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Title:	Towards mitigating explosive threats using quantum controlled initiation
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Towards mitigating explosive threats using quantum controlled initiation

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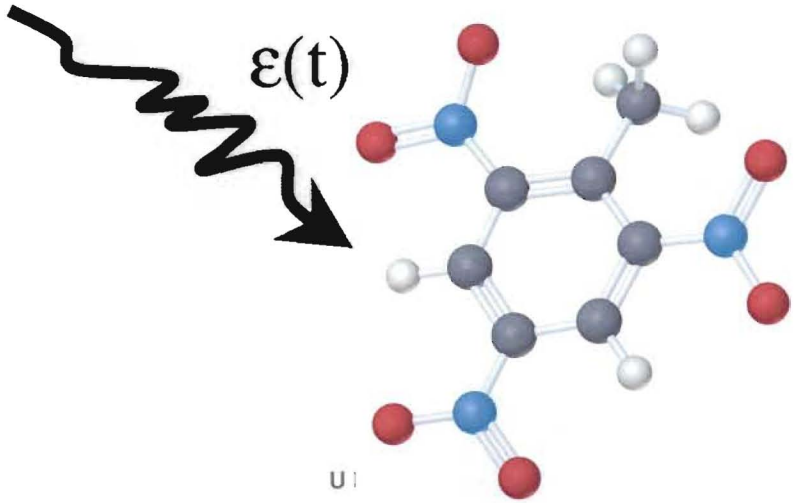


Towards mitigating explosives threats using quantum controlled initiation

Quantum control of localized energy deposition into an energetic material is being investigated as a method to allow controlled initiation and propagation of reaction without transition to detonation. Quantum controlled initiation (QCI) of explosives utilize time dependent phase shaped ultraviolet (UV) electric fields to drive the energetic chemical systems towards reaction. QCI searches for an optimally shaped ultrafast laser pulse that will guide energy flow along a desired path. QCI can be exploited as a stand-off mitigation technology that strives to reduce the impact of explosive blasts on people and property by initiating controlled low order reaction.

Quantum controlled initiation experiments require: 1) optimally shaped light pulses, 2) pulse shaping measurement, 3) feedback control algorithms, and 4) feedback measurement methods. Femtosecond laser pulse shaping in the UV at 400 nm employs a fused silica acousto-optic modulator (AOM) pulse shaper that consists of a 4-f zero dispersion compressor. Transient absorption spectroscopy is used to measure the pulse shaper 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. The 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 to obtain fully optimized complex control.

What is the threat?

Mitigate or detect trace or bulk amounts of explosives at **stand-off distances**

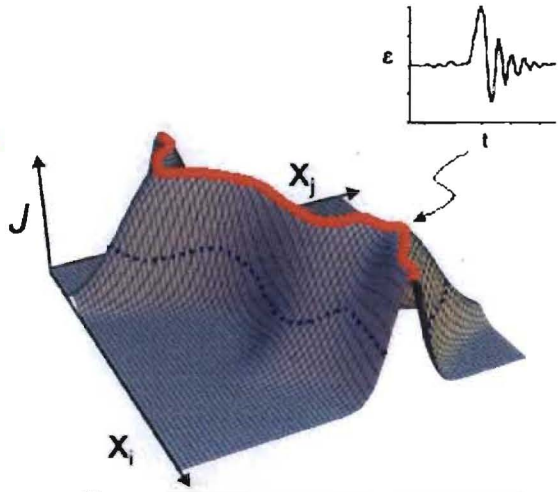
- Line of sight (laser)
  - Bulk (initiation)
  - Trace surface amounts
    - Outsides of casings, car doors, fingers
- Non-line of sight (acoustic)
  - Bulk
    - Stimulate signatures- thermal, vapor, RF
      - Buried, well sealed (clean)

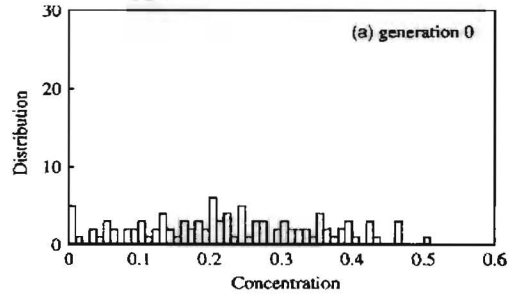
Problem: Cluttered background

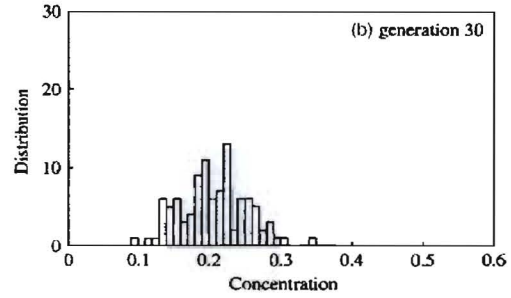
Solution: Optimal control

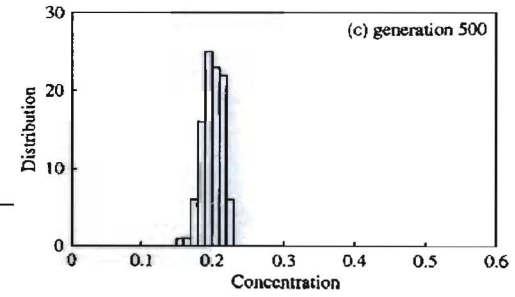
Optimal Control Landscape

- 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.
- Optimization is efficient and fast
  - Tens of generations using genetic algorithms
  - Tens of thousands of experiments per hour
- Optimal control is a smart machine
  - Uses science, engineering, and technology in the most efficient way





