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Title: Ultrafast laser based coherent control methods for explosives detection

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Ultrafast laser based coherent control methods for explosives detection

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.

Ultrafast laser based coherent control methods for explosives detection

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with:
Herschel Rabitz et al., Princeton University

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Outline

- Stand-off detection methods in use
- Why another method?
- Optimal Dynamic Detection
  - ODD basics
  - Bandwidth broadening / vibronic control
  - Multiplex CARS / mixtures
  - Use of excited electronic state dynamics
  - Summary

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Standoff Methods Demonstrated

- Raman
- Laser Induced Breakdown Spectroscopy - LIBS
- Laser Induced Fluorescence – LIF
- Laser Evaporation + LIF
- Infrared Spectroscopy
  - Vapor phase
    - Active and Passive
  - Solid phase
    - Mid-IR Quantum Cascade Laser Imaging
    - Mid-IR QCL photothermal detection
    - Pulsed laser fragmentation with mid-IR QCL
    - Passive IR imaging
- THz Spectroscopy and Imaging
- Coherent anti-Stokes Raman

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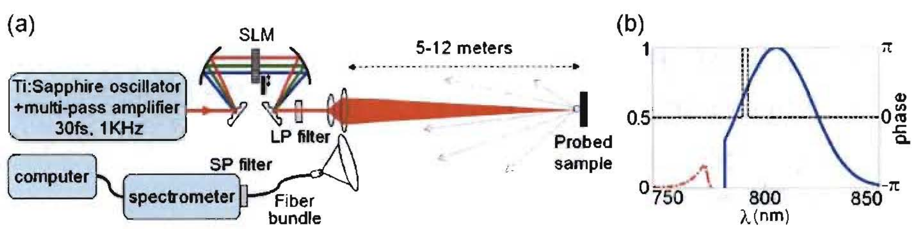
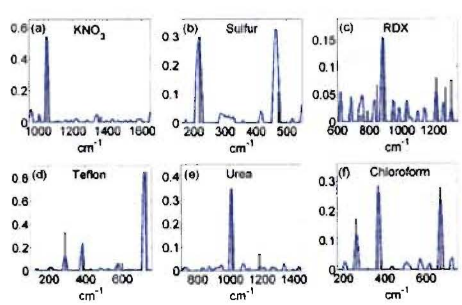
Coherent Anti-Stokes Raman Spectroscopy - CARS



O. Katz et al., Appl. Phys. Lett. 92:171116 (2008)



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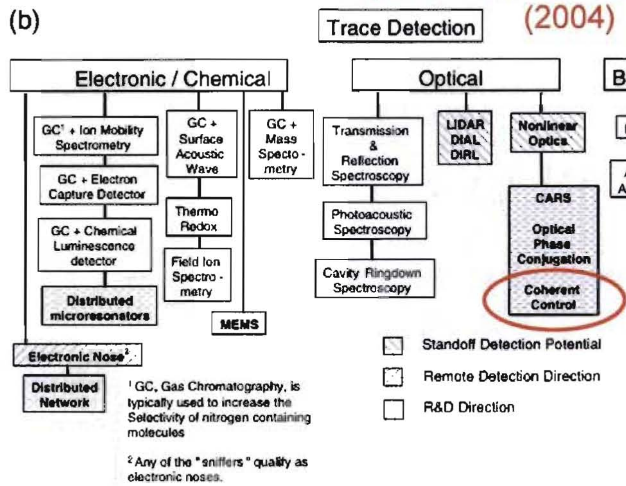
NRC Review

Existing and Potential Standoff Explosives Detection Techniques (2004)

(b)



