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Title: Towards understanding initiation reactions of explosives via ultrafast laser quantum control

Author(s): Margo Greenfield
Shawn McGrane
David Moore
R. Jason Scharff

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Los Alamos
NATIONAL LABORATORY
EST. 1943

Towards understanding initiation reactions of explosives via ultrafast laser quantum control

Optimal control can be utilized to control the initiation reaction of explosives, where time dependent phase shaped electric fields drive the chemical systems towards a desired state. For quantum controlled initiation (QCI) of explosives a pulse is created which seeks to achieve initiation by employing shaped ultraviolet light. QCI will enhance the understanding of energetic material reactions by yielding insight into the characteristics, such as critical "hot spot" size and reaction dynamics, necessary for initiation.

Quantum control experiments require the ability to: 1) phase and amplitude shape an ultrafast laser pulse, 2) measure the effect of pulse shape, and 3) optimize the desired outcome. Pulse shaping is performed with a 4-focal length dispersed fused silica acousto-optic modulator (AOM) at 400 nm in the ultraviolet (UV). Transient absorption spectroscopy is used to measure the pulse shape effects. Both global and local optimization search routines such as genetic algorithm, differential evolution, and downhill simplex are used to search for the optimal pulse shape.

Hexanitroazobenzene (HNAB), Trinitroanaline (TNA) and Diaminoazozyfurazan (DAAF) are excited to the first electronic state with 400 nm light. Our initiation experiments are studying the effect of phase shaped 400 nm pulses on HNAB, TNA and DAAF. Novel transient absorption spectra for each material have been obtained and note worthy regions further investigated with single parameter control (second order spectral phase and energy). Many systems have simple intensity control such as that shown by DAAF. TNA and HNAB have spectral features that are not single parameter driven and are being further investigated with complex control.

Towards understanding initiation reactions of explosives via ultrafast laser quantum control

Margo Greenfield, Shawn McGrane,

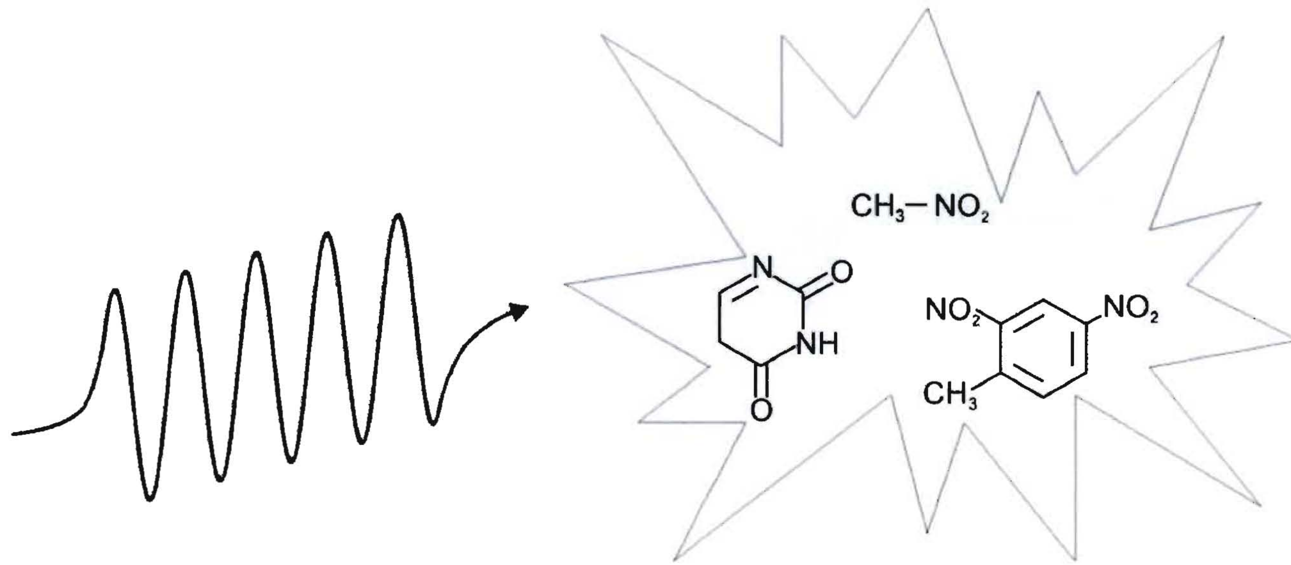
Jason Scharff, David Moore

Why coherent/optimal control?

- Actively control the chemical dynamics of condensed phase systems
  - Liquid and Solid
  - Time dependent shaped electric fields:
    - **OPTIMAL SHAPE**
- Ultimate Goal
  - **Optimal controlled initiation of explosives**
    - Initiation of energetic material
      - Control of photodynamics
    - Increased understanding of the dynamics of energetic materials of interest
      - **Experiment coupled with calculation**

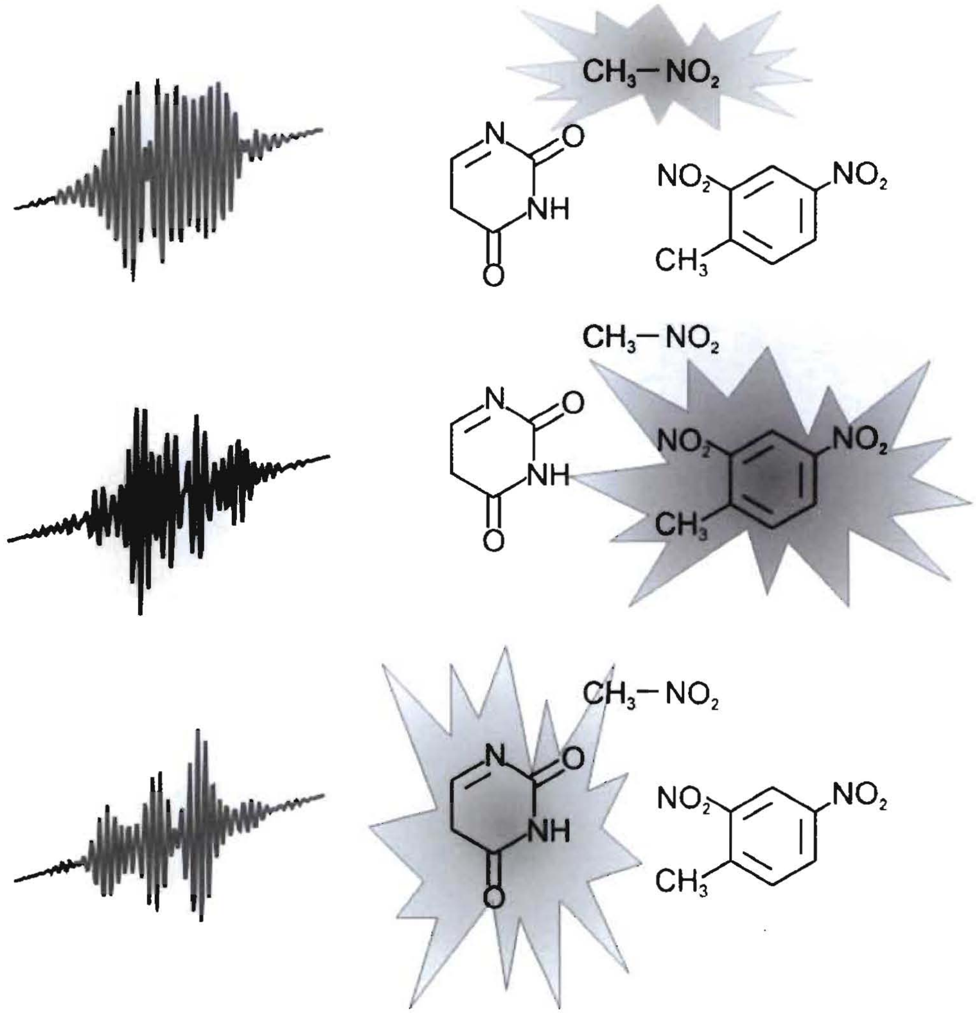
Linear spectroscopy - unshaped pulses

- Conventional steady-state or linear spectroscopy using unshaped pulses
  - **Poor** molecular discrimination



