Coulomb Explosion Imaging of Molecular Fragmentation in Femtosecond Pump-Probe Experiment

LOGAN RINEY

MENTORS: DR. DANIEL ROLLES,

DR. ARTEM RUDENKO, AND FARZANEH ZIAEE

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Brief Overview

- Motivation
- Experimental Setup
- Coulomb Explosion Simulation
- Results
- Conclusion

Motivation

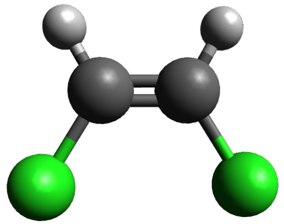
- Goal: Use Coulomb Explosion Simulation to model the repulsion of ionized fragments of a molecule.

- Would like to see if Coulomb Explosion Imaging can distinguish molecular isomers.

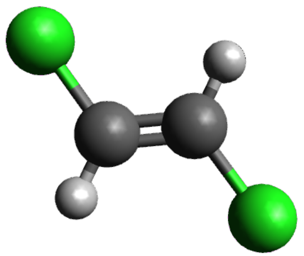
- The molecule of interest is cis-,trans-dichloroethene

- This model is used to find the kinetic energies of the ionic fragments for a given channel

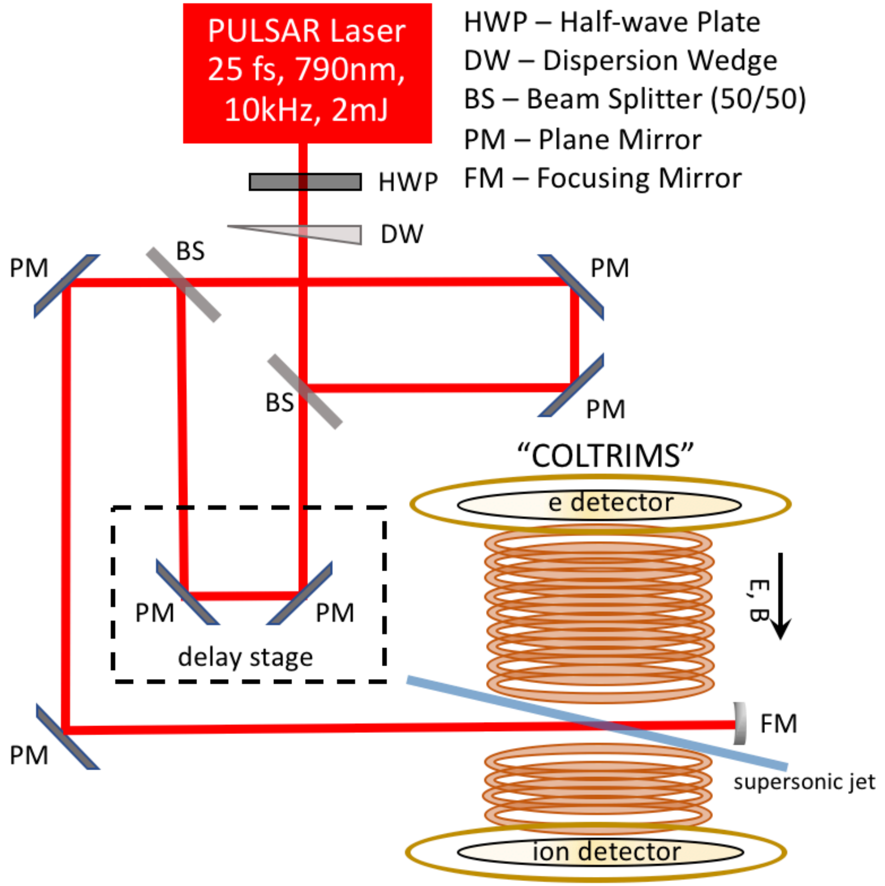
cis-$C_2H_2Cl_2$



trans-$C_2H_2Cl_2$



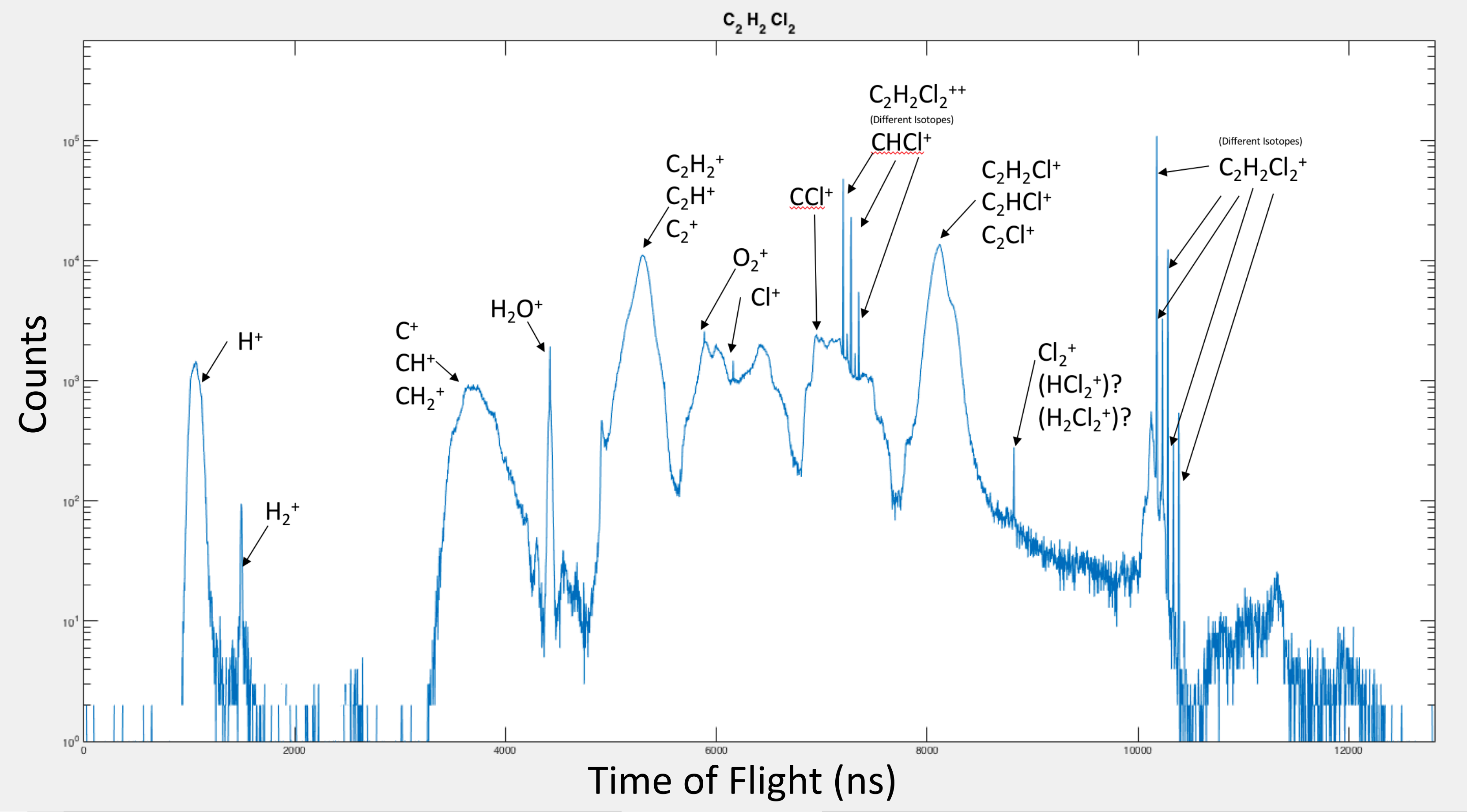
Experiment and Setup



Intensity: 4.25x10^{14} W/cm^2

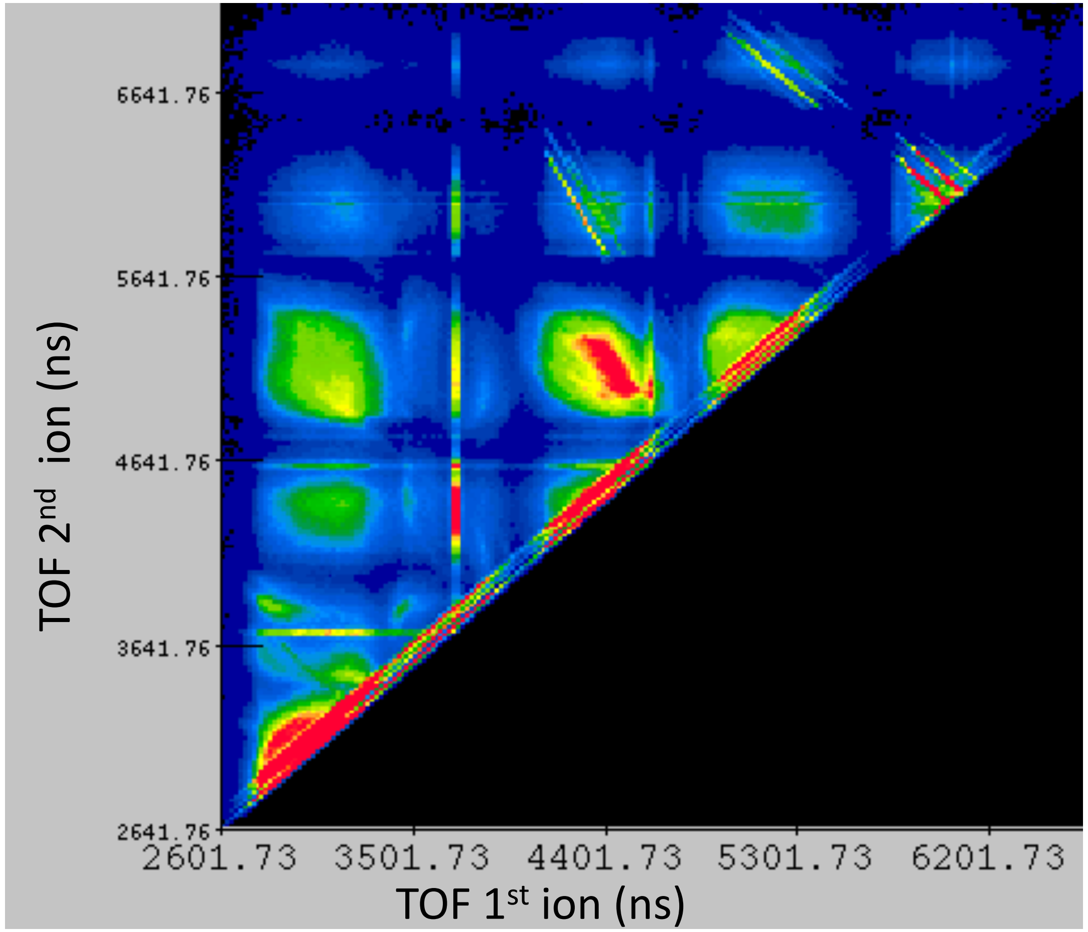
- Supersonic Jet of $C_2H_2Cl_2$ molecules is directed into the beam path to be photo-ionized.
- The molecules are photo-ionized by 790nm, 25 fs near-infrared laser pulses
- The resulting fragments are then directed by the E-field to the ion detector to measured by coincident ion momentum imaging.

Experiment and Setup



- The detector can measure the Time of Flight (TOF) of each ion.
- $TOF \propto \sqrt{\frac{Mass}{Charge}}$

