Characterization of a UV prism compressor for UV-IR pump-probe experiments

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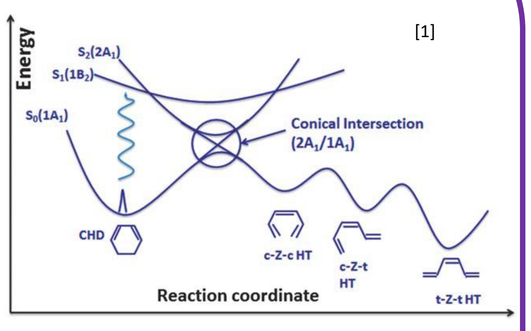
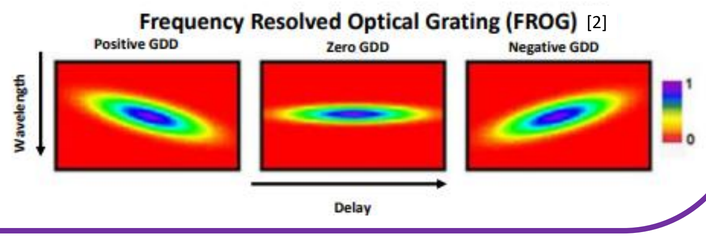
Motivation

Studying molecular dynamics with short pulses

- Dissociation of Cyclohexadiene (CHD) by ultrashort UV (266 nm) laser pulses
  - Under single UV-photon absorption, CHD excited to a repulsive $^3Q_0$ potential energy curve
  - Resulting channel Hexatriene (HT) has a conical intersection reached in 100 fs or less after excitation [1], so short pulse durations necessary to study these dynamics.



- UV pulse generation and characteristic
  - Third-Harmonic-Generation (THG) of a Ti:Sapphire laser (800 nm, 10 kHz, and 25 fs pulses)
  - Non-linear process that causes *positive* Group Delay Dispersion (GDD)
  - Compensate with *negative* GDD through tunable prism pair setup



THG Dispersion and Compensation

Reducing Dispersion from THG

- Dispersive properties of non-linear BBO crystals used for THG

Group delay dispersion (GDD) → Higher order expansion of spectral phase.
→ Varying propagation speeds of light pulse components due to wavelength dependent refractive index will change pulse duration
→ Using thinner crystals length (L_c) reduces GDD

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

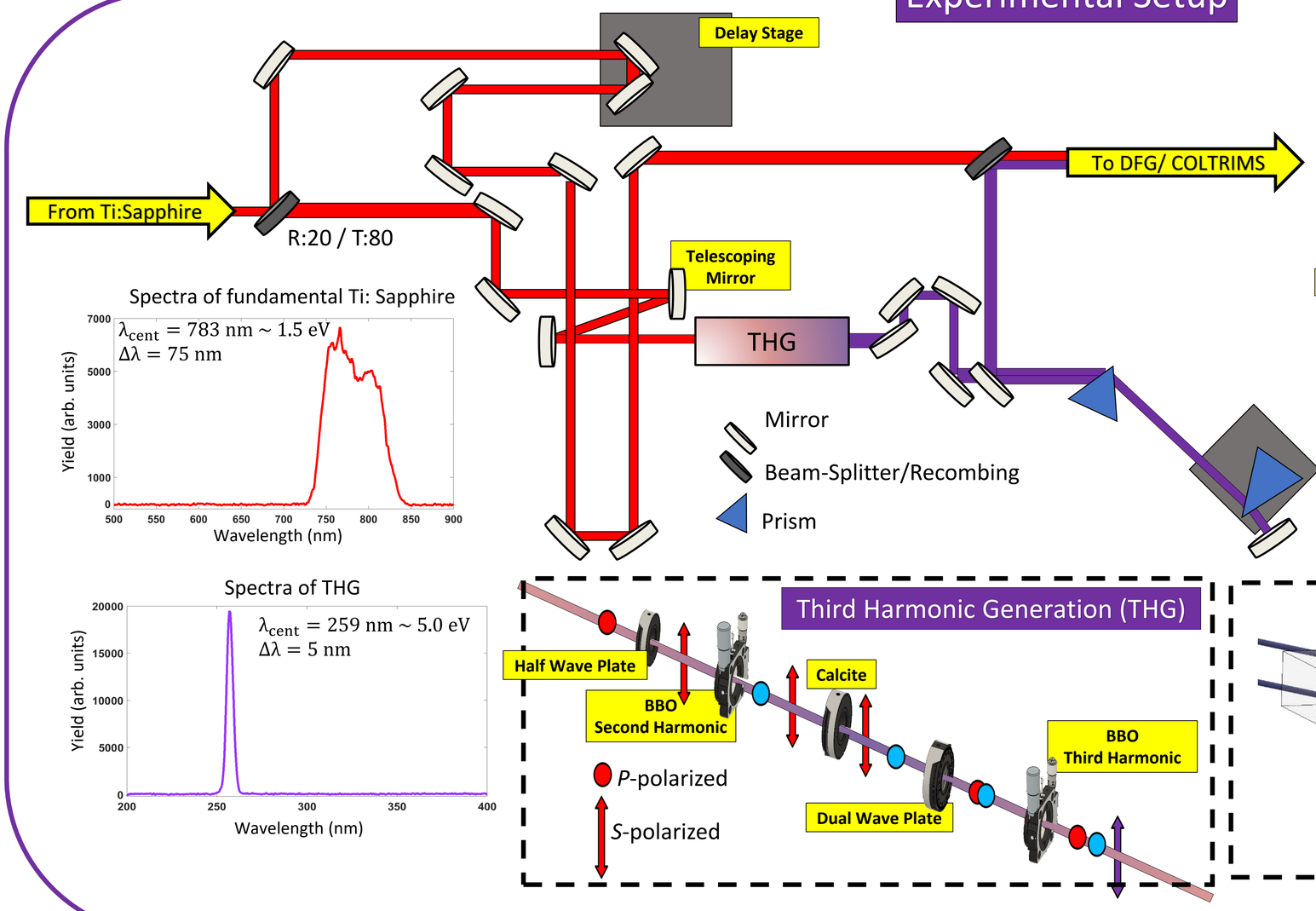
Prism Compressor → Introduces angular dispersion to counter positive GDD from the propagation through air, spectrometer entrance window, and BBO crystals [3,4].
→ Prism pair gives negative GDD that can be optimized by tuning separation distance l.

