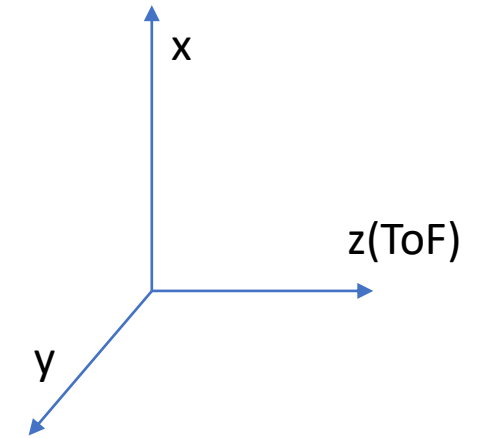
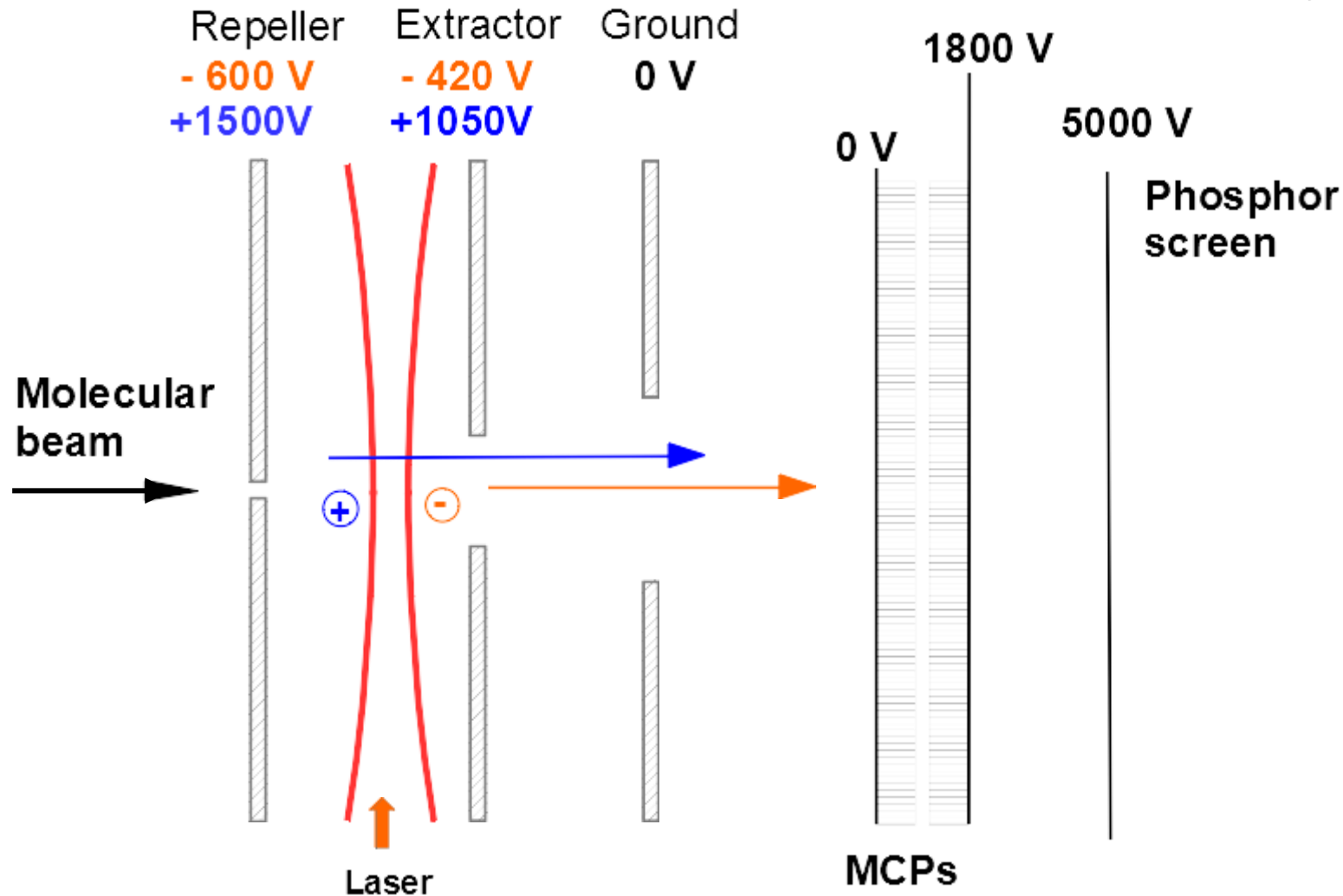


Introduction to covariance analysis technique – in contrast to coincidence analysis

Chuan Cheng

2021/09/14

1. Power of coincidence analysis

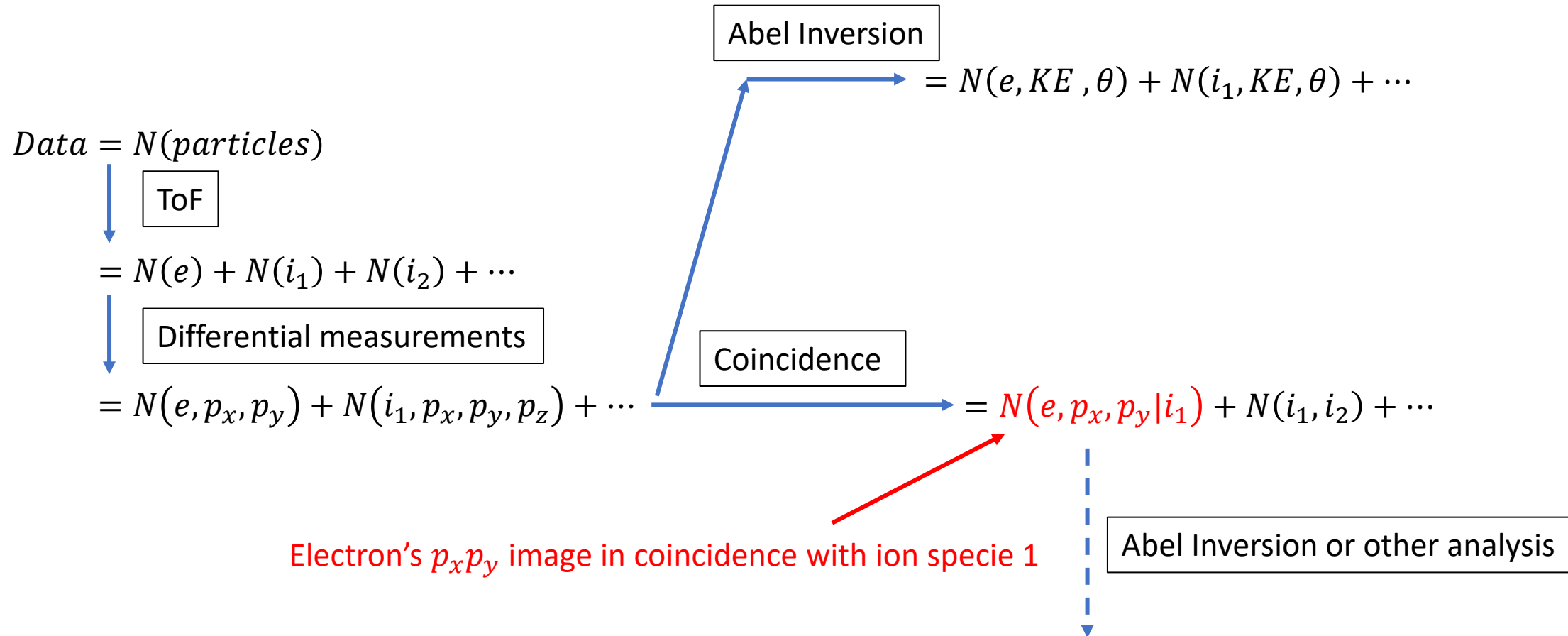


- Multiple particles coincidence
- multiple dimensional measurements

$$i^+ : p_x, p_y, p_z \left(\frac{m}{q} \right)$$

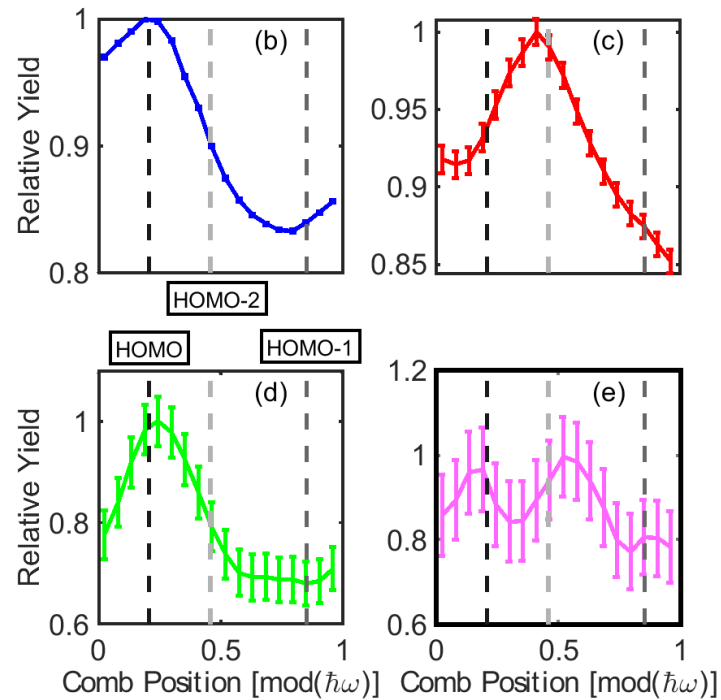
$$e^- : p_x, p_y$$

1. Power of coincidence analysis

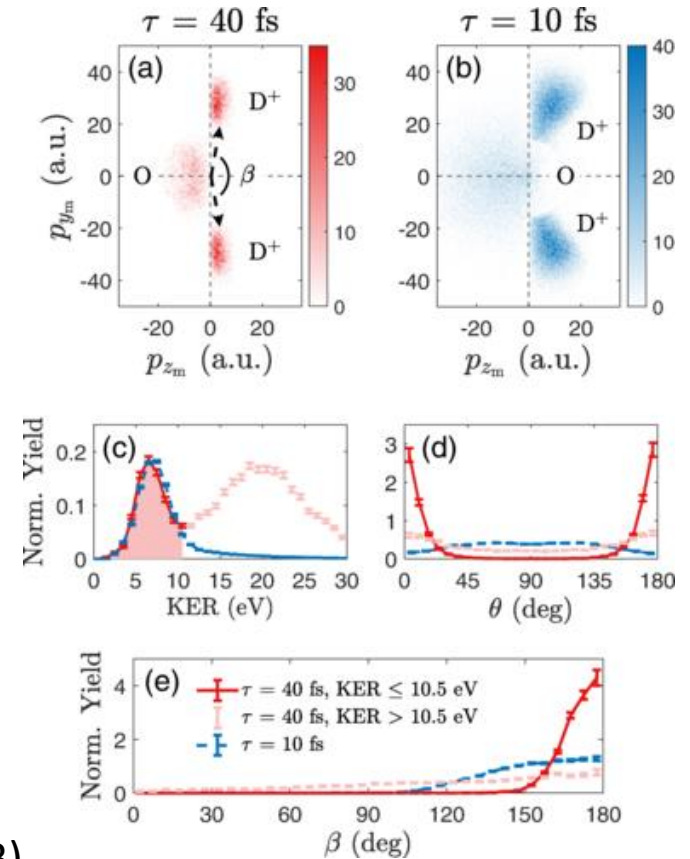
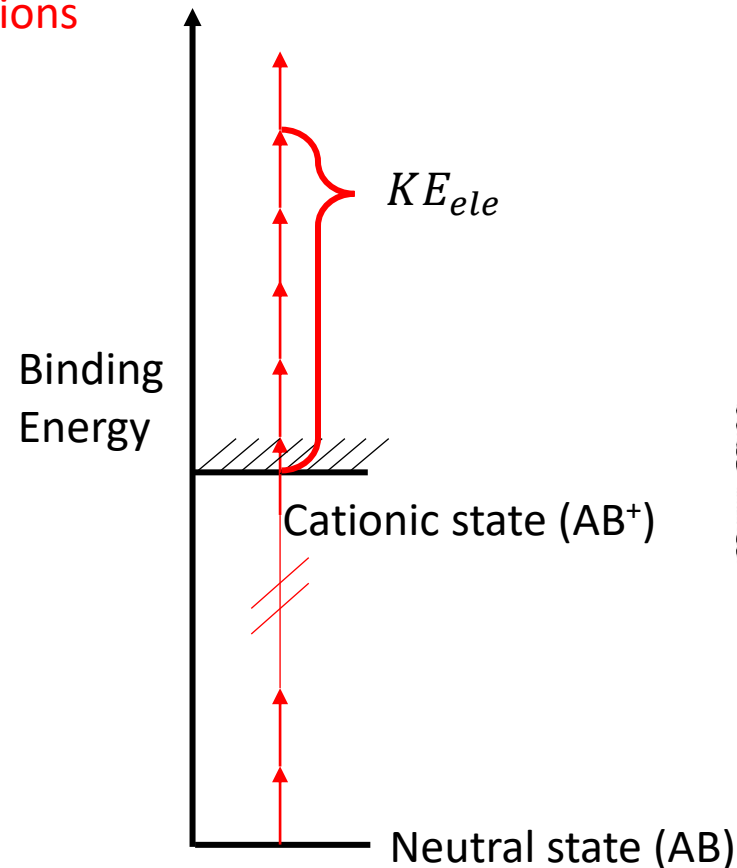


