Phase Control of Laser-Matter Interactions and Applications

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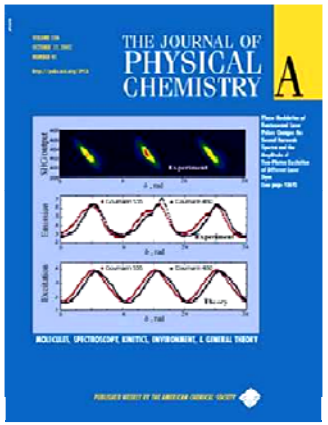
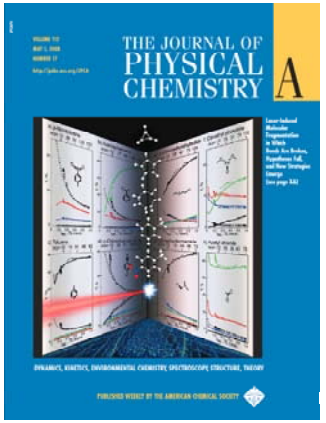
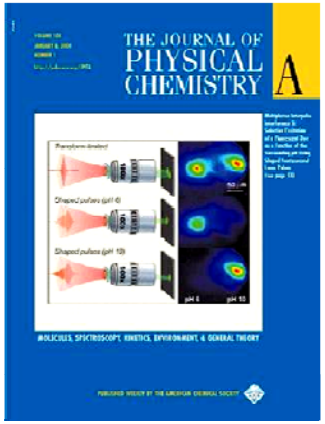
Jay Shah
Arkaprabah Konar
Marshall Bremer
Xin Zhu
Jess Gun
Paul Wrzesinski
Johanna De la Cruz
Janelle Shane
Nelson Winkler
Matthew Comstock

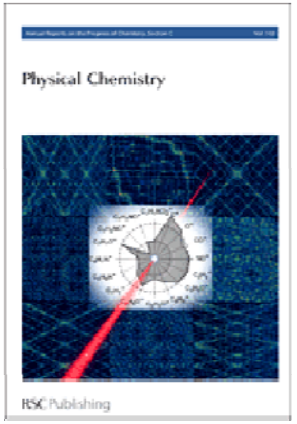




Biophotonic SOLUTIONS INC
ULTRAFAST PULSE SHAPING & COMPRESSION

Dr. Igor Pastirk
Dr. Tissa Gunaratne
Dr. Bingwei Xu
Dr. Dmitry Pestov





NSF
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NATIONAL INSTITUTES OF HEALTH
U.S. DEPARTMENT OF HOMELAND SECURITY
AIR FORCE OFFICE OF SCIENTIFIC RESEARCH
UNITED STATES AIR FORCE
MICHIGAN ECONOMIC DEVELOPMENT CORPORATION
MICHIGAN STATE UNIVERSITY

COI Disclosure: Dantus is the founder and president of Biophotonic Solutions Inc.

2001

A 6fs pulse Bardeen and Shank in 1987; Sub-6fs Baltuska and Weirsma 1997

Computer-controlled phase amplitude pulse shapers demonstrated 1994, Commercially available after 2001.

NEW VOICES IN CHEMISTRY

MARCOS DANTUS

LASER CONTROL OF CHEMICAL REACTIONS

Scientific and technological breakthroughs breathe renewed life into area of study

*"Smart photons, from ultrafast shaped pulses, could revolutionize techniques such as **laser machining**, laser deposition, and laser desorption."*

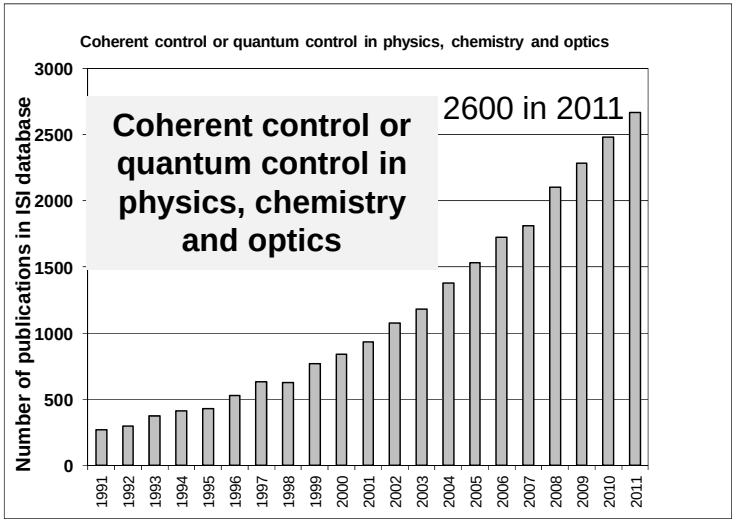
*"The ultimate goal of this research is to produce an **automated protein-sequencing** apparatus."*

Also accomplished in early 2000's

- Automated pulse compression
- Coherent communications with fs pulses
- Functional imaging to detect cancer
- Detection of warfare agents

M. Dantus, Chemical & Engineering News **79**, 191-191 (2001)

Outline:



1. Introduction

2. Progress in Quantum Control

a. Wave packet manipulation
b. Interference between laser pulses and wave packets
c. Selective two-photon excitation, CARS, SRS, electronic and vibrational control

3. Challenges in Chemistry

a. Chemical reactions
b. Coherent Control of Electrons, Electronic States and Electronic Transitions
c. Breaking strong chemical bonds while leaving weak ones intact
d. Determining the role of coherent quantum processes in nature

4. Future Outlook and Applications

a. Phase Control of Advanced Sources
b. Quantum control-based molecular sensing and diagnostics
c. Material Synthesis and Material Processing
d. Connecting the Macro-Micro-and-Nano Worlds
e. Coherent control and communications and information processing. Internet is incoherent

A Brief History of Quantum Phase Control

- Young's double-slit experiment (1803), although it is just linear interference

$$P \propto \left| e^{i\phi_1} + e^{i\phi_2} \right|^2 \propto \cos\left(\phi_1 - \phi_2\right)$$

- Brumer and Shapiro propose one vs. three-photon quantum control (1986)

$$P \propto \left| e^{i\phi_3} + e^{i3\phi_1} \right|^2 \propto \cos\left(\phi_3 - 3\phi_1\right)$$ (One final $3\omega_0$ narrow frequency)

- Weiner demonstrates Optical Code Division Multiple Access (CDMA) (1988, 1990, 1998)

(using inversion symmetry of binary phase codes to maximize SHG at $2\omega_0$)

- Gordon Experimental demonstration of one vs. three-photon control (1997)

- Silberberg demonstrates multi-chromatic two-photon control (1998)

$$P\left(\Delta\right) \propto \left| \int e^{i\left[\phi(\omega-\Delta)-\phi(\omega+\Delta)\right]} d\omega \right|^2$$ (One final Δ narrow frequency)

$$P\left(2\omega_0\right) = \left| \int e^{i\left[\phi(\omega_0-\Delta)+\phi(\omega_0+\Delta)\right]} d\Delta \right|^2$$ (One final narrow frequency $2\omega_0$)

- Weinacht, Ahn and Bucksbaum "write-read" on sculpted Rydberg wave packet (1998)

MII for other nonlinear processes

Two-photon excitation

$$E^{(2)}(2\omega) \propto \int_{-\infty}^{\infty} \left|E(\omega+\Omega)\right|\left|E(\omega-\Omega)\right| \exp\left[i\left\{\varphi(\omega+\Omega)+\varphi(\omega-\Omega)\right\}\right] d\Omega.$$

N-photon excitation

$$E^{(n)}(n\omega) \propto \int_{-\infty}^{\infty} \ldots \int_{-\infty}^{\infty} d\Omega_1 \ldots d\Omega_{n-1} \left|E(\omega+\Omega_1)\right| \ldots \left|E(\omega+\Omega_{n-1})\right| \left|E(\omega-\Omega_1)\right| \ldots \left|E(\omega-\Omega_{n-1})\right| \times$$
$$\exp\left\{i\left[\varphi(\omega+\Omega_1)+\ldots+\varphi(\omega+\Omega_{n-1})+\varphi(\omega-\Omega_1)\ldots+\varphi(\omega-\Omega_{n-1})\right]\right\}$$

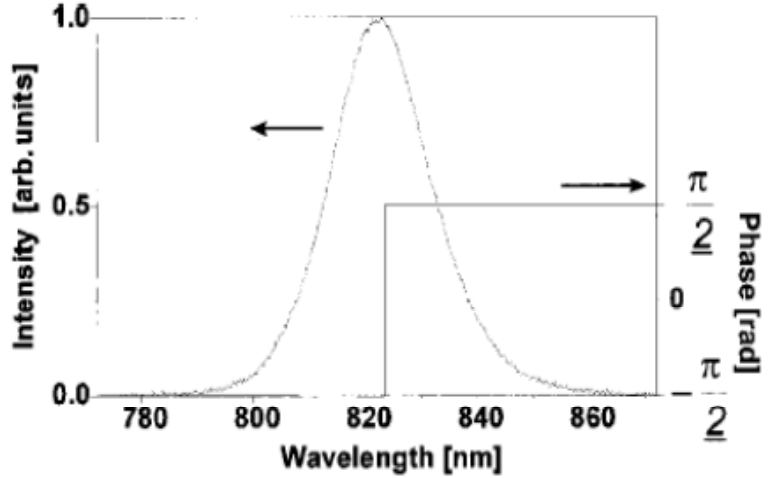
Selective two-photon excitation, SHG or SRS using Binary Phases

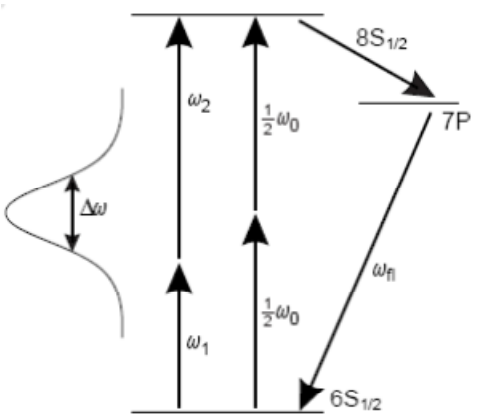
$$E^{(2)}(2\omega) \propto \int_{-\infty}^{\infty} E(\omega+\Omega)E(\omega-\Omega)d\Omega = \int_{-\infty}^{\infty} \exp\left[i\left\{\varphi(\omega+\Omega)\right\}\right] \exp\left[i\left\{\varphi(\omega-\Omega)\right\}\right] d\Omega = \sum E_i E_j$$

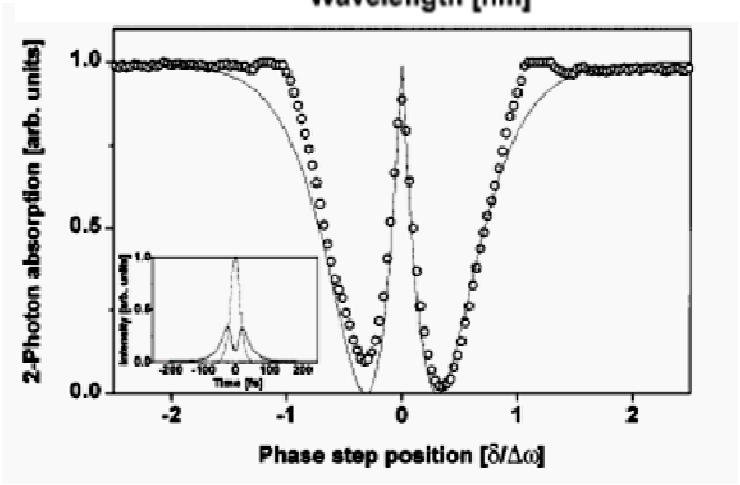
± 1

J.Phys. Chem. A, 106, 9369, 2002. J.Chem. Phys. **118**, 3187 (2003) Opt. Express, 12, 1061 (2004). ChemPhysChem, 6, 1952 (2005), Physical Review A **74**, 041805(R) (2006)

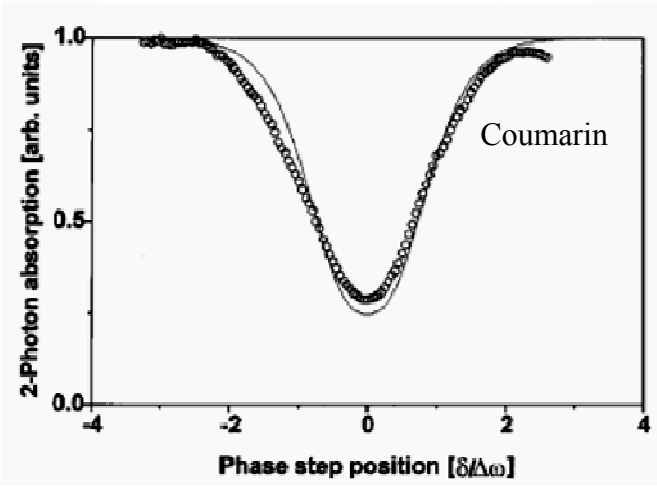
"*An atomic switch!*" (P. Bucksbaum, Nature)