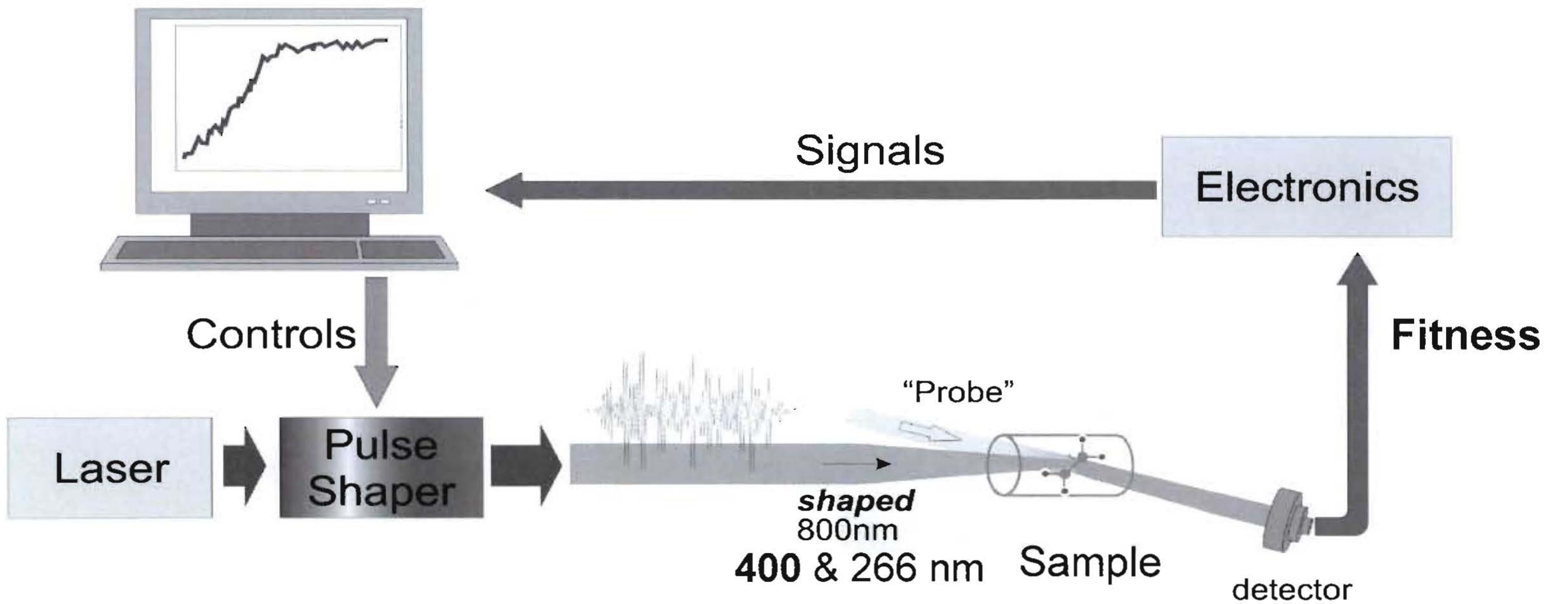
Quantum Optimal Dynamic Discrimination

- **Concept**: Optimally tailored laser pulses (photonic reagents)
  - Enables **selective** addressing of different species



Control by closed loop optimization

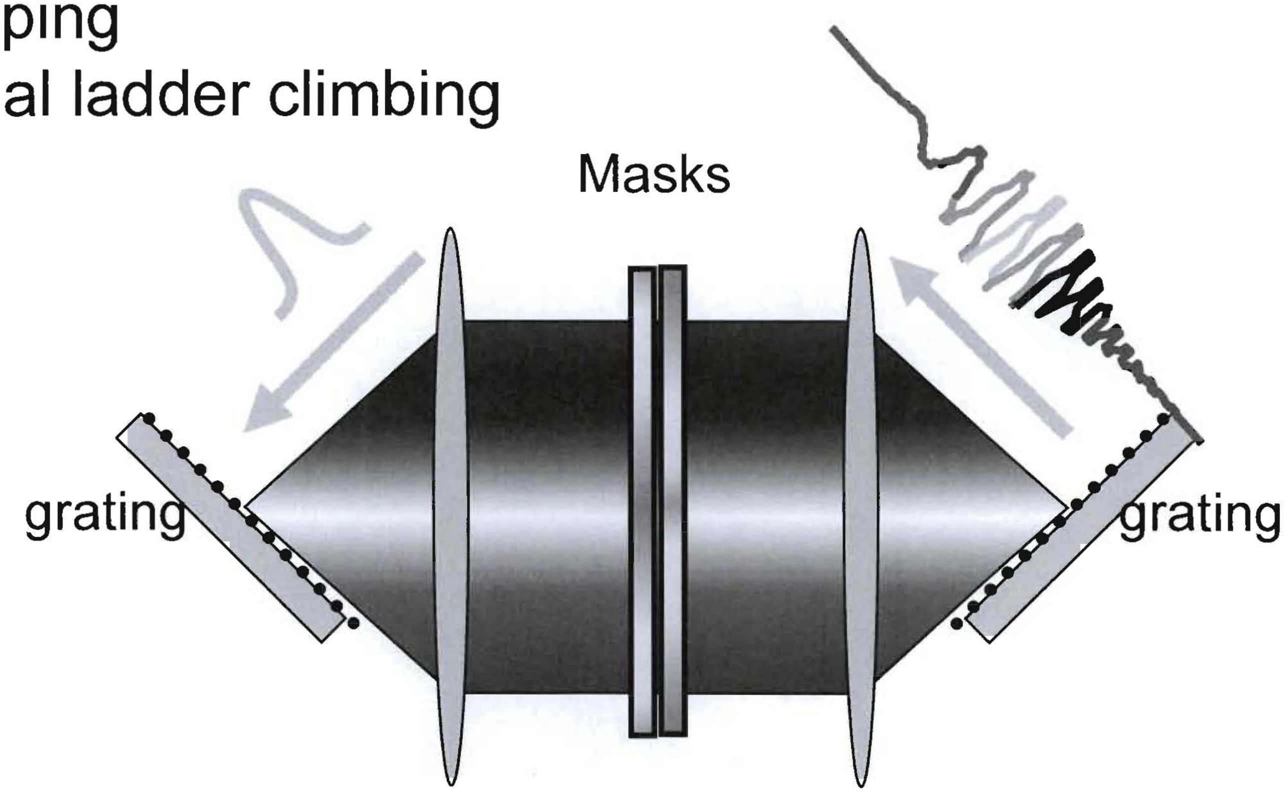
- Learning Loop
  - Searches for optimal pulse using feedback control
    - Initiation experiment: Transient absorption spectrum
  - Can use many optimization routines



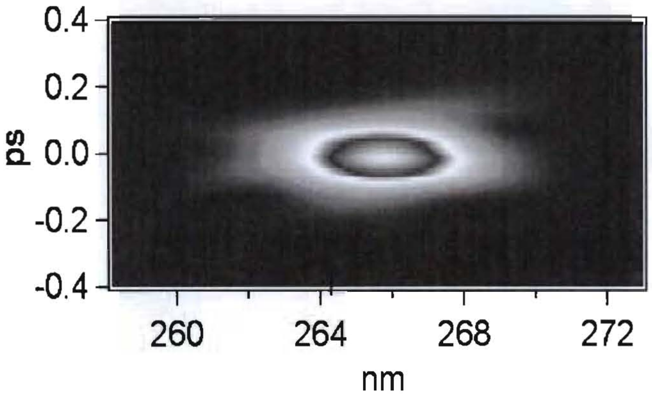
- Shaped 400 nm pump
- Broadband 325-750 nm super continuum probe for transient absorption

