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Title: Optimal Dynamic Detection of Explosives (ODD-Ex)

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Optimal Dynamic Detection of Explosives (ODD-Ex)

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Abstract

The detection of explosives is a notoriously difficult problem, especially at stand-off, due to their (generally) low vapor pressure, environmental and matrix interferences, and packaging. We are exploring Optimal Dynamic Detection of Explosives (ODD-Ex), which exploits the best capabilities of recent advances in laser technology and recent discoveries in optimal shaping of laser pulses for control of molecular processes to significantly enhance the standoff detection of explosives. The core of the ODD-Ex technique is the introduction of optimally shaped laser pulses to simultaneously enhance sensitivity to explosives signatures while dramatically improving specificity, particularly against matrix materials and background interferences. These goals are being addressed by operating in an optimal non-linear fashion, typically with a single shaped laser pulse inherently containing within it coherently locked control and probe sub-pulses. Recent results will be presented.

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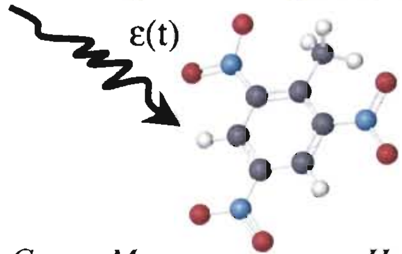
Optimal Dynamic Detection of Explosives (ODD-Ex)



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Homeland Security

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Participants and Outline

- **Princeton:**
  - Herschel Rabitz; Jon Roslund
- **Los Alamos:**
  - David Moore; Shawn McGrane; Jason Scharff; Margo Greenfield, Robert Chalmers

Outline

- Background
- ODD basics
- Bandwidth broadening / vibronic control
- Multiplex CARS / mixtures
- Multiobjective optimization
- Summary

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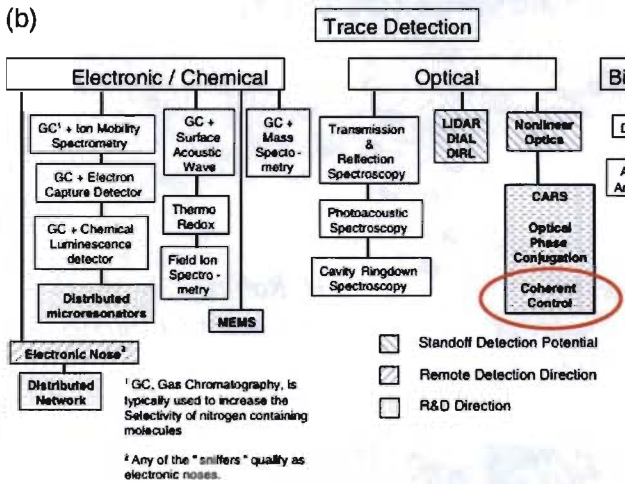
NRC Review

Existing and Potential Standoff Explosives Detection Techniques (2004)



"Using a new research technique called coherent control could further enhance molecular selectivity…one could choose to selectively ionize or dissociate particular explosive molecules…"

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DHS Workshop

Transformational Breakthroughs - Physics Approaches (LLNL - 2005)

Laser Spectroscopy Topic Area - Recommendations

- Tailored desorption methods to increase vapor phase concentration and suppress substrate lift-off
  - "spatial and temporal laser pulse shaping"
- Expand detected emission spectral range, especially for LIBS
  - LIBS is destructive of both the explosive molecule and the surface
  - Pulse shaping (quantum control) should allow use of much lower laser energies as well as lead to expanded emission spectral range
- Non-linear optical methods
  - Pulse shaping can enhance molecular resonances allowing long distance stand-off detection (i.e., force target molecules to spill out their signatures, or be strong emitters with a unique signature)