Two-photon excitation of Cs atoms vs. position of a step phase function



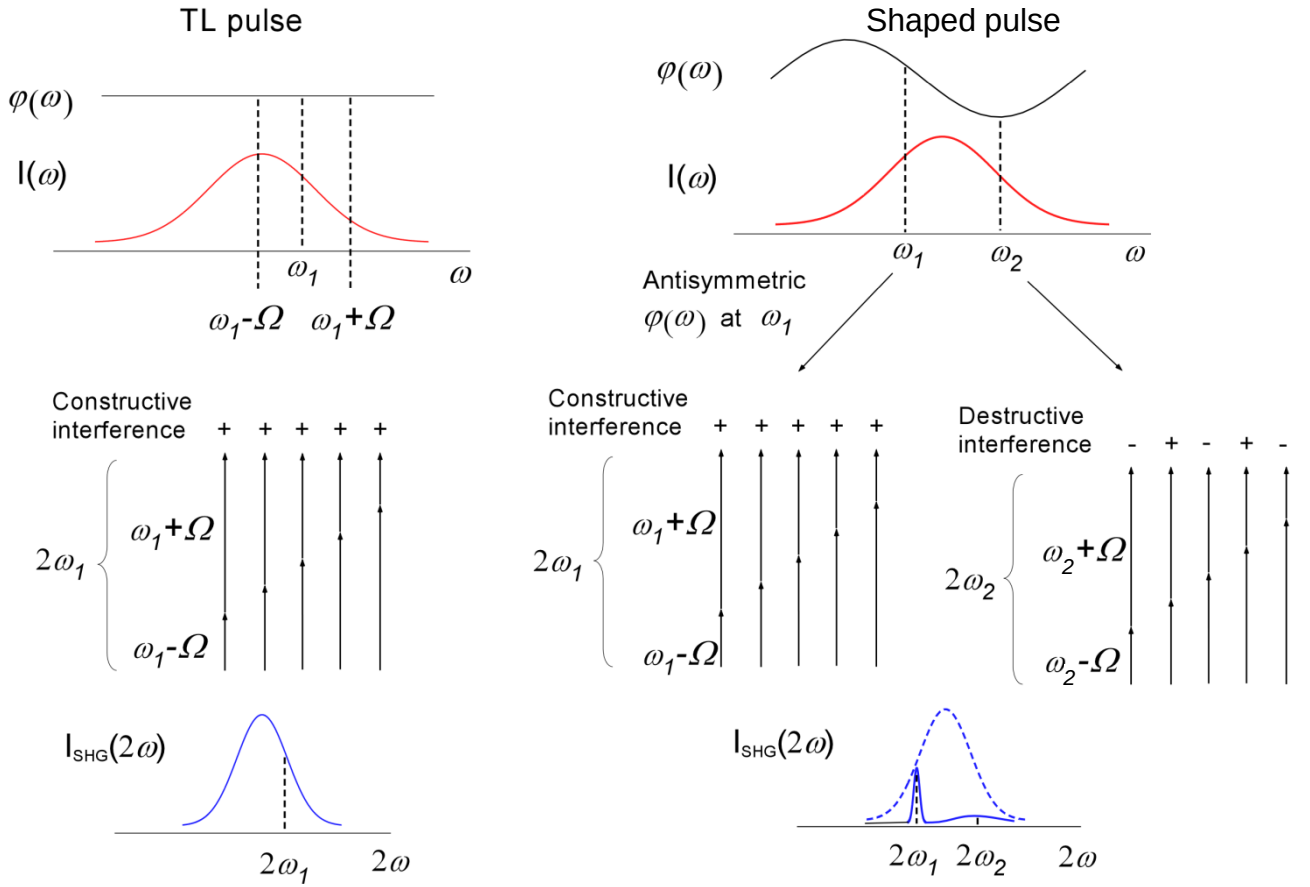
Two-photon excitation vs. position of a step phase function. No enhancement observed at 0 detuning.

D. Meshulach, Y. Silberberg, *Nature* 1998, 396, 239-242

Lozovoy and Dantus multichromatic multifrequency n-photon control (2002-)

Multiphoton Intrapulse Interference

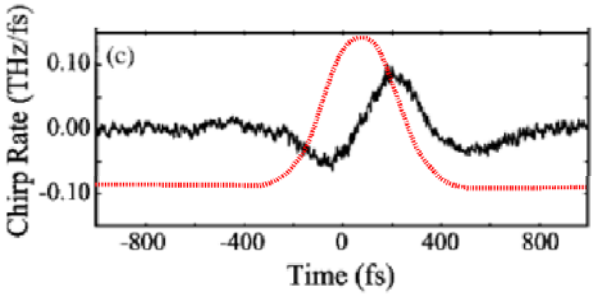
$$I_{SHG}(2\omega) \propto \left| \int \left| E_0(\omega+\Omega) \right| \left| E_0(\omega-\Omega) \right| \exp\left[i\left(\varphi(\omega+\Omega) + \varphi(\omega-\Omega) \right) \right] d\Omega \right|^2$$



J.Phys. Chem. A, 106, 9369, 2002. J.Chem. Phys. **118**, 3187 (2003) Opt. Express, 12, 1061 (2004). ChemPhysChem, 6, 1952 (2005), Physical Review A **74**, 041805(R) (2006)

Brief Review of Pulse Shaping Technology

Time Domain



Directly shape $E(t)$

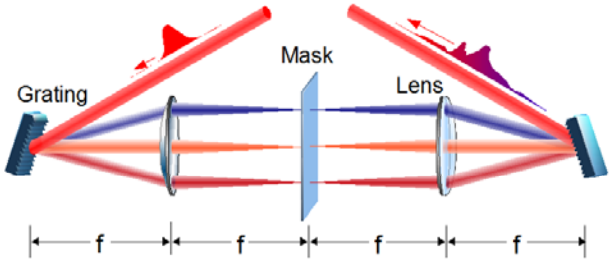
using ultrafast processes

Stavola, Agostinelli, Sceats, *Appl. Opt.* **18**, 4101 (1979)

J. G. Fujimoto and E. P. Ippen, *Opt. Lett.* **8**, 446 (1983)

Bartels,Weinacht,Wagner,BaertschyGreen, MurnaneKapteyn, *Phys. Rev. Lett.* **88**, 013903 (2002)

Frequency Domain



$$E(t) \xrightarrow{\text{Fourier Transform}} E(\omega)$$

Shape

$$E_{out}(\omega) \xrightarrow{\text{Inverse Transform}} E_{out}(t)$$

Desbois, Gires, Tournois, *IEEE J. Quantum Electron*.**QE-9**, 213 (1973)

Agostinelli, Harvey, Stone, Gabel, *Appl. Opt.* **18**, 2500 (1979)

Froehly, *J. Opt. (Paris),* **12**, 25 (1981) "The Fourier Plane"

Weiner and Heritage, *Rev. Phys. Appl.* **22** 1619 (1987)

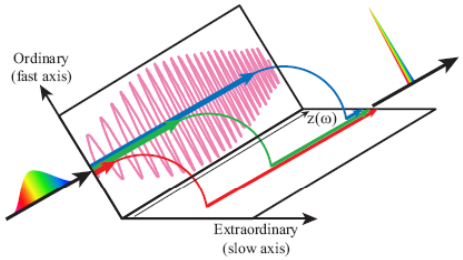
Weiner, Leaird, Patel, Wullert, *Opt. Lett.* **15**, 326 (1990)

Wefers, Nelson *Opt. Lett. **20**, 1047*(1995)

Commercially available:

Biophotonic Solutions Inc.

Freq. and Time Domain



Shape a combination of

$E(t)$ and $E(\omega)$

Haner and Warren, *Appl. Phys. Lett.* **52**, 1458 (1988)

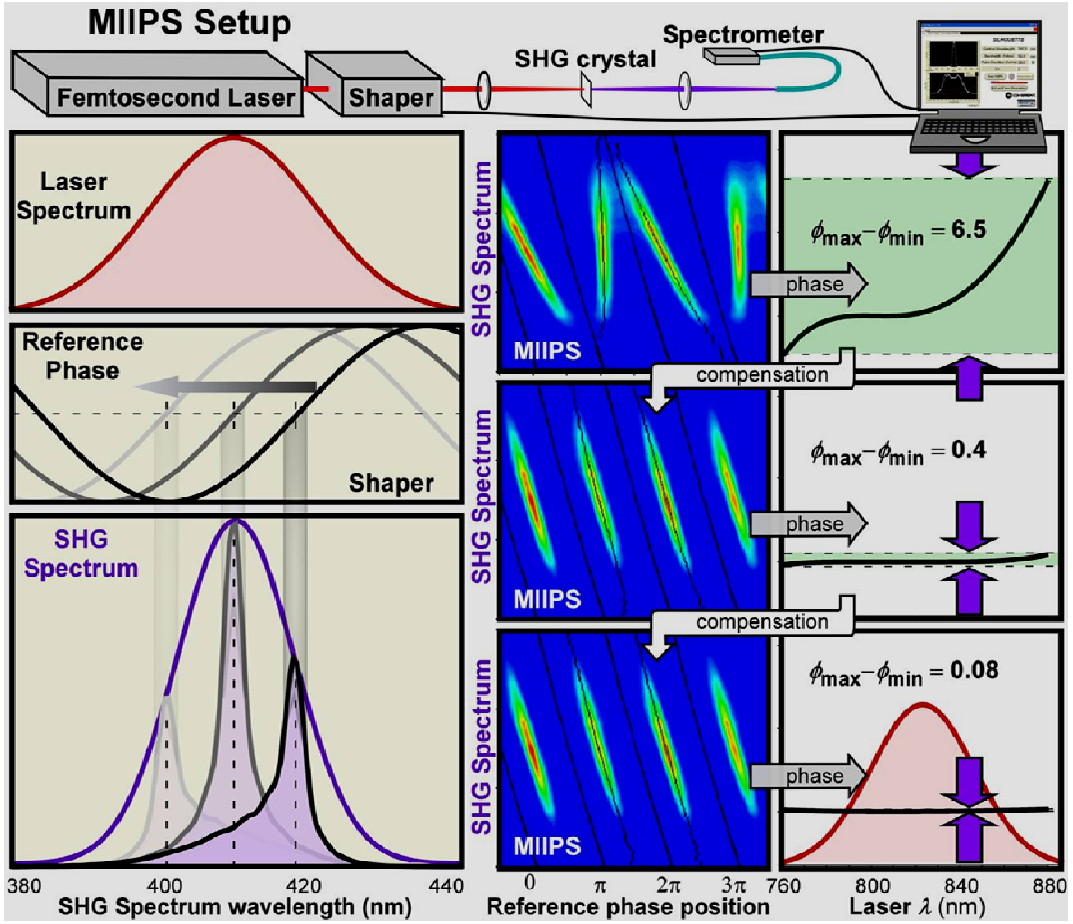
Fermann, da Silva, Smith, Silberberg, Weiner, *Opt. Lett.* **18**, 1505 (1993)

Verluise, Laude, Cheng, Spielmann, Tournois, *Opt. Lett.* **25**, 575 (2000)

Commercially available:

Fastlite

Multiphoton Intrapulse Interference Phase Scan (MIIPS)



Measures

Corrects

Done!

Opt. Lett. 29, 775 (2004)

JOSA B 23, 750 (2006)

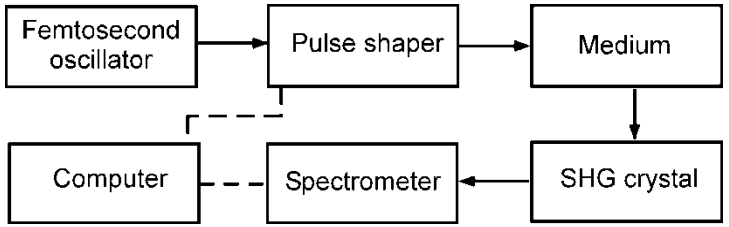
Laser Focus World 43, 101, (2007)

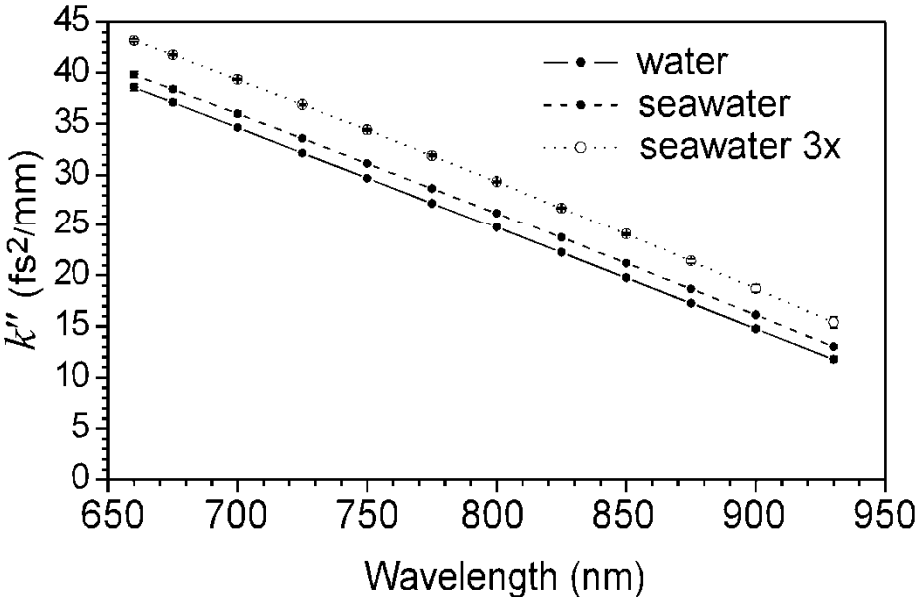
JOSA B 25, A140 (2008)

MIIPS based techniques and their applications are protected by 8 issued and 25 patents pending.