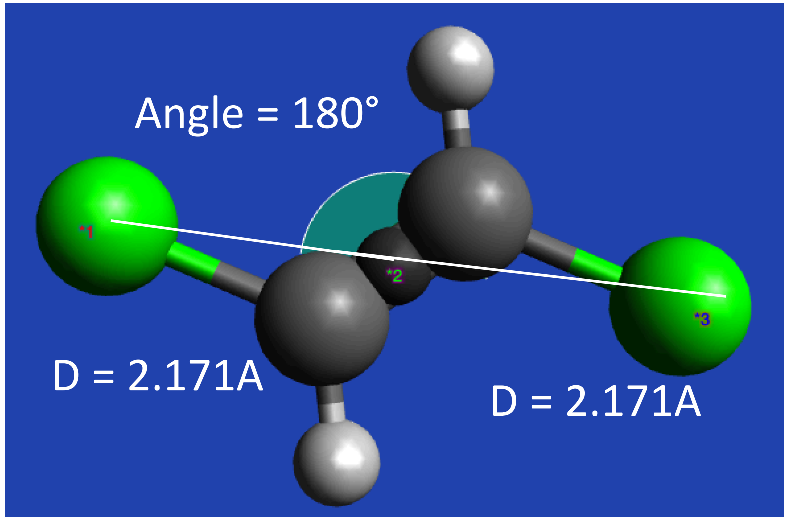
Experiment and Setup

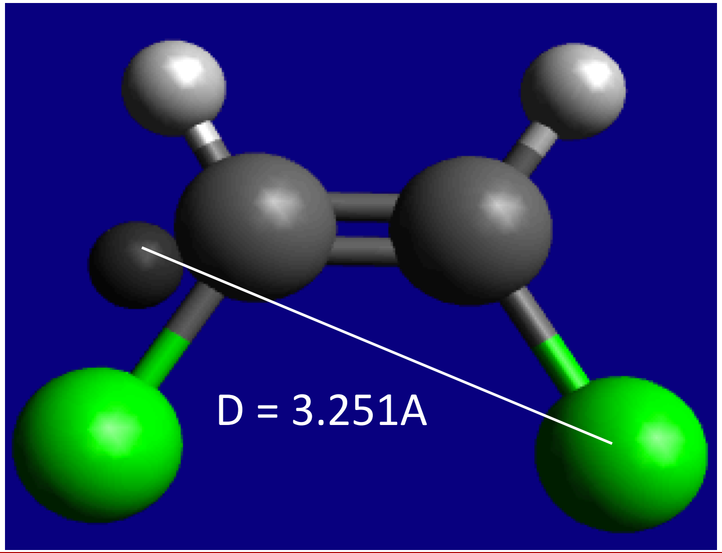


- The detector also measures photo-ions that are in coincidence (PIPICO).
- The diagonal stripes in PIPICO spectrum is due to conversation of p_z momenta of corresponding coincident ions.

Coulomb Explosion Simulation

- The fragment paths are calculated using the 8th order Runge-Kutta numerical method.
- The initial conditions for the molecule is calculated by using Avogadro to find the equilibrium geometry for the different breakup channels.





Coulomb Explosion Simulation

- For the $C_2H_2^+ + Cl^+ + Cl^+$ channel the following system was solved:

$$F_{C_2H_2^+}(r_1) \propto \frac{1}{R_{12}^2} + \frac{1}{R_{13}^2}$$

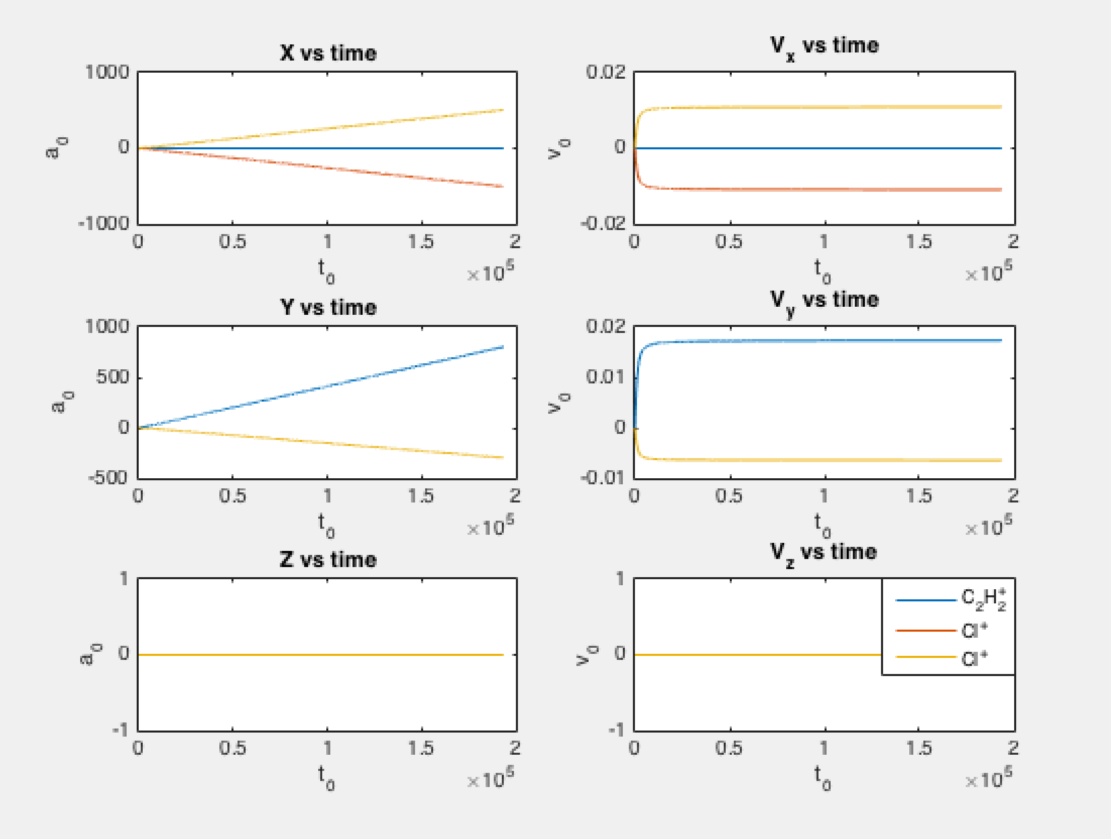
$$F_{Cl^+}(r_2) \propto \frac{1}{R_{21}^2} + \frac{1}{R_{32}^2}$$

