# Ultrafast Molecular Imaging Using 4-Fold Covariance: Coincidence Insight with Covariance Speed

**Chuan Cheng**, Gönenç Moğol, Thomas Weinacht @Stony Brook
Felix Allum, Andrew J. Howard, Ruaridh Forbes, Philip H. Bucksbaum @Stanford/SLAC
Mark Brouard @Oxford
Leszek J. Frasinski @ICL
Daniel Rolles @KSU

# Outline

1. Introduction

- Multiparticle imaging with Coulomb Explosion Imaging (CEI)
- Coincidence vs covariance

2. Results: strong field ionization of deuterated formaldehyde $CD_2O$

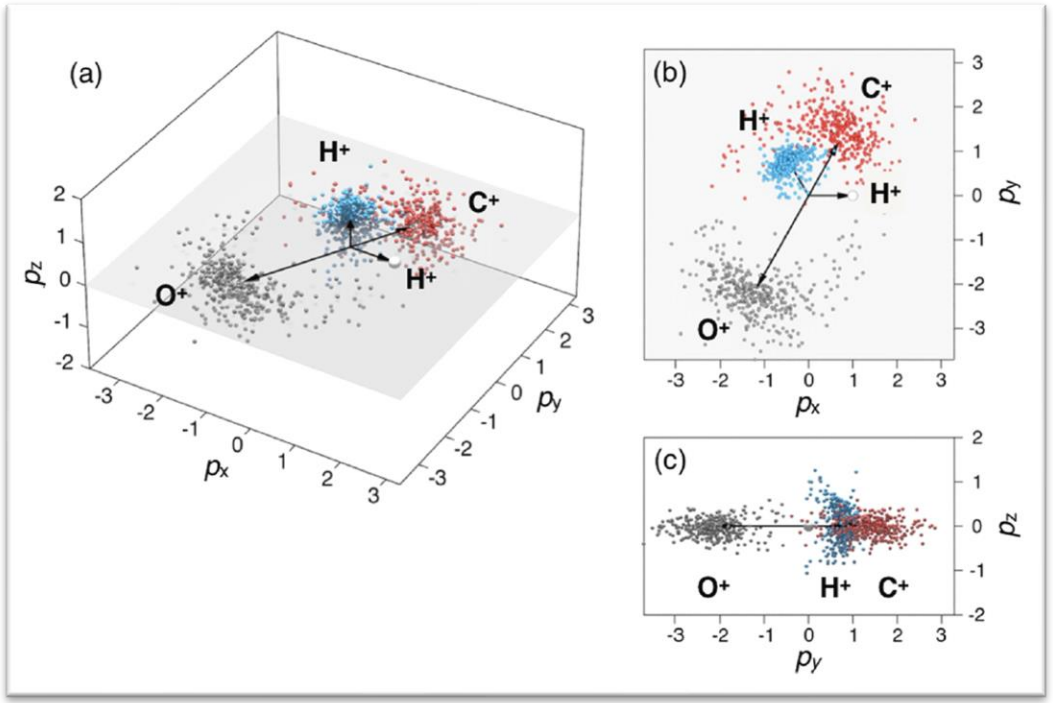
- 2-fold and 3-fold covariance
- 4-fold covariance

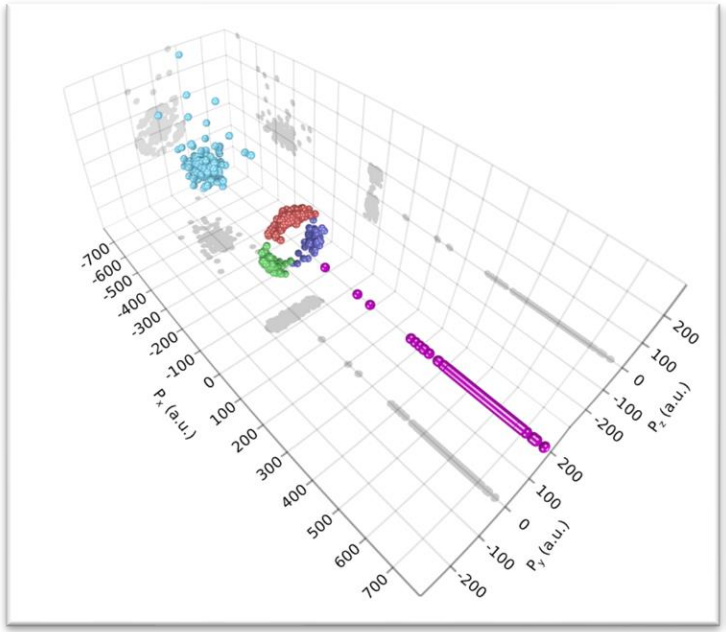
3. Summary

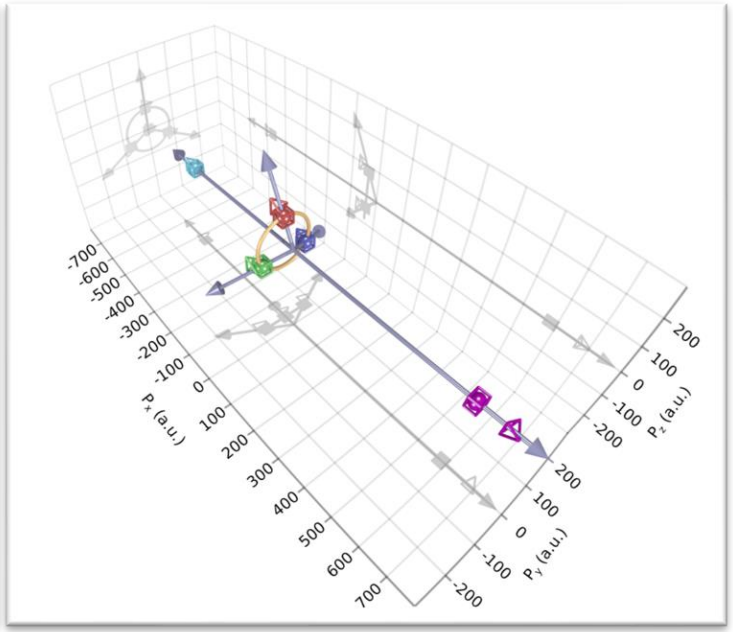
# 1. Introduction: Multiparticle imaging with Coulomb Explosion Imaging (CEI)

Target molecule: formaldehyde ($CH_2O$)
Light source: 800nm strong field ionization

Target molecule: iodomethane ($CH_3I$)
Light source: X-ray multiphoton ionization





*Li, Xiang, et al.* ***Physical Review Research 4.1 (2022): 013029.***
*Tseng, Chien-Ming, et al.* ***Journal of Electron Spectroscopy and Related Phenomena 228 (2018): 25-30.***

# 1. Introduction: coincidence

Channel (A,B,C,D) could come from:

- (A
- (B from
- (D fro
- (A, B from mo
- ……

1. Go for higher repetition rate -> 100kHz (currently 1kHz) -> more shots per time
2. Go for clever analysis -> covariance analysis