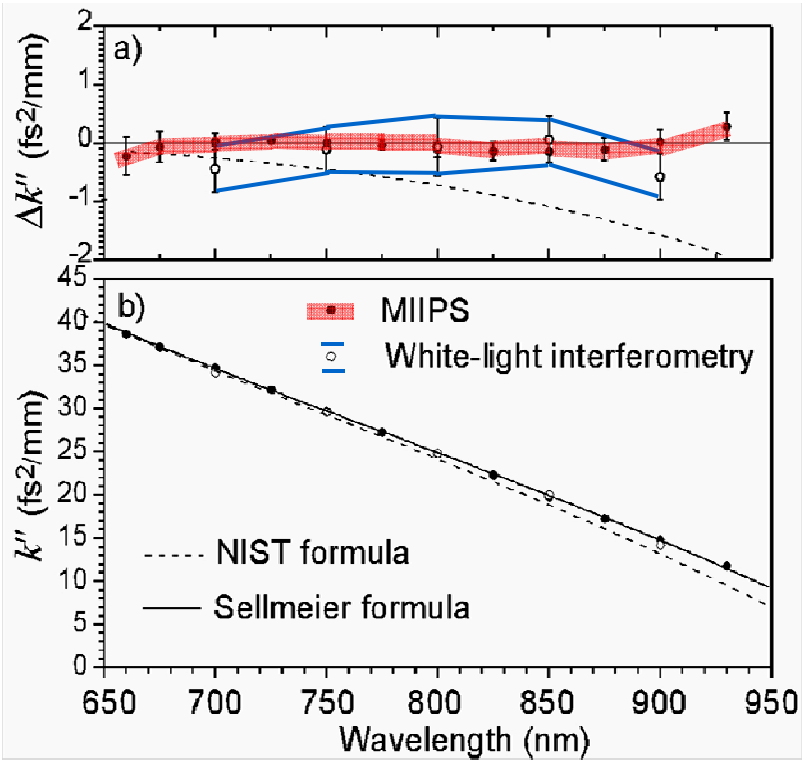
Multiphoton Intrapulse Interference Phase Scan (MIIPS)

GVD Measurements With ±0.1 fs^2 Accuracy and Precision





Dispersion of water



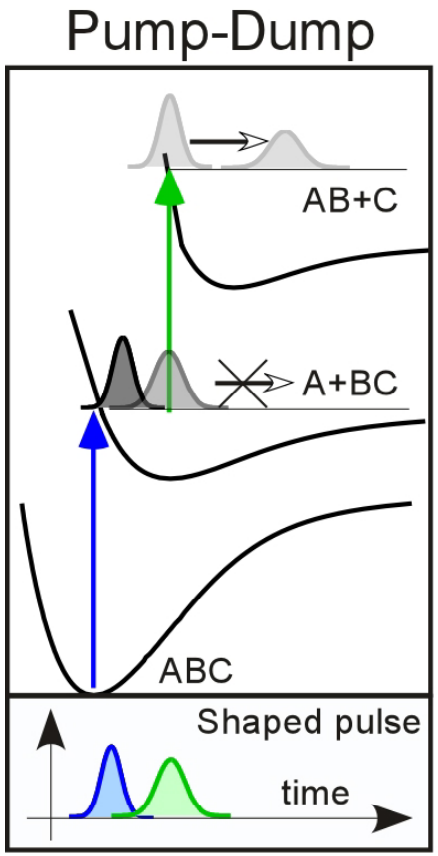
Coello *et al.*, *Applied Optics* **46**, 8394 (2007), Water and Sea Water

Devi, et al. *AIP Advances* **1**, 032166 (2011), Common Solvents

Wrzesinski, et al. *Opt. Express* **19**, 5163 (2011), Atmospheric and Combustion Gases

2. Progress in Quantum Control

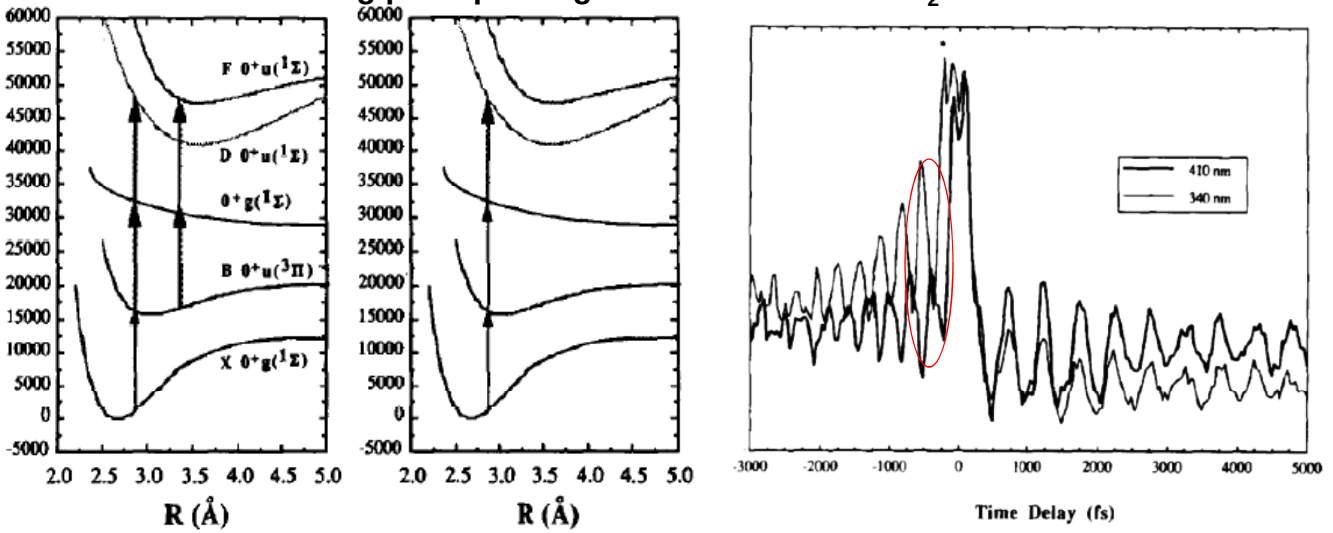
a. Wave Packet Manipulation and Role of Homogeneous and Inhomogeneous Broadening



Rice, Tannor, Kosloff
Dantus, Zewail, Gerber,
1985-88

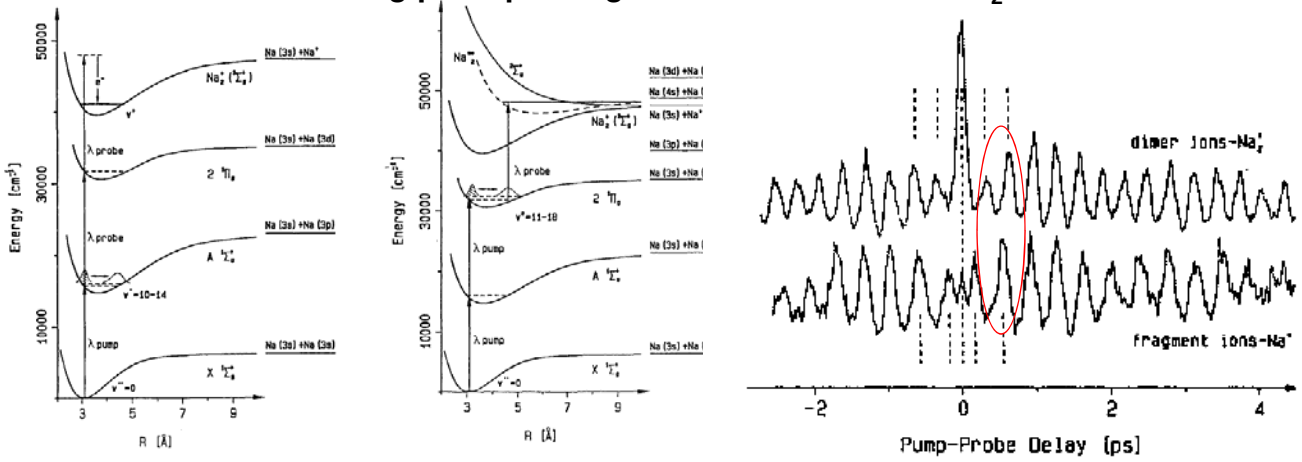
Time domain approach,
intuitive results.

1-photon pump 2-photon probe
Inner vs. outer turning point probing to different states of I_2



Bowman Dantus Zewail, Chem. Phys. Lett. 174, 546 (1990)

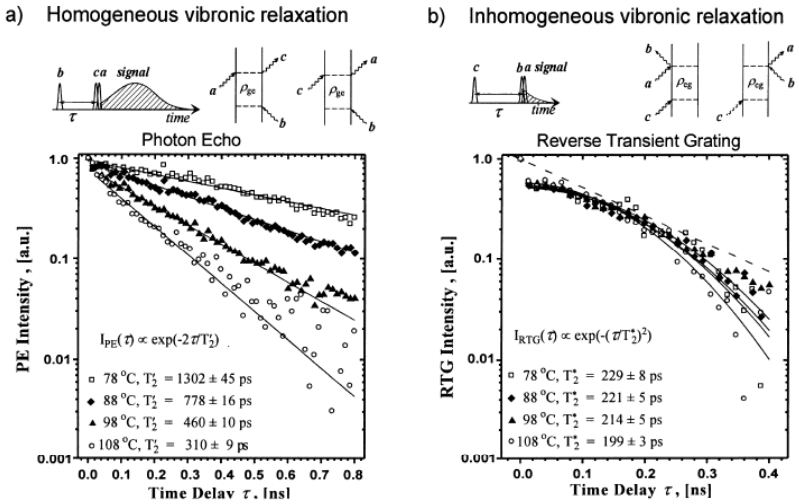
2-photon pump1-photon probe
Inner vs. outer turning point probing to different states of Na_2

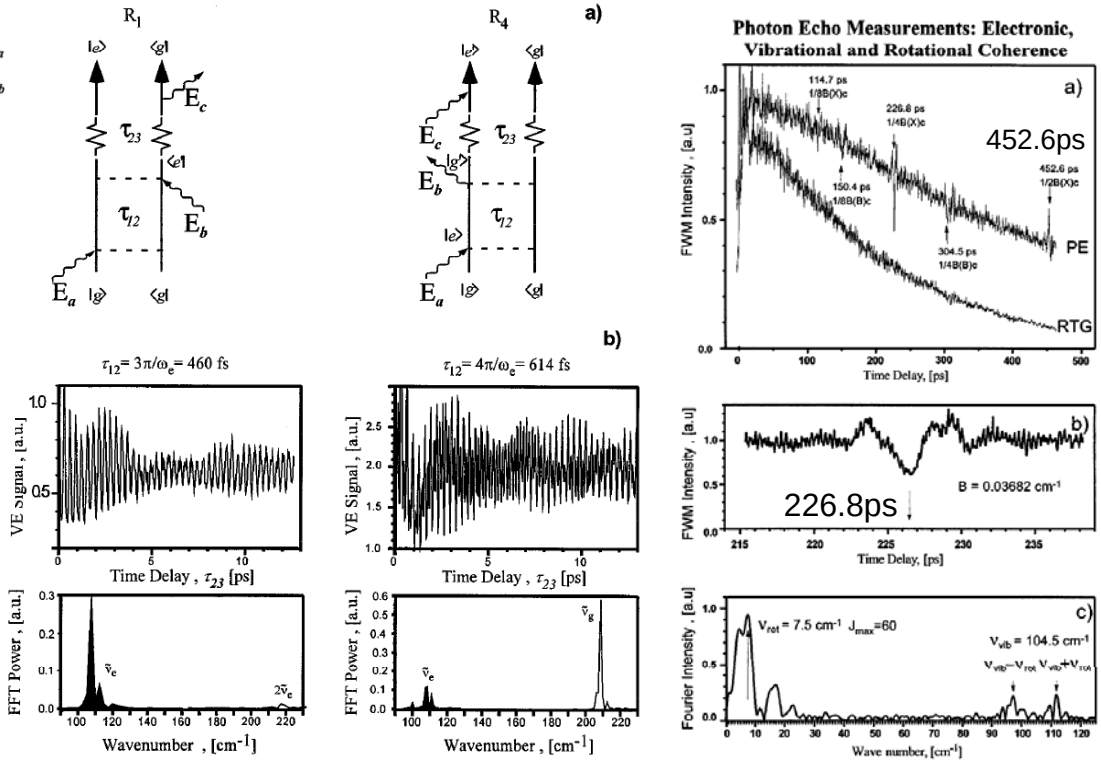


Na^+ vs Na_2^+ Baumert, Gerber, Isr.J.Chem. 34, 103 (1994)

2. Progress in Quantum Control

a. Control of ground and excited state coherence cancelling inhomogeneous broadening at room temp.



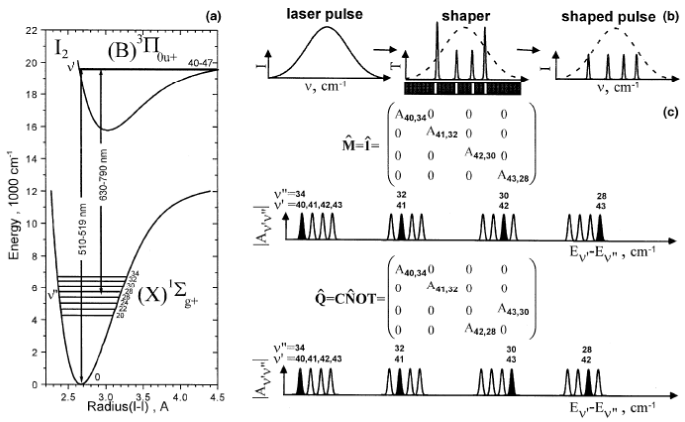


Pastirk et al. Chem.Phys.Letts. **333**, 76 (2001)

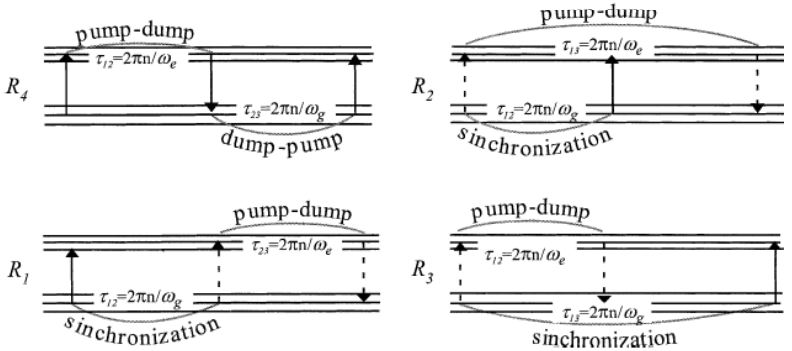
Brown et al. J. Chem. Phys. **111**, 3779 (1999)

Dantus, Annu. Rev. Phys. Chem. **52**, 639 (2001)

Lozovoy Dantus Chem.Phys.Letts. **351**, 213 (2002)