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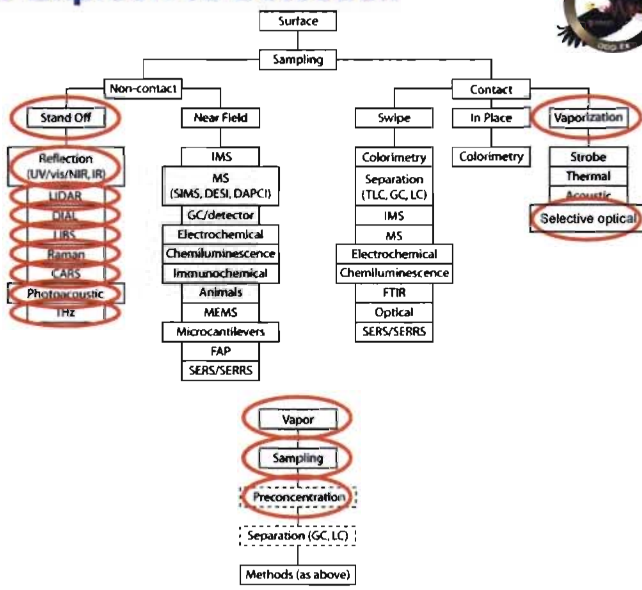
Application to Explosives Detection

- We foresee a large number of applicable areas for ODD
  - Circled in red
  - One can imagine a large number of spectroscopies with vastly improved characteristics



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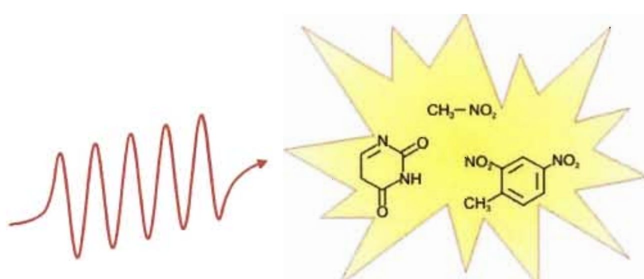
Linear spectroscopy - unshaped pulses

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



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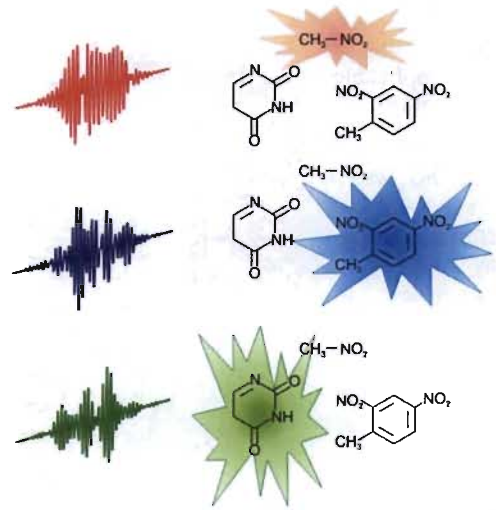
Quantum Optimal Dynamic Discrimination (ODD)

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



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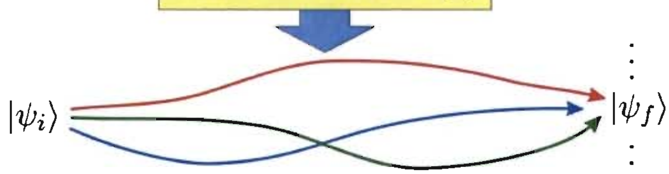
Control of Quantum Systems

- **Customization** of molecular Hamiltonian by optimally shaped field

$$H(t) = H_0 - \mu\epsilon(t)$$

- **Optimally drive** quantum system towards desired final state

Laser Control Field $\epsilon(t)$



- Constructive interference for $|\psi_f\rangle$
- Destructive interference for $|\psi_{f'}\rangle \neq |\psi_f\rangle$

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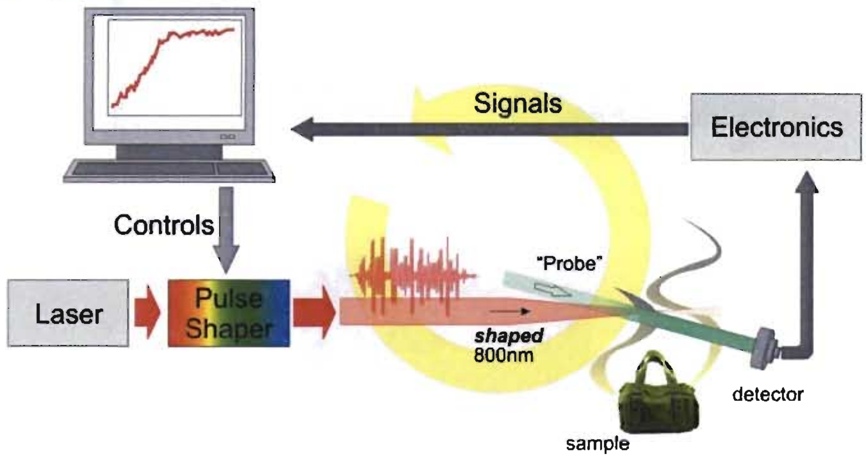
Discovery of Optimal Photonic Reagents