1. Suppress rate of ionizing two molecules

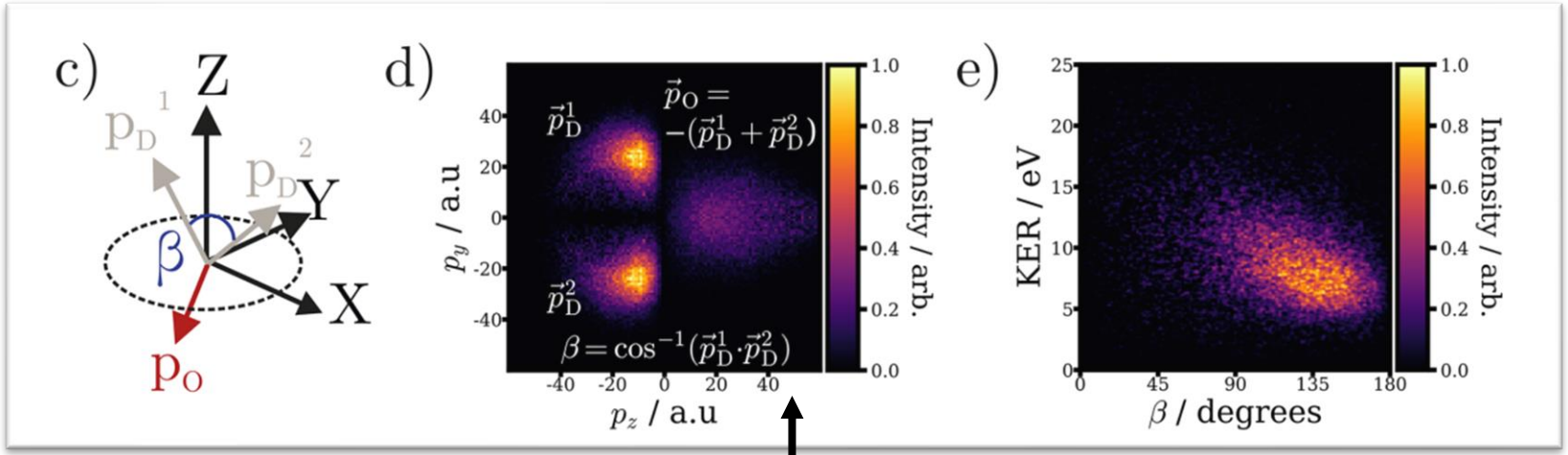
# 1. Introduction: covariance

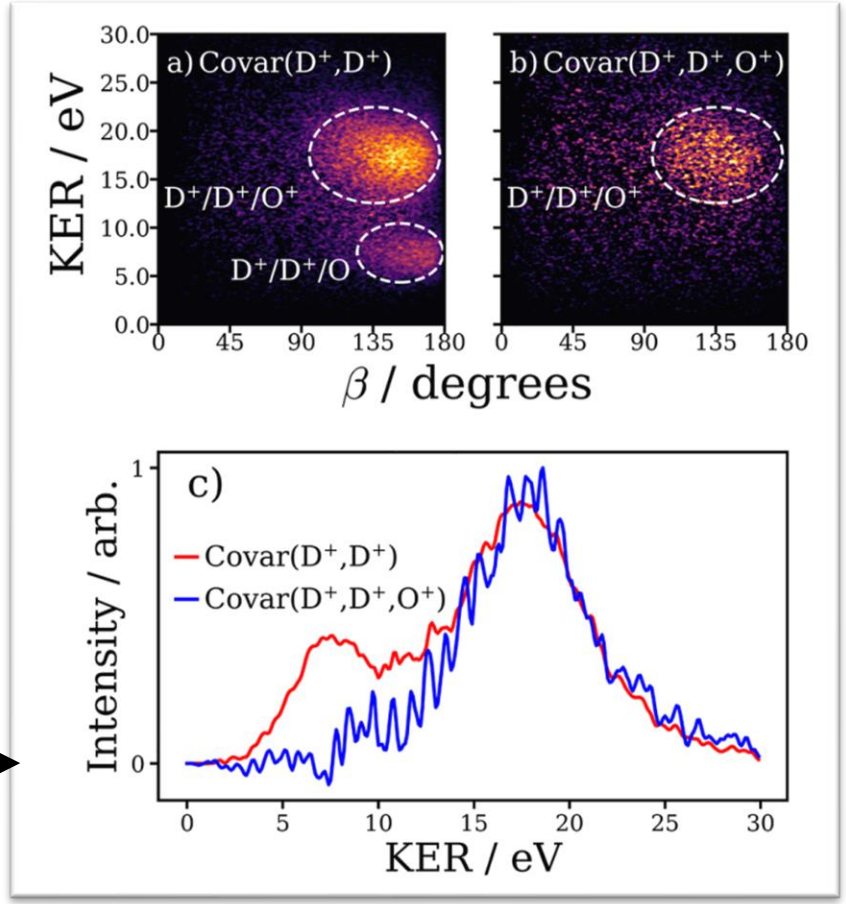
- $Cov(N_A, N_B) = \langle (N_A - \langle N_A \rangle)(N_B - \langle N_B \rangle) \rangle = \langle N_A N_B \rangle - \langle N_A \rangle \langle N_B \rangle$
- Where $N_{A(B)}$ = number of particle A (B) in each shot

$$\langle N_A \rangle = \frac{\sum_{all\ shots} N_A}{total\ shots}$$

- $Coin(N_A, N_B) = \langle N_A N_B \rangle|_{(N_A=1\ and\ N_B=1)}$

# 1. Introduction: covariance on deuterated water ionization




- Refer 3rd fragment from the 2 D+ ions momentum
- Direct comparison of 3 ions VS 2 ions
- How about higher charge ionization?



*Allum, Felix, et al.* ***The Journal of Physical Chemistry Letters 12.34 (2021): 8302-8308.***