Intra- and intermolecular coherence, the role of local fields

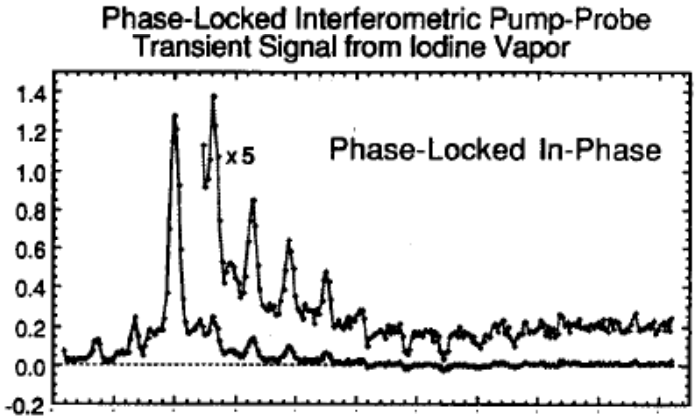


Lozovoy et al, J. Chem. Phys. 267, 99 (2001)

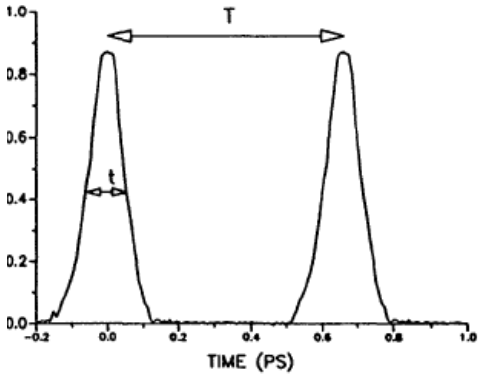
2. Progress in Quantum Control

b. Interference Between Laser Pulses and Wave Packets

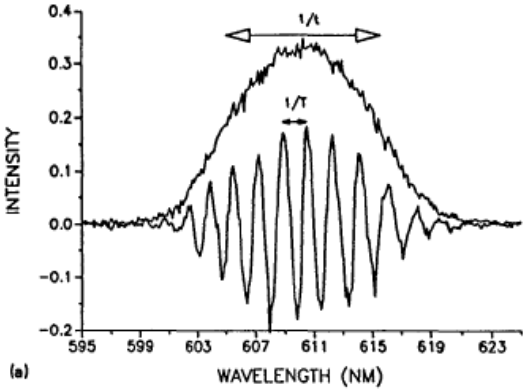
Scherer et al. JCP 93, 856 (1990), JCP 95, 1487 (1991)



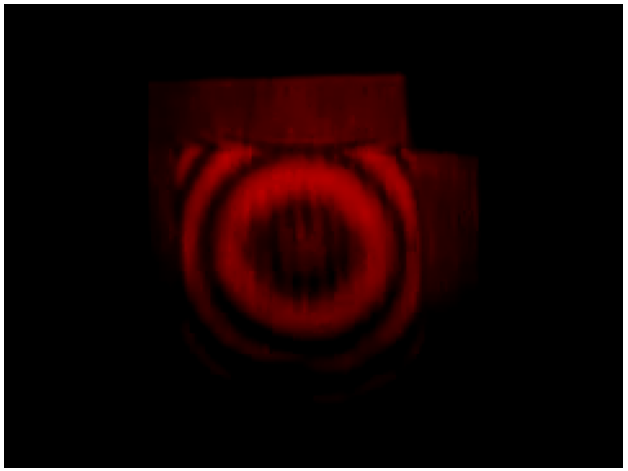
The two optical pulses interfere



The spectrum is modulated

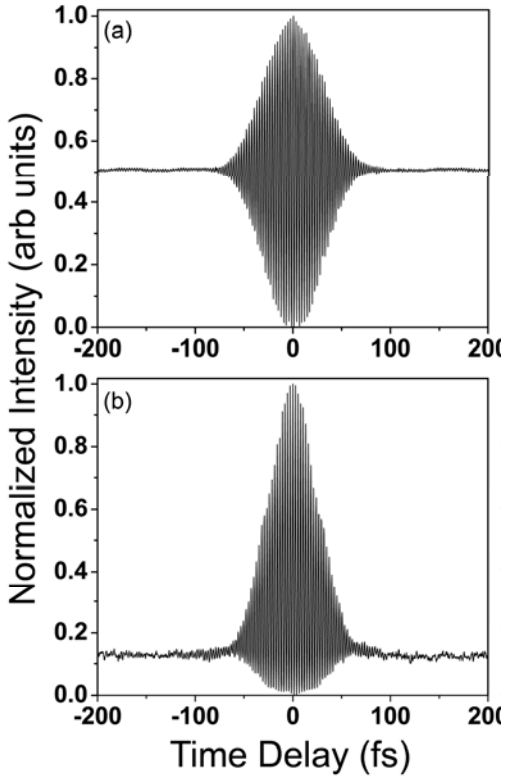


Michelson Interferometer



1st order autocorrelation

2nd order autocorrelation

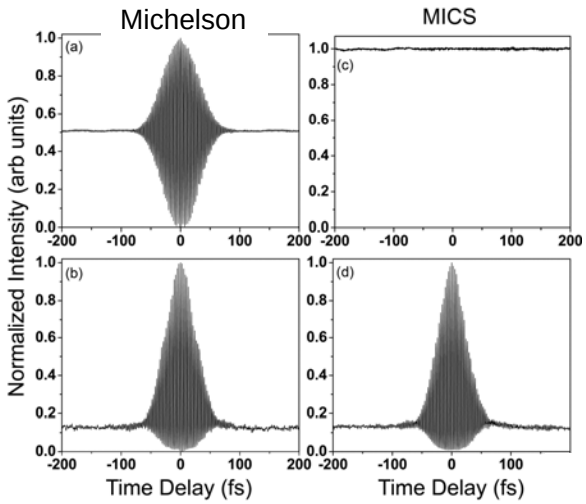


2. Progress in Quantum Control

b. Interference Between Laser Pulses and Wave Packets

Multiple independent comb shaping (MICS)

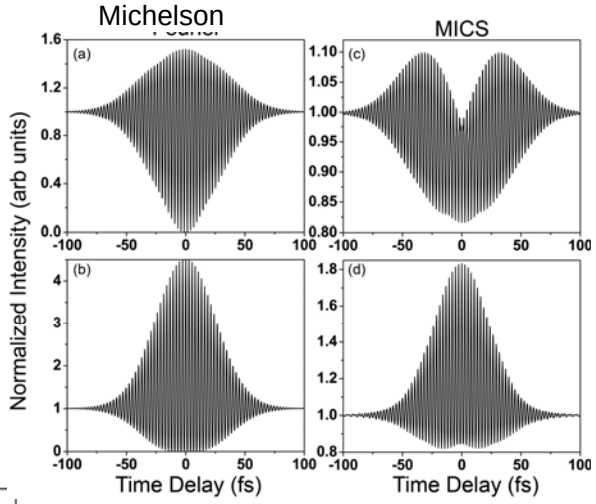
Avoiding linear interference by pulse shaping



MICS: Pestov et al. *Opt. Exp.* 17, 14351 (2009)

IR144: Konar et al. *J. Phys. Chem.* (2012)

Theory

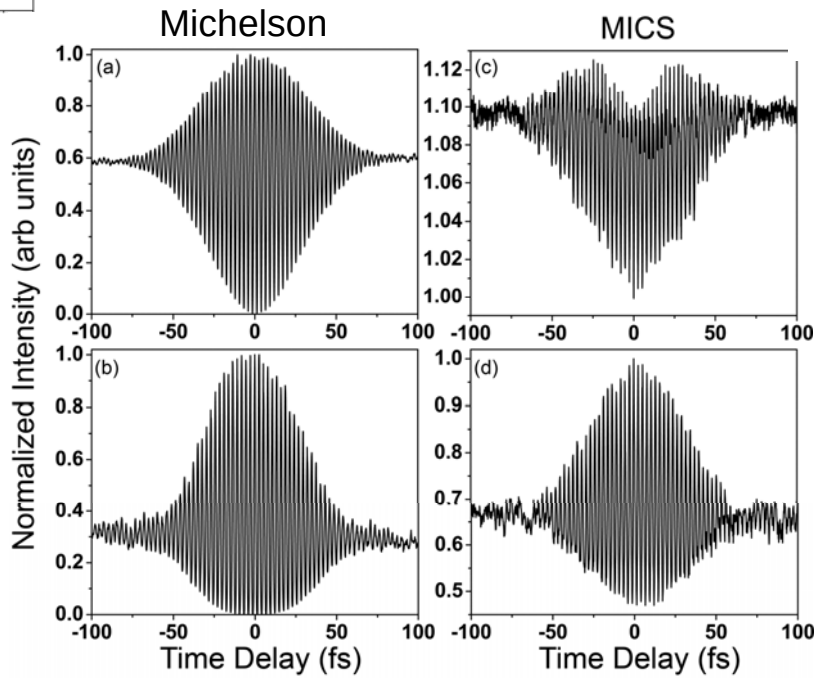


Experimental

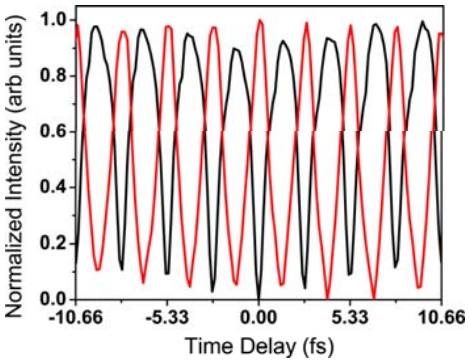
IR144 in solution

Fluorescence

Stim. Emission



Stim. Emission

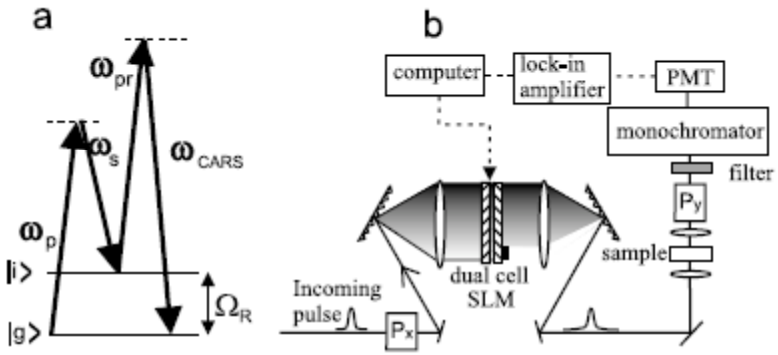


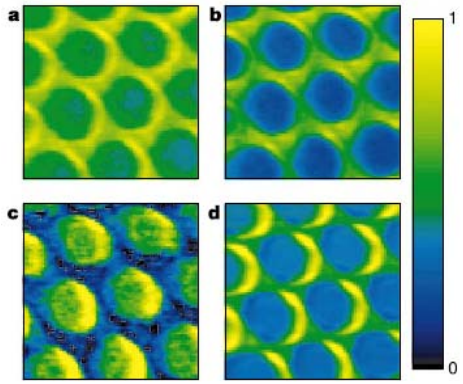
Fluorescence

2. Progress in Quantum Control

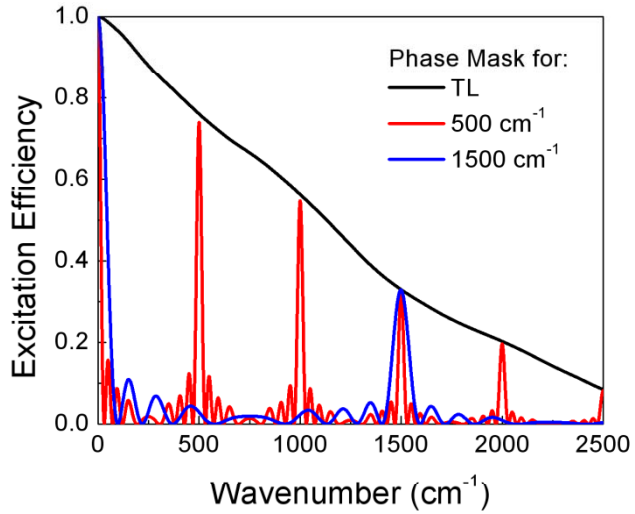
c. Selective Two-Photon Excitation, CARS, SRS, Electronic and Vibrational Control

Single-pulse CARS with selective vibrational mode excitation: Dudovich Oron Silberberg Nature 418, 512 (2002)

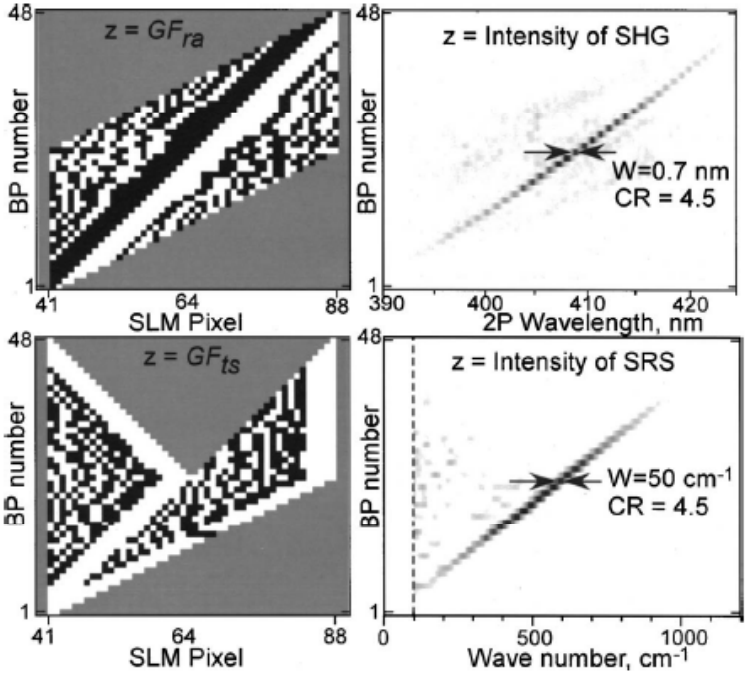




Sinusoidal modulation

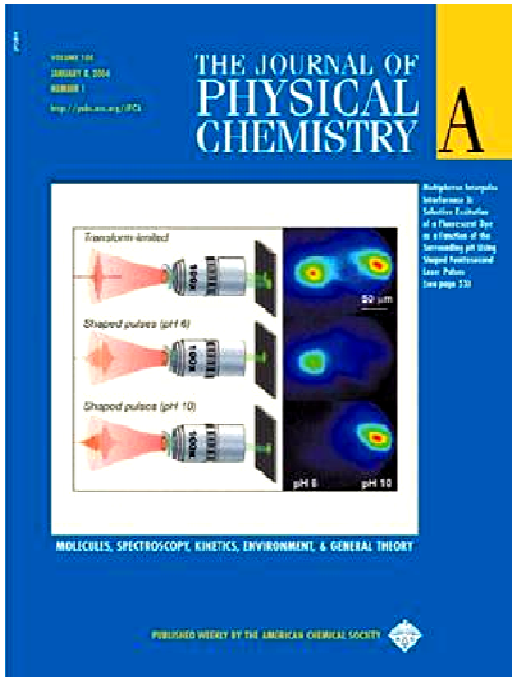


Binary Phase Modulation: Physical Review A **74**, 041805(R) (2006)