Cutting edge pulse shaping

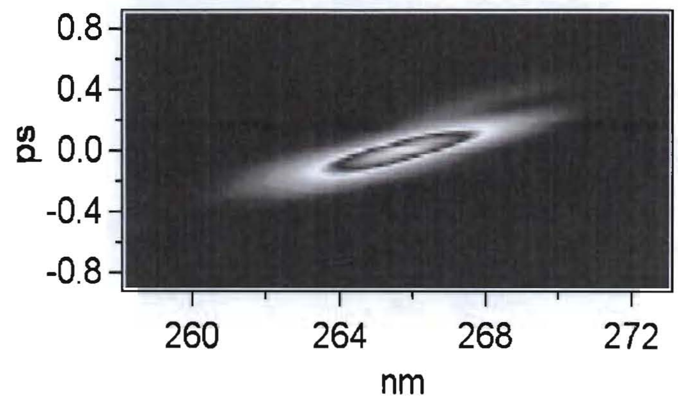
- 400 and 266nm ultraviolet (UV) shaping
  - Allows for single photon processes
  - In the low field limit
- 800 nm shaping
  - Vibrational ladder climbing



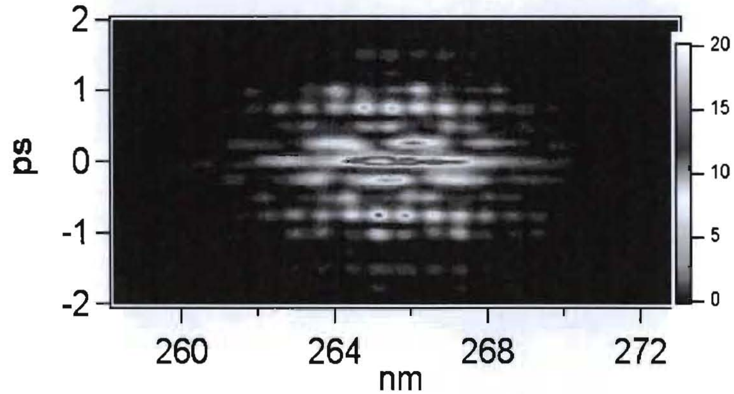
Examples of shaped pulses



Transform limited ~150fs



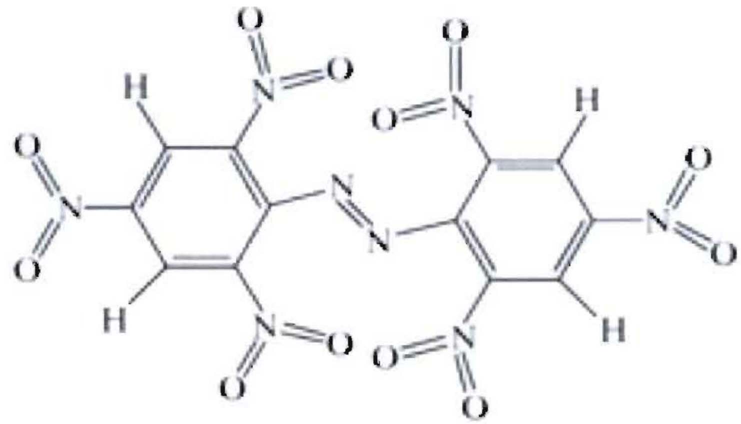
Simple linear chirp



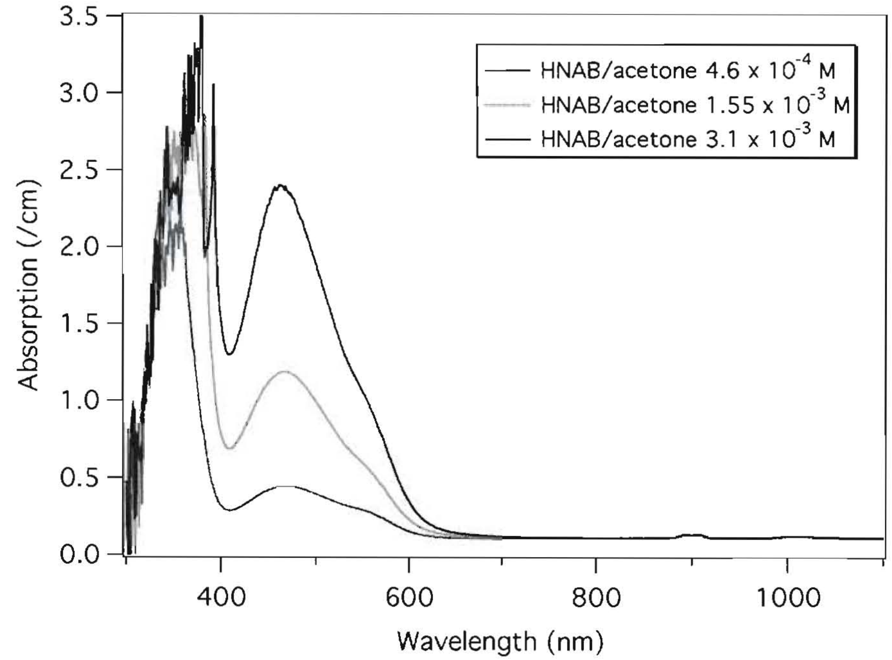
Dual sine waves

Towards optimal control of explosives

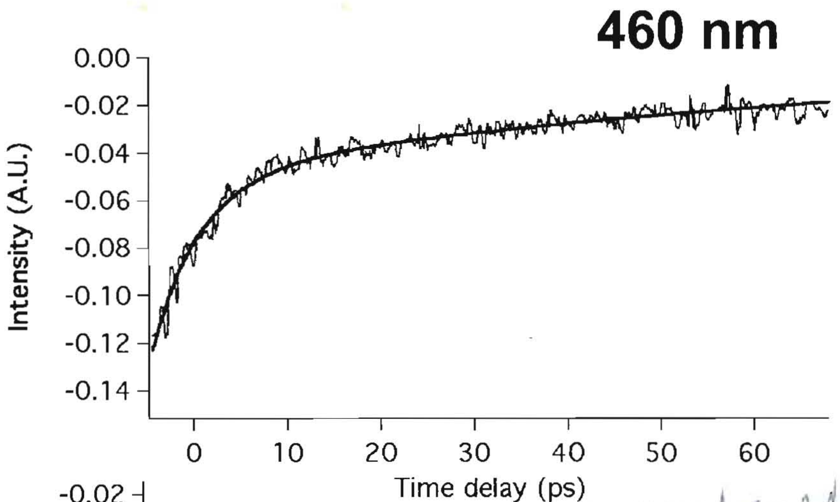
- Potential energy surface unknown
- HNAB absorbs well around 400 nm
- Good starting point for control