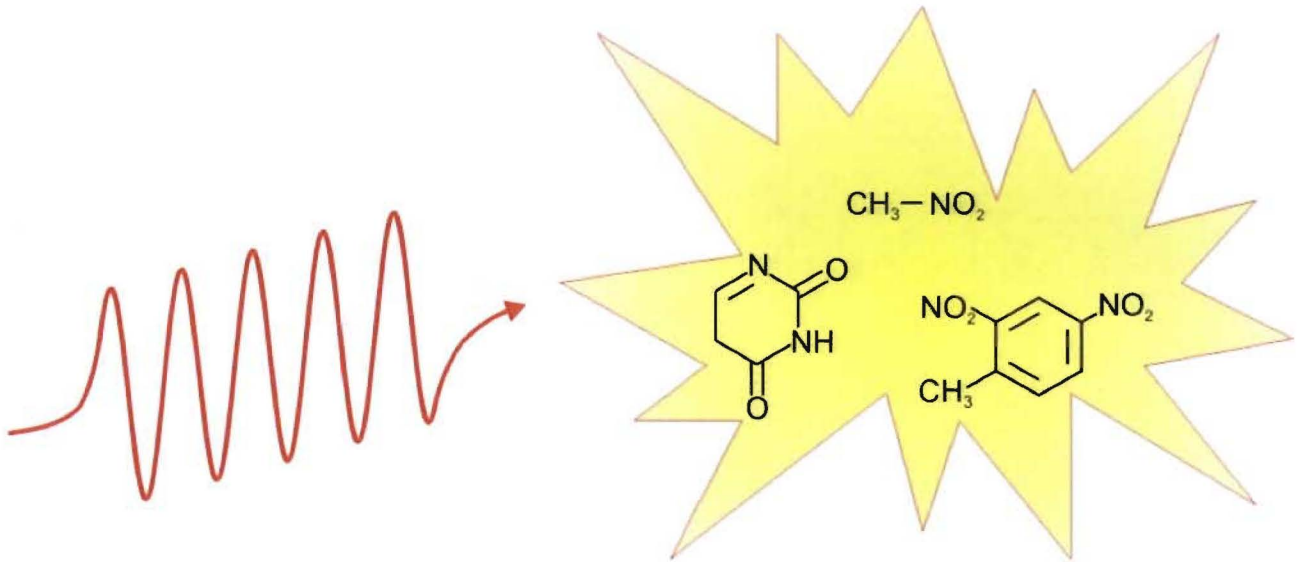




Use optimal control for smarter MITIGATION and detection of explosives

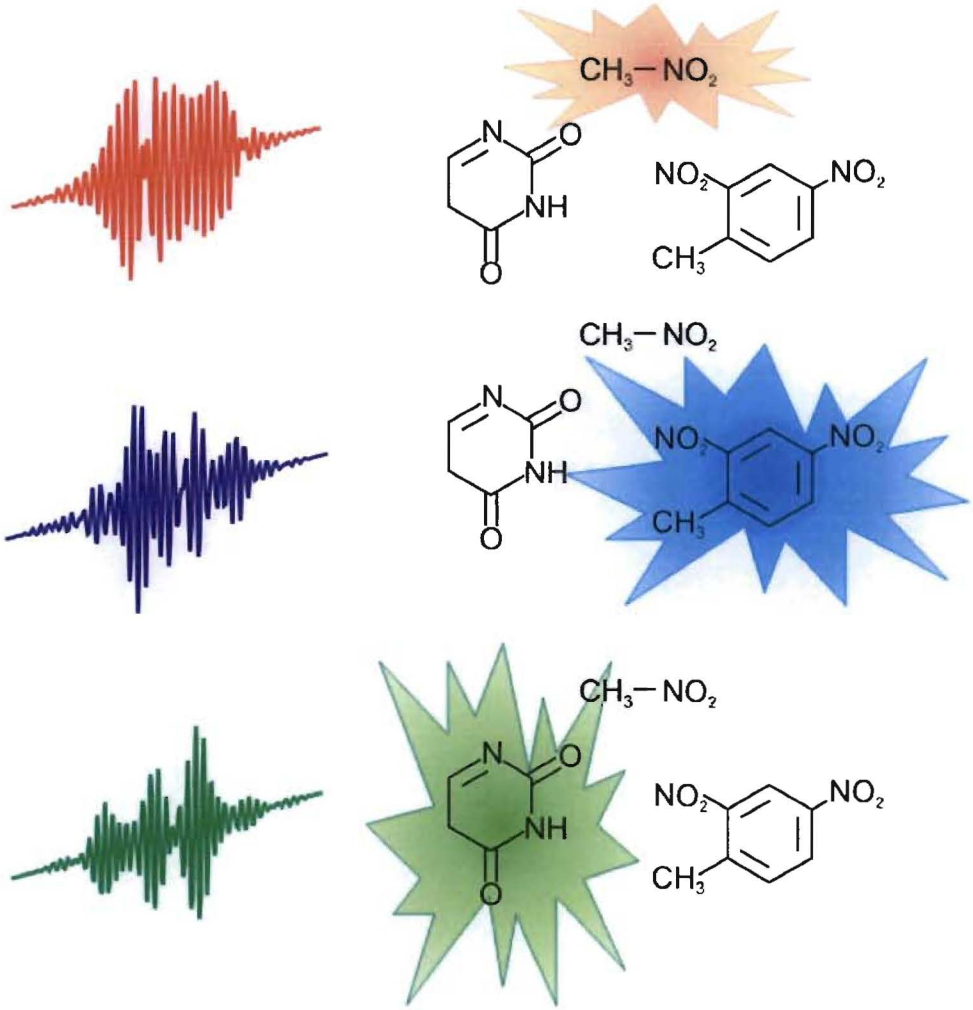
Linear spectroscopy - unshaped pulses

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



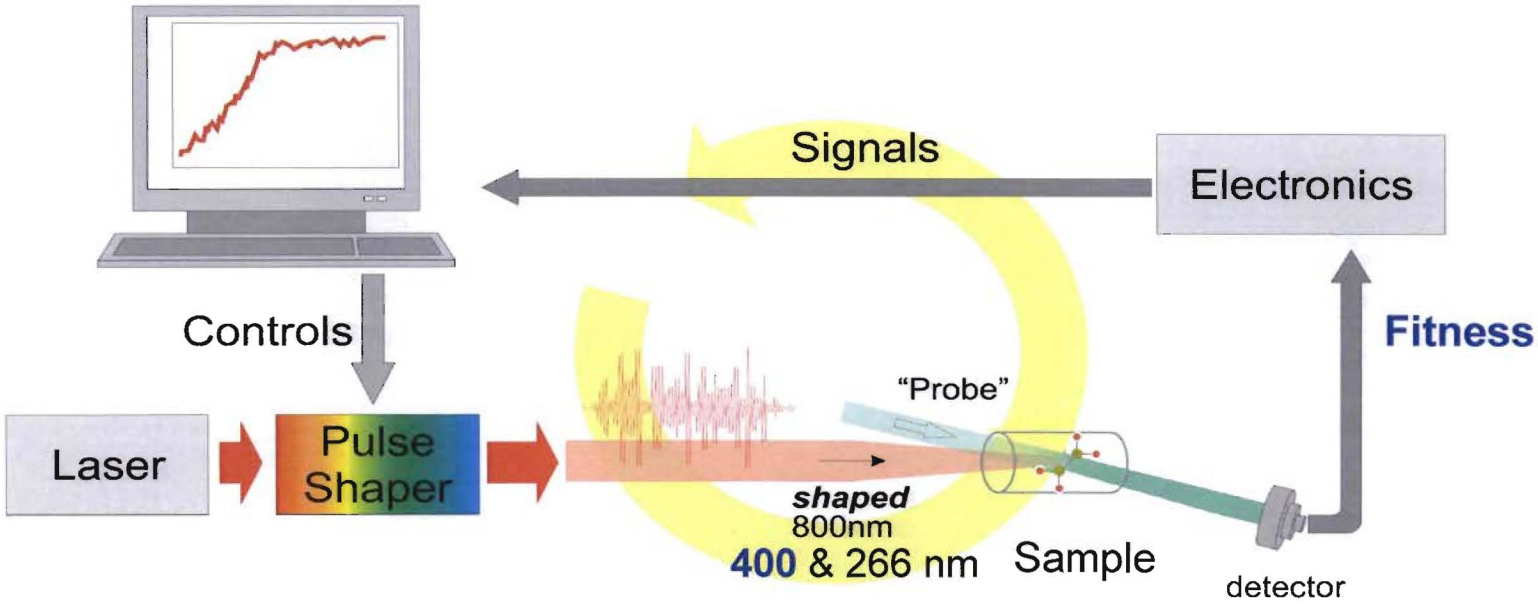
Quantum Optimal Dynamic Discrimination

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



Control by closed loop optimization

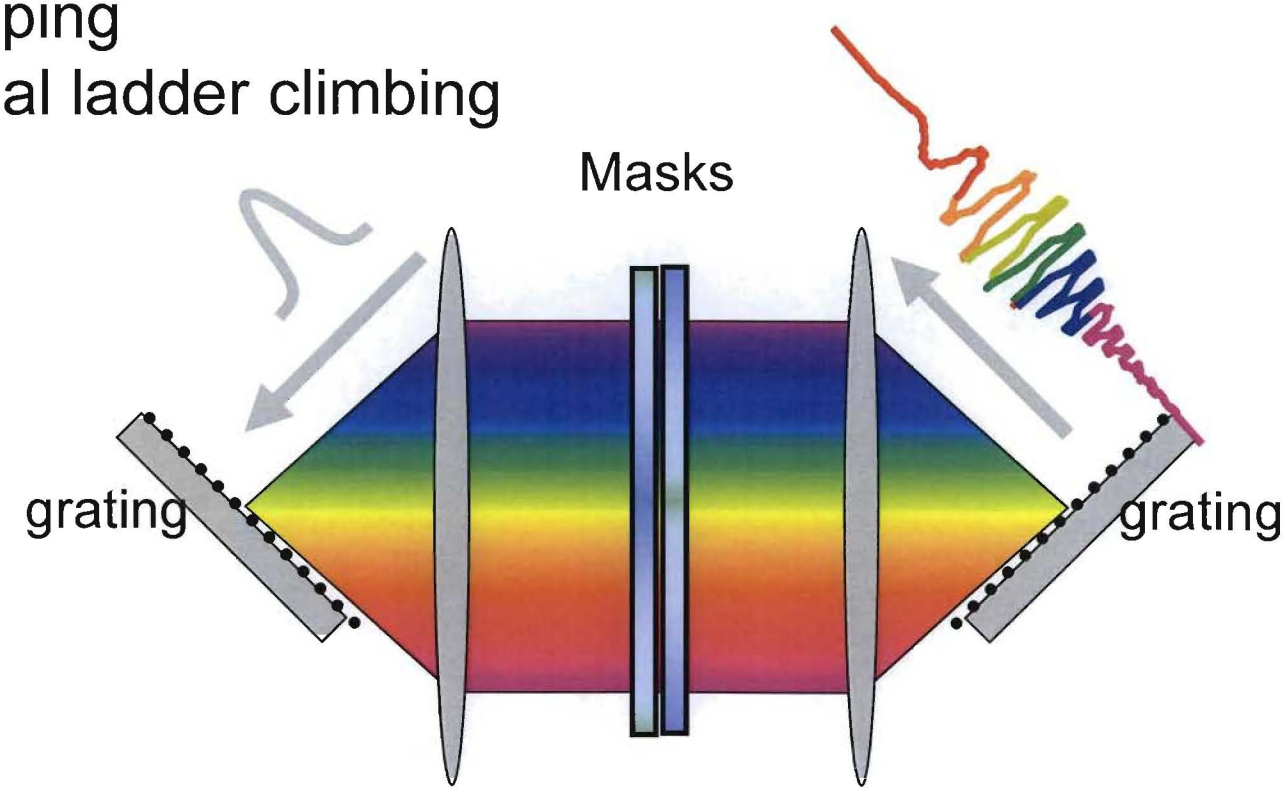
- Learning Loop
  - Searches for optimal pulse using feedback control
    - Initiation experiment: Transient absorption spectrum
  - Can use many optimization routines



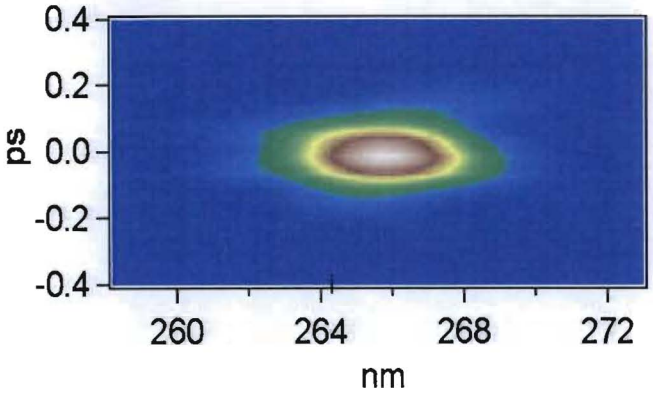
- Shaped 400 nm pump
- Broadband 325-750 nm super continuum probe for transient absorption

Cutting edge pulse shaping

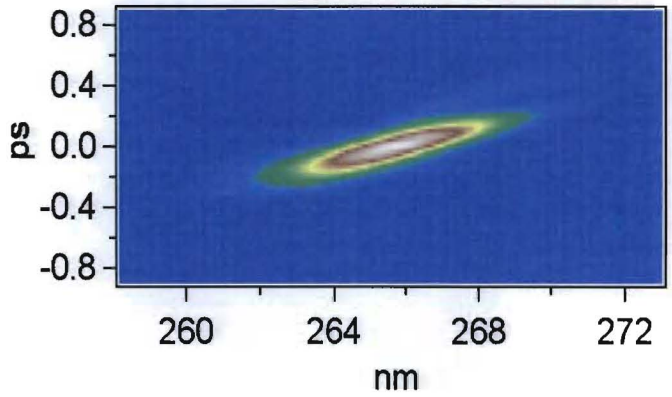
- 400 and 266nm ultraviolet (UV) shaping
  - Allows for single photon processes
  - In the low field limit
- 800 nm shaping
  - Vibrational ladder climbing