1. Power of coincidence analysis

Photoelectron in coincidence with different ions



Photoion correlations arising from multi-ionization



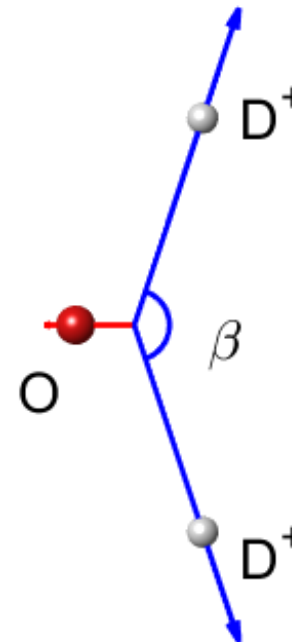
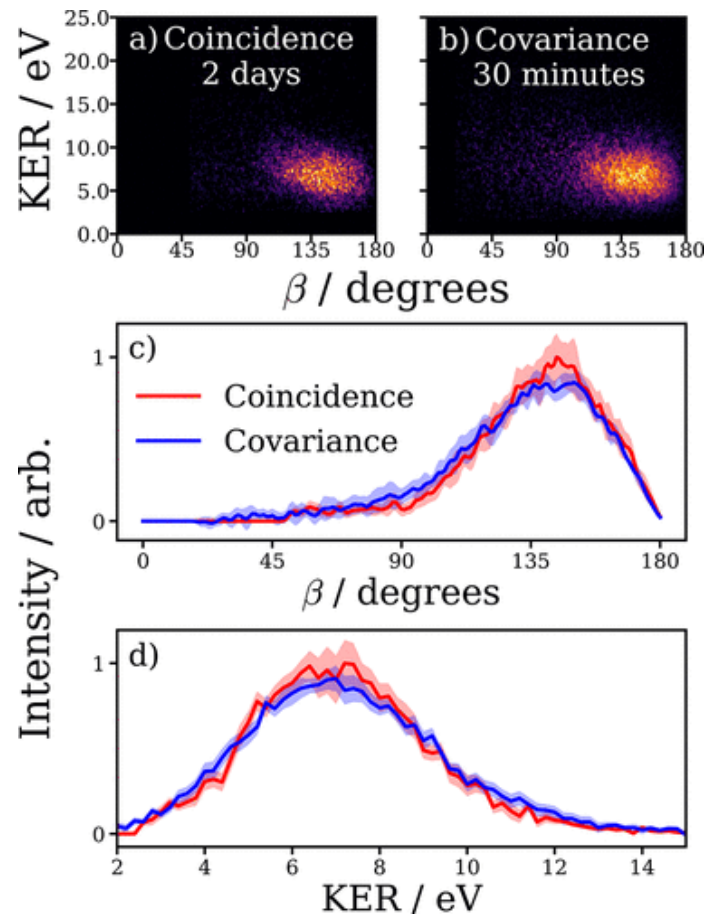
Cheng, Chuan, et al. "Momentum-resolved above-threshold ionization of deuterated water." *Physical Review A* 102.5 (2020): 052813

9/15/2021

Howard, A. J., et al. "Strong-field ionization of water: Nuclear dynamics revealed by varying the pulse duration." *Physical Review*

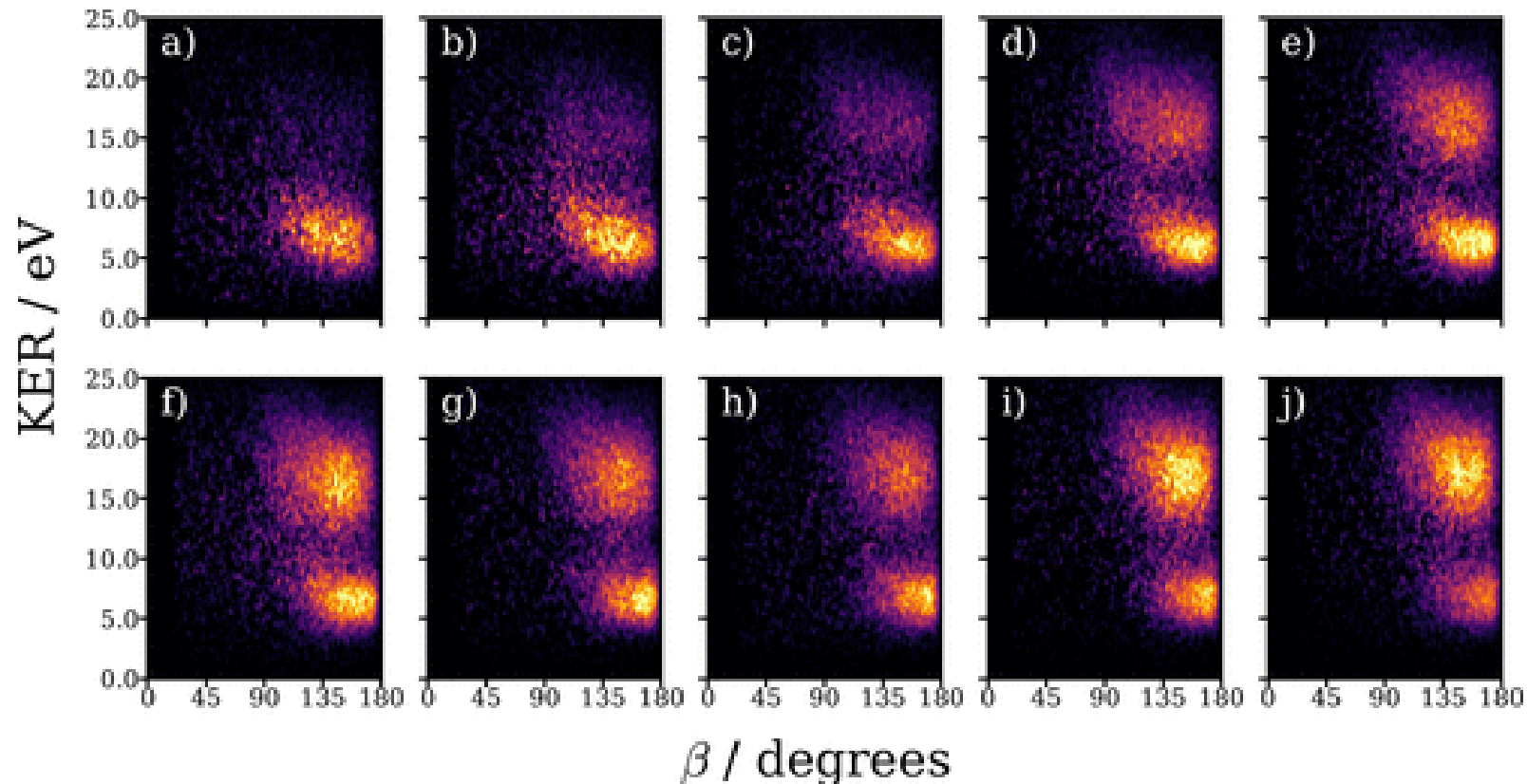
A 103.4 (2021): 043120.

1. Power of coincidence analysis – coincidence power for covariance price



- Coin vs covar analysis in ion-ion events
- Boost factor = $2\text{days}/30\text{mins} \approx 100$

1. Power of coincidence analysis – coincidence power for covariance price



- Coin vs covar analysis in ion-ion events

- Boost factor = 2days/30mins ≈ 100

- Intensity scan = 20mins*10 ≈ 3 hrs

- Question: why it seems to work so well?

Based on agreement between coincidence and covariance

- Coincidence analysis
- Agreement between coincidence and covariance



- Expectation of coincidence (math)
- Constraints in speed

2. Expectation in coincidence analysis

- The yield of (e, i_1) events should follow:

Normalized channel branching ratio



$$W_{coin} = \nu_0 * f(e, i_1) * \xi_e \xi_{i_1}$$

← Detection efficiency for electrons and ions

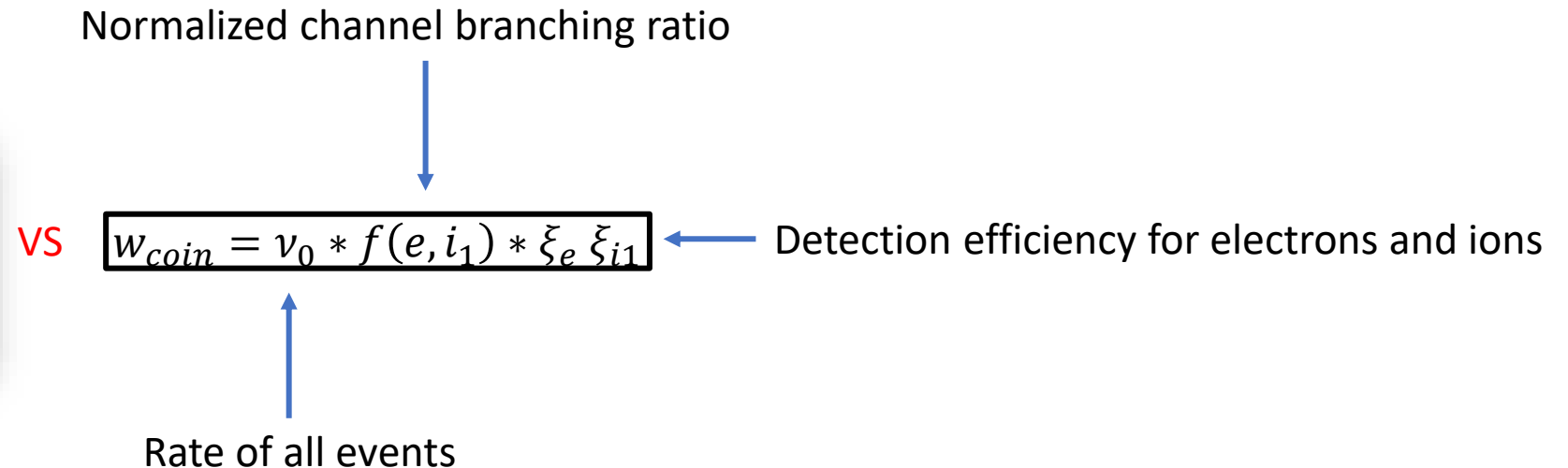


Rate of all events

2. Expectation in coincidence analysis

- The yield of (e, i_1) events should follow:

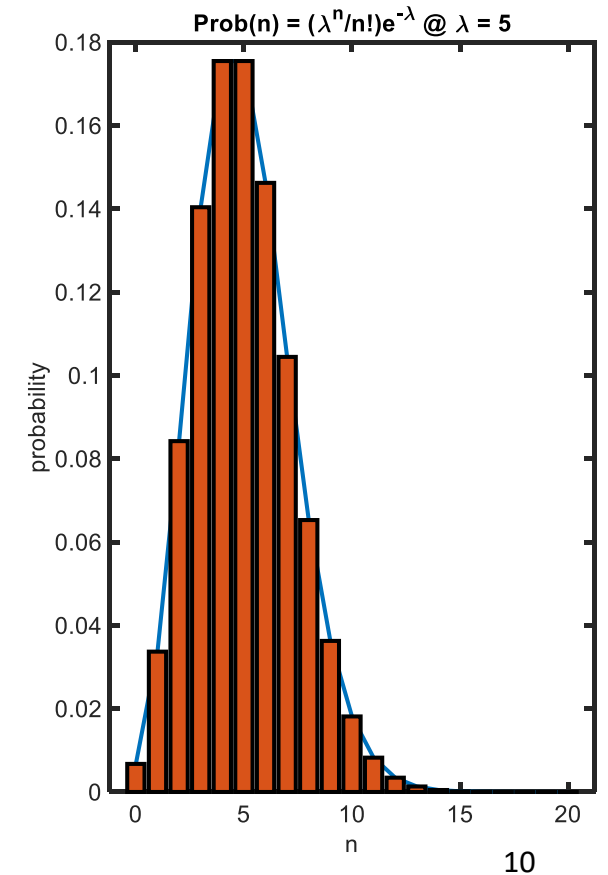
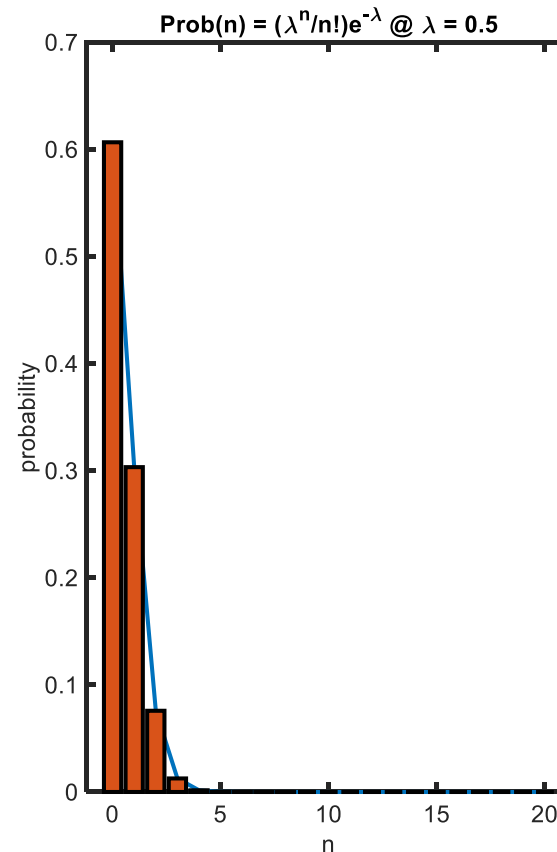
$$w^{(t)}(M, E) = f(M) f_M(E) p,$$
$$p = \xi_i \xi_e \nu_0 e^{-q},$$
$$q = \nu_0 (\xi_i + \xi_e - \xi_i \xi_e)$$



2. Expectation in coincidence analysis

- In the exp, the data (ionization event) may fluctuate following Poisson distribution:

Shot #	e-	H+
1	1	1
2	0	0
3	1	1
4	1	1
5	0	0
6	3	3
7	1	1
8	0	0
...