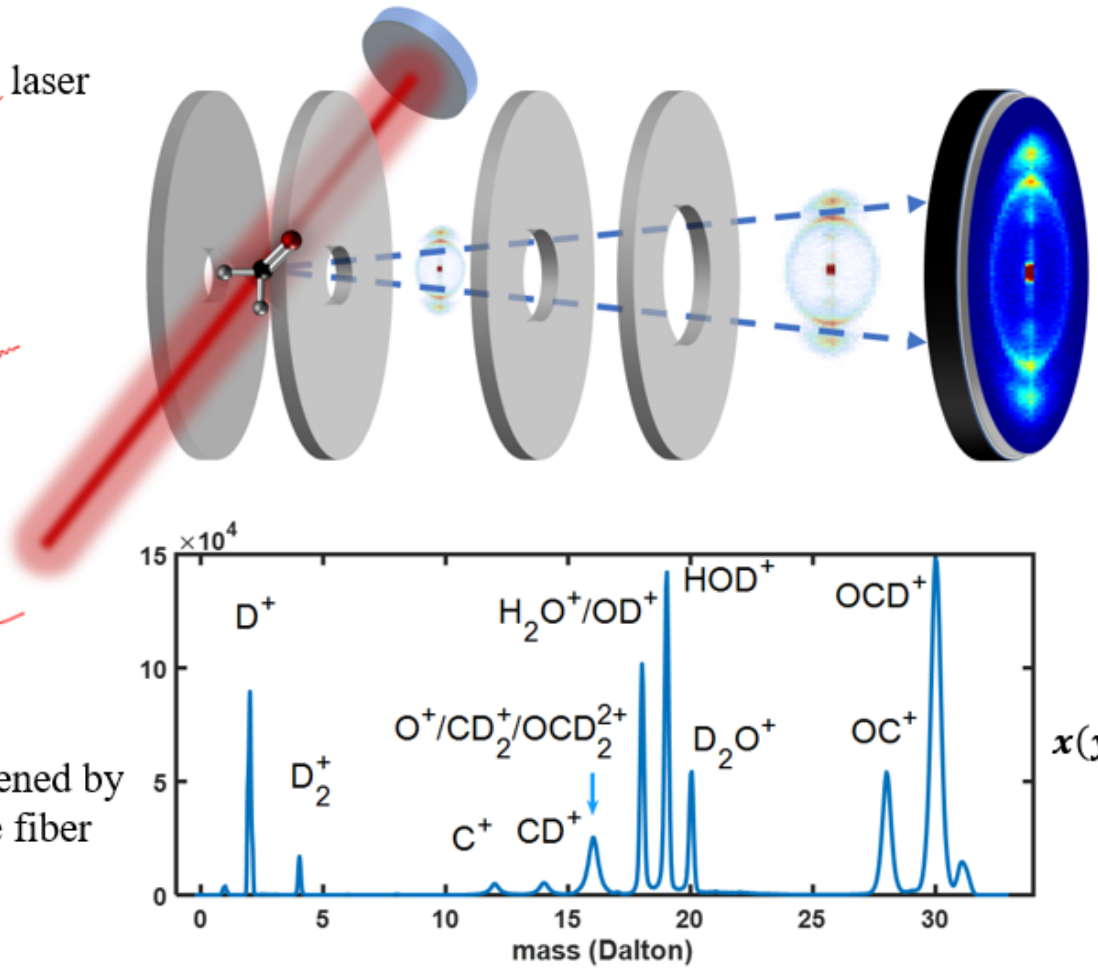
# 2. Experimental set up

Gönenç Moğol Z07.00007: 3D Velocity Map Imaging of Electrons

Ti:Sapphire laser
780nm
30fs
5-30mW

Or

Laser broadened by
Hollow core fiber
780nm
6fs
5-45mW [1]