2. Progress in Quantum Control

c. Selective Two-Photon Excitation, CARS, SRS, Electronic and Vibrational Control

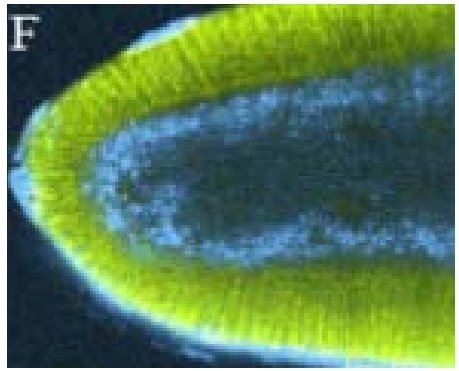


J. Phys. Chem. 108, 53 2004

Selective two-photon Microscopy
Optics Express 11, 1695 (2003)

Live fluorescently labeled *Drosphila* embryos

Jennifer P. Ogilvie et al.
Optics Express 14, (2006)

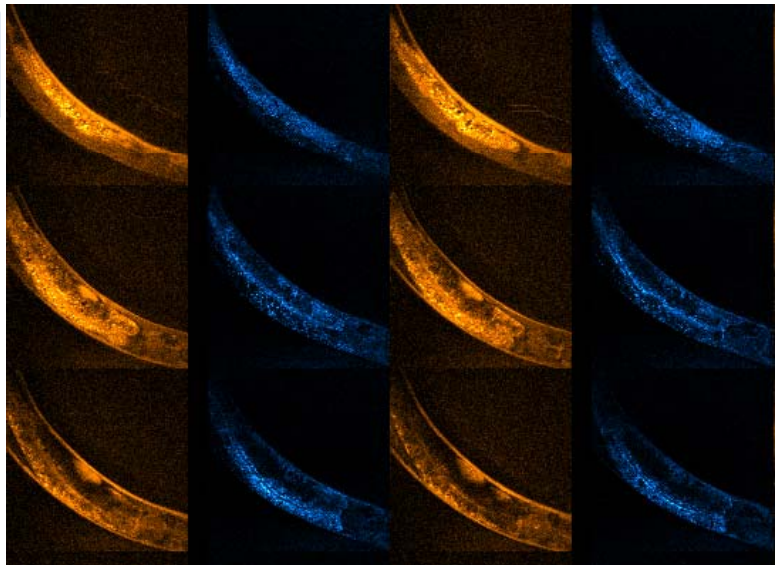


nature photonics
ARTICLES
PUBLISHED ONLINE: 16 JANUARY 2011 | DOI: 10.1038/NPHOTON.2010.294

Highly specific label-free molecular imaging with spectrally tailored excitation-stimulated Raman scattering (STE-SRS) microscopy

Christian W. Freudiger[1,2], Wei Min[2], Gary R. Holtom[2], Bingwei Xu[3], Marcos Dantus[4] and X. Sunney Xie[2]*

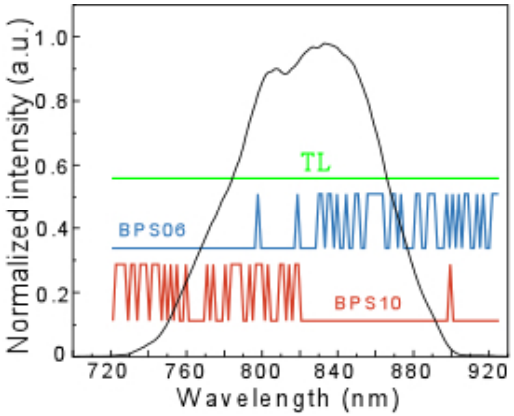
Nature Photonics, 294 (2010)



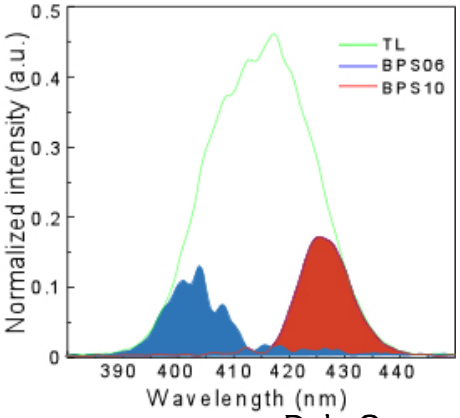
2. Progress in Quantum Control

c. Selective Two-Photon Excitation, CARS, SRS, Electronic and Vibrational Control

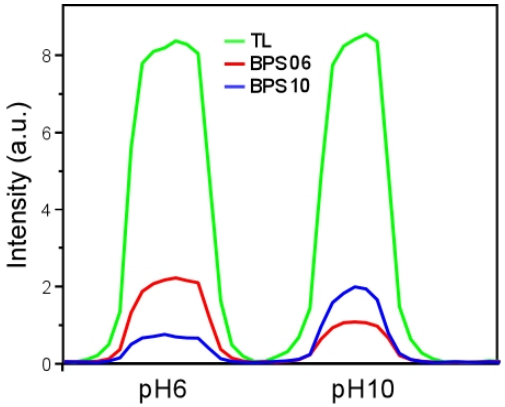
Binary pulse shaping



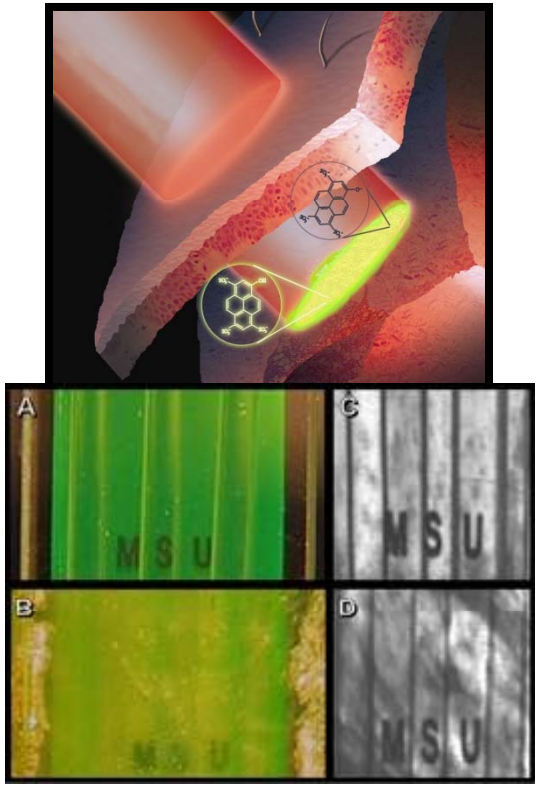
Testing SHG spectra



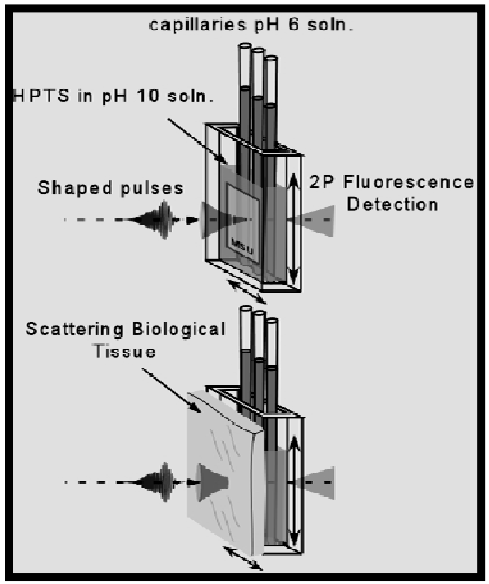
Testing selective excitation



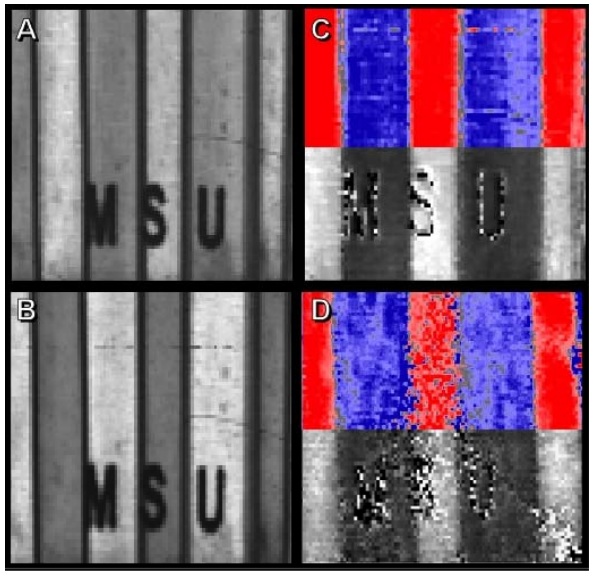
Dela Cruz et al. *Opt. Express*, **12**, 4144 (2004)



Experimental Setup



Quantum Control-Based Functional Imaging Through Biological Tissue

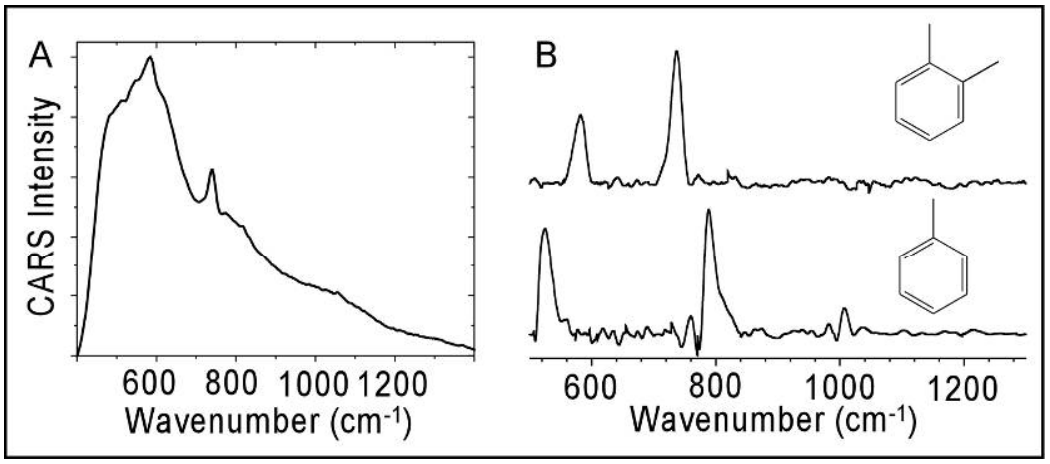


Dela Cruz et al. *PNAS* **101,** 16996 (2004)

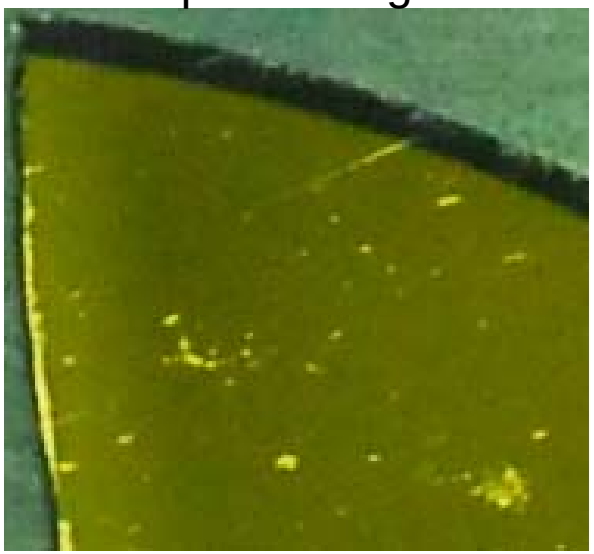
2. Progress in Quantum Control

c. Selective Two-Photon Excitation, CARS, SRS, Electronic and Vibrational Control

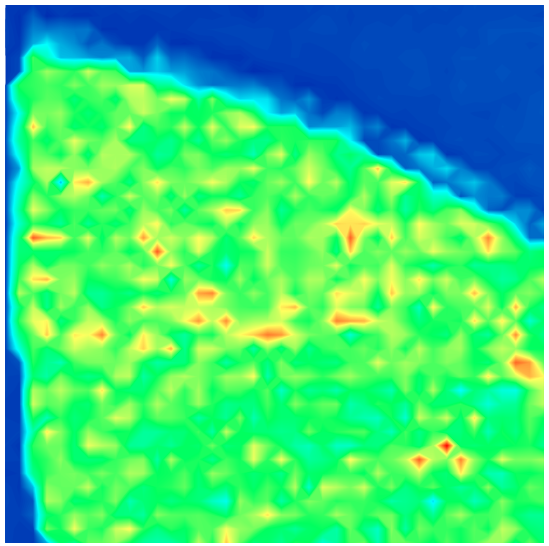
Data obtained from a scattering surface at 12m standoff distance, 10μJ/pulse, ~100μm neat liquid film ~10mg/cm^2 Li, Harris, Wrzesinski, Xu, Lozovoy and Dantus, Opt. Express, 16, 592 (2008)