- **Fully automated** high duty cycle closed-loop operation



- **High finesse control** of system without *a priori* model of the physical sample

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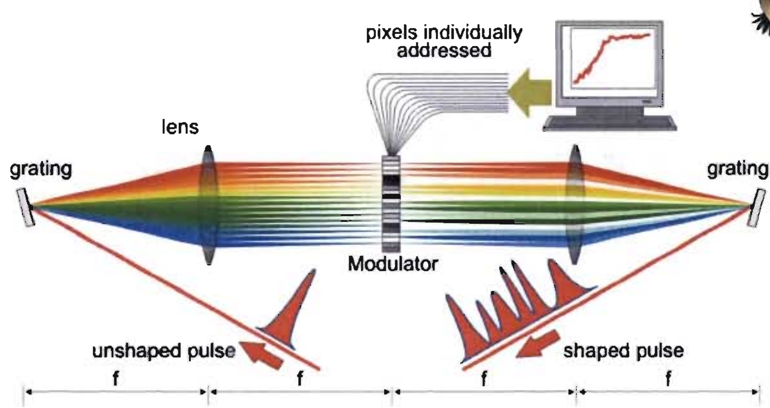
Creating Photonic Reagents on Demand



- Start with raw, featureless, ultrafast laser pulse (30-100 fs)
- Filter spectral amplitude and phase (SLM or AOM)
- Fully automated computer generation of photonic reagents

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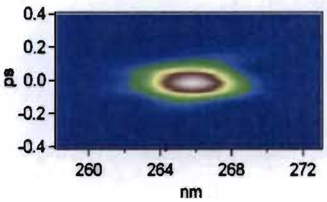
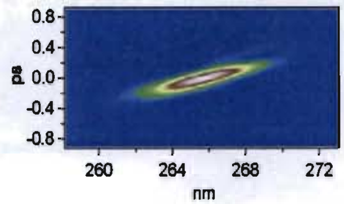
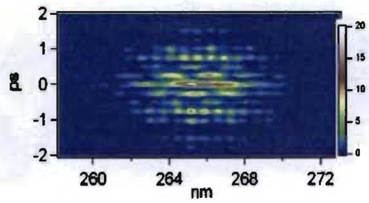
Examples of Shaped Pulses



Transform limited ~150fs



Simple linear chirp



Dual sine waves

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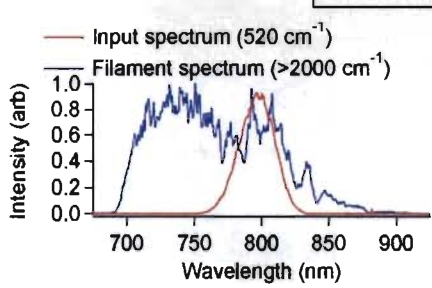
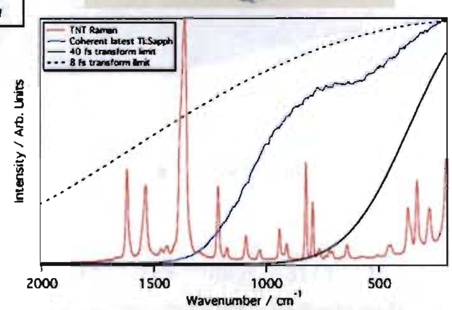
Increasing the Control Bandwidth