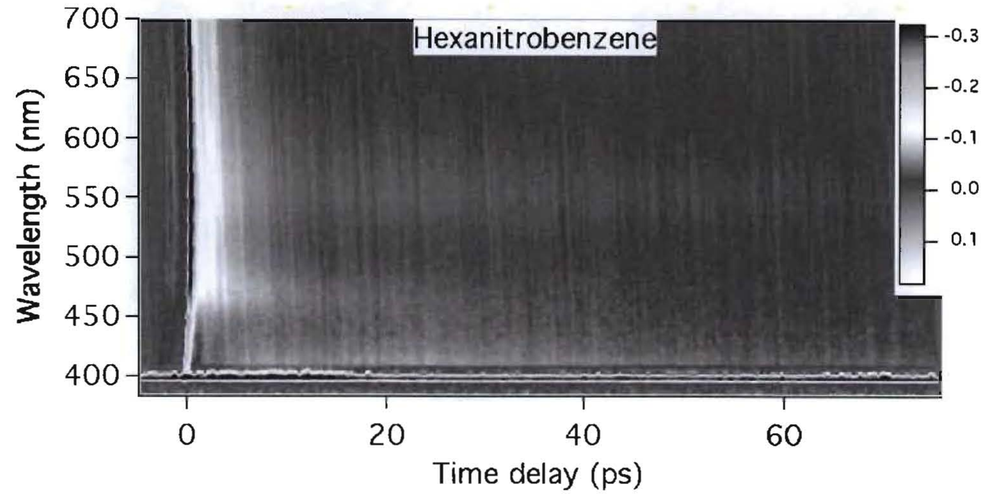


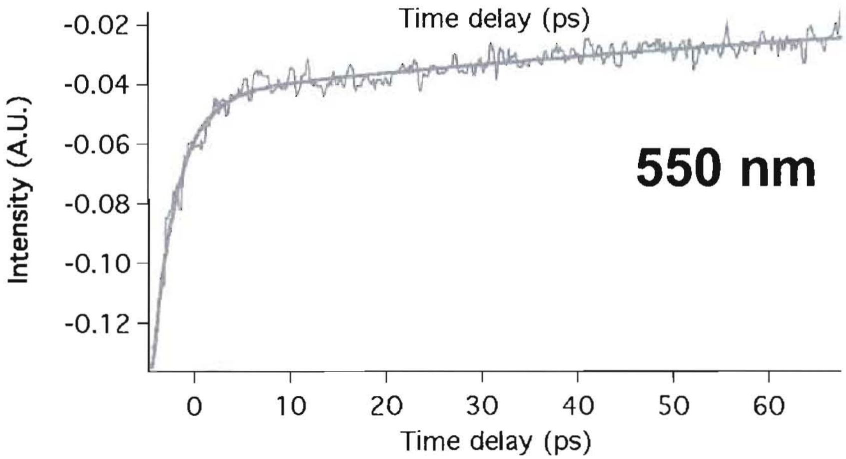
hexanitroazobenzene



HNAB 1.38 mM solution in Acetone







- Biexponential fits
  - **460 nm**

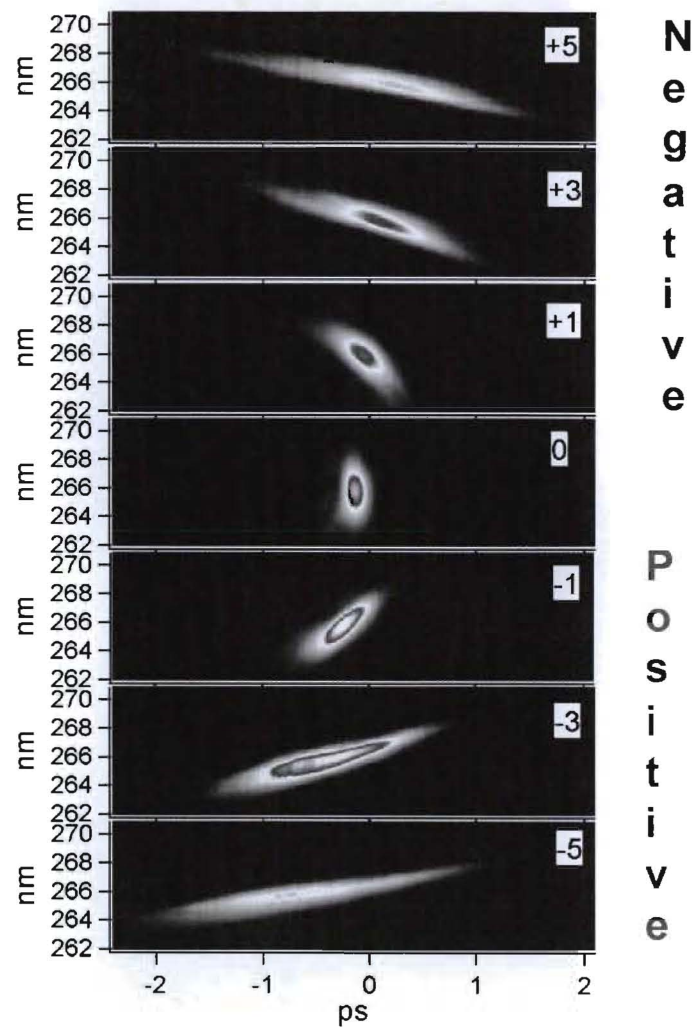
    τ_1=8ps τ_2=130ps
  - **550 nm**

    τ_1=17ps τ_2=223ps

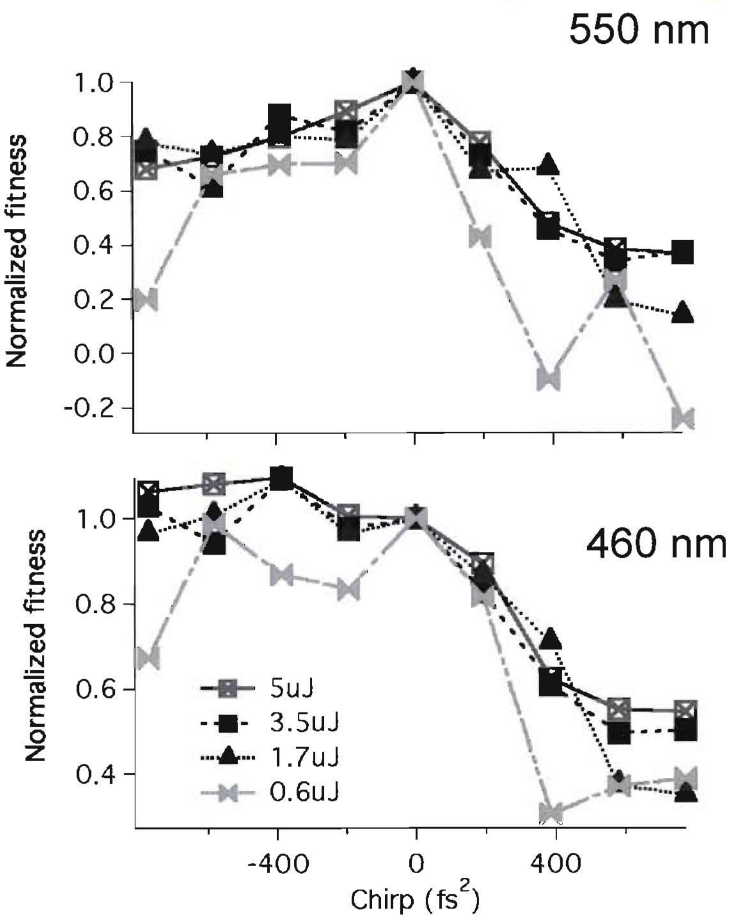
Single parameter control

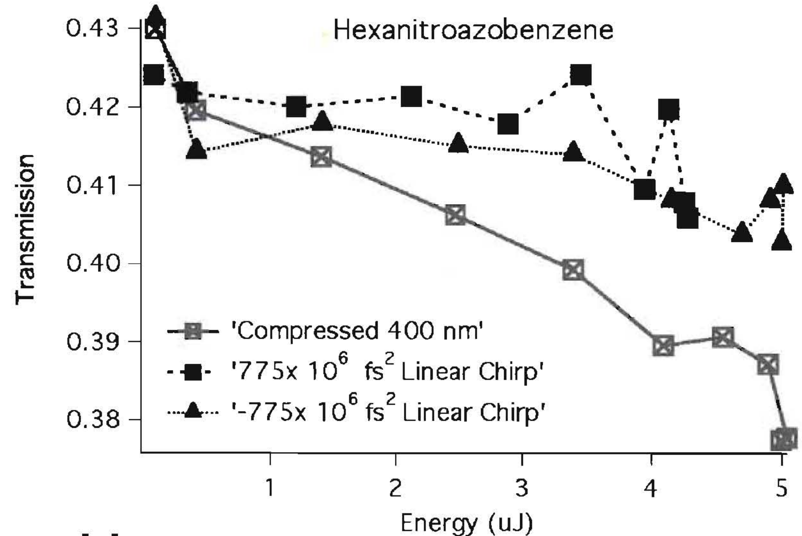
- **Linear chirp**
  - Linear relationship of laser frequency with time

