STUDYING ULTRAFAST MOLECULAR DYNAMICS IN PUMP-PROBE EXPERIMENTS WITH FEMTOSECOND LASERS

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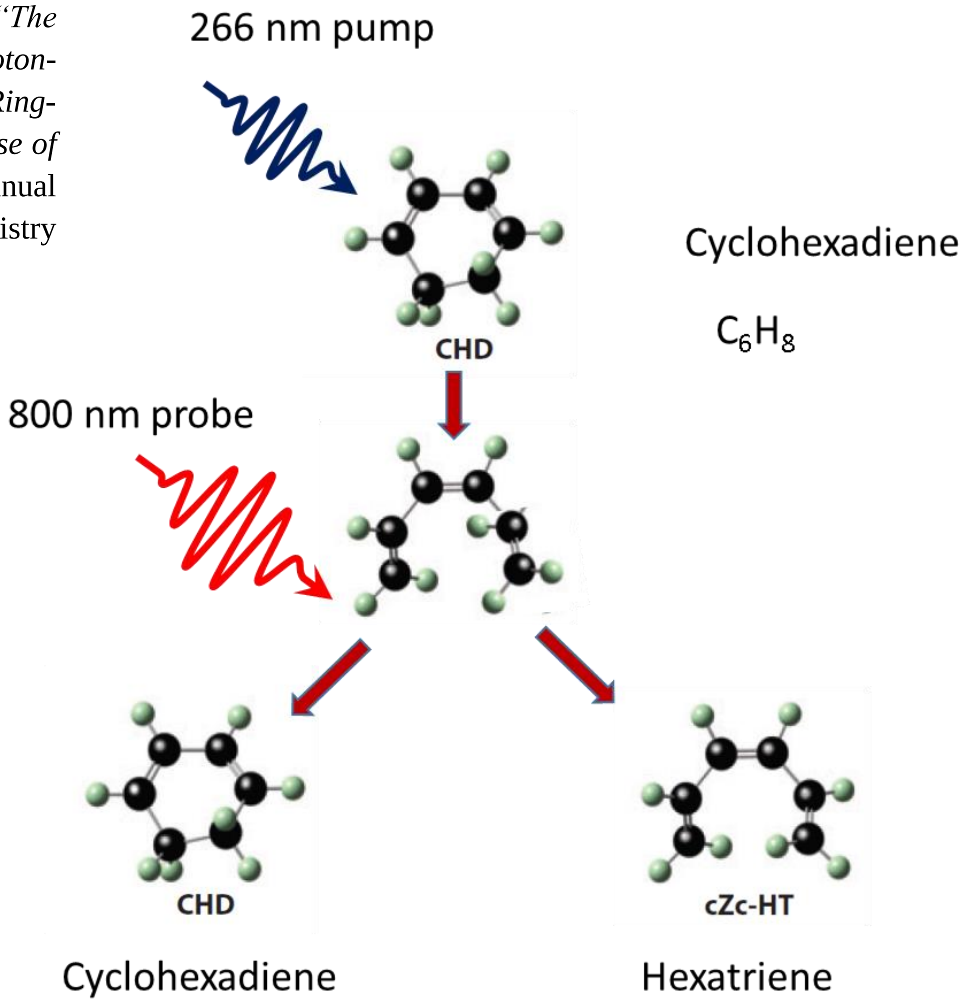
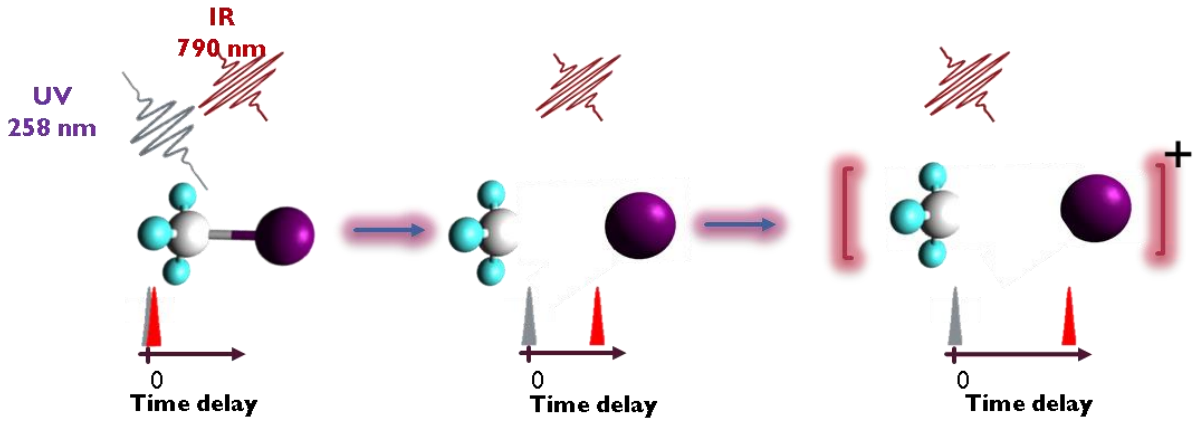
MOTIVATION FOR RESEARCH

- Further understanding
- Optimize reactions
- Faster electronics

S. Deb and P.M. Weber, *"The Ultrafast Pathway of Photon-Induced Electrocyclic Ring-Opening Reactions: The Case of 1,3-Cyclohexadiene"*, Annual Review of Physical Chemistry **62**, 19 (2011).

