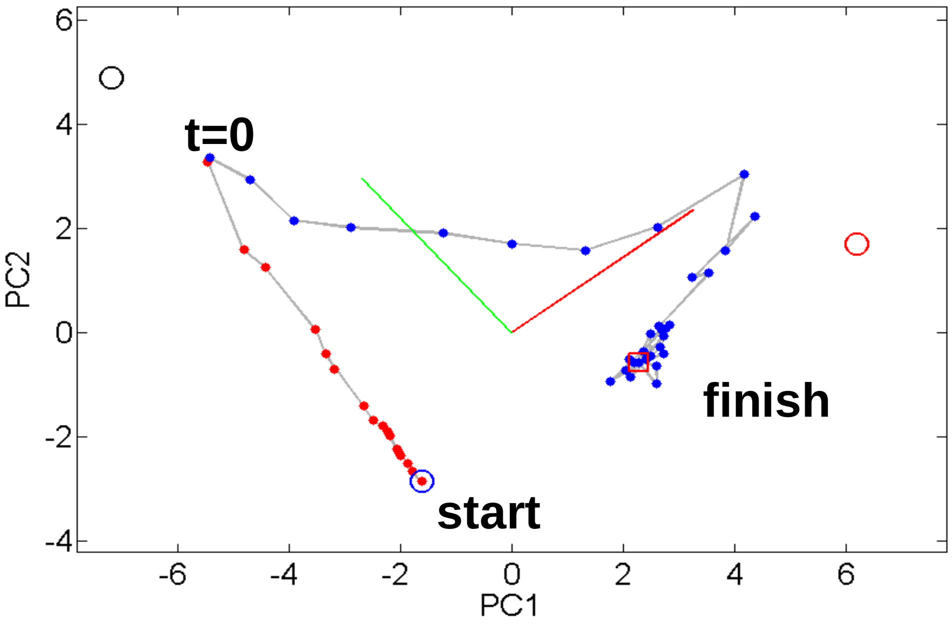
Tracing the ring opening using patterns of fragmentation





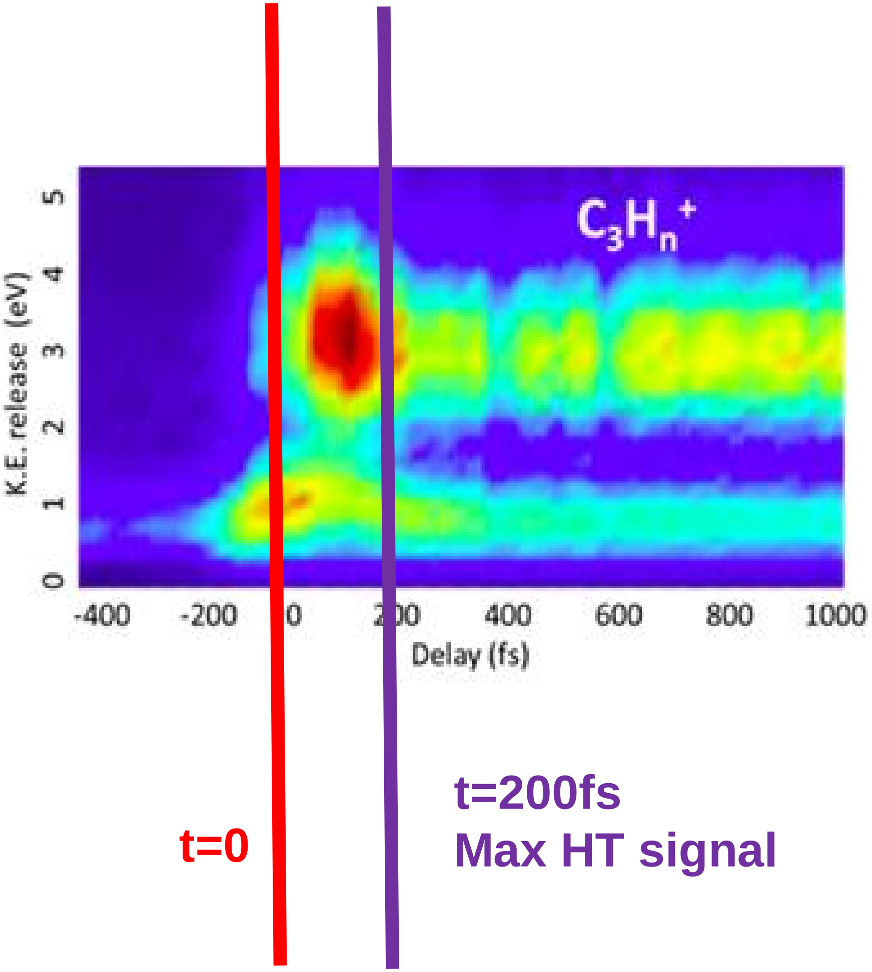
Principal component analysis shows that only three orthogonal spectral patterns account for 90% of the fragmentation.

Apparent excess in the HT channel at 200 fs



J. L. White, et al., JCP 136, 054303 (2012).

Kinetic energy analysis shows that the increased signal is correlated with higher charge states



t=0

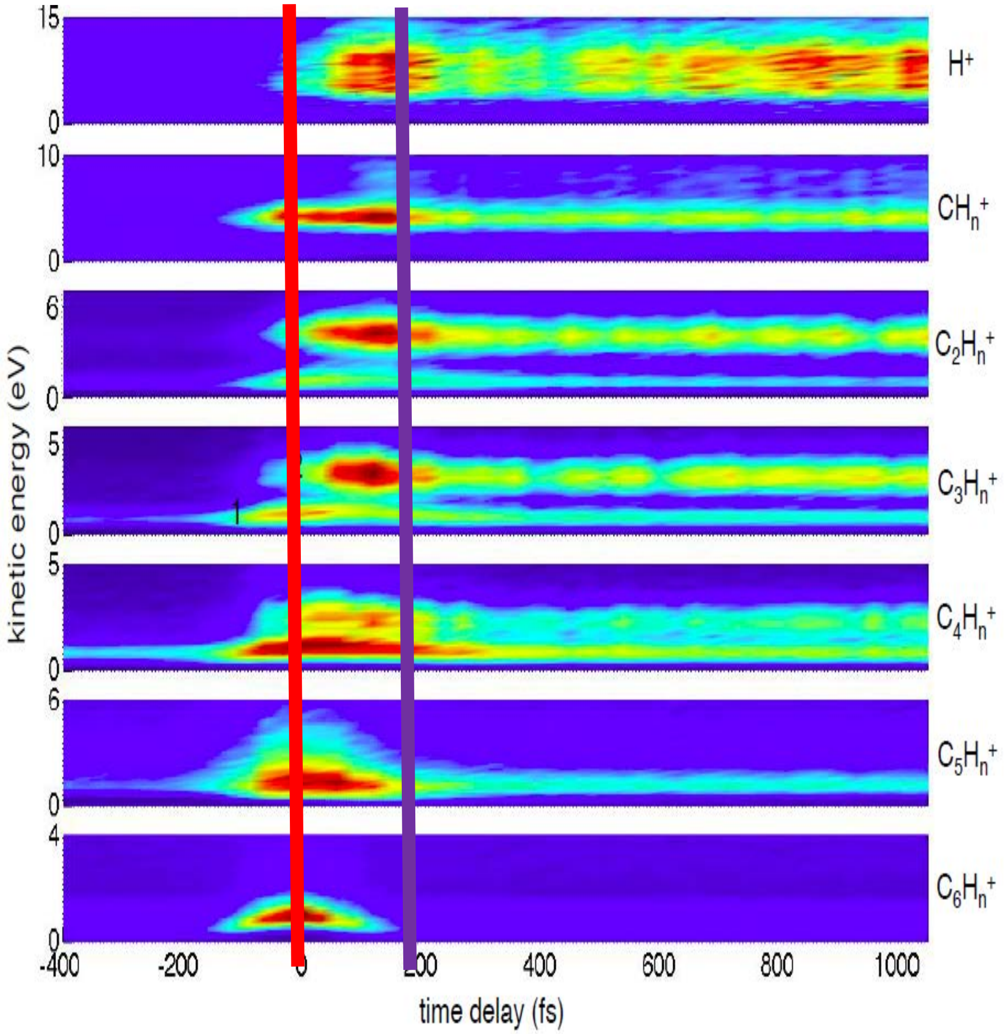
t=200fs
Max HT signal

$AB^{++} \rightarrow A^+ + B^+$

$AB^+ \rightarrow A^+ + B$

Bucksbaum and Petrovic,
Faraday Discussion 163

Kinetic energy analysis shows that the excess HT may be correlated with higher charge states