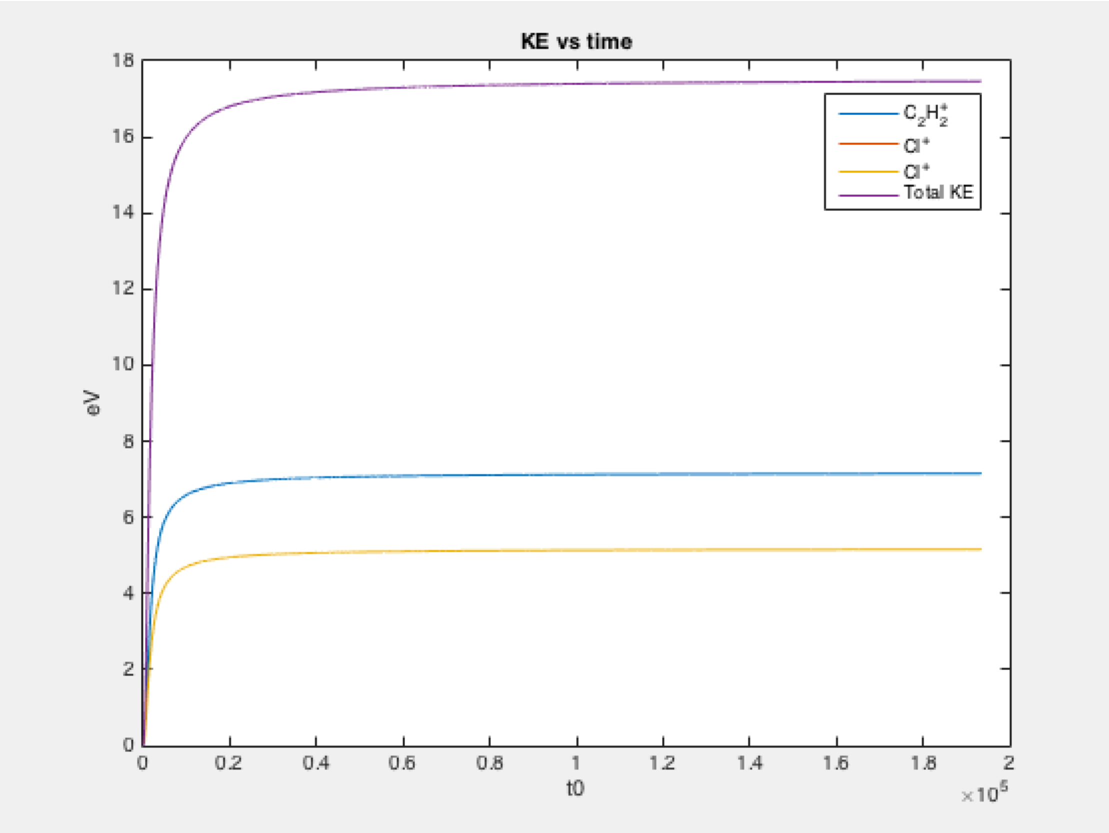
$$F_{Cl^+}(r_3) \propto \frac{1}{R_{31}^2} + \frac{1}{R_{32}^2}$$

- 8th Order Runge-Kutta gives higher precision than other commonly used methods like the 4th Order Runge-Kutta method.
- A similar, more simple set of equations is used to calculate 2 Body Breakups.

Coulomb Explosion Simulation

- After solving a similar system





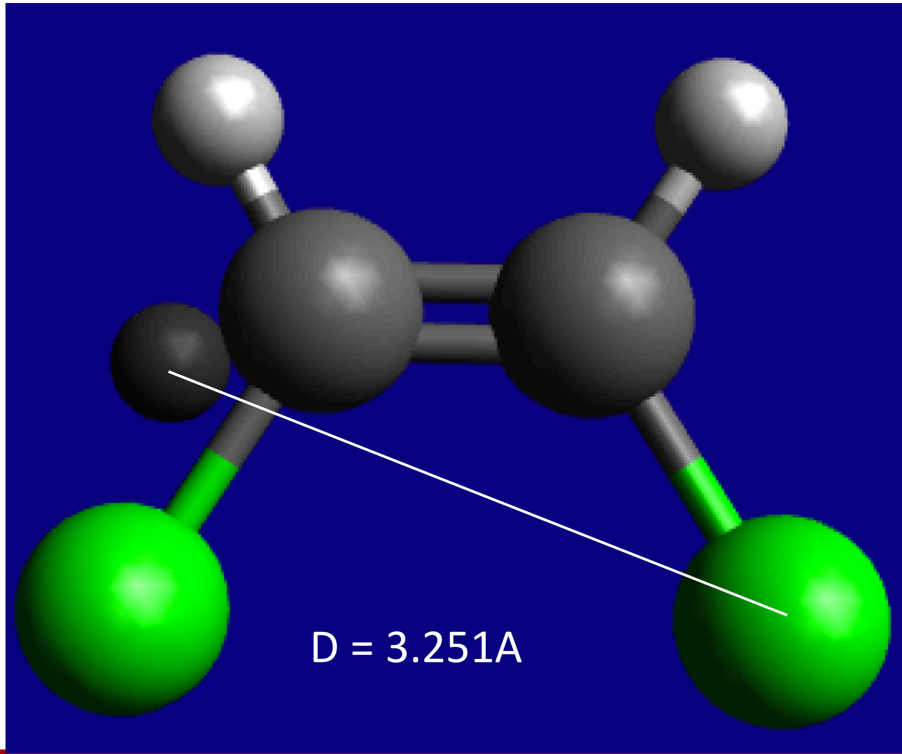
Results

- The Coulomb Explosion simulation works well for both the cis- and trans- parent molecule.