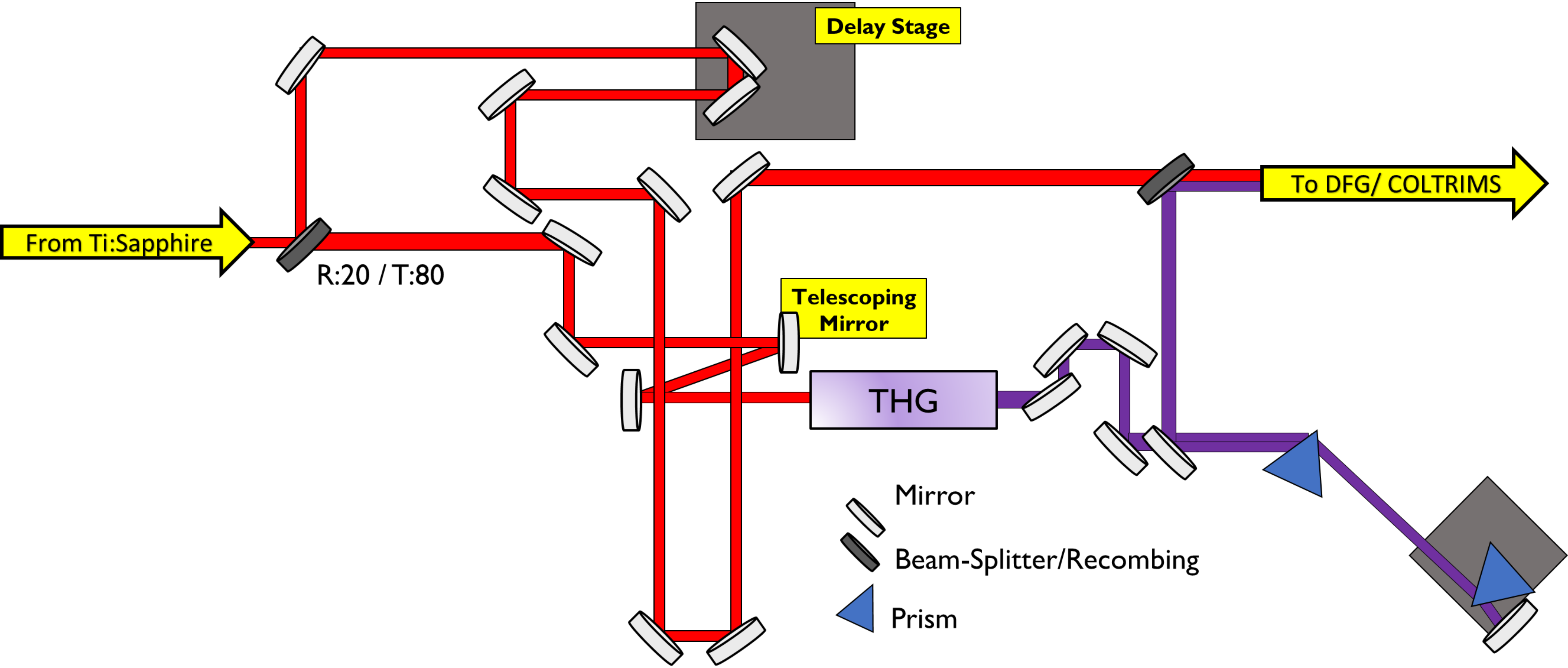


Iodomethane

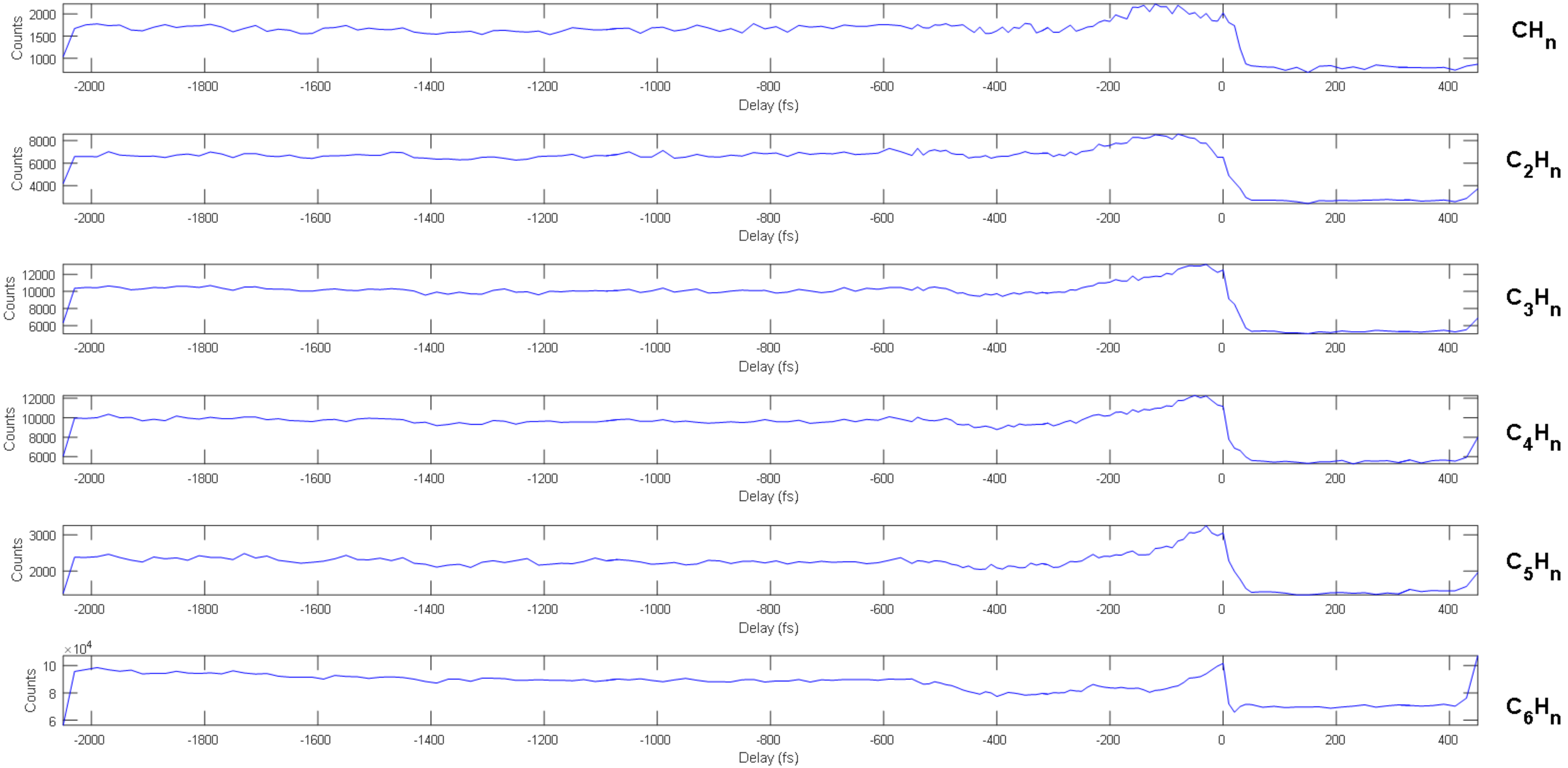


J. Durá, R. de Nalda, G. A. Amaral, and L. Bañares, *"Imaging transient species in the femtosecond A-band