Filamentation



New Laser Technology



~2000 cm^{-1} bandwidth is comparable to vibrational fingerprint region

- Allows coherent Raman spectroscopies
  - and vibronic control of emission

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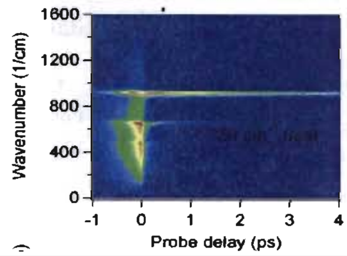
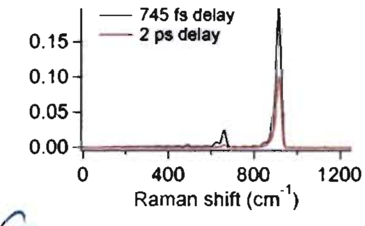
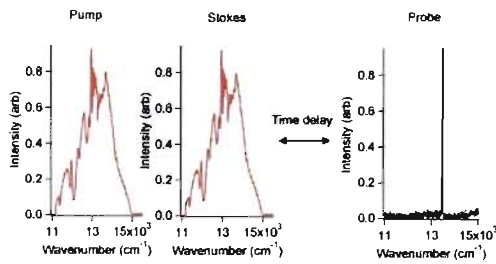
Broadband Coherent Raman

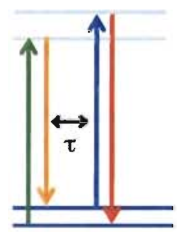
Multiplex CARS in nitromethane



Records full spectrum each laser pulse







Coherence created with first 2 pulses probed by 3rd pulse induces emission of 4th pulse

Pump + Stokes delay τ Probe+CARS

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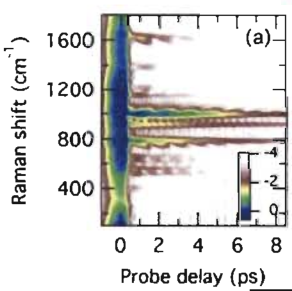
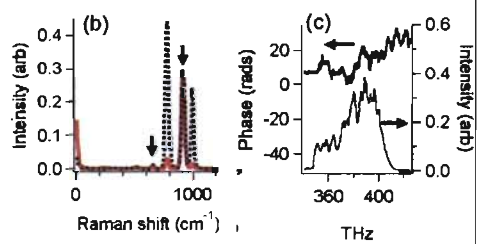
Controlled Detection of Mixture Components

Selectivity through pulse shaping

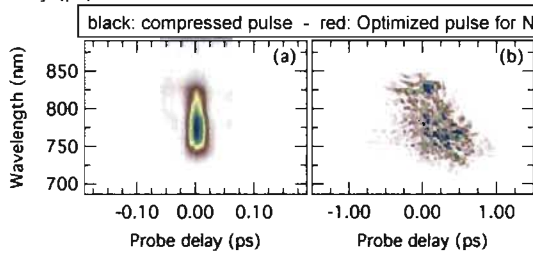
CARS of mixture: toluene; acetone; nitromethane



Optimize at 1.25 ps



black: compressed pulse - red: Optimized pulse for NM



Optimized pulse retains NM peaks while suppressing solvent peaks

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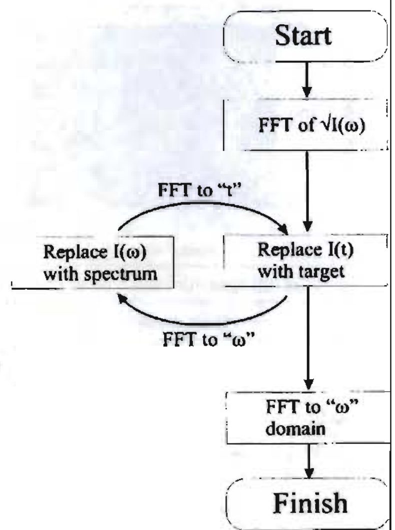
Faster Optimization

Use of Gerchberg-Saxton Algorithm

- GS is independent of target complexity, pulse shaper resolution; no cost functions, weight factors or optimization parameters.
- Only requires known target spectrum and the raw laser spectrum
- Algorithm steps:
  - FFT of the laser pulse (spectrum), starting with random phase
  - Replace amplitude with target spectrum, retain phase
  - IFFT back to spectral domain, replace amplitude with laser spectrum, retain phase
  - Iterate until minimal changes