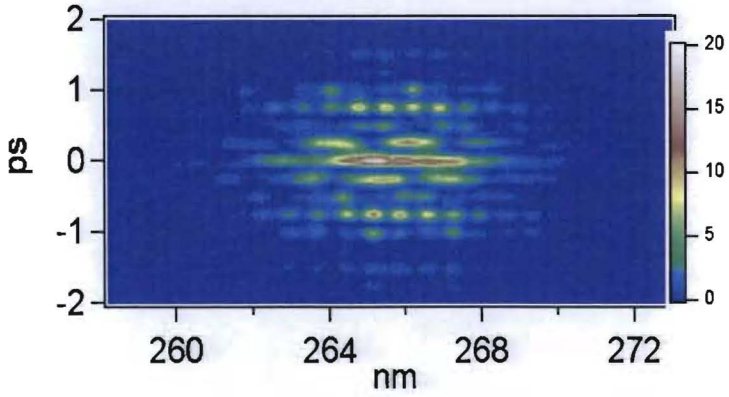
Examples of shaped pulses



Transform limited ~150fs



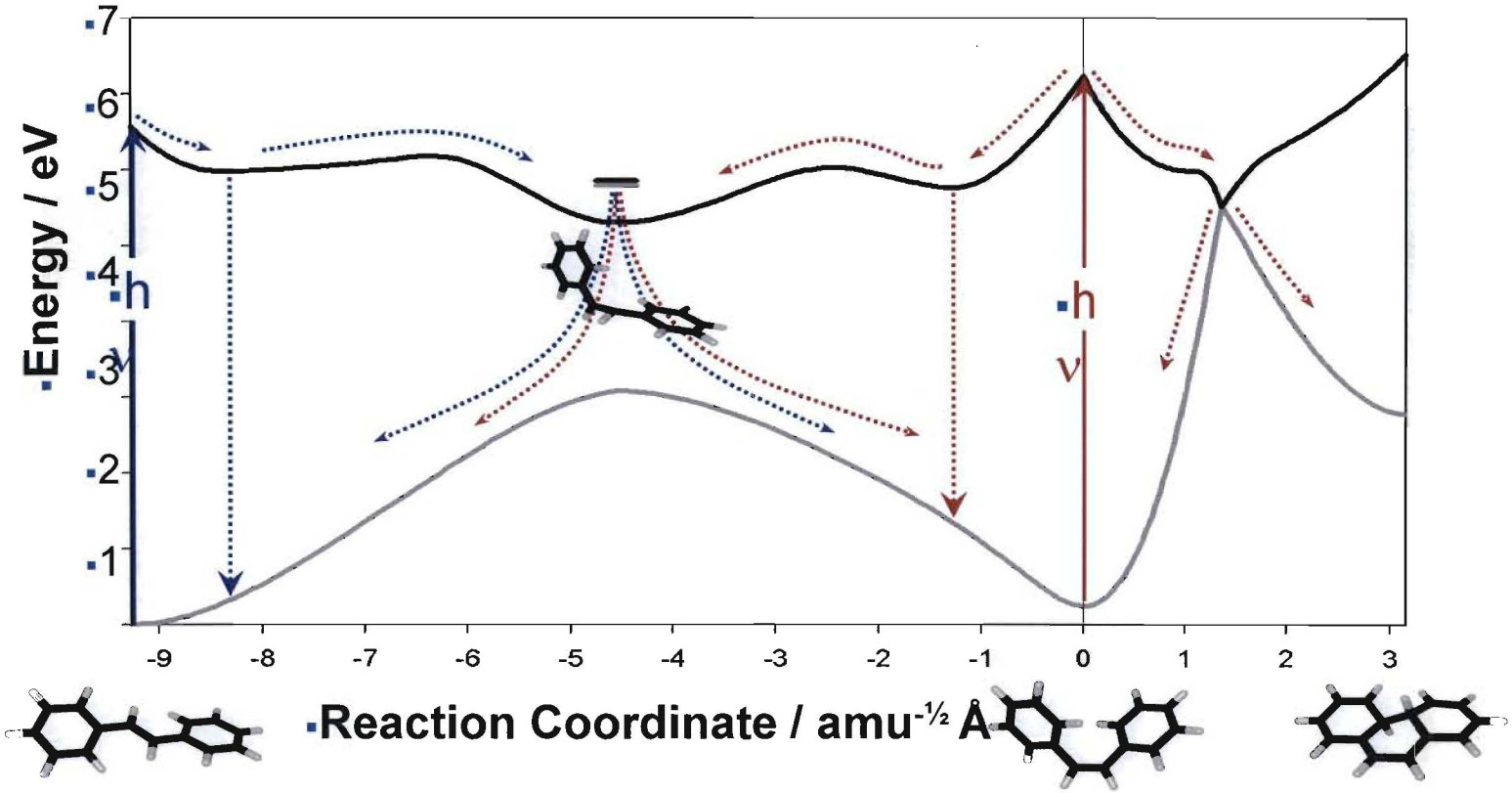
Simple linear chirp



Dual sine waves

Towards optimal controlled initiation

Experimental test bed: Cis-stilbene

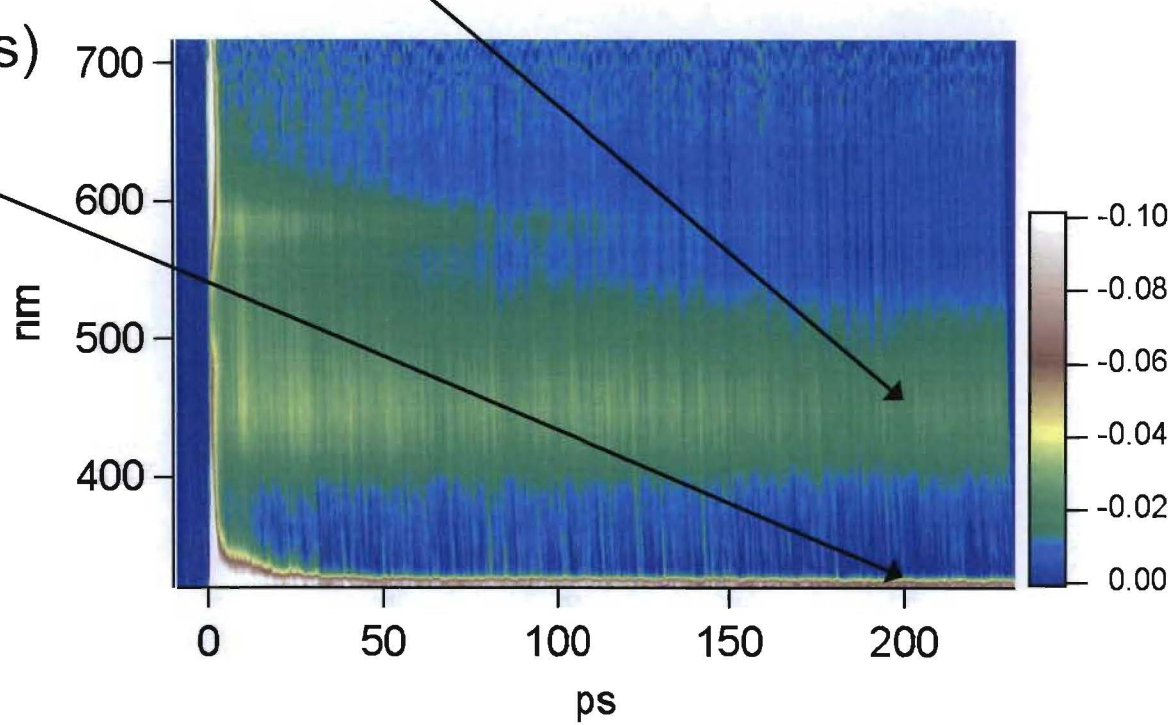


Optimize *trans*-stilbene or DHP product

- **Feedback information**
  - Obtained from transient absorption spectrum of cis-stilbene

- Cyclization (cis→DHP 4a,4b-Dihydrophenanthrene) 400-500 nm

- Isomerization (cis→trans)
  - 320-330 nm

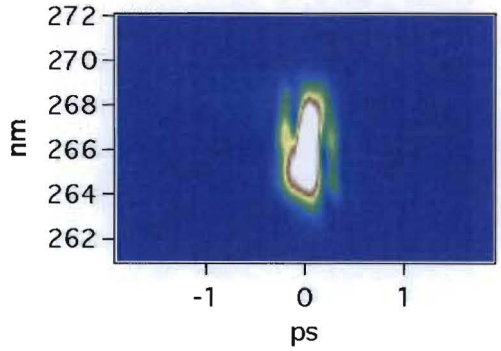


Optimize trans-stilbene product

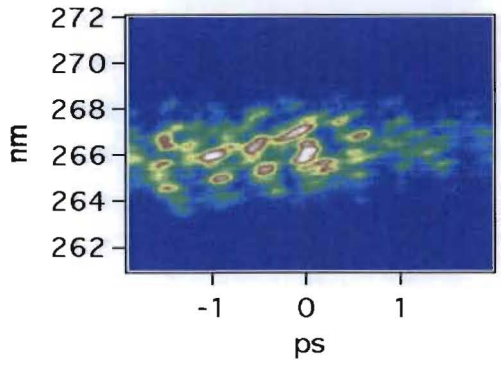
Achieved 2.08x increase above normal yield

Compressed pulse:

normal yield (1x)