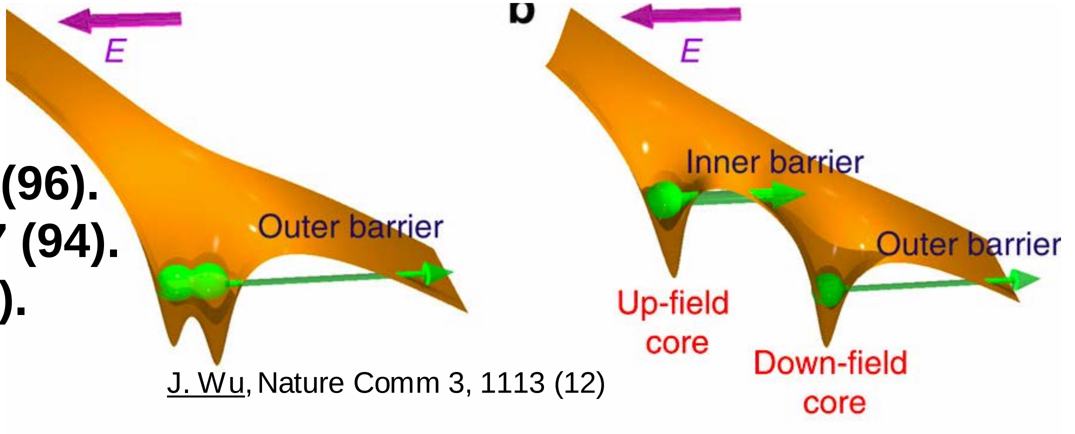


The strong laser field induces multiple ionization during the ring-opening.

Petrovic et al., arXiv:1212.3728 [physics.chem-ph]

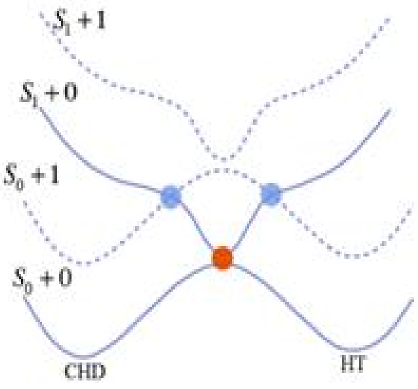
Possible mechanisms: enhanced ionization, quantum resonance rings

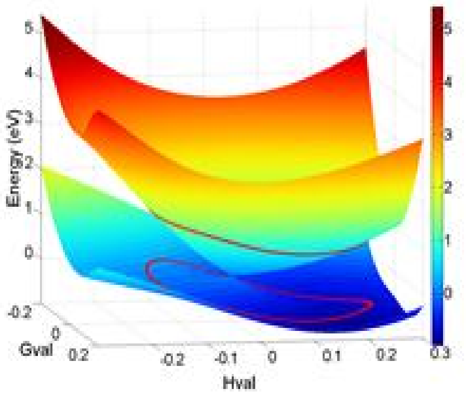
<u>Enhanced ionization</u>
E. Constant et al. PRL 76, 4140 (96).
M. Schmidt, et al., PRA 50, 5037 (94).
W. T. Hill et al. PRL 69, 2646 (92).



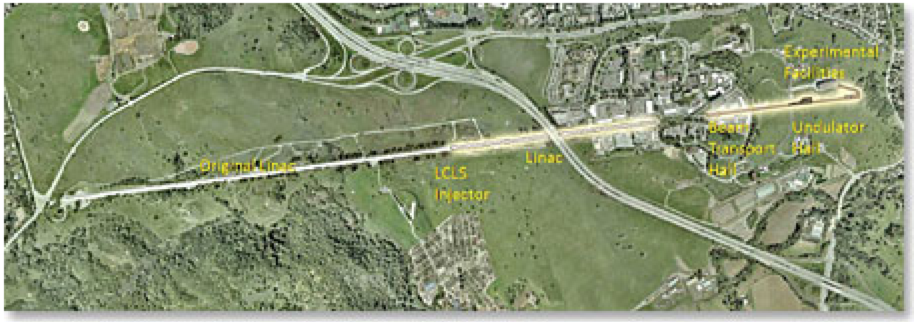
J. Wu, Nature Comm 3, 1113 (12)

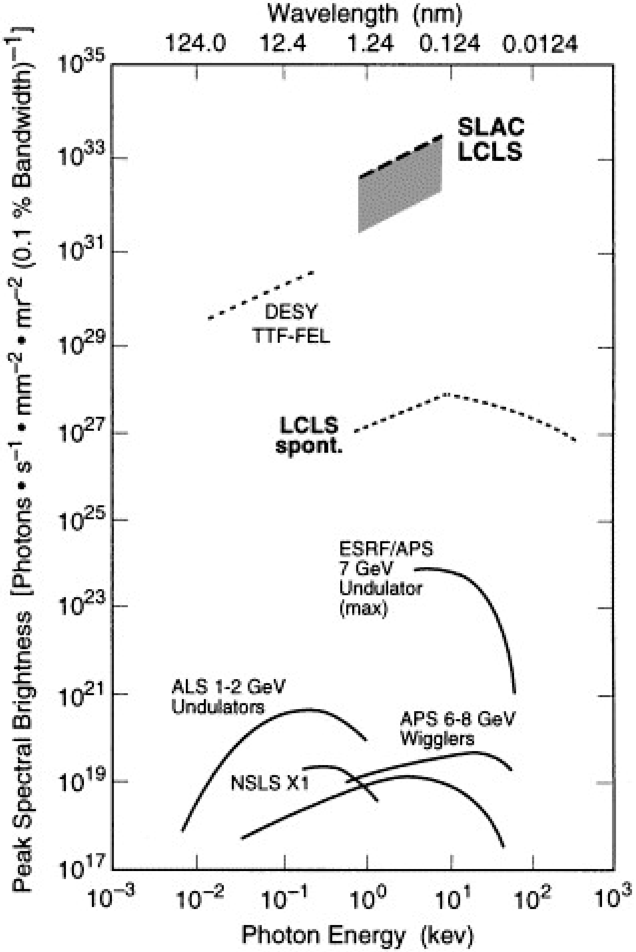
<u>Quantum resonance rings</u>
Kim, et al.
J. Kim, J. Phys. Chem A 116 2758 (11).





X-ray Free Electron Laser (LCLS)

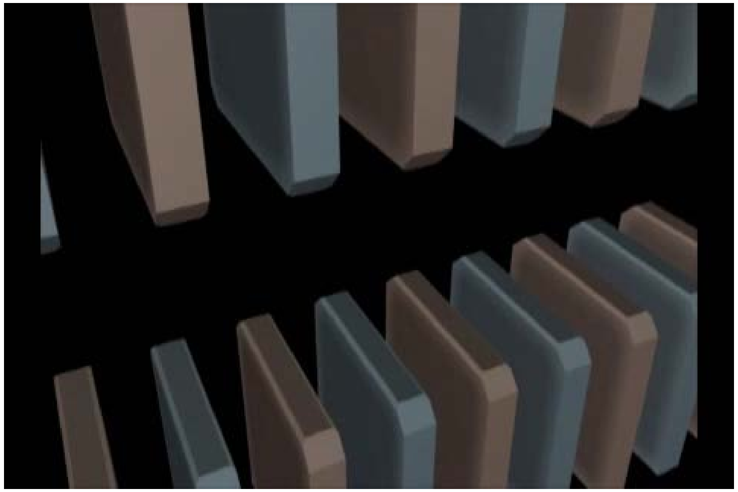




Pulse energies: millijoules

Strong field x-ray source: LCLS

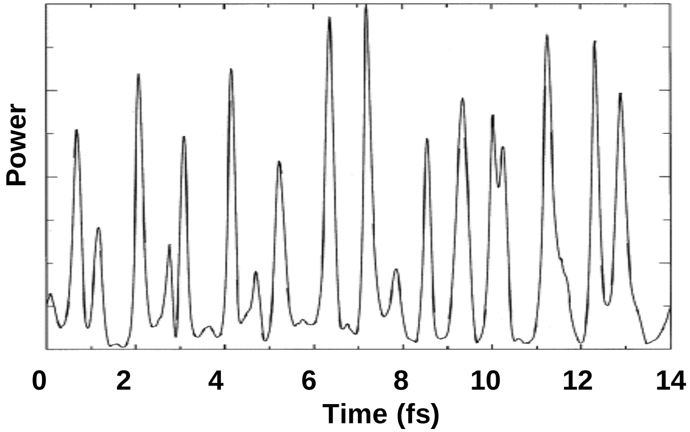
**Relativistic electrons
in strong magnetic fields.**