- **Energy relationship**
  - % transmission of 400 nm with energy



Single parameter control





$$\text{Norm Fit} = \frac{\text{chirped fitness}}{\text{compressed fitness}}$$

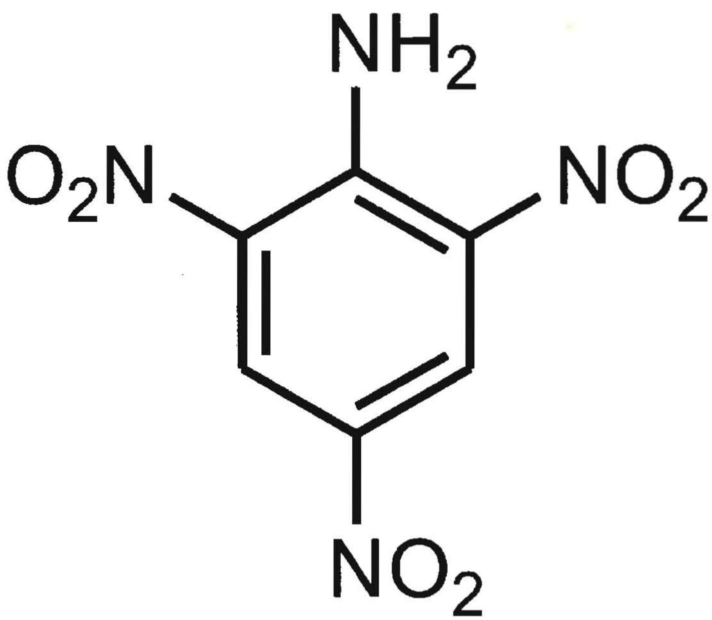
Linear chirp

- 550 nm
  - Symmetric about compressed pulse
  - Intensity control
- 460 nm
  - Asymmetric about compressed pulse
  - Possible complex control

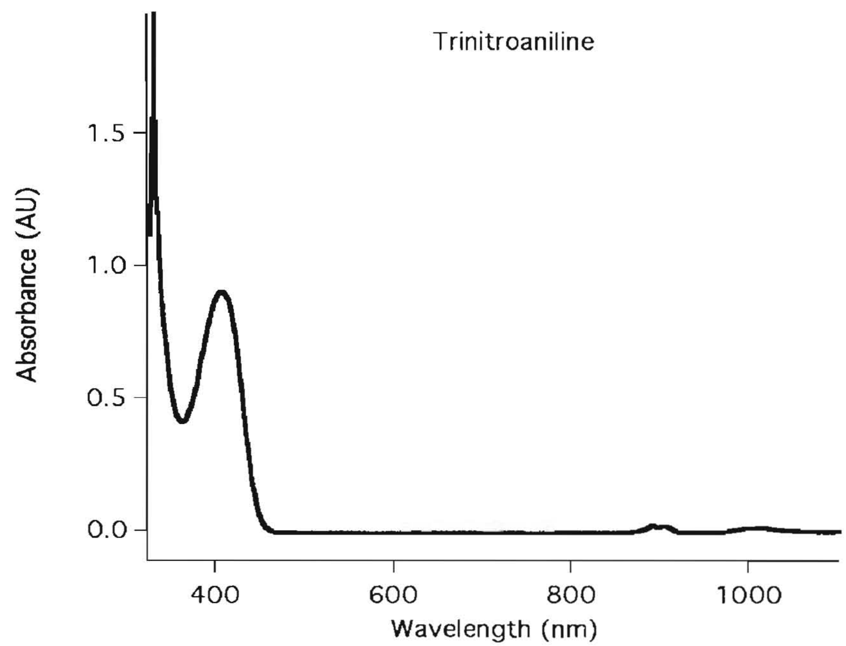
Energy-

- Decreased % transmission with increased energy

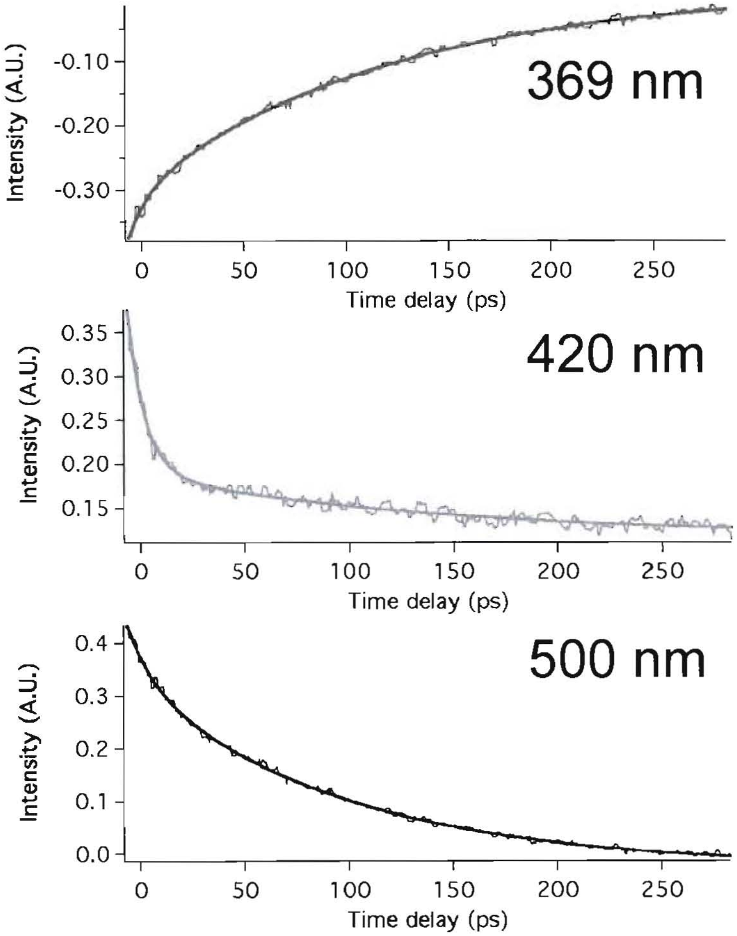
Trinitroaniline (TNA)

