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Title:	Coherent Control Methods for Detection of Explosives
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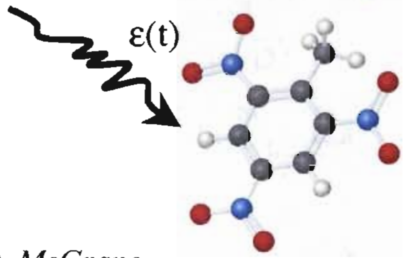
Coherent control methods for detection of explosives

D.S. Moore, S.D. McGrane, M.T. Greenfield

OSA Laser applications to chemical, security and environmental analysis (LACSEA) Conference,
San Diego, CA 29 Jan – 1 Feb 2012

Abstract:
We are exploring optimal dynamic detection of explosives (ODD-Ex) to exploit recent advances in optimal shaping of laser pulses for control of molecular processes to significantly enhance the standoff detection of explosives.

Coherent Control Methods for Detection of Explosives



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Participants and Outline

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

Outline

- Background
- Optimal Control
- Bandwidth broadening / vibronic control
- Multiplex CARS / mixtures
- Other applications
- Summary

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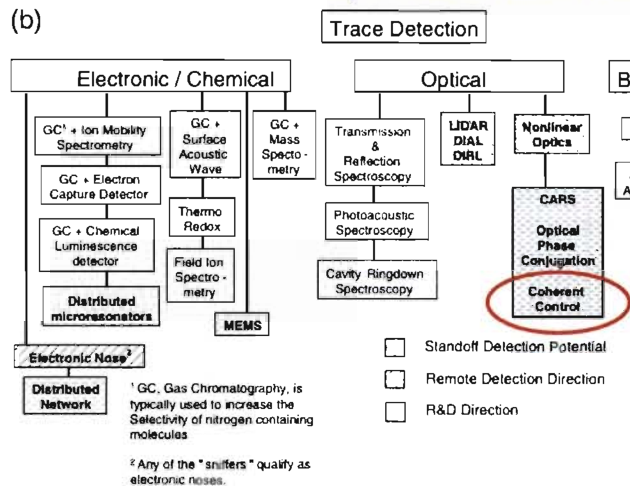
NRC Review

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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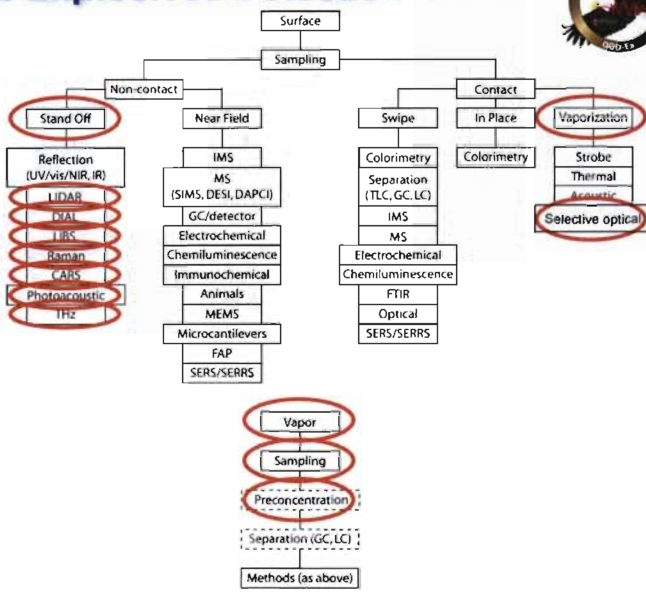
Application to Explosives Detection

- 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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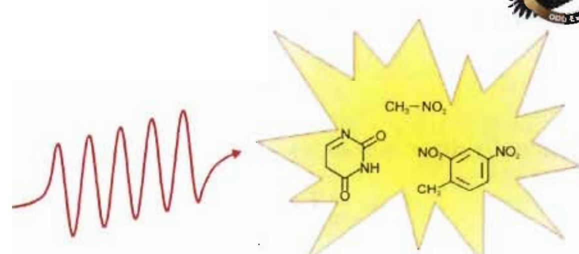
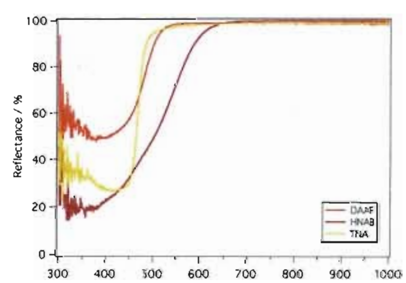
Linear spectroscopy - unshaped pulses