"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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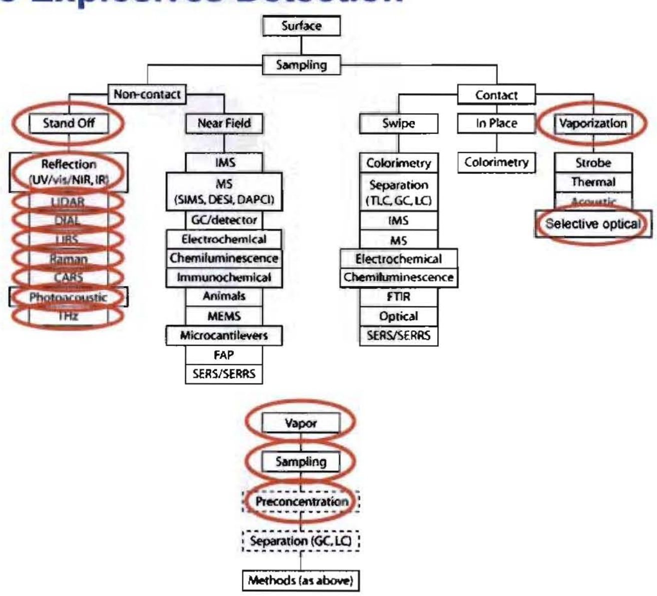
Application to Explosives Detection

- 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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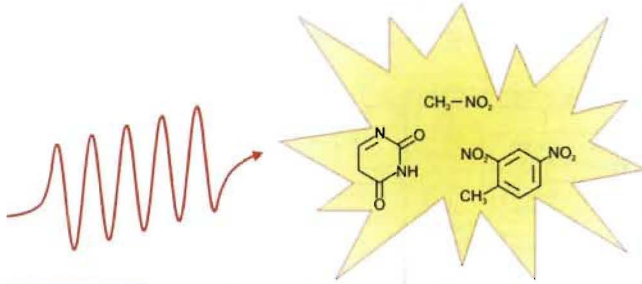
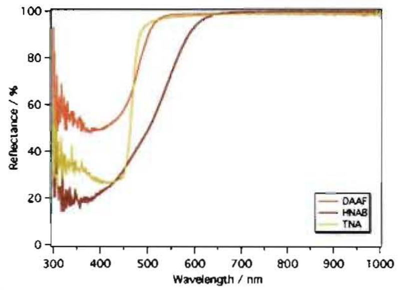
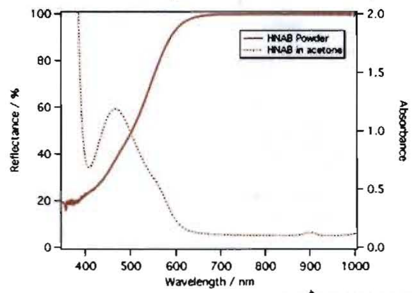
Linear spectroscopy - unshaped pulses

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







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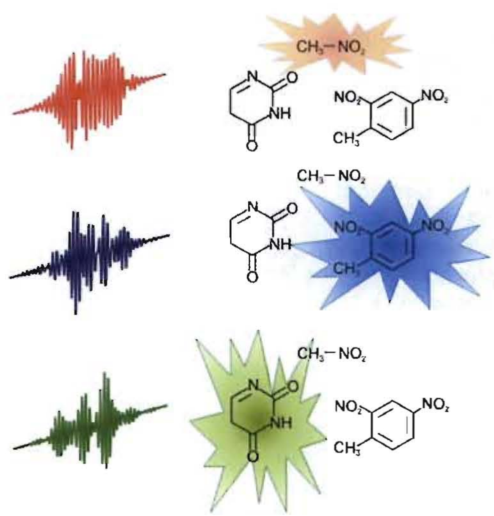
Quantum Optimal Dynamic Discrimination (ODD)

- **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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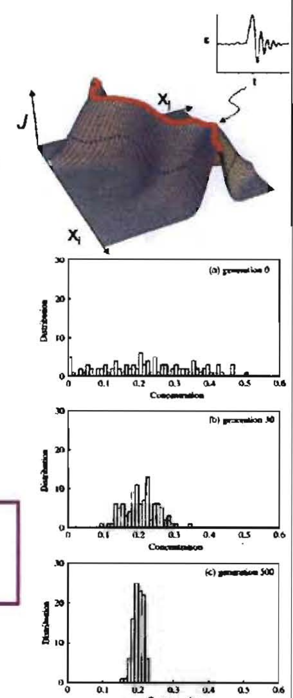
Optimal control landscape