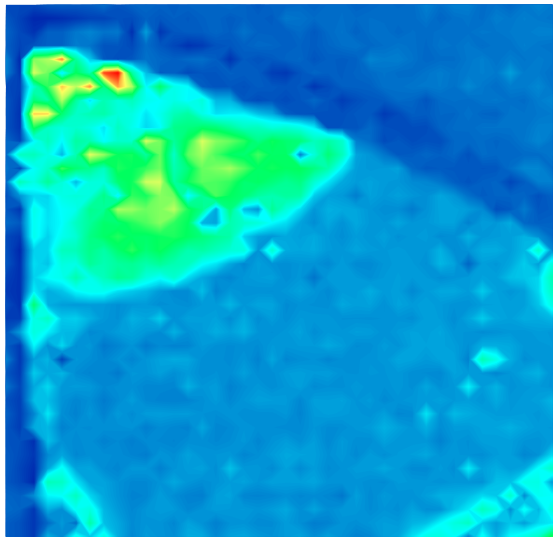
Optical Image



Selective CARS for PS



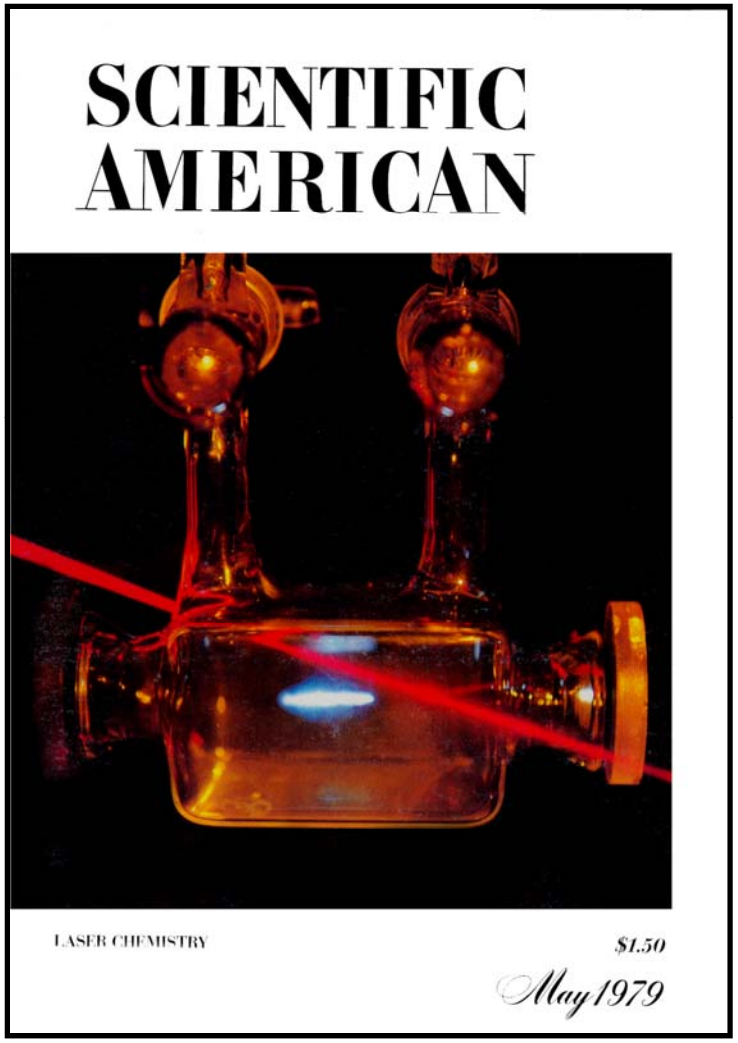
Selective CARS for DNT



"Highly Selective Standoff Detection and Imaging of Trace Chemicals in a Complex Background using Single-Beam Coherent Anti-Stokes Raman Scattering",
App. Phys. Letters **99**, 101109 (2011)

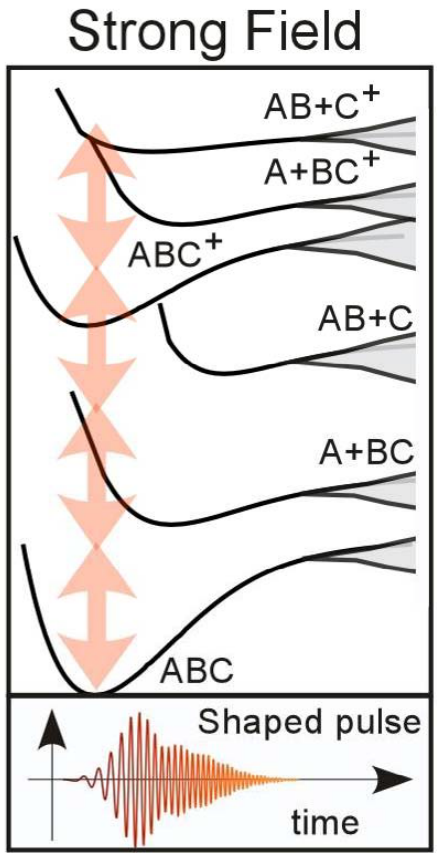
3. Challenges in Chemistry

a. Chemical Reactions



LASER CHEMISTRY
J. Chem. Educ. 43, 566 (1966)

LASER CHEMISTRY
SCI. AMER. 240, 114 (1979)



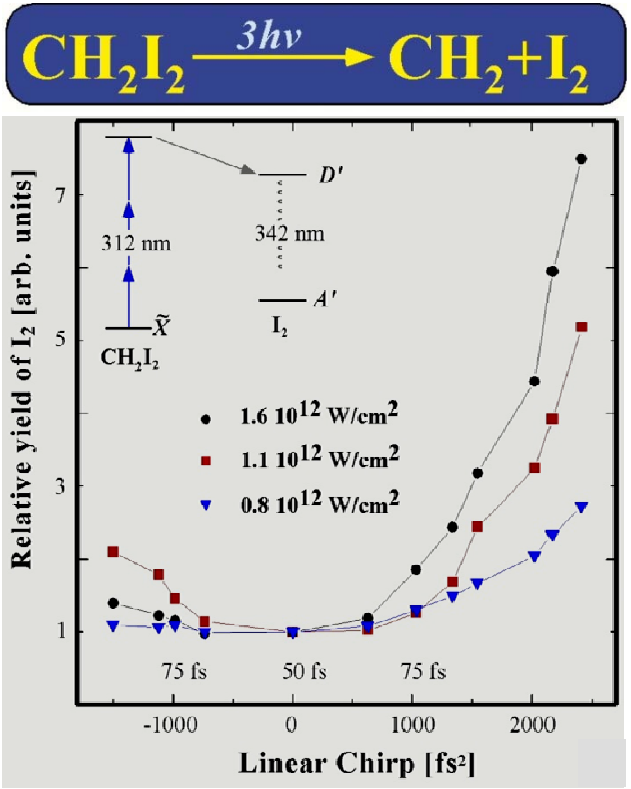
Kosloff, Rice, Gaspard, Tersigni, and Tannor propose achieving chemical selectivity by shaping of intense femtosecond pulses, **introduce an iteration procedure for optimization of the pulse shapes** (1989).

Judson and Rabitz propose using **a learning algorithm** to control molecules (1992)

Assion, Baumert, Bergt, Brixner, Kiefer, Seyfried, Strehle, Gerber Control of chemical reactions by feedback-optimized phase shaped laser pulses (1998)

3. Challenges in Coherent Control

a. Chemical Reactions

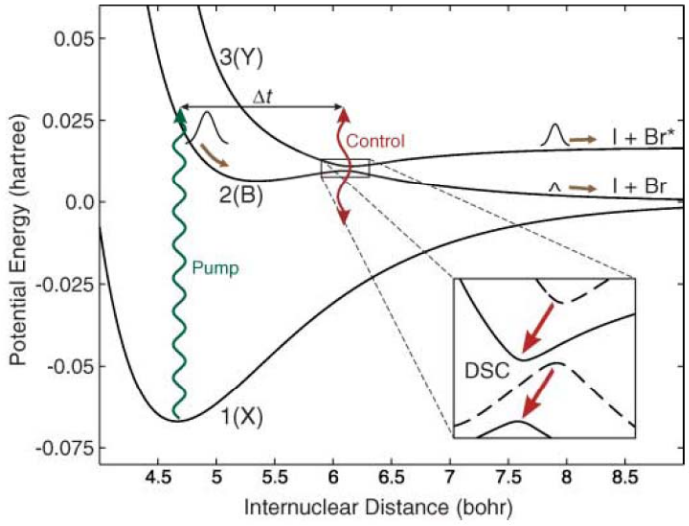


Pastirk and Dantus JCP 108, 4375 (1998)

Quantum control of a concerted chemical reaction

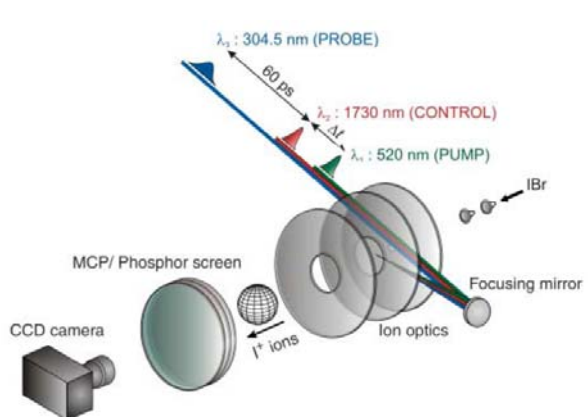
Dynamic Stark Control of Photochemical Process

Sussman et al. Science 314, 278 (2006)