2. Expectation in coincidence analysis

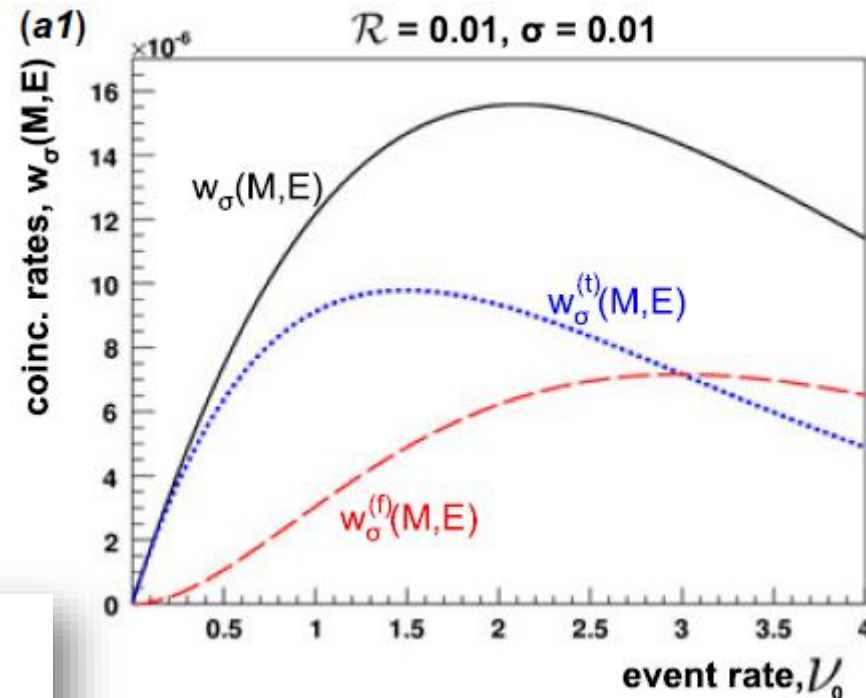
- In reality, there are more fragments (different events) in each shot

Shot #	e-	H+	O+	OH+	H2O+	...
1	3(1)	1	1	0	1	...
2	1(0)	0	0	0	1	...
3	1(1)	1	0	0	0	...
4	7(1)	1	1	0	5	...
5	3(0)	0	0	1	2	...
6	9(3)	3	0	2	4	...
7	7(1)	1	1	2	3	...
8	7(0)	0	0	0	7	...
...

2. Expectation in coincidence analysis

$$w(M, E) = \text{true} + \text{false} = w^{(t)}(M, E) + w^{(f)}(M, E)$$

- All = true + false
- Event rate -> intermediate ν_0



$$w^{(t)}(M, E) = f(M) f_M(E) p,$$

$$p = \xi_i \xi_e \nu_0 e^{-q},$$

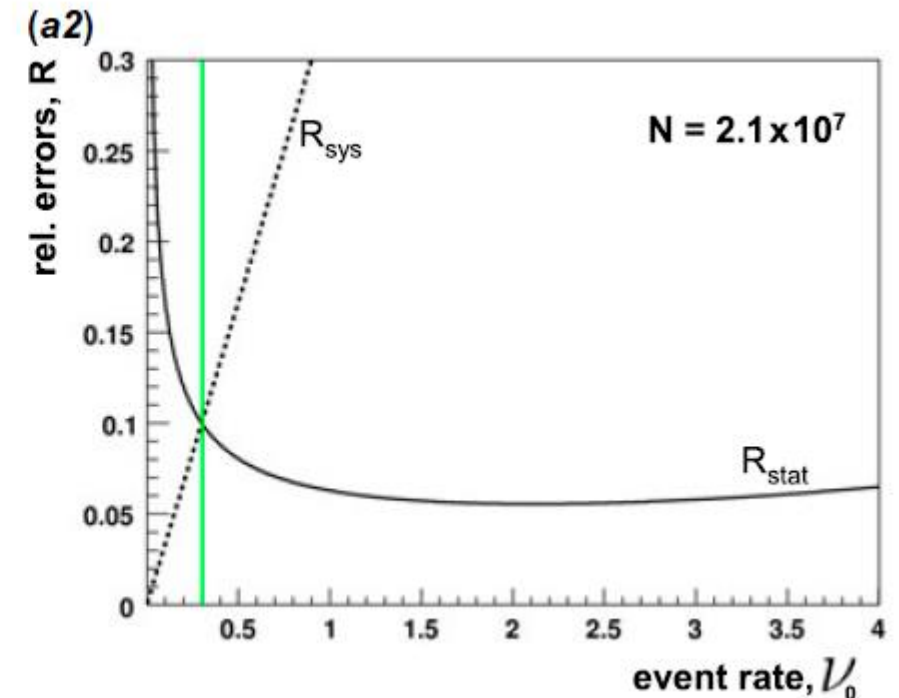
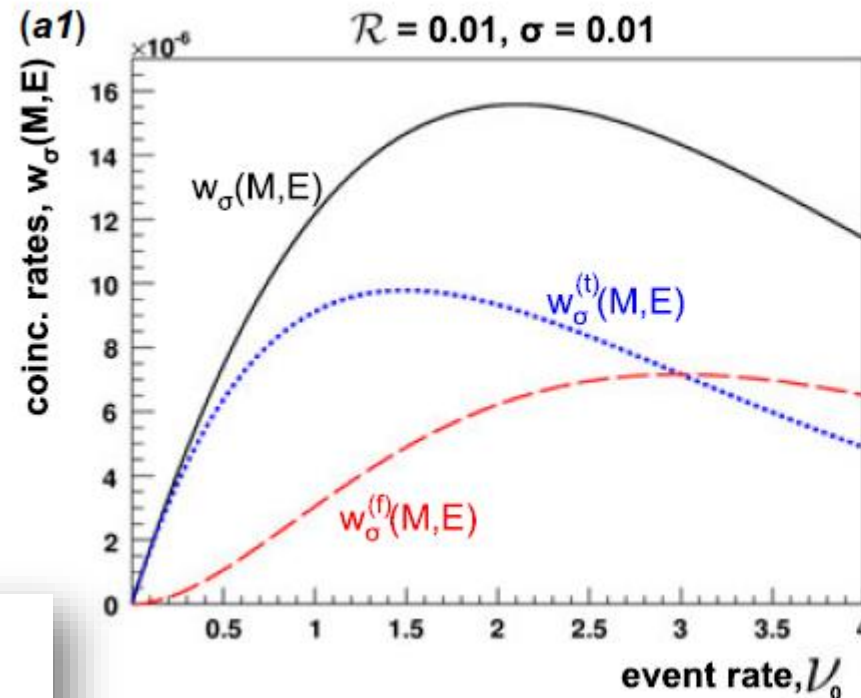
$$q = \nu_0 (\xi_i + \xi_e - \xi_i \xi_e)$$

2. Expectation in coincidence analysis

$$w(M, E) = \text{true} + \text{false} = w^{(t)}(M, E) + w^{(f)}(M, E)$$

$$R_{\text{sys}} = \frac{\text{false}}{\text{all}} = \frac{w^{(f)}(M, E)}{w(M, E)}$$

- All = true + false
- Event rate -> intermediate ν_0
- Sys error -> small ν_0



$$w^{(t)}(M, E) = f(M) f_M(E) p,$$

$$p = \xi_i \xi_e \nu_0 e^{-q},$$

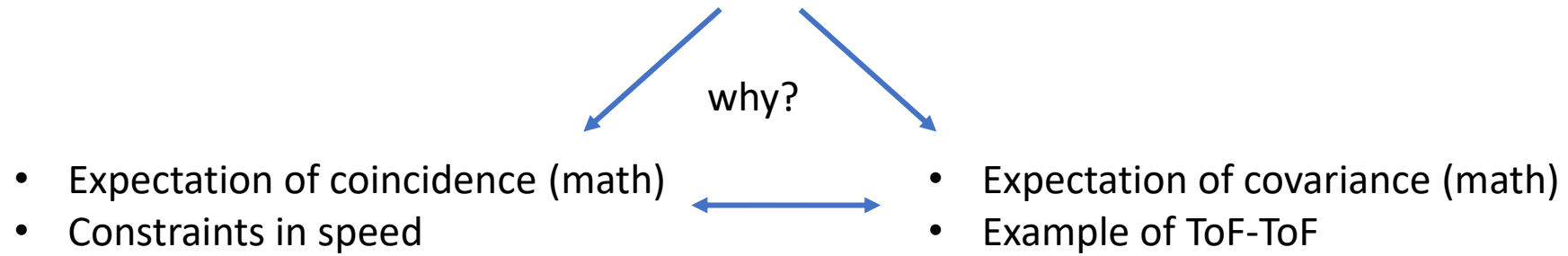
$$q = \nu_0 (\xi_i + \xi_e - \xi_i \xi_e)$$

2. Expectation in coincidence analysis

- Typical acquisition times while **keep low sys error**:
 1. Coincidence for (e, i) takes $\sim 1\text{hr}$
 2. Coincidence for $(2e, 2i)$ takes $\sim 10\text{days}$
- Solutions:
 1. Go for higher repetition rate $\rightarrow 100\text{kHz}$ (currently 1kHz) \rightarrow more shots per time
 2. Go for clever analysis \rightarrow **covariance analysis**

Shot #	e-	H+	O+	OH+	H2O+	...
1	3(1)	1	1	0	1	...
2	1(0)	0	0	0	1	...
3	1(1)	1	0	0	0	...
4	7(1)	1	1	0	5	...
5	3(0)	0	0	1	2	...
6	9(3)	3	0	2	4	...
7	7(1)	1	1	2	3	...
8	7(0)	0	0	0	7	...
...

- Coincidence analysis
- Agreement between coincidence and covariance



3. Carry out covariance – brief math Poisson distribution

- For Poisson distribution, the expectation values are:

- $P(N) = \frac{\lambda^N}{N!} e^{-\lambda}, P(N = 1) = \lambda e^{-\lambda}$

- $\langle N \rangle = \sum N P(N) = \sum N * \frac{\lambda^N}{N!} e^{-\lambda} = \lambda$

- $\langle N^2 \rangle = \sum N^2 P(N) = \sum N^2 * \frac{\lambda^N}{N!} e^{-\lambda} = \lambda^2 + \lambda$

- $Cov(N, N) = Var(N) = \langle N^2 \rangle - \langle N \rangle^2 = \lambda$

$$w^{(t)}(M, E) = f(M) f_M(E) p,$$

$$p = \xi_i \xi_e \nu_0 e^{-q},$$

$$q = \nu_0 (\xi_i + \xi_e - \xi_i \xi_e)$$

vs

w_{covar}

3. Carry out covariance – brief math

$$\begin{aligned} \text{Cov}(N_M, N_E) &= \langle N_M N_E \rangle - \langle N_M \rangle \langle N_E \rangle \\ &= P_1 \nu_0 + \sigma^2 \nu_0^2 (P_1 + P_2)(P_1 + P_3) \end{aligned}$$

$$P_1 = f(M) f_M(E) \xi_i \xi_e$$

Term proportional to fluctuation.
For our laser it is small (~2%).

$$\begin{aligned} w^{(t)}(M, E) &= f(M) f_M(E) p, \\ p &= \xi_i \xi_e \nu_0 e^{-q}, \\ q &= \nu_0 (\xi_i + \xi_e - \xi_i \xi_e) \end{aligned}$$

VS

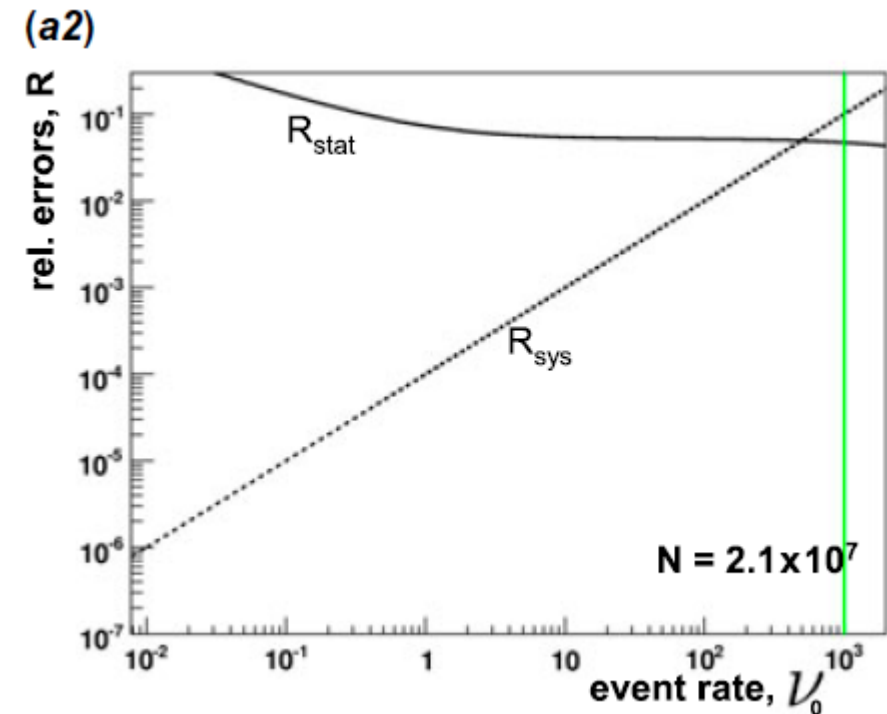
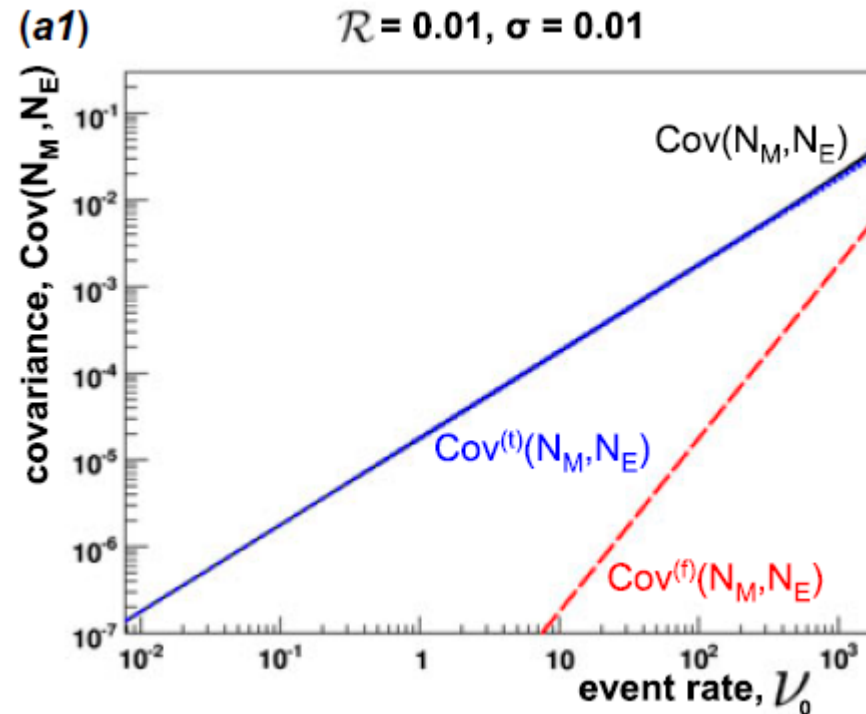
W_{covar}

3. Carry out covariance – simulation

$$w(M, E) = \text{true} + \text{false} = w^{(t)}(M, E) + w^{(f)}(M, E)$$

$$R_{\text{sys}} = \frac{\text{false}}{\text{all}} = \frac{w^{(f)}(M, E)}{w(M, E)}$$

- All = true + false
- Event rate -> the higher ν_0 the better!!
- Sys error -> large ν_0 !!