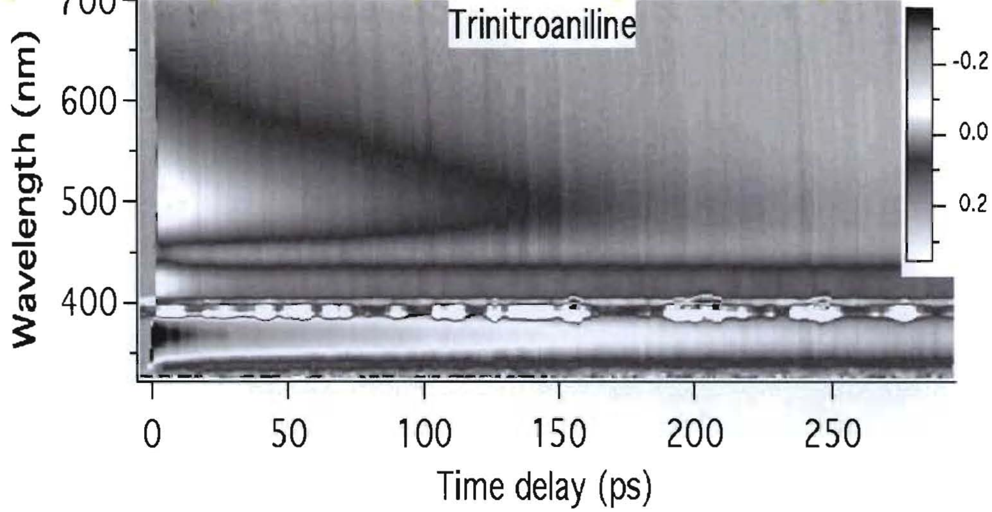


- Also known as picramide
- Absorbs well around 400 nm



TNA 0.3 mM solution in Acetone





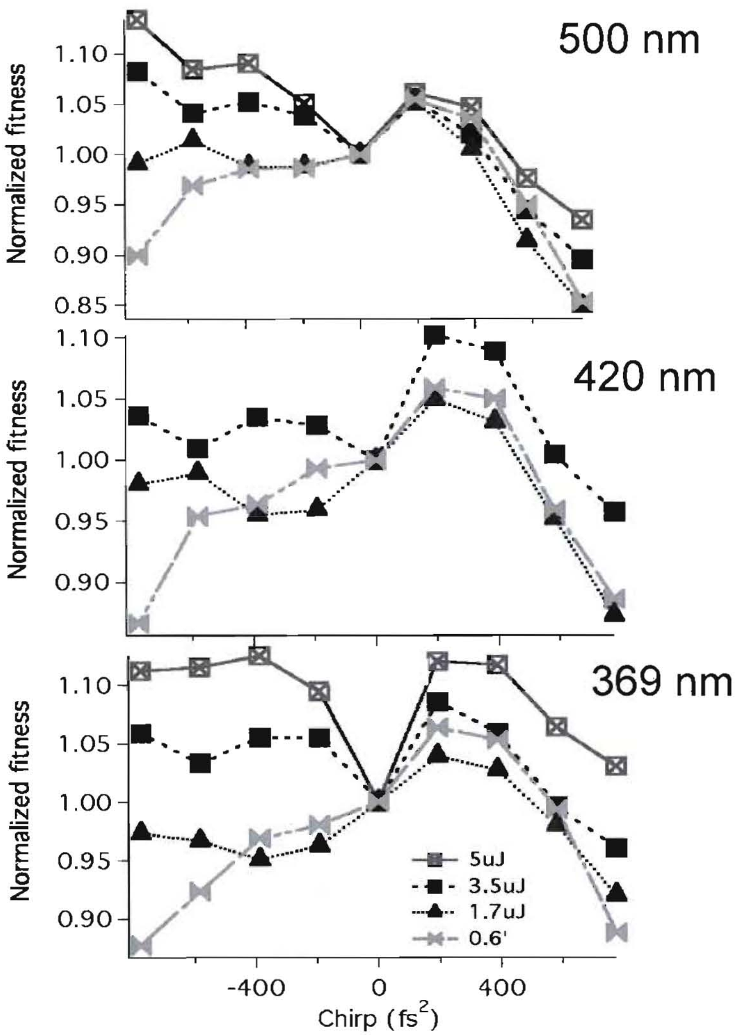
- Biexponential fits
  - **369 nm** (absorption)

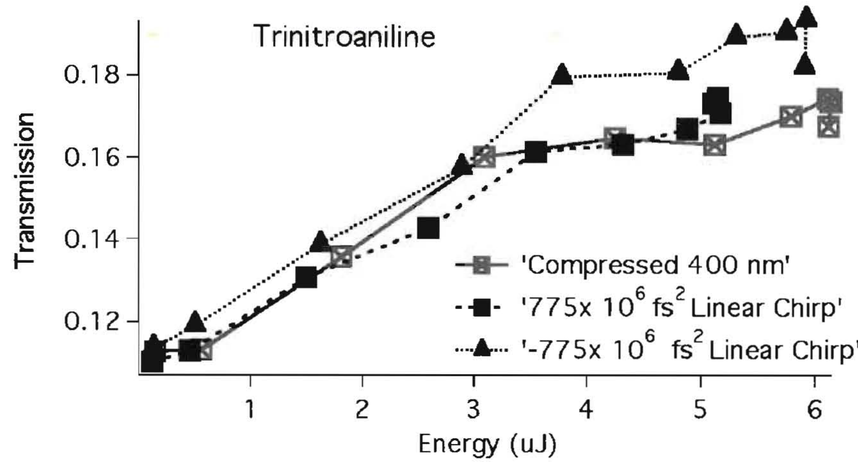
    τ_1=11ps τ_2=126ps
  - **420 nm** (ground state bleach/ stimulated emission)

    τ_1=9ps τ_2=141ps
  - **500 nm** (stimulated emission)

    τ_1=12ps τ_2=102ps

Single parameter control





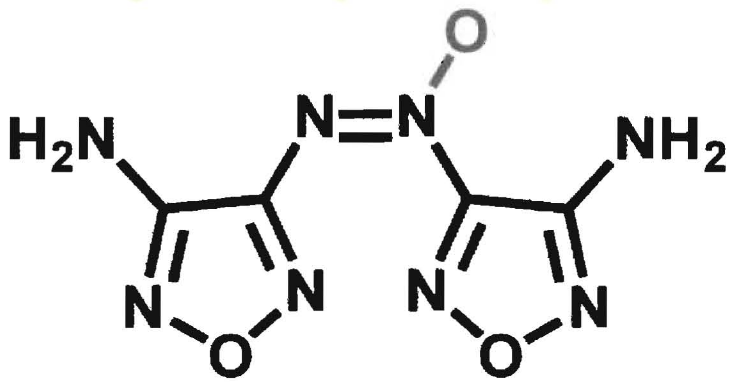
Linear chirp

- 550, 420, 369 nm
  - Positively chirped pulses
    - Intensity decreases with chirp
  - Negatively chirped pulses
    - Energy dependence
    - Increased energy—increased signal