photodissociation of CH_3I", J. Chem. Phys.* ***131****, 134311 (2009).*

OPTICAL SET-UP

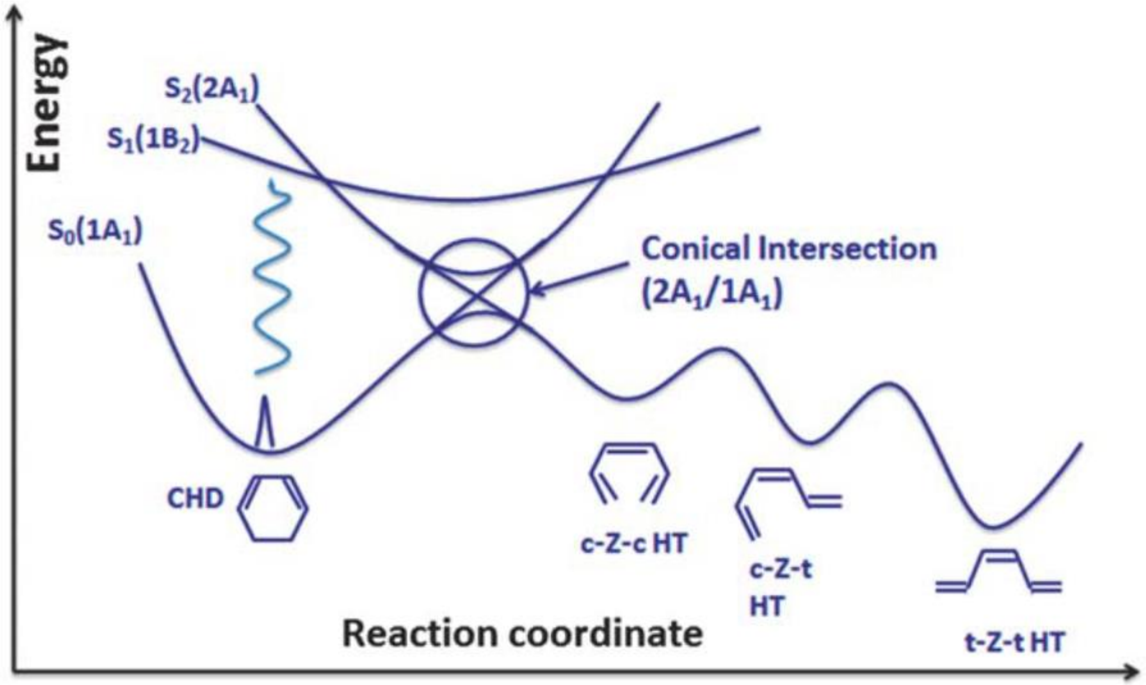


DELAY STAGE FOR TEMPORAL OVERLAPPING



ISOMERIZATION OF CYCLOHEXADIENE (CHD)