3. Carry out covariance – e.g. ToFToF

$$\text{Cov}(N^a, N^b) \stackrel{\text{def}}{=} \langle N^{t_1} N^{t_2} \rangle - \langle N^{t_1} \rangle \langle N^{t_2} \rangle$$

i denotes shot number

$$= \frac{\sum N_i^{t_1} N_i^{t_2}}{\text{Shots}} - \frac{\sum N_i^{t_1} \sum N_i^{t_2} - \sum N_i^{t_1} N_i^{t_2}}{\text{Shots}(\text{Shots} - 1)}$$

$$= \frac{\sum N_i^{t_1} N_i^{t_2}}{\text{Shots} - 1} - \frac{\sum N_i^{t_1} \sum N_i^{t_2}}{\text{Shots}(\text{Shots} - 1)}$$

$$\approx \frac{\sum N_i^{t_1} N_i^{t_2}}{\text{Shots}} - \frac{\sum N_i^{t_1} \sum N_i^{t_2}}{\text{Shots}^2}$$

Goal is to compute these two terms summed over *i*

3. Carry out covariance – e.g. ToFToF

Language of computing the fluctuation

$$= \langle (N^{t_1} - \langle N^{t_1} \rangle) (N^{t_2} - \langle N^{t_2} \rangle) \rangle$$

$$\text{Cov}(N^a, N^b) = \langle N^{t_1} N^{t_2} \rangle - \langle N^{t_1} \rangle \langle N^{t_2} \rangle$$

$$= \frac{\sum N_i^{t_1} N_i^{t_2}}{\text{Shots}} - \frac{\sum N_i^{t_1} \sum N_i^{t_2} - \sum N_i^{t_1} N_i^{t_2}}{\text{Shots}(\text{Shots} - 1)}$$

$$= \frac{\sum N_i^{t_1} N_i^{t_2}}{\text{Shots} - 1} - \frac{\sum N_i^{t_1} \sum N_i^{t_2}}{\text{Shots}(\text{Shots} - 1)}$$

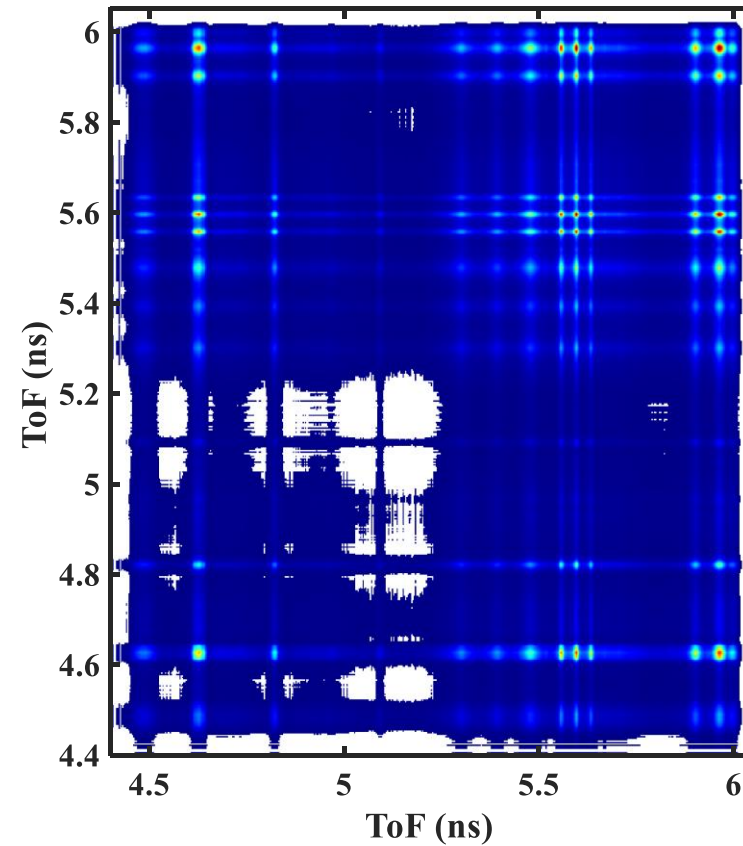
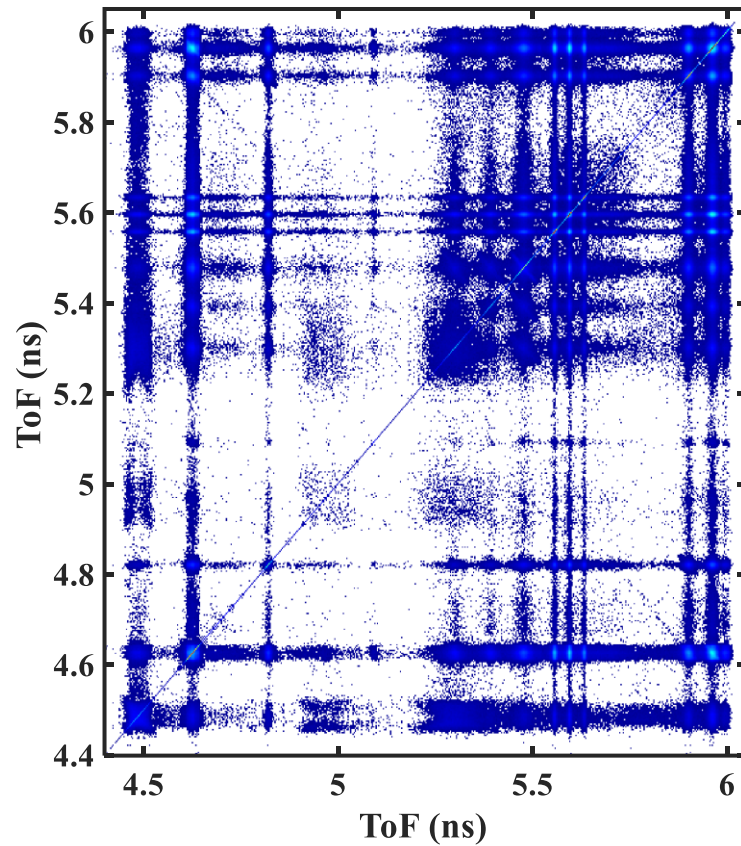
$$\approx \frac{\sum N_i^{t_1} N_i^{t_2}}{\text{Shots}} - \frac{\sum N_i^{t_1} \sum N_i^{t_2}}{\text{Shots}^2}$$

3. Carry out covariance – e.g. ToFToF

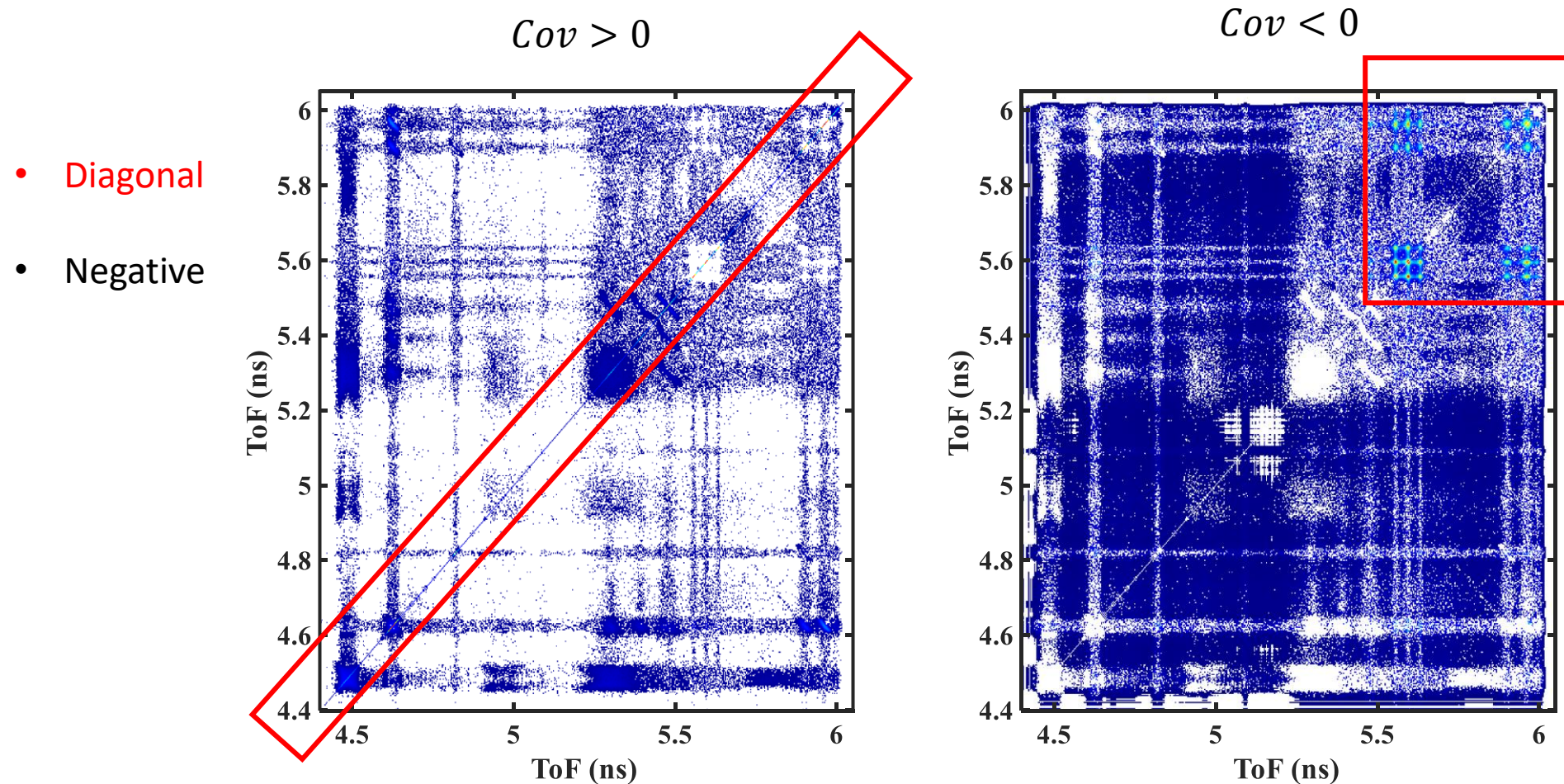
$$S_{12} = \frac{\sum N_i^{t_1} N_i^{t_2}}{\text{Shots}}$$

–

$$S_1 S_2 = \frac{\sum N_i^{t_1} \sum N_i^{t_2}}{\text{Shots}^2}$$



3. Carry out covariance – e.g. ToFToF



3. Carry out covariance – diagonal: $t_1 = t_2 = t$

$$\begin{aligned}
 \text{Cov}(N^{(a)}, N^{(a)}) &= \langle N^{t_1} N^{t_2} \rangle - \langle N^{t_1} \rangle \langle N^{t_2} \rangle \\
 &= \frac{\Sigma(N_i^t)^2}{\text{Shots}} - \frac{\Sigma N_i^t \Sigma N_i^t - \Sigma(N_i^t)^2}{\text{Shots}(\text{Shots} - 1)} \\
 &= \frac{\Sigma(N_i^t)^2}{\text{Shots} - 1} - \frac{\Sigma N_i^t \Sigma N_i^t}{\text{Shots}(\text{Shots} - 1)}
 \end{aligned}$$

Also counting identical pairs

An event rate is λ , produces k fragments that has identical detection efficiency η :

$$\begin{aligned}
 \langle N \rangle &= \lambda k \eta \\
 \text{Cov}(N, N) &= \lambda(2C_k^2 \eta^2 + k\eta)
 \end{aligned}$$