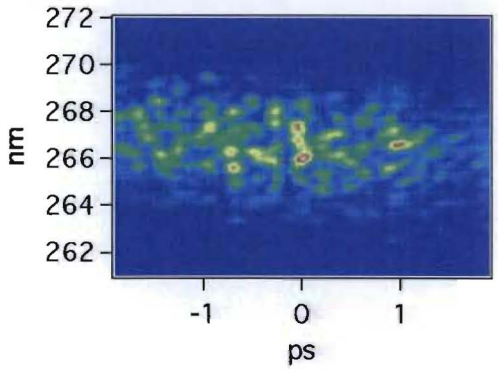


- Pulse shapes that achieved higher trans-stilbene product



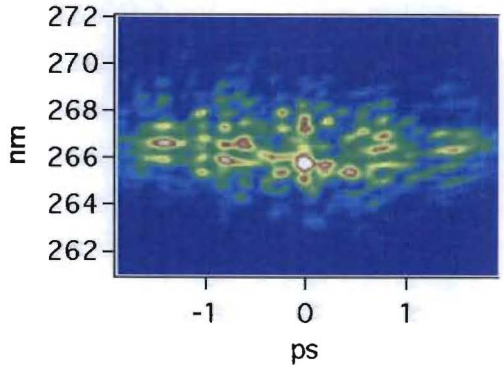
Downhill simplex

2.08 x



Differential Evolution

1.91 x

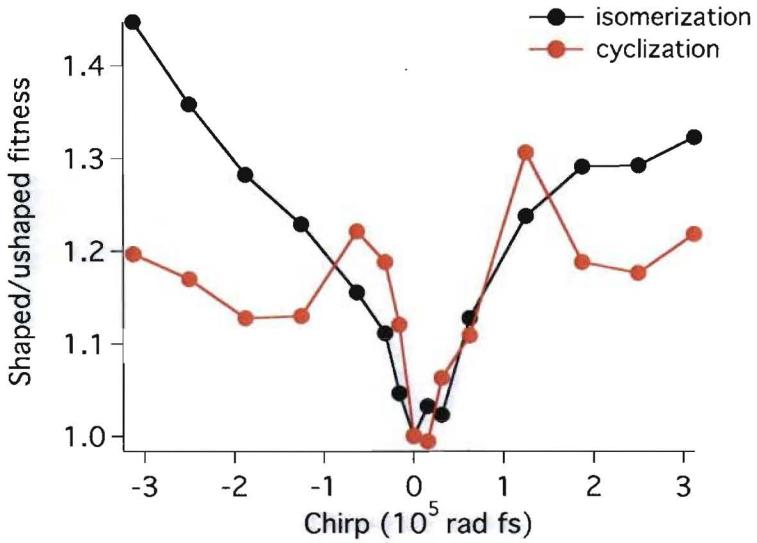


Genetic Algorithm

1.78 x

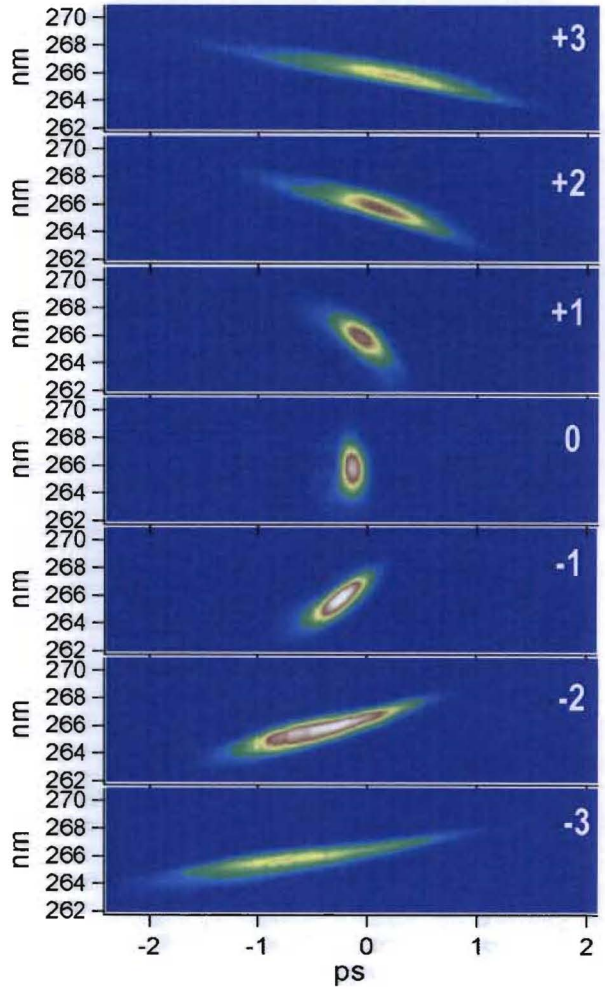
Single parameter control: linear chirp

Can we increase yield with simpler pulses, such as linear chirp?



YES!

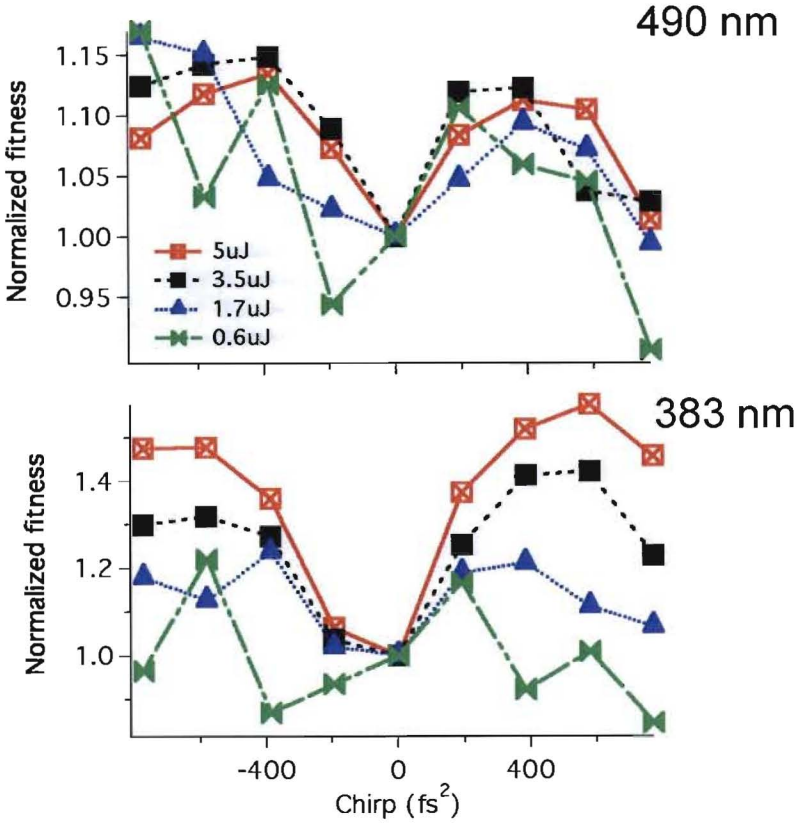
- Optimization at high energy recovers signal yield
- High intensity accessing multiphoton transitions to higher excited states

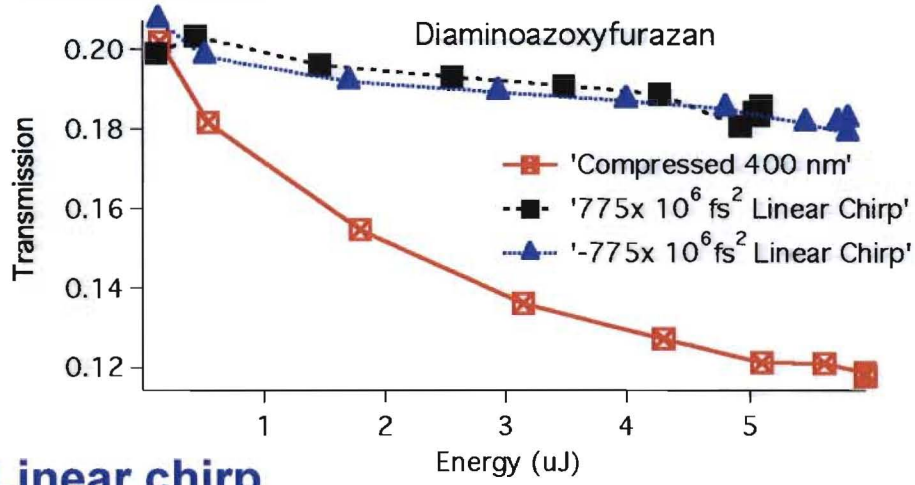


Optically controlled initiation for mitigation

- Control through single parameter
- Bandwidth key for control

Single parameter control





Linear chirp

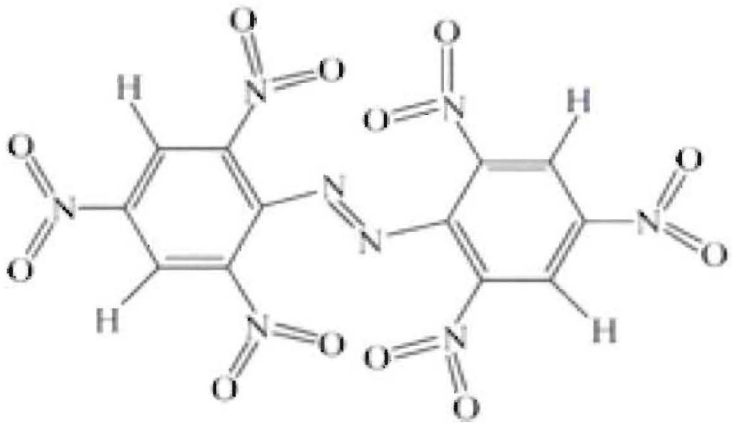
- 490, 383nm
  - Inflection point
    - Population of ≥ 3 excited states
- 383 nm
  - Intensity controlled
    - Higher energy ➔more chirp control

Energy

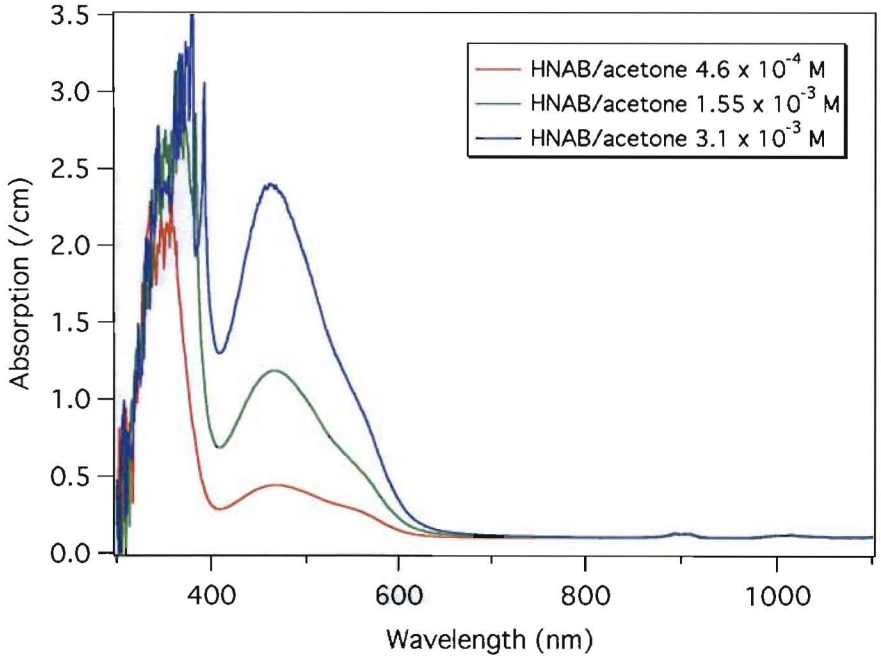
- Increased % transmission with decreased energy
- Greatest effect seen on compressed pulse