Results: 2 Body - $C_2H_2Cl^+ + Cl^+$

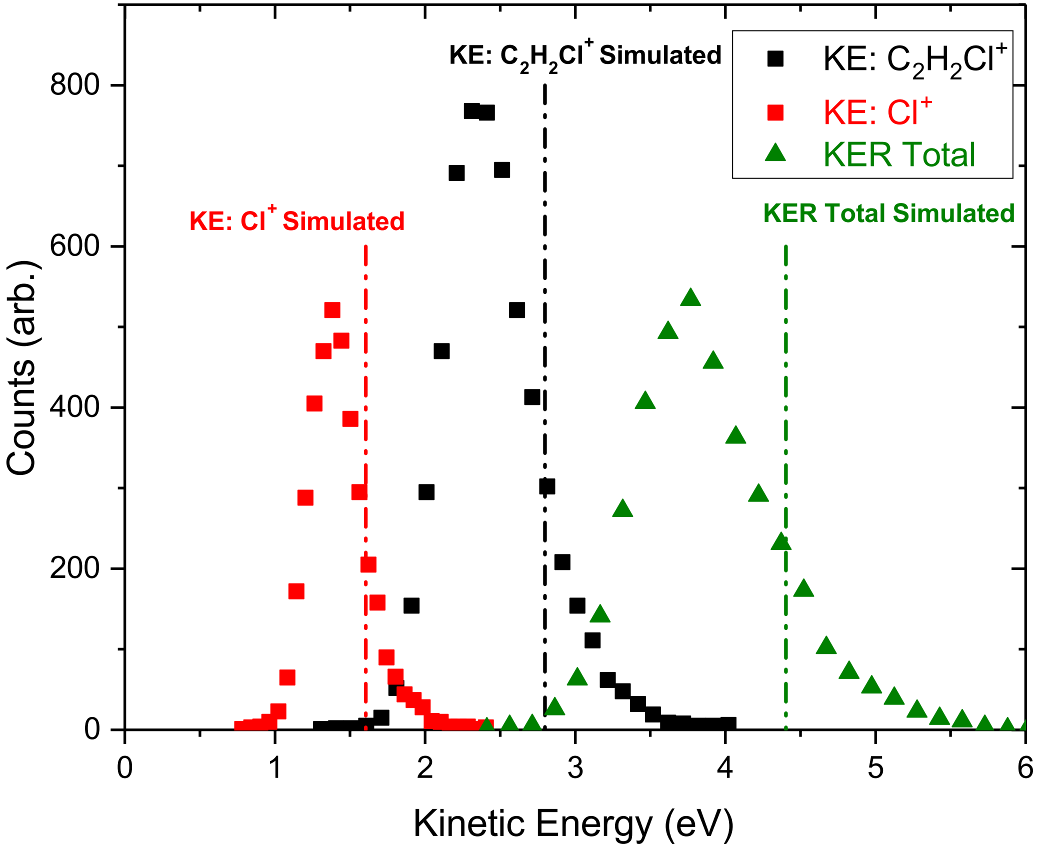
- For cis-$C_2H_2Cl_2$

- The initial geometry calculated by Avogadro



Results: 2 Body - $C_2H_2Cl^+ + Cl^+$

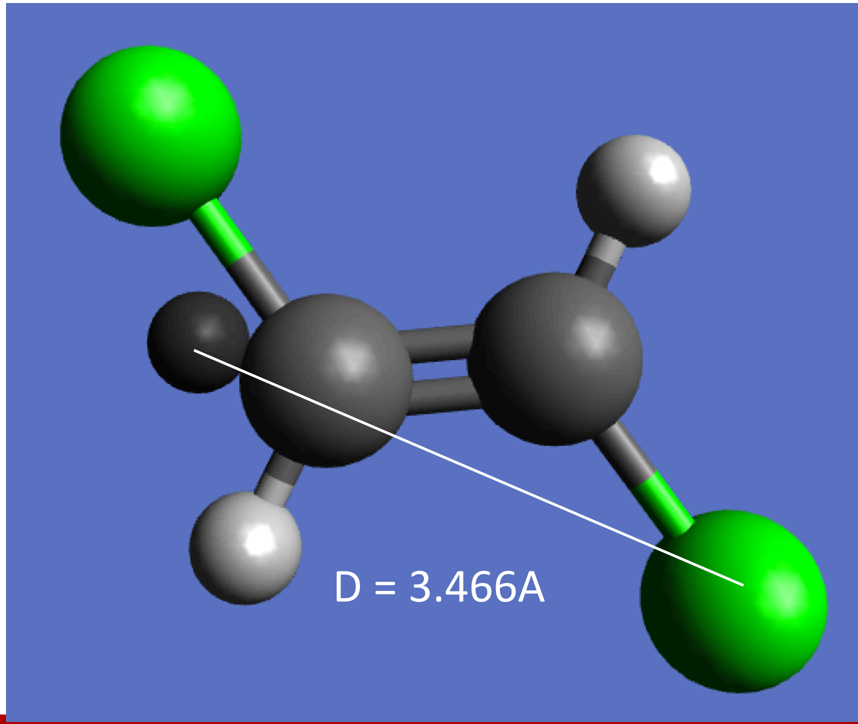
- For cis-$C_2H_2Cl_2$



	$C_2H_2Cl^+$	Cl^+	Total KER
Experimental Energy	2.3618eV	1.4028eV	3.7185eV
Simulated Energy	2.7979eV	1.6053eV	4.4032eV