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





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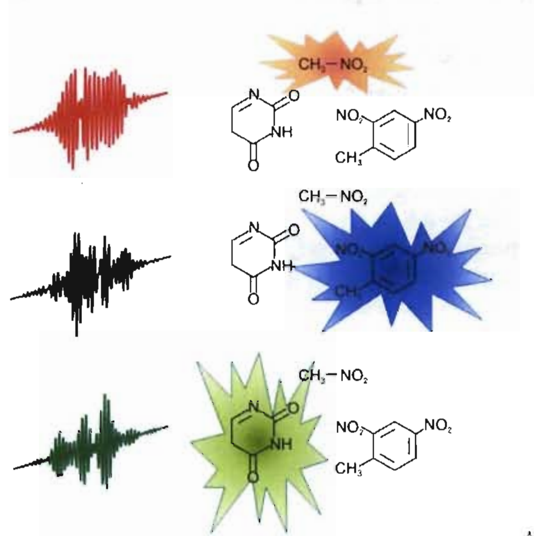
Quantum Optimal Dynamic Discrimination (ODD)

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



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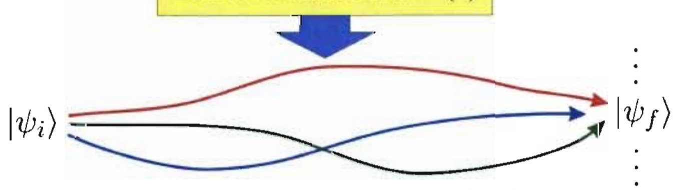
Control of Quantum Systems

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

$$\boxed{H(t) = H_0 - \mu\epsilon(t)}$$

- **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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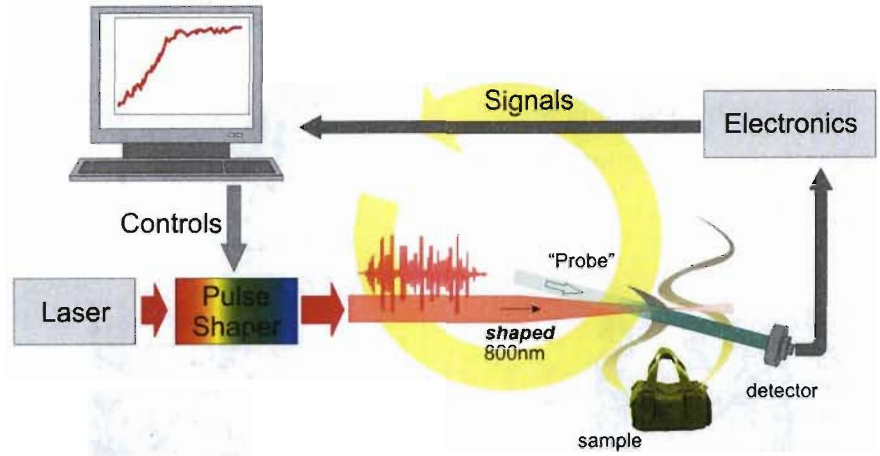
Discovery of Optimal Photonic Reagents

- **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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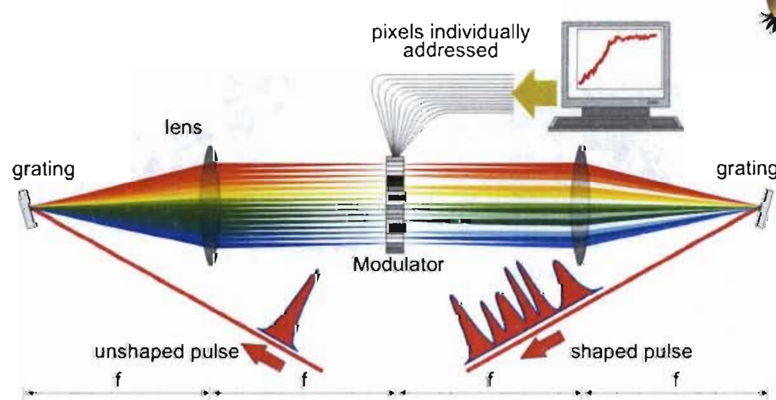
Creating Photonic Reagents on Demand