Towards optimal control of explosives

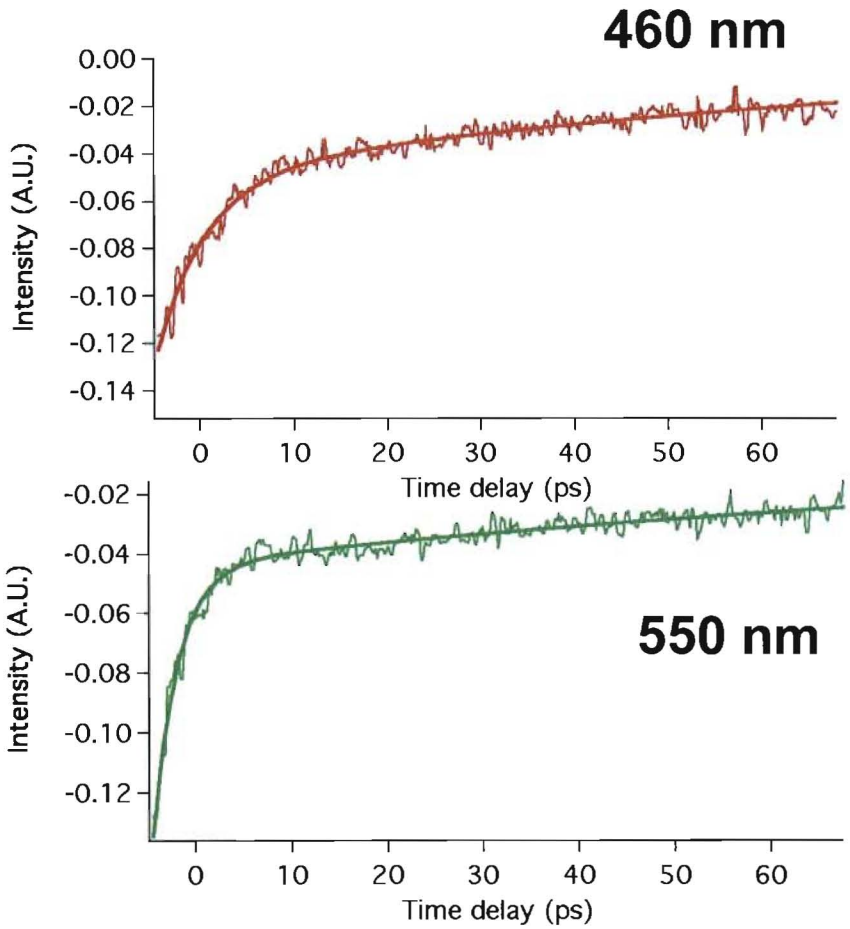
- Potential energy surface unknown
- HNAB absorbs well around 400 nm
- Good starting point for control