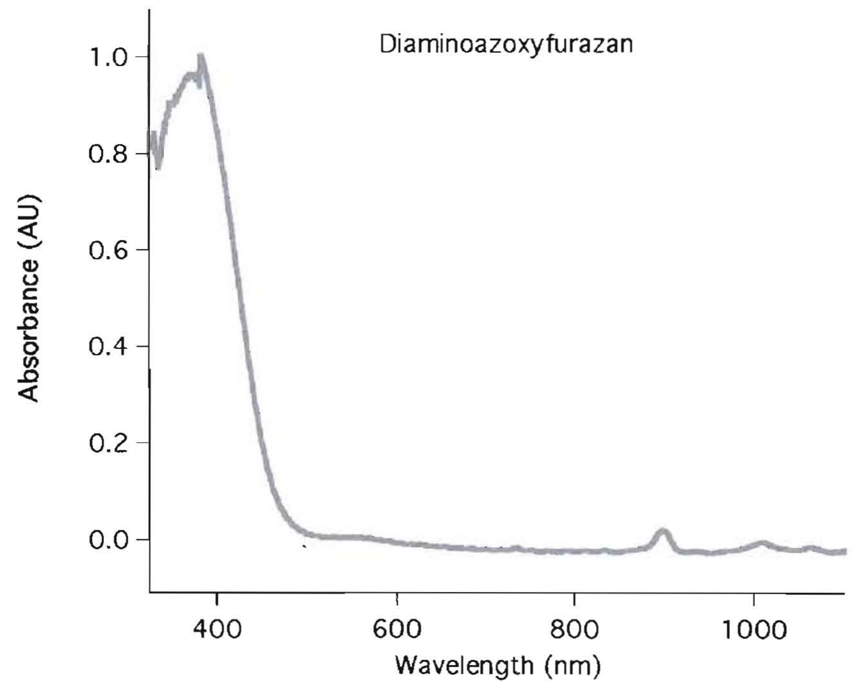
Energy

- Decreased % transmission with decreased energy

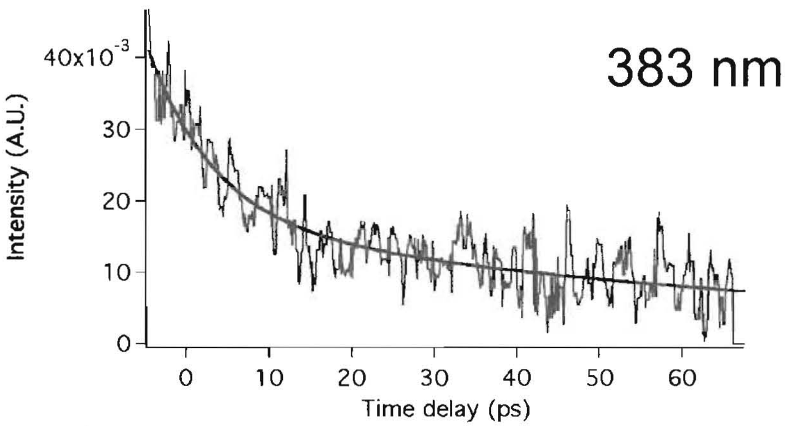
Diaminoazoxyfurazan (DAAF)

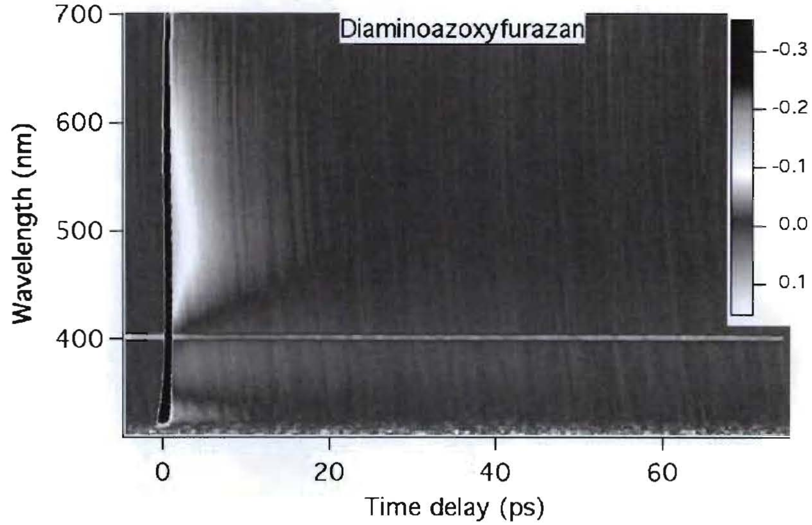


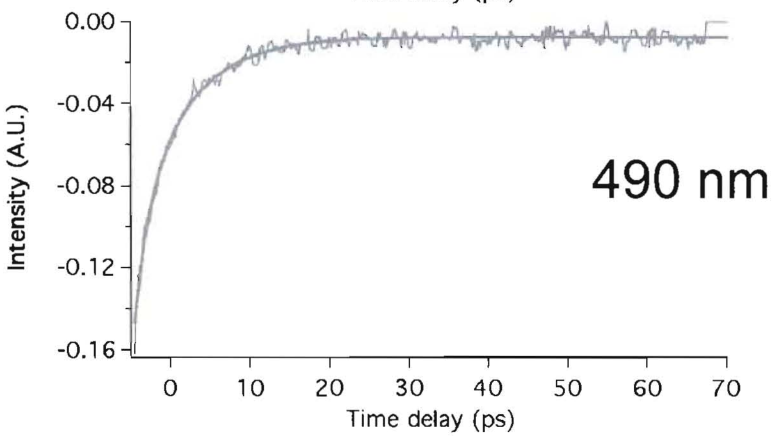
- Insensitive explosive
- Produced in "green" synthesis
- Absorbs well around 400 nm