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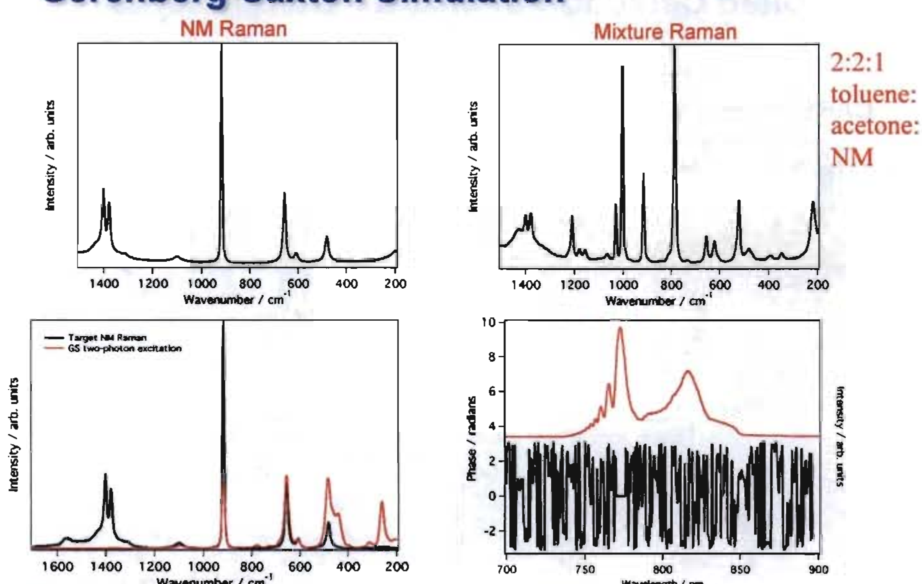
Gerchberg-Saxton Simulation



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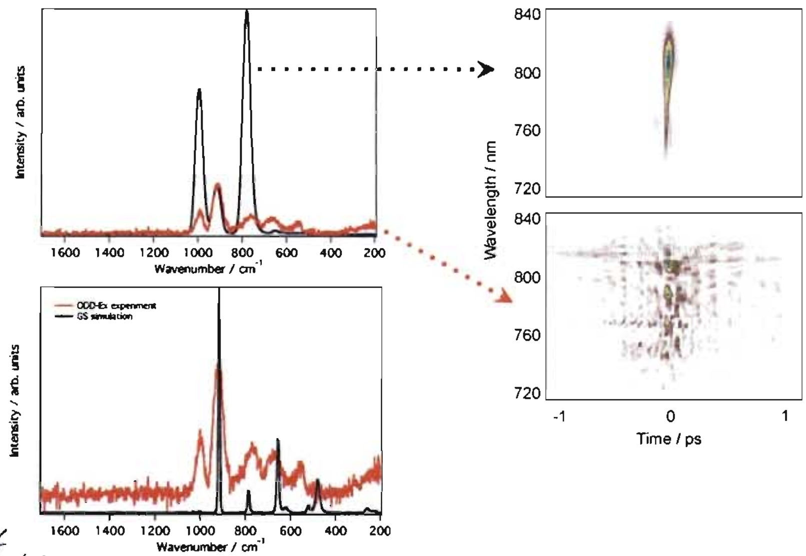
ODD-Ex with Gerchberg-Saxton Phase



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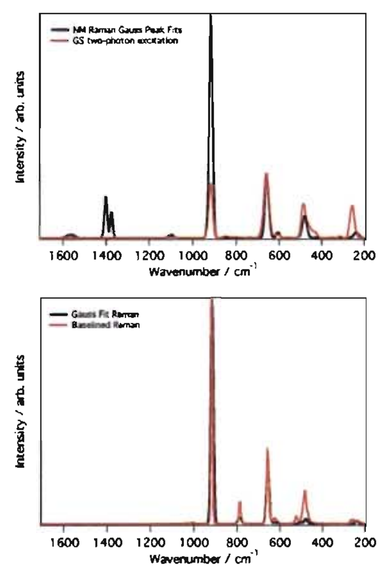
Gerchberg-Saxton Improvements

- Noise reduction in target spectrum
  - Use peak fits instead of baseline removal to produce zeros between peaks
- Establish iteration end criteria
  - Look for minimal change - define



