Opining on … well everything

Multi-color, multi-polarization, soft / hard… what might we imagine?

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U.S. DEPARTMENT OF ENERGY
Office of Science

SLAC NATIONAL ACCELERATOR LABORATORY

Outline

- The elephant in the room
- Coincidental alternatives
- Infrastructure
- Short term memory loss

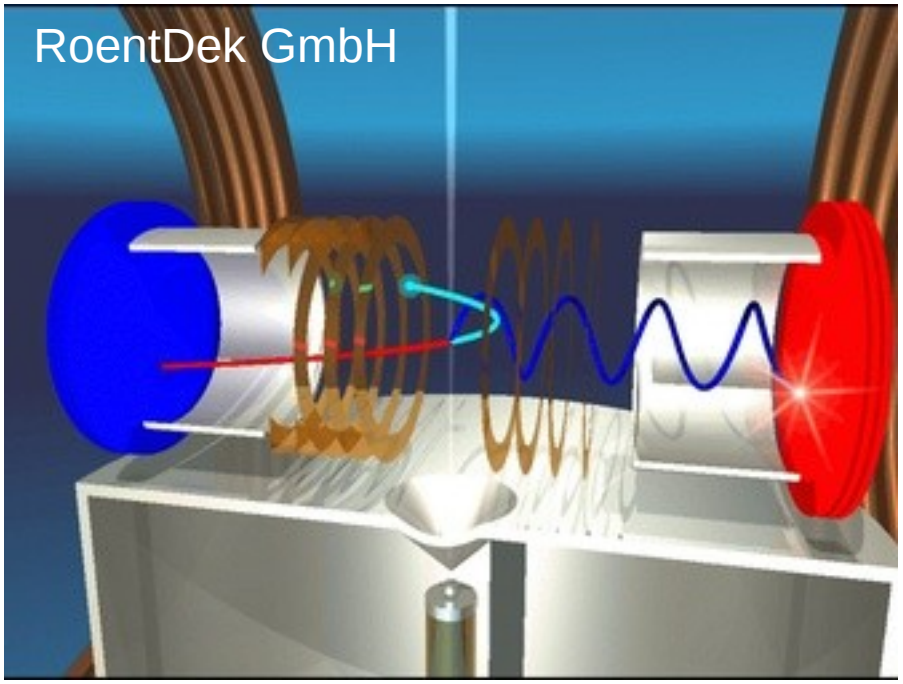
The Elephant in the room

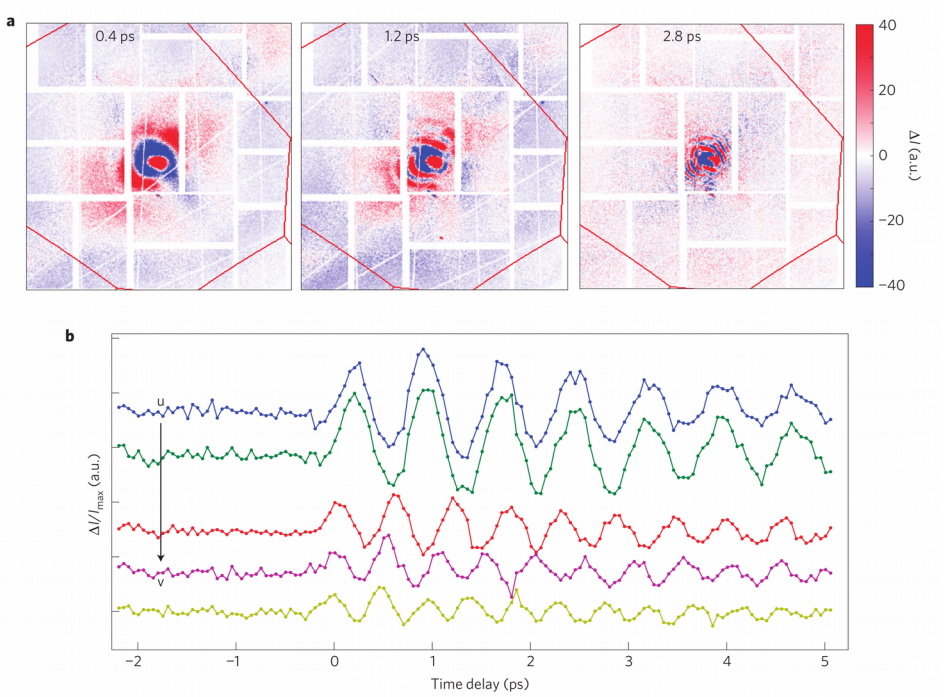
Outline

- The elephant in the room
- Coincidental alternatives
- Infrastructure
- Short term memory loss

Is there another way?

- Can we find something that doesn't care about axial recoil?
- Do we really need complete reconstruction?
- Is it time to start playing dirty?

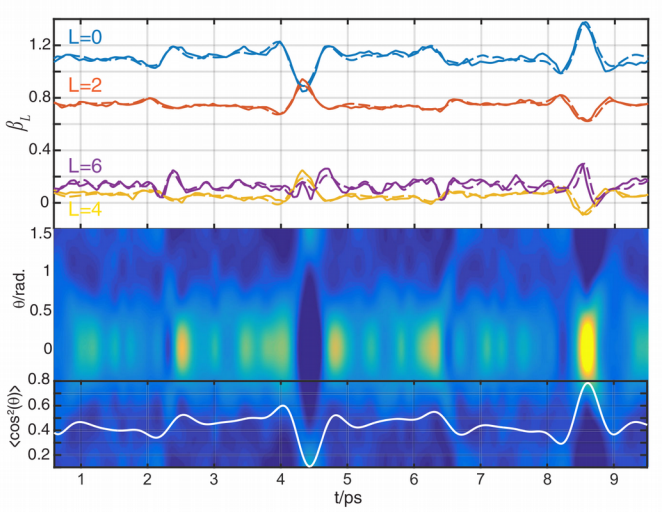




Trigo *et al.*, Nat. Phys. **9**, 790 (2013)

Is there another way?

- Asymmetric Top molecules are everywhere
- Record whole pattern through the 3D alignment/orientation revival
- Tumbling pattern should be “calculable”



PRL 119, 083401 (2017) PHYSICAL REVIEW LETTERS week ending 25 AUGUST 2017

Molecular Frame Reconstruction Using Time-Domain Photoionization Interferometry

Claude Marceau,[1] Varun Makhija,[2] Dominique Platzer,[1] A. Yu. Naumov,[1] P. B. Corkum,[1] Albert Stolow,[2,3,4] D. M. Villeneuve,[1] and Paul Hockett[4,*]

[1]*Joint Attosecond Science Laboratory, National Research Council of Canada and University of Ottawa, 100 Sussex Drive, Ottawa, Ontario K1A 0R6, Canada*
[2]*Department of Physics, University of Ottawa, 150 Louis Pasteur, Ottawa, Ontario K1N 6N5, Canada*
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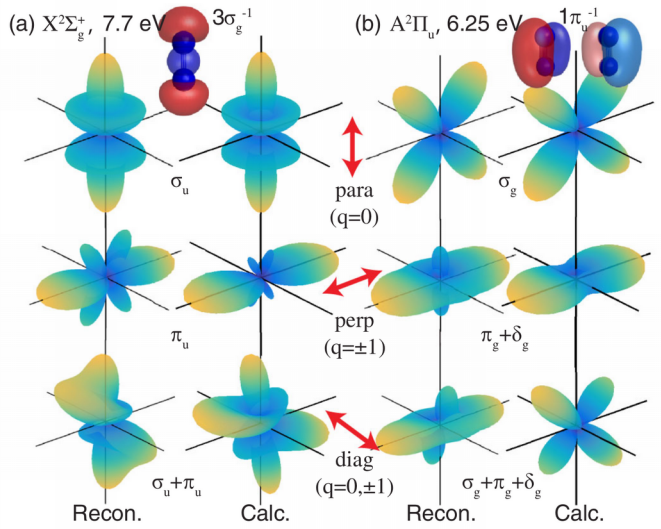
(Received 30 January 2017; published 22 August 2017)

Photoionization of molecular species is, essentially, a multipath interferometer with both experimentally controllable and intrinsic molecular characteristics. In this work, XUV photoionization of impulsively aligned molecular targets (N_2) is used to provide a time-domain route to “complete” photoionization experiments, in which the rotational wave packet controls the geometric part of the photoionization interferometer. The data obtained is sufficient to determine the magnitudes and phases of the ionization matrix elements for all observed channels, and to reconstruct molecular frame interferograms from lab frame measurements. In principle, this methodology provides a time-domain route to complete photoionization experiments and the molecular frame, which is generally applicable to any molecule (no prerequisites), for all energies and ionization channels.

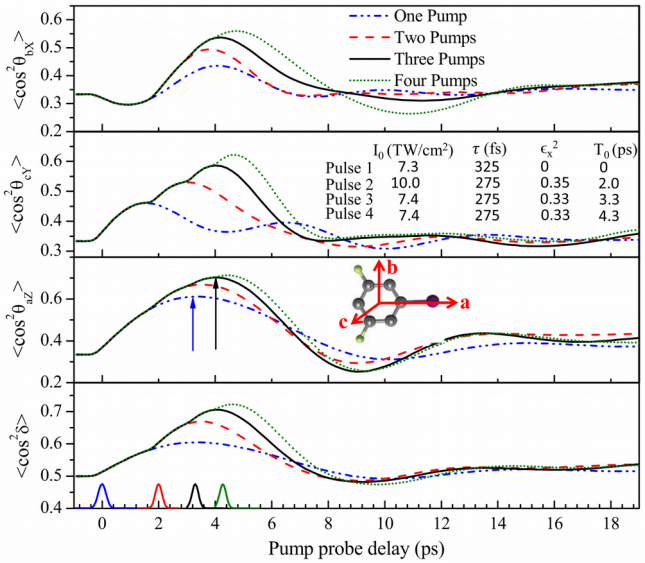
DOI: 10.1103/PhysRevLett.119.083401

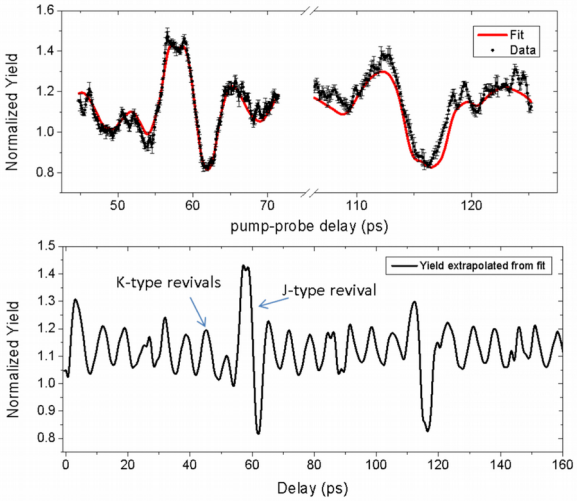


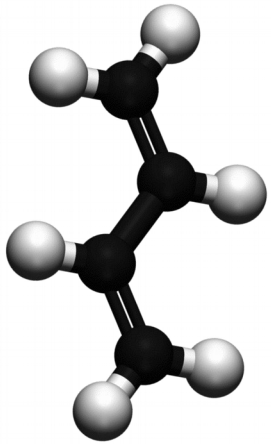
Is there another way?