- 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 detection of explosives

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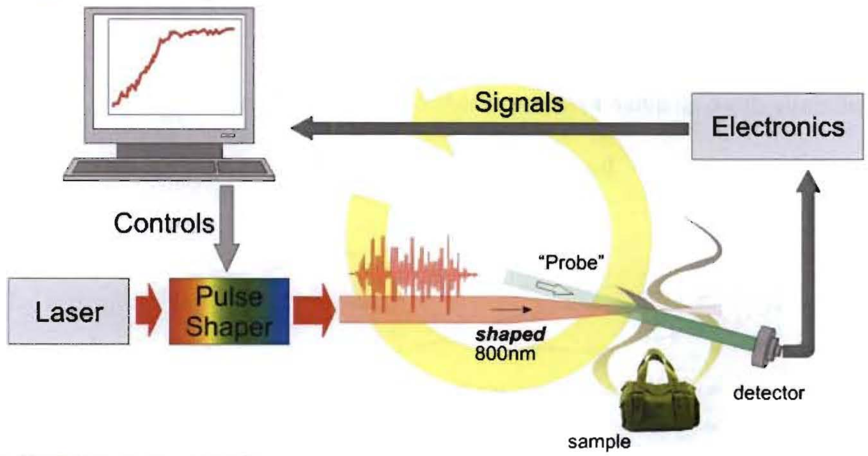
Discovery of optimal photonic reagents

- **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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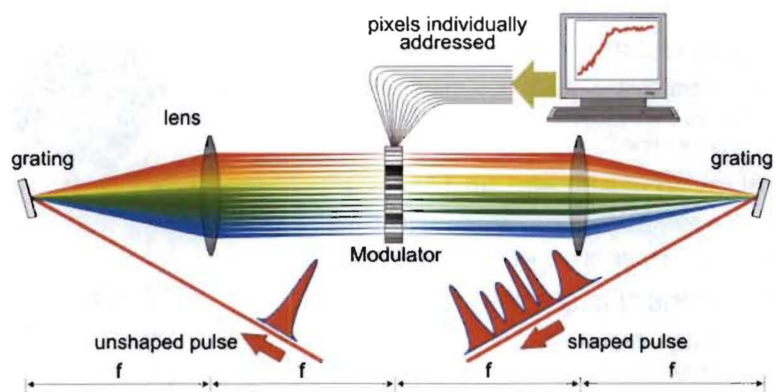
Creating photonic reagents on demand



- 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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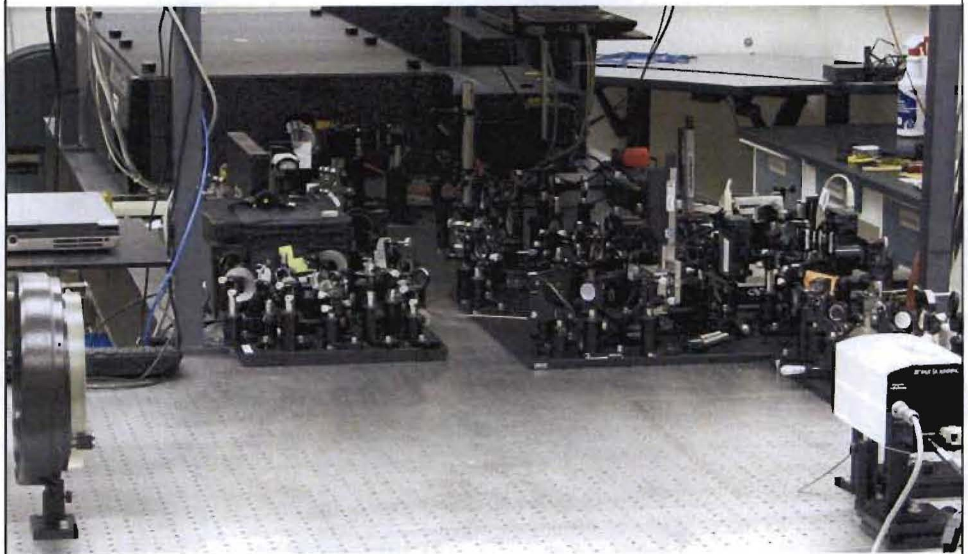
What does this actually look like?