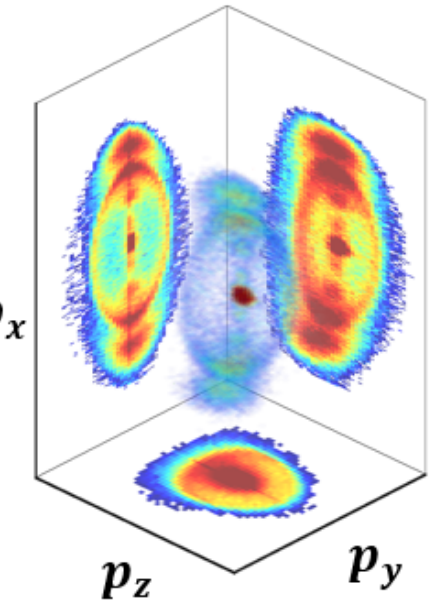






$$x(y) = \frac{L}{\sqrt{2mqU}} p_x(p_y) \propto p_x(p_y)$$

$$\Delta t = \frac{d}{qU_0} p_z \propto p_z \text{ [2]}$$

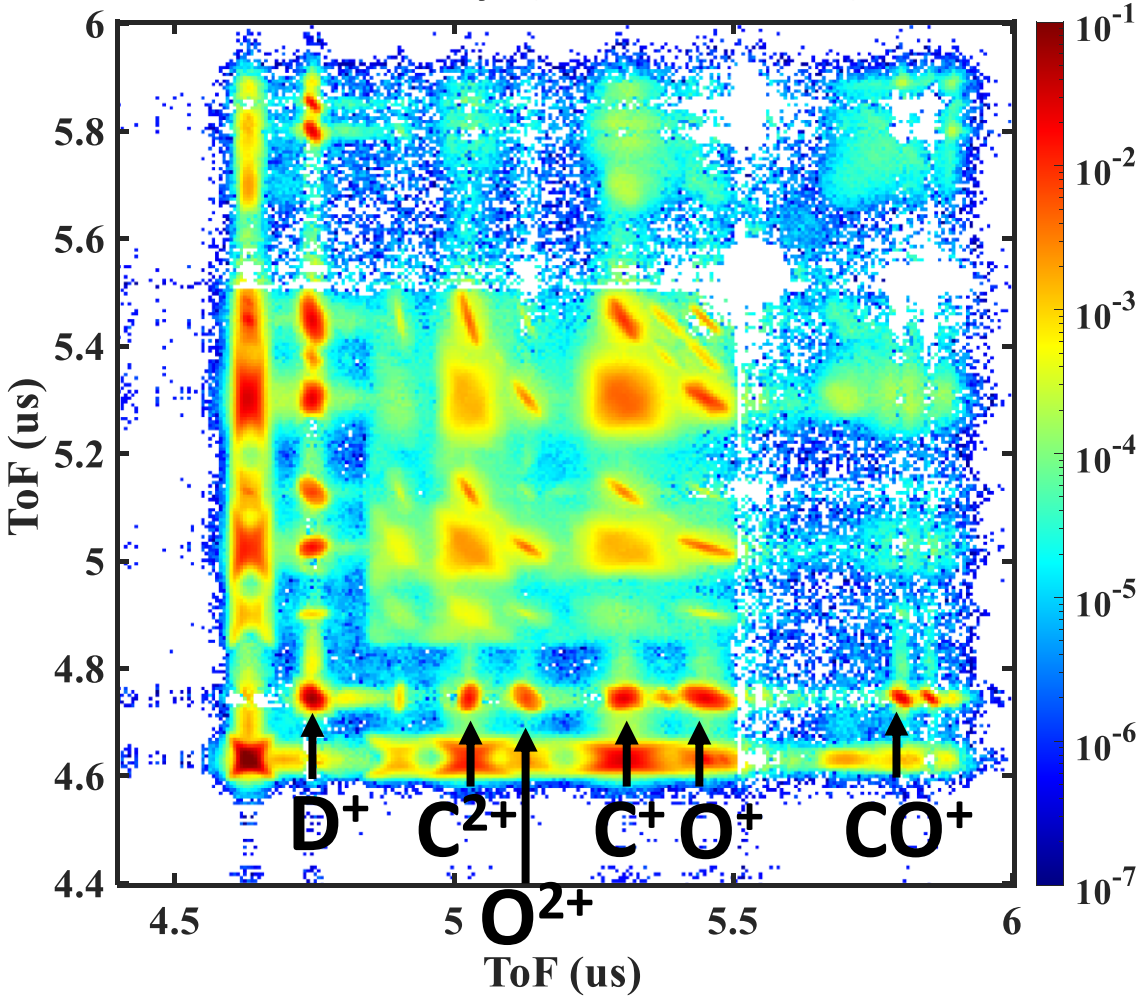




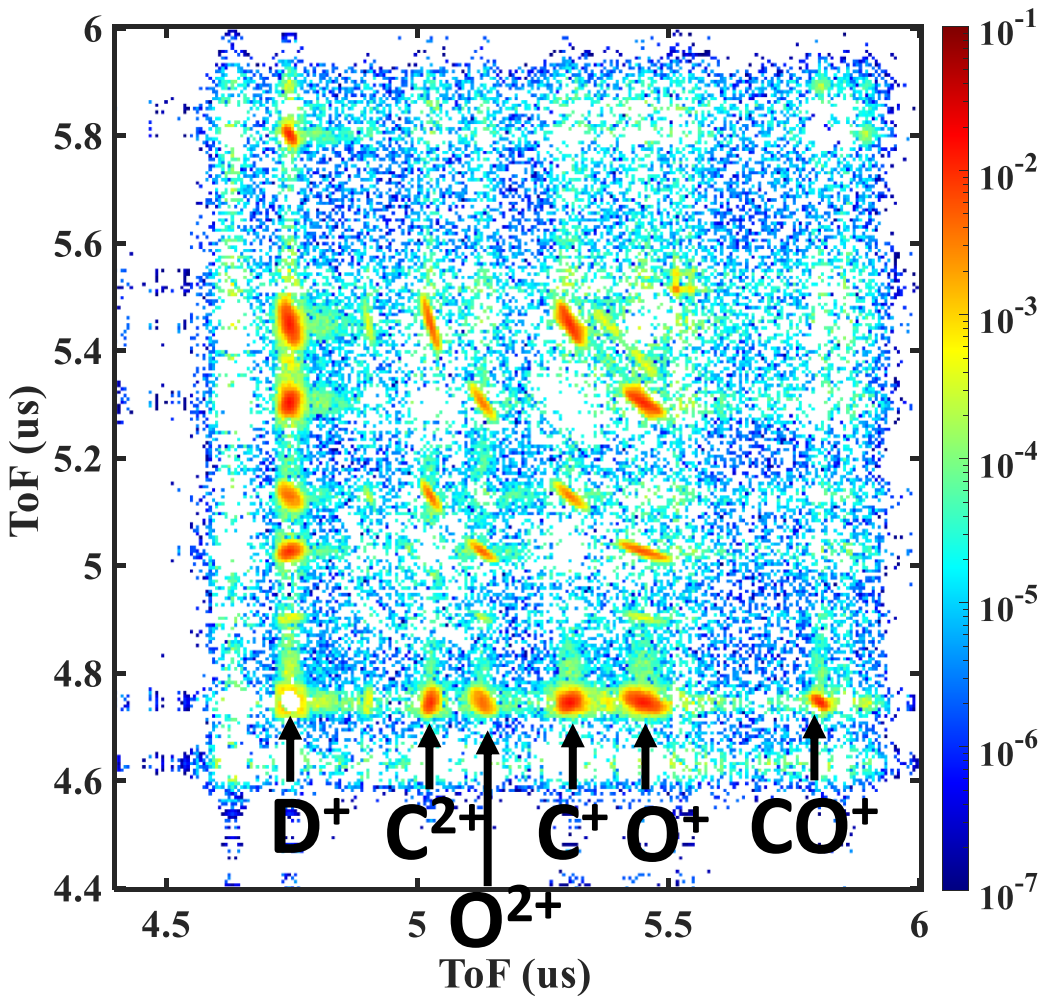
*[1] Catanese, Anthony, et al.* ***OSA Continuum 4.12 (2021): 3176-3183.***
*[2] Cheng, Chuan, et al.* ***Review of Scientific Instruments 93.1 (2022): 013003.***

# 2. Results: ToF-ToF covariance ($D^+$, $D^+$, $C^{n+}$, $O^{n+}$)

2body (ion1, ion2)




3body (ion1, ion2| $D^+$)



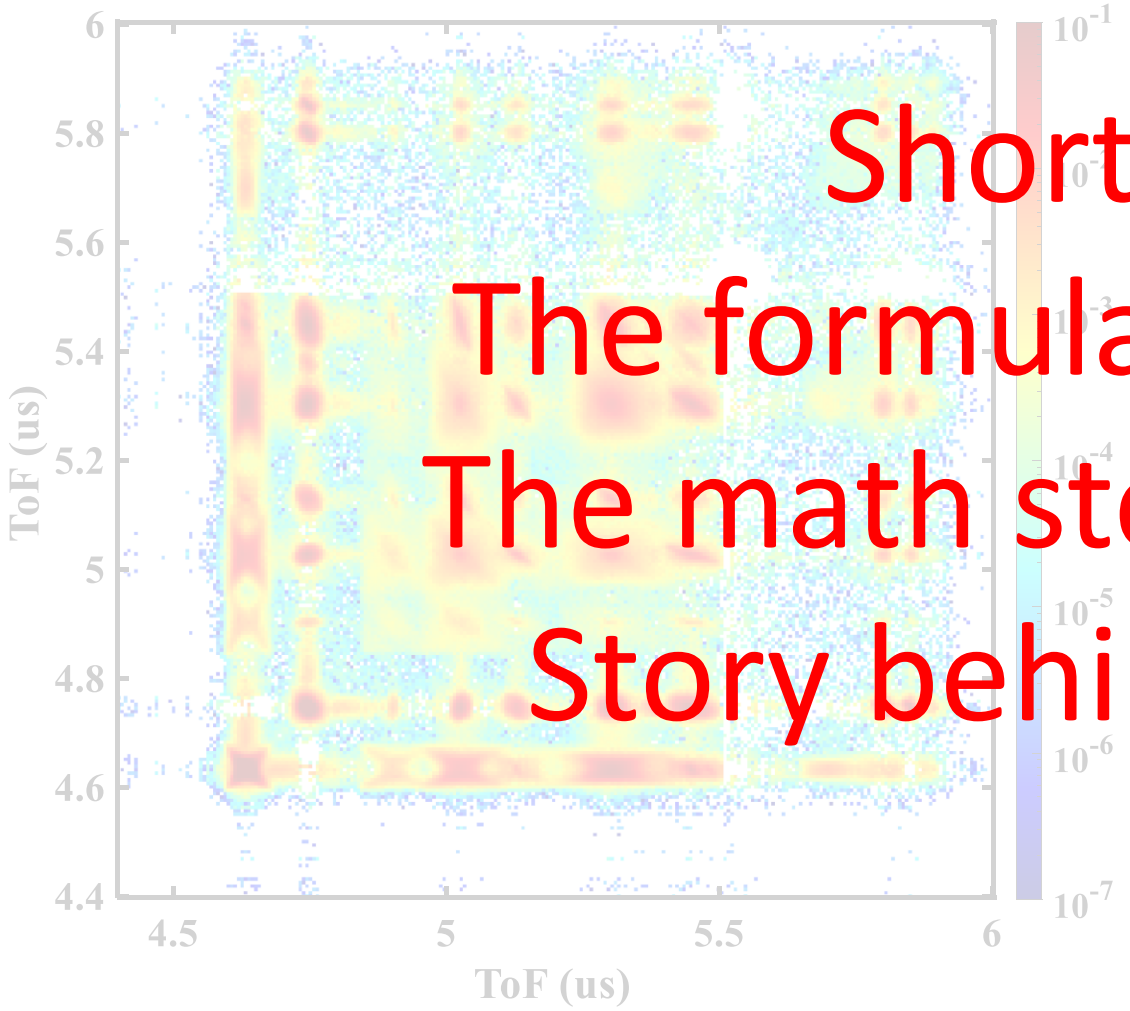
# 2. Results: ToF-ToF covariance

- 2-fold: $Cov(N_A, N_B) = \langle (N_A - \langle N_A \rangle)(N_B - \langle N_B \rangle) \rangle$
- 3-fold: $Cov(N_A, N_B, N_C) = \langle (N_A - \langle N_A \rangle)(N_B - \langle N_B \rangle)(N_C - \langle N_C \rangle) \rangle$

- So:
- N-fold: $Cov(N_A, N_B, N_C, \dots) = \langle (N_A - \langle N_A \rangle)(N_B - \langle N_B \rangle)(N_C - \langle N_C \rangle) \dots \rangle$
- 4-fold: $Cov(N_A, N_B, N_C, N_D) = \langle (N_A - \langle N_A \rangle)(N_B - \langle N_B \rangle)(N_C - \langle N_C \rangle)(N_D - \langle N_D \rangle) \rangle$

# 2. Results: ToF-ToF covariance

## 4body (ion1, ion2| $D^+$, $D^+$)




Short Story is:

The formula is incomplete.