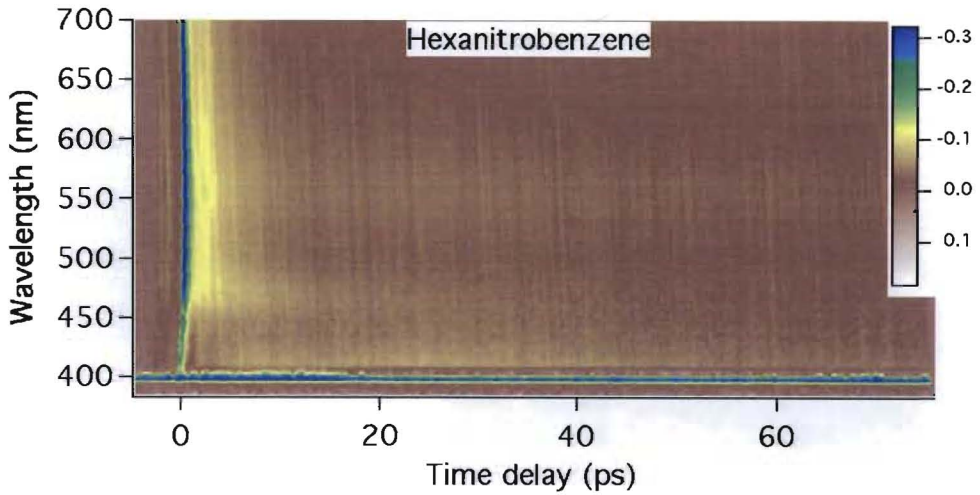


hexanitroazobenzene



HNAB 1.38 mM solution in Acetone





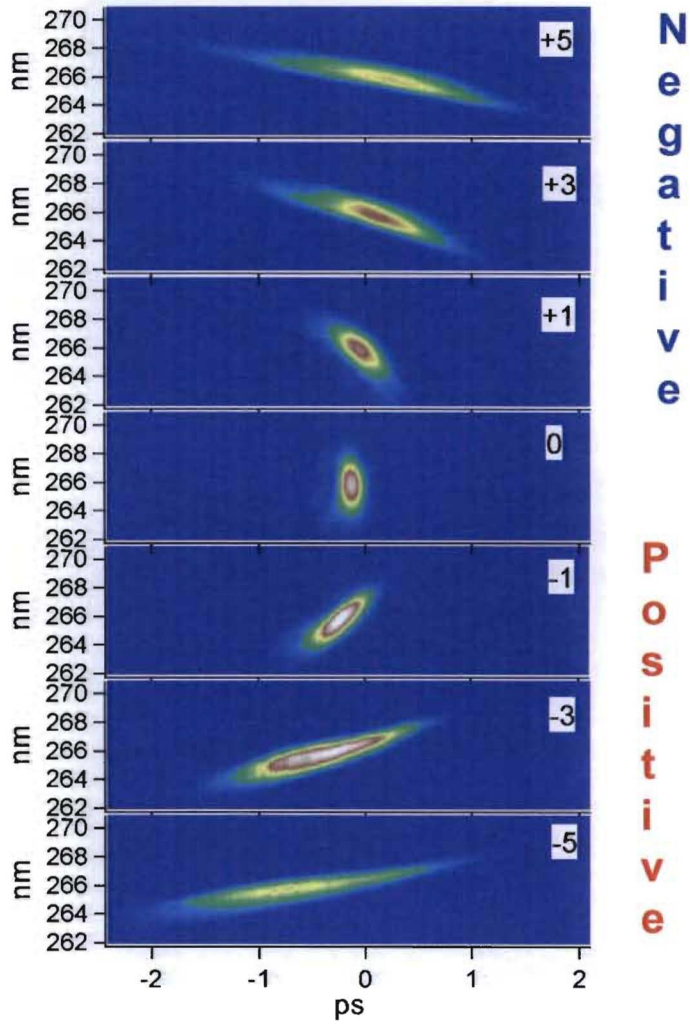
- Biexponential fits
  - **460 nm**

    τ_1=8ps τ_2=130ps
  - **550 nm**

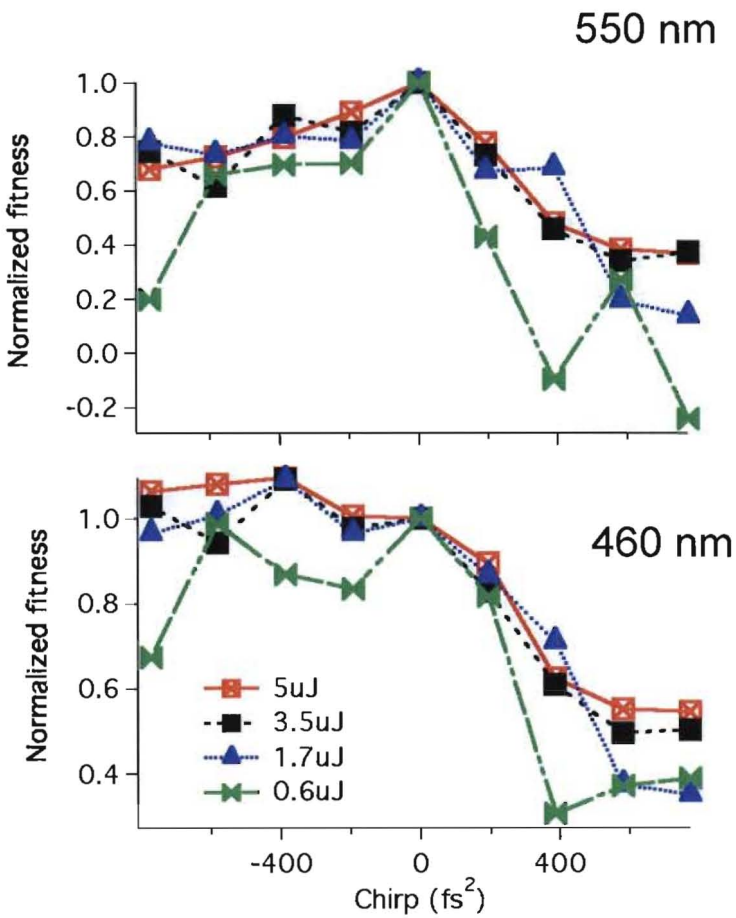
    τ_1=17ps τ_2=223ps

Single parameter control

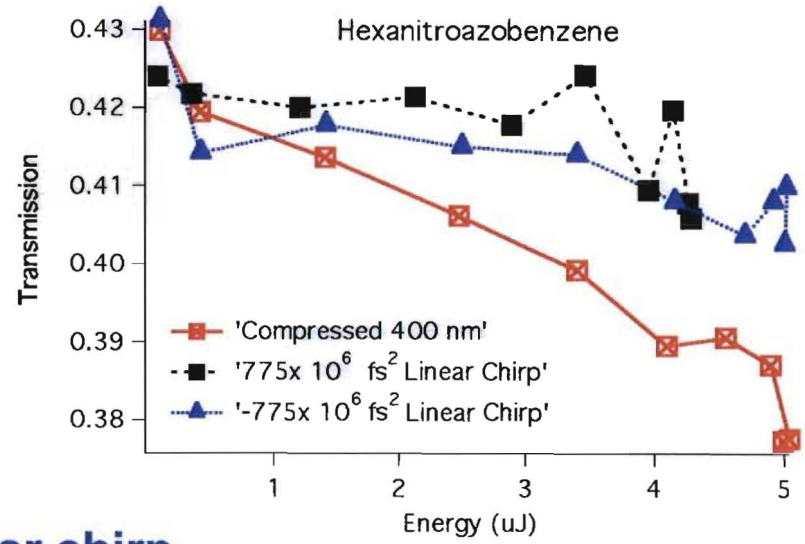
- **Linear chirp**
  - Linear relationship of laser frequency with time
- **Energy relationship**
  - % transmission of 400 nm with energy



Single parameter control



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



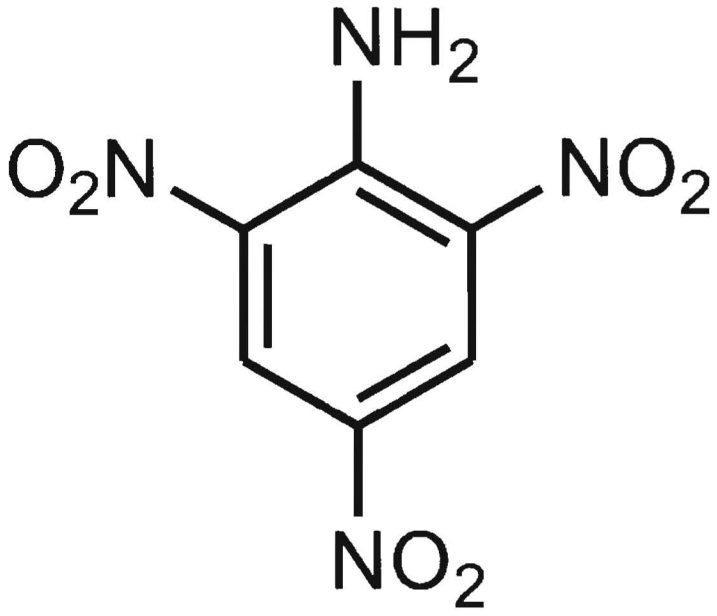
Linear chirp

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

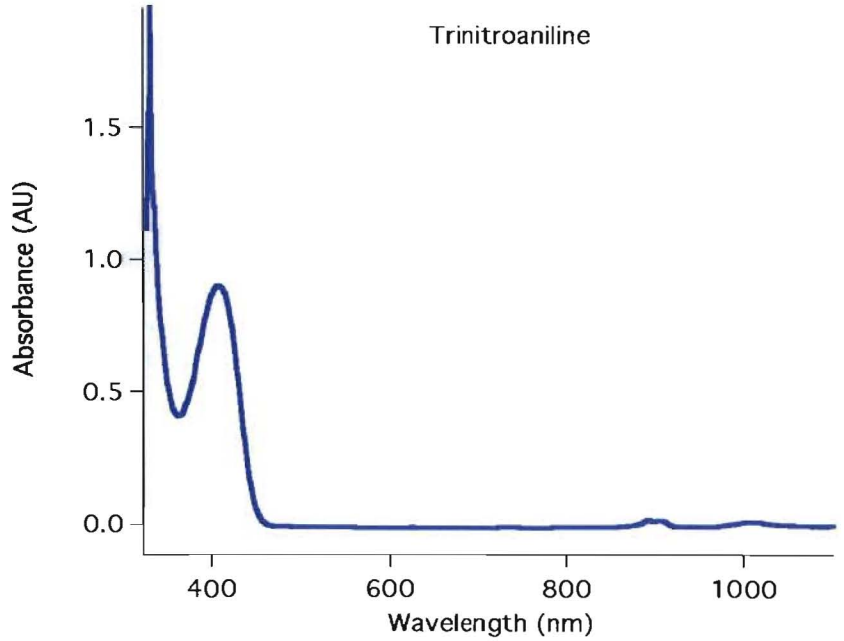
Energy-

- Decreased % transmission with increased energy

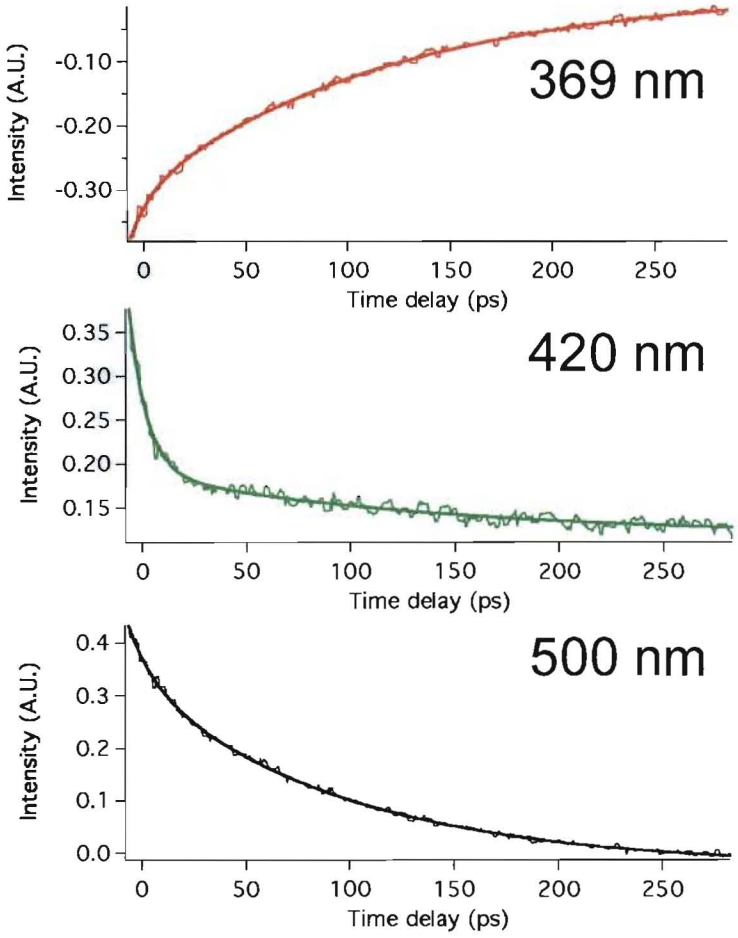
Trinitroaniline (TNA)

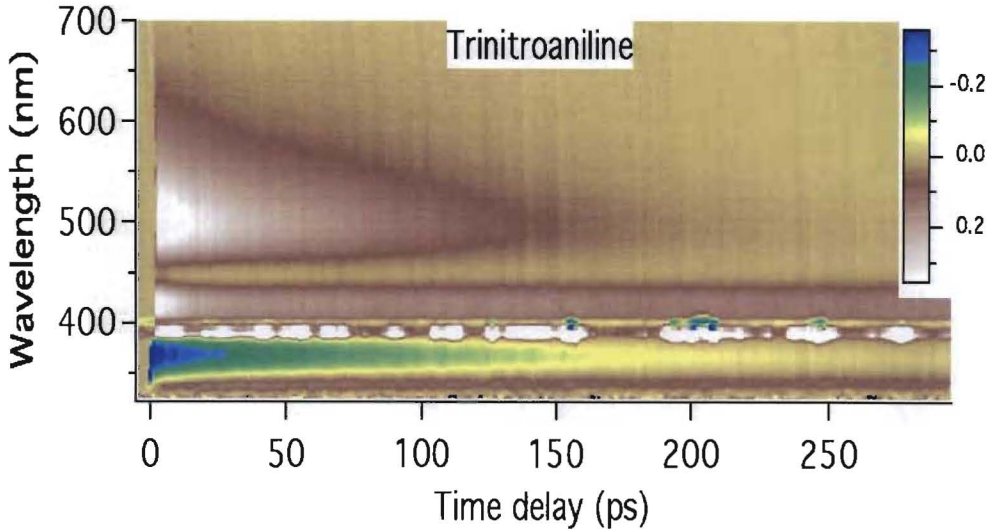


- Also known as picramide
- Absorbs well around 400 nm



TNA 0.3 mM solution in Acetone





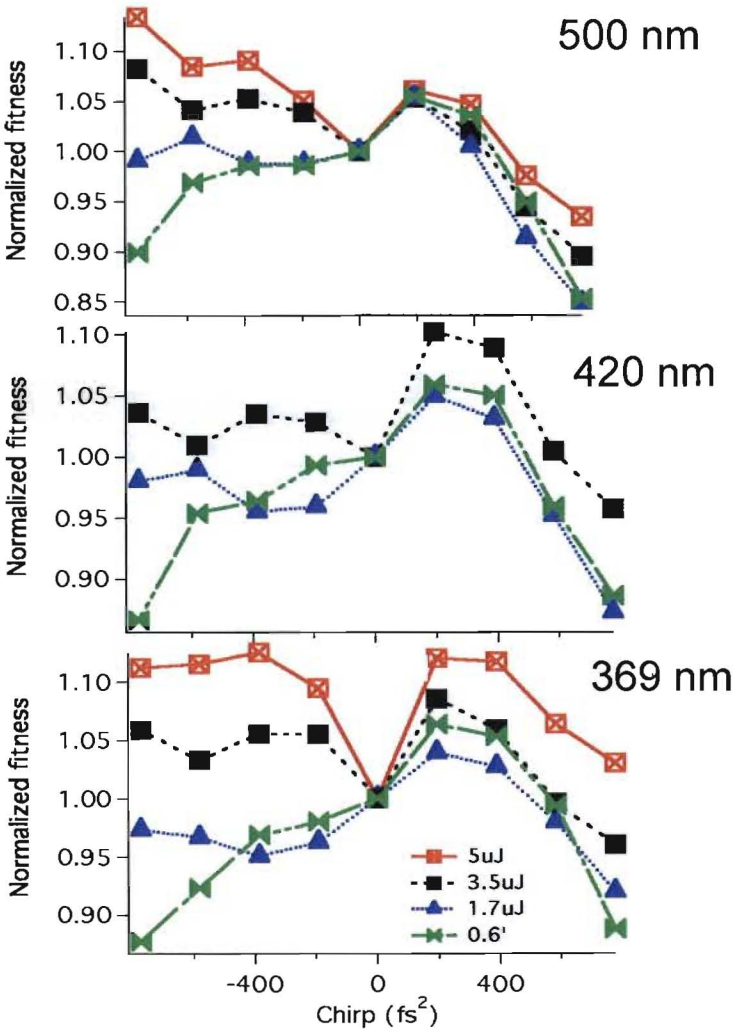
- Biexponential fits
  - **369 nm** (absorption)

