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

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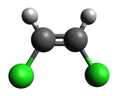
Abstract:

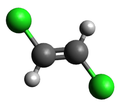
The Coulomb Explosion Imaging technique [1,2] is used to study cis- and trans-dichloroethene molecules in the gas phase. The molecules are photo-ionized by 790nm, 25 fs near-infrared laser pulses, and the resulting fragments are measured by coincident ion momentum imaging. The experimental data is then compared to numerical simulations of the Coulomb Explosion process carried out by utilizing the 8th order Runge-Kutta method.

Motivation:

The goal of this project is to use Coulomb Explosion Simulation to model the repulsion of ionized fragments of cis-$C_2H_2Cl_2$ and trans-$C_2H_2Cl_2$ after being photo-excited by femtosecond laser pulses. This model is then used to find the the kinetic energies of the ionic fragments for a given channel.



cis-dichloroethene



trans-dichloroethene

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

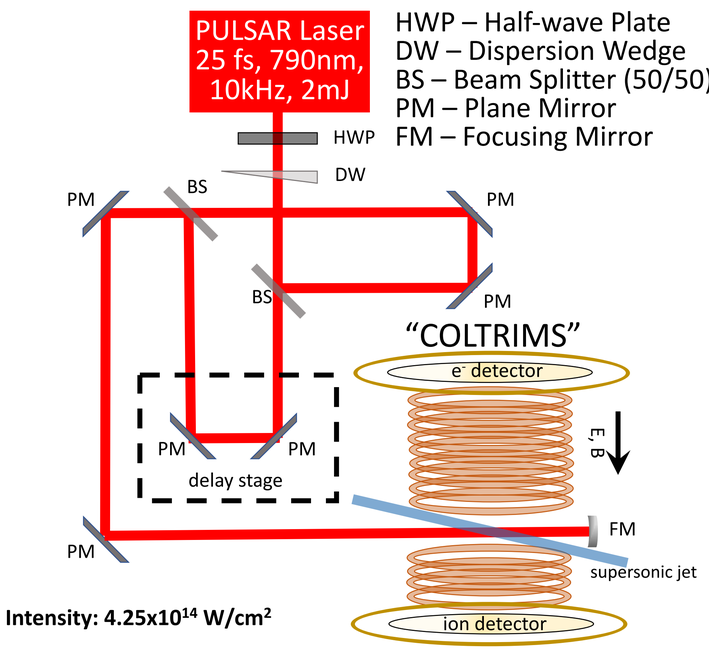
$$F_{C_2H_2^+}(r_1) \propto \frac{1}{R_{12}^2} + \frac{1}{R_{13}^2}$$

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

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

- 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.

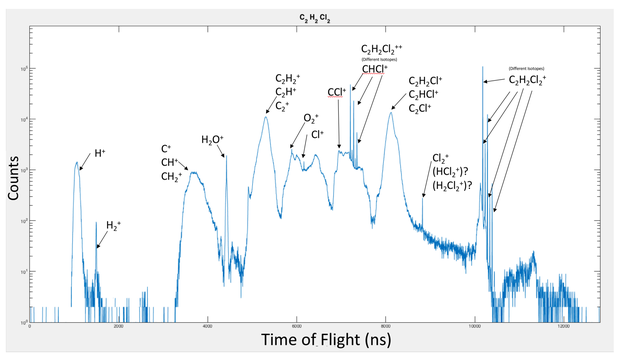
Experiment and Setup:



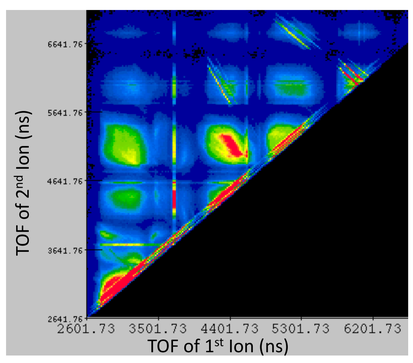
Intensity: 4.25×10^{14} W/cm^2

- Supersonic Jet of $C_2H_2Cl_2$ molecules is directed into the beam path to be photo-ionized.
- The E-field then directs the ions to the ion detector
- The detector can measure the Time of Flight (TOF) of each ion.
- The detector also measures photo-ions that are in coincidence (PIPICO).
- These coincidence channels show the fragments the molecule dissociates into.