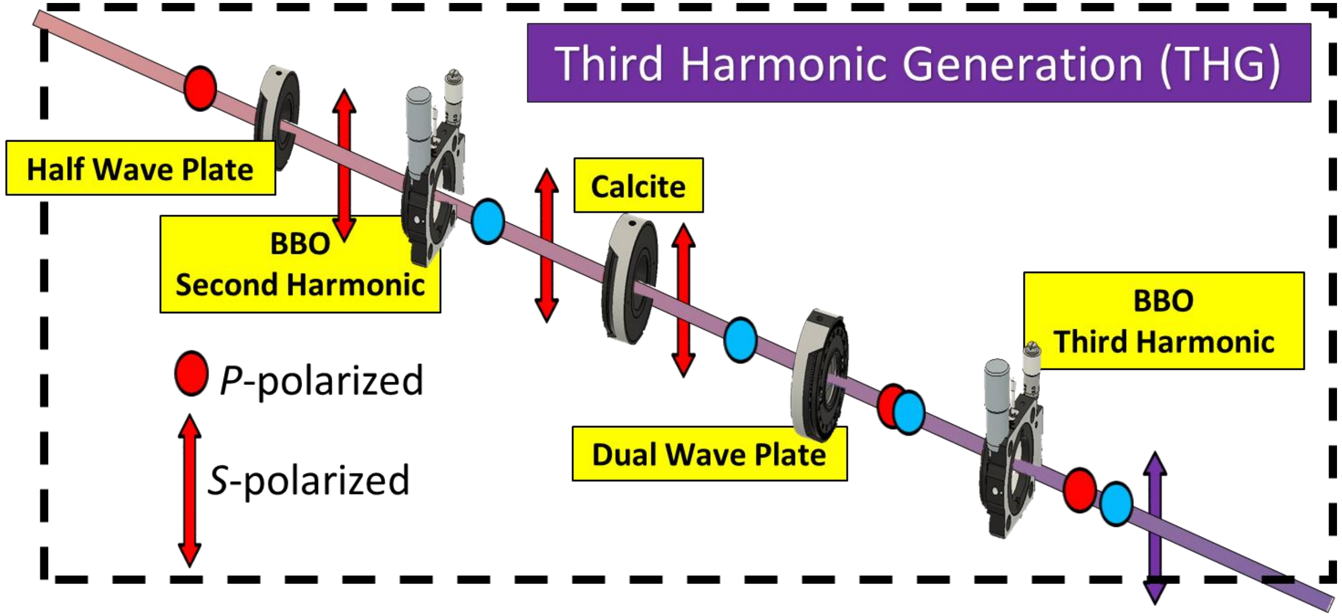
- Excite CHD molecule with single photon UV
- Within 100 fs or less after excitation CHD reaches the conical intersection.
- Short pulses are required to be able to analyze the event effectively.



Bucksbaum and Petrovic *Faraday Discuss.*, **163**, 475–484 (2013)

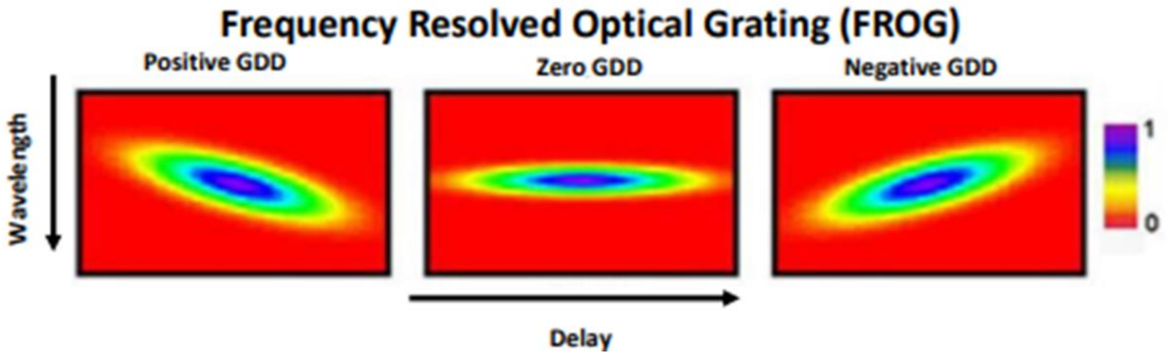
3RD HARMONIC GENERATION

- $c = \lambda \nu$
- First we double the frequency of the IR beam
- Using sum frequency generation, we can “beat” the two frequencies together
- Frequency and wavelength are indirectly proportional
- Causes a positive group delay dispersion (GDD)