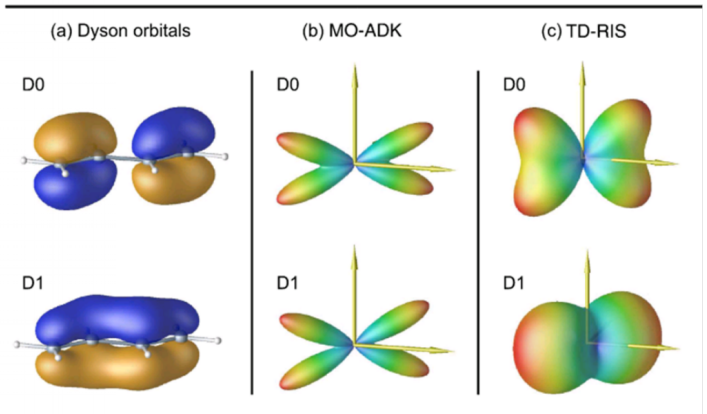
- Asymmetric Top molecules are everywhere
- Record whole pattern through the 3D alignment/orientation revival
- Tumbling pattern should be “calculable”

Xiaoming Ren, Varun Makhija, and Vinod Kumarappan, PRL **112**, 173602 (2014)









Varun Makhija … Spanner & Stolow, *in prep.*

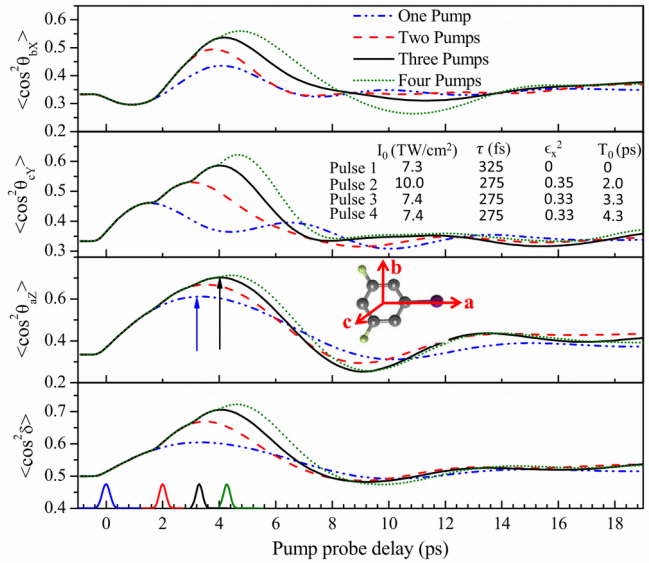
Is there another way?

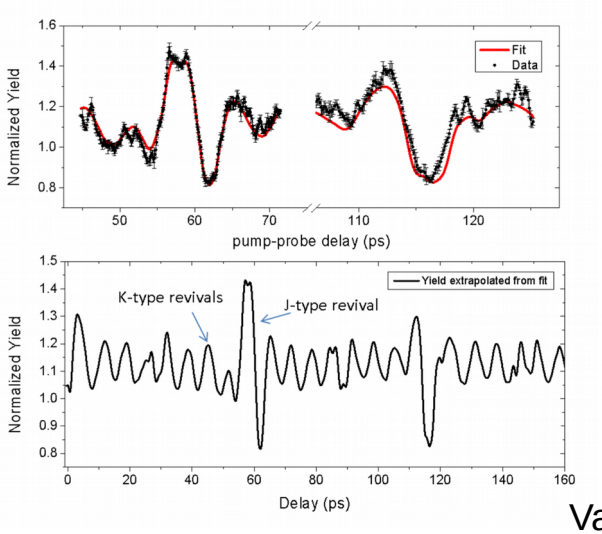
Xiaoming Ren, Varun Makhija, and Vinod Kumarappan, PRL **112**, 173602 (2014)

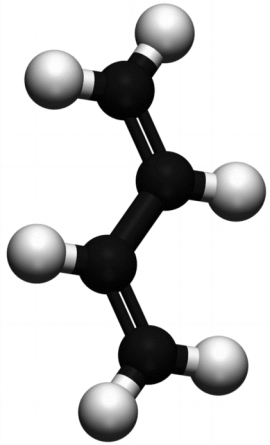
the measured time dependent ionization probability,

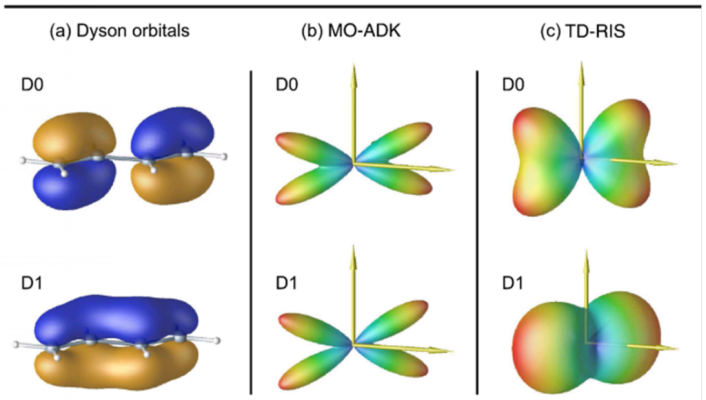
$$\langle S \rangle (t) = \sum_{jk} C_{jk} \left(\left\langle D^j_{0k} \right\rangle (t) + \left\langle D^j_{0-k} \right\rangle (t) \right).$$

where the angle brackets indicate an average over the molecular axis distribution. The above equation is linear in the C_{jk} and easily (and uniquely) solvable if the $\left\langle D^j_{0k} \right\rangle (t)$ can be calculated. We solve the time dependent Shcrödinger equation (TDSE) for an asymmetric rigid rotor interacting with the laser pulse and calculate and store the $\left\langle D^j_{0k} \right\rangle (t)$ up to $j, k = 4$ for numerous laser intensities and pulse durations and a large number of initial rotational states.





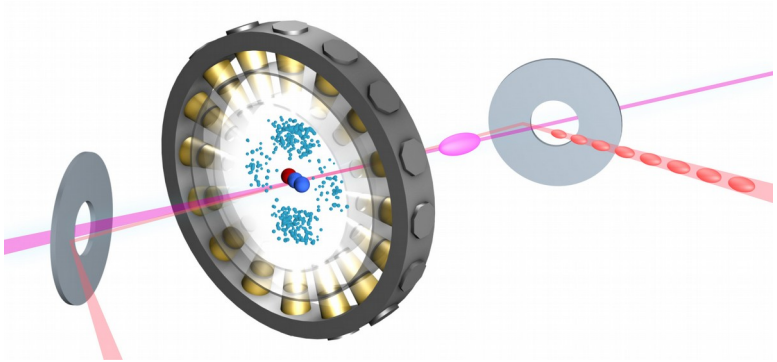


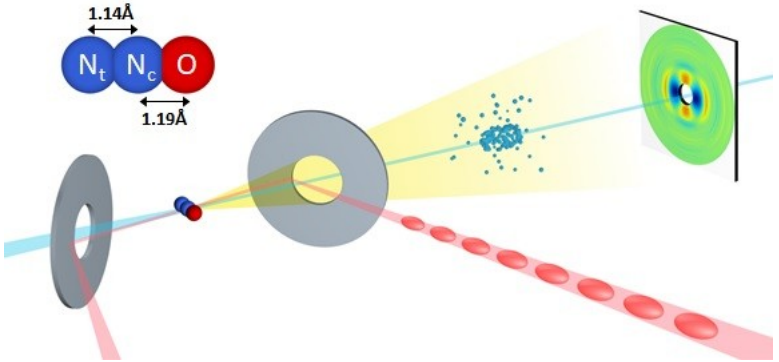


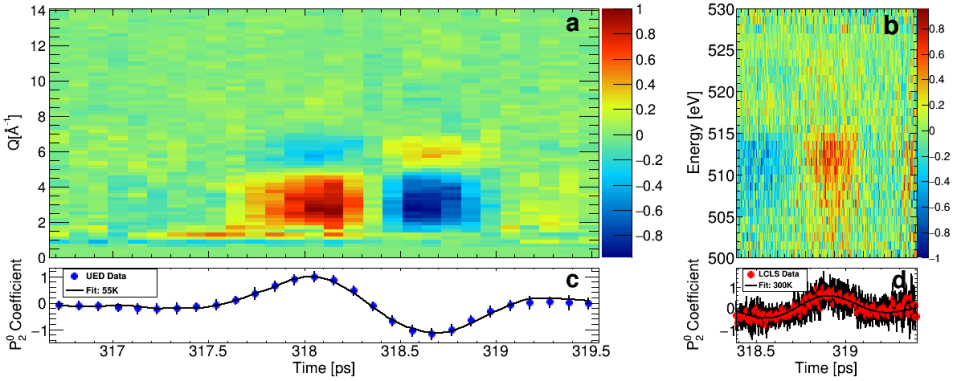
Varun Makhija … Spanner & Stolow, *in prep.*

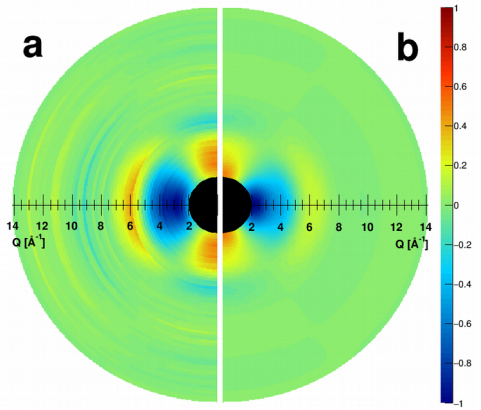
Is there another way?