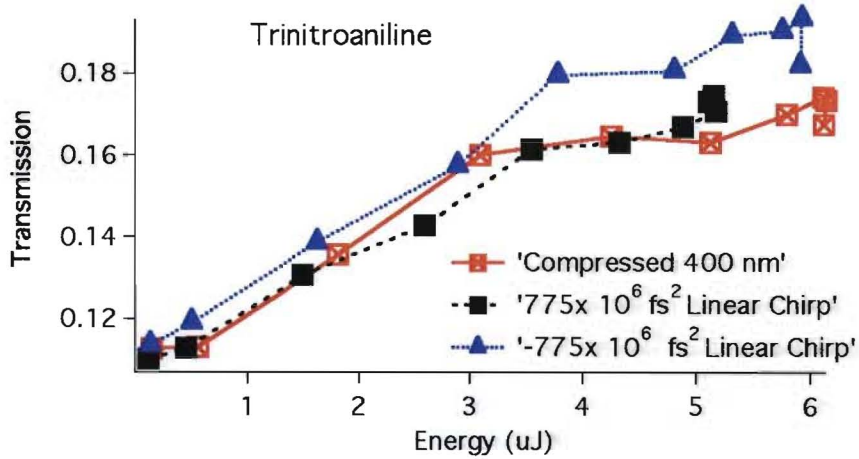
    τ_1=11ps τ_2=126ps
  - **420 nm** (ground state bleach/ stimulated emission)

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

    τ_1=12ps τ_2=102ps

Single parameter control





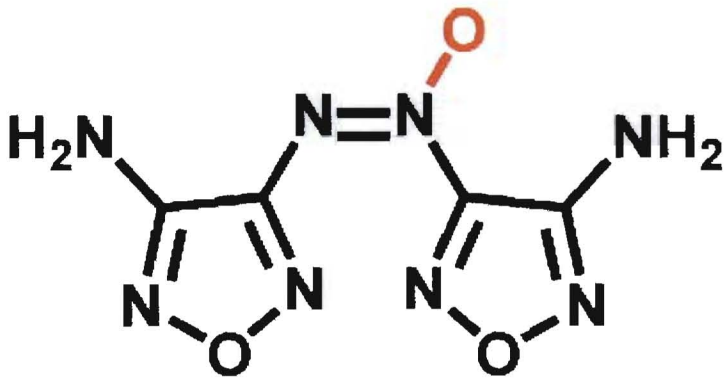
Linear chirp

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

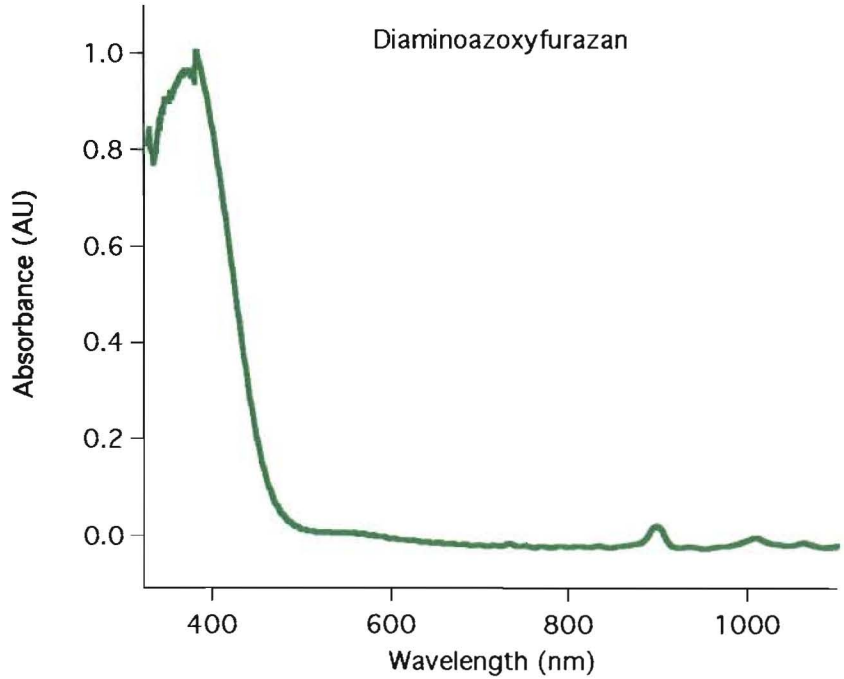
Energy

- Decreased % transmission with decreased energy

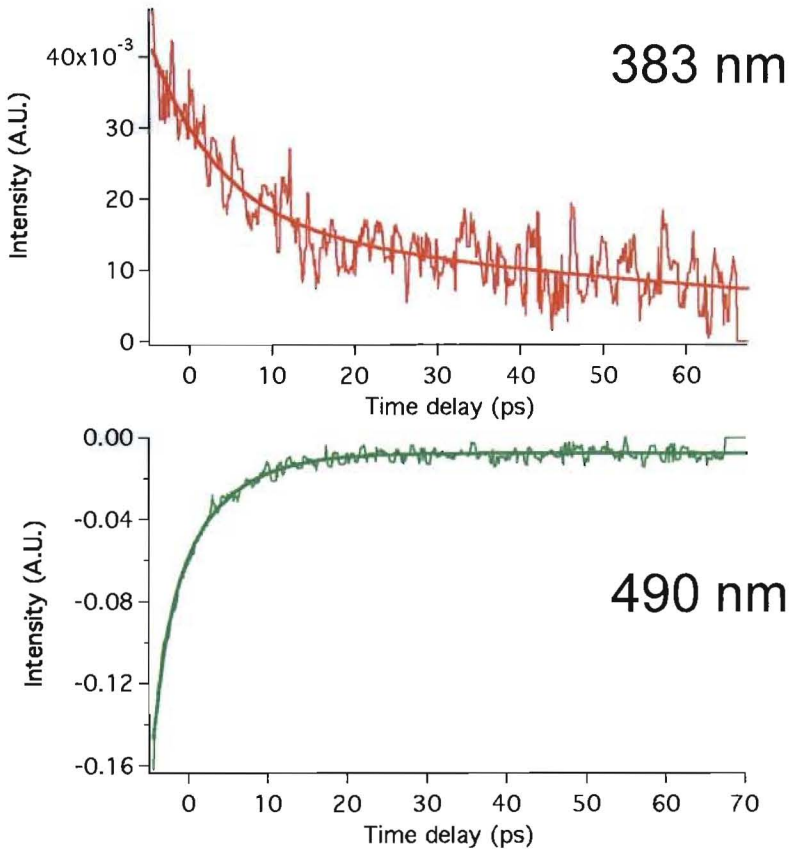
Diaminoazoxyfurazan (DAAF)

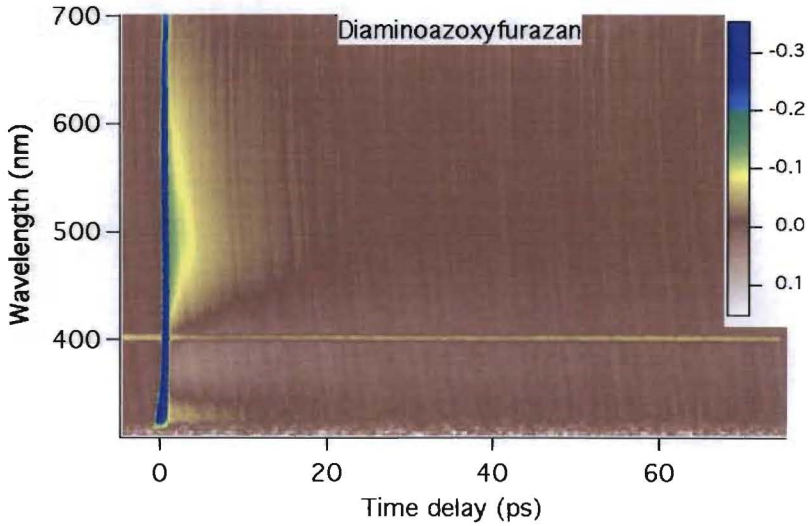


- Insensitive explosive
- Produced in "green" synthesis
- Absorbs well around 400 nm