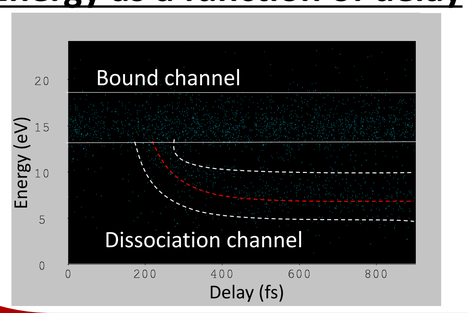
TOF Spectra



PIPICO



Energy as a function of delay



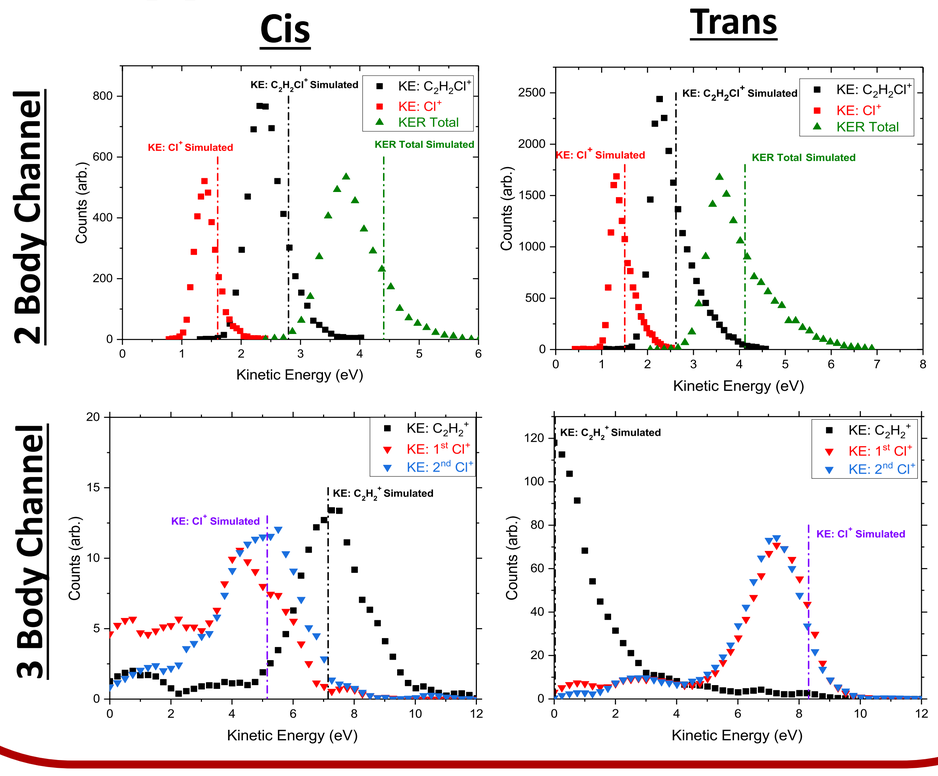
- This channel denotes that there is dissociation of the parent molecule.
- Coincident pairs will only occur after Coulomb Explosion.

Results:

- The Coulomb Explosion simulation works well for both the cis- and trans- parent molecule.
- The photo-ionized channels of interest:

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

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



Conclusion:

Coulomb Explosion Imaging is a useful technique to distinguish molecular isomers. Numerical Coulomb Explosion simulations agree well with the measured kinetic energies. There is a clear difference in the energies for cis- and trans-dichloroethene and this method can easily differentiate between them.

Acknowledgements:

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References:

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[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).