- Let's induce a rotational coherence
- Let's look for that pattern in every pixel of our "detector"







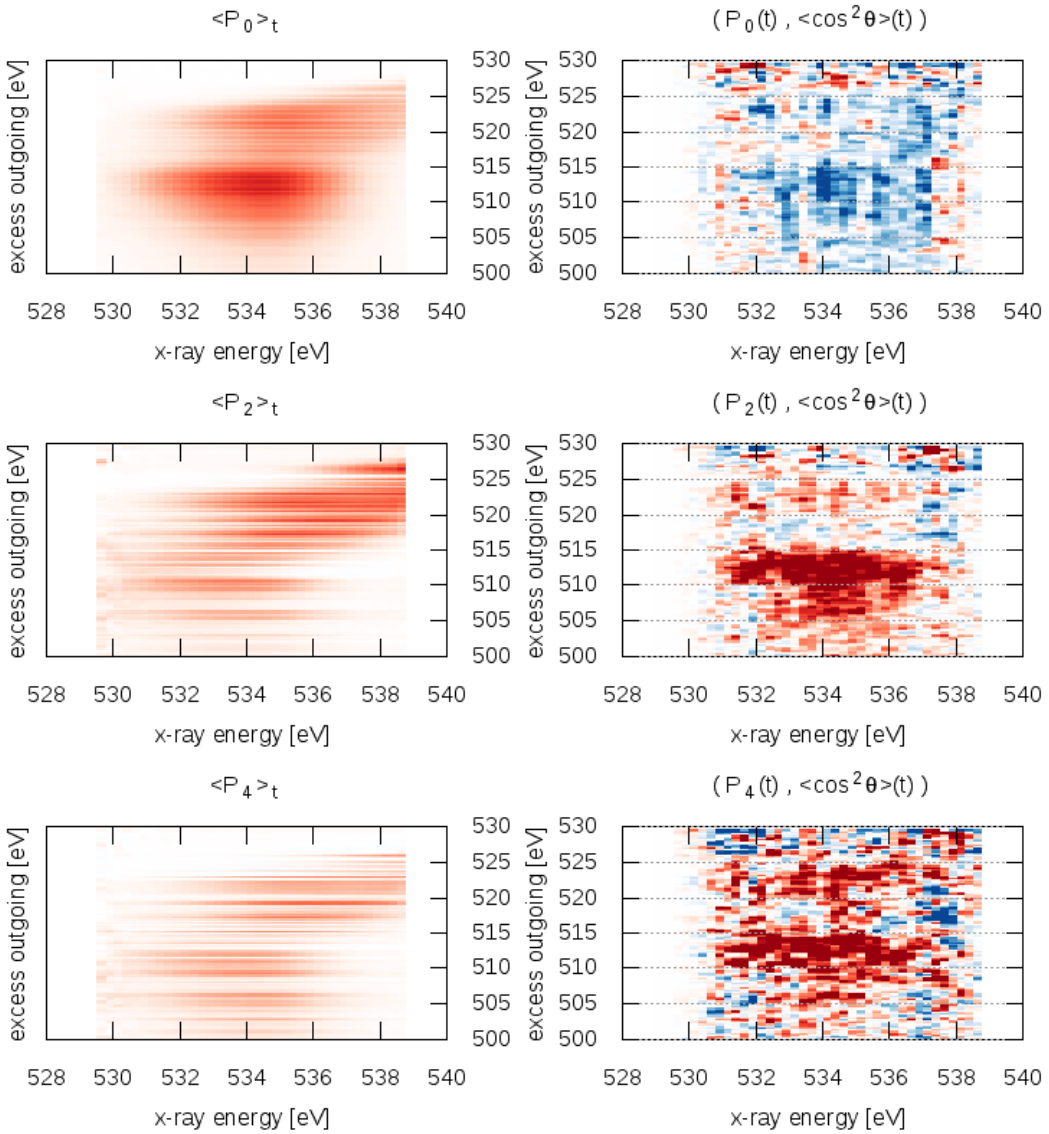


Hegazy *et al.*, in prep

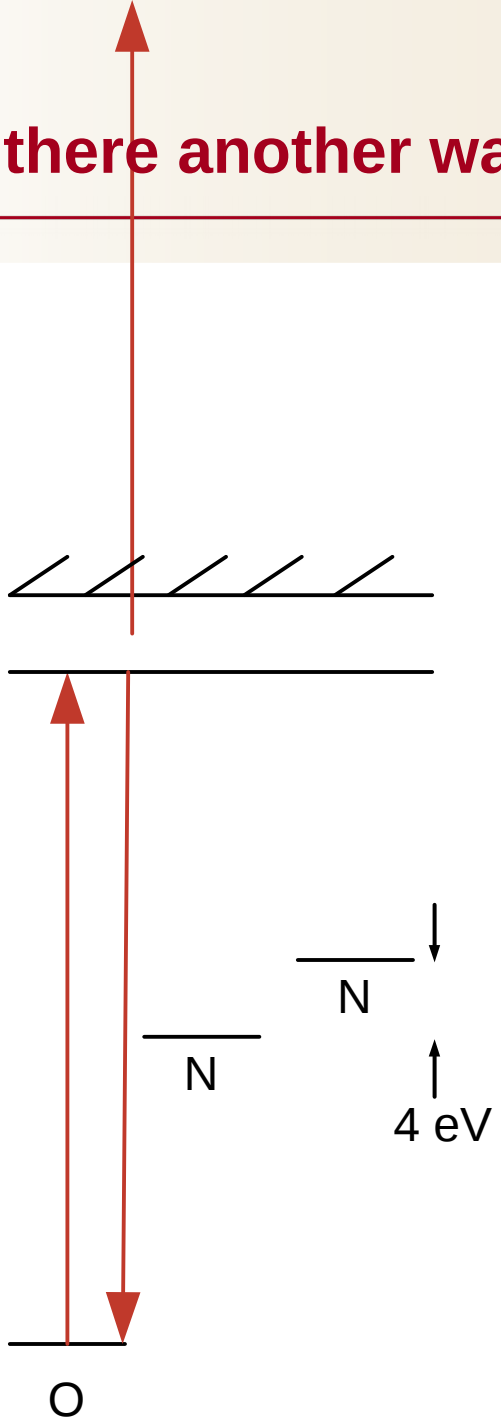
Is there another way?

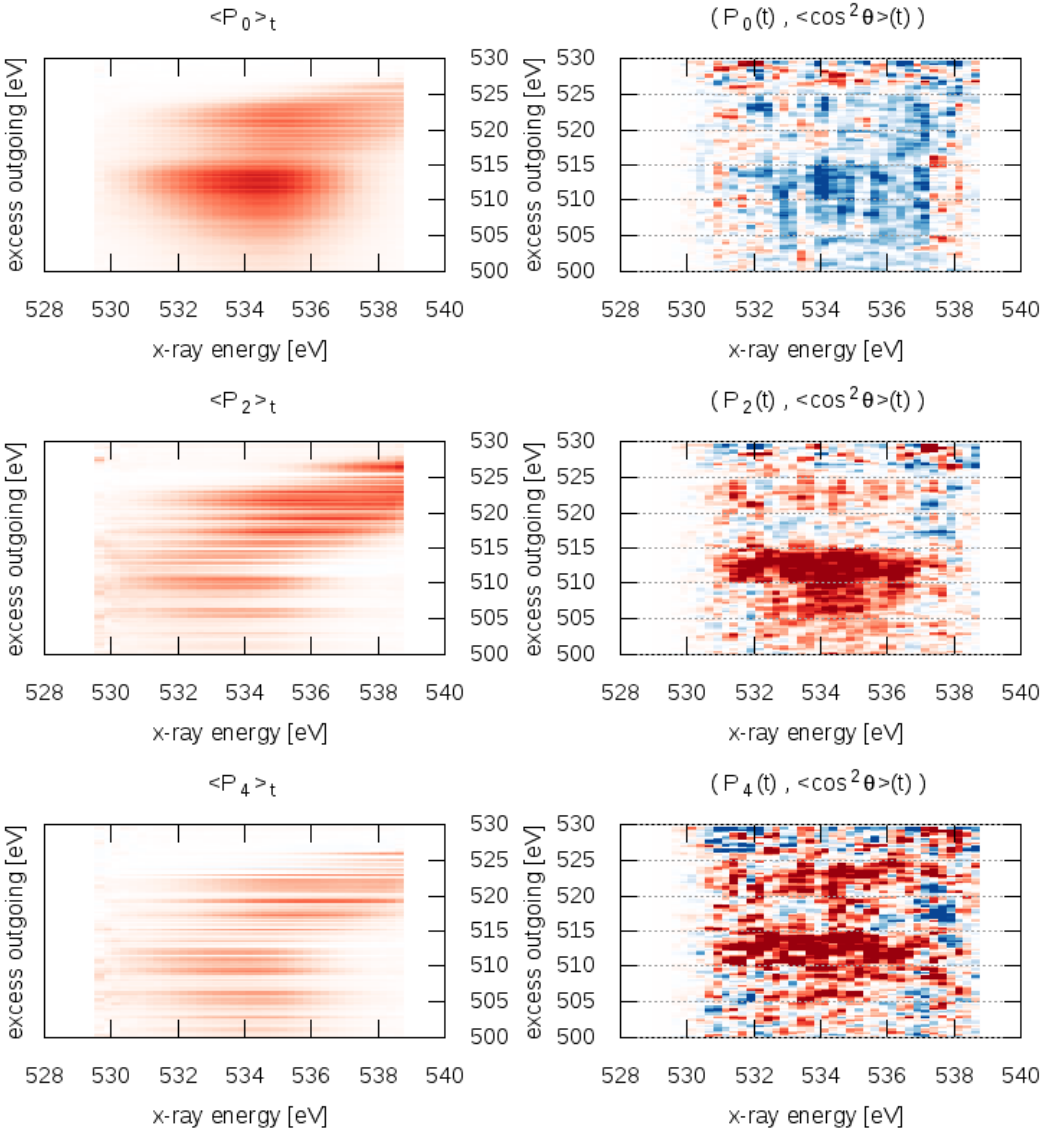
- High dimensional parameter space
  - Xray spectrum
  - Xray/optical delay
  - Outgoing energy
  - Outgoing angular pattern
- Desire a “foveated” distribution of statistics
  - Data router
  - Suggestion engine for machine controls

Subtlety is the next step for high velocity data in science



Is there another way?