- 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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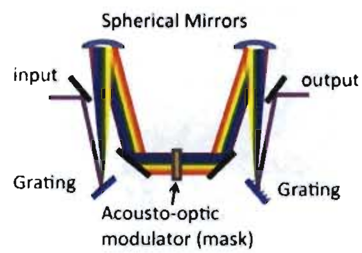
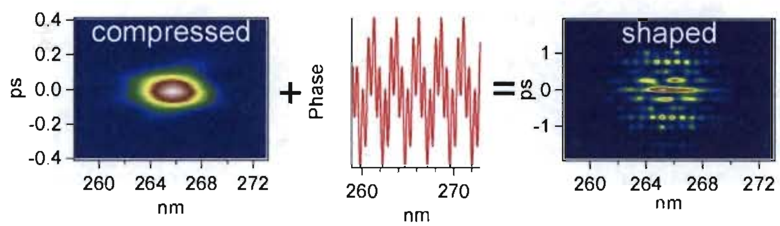
Pulse Shaping : Time Dependent Electric Field

- **Femtosecond optical pulse shaping**
  - Shape E(t) by adding phase to E(ω) and Fourier transforming
  - $E(t) = a(t)e^{i\varphi(t)}$





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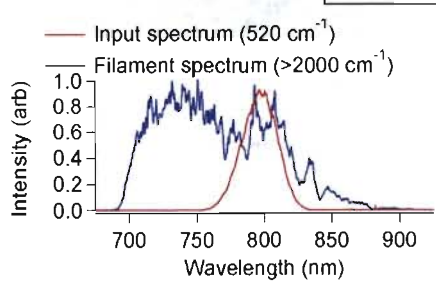
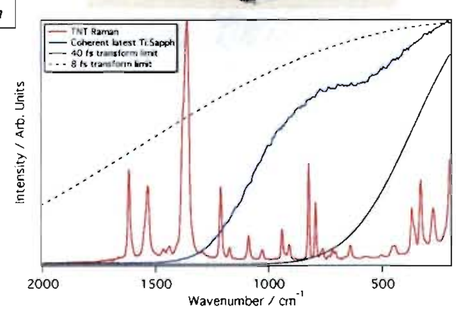
Increasing the Control Bandwidth

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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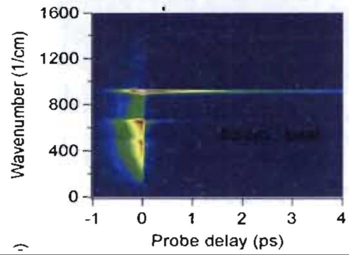
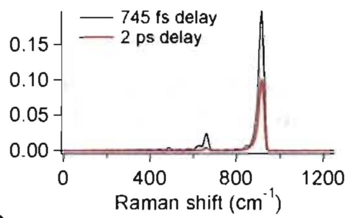
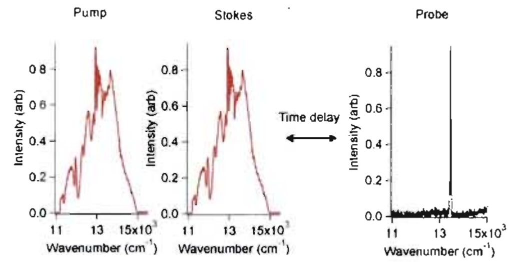
Broadband Coherent Raman