Contribution of recounting

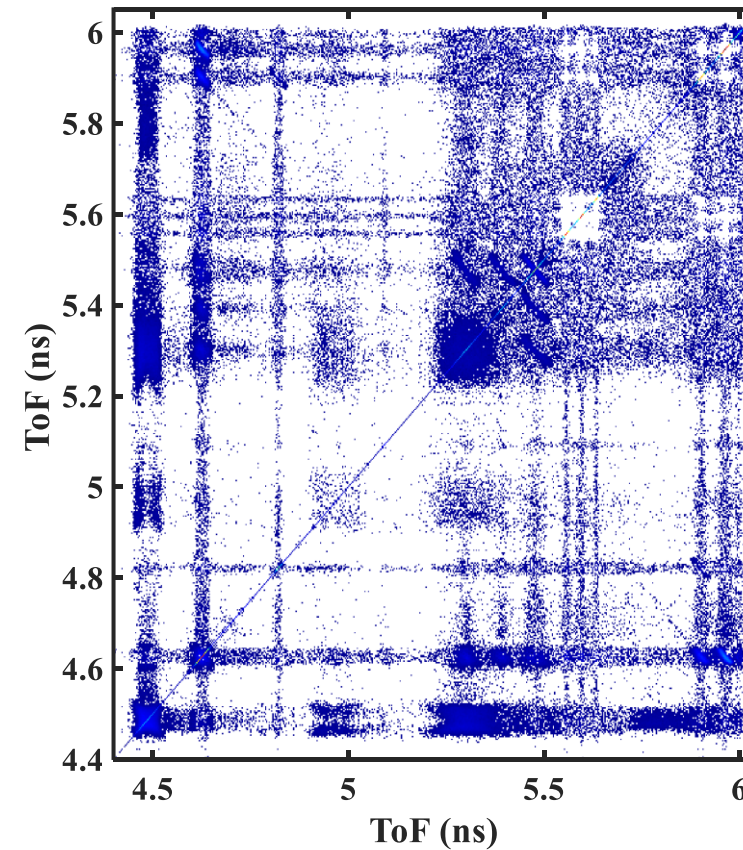
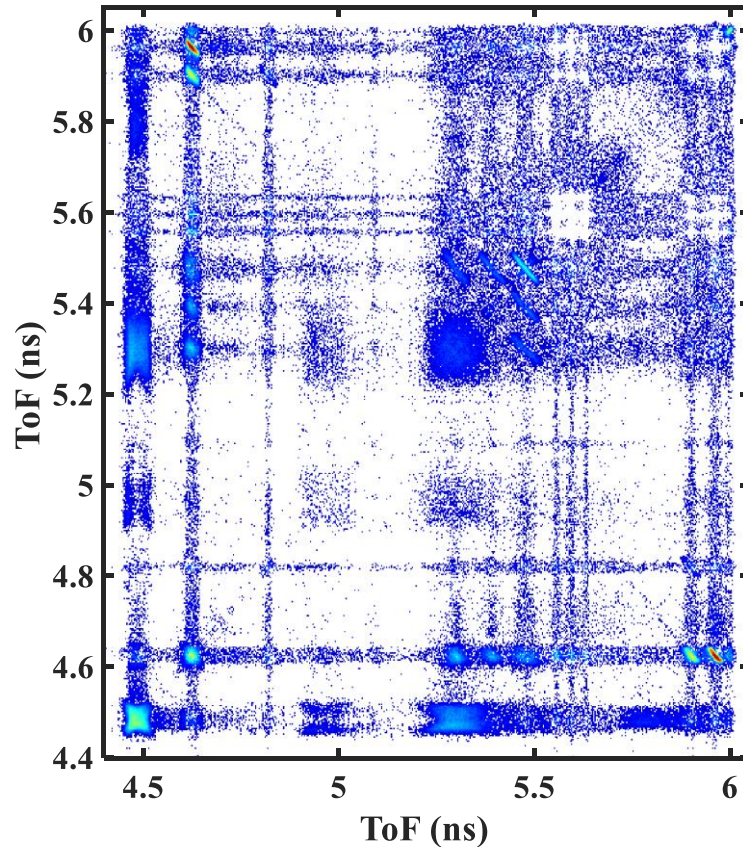
$$\begin{array}{ccc}
 \text{Cov}(N^{(a)}, N^{(a)}) & \longrightarrow & \text{Cov}(N^{(a)}, N^{(a)}) - \langle N^{(a)} \rangle \\
 \text{Cov}(N^{(a)}, N^{(b)}) & & \text{Cov}(N^{(a)}, N^{(b)}) - \langle N^{(a)} \cap N^{(b)} \rangle
 \end{array}$$

3. Carry out covariance – e.g. ToFToF

$$\text{Cov}(N^a, N^b) - \langle N^a \cap N^b \rangle > 0$$

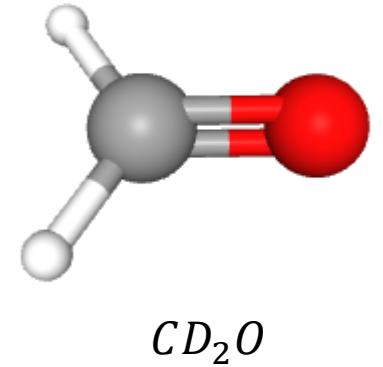
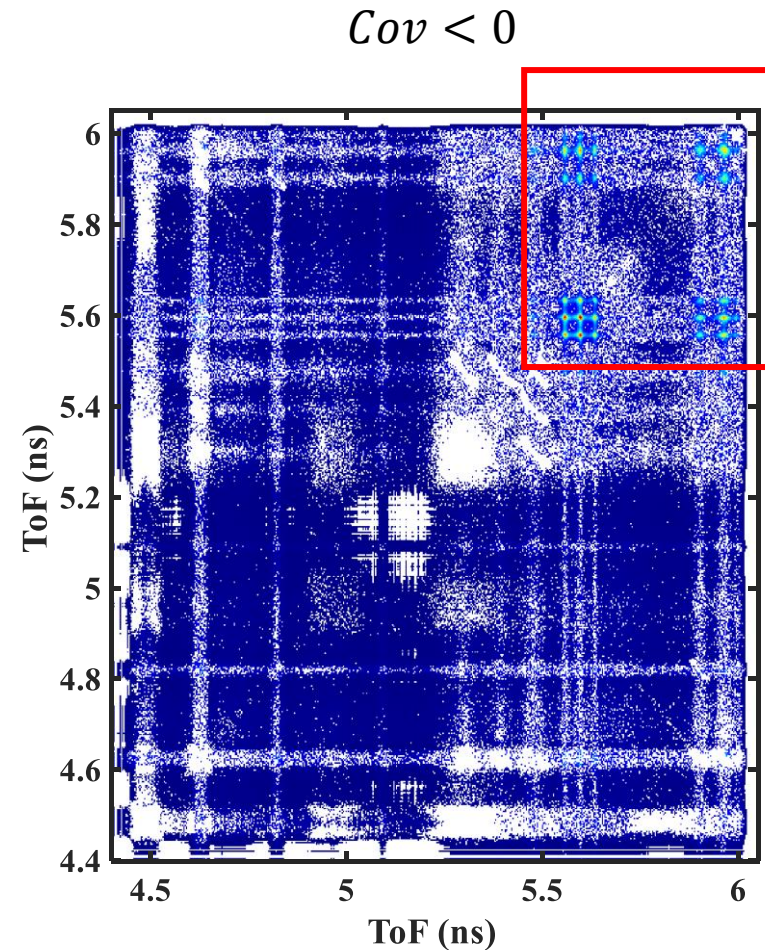
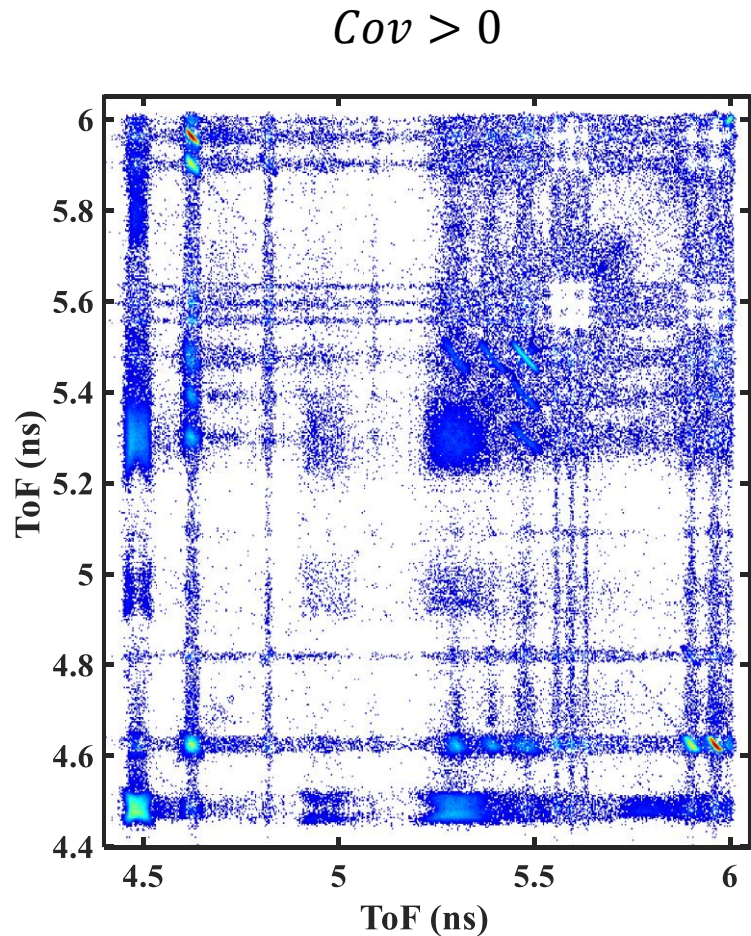
$$\text{Cov}(N^a, N^b) > 0$$

- Diagonal
- Negative



3. Carry out covariance – e.g. ToFToF

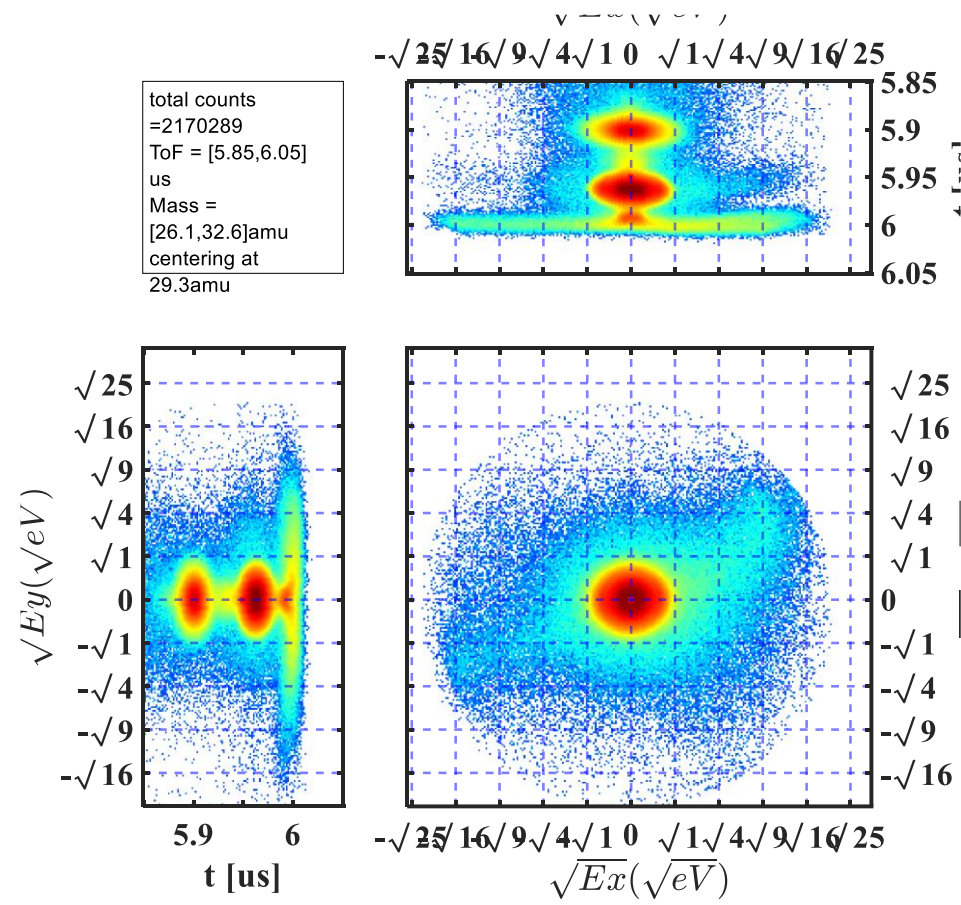
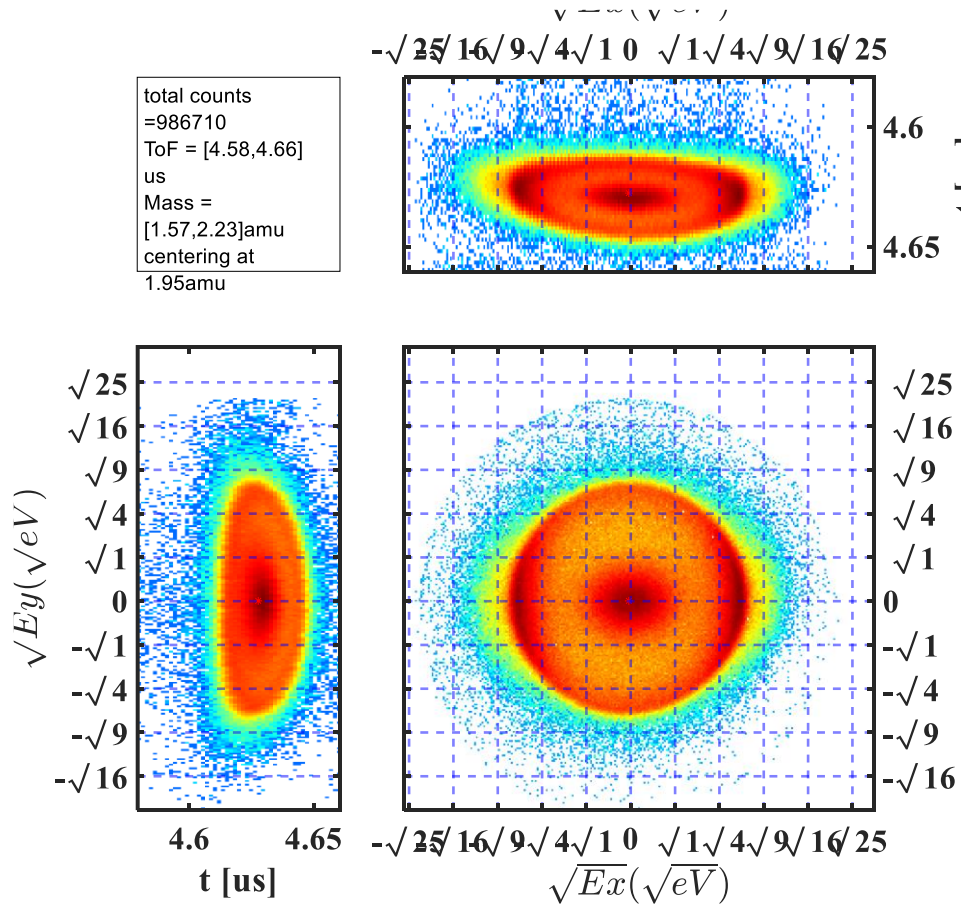
- Diagonal
- Negative