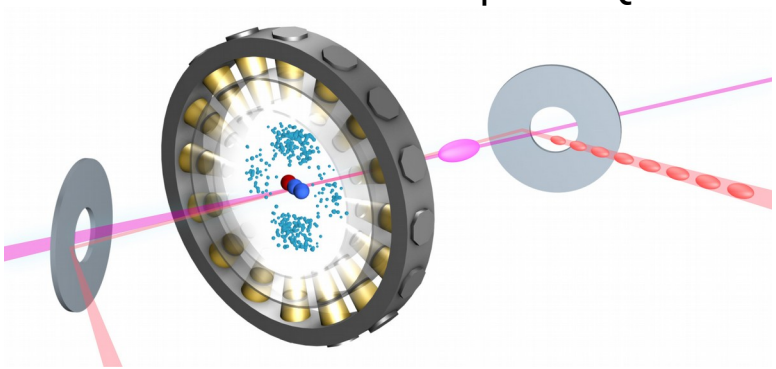


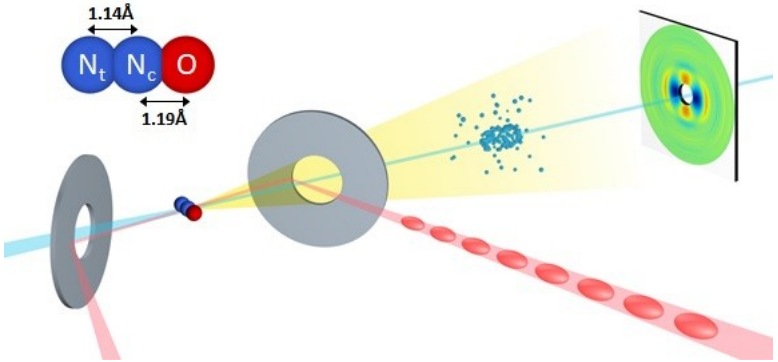
Outline

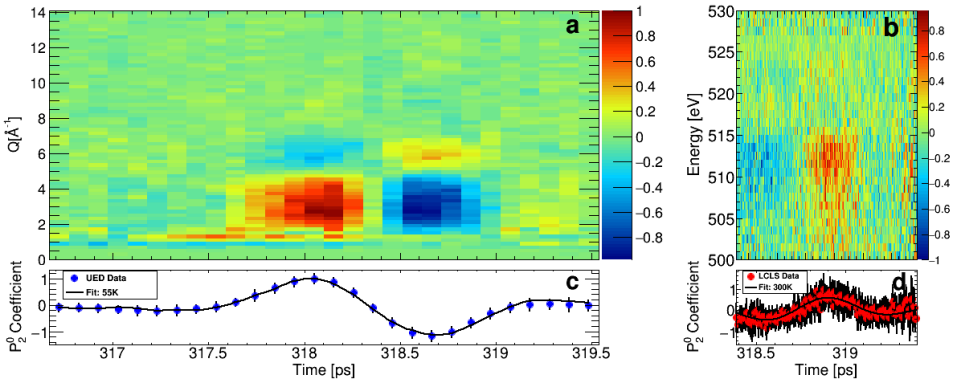
- The elephant in the room
- Coincidental alternatives
- Infrastructure
- Short term memory loss

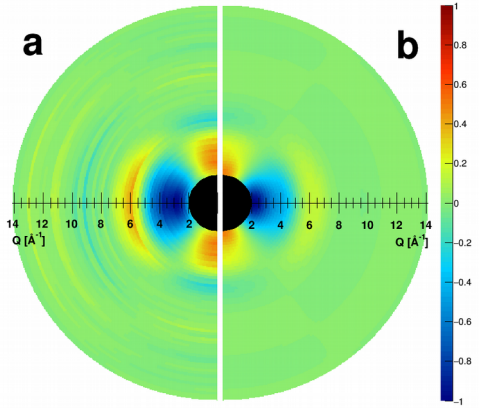
Using “wiggles”

- More “wiggles” in diffraction locks data and calculation more tightly
- Electronic symmetry breaking – 0.5-2 fs duv & x-ray pulses with 0.5-2 fs time sorting
  “Give an inch, they’ll take a mile”
- Corresponding structural deformation – 5 fs pulses with ~5 fs time-sort resolution,
  Nyquist for 3000 cm^{-1}
- Partner diffraction setup with Q > 20 $Å^{-1}$





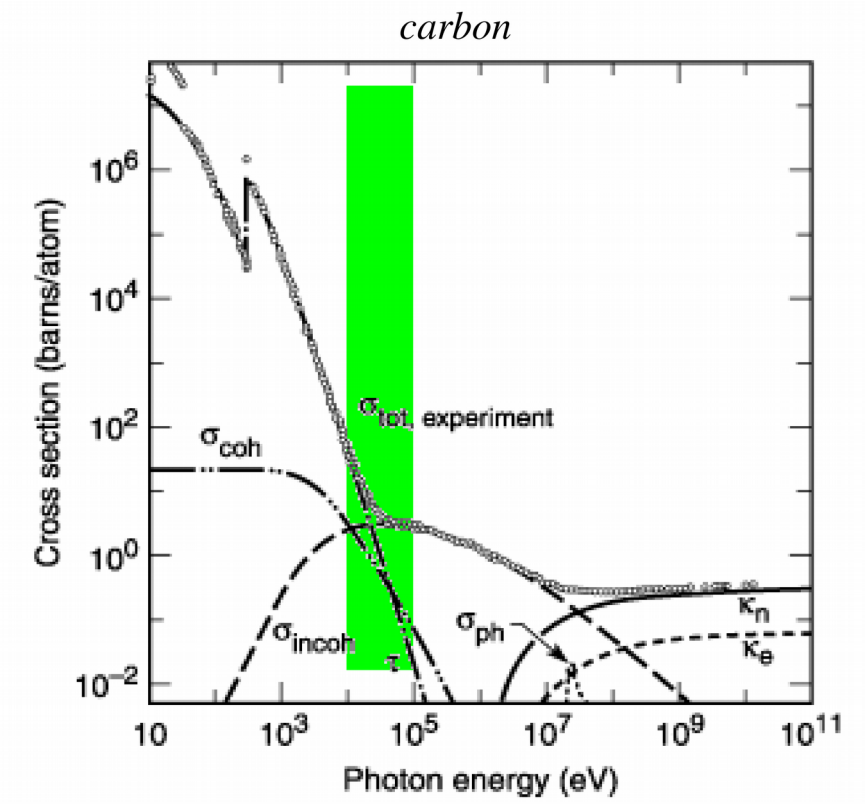


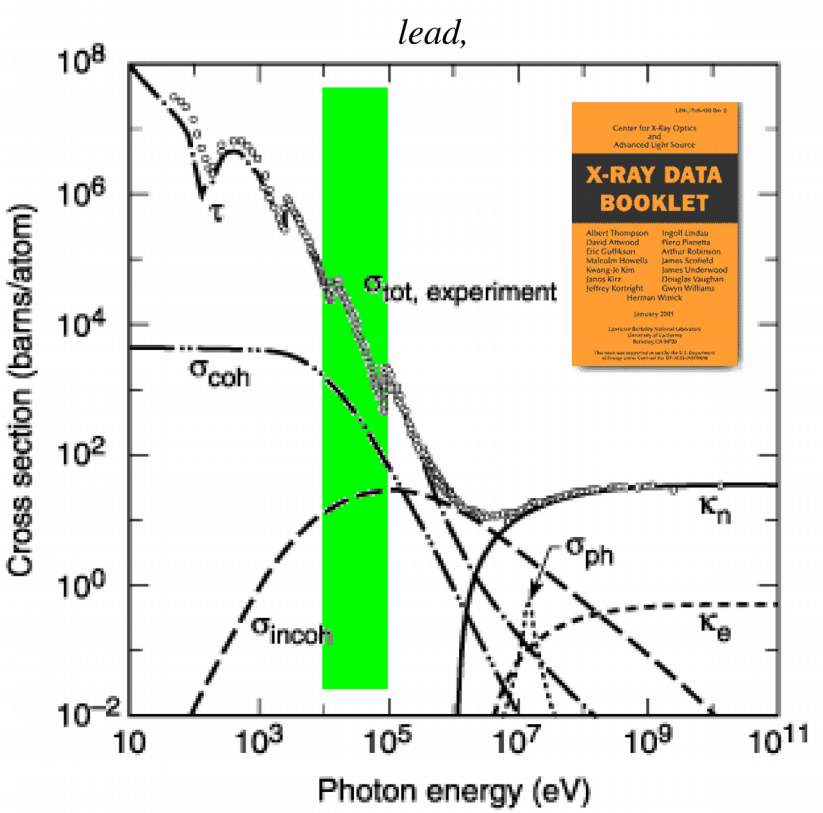


Hegazy *et al.*, in prep

Hard X-rays… how hard?