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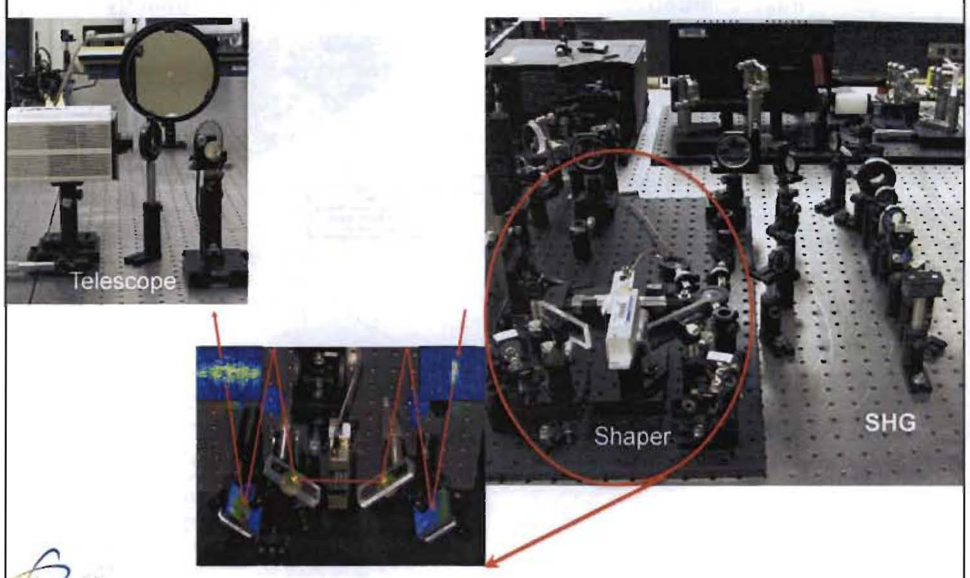

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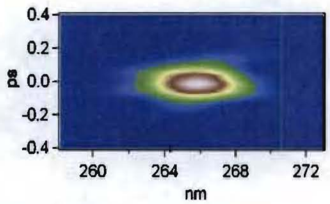
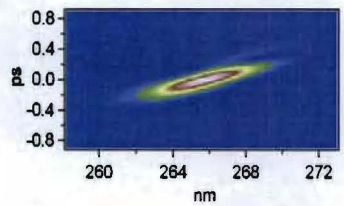
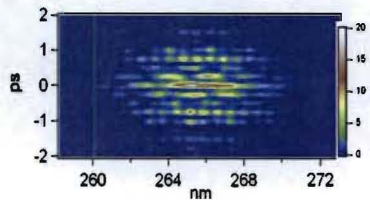
Examples of shaped pulses



Transform limited ~150fs



Simple linear chirp



Dual sine waves

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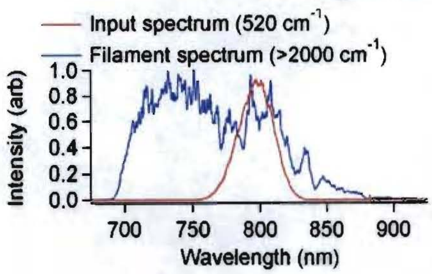
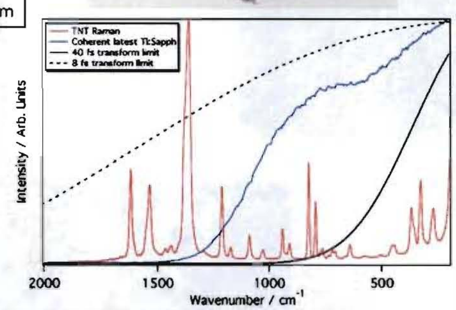
Increasing the Control Bandwidth