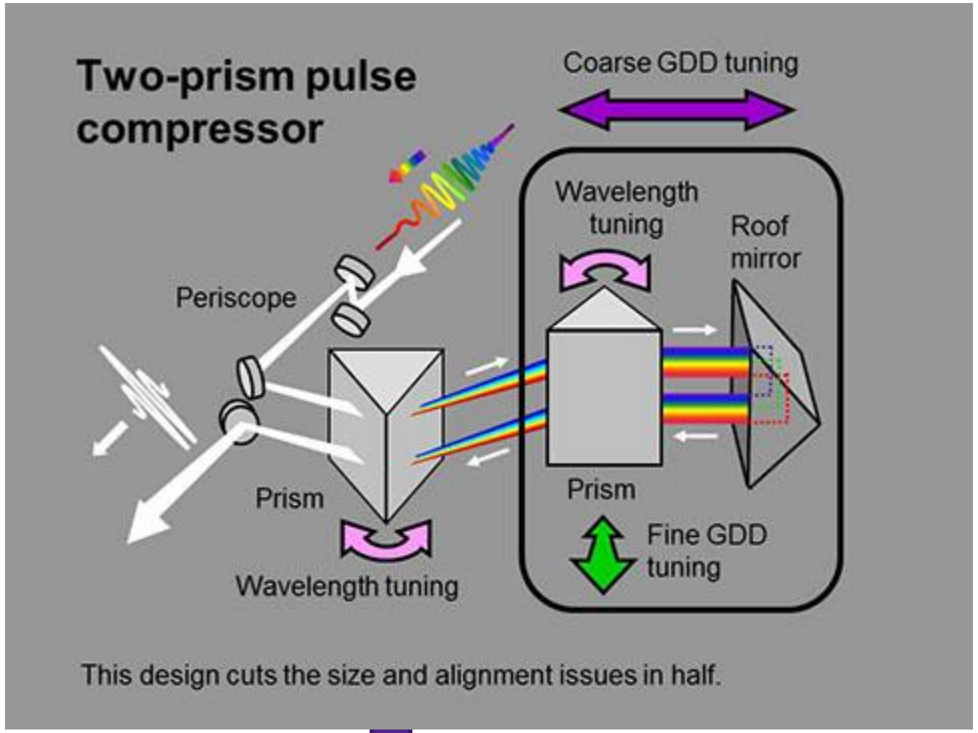


USING PRISM COMPRESSOR TO COMPENSATE FOR GDD

$$GDD = \frac{\lambda^3}{2\pi c^2}\left(\frac{d^2n}{d\lambda^2}\right) * L_c$$



R. Trebino *et. al.*, Review of Scientific Instruments **68(9)**, 32777 (1997)

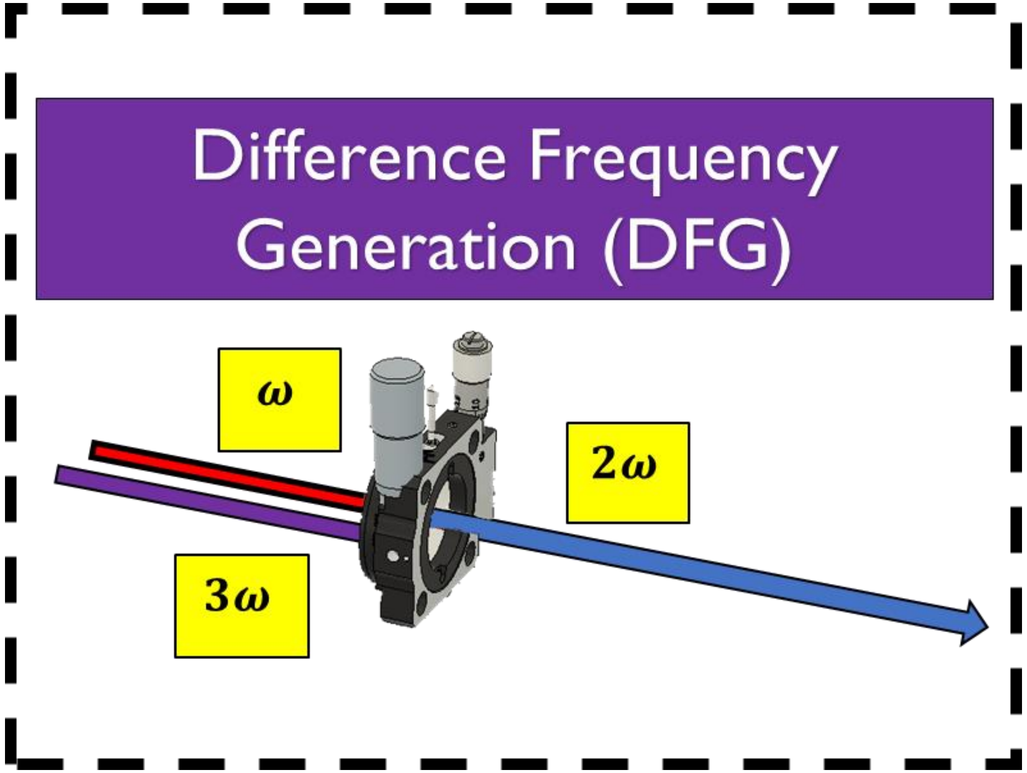


http://frog.gatech.edu/pulse-compression.html