3. Carry out covariance – negative parts of covariance

D^+ : x-y, x-t, y-t plots

CO^+ , CDO^+ , CD_2O^+ : x-y, x-t, y-t plots



- Single ionization dissociation low KE
- $p_1 + p_2 = 0$
- $KE_1:KE_2 = m_2:m_1$
- Overlapping hits issue

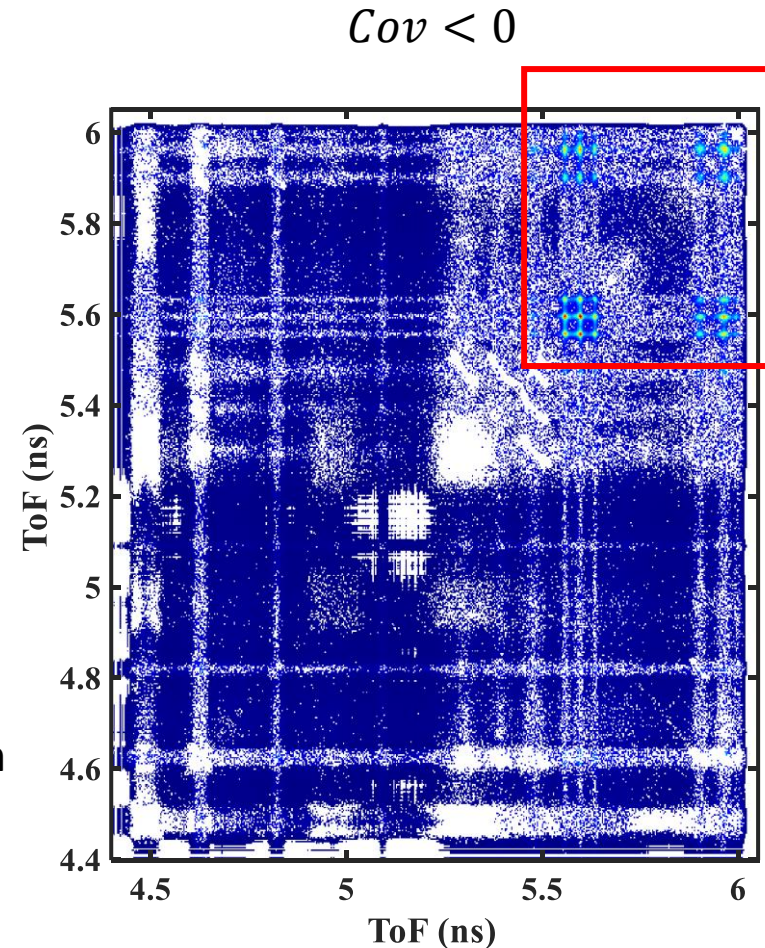
3. Carry out covariance – negative parts of covariance

$$p(N) = \begin{cases} 1 - p = p_1(0) & N = 0 \\ p = \sum_{n \geq 1} p_1(n) & N = 1 \end{cases}$$

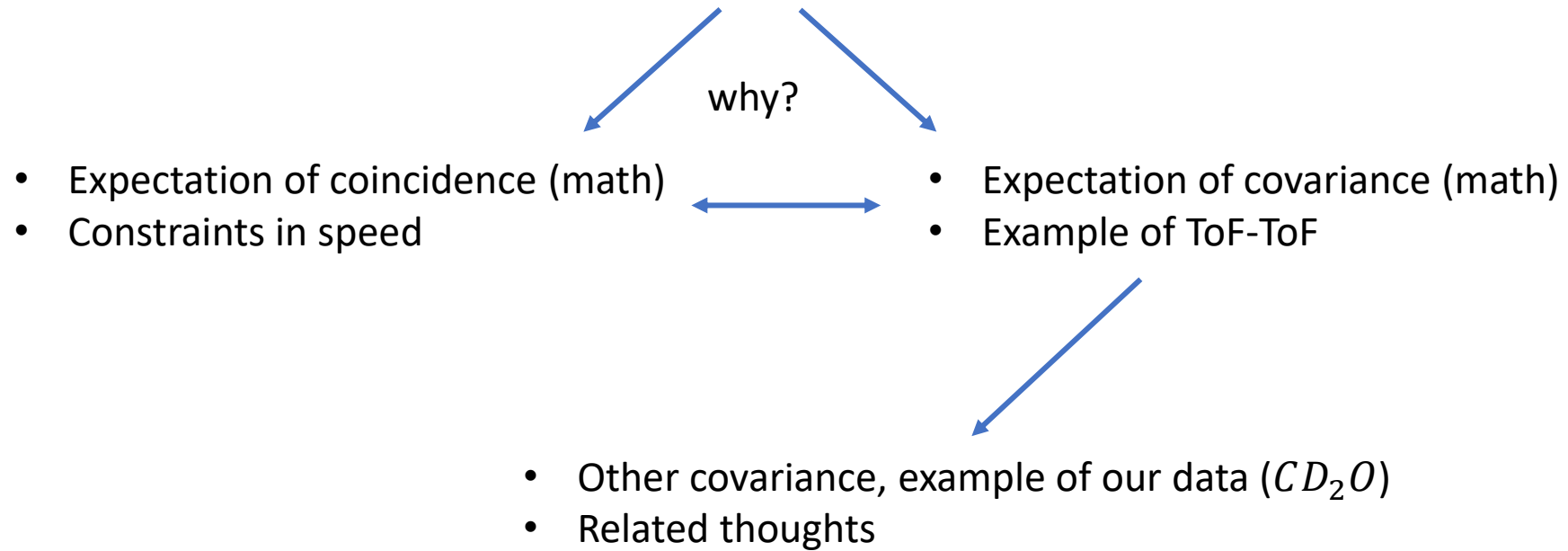
And the related statistics are:

$$\begin{aligned} \langle N \rangle &= p \\ \text{Cov}(N, N) &= p(1 - p) \\ \text{Cov}(N, N) - \langle N \rangle &= -p^2 \end{aligned}$$

- Any imaging-centroiding detector will have this problem



- Coincidence analysis
- Agreement between coincidence and covariance



4. More about covariance – matrix

- $Cov(N^{i_1}, N^{i_2})$

- Matrix of covariance

- $Cov(N^{i_1:t_1}, N^{i_2:t_2})$

- $N^{i_1} = \sum_{t_1} N^{i_1:t_1}$

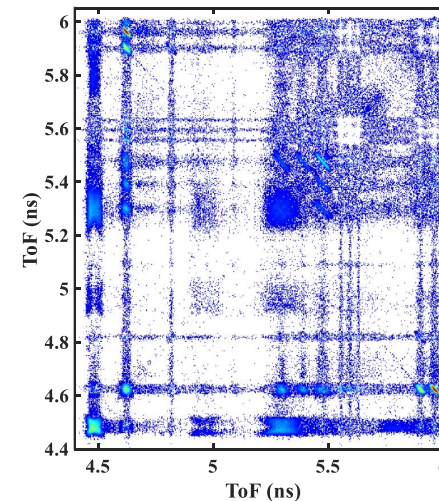
- Other observables like x, y

From Wikipedia:

$$K_{\mathbf{X}\mathbf{X}} = \begin{bmatrix} E[(X_1 - E[X_1])(X_1 - E[X_1])] & E[(X_1 - E[X_1])(X_2 - E[X_2])] & \cdots & E[(X_1 - E[X_1])(X_n - E[X_n])] \\ E[(X_2 - E[X_2])(X_1 - E[X_1])] & E[(X_2 - E[X_2])(X_2 - E[X_2])] & \cdots & E[(X_2 - E[X_2])(X_n - E[X_n])] \\ \vdots & \vdots & \ddots & \vdots \\ E[(X_n - E[X_n])(X_1 - E[X_1])] & E[(X_n - E[X_n])(X_2 - E[X_2])] & \cdots & E[(X_n - E[X_n])(X_n - E[X_n])] \end{bmatrix}$$

The definition above is equivalent to the matrix equality

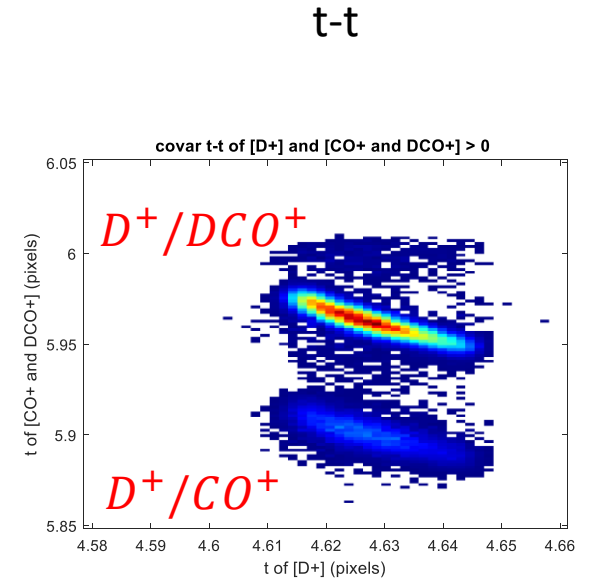
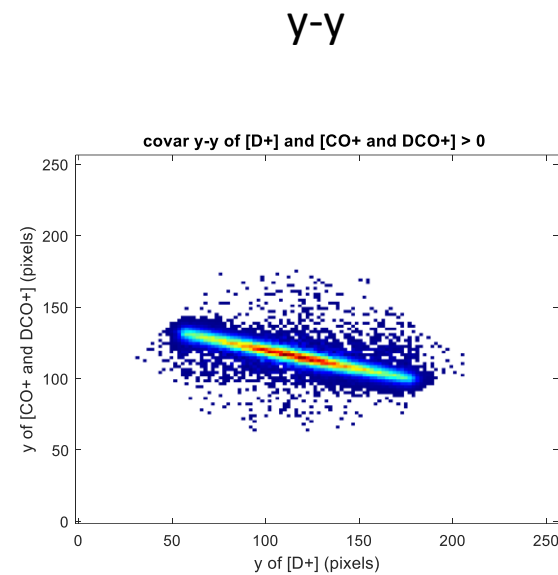
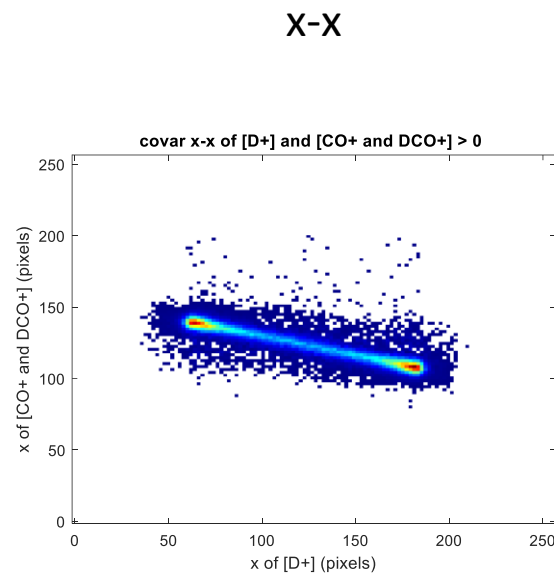
$$K_{\mathbf{X}\mathbf{X}} = \text{cov}[\mathbf{X}, \mathbf{X}] = E[(\mathbf{X} - \mu_{\mathbf{X}})(\mathbf{X} - \mu_{\mathbf{X}})^T] = E[\mathbf{X}\mathbf{X}^T] - \mu_{\mathbf{X}}\mu_{\mathbf{X}}^T \quad (\text{Eq.1})$$



4. More about covariance – momentum correlation

Particle 1 choose D^+ , look at its x direction
Particle 2 choose CO^+/DCO^+ , look at its x direction

- $Cov(N^{D^+:x}, N^{CO^+/DCO^+:x})$
- Momentum conservation
- Coincidence feature
- Angular distribution



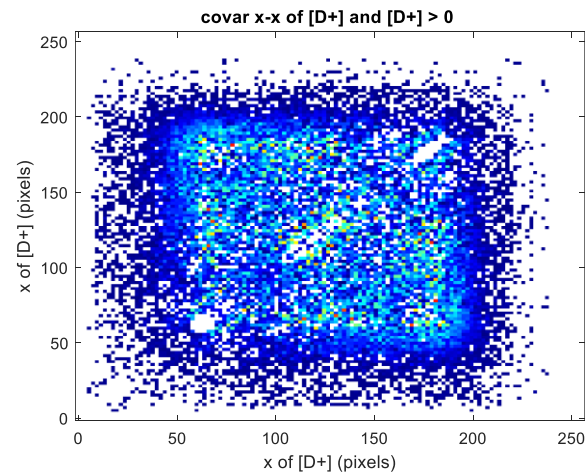
4. More about covariance – momentum correlation

Particle 1 choose D^+ , look at its x direction
Particle 2 choose D^+ , look at its x direction