DAAF 0.44 mM solution in Dimethylsulfoxide





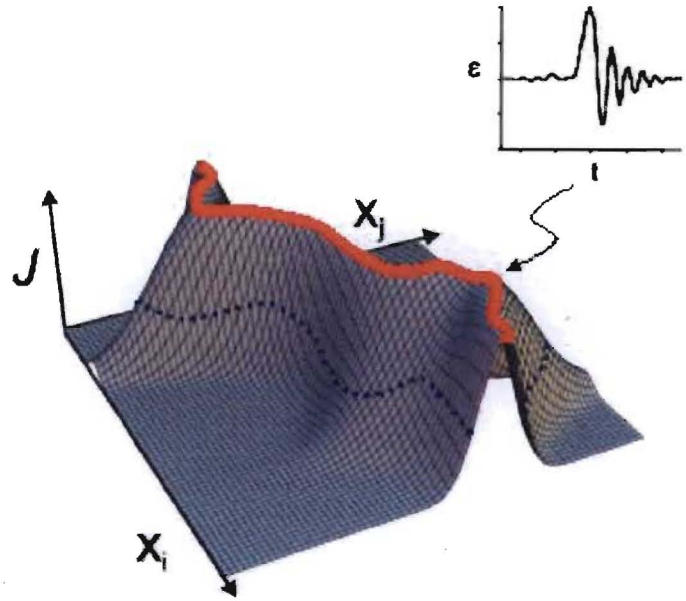
- Biexponential fits
  - **383 nm**

    τ_1=8ps τ_2=83ps
  - **490 nm**

    τ_1=0.3ps τ_2=6ps

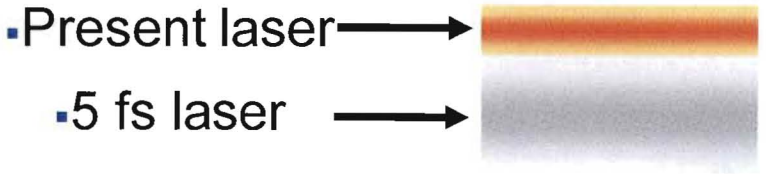
Optimal Control Landscape

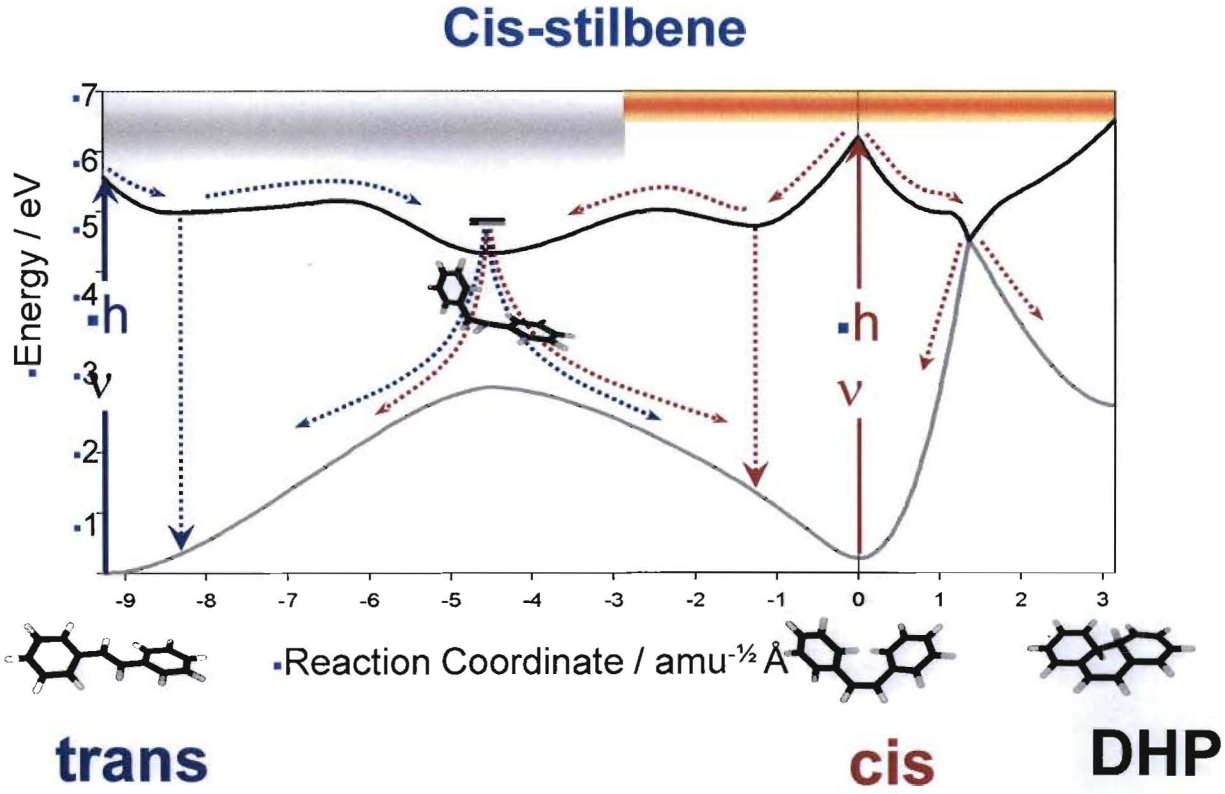
- Control landscape has no traps
  - Given a controllable quantum system, there is always a trap free pathway to the top of the control landscape from any location.
    - **Bandwidth dependent**
      - Unlimited bandwidth
      - Unlimited control
    - **Experimental limitations**



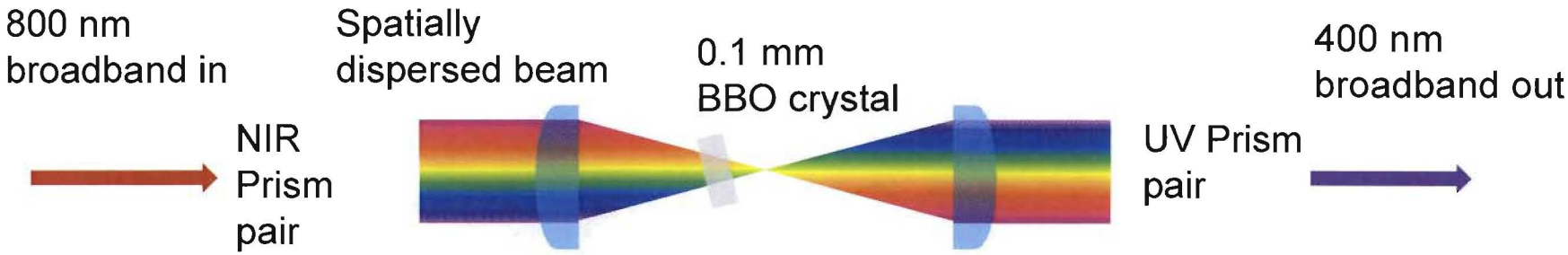
More bandwidth needed for control

- Present lasers allow for control but the extent is limited
- More bandwidth gives more control over total energy space

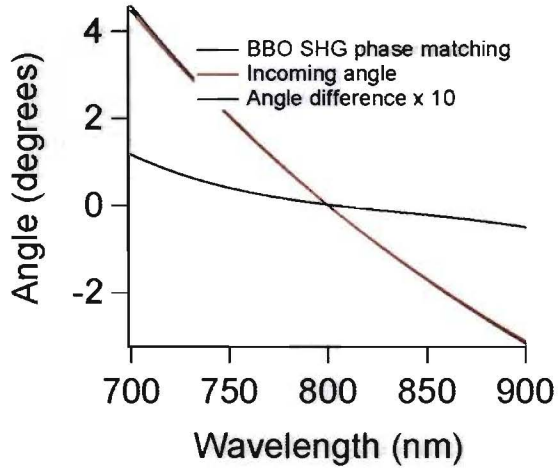




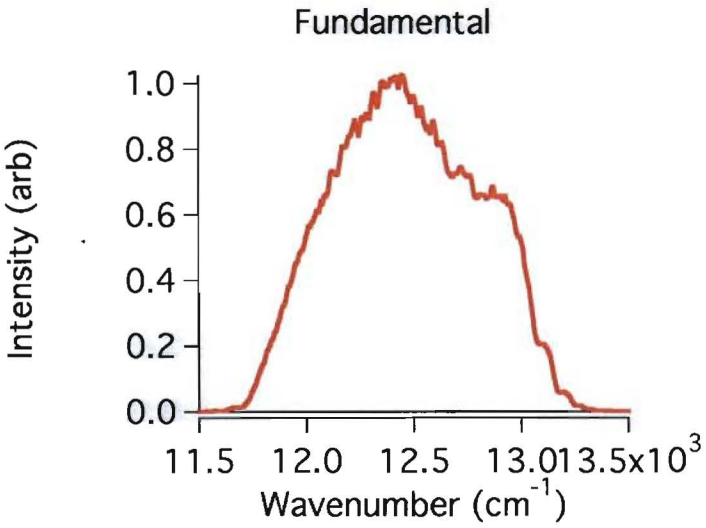
Achromatic second harmonic generation (ASHG) will allow vibrational control with electronic resonance

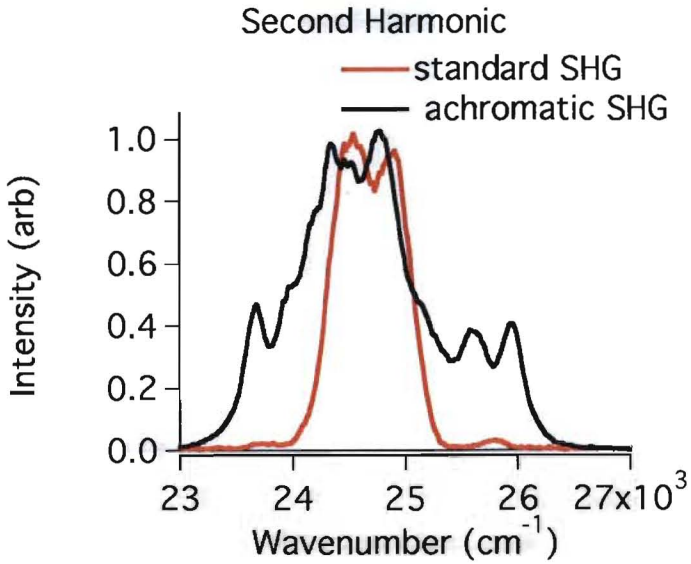


Focus angle = phase matching angle



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
    - Of non simple parameter response in HNAB and TNA
    - DAAF- further investigate degree of intensity control
    - Trinitrotoulene (TNT)
  - **Achromatic phase matching**