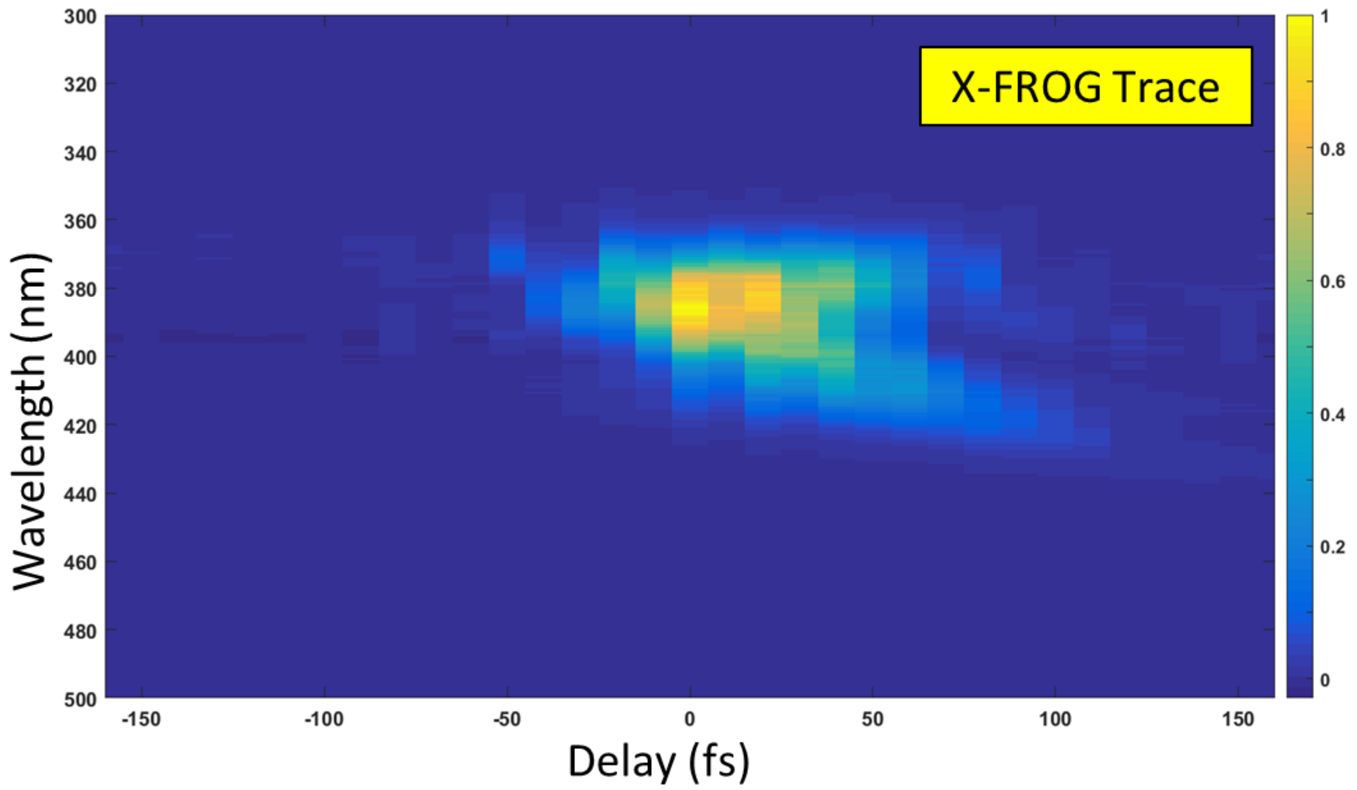
- The speed of light in most materials is different for different wavelengths
- When light travels through a medium the different colors composing the pulse arrive at different times (stretched pulse)
- We make each color travel a slightly longer or shorter path length, such that they all arrive at the same time again