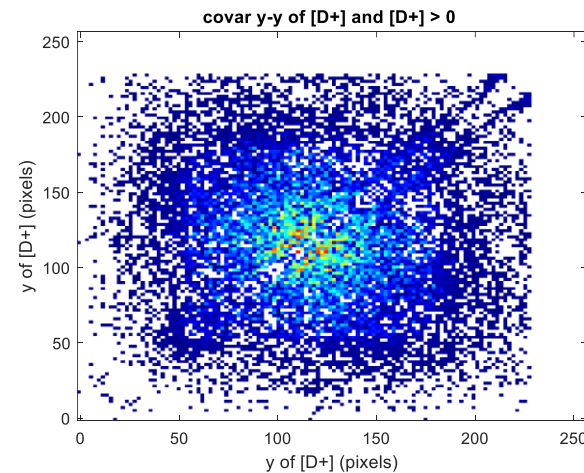


- $Cov(N^{D^+:x}, N^{D^+:x})$
- 3-body or 4-body dissociation
- Diagonal missing

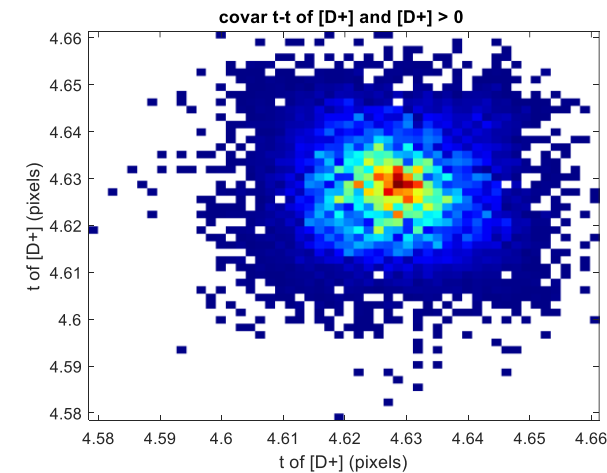
X-X



y-y



t-t



4. More about covariance – $Cov(N_{xy}, N)$

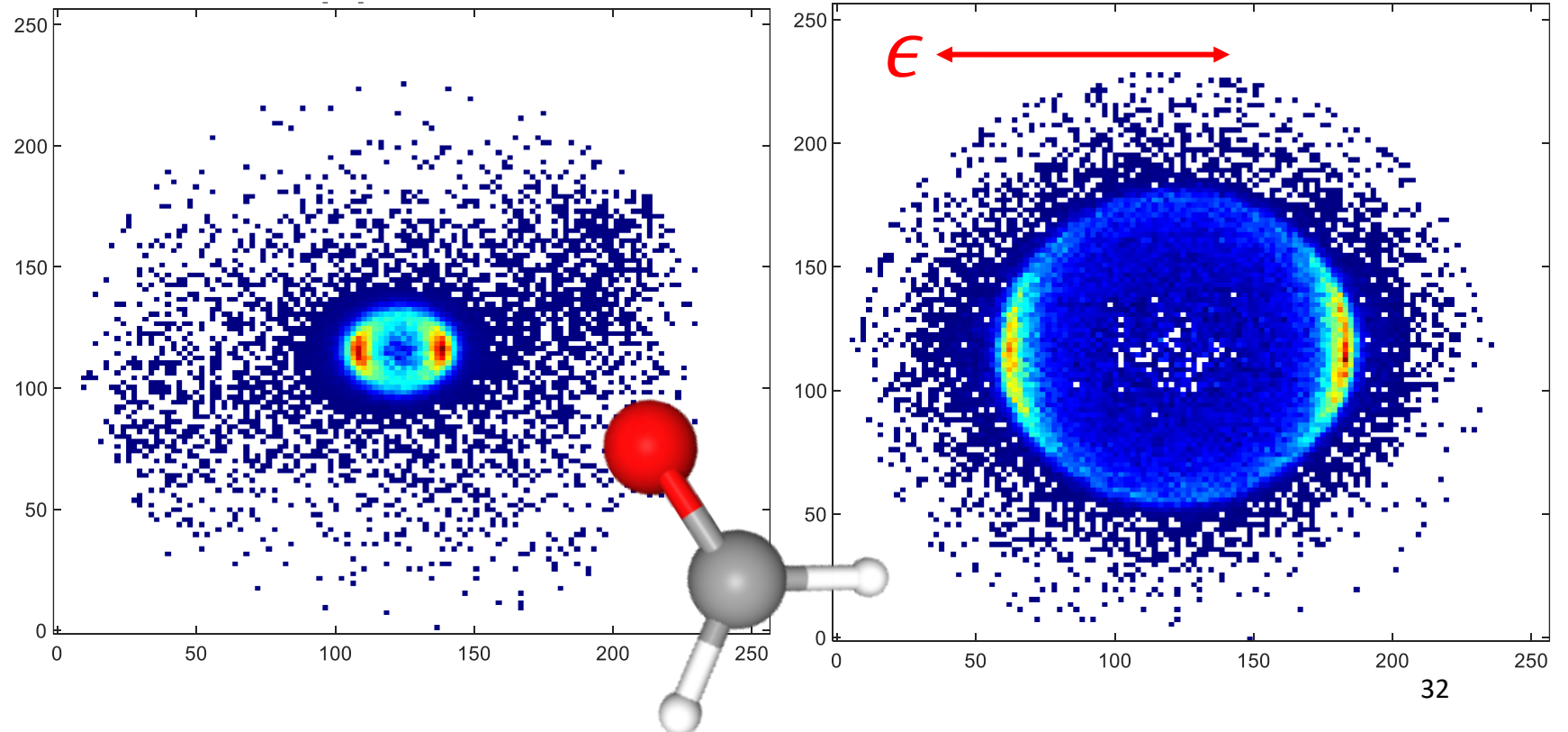
Particle 1 choose CO^+/DCO^+ , look at its xy image
 Particle 2 choose D^+ , look at its $sum\ yield$

Particle 1 choose CO^+/DCO^+ , look at its $sum\ yield$
 Particle 2 choose D^+ , look at its xy image

$$Cov(N^{CO^+/DCO^+:xy}, N^{D^+})$$

$$Cov(N^{CO^+/DCO^+}, N^{D^+:xy})$$

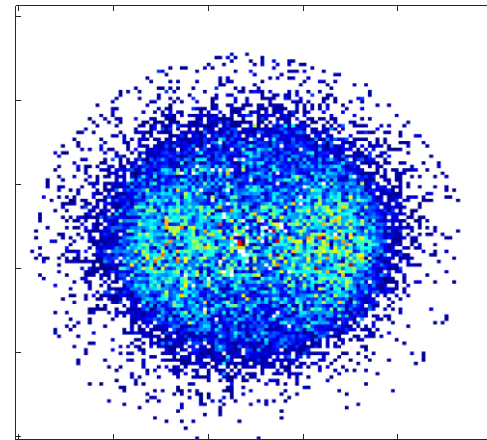
- $p_1 + p_2 = 0$
- $KE_1:KE_2 = m_2:m_1$
- 2-body dissociation
- Angular distribution (alignment)



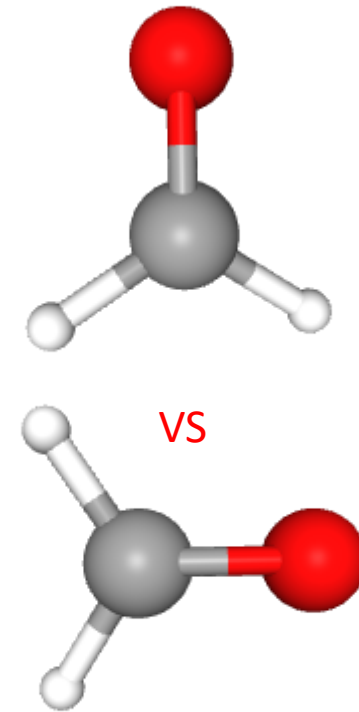
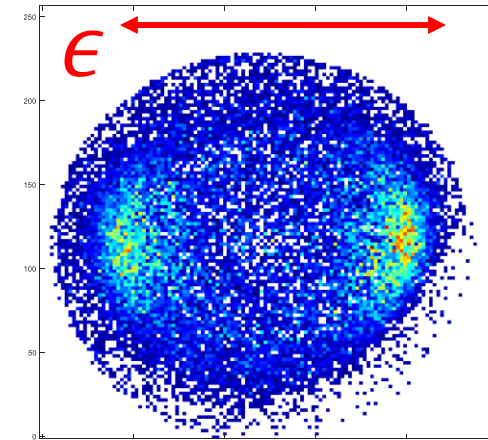
4. More about covariance – 3-body dissociation

- vague angular distribution
- No clear momentum conservation
- Has to do 3-body covariance

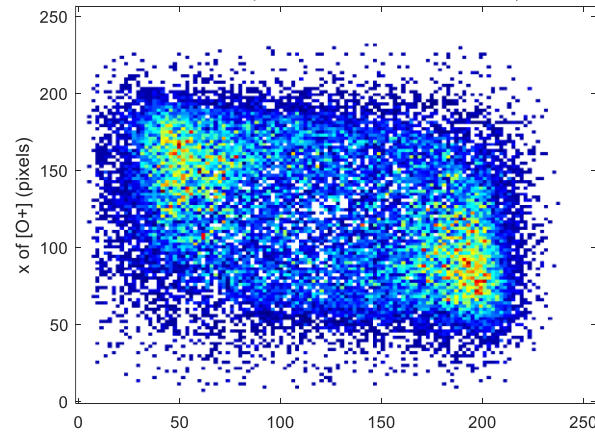
$Cov(N^{O^+:xy}, N^{D^+})$



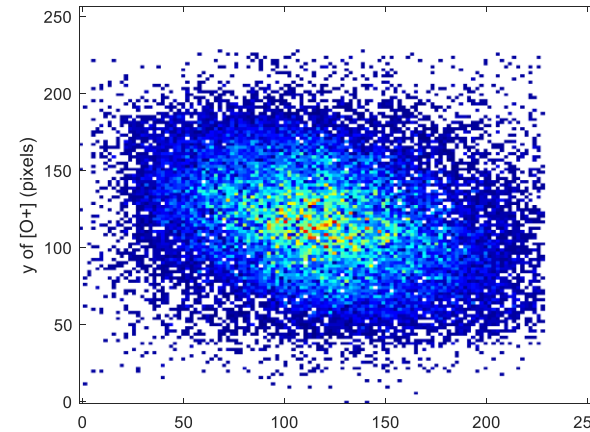
$Cov(N^{O^+}, N^{D^+:xy})$



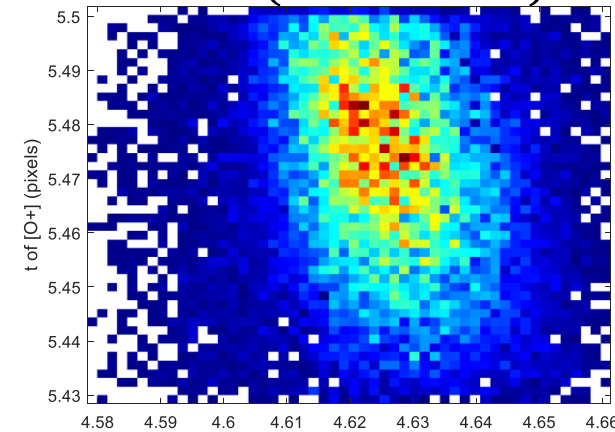
$Cov(N^{O^+:x}, N^{D^+:x})$



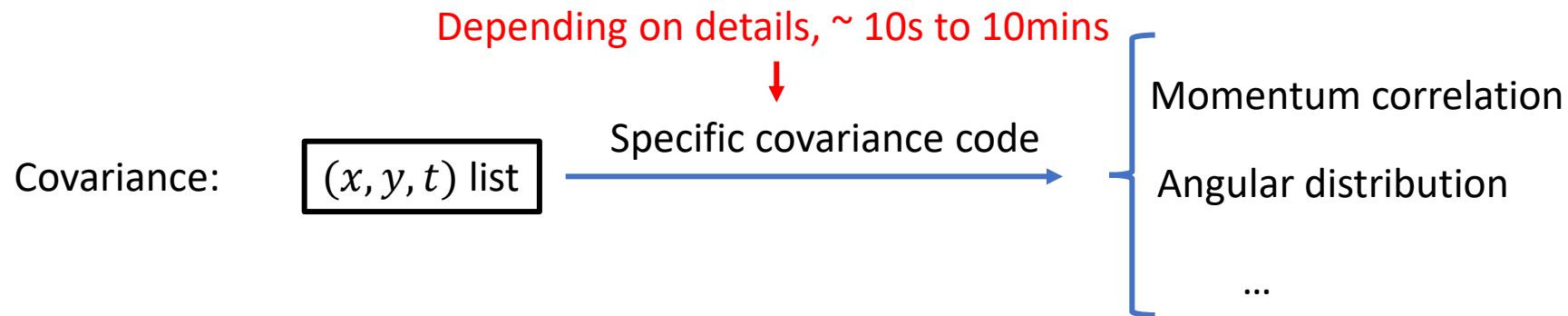
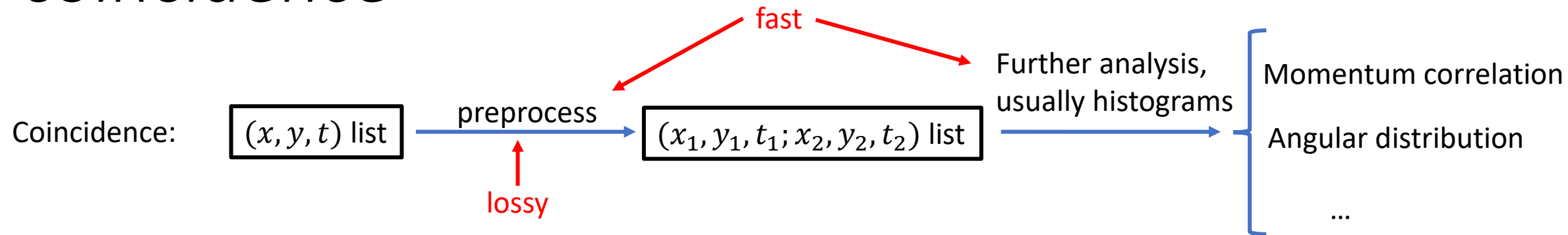
$Cov(N^{O^+:y}, N^{D^+:y})$



$Cov(N^{O^+:t}, N^{D^+:t})$



4. More about covariance – compared to coincidence




We should be doing covariance 10 years ago! $O(n \cdot n)$

4. More about covariance – related ideas

- The pressure should be proportional to the event rate so $Cov(N_e(t), P) \propto \langle N_e \rangle$?
- For a pump probe system like UV-VUV, would it be possible if: $Cov(N_e(t), I_{UV}I_{VUV}) \propto \langle N_e \rangle$ assuming $N_e(t) \propto I_{UV}I_{VUV}$?
- Use ion counts together with photoelectrons
- Partial covariance may help clean some noise as well

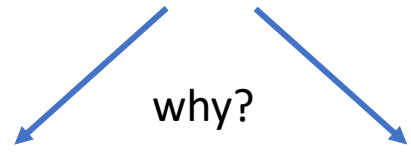
- Thanks for joining. Any thoughts or questions?