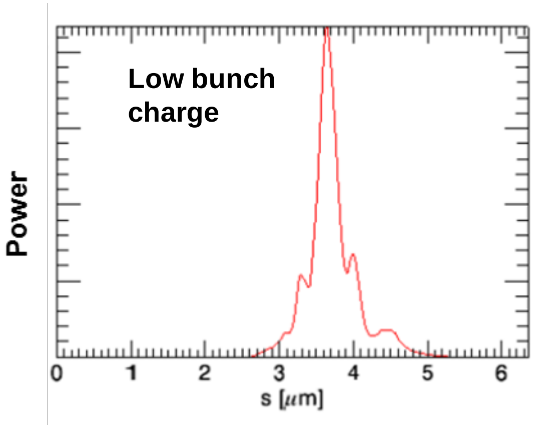


- Electron energy= γmc^2
- Wavelength = λ
- Undulator period =λ_u
- Maximum deviation angle of the electrons = K/γ

$$\lambda_{FEL} = \frac{\lambda_u}{2\gamma^2}\left(1+\frac{K^2}{2}\right) \sim 0.1-4nm$$

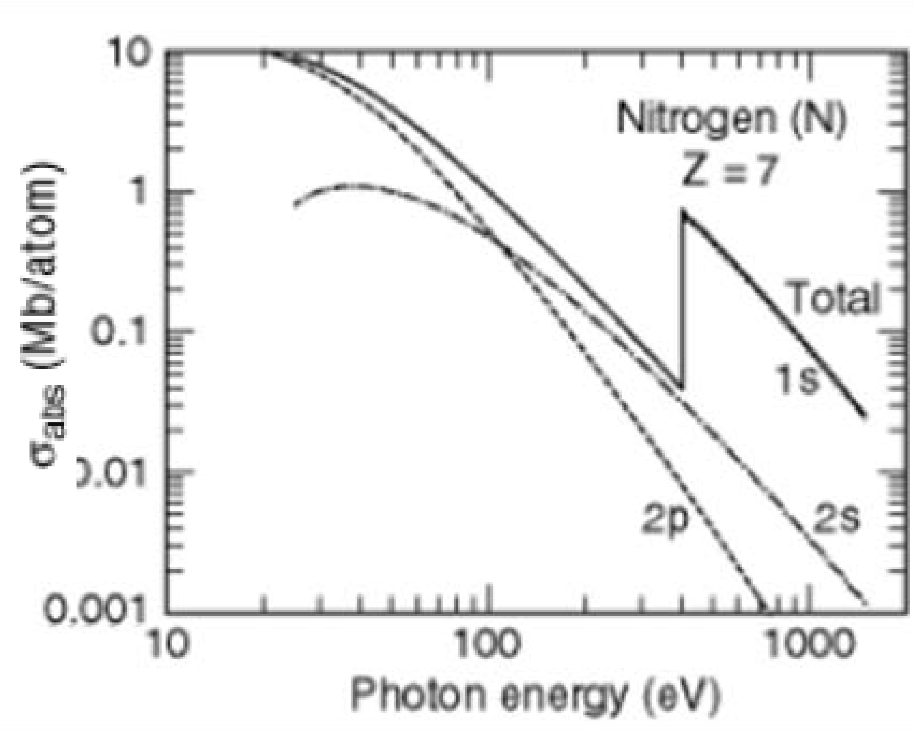
Sub-fs spikes, SASE

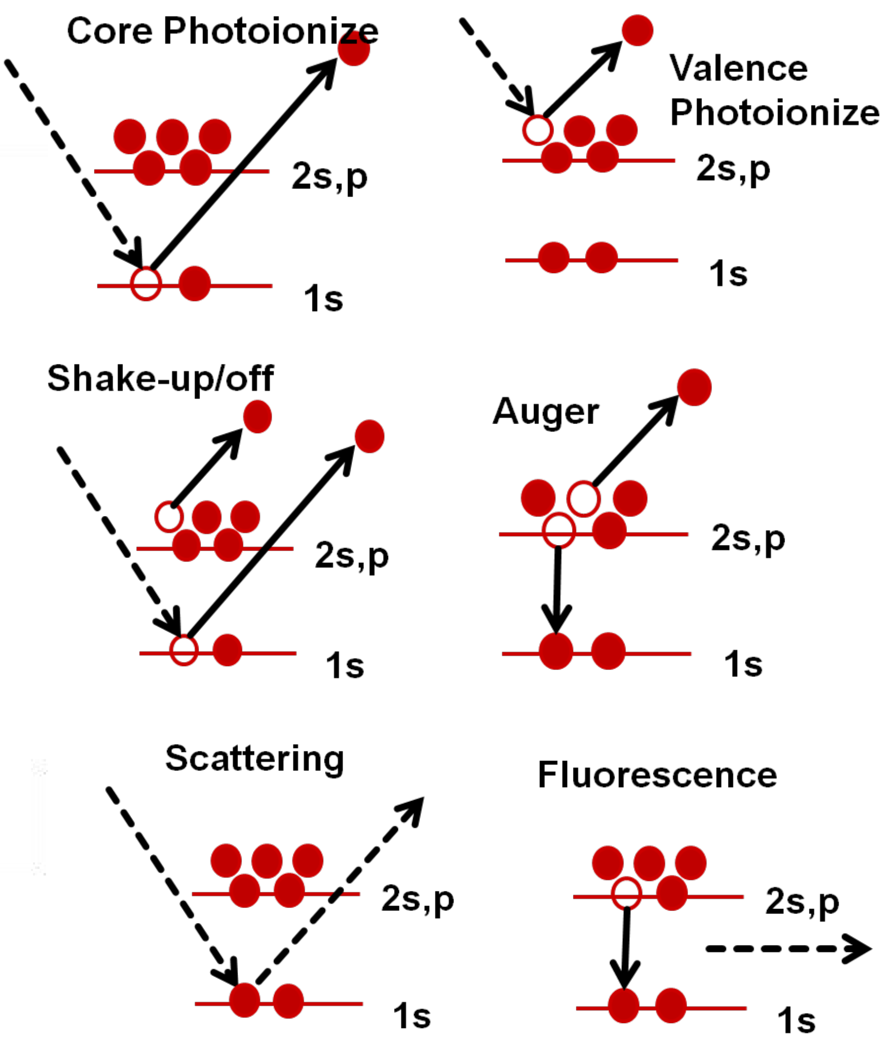




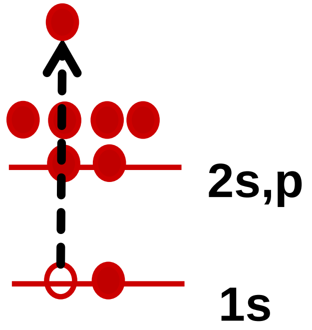
Nuhn, NIM A 429, 249 (1999)
Y. Ding, PRL 102, 254801(2009)

X-ray interactions with atoms





Strong fields: 1-photon, 1-electron ionization



photoionization

consider a 1-photon K-shell transition:

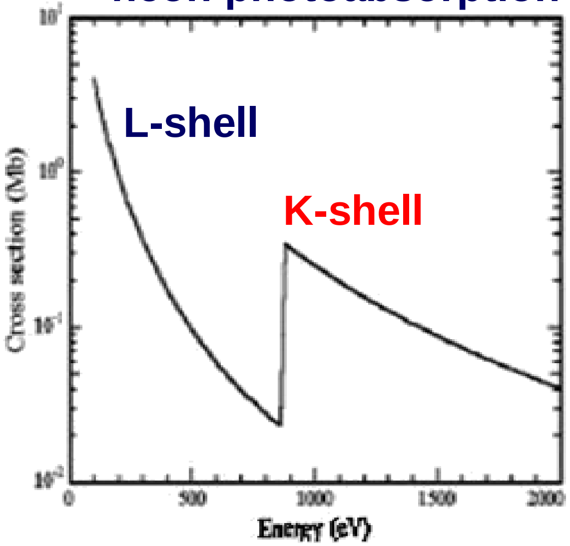
$\sigma_K \approx 10^{-18}\ \mathrm{cm}^2$

$\Gamma_K = \sigma_K F_{lcls} \approx 10^{15}\ \mathrm{s}^{-1}$ **(*saturated*)**