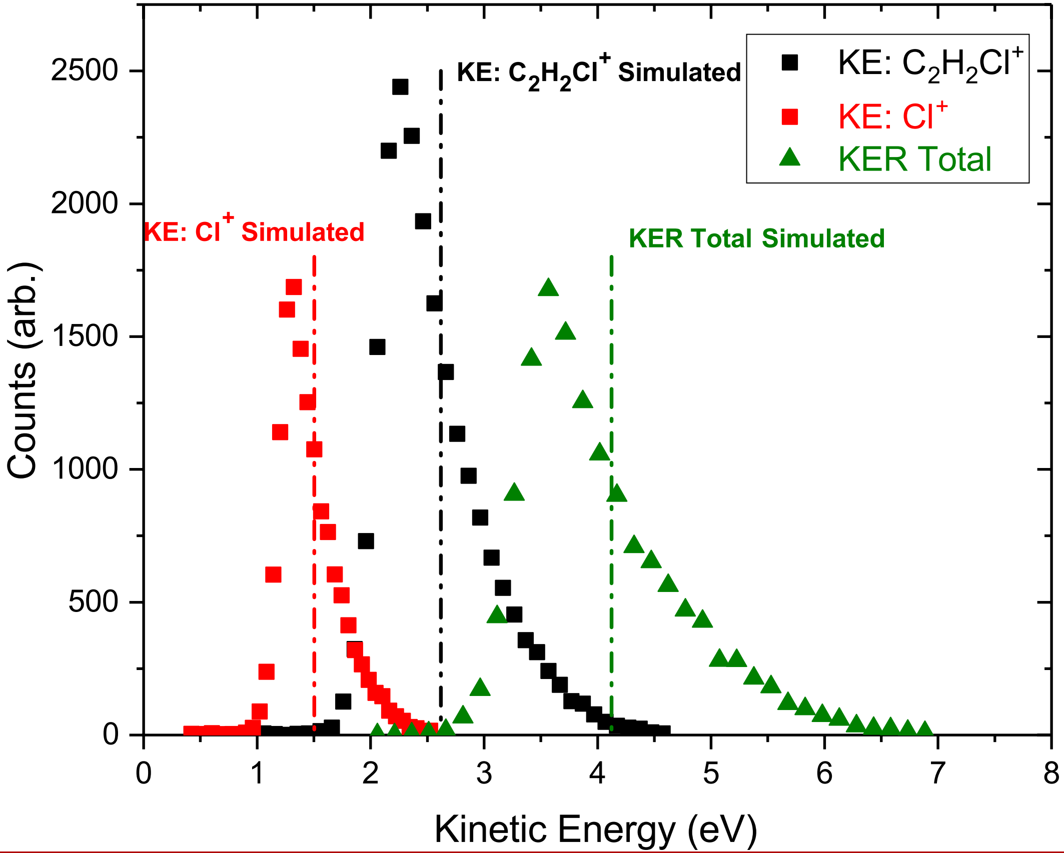
Results: 2 Body - $C_2H_2Cl^+ + Cl^+$

- For trans-$C_2H_2Cl_2$
- The initial geometry calculated by Avogadro



Results: 2 Body - $C_2H_2Cl^+ + Cl^+$

- For trans-$C_2H_2Cl_2$

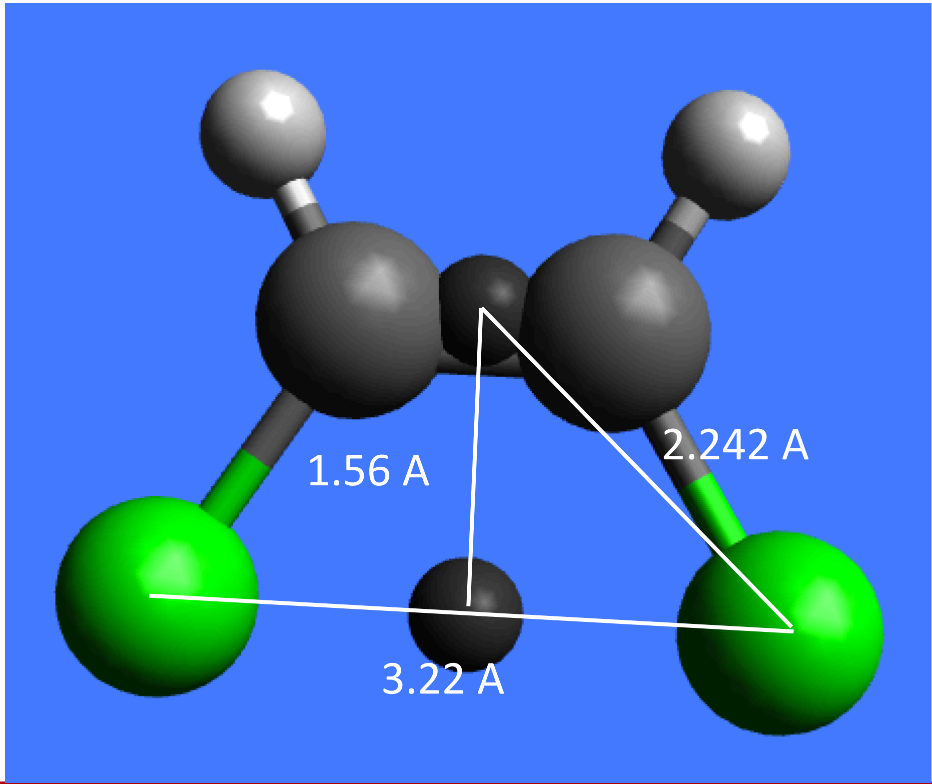


	$C_2H_2Cl^+$	Cl^+	Total KER
Experimental Energy	2.2613eV	1.3026eV	3.5678eV
Simulated Energy	2.6198eV	1.5032eV	4.1230eV