Check out our paper:

Allum, Felix, et al. "Multi-Particle Three-Dimensional Covariance Imaging: "Coincidence" Insights into the Many-Body Fragmentation of Strong-Field Ionized D₂O." *The Journal of Physical Chemistry Letters* 12 (2021): 8302-8308.

- Coincidence analysis
- Agreement between coincidence and covariance



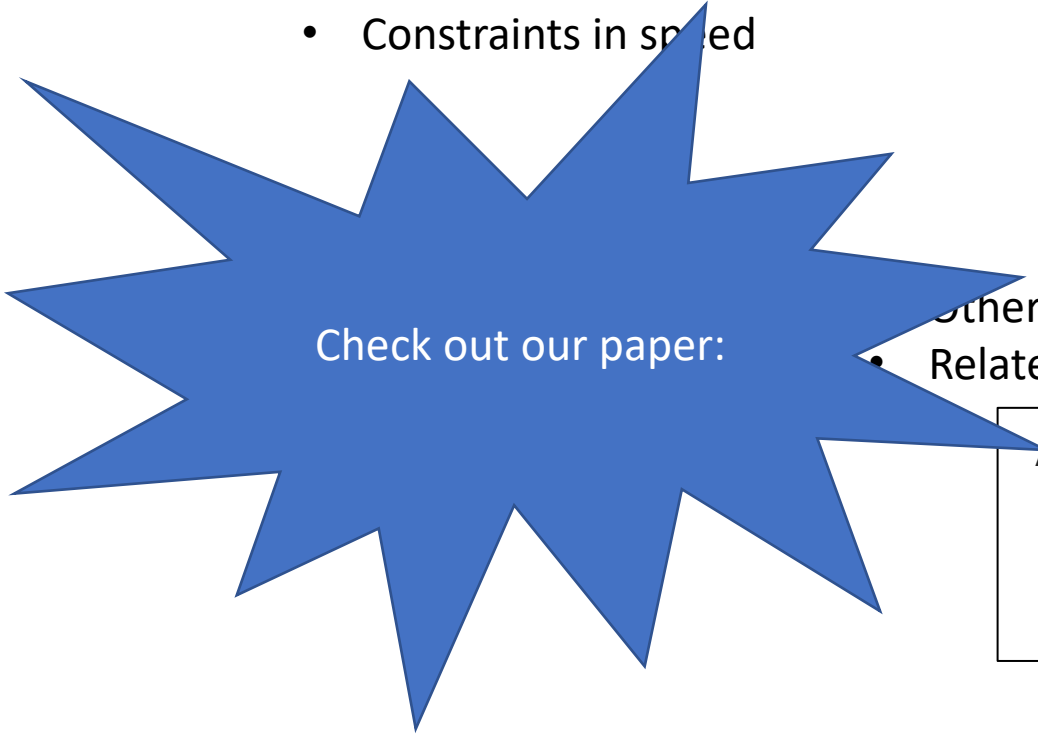
- Expectation of coincidence (math)
- Constraints in speed



- Expectation of covariance (math)
- Example of ToF-ToF



- Other covariance, example of our data (CD_2O)
- Related thoughts



Allum, Felix, et al. "Multi-Particle Three-Dimensional Covariance Imaging: "Coincidence" Insights into the Many-Body Fragmentation of Strong-Field Ionized D2O." *The Journal of Physical Chemistry Letters* 12 (2021): 8302-8308.

Some reading materials

<https://journals.aps.org/pr/abstract/10.1103/PhysRevA.89.011401>

<https://journals.aps.org/pr/abstract/10.1103/PhysRevA.91.053424>

<https://journals.aps.org/prl/abstract/10.1103/PhysRevLett.113.073005>

<https://aip.scitation.org/doi/full/10.1063/1.4947551>

<https://aip.scitation.org/doi/full/10.1063/1.5041381>

<https://pubs.rsc.org/en/content/articlehtml/2020/fd/d0fd00115e>

<https://www.nature.com/articles/s42004-020-0320-3>

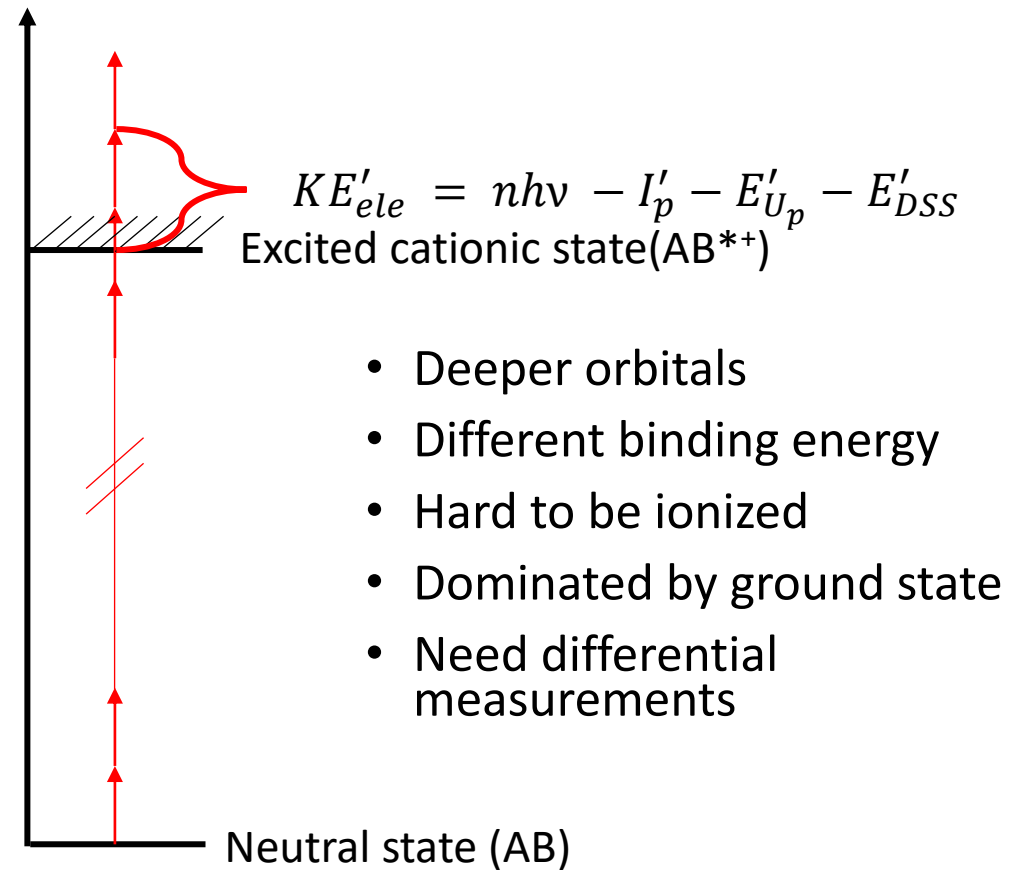
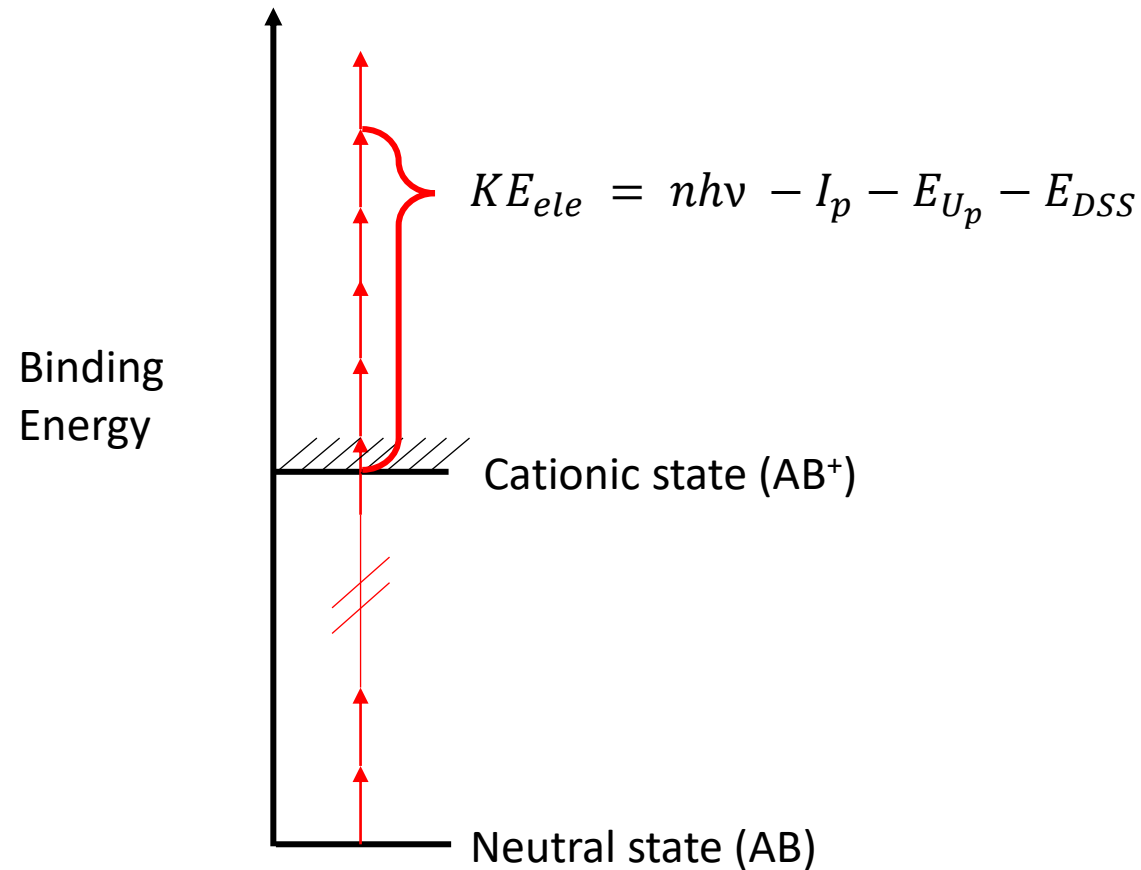
<https://journals.aps.org/pr/abstract/10.1103/PhysRevA.89.053418>

Mainly focus on the following papers:

<https://www.tandfonline.com/doi/full/10.1080/09500340.2013.839837>

<https://www.tandfonline.com/doi/full/10.1080/09500340.2013.839840>

Appendix 1: CRATI



Appendix 3: ToFToF computation

$$\frac{\sum N_i^{t_1} N_i^{t_2}}{Shots}$$

```
for ii = 1:1:length(trigindex) - 1
    for jj1 = trigindex(ii):trigindex(ii + 1) - 1
        for jj2 = trigindex(ii):trigindex(ii + 1) - 1
            ToFToF(ToFIndexes(jj1),ToFIndexes(jj2)) = ToFToF(ToFIndexes(jj1),ToFIndexes(jj2)) + 1;
        end
    end
end
```

Appendix 4: more poisson

- Poisson distribution and multiple events

- Event n follows Poisson distribution:

- $$P(N_n) = \frac{\lambda_n^{N_n}}{N_n!} e^{-\lambda_n}$$

- Simple calculation shows that, summing up all m events to get everything $N = N_1 + N_2 + \dots + N_m$, N follows:

- $$P(N) = \frac{\lambda^N}{N!} e^{-\lambda} \text{ where } \lambda = \lambda_1 + \lambda_2 + \dots + \lambda_m$$

- Which tells us that: everything is Poisson style.

Appendix 4: more poisson

- For Poisson distribution, the expectation value is:
- $\langle N \rangle = \sum N P(N) = \sum N * \frac{\lambda^N}{N!} e^{-\lambda} = \lambda$
- $\langle N^2 \rangle = \sum N^2 P(N) = \sum N^2 * \frac{\lambda^N}{N!} e^{-\lambda} = \lambda^2 + \lambda$
- $\langle N^3 \rangle = \sum N^3 P(N) = \sum N^3 * \frac{\lambda^N}{N!} e^{-\lambda} = \lambda^3 + 3\lambda^2 + \lambda$
- $\sqrt{Var(N)} = \sqrt{\langle (N - \langle N \rangle)^2 \rangle} = \sqrt{\langle N^2 \rangle - \langle N \rangle^2} = \sqrt{\lambda}$
- $\frac{\langle N \rangle}{\sqrt{Var(N)}} = \sqrt{\lambda}$

Appendix 4: more poisson

- **Another view of the covariance: fluctuation between two particles**

- Imagine our detection efficiency is 100% for both ele and ion.

- $Cov(N_e, N_M) = Cov(N_{M1} + N_{M2} + \dots, N_M) = Cov(N_M, N_M) = N_M$

Expand the sources of
electrons: with different ions

Nature of irrelevant events the
covariance calculation. Note 1
ionization is not going to generate
so many different ions!

Nature of Poisson style
distribution

Appendix 4: more poisson

- Another view of the covariance: fluctuation between two particles
- Imagine our detection efficiency is 100% for both ele and ion.

- $Cov(N_e, N_M) = Cov(N_{M1} + N_{M2} + \dots, N_M) = Cov(N_M, N_M) = N_M$

Note that with the help of N_M the N_e has been filtered out to only include events involve N_M !!

- $Cov(\xi_e N_e, \xi_i N_M) = \dots = Cov(\xi_e N_M, \xi_i N_M) = \xi_e \xi_i N_M$

1. Strong of coincidence

- ~~0. the multiple particle nature of the measurement~~
- ~~1. MRATI~~
- ~~2. short VS long~~
- ~~3. example of covariance to fit coincidence~~

2. Coincidence expectation

- ~~1. from intuition~~
- ~~2. from real math~~
- ~~3. the boundary → acquisition time (stat table) → MRI and covariance~~

3. Carry out covariance

- ~~1. covar math brief~~
- ~~2. covar expectation plot~~
- ~~3. example: ToFToF~~

4. More about covariance

- ~~1. different covar (ToFToF, Nnxy, NxNy, betaKER) speed and back to coincidence~~
- ~~2. possible covar params/apps – UV with ions or laser intensity~~
- 3. covar constraints** -- comparison and speed boost math