The math story is a bit long.

Story behind derivation.

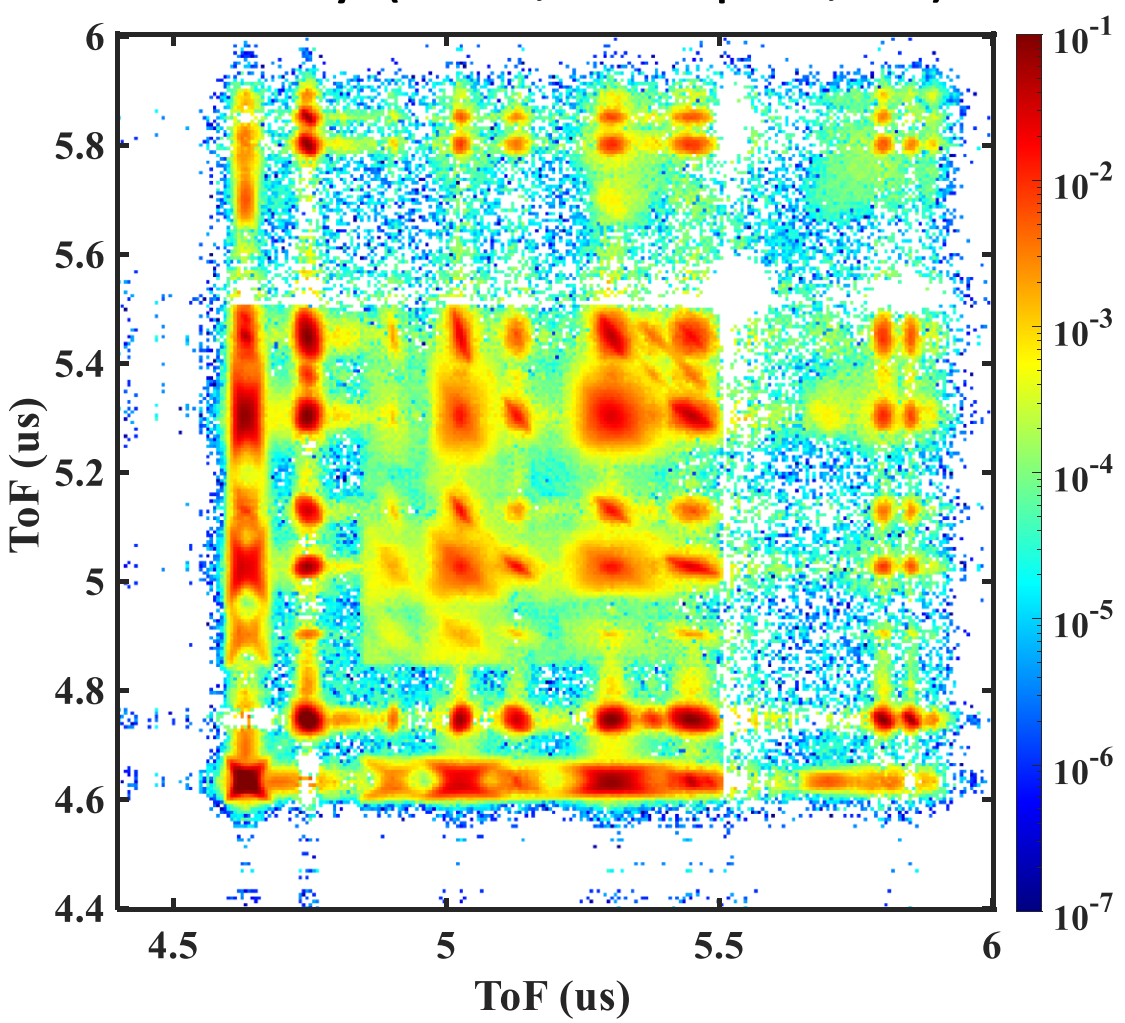
Why more peaks than 3-fold?

Why do peaks look blurred?

Frasinski, Leszek J. ***arXiv preprint arXiv:2204.03709*** **(2022).**

# 2. Results: 4-fold ToF-ToF covariance

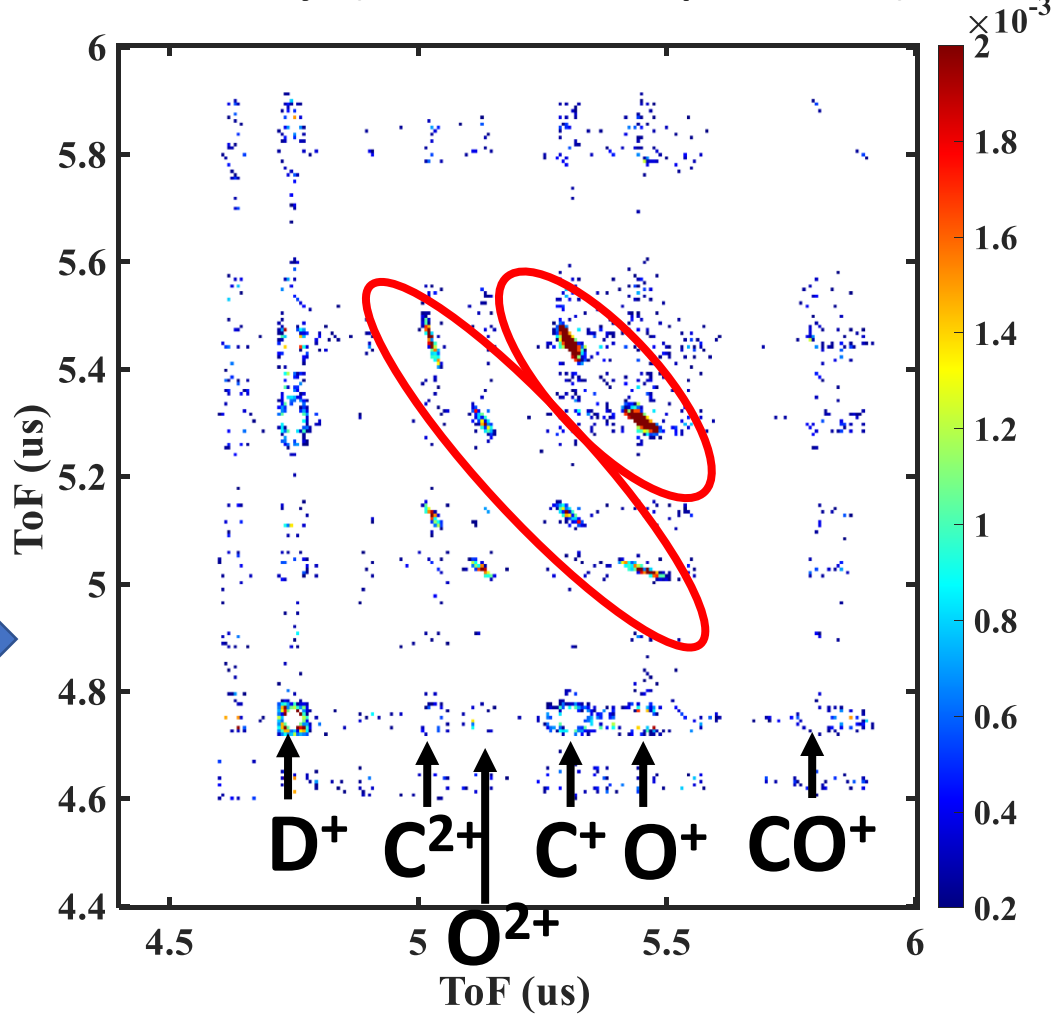
4body (ion1, ion2| $D^+, D^+$)