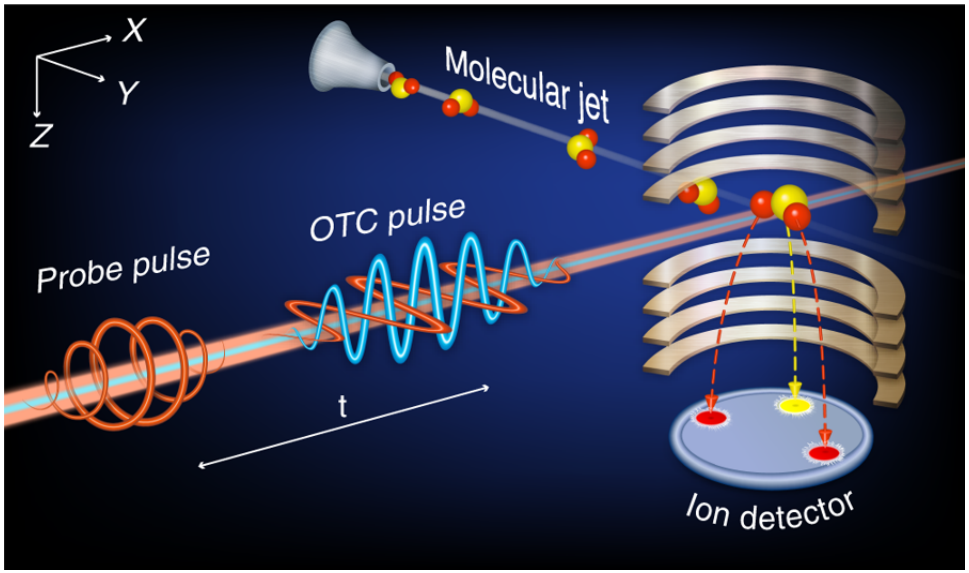
- 10-15 keV Coherent/Compton is around 1:1 for 2nd row elements
- 50-100 keV for 6th row : Lu, W, Ir, Pt, Au, Hg, Pb, Bi

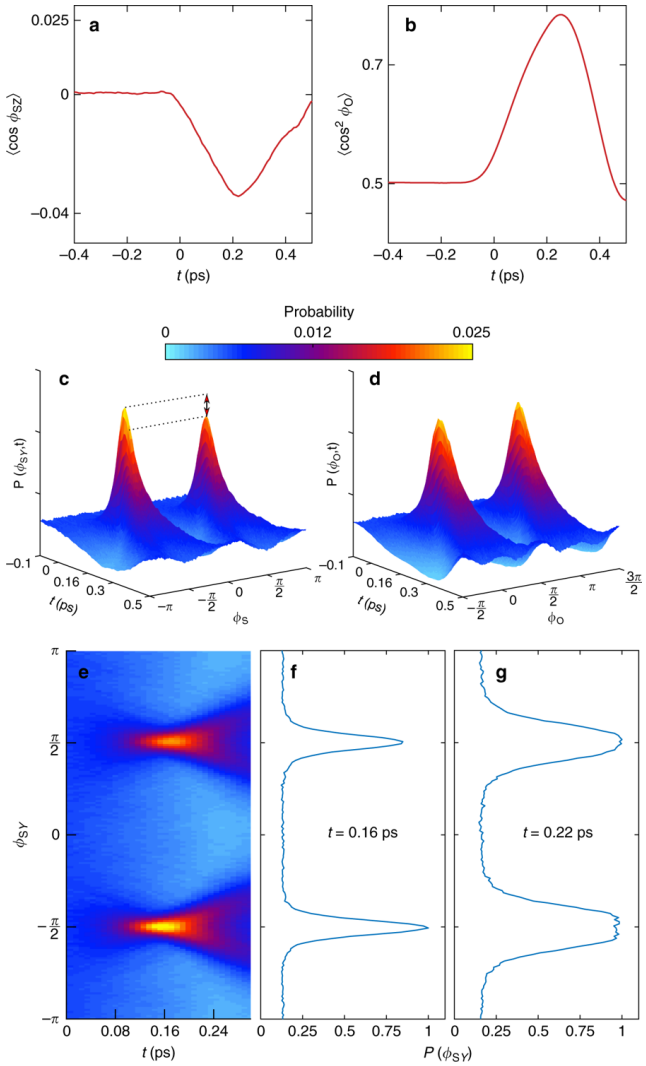




Shaping “setup” pulses

- Asymmetric Top molecules are everywhere
- Record whole pattern through the 3D alignment/orientation revival
- Tumbling pattern should be “calculable”





Lin, … Averbukh & Wu, Nat. Comm. **9**, 5134 (2018)

Shaping “setup” pulses

- Anders, “Waggling” of the CO on Ru is likely very close to the bend mode in CO_2, so the THz could resonantly pump the libration
- So might the beat note between two IR pulses that have spectral phase control, ala Molecular Centrifuge

VOLUME 65, NUMBER 19 PHYSICAL REVIEW LETTERS 5 NOVEMBER 1990

Efficient Molecular Dissociation by a Chirped Ultrashort Infrared Laser Pulse

Szczepan Chelkowski,[(1)] André D. Bandrauk,[(1)] and Paul B. Corkum[(2)]
[(1)]*Département de Chimie, Faculté des Sciences, Université de Sherbrooke, Sherbrooke, Québec, Canada J1K 2R1*
[(2)]*Division of Physics, National Research Council, Ottawa, Ontario, Canada K1A 0R6*
(Received 11 June 1990)

VOLUME 82, NUMBER 17 PHYSICAL REVIEW LETTERS 26 APRIL 1999

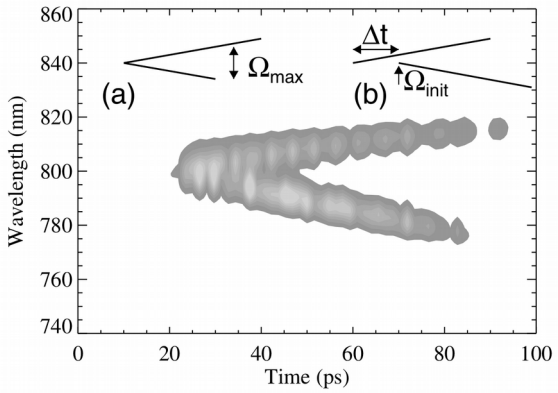
Optical Centrifuge for Molecules

Joanna Karczmarek,[1] James Wright,[2] Paul Corkum,[1] and Misha Ivanov[1]
[1]*SIMS NRC, 100 Sussex Drive, Ottawa, Ontario, Canada K1A 0R6*
[2]*Ottawa-Carleton Chemistry Institute, Carleton University, Ottawa, Ontario, Canada K1S 5B6*
(Received 5 October 1998)

VOLUME 85, NUMBER 3 PHYSICAL REVIEW LETTERS 17 JULY 2000

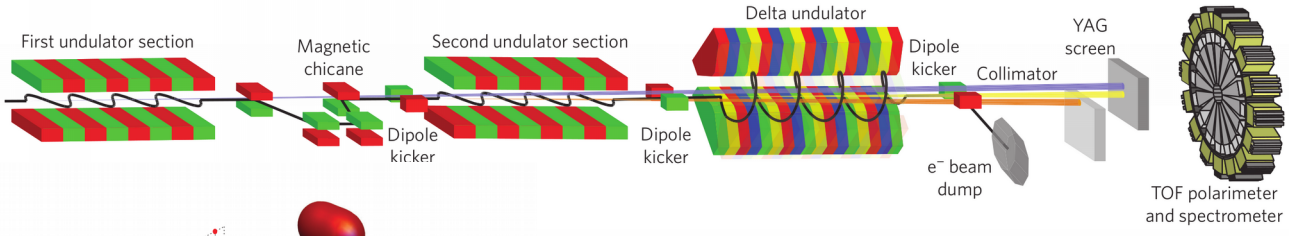
Forced Molecular Rotation in an Optical Centrifuge

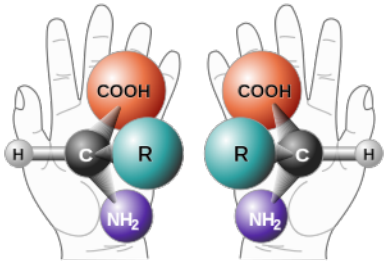
D. M. Villeneuve,[1,*] S. A. Aseyev,[1] P. Dietrich,[1,2] M. Spanner,[1] M. Yu. Ivanov,[1] and P. B. Corkum[1]
[1]*National Research Council of Canada, 100 Sussex Drive, Ottawa, Ontario, Canada K1A 0R6*
[2]*Freie Universität Berlin, Arnimallee 14, D-14195 Berlin, Germany*
(Received 17 February 2000)

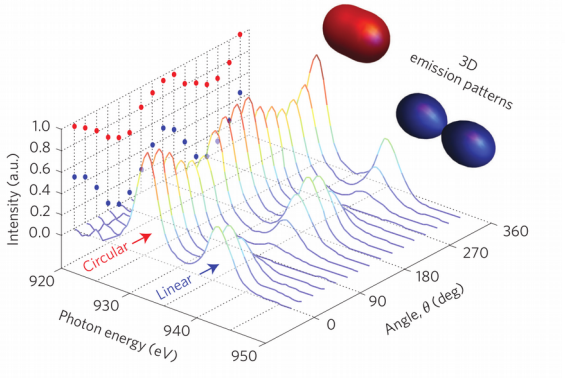


Shaping “setup” pulses

Lutman *et al.,* Nat. Photon. **10**, 468 (2016)





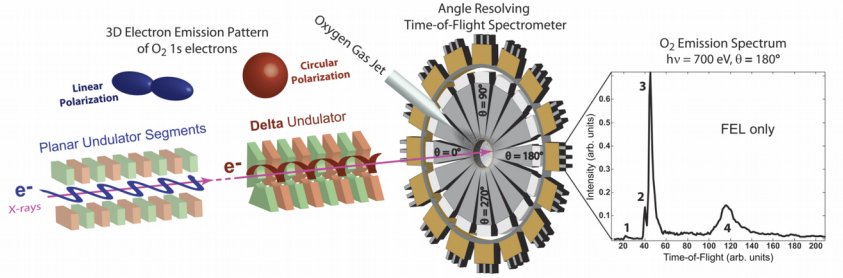


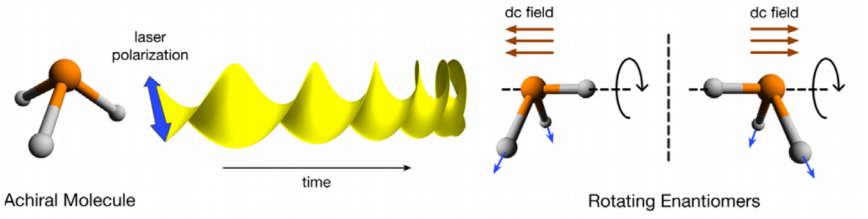
PHYSICAL REVIEW LETTERS **121,** 193201 (2018)

Climbing the Rotational Ladder to Chirality

Alec Owens,[1,2] Andrey Yachmenev,[1,2,*] Sergei N. Yurchenko,[3] and Jochen Küpper[1,2,4]
[1]*Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Notkestraße 85, 22607 Hamburg, Germany*
[2]*The Hamburg Center for Ultrafast Imaging, Universität Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany*
[3]*Department of Physics and Astronomy, University College London, Gower Street, WC1E 6BT London, United Kingdom*
[4]*Department of Physics, Universität Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany*

