Coulomb Explosion Imaging and Ionization of CH_2I_2 in Strong Laser Pulses

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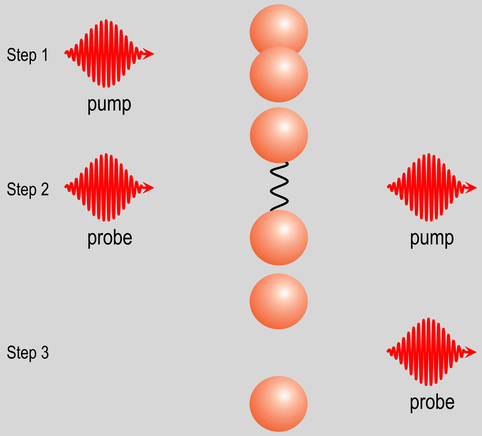
Abstract

Halomethanes, including the diiodomethane (CH_2I_2) molecule, are responsible for the production of reactive halides leading to ozone destruction.[1] Through exploitation of an ultrafast, 25 fs pulse pump-probe setup and high-resolution reaction microscope, time-resolved Coulomb explosion (CE) imaging was established. As a result, ion fragments and dissociation channels subsequent to CH_2I_2 ionization and Coulomb explosion were classified. Furthermore, vibrations within the molecule's structure after excitation were observed.

Pump-Probe Concept

The objective of a pump-probe experiment is to capture a "snapshot" of a structure's molecular motion. The process begins as a "pump" pulse excites vibrations in a molecule. Subsequently, a "probe" pulse further ionizes and, due to Coulomb repulsion, the molecule explodes, see figure below.



The distance between the fragments is reconstructed from the measured fragment energies as

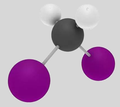
$$E \approx \frac{1}{R}.$$

Experimental Specifications

Pulsar Laser Specifications

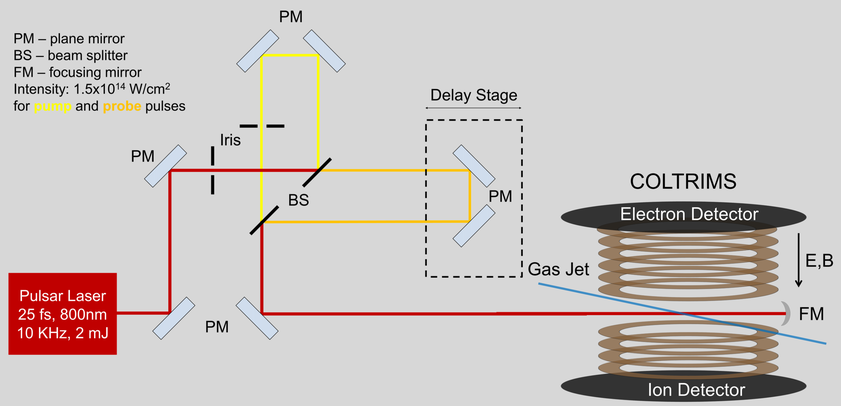
- 25 fs pulse
- Wavelength: 800 nm
- Repetition Rate: 10 kHz
- Focused Pulse Intensity: 1.5×10^{14}W/cm^2

Molecule Specifications



- Molecule: CH_2I_2
- Ionization Potential: 9.34 eV
- Vapor Pressure: 141 torr (0° C)
- Vibrational Periods: 300 fs

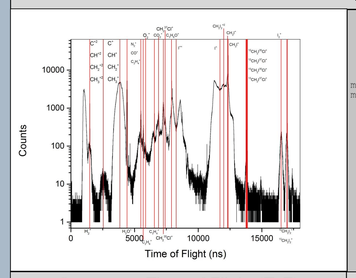
Experimental Setup



The experimental setup consists of an ultrafast, pulse laser directed through a Mach-Zehnder interferometer and into a COLTRIMS setup. The interferometer contains a delay stage responsible for the delay between pump and probe pulses. The COLTRIMS setup detects resulting ions after interactions between the laser and gas jet. This setup is displayed in greater detail in the COLTRIMS section.

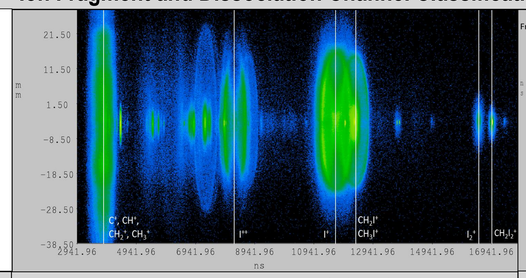
Data Analysis

Ion Fragment and Dissociation Channel Classification



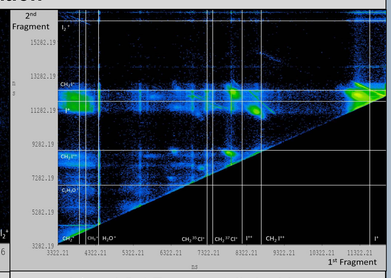
Counts vs. Time of Flight (ToF)

The yield, or count, of each potential ion was plotted as a function of the ion's ToF after ionization. The ion's measured ToF and corresponding mass to charge ratio was utilized to determine possible classifications for each significant ion fragment.