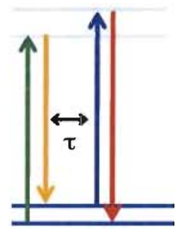
Multiplex CARS in nitromethane



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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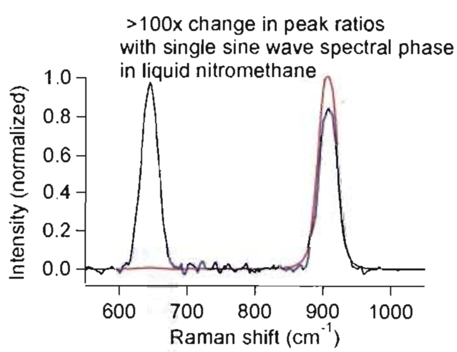
Coherent control of a single peak is easy



- Same solution, same pulse energy, same pulse spectrum
- Different emission spectra

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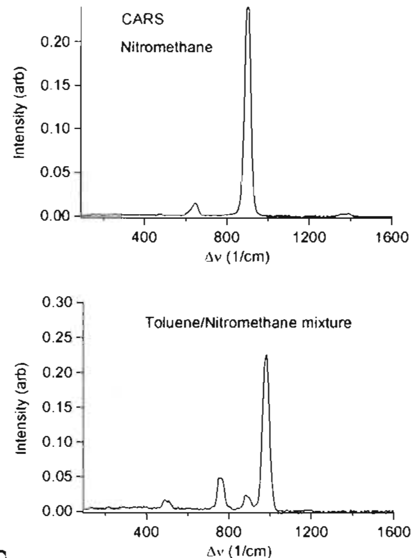
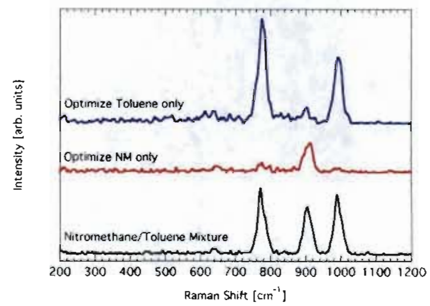
Coherent control of a few peaks is less easy





- Optimize the nonlinear excitation of multiple frequencies while simultaneously minimizing all other frequencies

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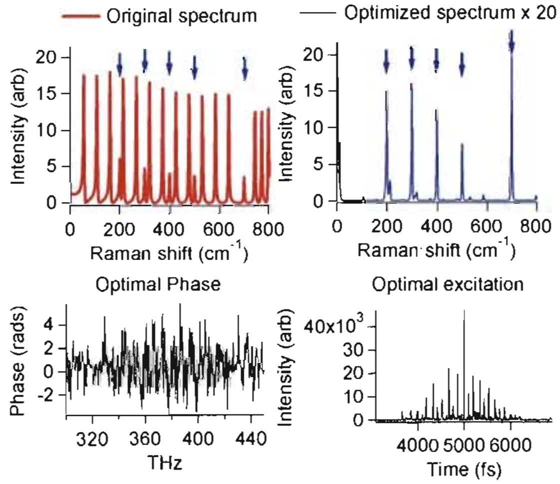
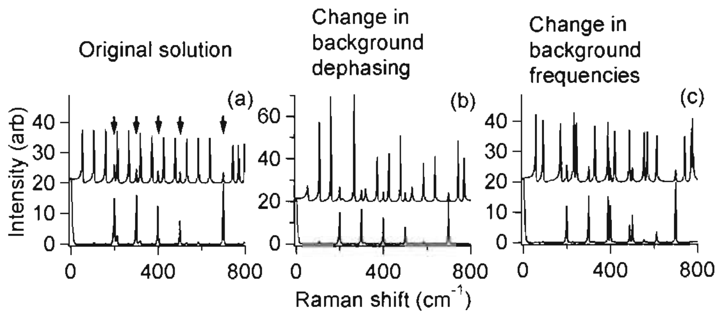
Coherent control of many peaks is more difficult

CARS multimode optimization simulation



- Results from a computer simulated GA optimization

However, the solution is robust to environmental changes



Since the mechanism is frequency based suppression, the optimal excitation can be robust to changing the interferent spectra.

This is ideal for selective Raman detection.