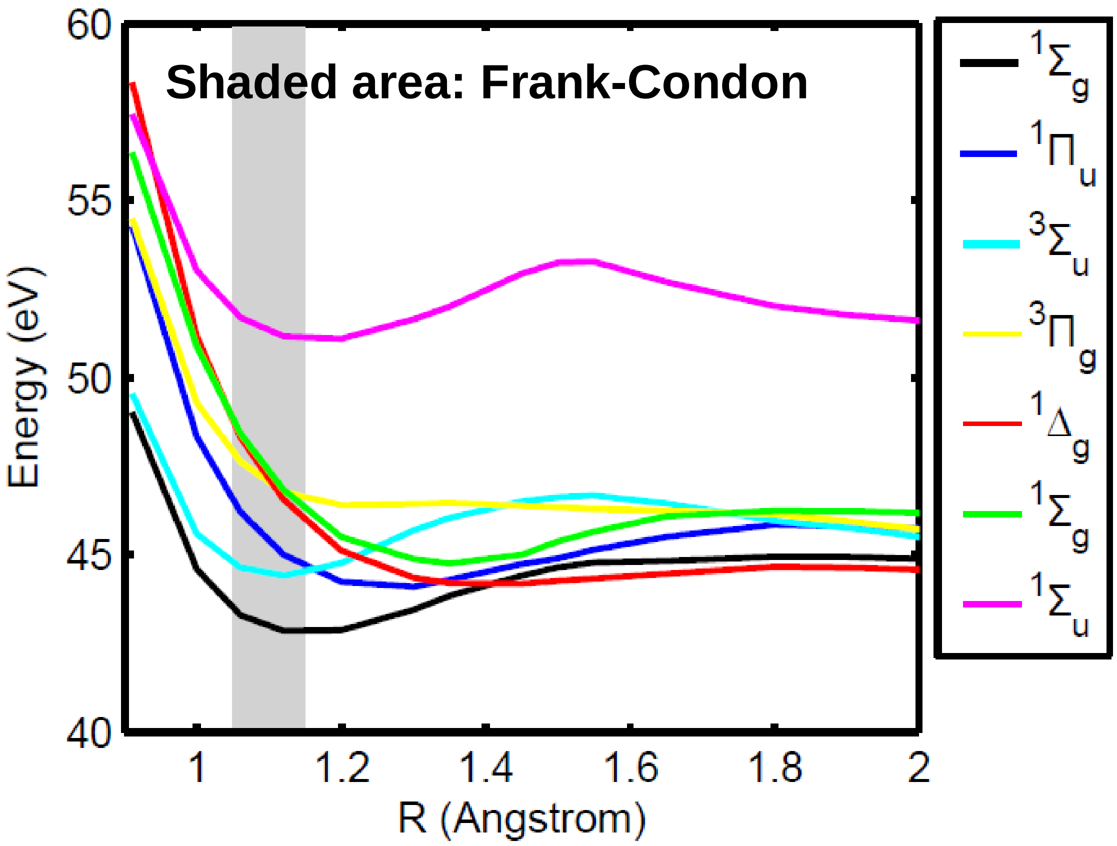
$t_K = 1/\,\Gamma_K = 1\ \mathrm{fs}$

- **rapid enough to ionize more than one electron.**
- **fast enough to compete with atomic relaxation.**

neon photoabsorption

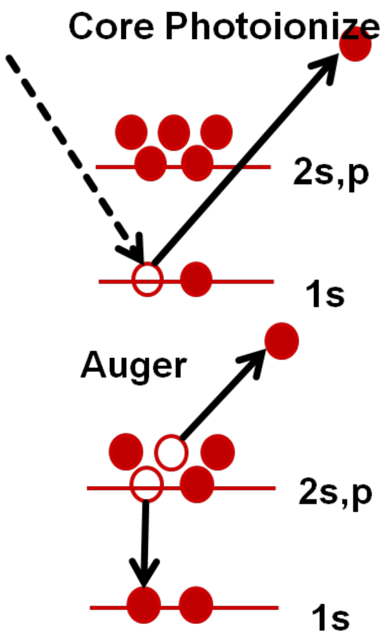


Transient state studies: N_2^{++} Potential Energy Surfaces



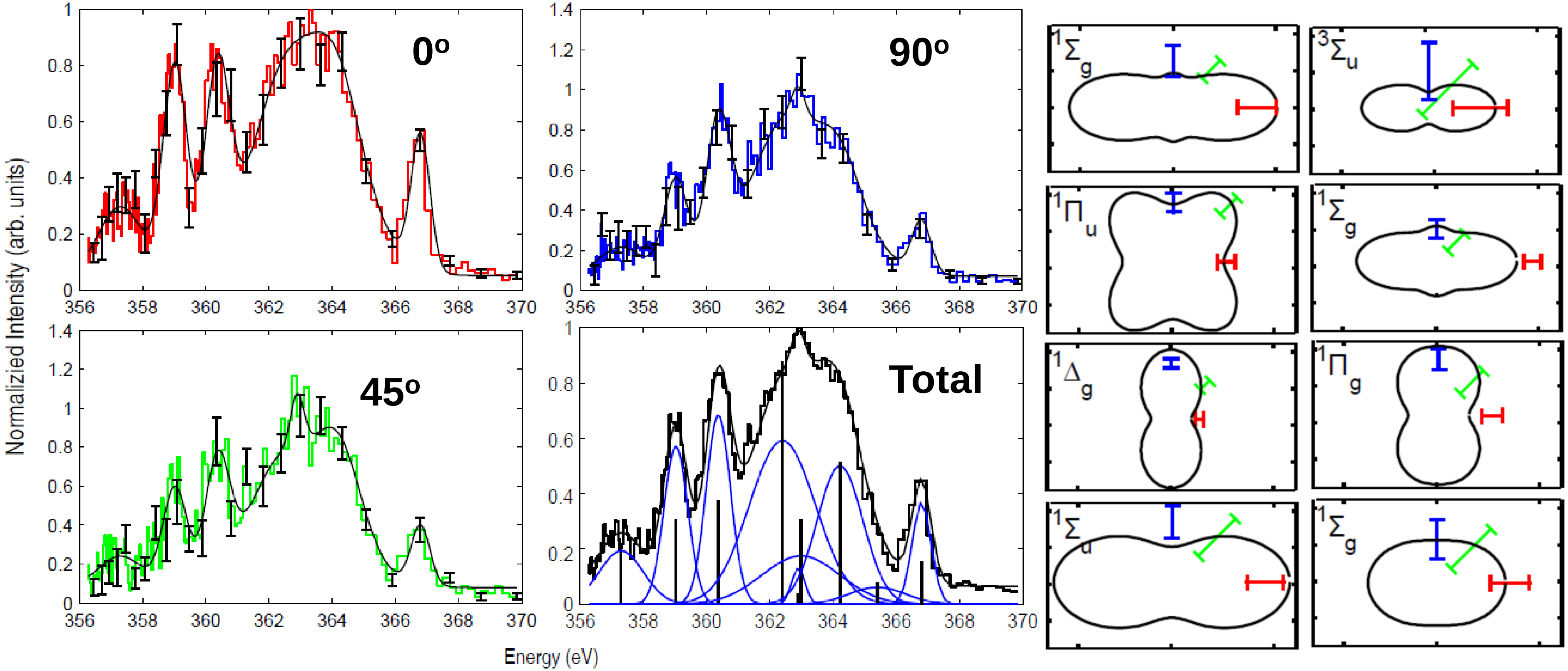
R. W. Wetmore and R. K. Boyd, J. Phys. Chem. 90, 5540 (1986).

$\Gamma_K = \sigma_K F_{lcls} \approx 10^{15}\ s^{-1}$



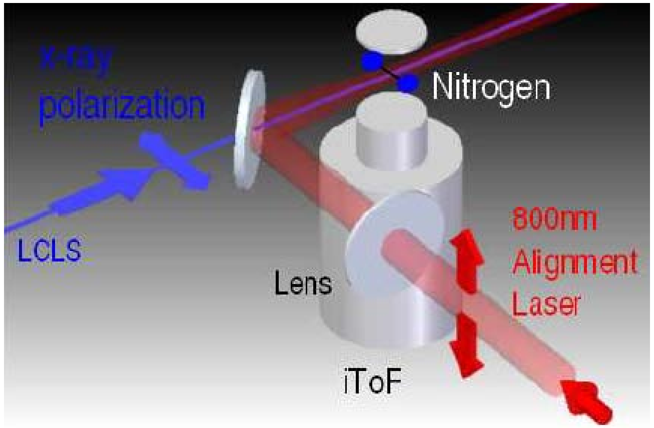
Impulsive dynamics
If Auger time
$t < h/\Delta E$

Auger electron energies observed in the molecular frame from $N_2 \rightarrow N_2^+ \rightarrow N_2^{2+}$ at 1.1keV

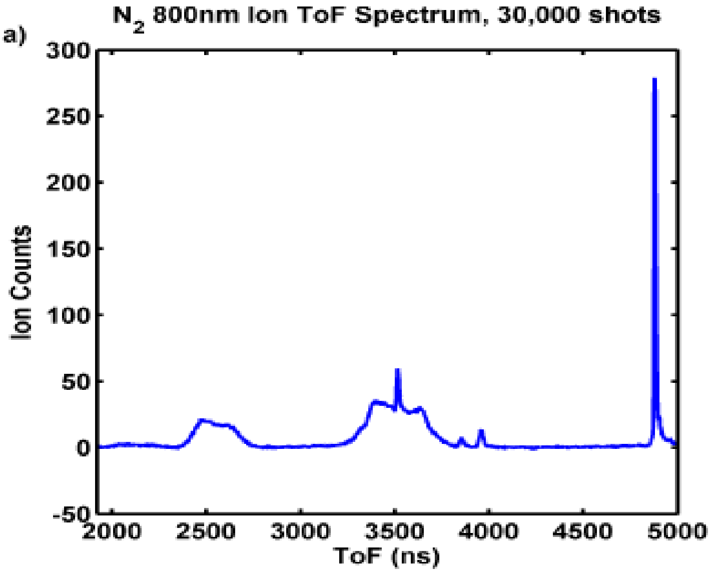


(Cryan et al, J. Phys. B 45 055601 (2012))