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

- Difference Frequency Generation (DFG) for cross-correlation measurement of pulse width

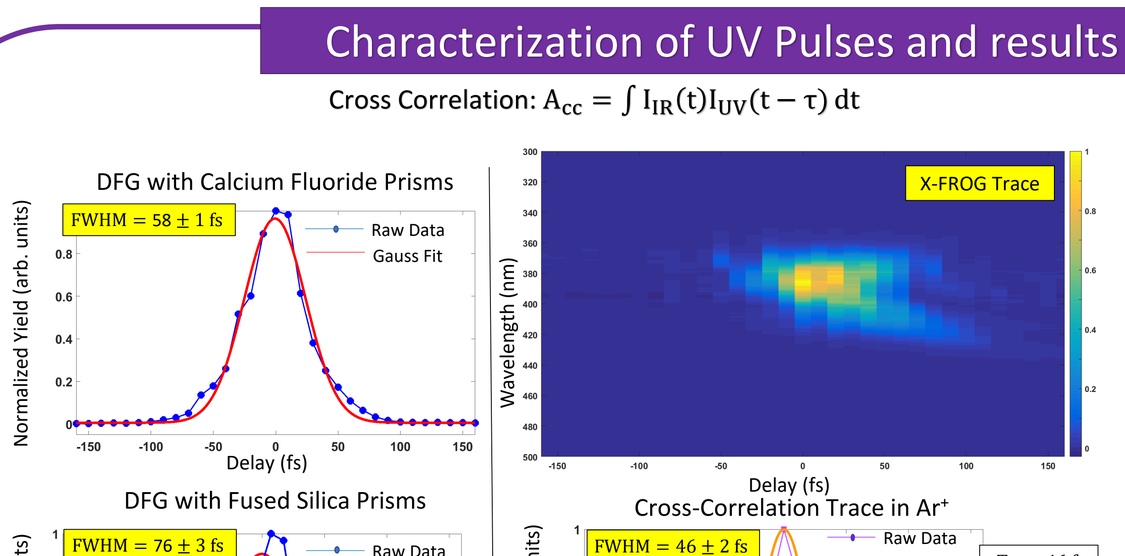
Phase matching conditions in non-linear crystals will transmit frequency that is difference between two pump beams. Can use this signal for cross-correlation to measure pulse-width of Third Harmonic

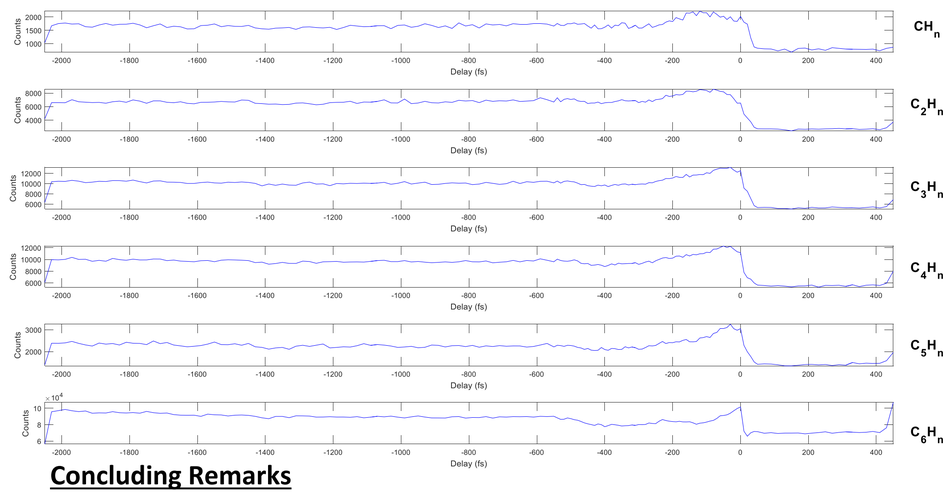
Experimental Setup



Characterization of UV Pulses and results from UV-Pump-IR Probe experiments on CHD

Cross Correlation: $A_{cc} = \int I_{IR}(t) I_{UV}(t-\tau)\, dt$





Concluding Remarks

- Pulse Characterization using cross correlation
  - Prism pair compressor in folded geometry is an effective way to reduce pulse duration
  - Switching material from *fused silica* to *calcium fluoride* improves performance
- Experimental results
  - Found good overlap with low power UV and mid-range power IR
  - X-FROG Trace shows a GDD of roughly zero
- Future goals
  - Extract kinetic energy release near conical intersection and compare to theoretical values
  - Angular distribution defined with angle between p-polarized laser field and dissociation vector

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