$$GDD_{prism} = \frac{\lambda^3}{2\pi c^2}\left[-4l\left\{2\left(\frac{dn}{d\lambda}\right)^2\right\} + 4\left(\frac{d^2n}{d\lambda^2}\right)\left(2D_{e^{-2}}\right)\right]$$

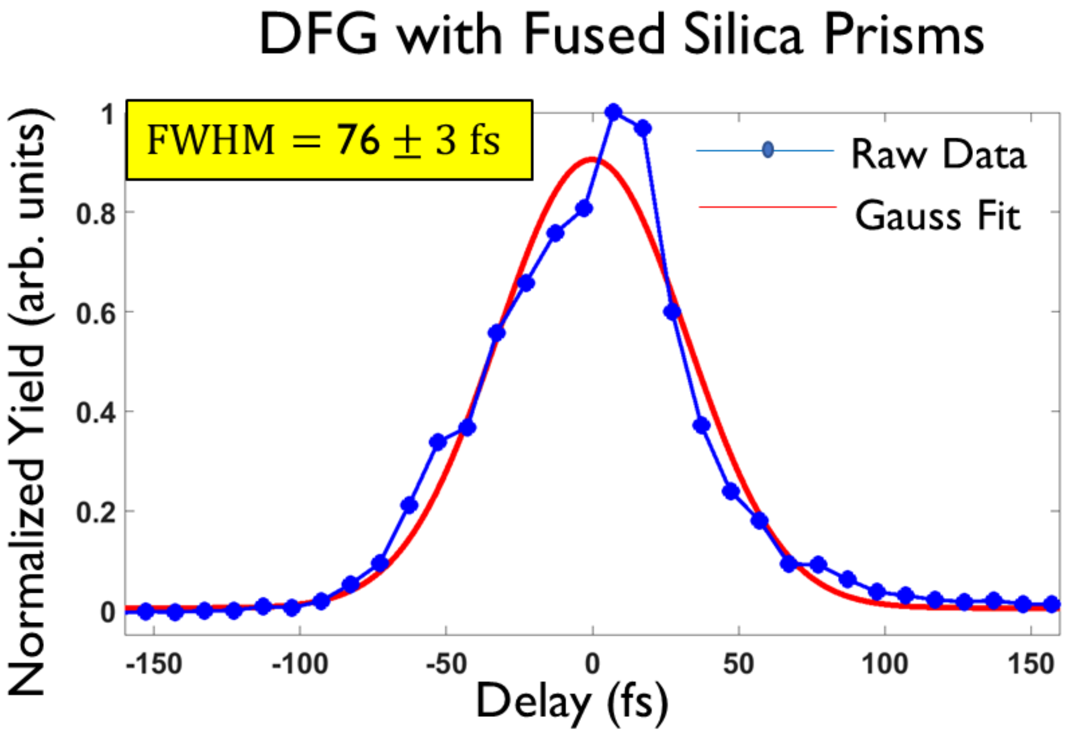
CHECKING THE COMPENSATION WITH DFG

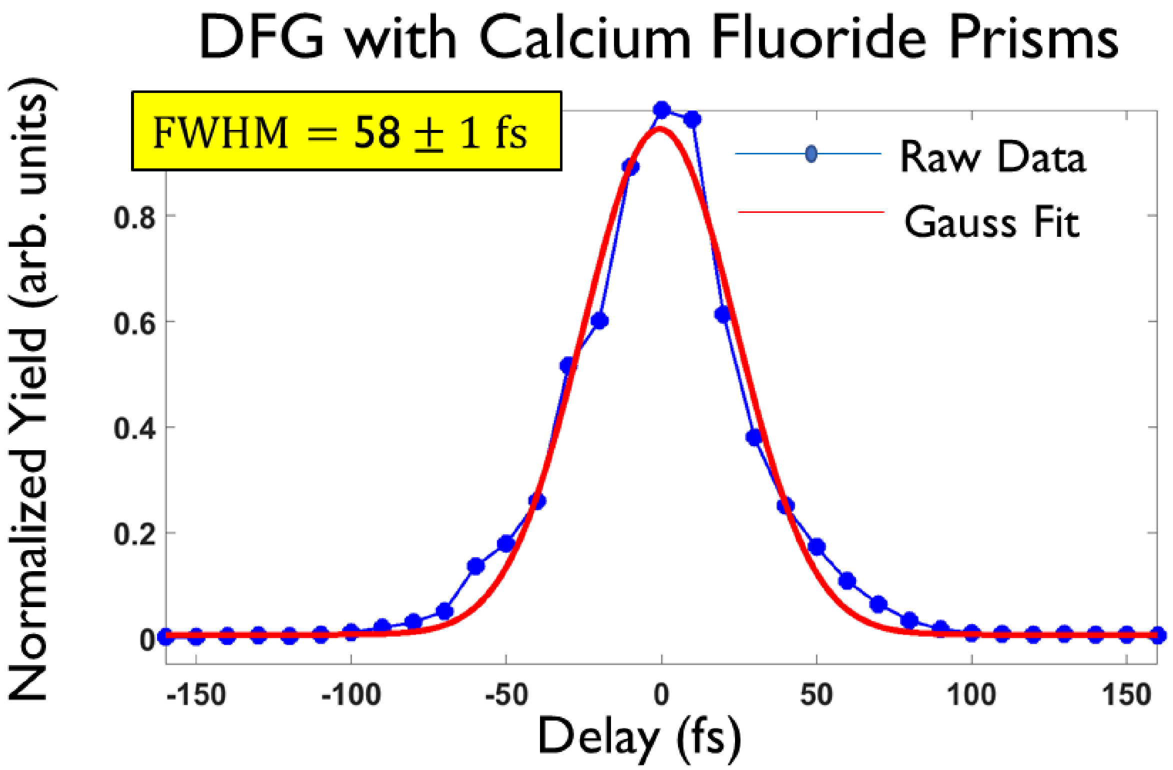


Cross-correlation Frequency Resolved Optical Gating



CHARACTERIZATION OF OUR PULSES





CONCLUSION

- Now that we have the characterization of the UV pulse we can send the laser into our COLTRIMS setup to analyze the cyclohexadiene molecule.
- Using the known strength of the electric field, time of flight, mass divided by the charge of the ion, and the final position of the ion, we hope to be able to reconstruct what the molecule looked like before the reaction.
- Begin to look at the kinetic energy of the fragments as a function of delay and separate low kinetic energy groups from high ones. Might allow us to determine the charge of CHD right before fragmentation.

ACKNOWLEDGEMENTS

I would like to thank Daniel, Artem, Kurtis, and Farzaneh for allowing me to observe, understand, and learn the research process. Also, the NSF for funding this research opportunity. Finally, the other REU students for making the time here memorable.

TIME OF FLIGHT

- Photodiode around the beam path is the trigger to start the time
- Microchannel plate is the trigger to stop the time
- Using kinematic equations we can solve for time