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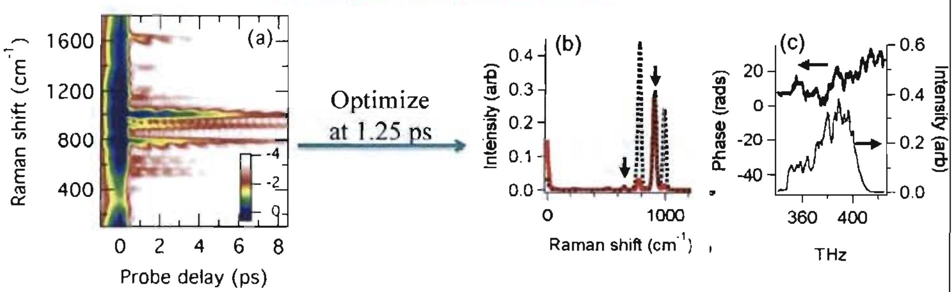
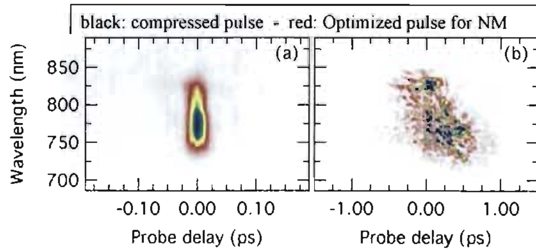
Controlled Detection of Mixture Components

Selectivity through pulse shaping

CARS of mixture: toluene; acetone; nitromethane



black: compressed pulse - red: Optimized pulse for NM



Optimized pulse retains NM peaks while suppressing solvent peaks

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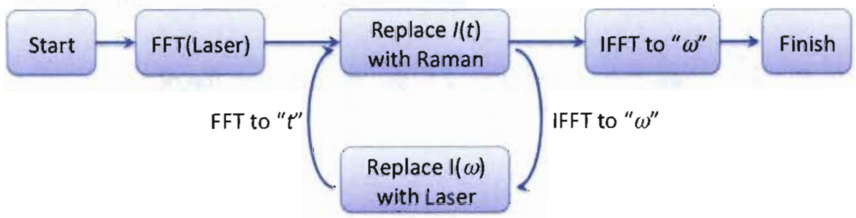
Faster Optimization

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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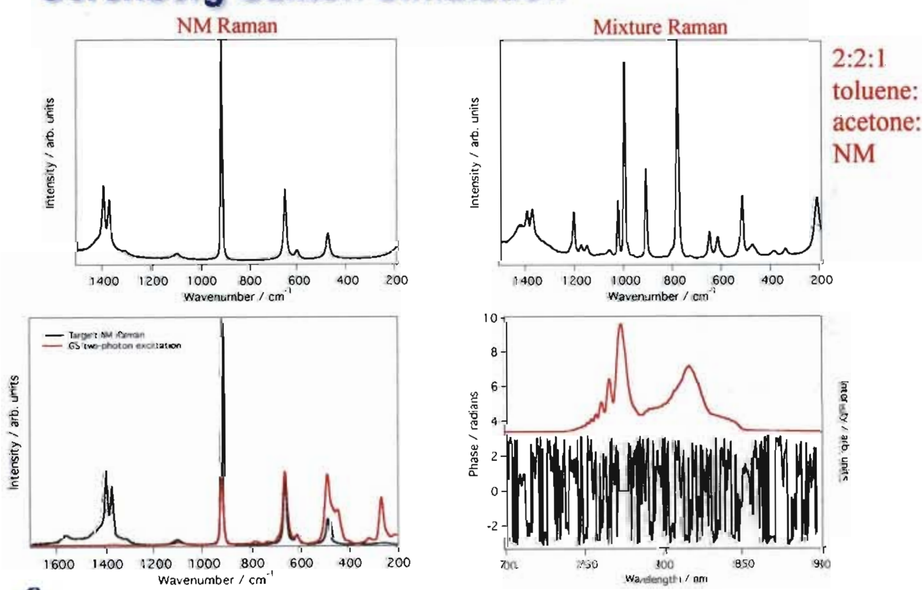
Gerchberg-Saxton Simulation