Detector Position vs. Time of Flight (ToF)

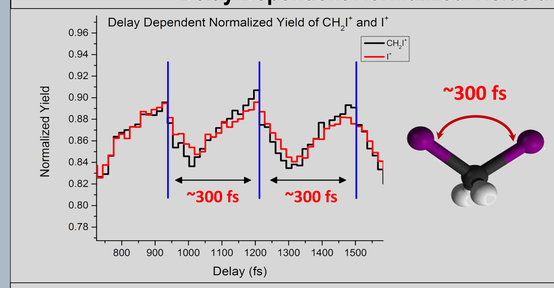
A two-dimensional representation of the ion ToF plot with respect to the detector frame. Each peak in the one-dimensional representation corresponds to an illuminated section in time and space represented in the two-dimensional image. Several peaks have been labeled to illustrate this relationship.



1st and 2nd Fragment Coincidence

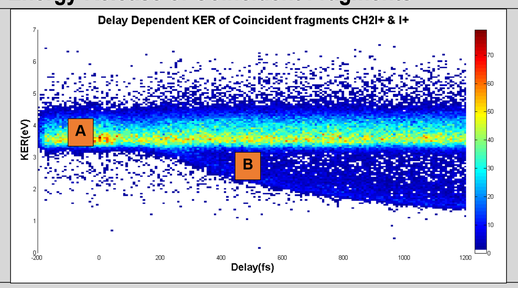
As a result of momentum conservation, plotting the 2nd fragment ToF against the 1st fragment ToF successfully displays two body dissociation channels as straight lines of negative slope. By comparing corresponding ToFs for each fragment with previously classified peaks, various dissociation channels can be established.

Delay Dependent Normalized Yields and Kinetic Energy Release of Coincident Fragments



Delay Dependent Normalized Yields of CH_2I^+ and I^+

Plotting the normalized yields of various ions, in this case CH_2I^+ and I^+, as a function of the delay between pulses allows one to easily observe vibrations, or scissor motions, within the molecule. Furthermore, it is clear that the period of each oscillation is approximately 300 fs.



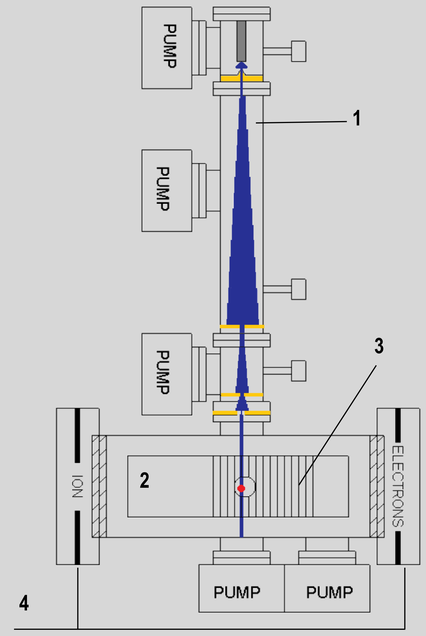
Delay Dependent KER of Coincident fragments CH_2I^+ and I^+

Plotting Kinetic Energy Release of the coincident fragments CH_2I^+ and I^+ as a function of the delay between pulses provides an additional illustration of the molecule's energy during dissociation. The **A-band** displays the delay independent bound wave packet. The **B-band** portrays the delay dependent unbound wave packet. So, as the delay time increases, the internuclear distance between ions and energy can be related by $E \approx \frac{1}{R}$.

COLTRIMS

Main Components:

1. Gas jet
2. Ultrahigh vacuum chamber (10^{-11} torr)
3. Spectrometer
4. Time and position detectors
5. Focusing mirror (not shown)



Original Image: Maharjan, C. M. *Momentum imaging studies of electron and ion dynamics in a strong laser field*. 2007.

Conclusion

Through the use of an ultrafast pump-probe setup and high-resolution techniques, Coulomb explosion imaging was achieved. We classified various ion fragments and dissociation channels after ionization of diiodomethane via pump and probe pulses. Furthermore, we observed bending vibrations within the molecular structure after dissociation. Future work will be conducted to establish a complete movie of the molecule's motion as well as study different light-induced reaction in similar molecules.

Work Cited

1. Nichols, S. R. *Strong field dynamics and control of molecular dissociation.* (ProQuest, 2008).

Acknowledgement:

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