$Cov(N_A, N_B, N_C, N_D)$

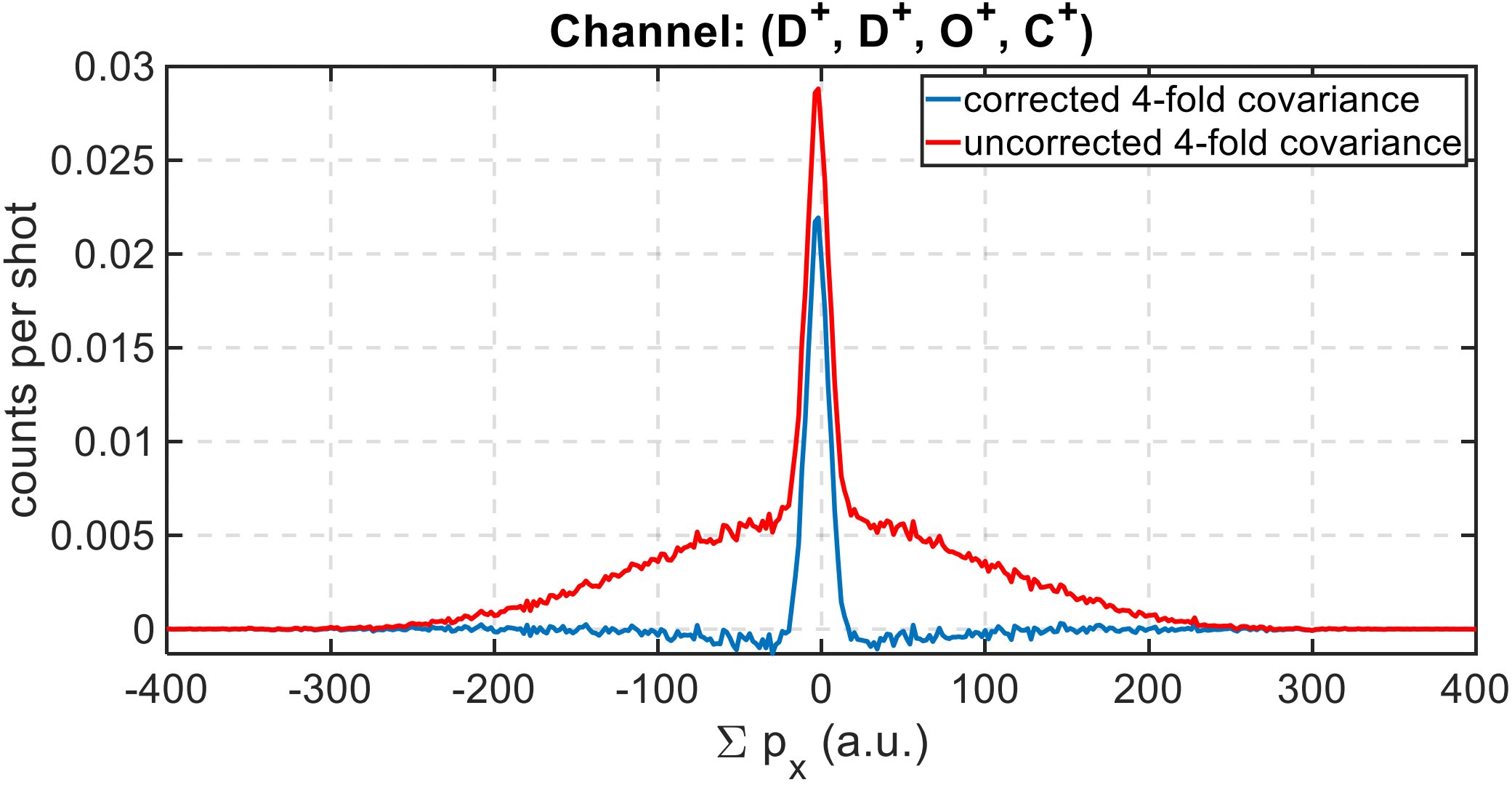
Cleaned out with right formula

4body (ion1, ion2| $D^+, D^+$)