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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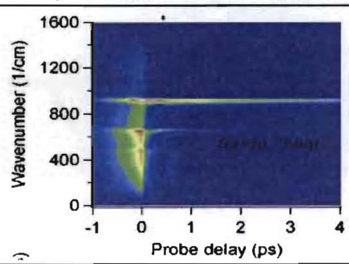
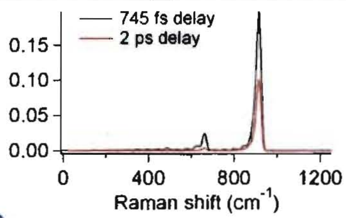
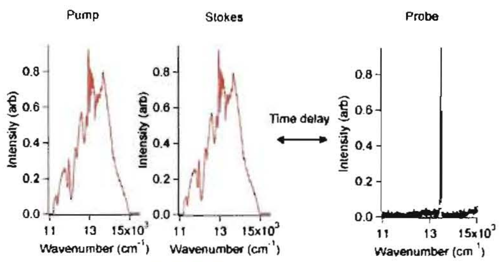
Broadband Coherent Raman

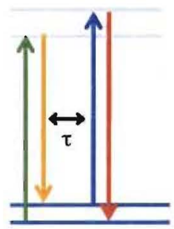
Multiplex CARS in nitromethane



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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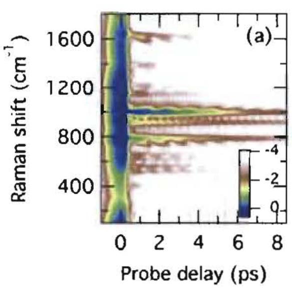
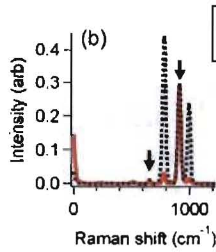
Controlled detection of mixture components

Selectivity through pulse shaping

CARS of mixture: toluene; acetone; nitromethane



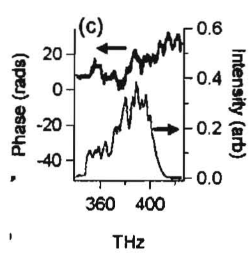
Optimize at 1.25 ps



black: compressed pulse
red: Optimized pulse for NM

Optimized pulse retains NM peaks while suppressing solvent peaks

Optimized pulse:
top: phase
bottom: intensity



Choice of fitness function is critical to success

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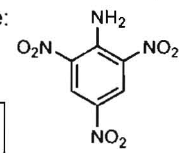
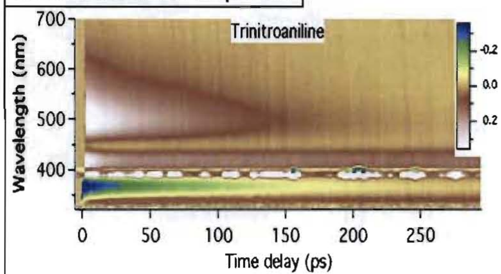
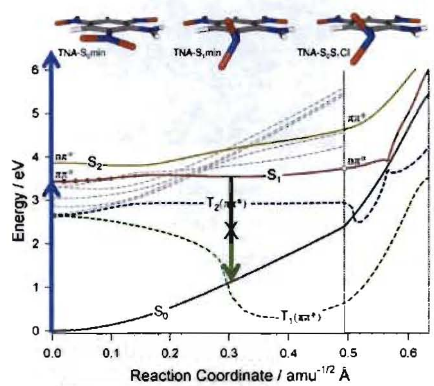
Can we use excited state dynamics?

- Calculations of excited states and trajectories indicate other detection routes
- Trinitroaniline example:



Excite at 400 nm
Measure transient spectra





Must be multiphoton
Calculate no osc. from S1 min

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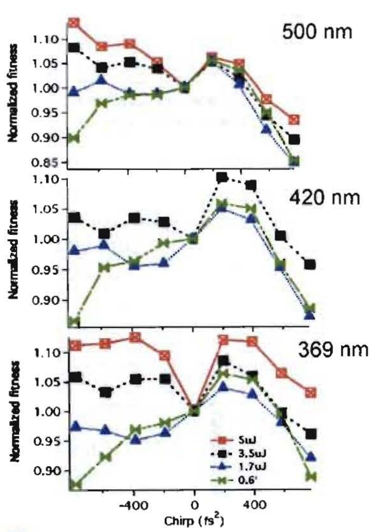
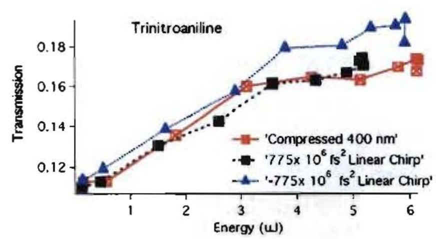
Is single parameter control available?





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

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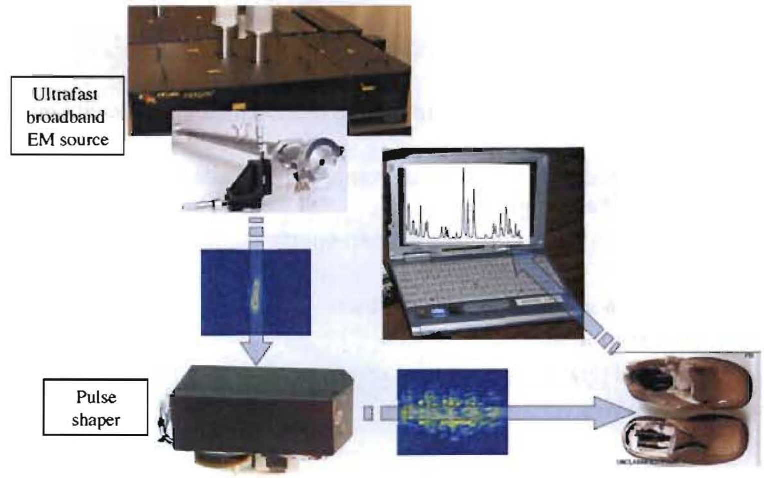
Building Blocks for ODD-Ex

- A compact, engineered ODD apparatus is in development



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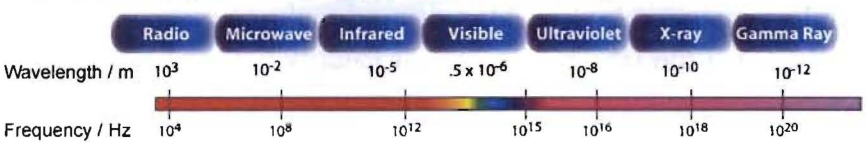
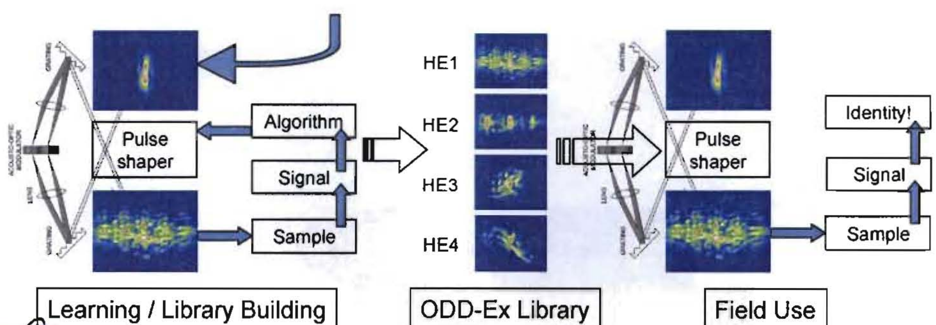
Optimal Control Space

- Not only UV/visible/NIR 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

- **Standoff Optical Methods** have been shown capable of measuring small amounts of explosives at many meters distance, but suffer from interferences or low specificity
- **Standoff Raman** improves specificity, but fluorescence and weak signals are still issues
- **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
- **Large bandwidth sources** allow coherent Raman spectroscopies and vibronic control of emission
- **Calculations** aid optimizations and fitness functions
- **Multiobjective optimization** allows
  - Discrimination against unwanted nonlinear effects or other interferences
  - Balancing e.g., selectivity versus sensitivity

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