Results: 3 Body - $C_2H_2^+ + Cl^+ + Cl^+$

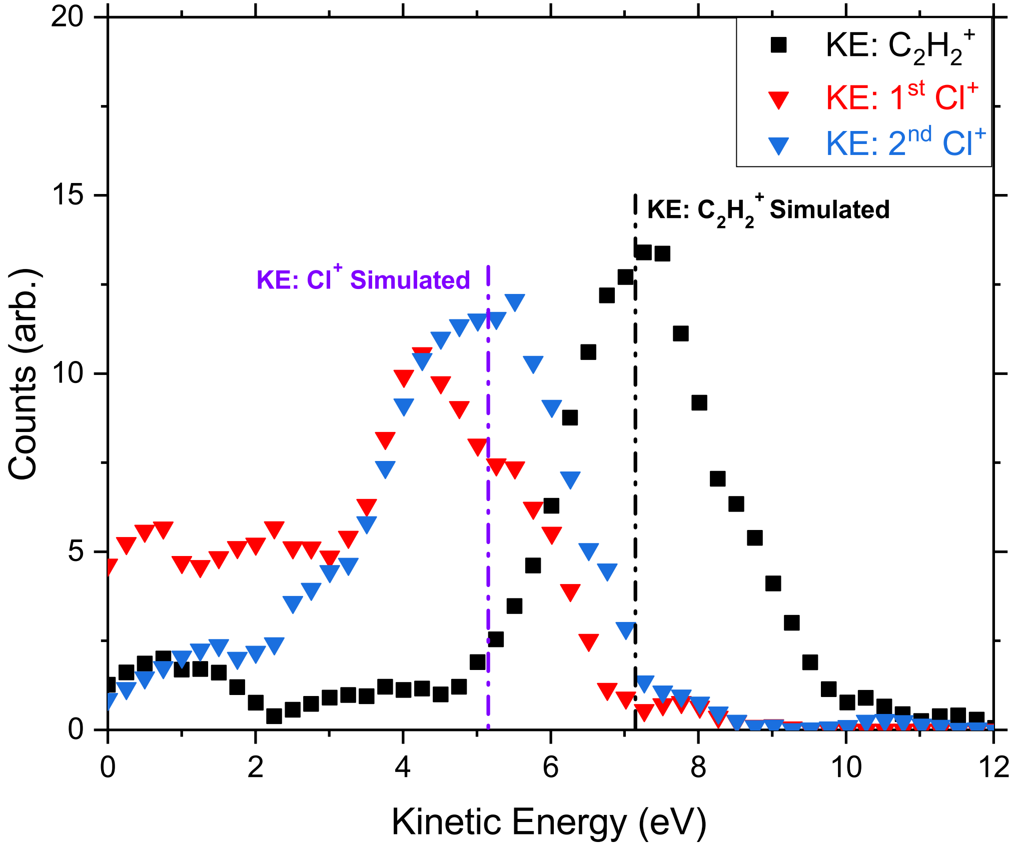
- For cis-$C_2H_2Cl_2$

- The initial geometry calculated by Avogadro



Results: 3 Body - $C_2H_2^+ + Cl^+ + Cl^+$

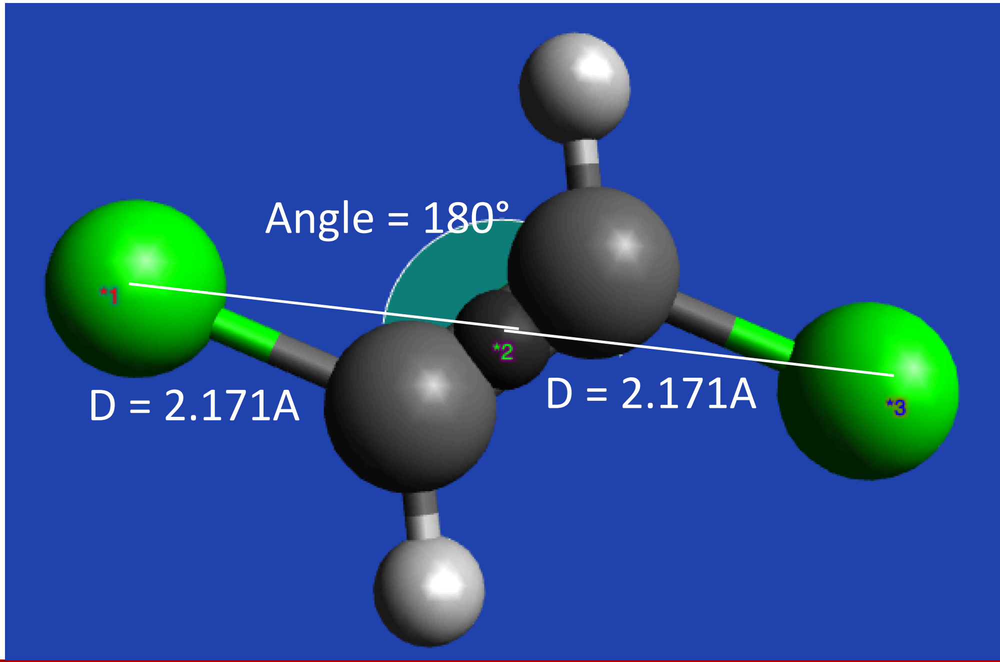
- For cis-$C_2H_2Cl_2$



	$C_2H_2^+$	Cl^+	Cl^+	Total KER
Experimental Energy	7.012eV	5.113eV	5.063eV	15.000eV
Simulated Energy	7.147eV	5.154eV	5.154eV	17.455eV

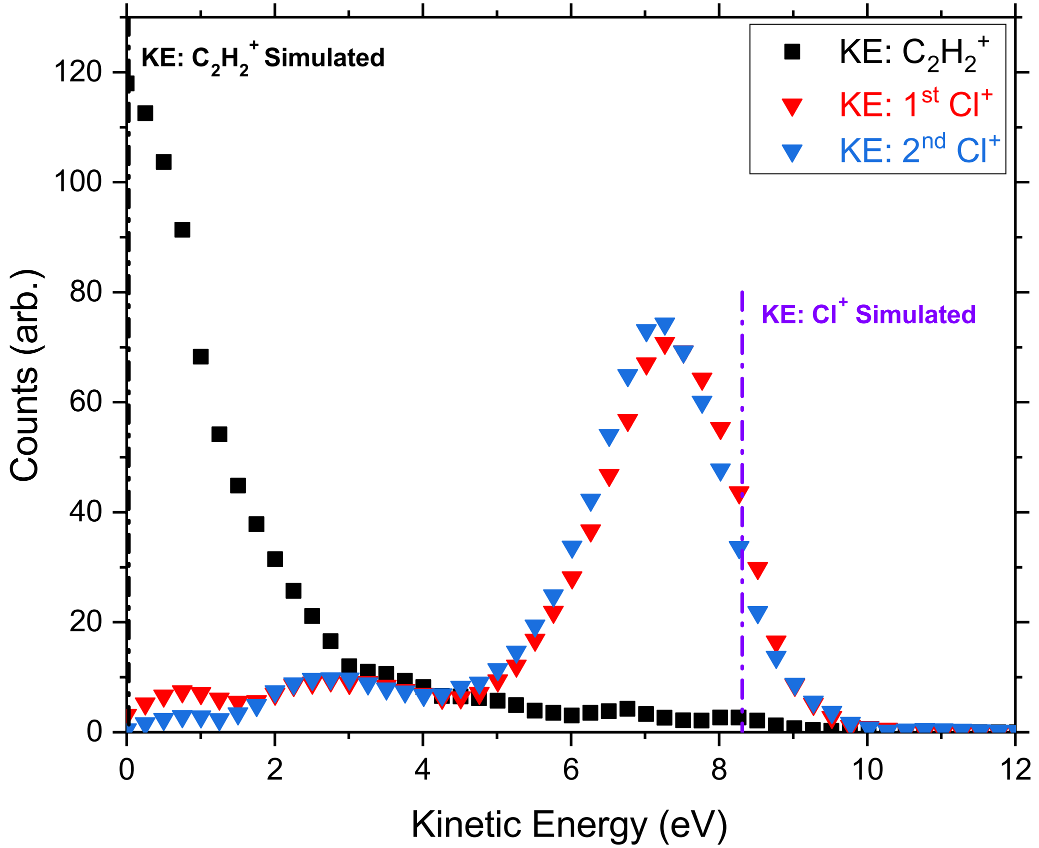
Results: 3 Body - $C_2H_2^+ + Cl^+ + Cl^+$