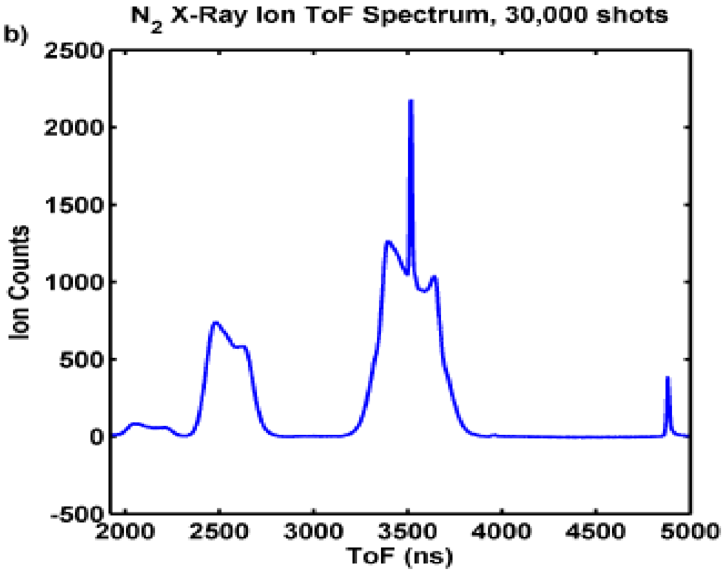
Fragmentation Time of flight spectra



800 nm

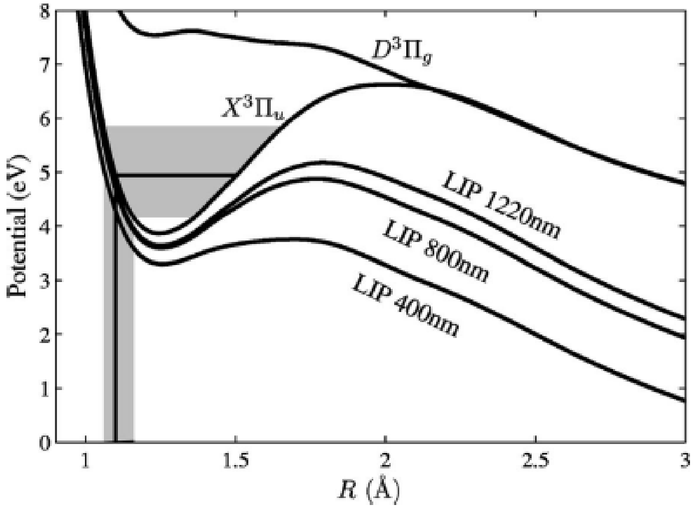


1.1 keV



M. Glownia, 2012

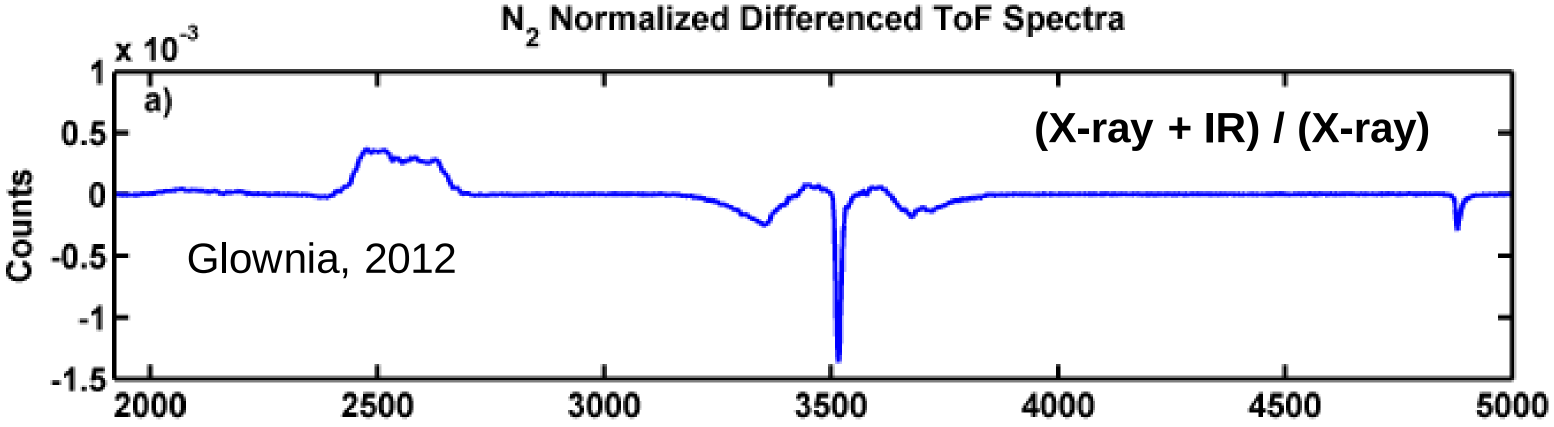
Strong field dissociation of Auger-produced dication states



R. Coffee et al., Phys. Rev. A, 73:043417 (2006).

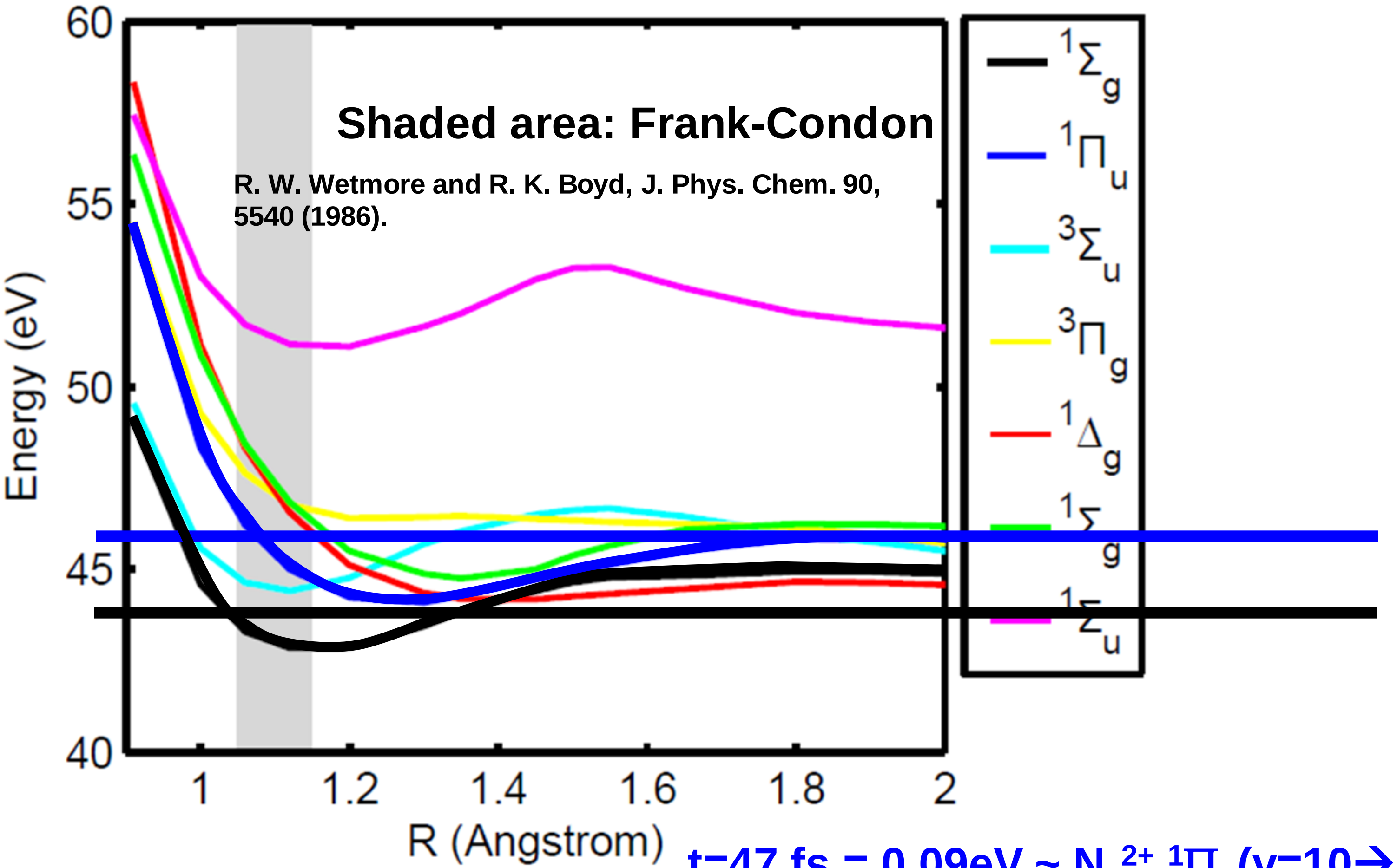
Auger-produced dication states can undergo bond softening in an intense laser "probe" field.