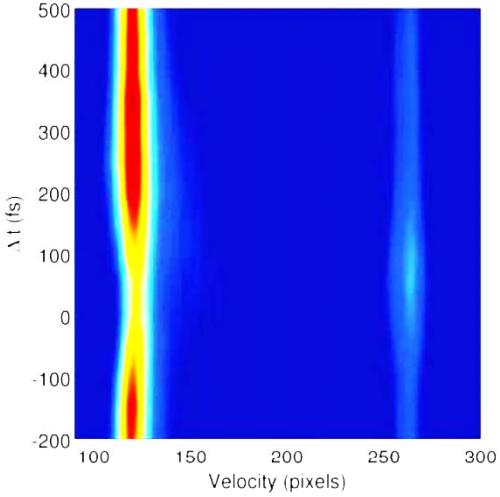


I+Br*

I+Br



I+Br* I+Br

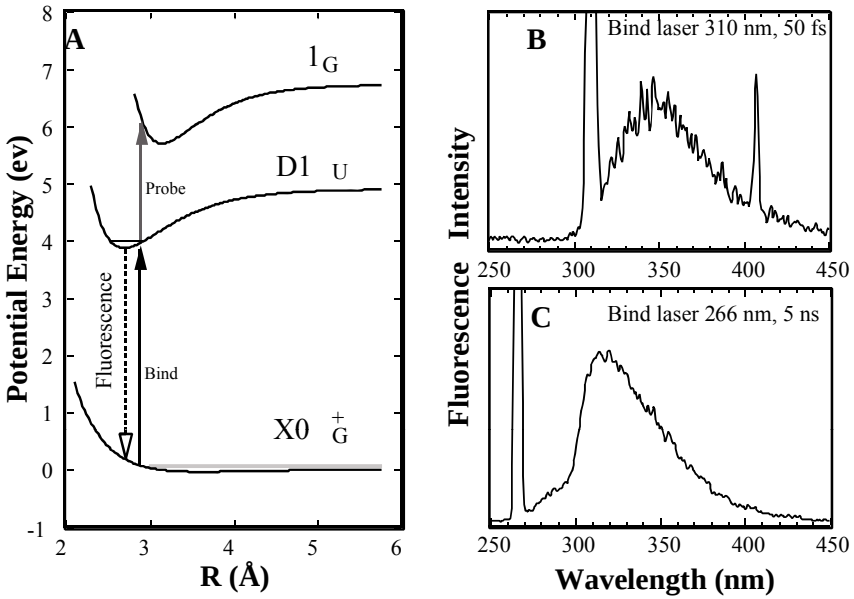


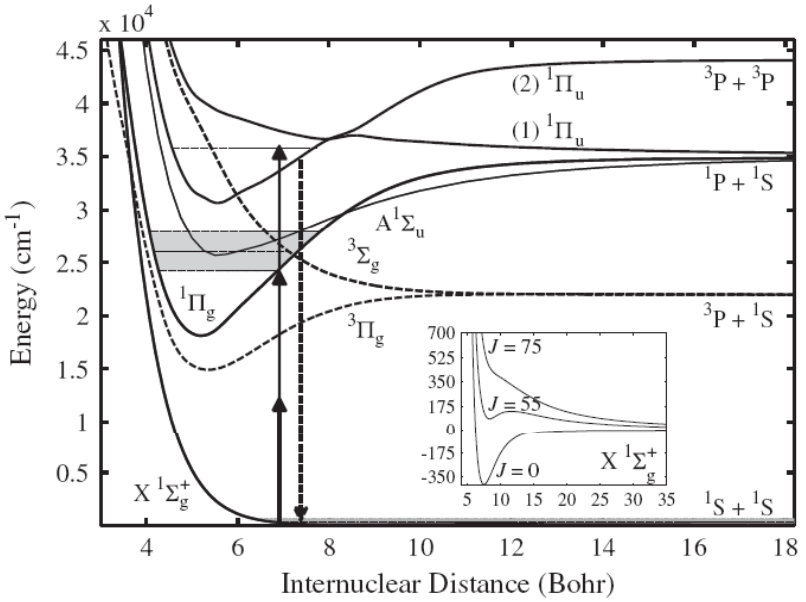
3. Challenges in Coherent Control

a. Chemical Reactions

iii. **Bimolecular reactions**

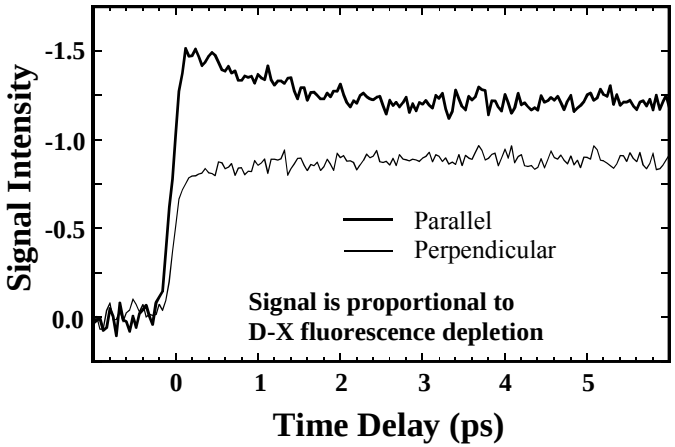
From atoms to molecules …





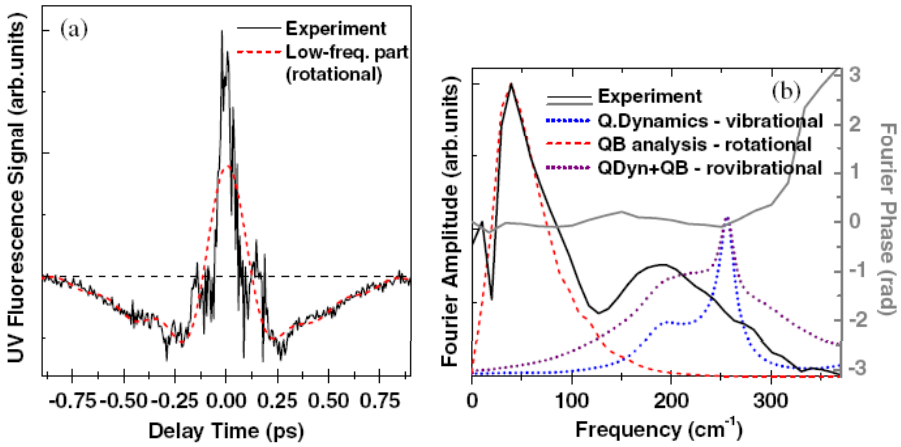
Femtosecond Photoassociation

$$Hg + Hg \xrightarrow{h\nu} Hg_2^*$$



Marvet et al. Chem. Phys. Lett. 245, 393 (1995)

Two-photon photoassociation of hot Mg atoms



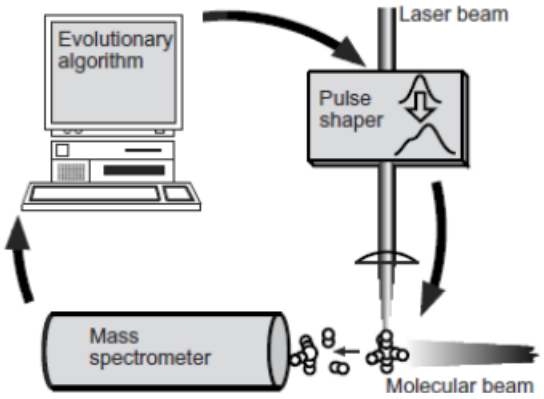
Rybak et al. PRL 107, 273001 (2011)

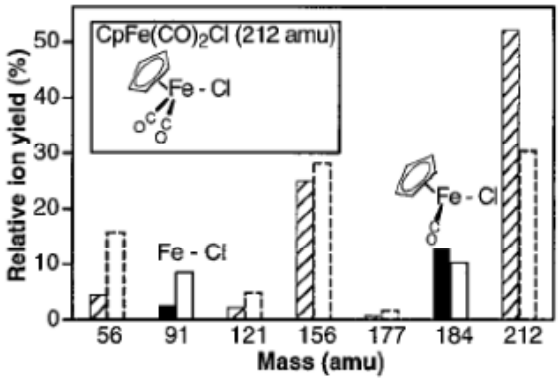
3. Challenges in Coherent Control

a. Chemical Reactions

i. **A mol is a very large quantity**

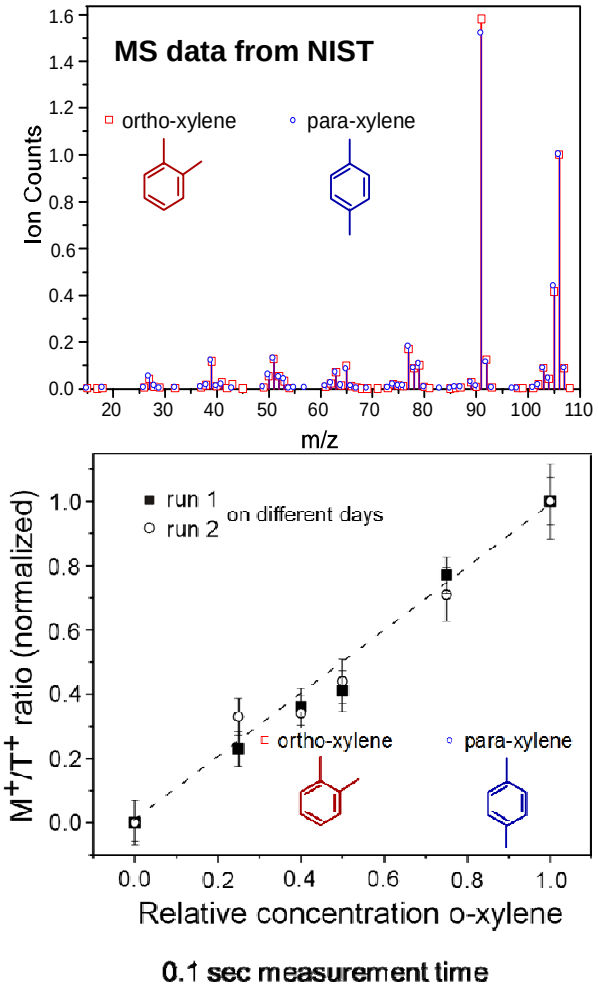
ii. **Coherent vs. Incoherent**





Science **282**, 919 (1998)

Analytical Chemistry



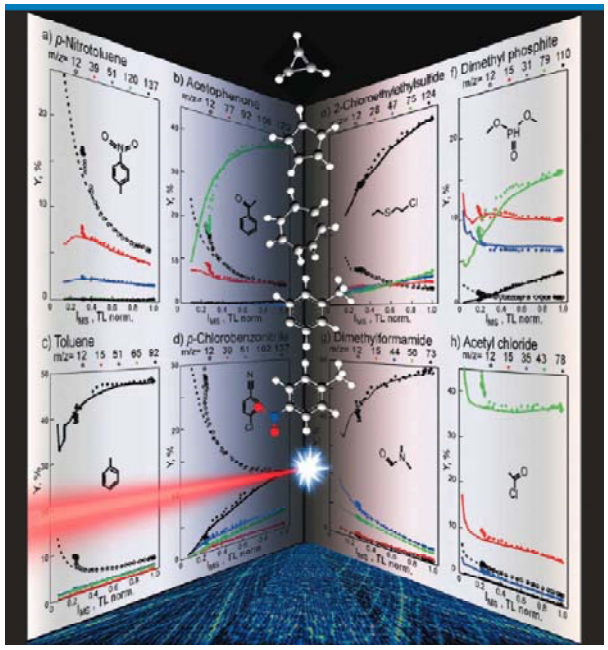
0.1 sec measurement time

Pastirk et al. J. Phys. Chem. A 109,2413 (2005)

Dela Cruz et al. J. Phys. Chem. A. 109, 8447 (2005)

Ann. Rep. Prog. Chem. C 102, 227 (2005)

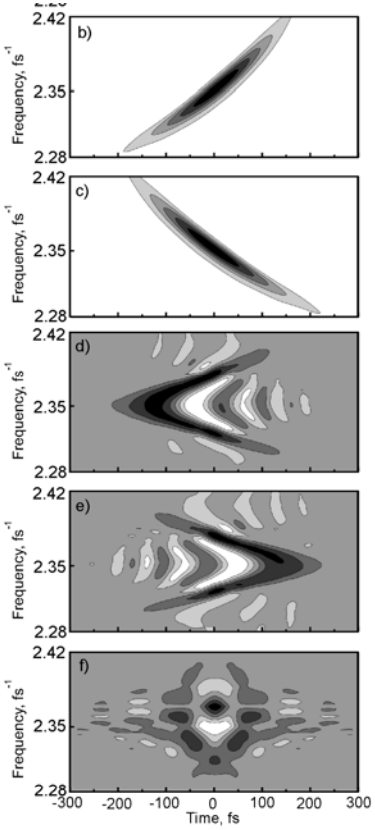
Either no coherence or very short lived.

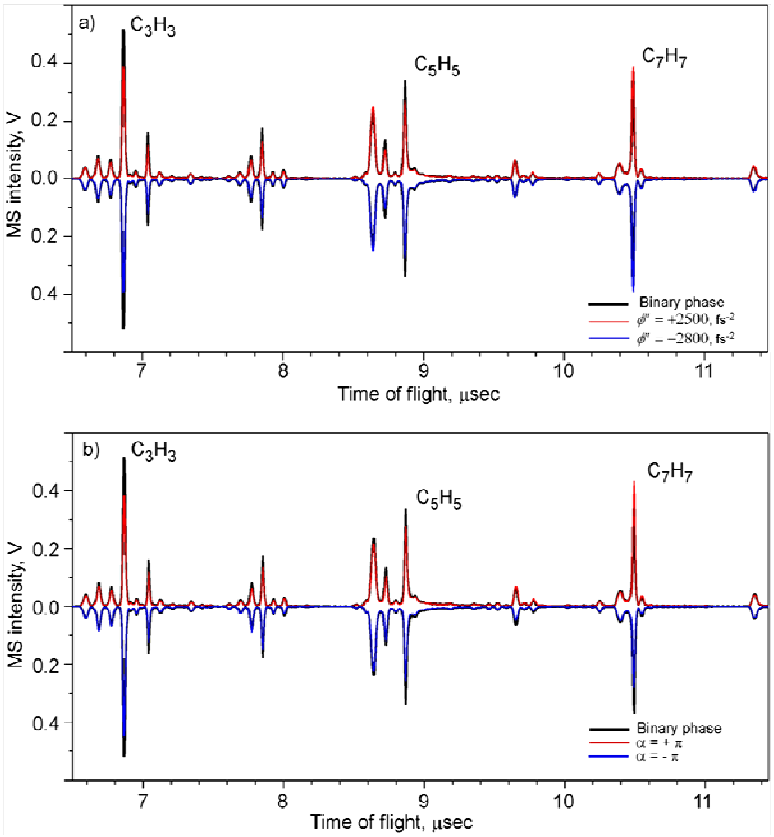


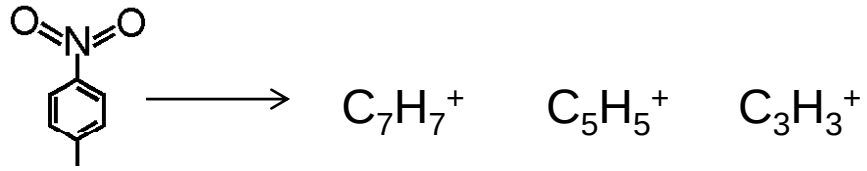
J. Phys. Chem. A. 112, 3789 (2008)

Is photofragmentation pattern phase dependent?

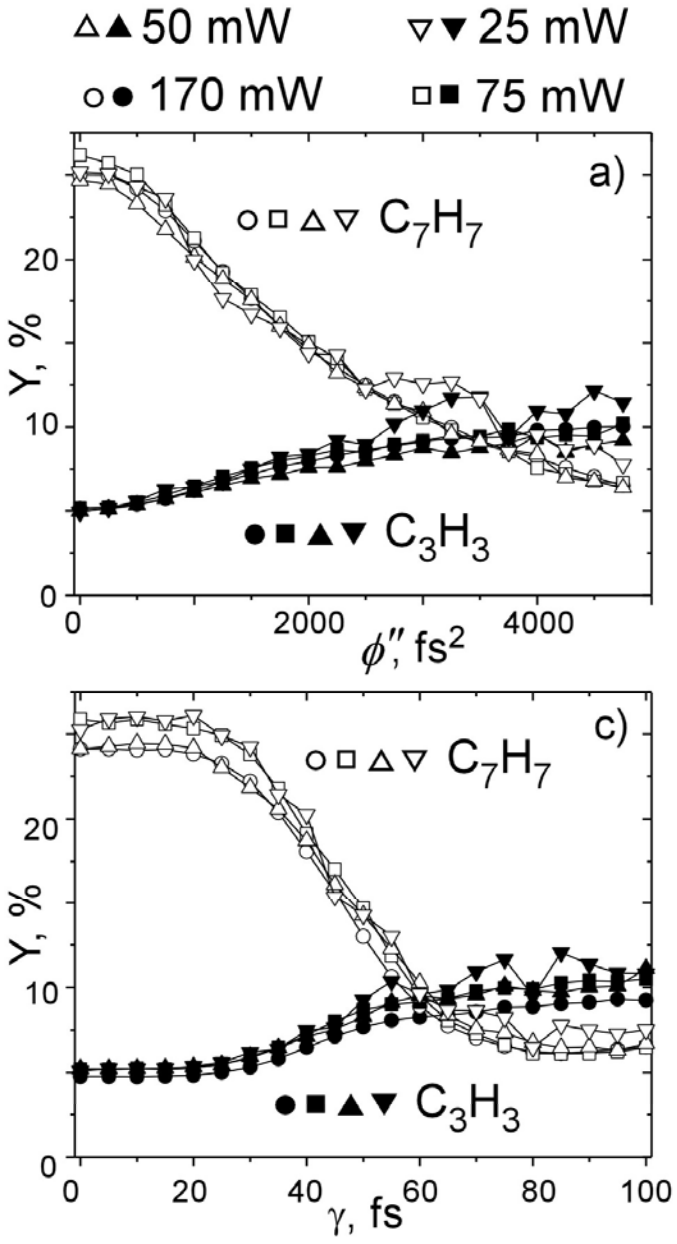
Comprehensive exploration on the role of phase coherence on the fragmentation of large molecules by intense shaped pulses