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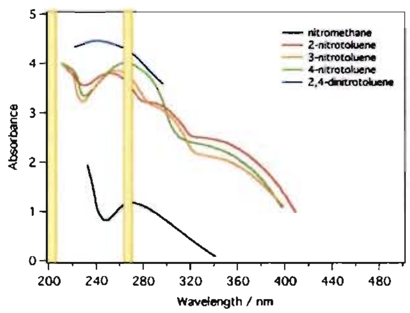
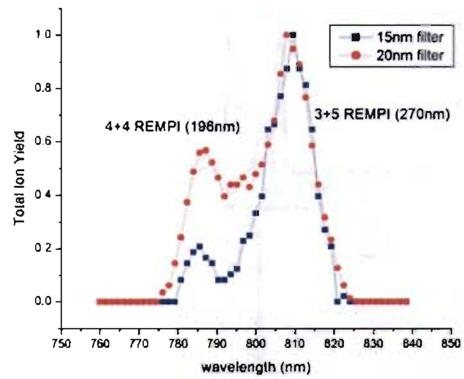
Use Electronic Resonances

- Nitrotoluenes have resonances near 266 nm and 200 nm
- Excellent fit to Ti:sapphire harmonics
- Measure multi-order REMPI spectrum
- Obvious 3 and 4 photon resonances





Exploit these two multiphoton resonances to generate interferences

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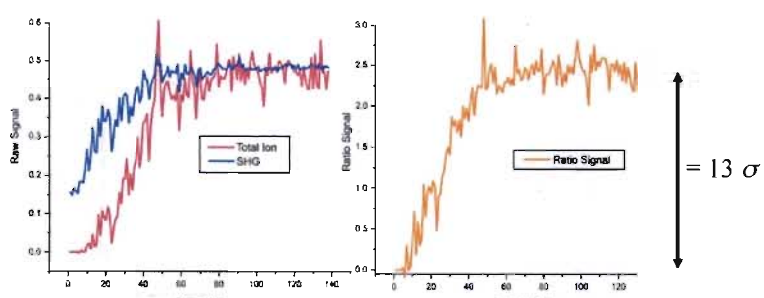
Multiobjective Optimization Needed

- Need to discriminate against intensity dependence - use fitness function:

$$f = \frac{\mathcal{J}_{Ion}}{SHG^{\alpha}}$$

- Use 60 nm bandwidth to cover both 3 and 4 photon resonances for enhanced discrimination
- Use MOTC algorithm: Derandomized Evolution Strategies (DES) with extractable covariance matrix information



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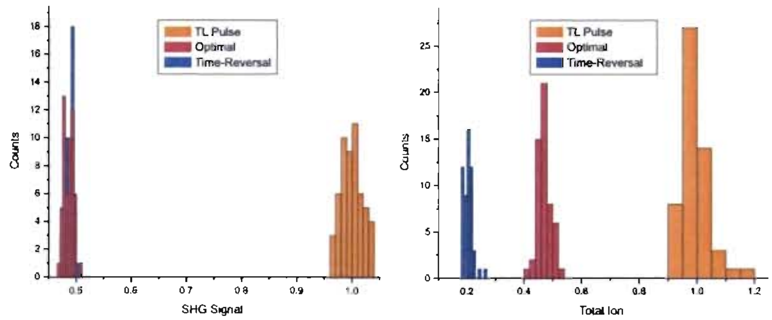
Is This Just an Intensity Effect?

- Time ordering of colors in shaped pulse doesn't matter for purely intensity dependent processes
- Test effect of reversed color ordering in optimal pulse:



Expected result for SHG, but…
Total Ion depends on color ordering!

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Summary

- **Optimal Dynamic Detection** offers a viable path to significant improvements in selectivity and sensitivity
- **Photonic reagents** are optimally tailored electromagnetic pulses that enable selective addressing of different species
- **Single pulse** photonic reagent can be designed to create a tailored wavepacket in the analyte excited state and interrogate the system by a stimulated signal
  - The optimally controlled multispectral stimulated signal is sensitive to detailed sample vibronic structure and dynamics
- **Large bandwidth sources** allow coherent Raman spectroscopies and vibronic control of emission
- **Multiobjective optimization** allows
  - Discrimination against unwanted nonlinear effects or other interferences
  - Balancing e.g., selectivity versus sensitivity

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