We also see strong evidence for laser-induced field ionization of the dication formed by Auger



Glownia, 2012

Transient state studies: N_2^{++} Potential Energy Surfaces



Shaded area: Frank-Condon

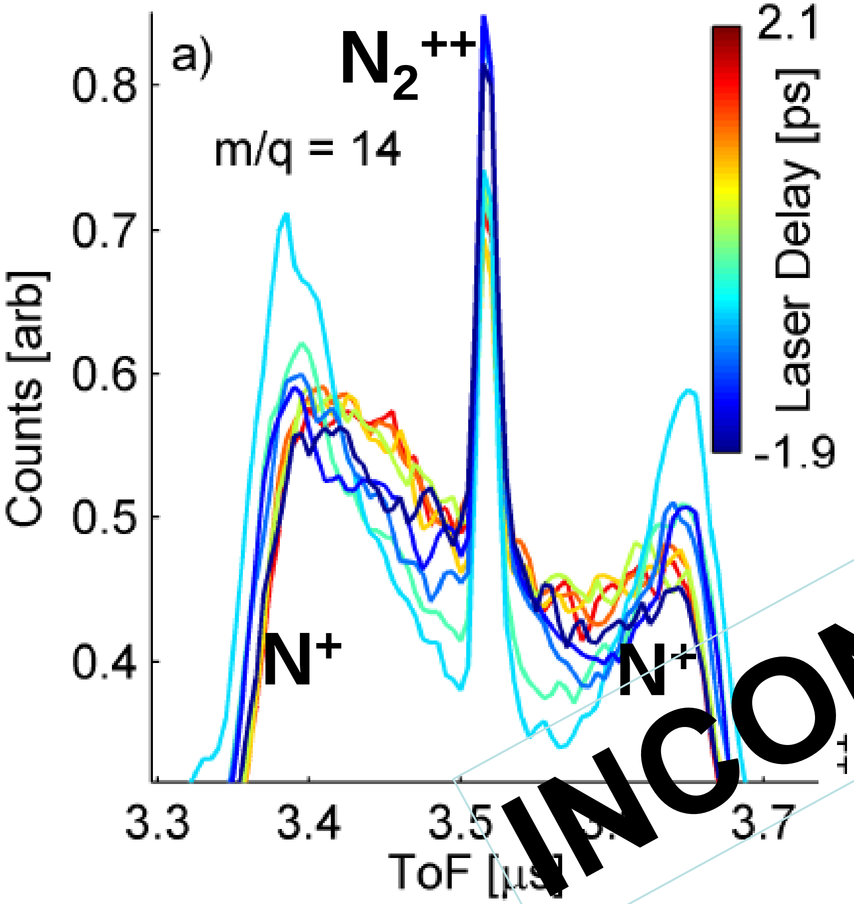
R. W. Wetmore and R. K. Boyd, J. Phys. Chem. 90, 5540 (1986).

t=47 fs = 0.09eV ~ N_2^{2+} $^1\Pi_u$(v=10→11)

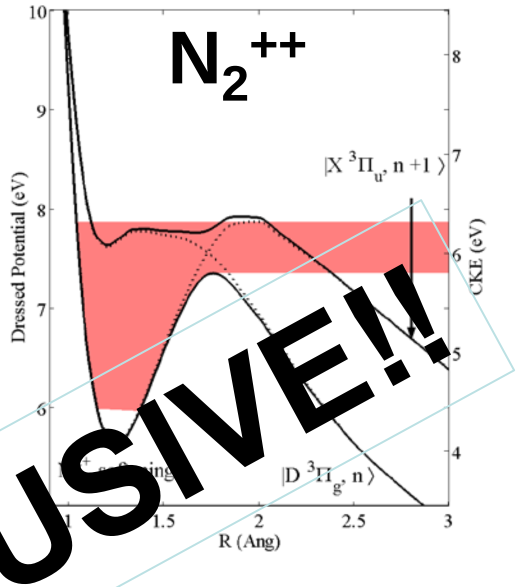
t=21 fs = 0.19eV ~ N_2^{2+} $X^1\Sigma_g$(v=3→4)

t=13 fs = 0.31eV ~ N_2^{+} $^2\Sigma_g$(v=0→1)

Search for N_2^{++} vibrational wave packets probed by 800 nm dissociation.



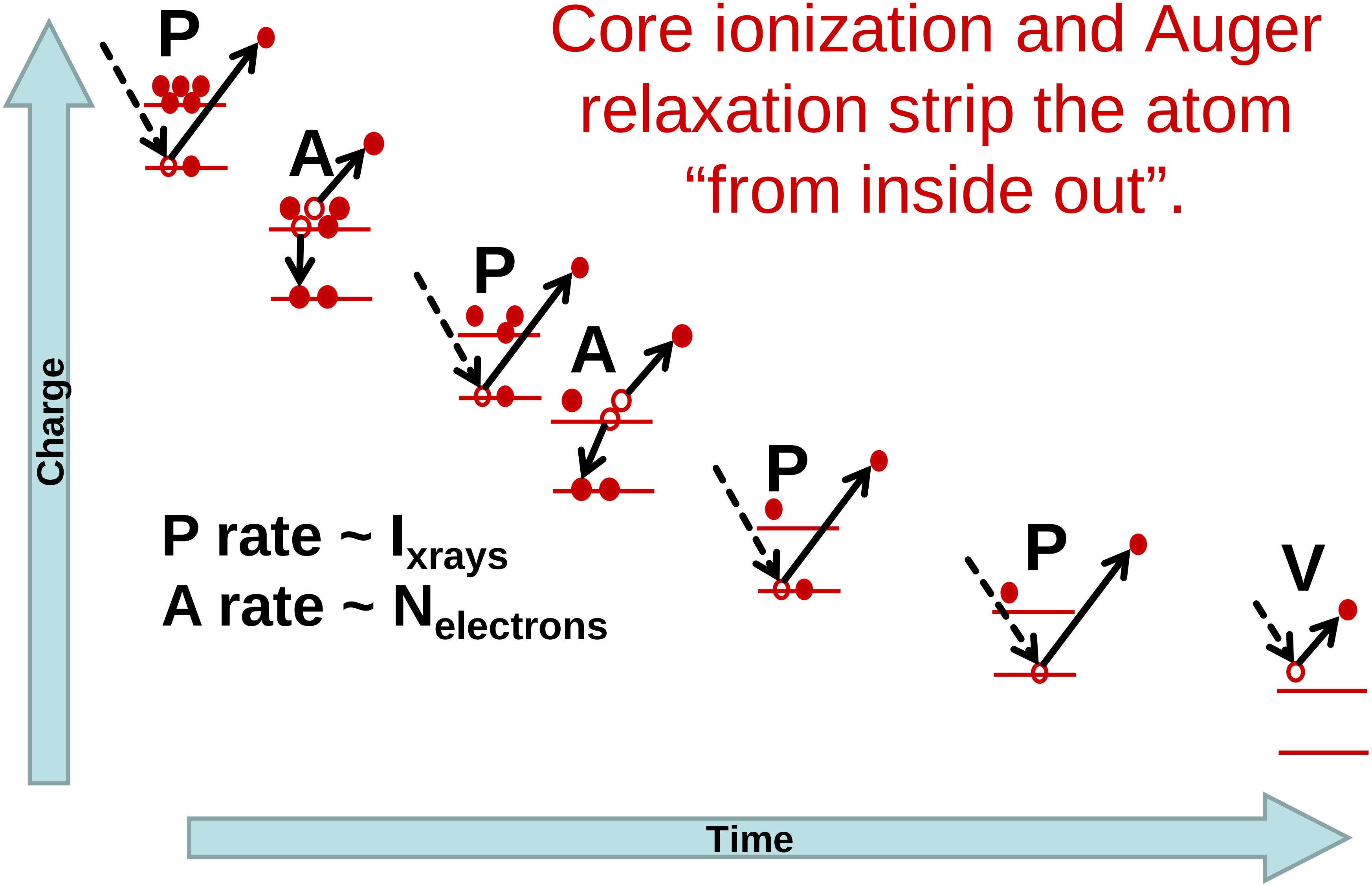
INCONCLUSIVE!!



Pump-probe jitter obscures the femtosecond dynamics

Glownia, J. M., et al. (2010). Opt. Express 18(17): 17620-17630.

Processes with more than 2 active electrons: Stripping atoms at LCLS



Core ionization and Auger relaxation strip the atom "from inside out".