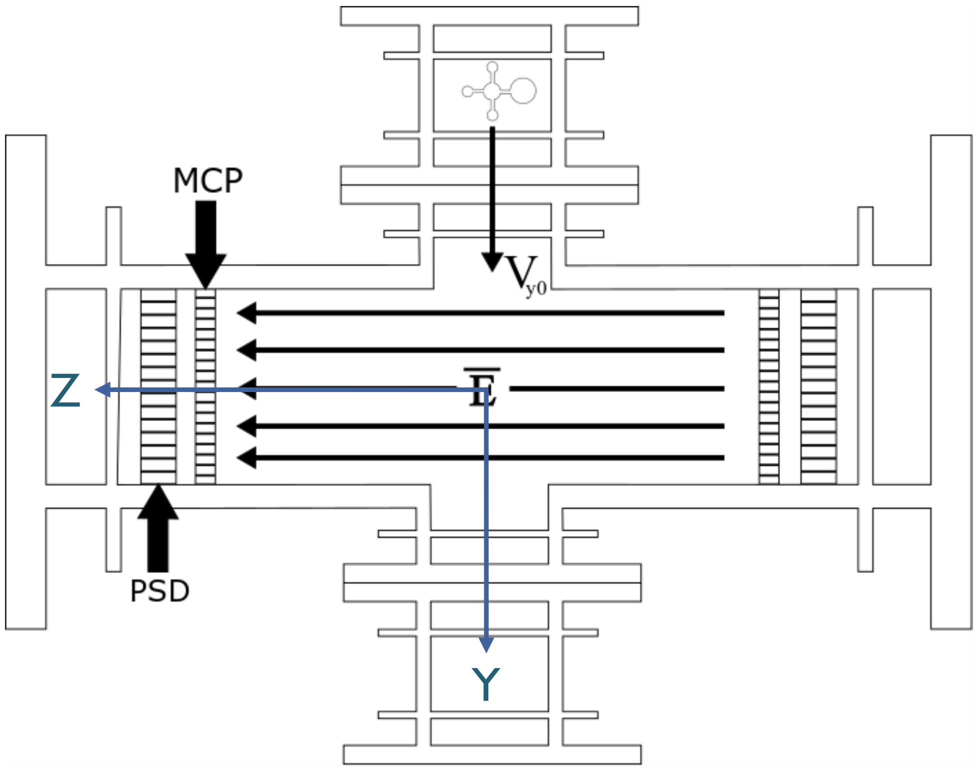
$$F = ma$$

$$F = qE$$

$$z - z_0 = \frac{1}{2} a_z t^2 + v_{z0} t$$

$$\frac{2z}{E} \cdot \sqrt{\frac{m}{q}} = t$$

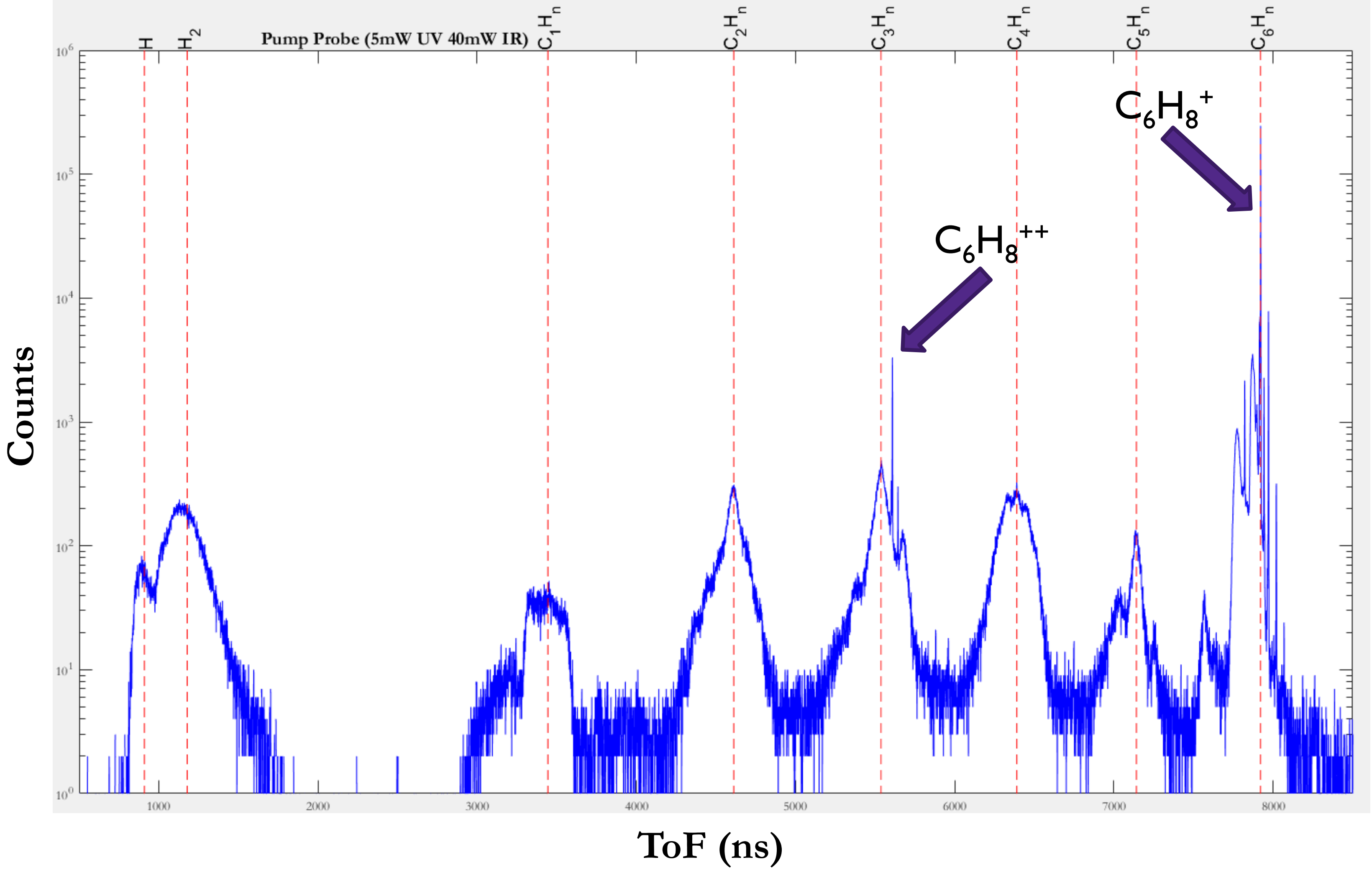
Ion Spectrometer



IDENTIFYING FRAGMENTS

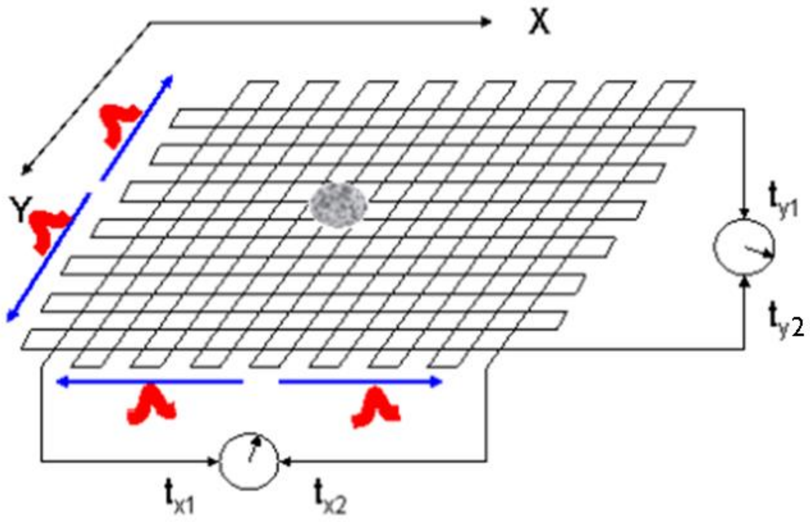
$$T.O.F_1 = C\sqrt{\frac{m_1}{q_1}} + t_0$$

$$T.O.F_2 = C\sqrt{\frac{m_2}{q_2}} + t_0$$



POSITION OF THE FRAGMENTS

- The position sensitive detector is a double spiral wire around a ceramic plate
- Time detectors on each of the four corners
- Using the time it takes a signal to reach the four corners



FRAGMENT TIME OF FLIGHTS AND ASSOCIATED POSITIONS