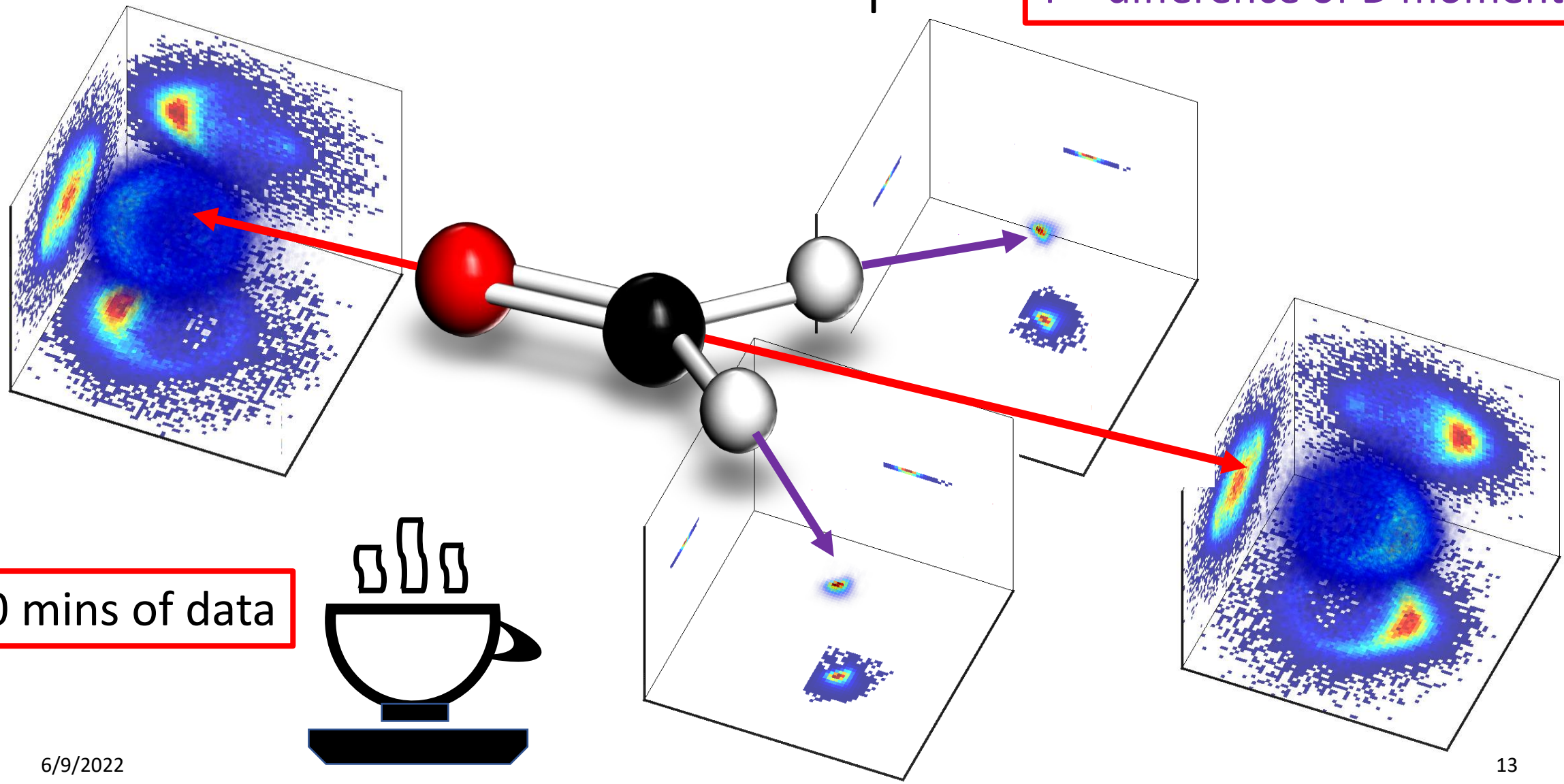

$Cov(N_A, N_B, N_C, N_D) - [Cov(N_A, N_B)Cov(N_C, N_D) + sym]$

# 2. Results: 4-fold momentum conservation



# 2. Results: 4-fold Newton plot

X+ = bisector of D momentum
Y = difference of D momentum



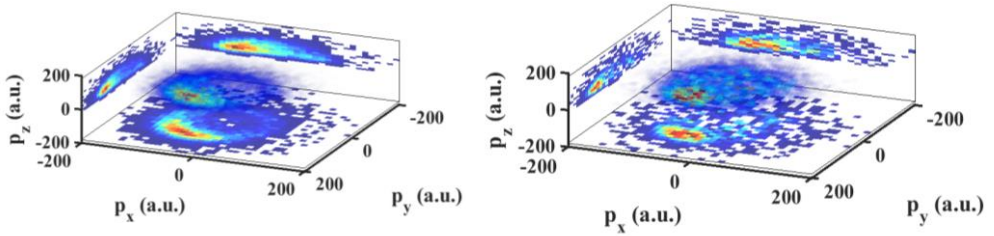
20 mins of data

# 3. outlook: speed of the code and further experiments

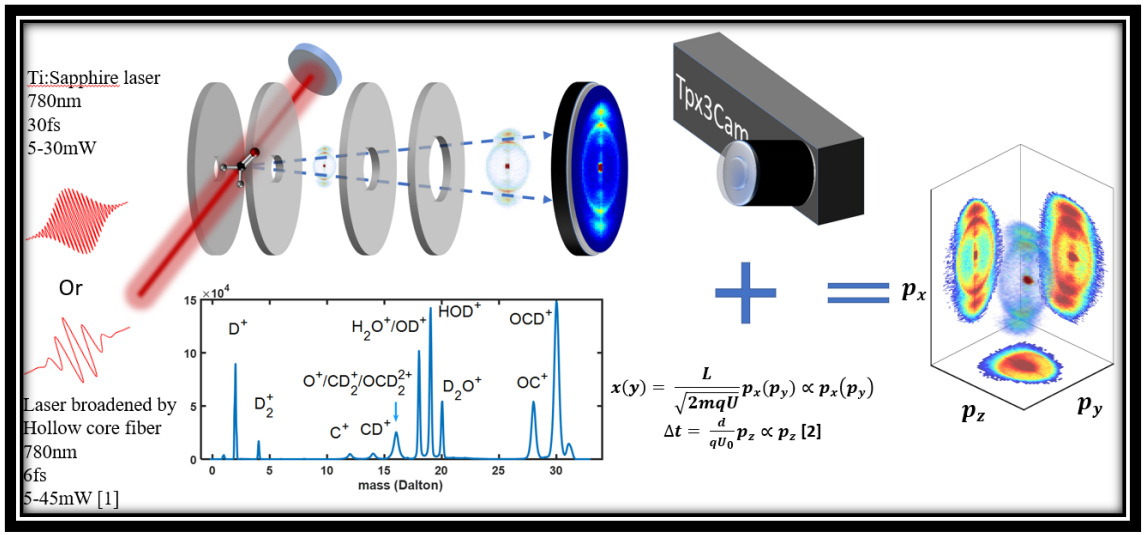
- Data stream time scales:
  - Initial data set: 20 mins
  - Centroiding and other pre-analysis: 20mins - 1 hour
  - Load data in Matlab and get 2/3/4-fold ToFToF covariance: 1 min
  - Do Newton plot type of analysis: 5 mins - 10 hours depending on algorithms, also memory consuming. May apply machine learning

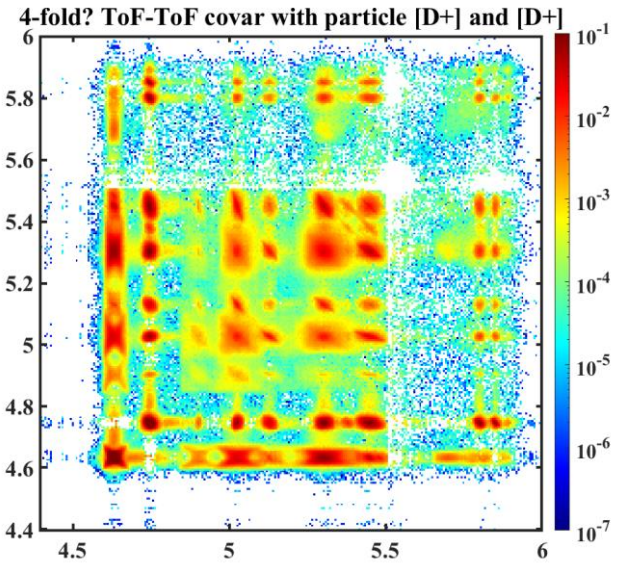
- Further experiments (4-fold covariance):
  - Long (30fs) vs short pulse (6fs)
  - 6fs pump probe
  - Isotope effects
  - Vary the count rate to test how much data needed