P rate ~ I_{xrays}

A rate ~ $N_{\mathrm{electrons}}$

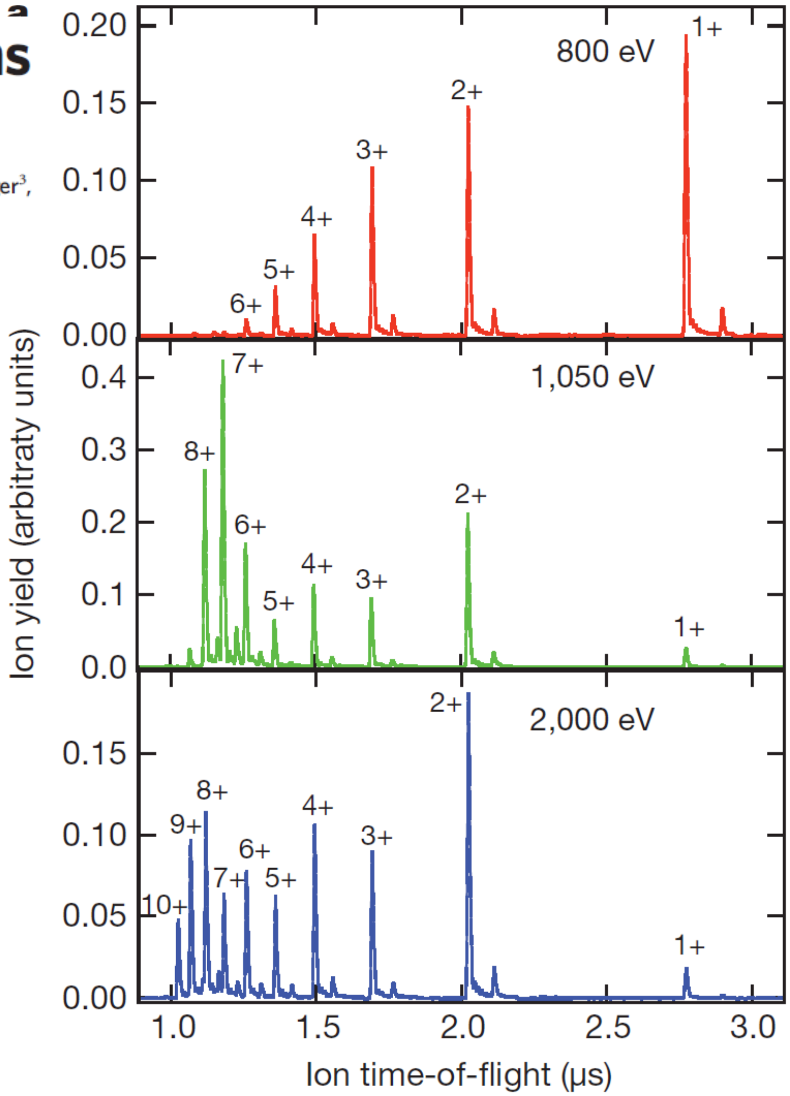
Ne^{10+} at the focus

Femtosecond electronic response of atoms to ultra-intense X-rays

L. Young[1], E. P. Kanter[1], B. Krässig[1], Y. Li[1], A. M. March[1], S. T. Pratt[1], R. Santra[1,2], S. H. Southworth[1], N. Rohringer[3], L. F. DiMauro[4], G. Doumy[4], C. A. Roedig[4], N. Berrah[5], L. Fang[5], M. Hoener[5,6], P. H. Bucksbaum[7], J. P. Cryan[7], S. Ghimire[7], J. M. Glownia[7], D. A. Reis[7], J. D. Bozek[8], C. Bostedt[8] & M. Messerschmidt[8]

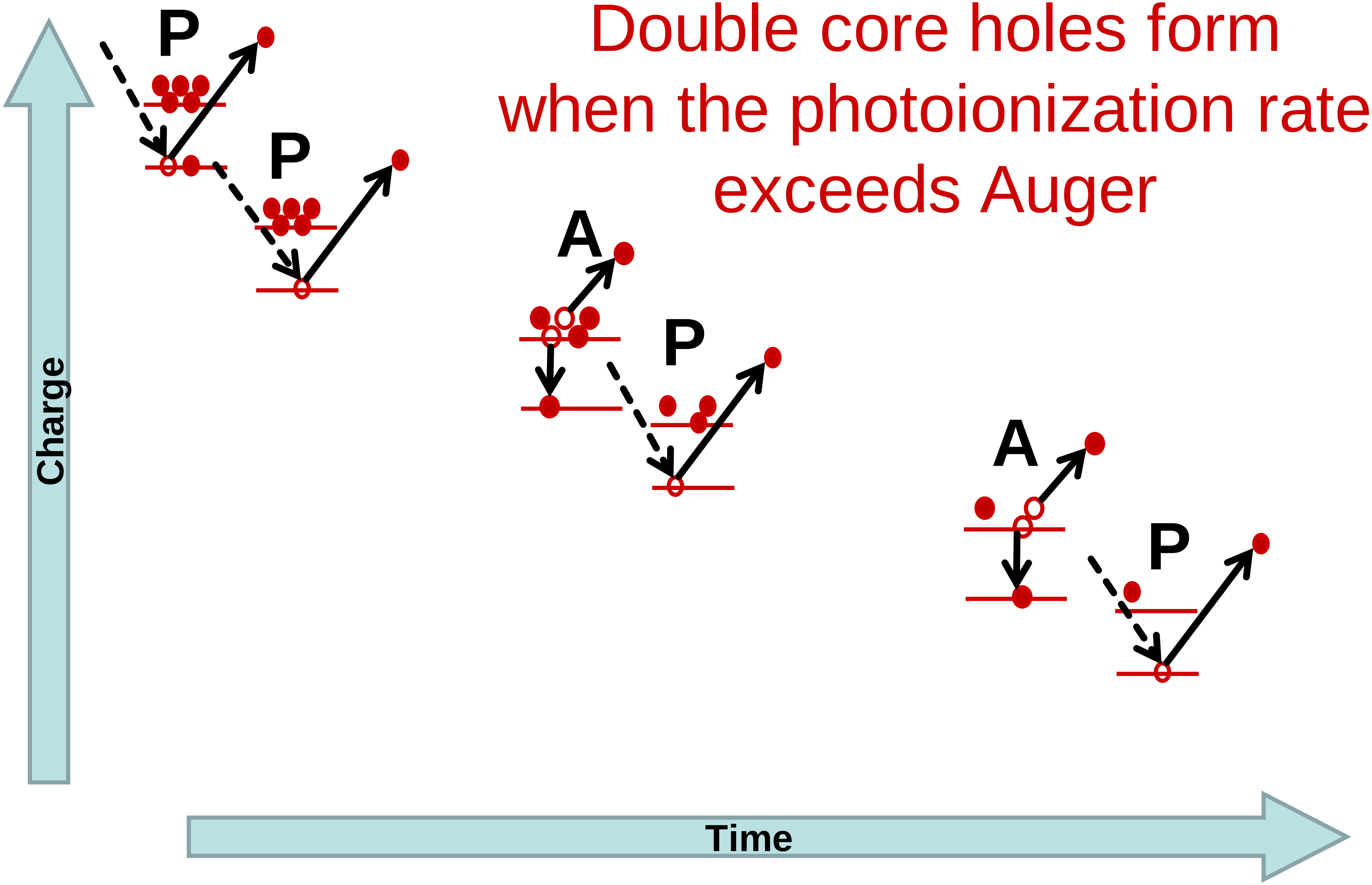
Nature 466, 56 (2010)