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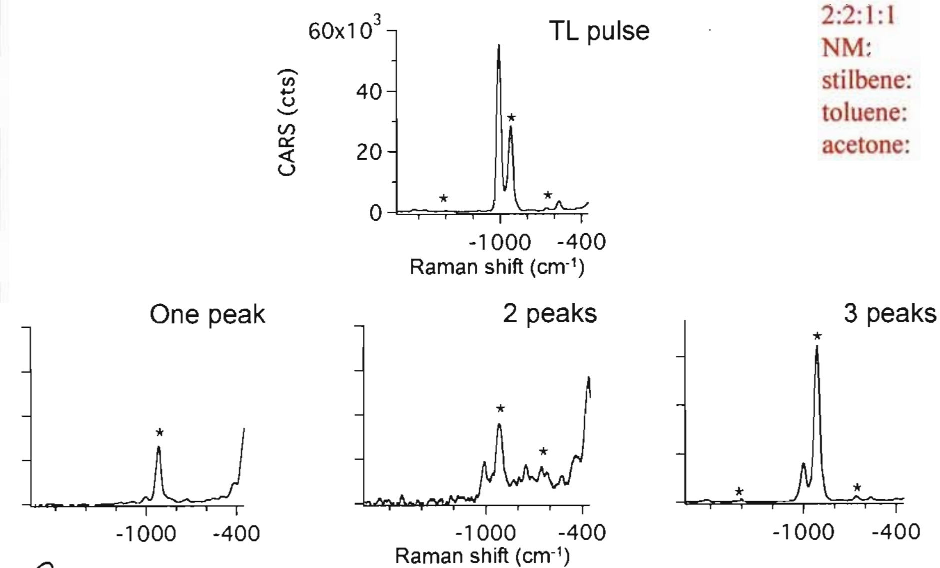
ODD-Ex with Gerchberg-Saxton Phase

2:2:1:1
NM:
stilbene:
toluene:
acetone:



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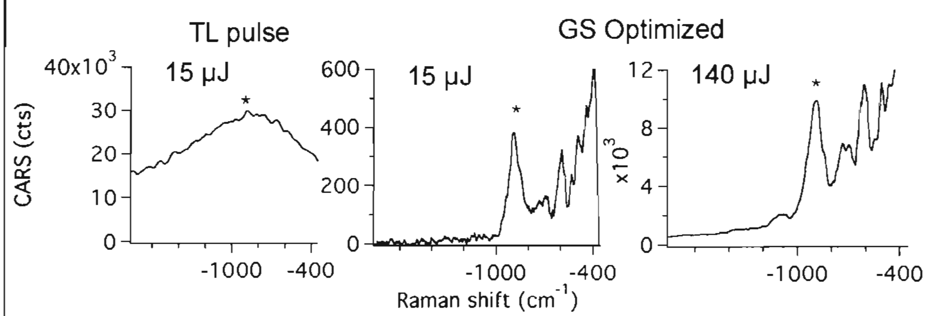
ODD-CARS in Other Samples

3% H_2O_2 in fluorescent mixture



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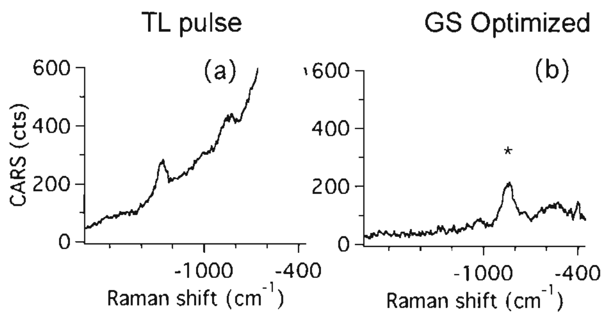
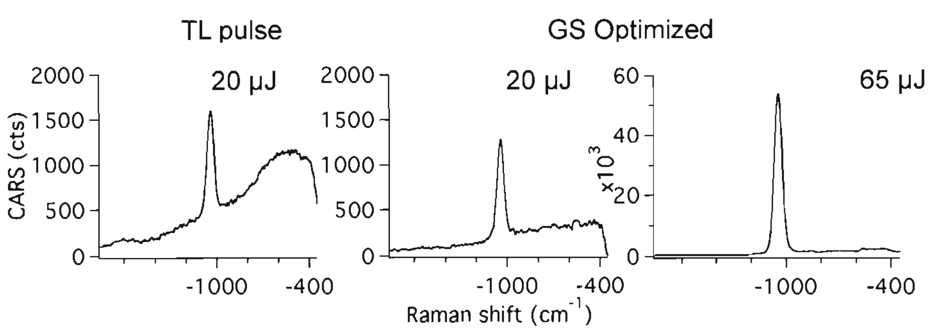
ODD-CARS in Other Samples