(Received 3 May 2018; revised manuscript received 2 September 2018; published 8 November 2018)

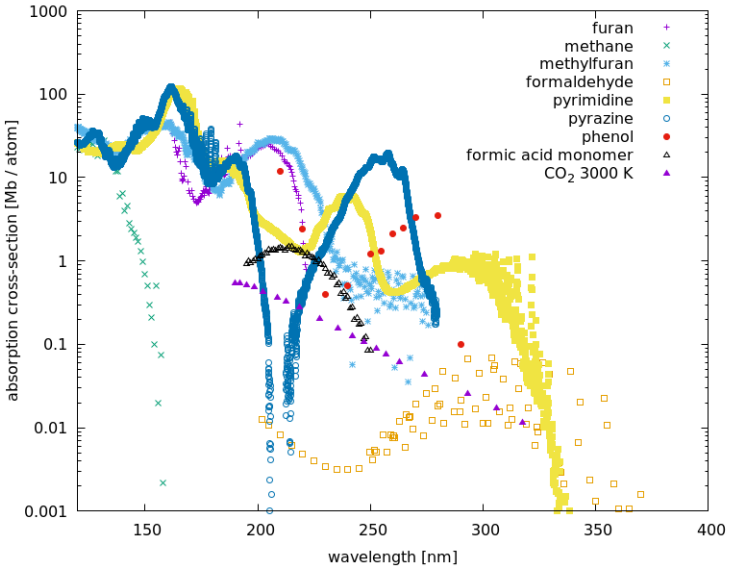


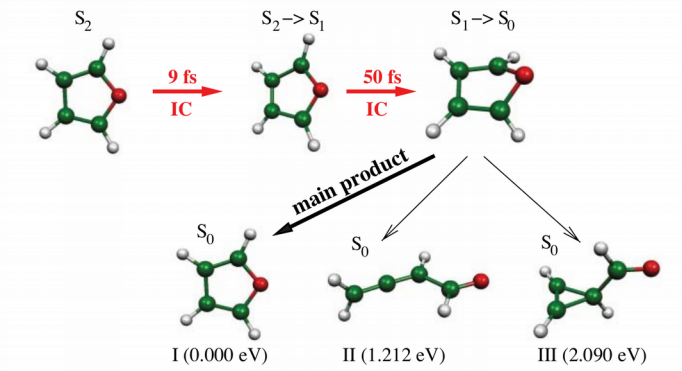


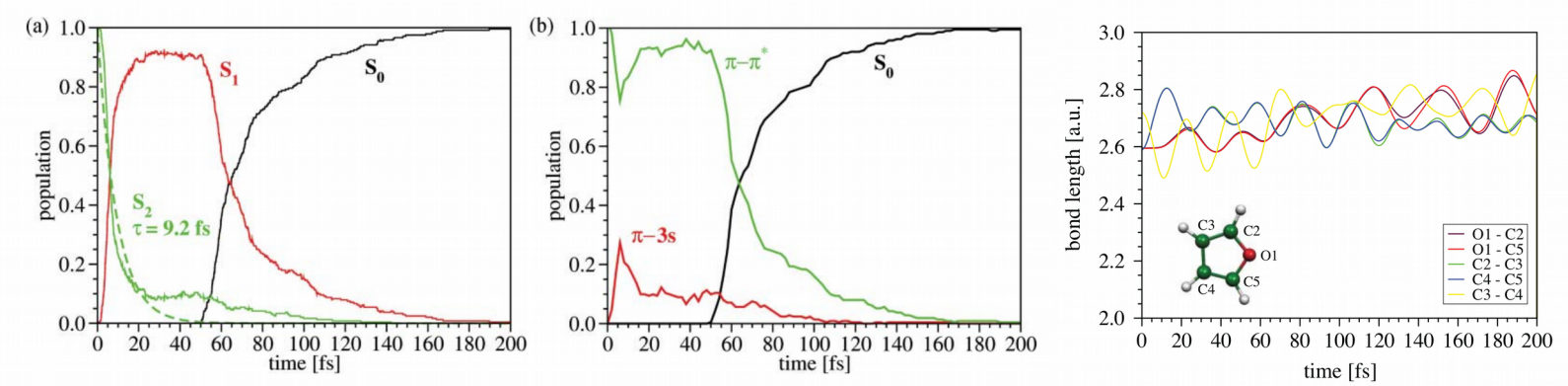
G. Hartmann *et al.,* RSI **87**, 083113 (2016)

Shaping “setup” pulses

- General, deep uv pumps bonding to anti-bonding
- “Electronic” transition is fast
- Resulting nuclear evolution is slow… like a whole 10 fs.

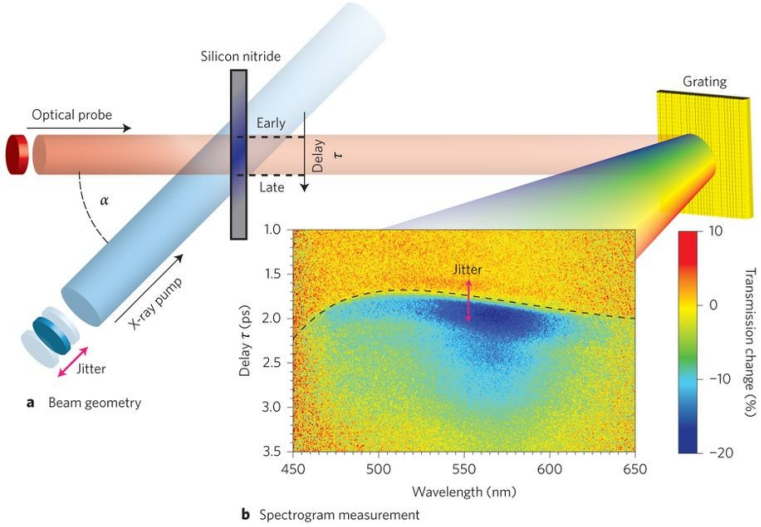


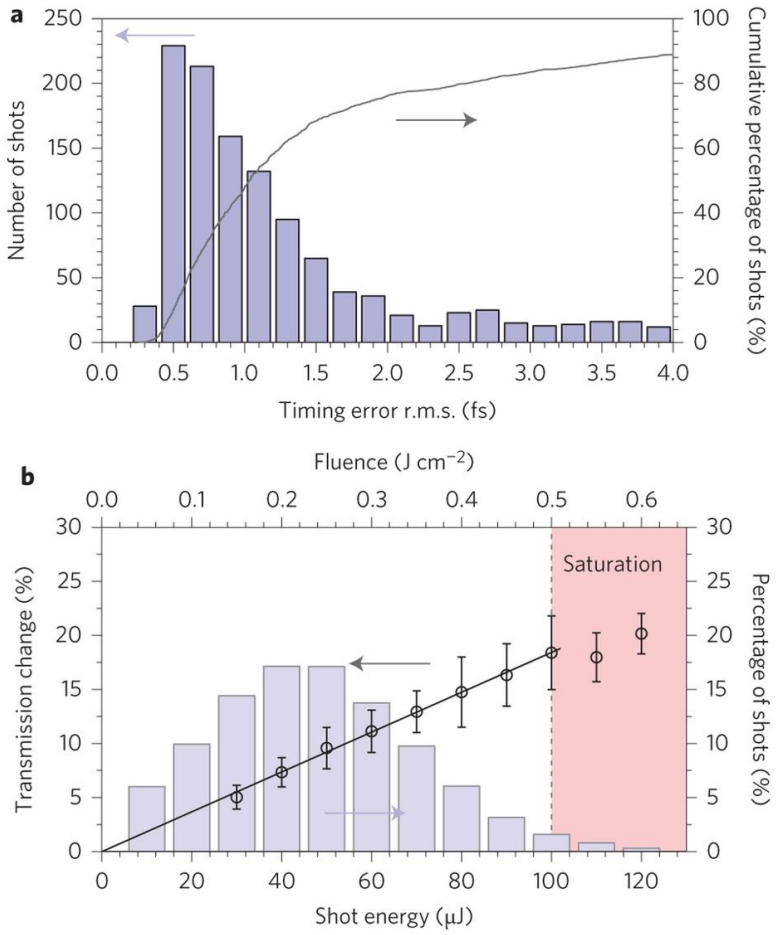




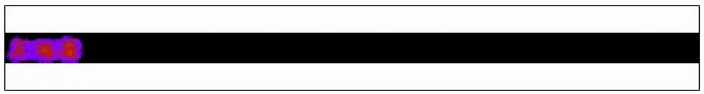
Fuji *et al.*, JCP **133**, 234303 (2010)

Ok… you don't have to believe me… but...