- **Full ionization of Ne is possible**



Strong and Ultrafast: Hollow atoms

Double core holes form
when the photoionization rate
exceeds Auger



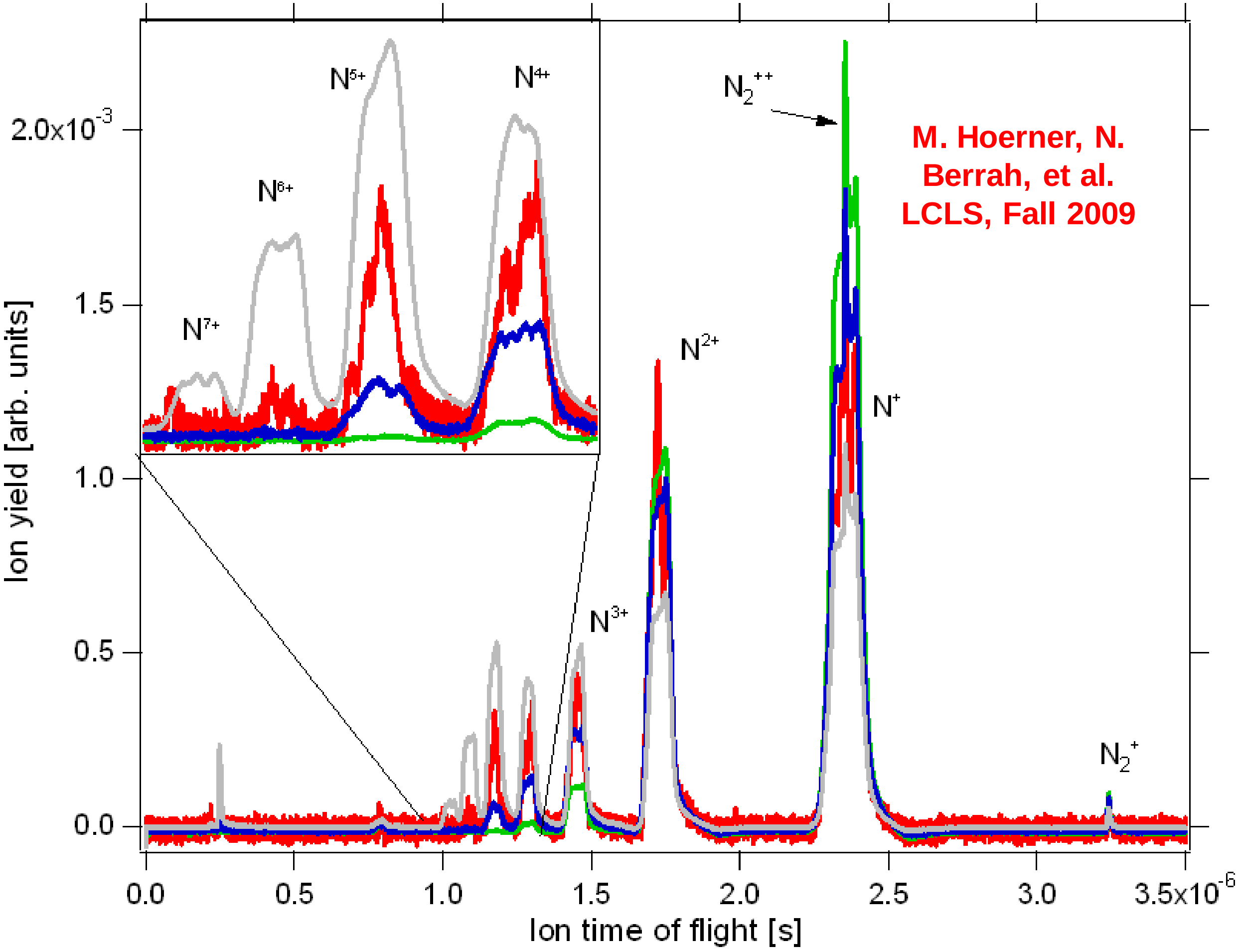
Hollow atom formation at LCLS

Hoener, M., L. Fang, et al. Phys. Rev. Lett. 104(25): 253002 (2010).

Frustrated ionization in N2

pulse duration,
pulse energy
🟥 280 fs, 0.26 mJ
🟦 80 fs, 0.27 mJ
🟩 5-10 fs, 0.26 mJ

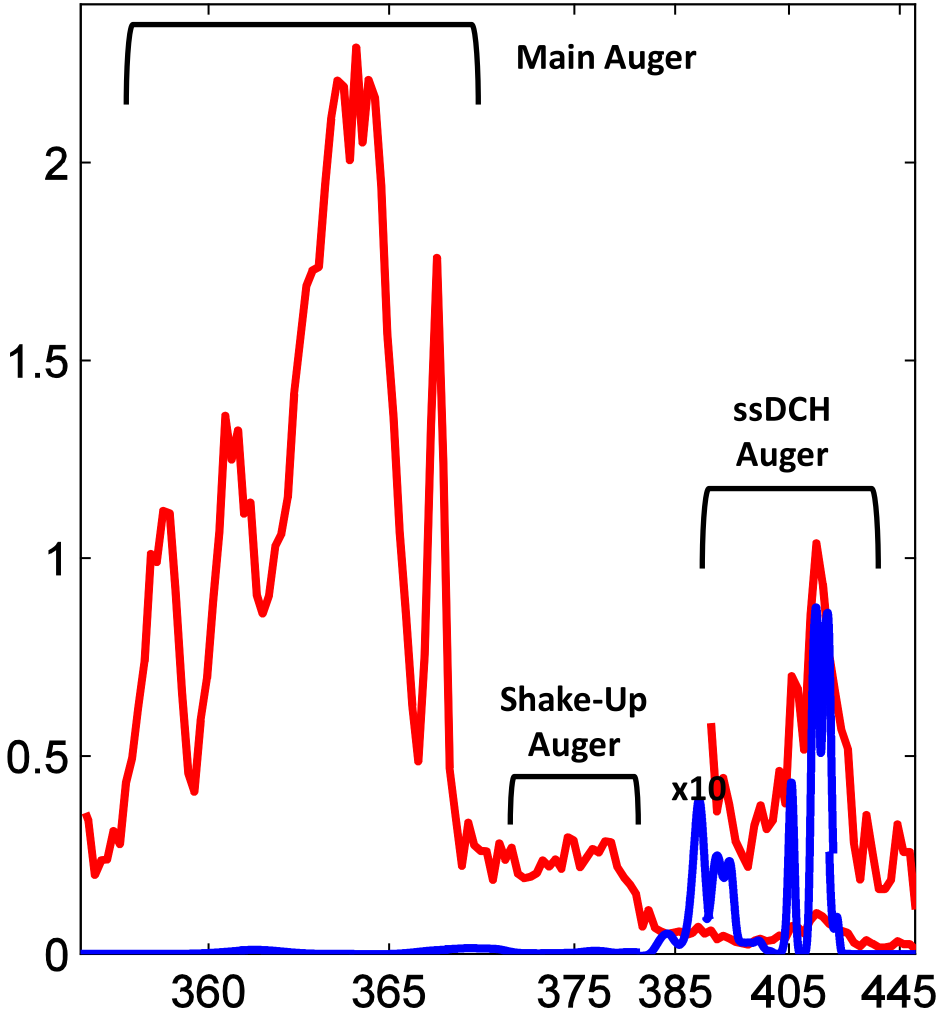
⬜ 280 fs, 2.2 mJ



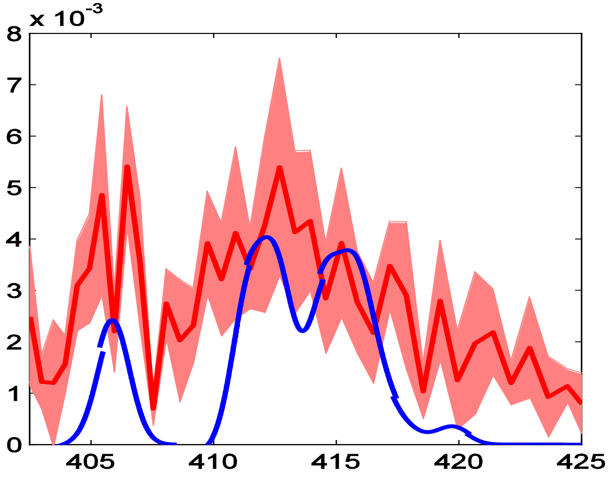
M. Hoerner, N. Berrah, et al.
LCLS, Fall 2009

Hoener, M., Fang, L., Kornilov, O., Gessner, O., Pratt, S. T., G\"Uhr, M., Kanter, E. P., Blaga, C., Bostedt, C., Bozek, J. D., Bucksbaum, P. H., Buth, C., Chen, M., Coffee, R., Cryan, J., Dimauro, L., Glownia, M., Hosler, E., Kukk, E., Leone, S. R., Mcfarland, B., Messerschmidt, M., Murphy, B., Petrovic, V., Rolles, D. & Berrah, N.

N_2 from LCLS shows Double core vacancies.



Single-site double core Auger spectrum



Cryan PRL 105, 083004 (2010)

Cryan, J. P., Glownia, J. M., Andreasson, J., Belkacem, A., Berrah, N., Blaga, C. I., Bostedt, C., Bozek, J., Buth, C., Dimauro, L. F., Fang, L., Gessner, O., Guehr, M., Hajdu, J., Hertlein, M. P., Hoener, M., Kornilov, O., Marangos, J. P., March, A. M., Mcfarland, B. K., Merdji, H., Petrovi\Ifmmode \Acutec\Else \'C\Fi, V. S., Raman, C., Ray, D., Reis, D., Tarantelli, F., Trigo, M., White, J. L., White, W., Young, L., Bucksbaum, P. H. & Coffee, R. N.

Beyond the single active electron

- X-ray induced core holes and transient states in molecules are two new paths to driven multi-electron dynamics.
- Strong field lasers can probe interelectron dynamics induced on the fastest time scales in molecules.
- Hollow atoms and Auger spectra offer new opportunities to study strong-field processes

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Thanks!