- For trans-$C_2H_2Cl_2$
- The initial geometry calculated by Avogadro



Results: 3 Body - $C_2H_2^+ + Cl^+ + Cl^+$

- For trans-$C_2H_2Cl_2$



	$C_2H_2^+$	Cl^+	Cl^+	Total KER
Experimental Energy	0.301eV	7.368eV	6.967eV	14.899eV
Simulated* Energy	0eV	8.312eV	8.312eV	16.624eV

*This is assuming the bond angle is exactly 180°

Conclusion

- Coulomb Explosion Imaging is a useful technique to distinguish molecular isomers.
- Numerical Coulomb Explosion simulations agree well with the measured kinetic energies.
- Because the simulation uses an instantaneous pulse, it gives an upper bound to the energy released from the Coulomb Explosion
- There is a clear difference in the energies for cis- and trans-dichloroethene and this method can easily differentiate between them.

What's Next

- Use this simulation to investigate other break up channels.
- Improve approximation method for initial condition of the molecules.
- Integrate pulse width into the simulation.
- Expand Coulomb Explosion simulation to work for 4 Body Break ups.

Acknowledgments

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Thank you!

QUESTIONS?

References

[1] Vager, Z., Naaman, R. & Kanter, E. P. Coulomb explosion imaging of small molecules. *Science* **244**, 426–431 (1989).

[2] I.A. Bocharova et al., "Time-resolved Coulomb-explosion imaging of nuclear wave-packet dynamics induced in diatomic molecules by intense few-cycle laser pulses", Phys. Rev. A 83, 013417 (2011).

[3] Ablikim et al. Identification of absolute geometries of *cis* and *trans* molecular isomers by Coulomb Explosion Imaging. *Scientific Reports* **6**, Article number: 38202 (2016).

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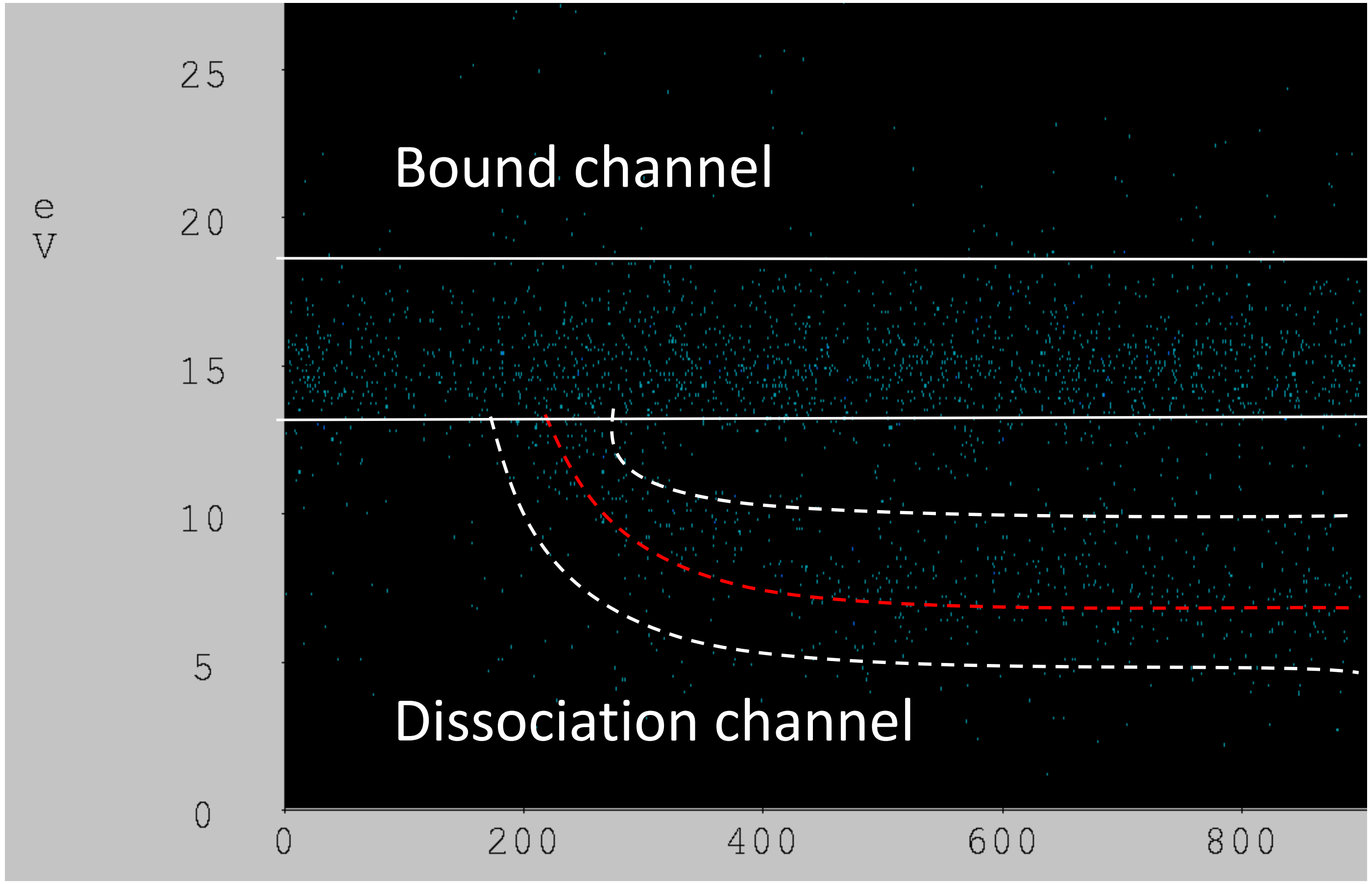
Experiment and Setup



- ➢This channel denotes that there is dissociation of the parent molecule.
- ➢Coincident pairs will only occur after a Coulomb Explosion event.