- It is at least conceivable to get sub-femtosecond
- I'm working on it… be patient…



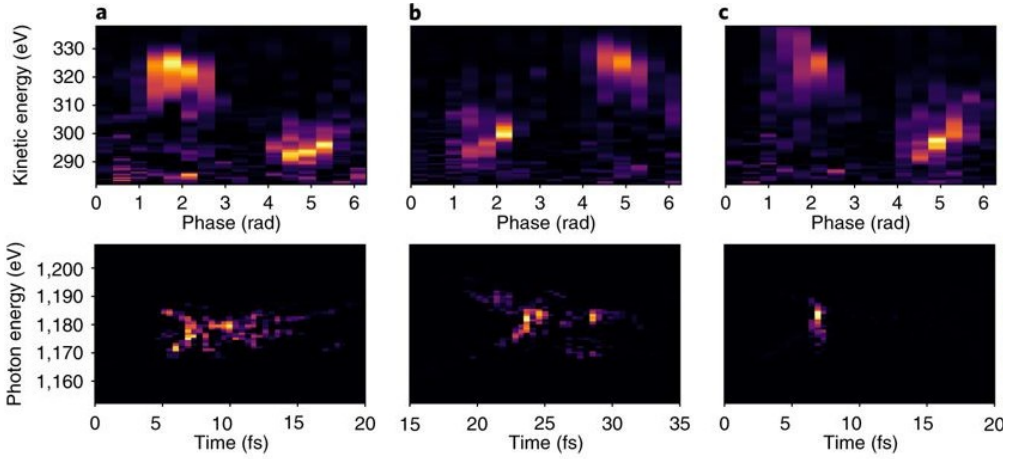
N. Hartmann *et al.,* Nat. Photon., **8**, 706 (2014)

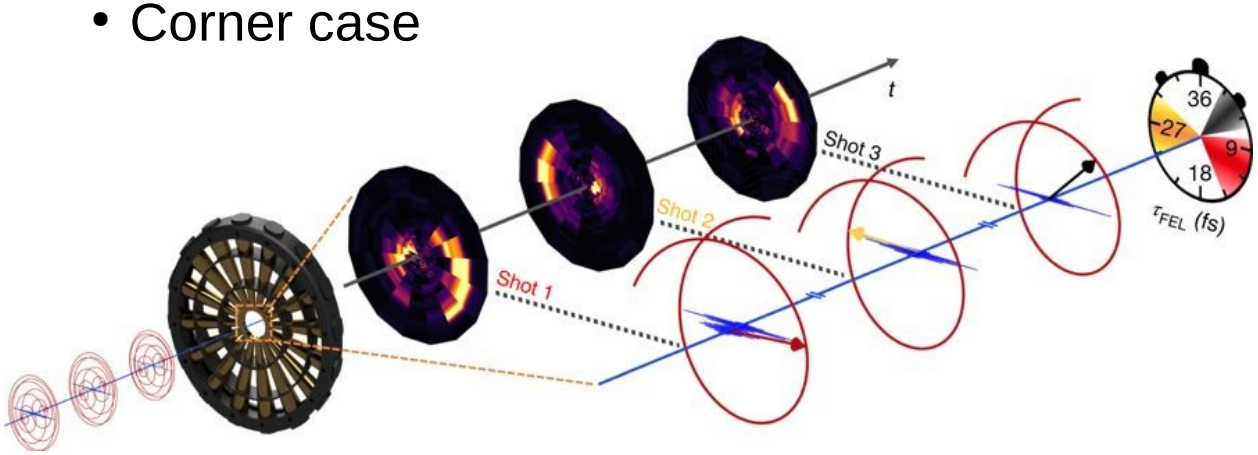
Outline

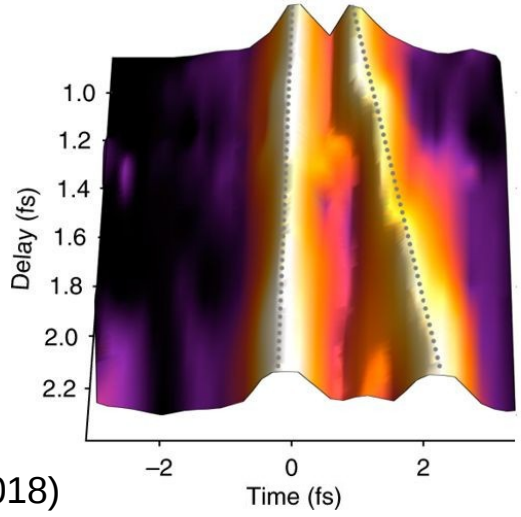
- The elephant in the room
- Coincidental alternatives
- Infrastructure
- Short term memory loss

A word on rare events … same new tricks

- Need the Time-Energy distribution of the x-ray pulse
- Forward simulation is trivial
- Inference engine can learn to solve the inverse problem
- Informs detector chain actions
  - Veto
  - Delay sorting
  - Corner case





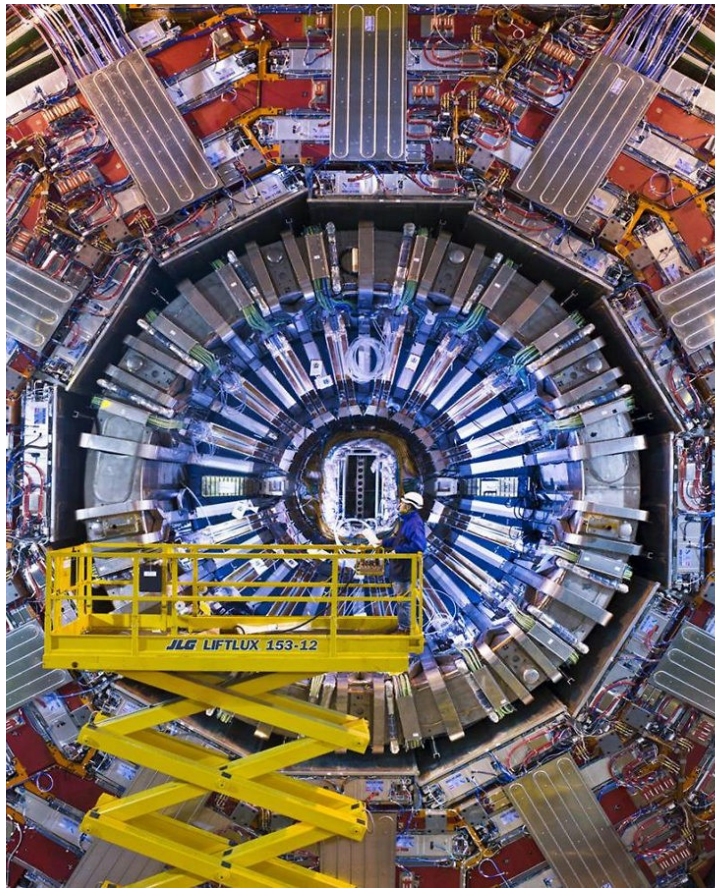


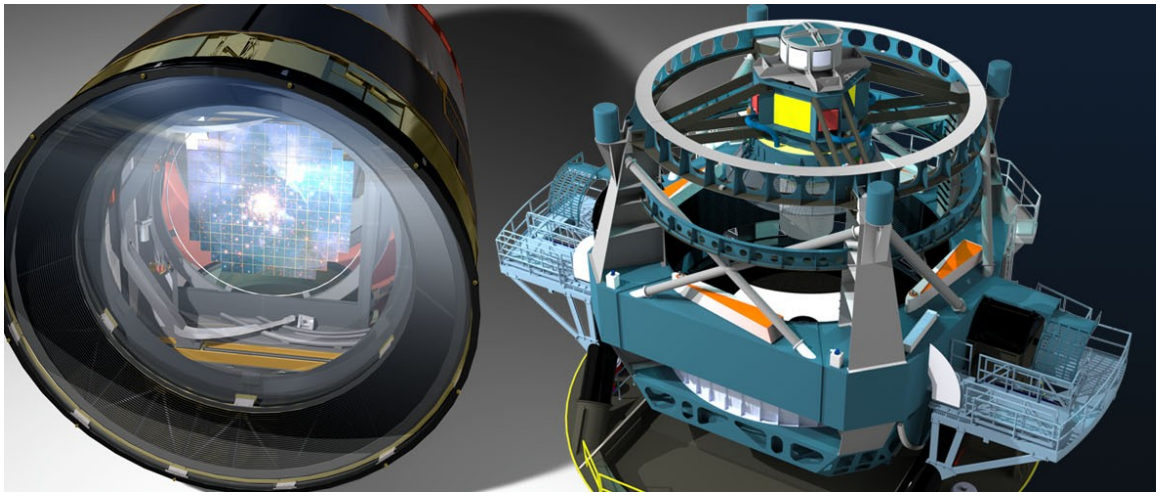
N. Hartmann *et al.,* Nat. Photon. **12** 215 (2018)

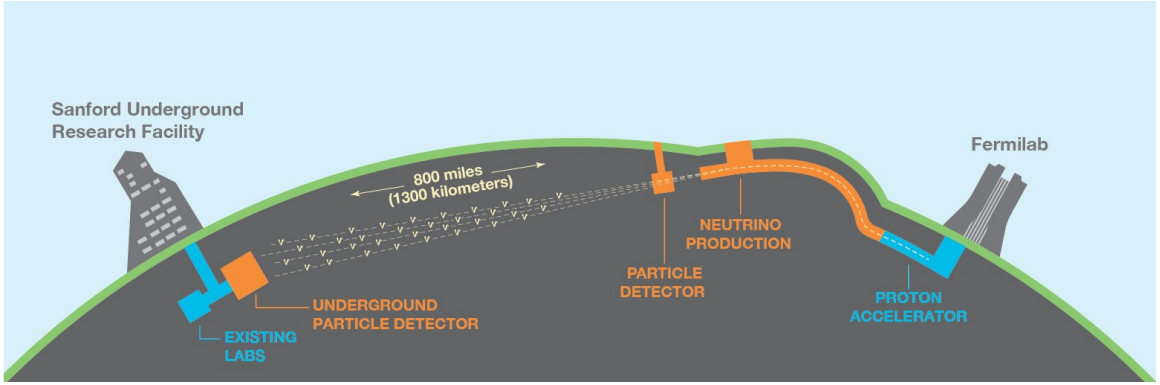
A word on rare events … same new tricks

SLAC

- Intelligent Triggers for ATLAS
- Context dependent models for LSST
- Streaming particle identification for DUNE