DAAF 0.44 mM solution in Dimethylsulfoxide





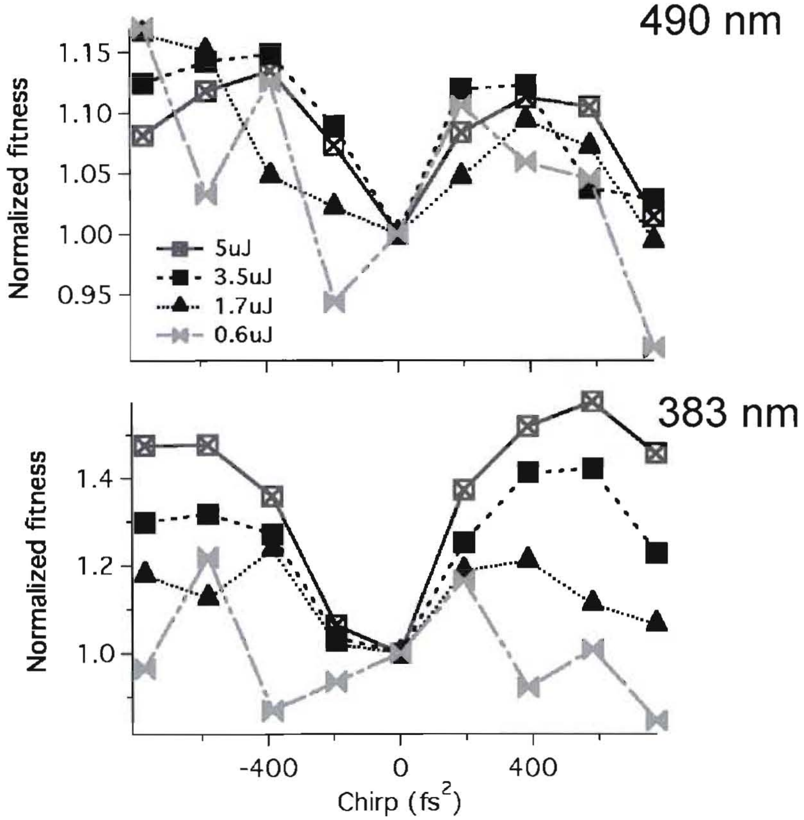


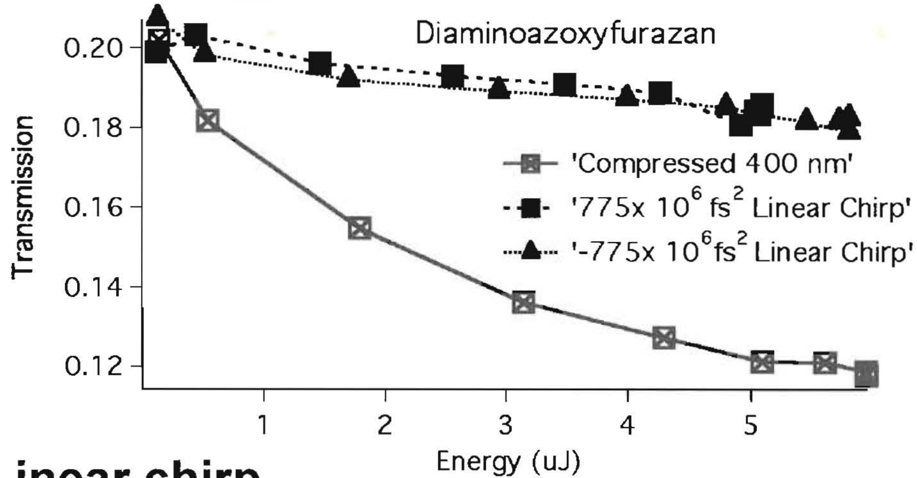
- Biexponential fits
  - **383 nm**

    τ_1=8ps τ_2=83ps
  - **490 nm**

    τ_1=0.3ps τ_2=6ps

Single parameter control





Linear chirp

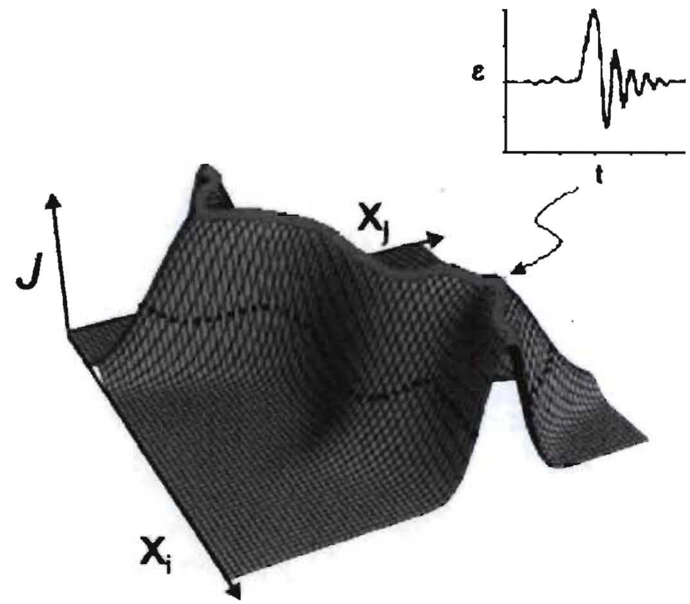
- 490, 383nm
  - Inflection point
    - Population of ≥ 3 excited states
- 383 nm
  - Intensity controlled
    - Higher energy → more chirp control

Energy

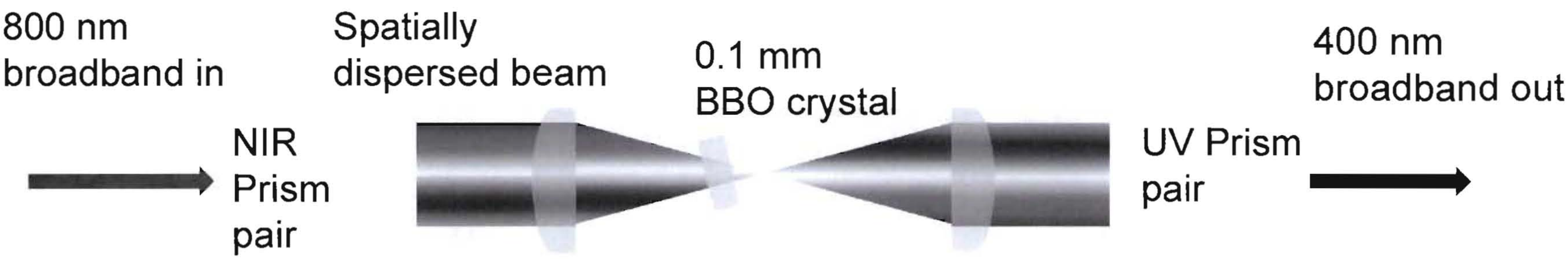
- Increased % transmission with decreased energy
- Greatest effect seen on compressed pulse

Optimal Control Landscape

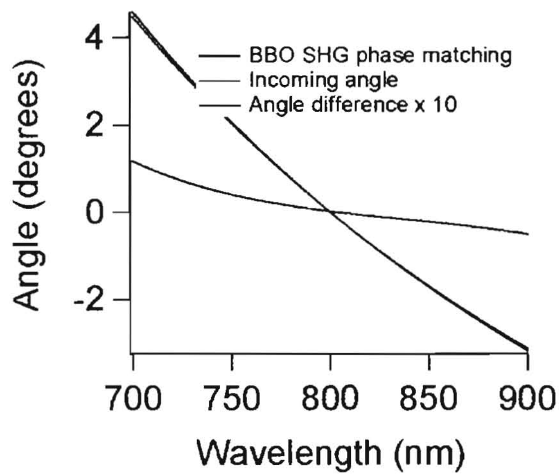
- Control landscape has no traps
  - Given a controllable quantum system, there is always a trap free pathway to the top of the control landscape from any location.
    - **Bandwidth dependent**
      - Unlimited bandwidth
      - Unlimited control
    - **Experimental limitations**



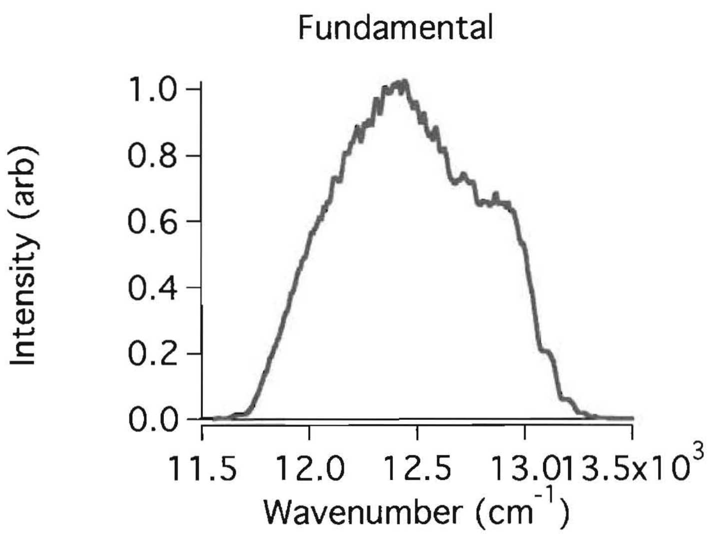
Achromatic second harmonic generation (ASHG) will allow vibrational control with electronic resonance

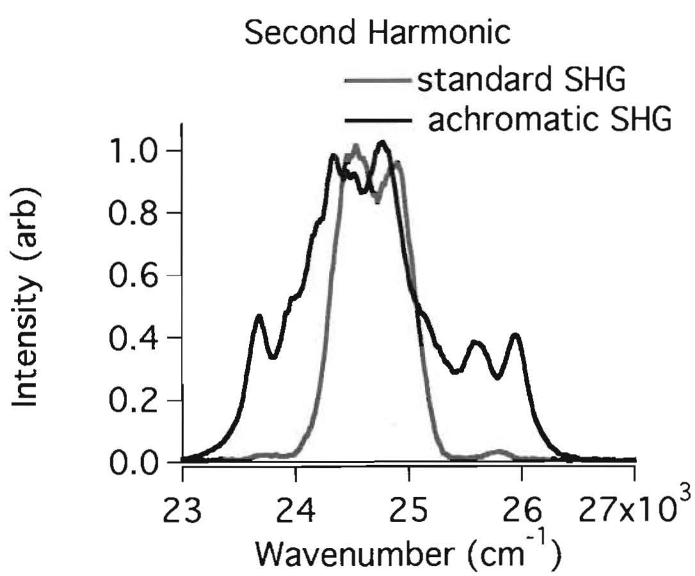


Focus angle = phase matching angle



ASHG doubles the frequency and frequency range





Not only the center frequency, but also the frequency range is doubled.

This allows impulsive Raman excitation and control over a larger range of vibrational frequencies.

Efficiency of ASHG is high (~40%)

ASHG is insensitive to spectral phase noise (pulse is not compressed)

Summary

- **HNAB, TNA, DAAF**
  - **Transient absorption data**
  - **Single parameter control**
- Future
  - Complex control
    - **Achromatic phase matching**
    - Of non simple parameter response in HNAB and TNA
    - DAAF- further investigate degree of intensity control
    - Trinitrotoulene (TNT)