Trace PETN on yellow painted surface



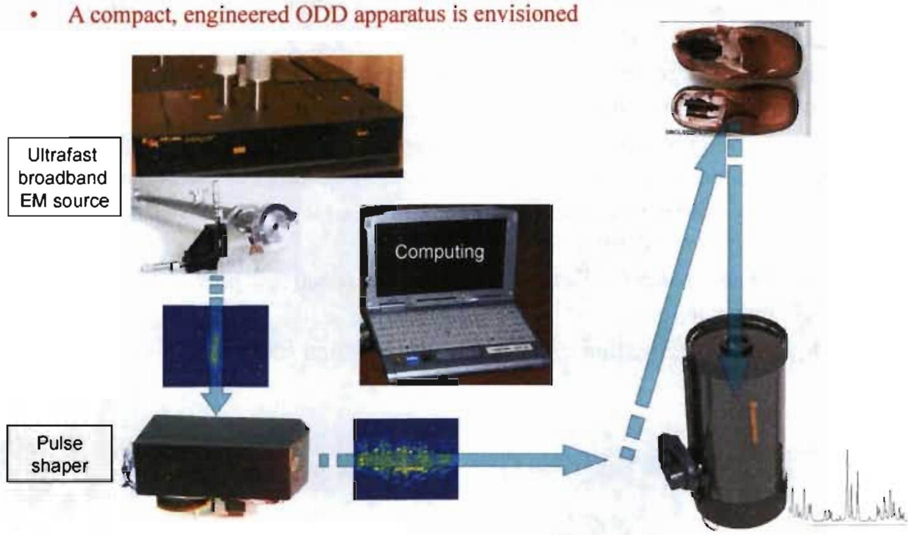
ODD-CARS in Other Samples

10% Ammonium nitrate
a fluorescing solid



Building Blocks for ODD-Ex

- A compact, engineered ODD apparatus is envisioned



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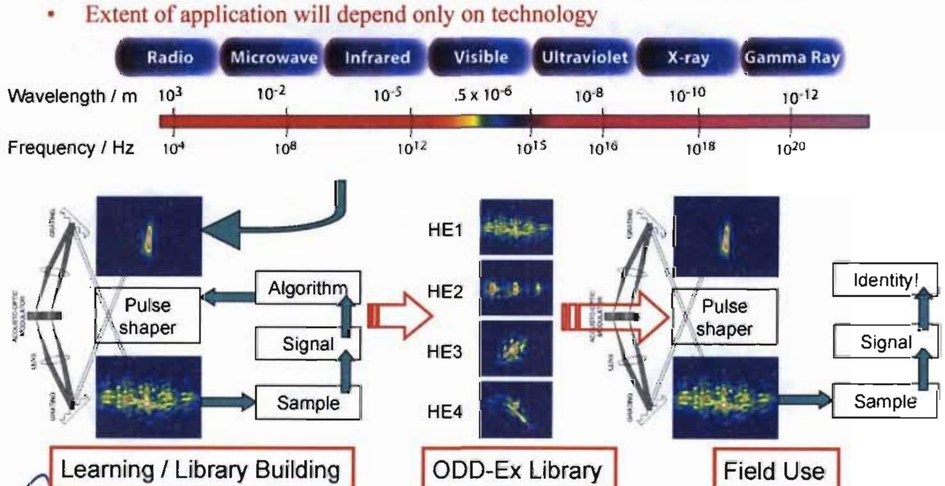
Optimal Control Space

- Not only UV/visible spectroscopic regions can be controlled, but *anywhere* in the entire EM spectrum *where source bandwidth is available for manipulation*
- Extent of application will depend only on technology



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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** to balance selectivity and sensitivity

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