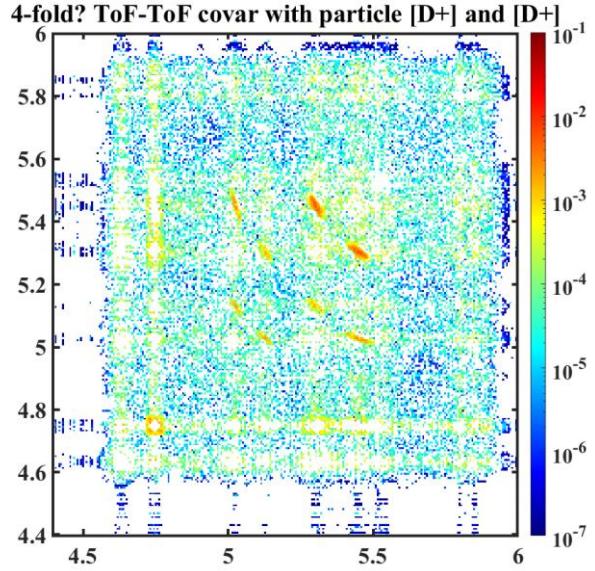
Andrew J Howard V01.00143: Imaging Molecular Motion During the Strong-Field Enhanced Ionization of Water



# 3. summary



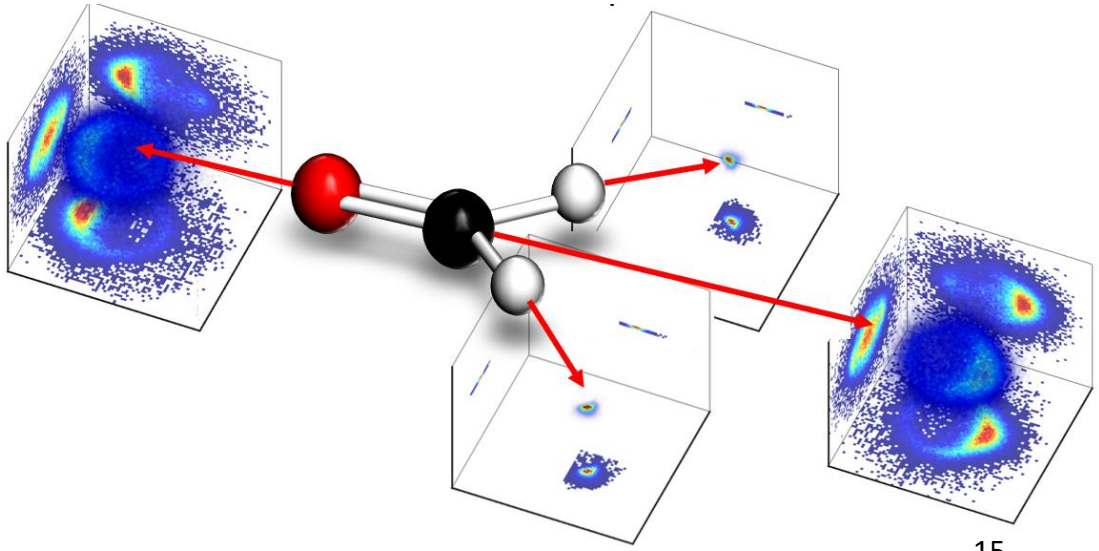



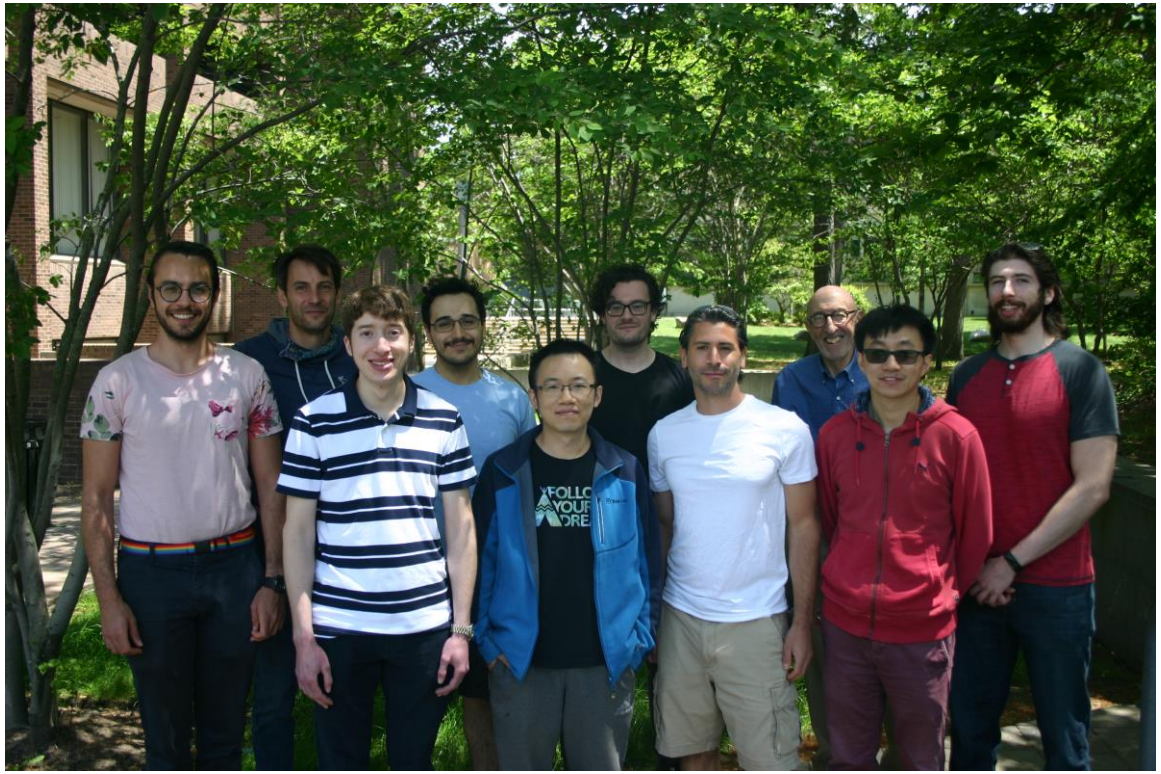




$Cov(N_A, N_B, N_C, N_D)$

Right recipe is crucial

$Cov(N_A, N_B, N_C, N_D) - [Cov(N_A, N_B)Cov(N_C, N_D) + sym]$





Thomas Weinacht Group @Stony Brook University
Back: Thomas Weinacht, Denis Aglagul, Eric Jones, Martin Cohen, Brian Kaufman
Front: Gönenç Moğol, Samuel McClung, Chuan Cheng, Anthony Catanese, Yusong Liu

@Stanford University and SLAC

Philip H. Bucksbaum

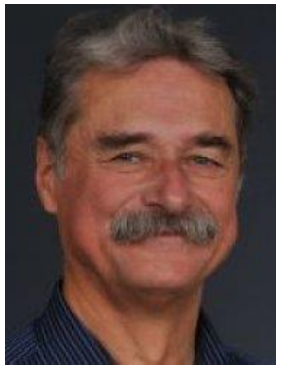
Ruaridh Forbes

Felix Allum

Andrew J. Howard

| | |
|---|---|
| Spiridoula Matsika | @Temple University |
| Vaibhav Singh | @Temple University |
| Michael Spanner | @Ottawa University |
| Zachary Streeter | @AMD Inc. |
| C. William McCurdy | @LBNL |
| Robert R. Lucchese | @LBNL |

Mark Brouard
@Oxford



Leszek J. Frasinski
@ICL

Daniel Rolles
@KSU

# Thanks for listening!
# Welcome for questions and more collaborations!