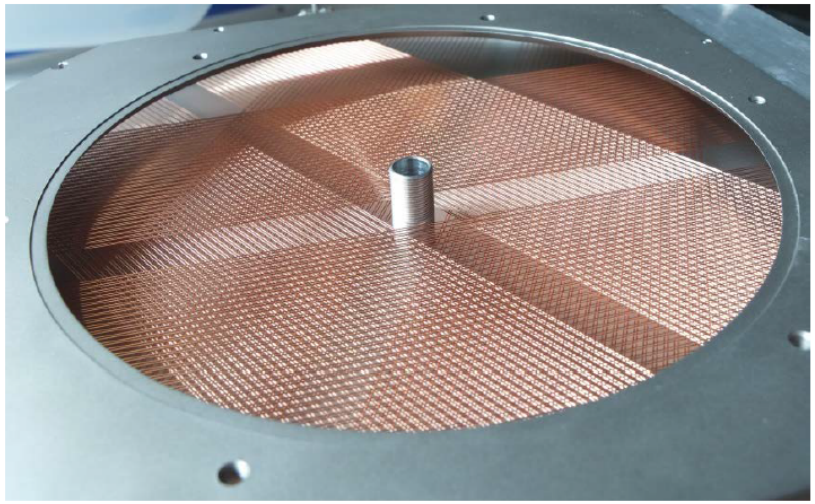




A word on rare events … same old clicks

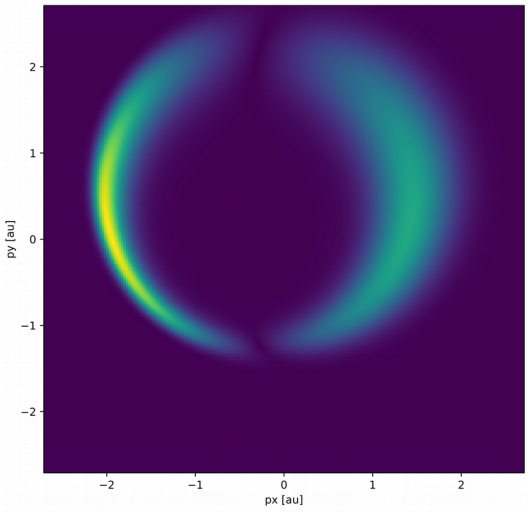
SLAC

RoentDek GmbH

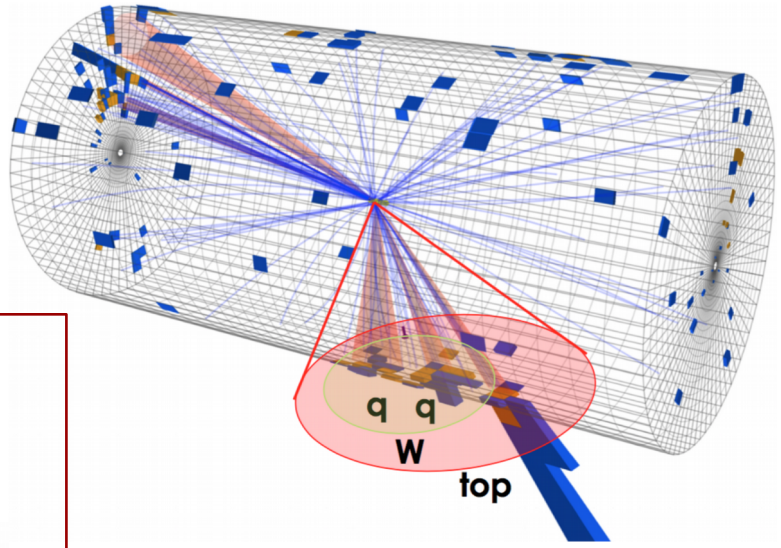


- Physicists make clicks on wires
- We have been quietly stealing from HEP for decades
  - Still weaving and winding wires
  - We moved our CFD+TDCs to ADCs + software out of näiveté
- Let's instead encourage a unified co-design body
  - Sensor design
  - Hardware
  - Software
  - Simulation
  - Science Domain

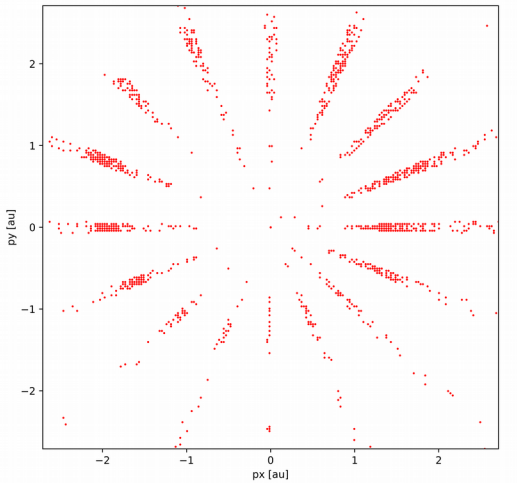
A word on rare events … same new tricks

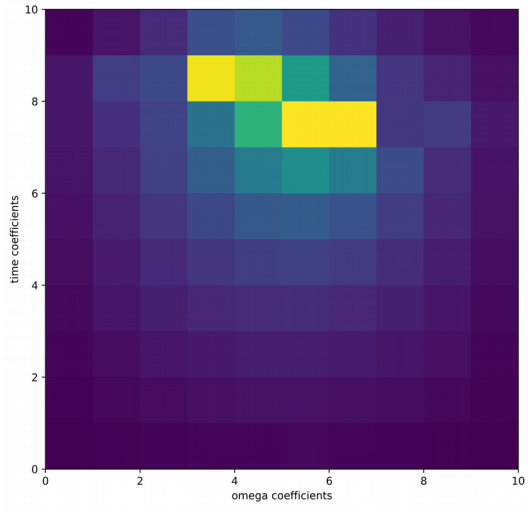


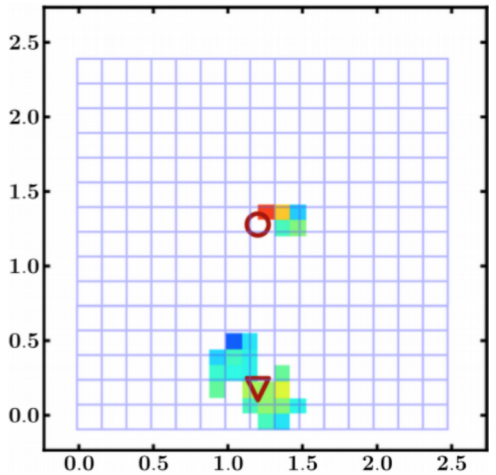
- Unwrapping LHC events



- Decompose into VN basis







Courtesy of Averell Gatton, Siqi Li, and James Cryan

Kagan, Schwartzman, *et al.,* 2016

Summary notes

SLAC

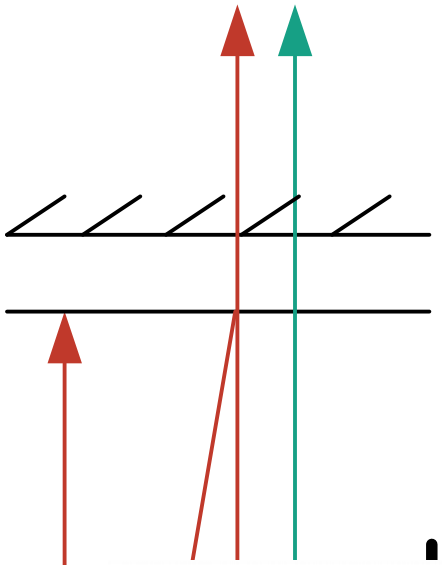
- I like using time-domain for energy and keeping spatial dimensions spatial
- Partner soft/hard beamlines (standard)
  - Common “setup” shaping optical laser tools
  - Common deep uv transform limited 0.5-2 fs pump pulses
- Complimentary soft/hard x-ray capabilities
  - 10 fs hard x-ray pulses, play with 6^{th} row elements
  - 0.5-2 fs soft x-ray pulses, multi-color, multi-polarization
- Every dimension (variational parameter) take a new view on the system, further constrains theory – delay, color, polarization
- Intelligent veto or anomoly detection to trigger on rare events
  - Always watch but rarely save
  - Likely needs some new thinking on detectors

Thank you!

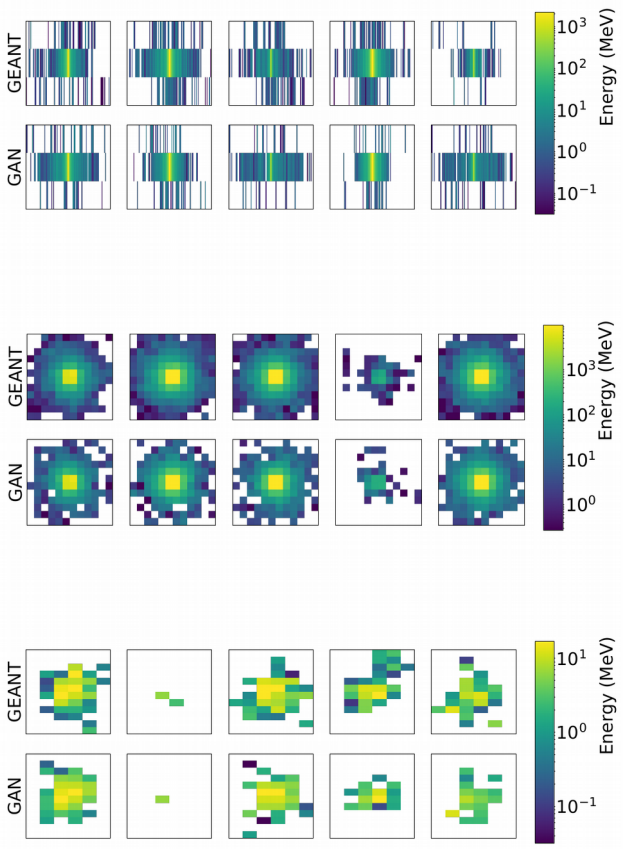
Serguei

Different domains … same new tricks

Paganini, de Oliveira, Nachman, PRL **120** 042003 (2018)



- Use Generative Adversarial Networks to turn cumbersome simulation into an "inference" problem
- The particle shower is a very deep decision tree



A Quantum Algorithm to Efficiently Sample from Interfering Binary Trees

Davide Provasoli,[1,*] Benjamin Nachman,[1,†] Wibe A. de Jong,[2,‡] and Christian W Bauer[1,§]

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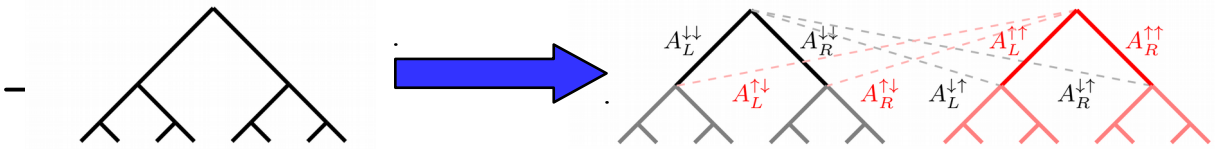
[2] *Computational Research Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA*

Quantum computers provide an opportunity to sample from probability distributions that include non-trivial interference effects between a large number of amplitudes of binary trees. Using a simple process wherein all possible state histories can be specified by a binary tree, we construct an explicit quantum algorithm that runs in polynomial time to sample from the process once. An interesting feature of these binary trees is that the they are not unitary, but can still be sampled on a quantum computer.



https://arxiv.org/pdf/1901.08148.pdf