$C_7H_7^+$ $C_5H_5^+$ $C_3H_3^+$

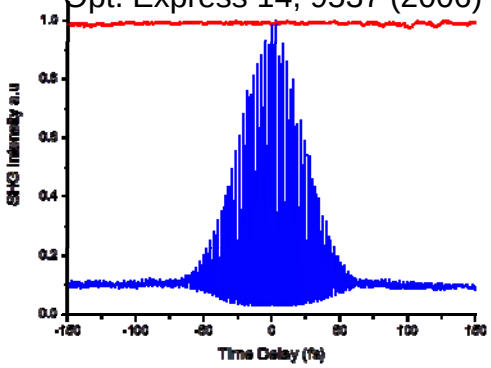


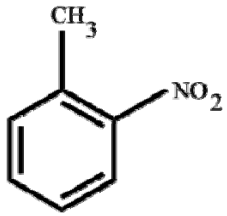
Zhu et al. J. Phys. Chem. A. 112, 3789 (2008)

3. Challenges in Coherent Control

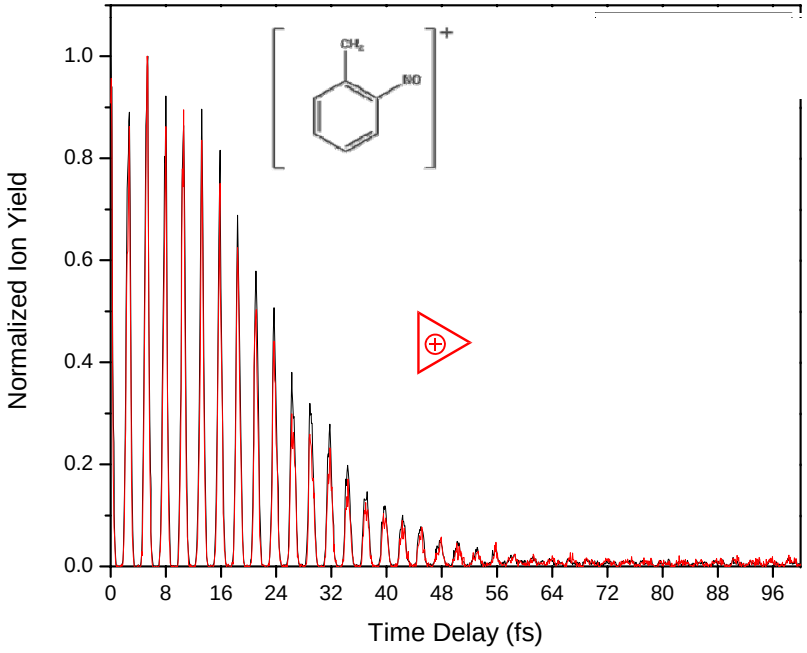
b. Coherent Control of Electrons, Electronic States and Electronic Transitions

Opt. Express 14, 9537 (2006)

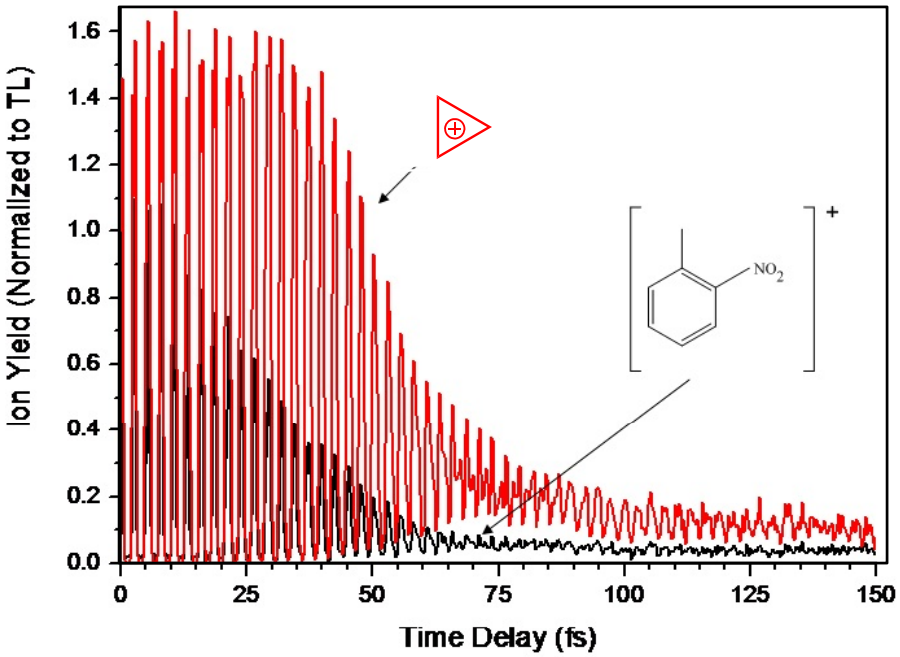




ortho-Nitrotoluene



Peak Intensity = 8×10^{13} W/cm^2

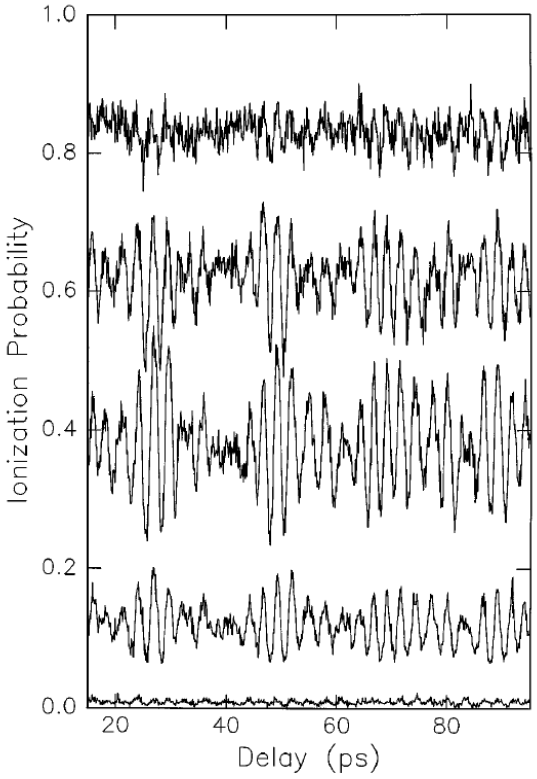


Peak Intensity = 5×10^{14} W/cm^2

3. Challenges in Coherent Control

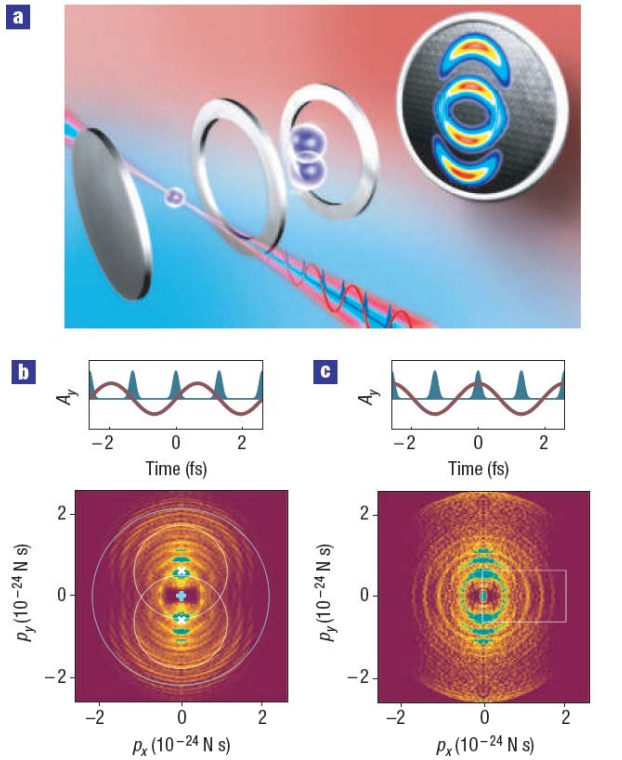
b. Coherent Control of Electrons, Electronic States and Electronic Transitions

Electronic Wave Packets using ½-cycle pulses

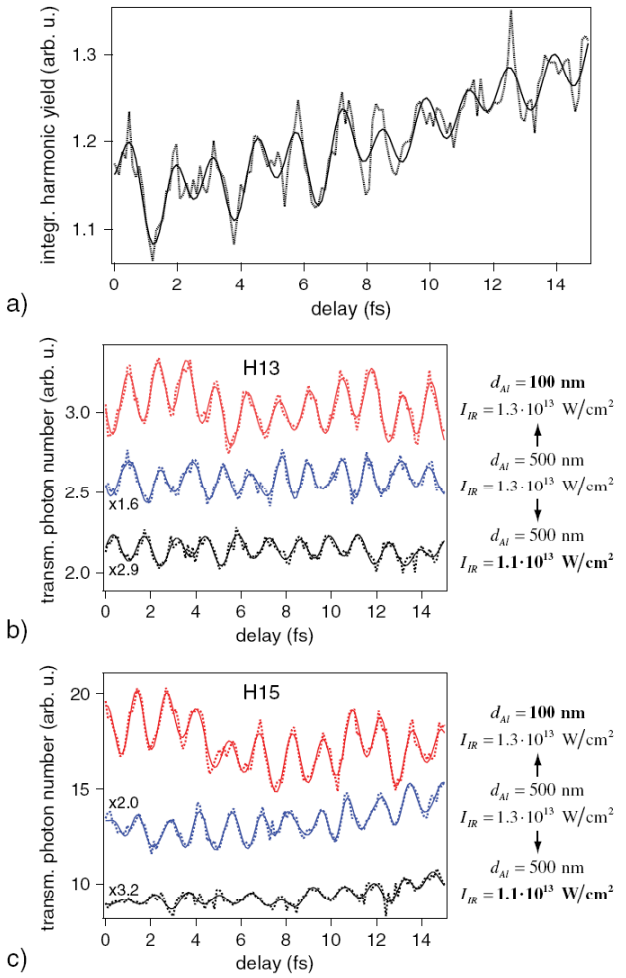


R. R. Jones, PRL 76, 3927 (1996)

Attosecond electronic wave packet interferometry



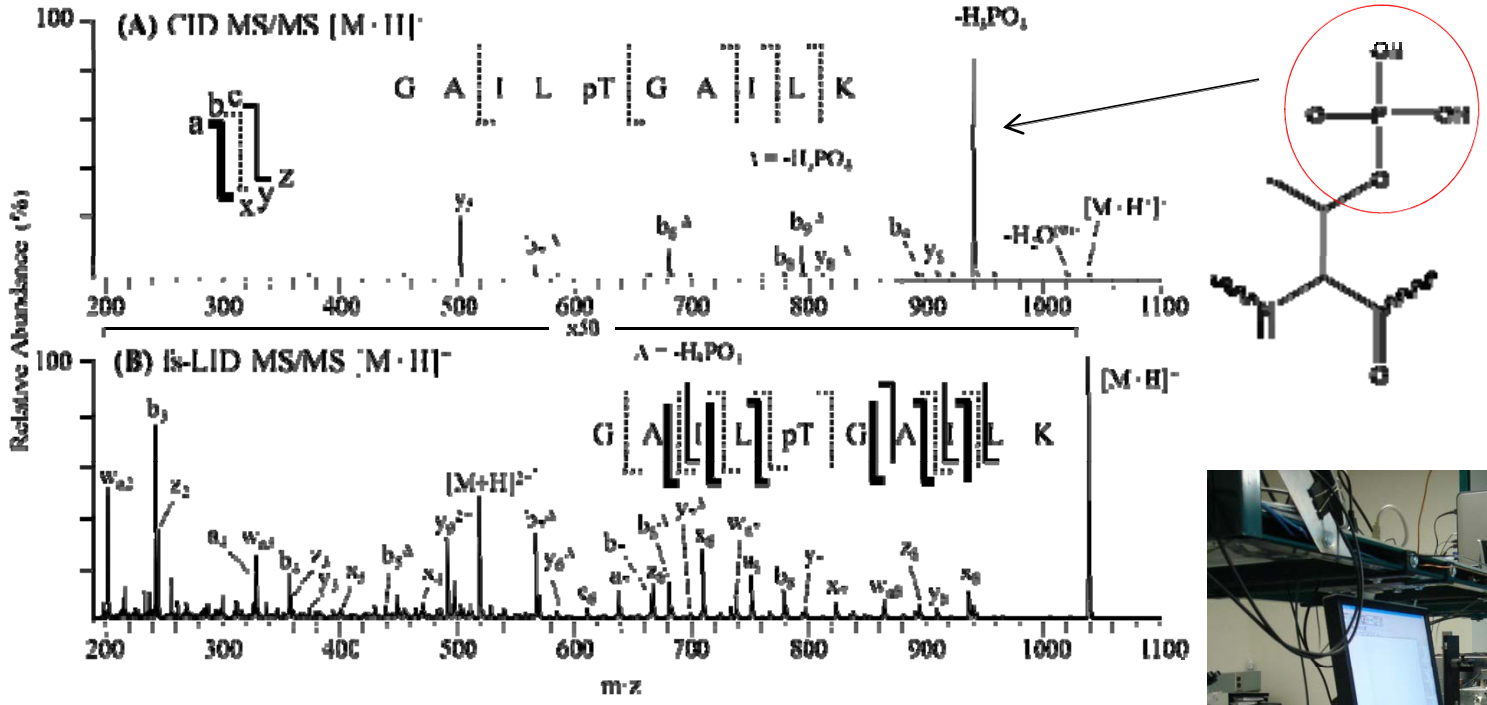
Remetter et al, Nature Phys. 2, 323 (2006)



Holler, PRL 106, 123601 (2011)

3. Challenges in Coherent Control

c. Breaking strong chemical bonds while leaving weak ones intact



Kalcic *et al. J. Am. Chem. Soc.***131**, 940 (2009)

Smith et al. *J Am Soc Mass Spectrom,* **21**, 2031 (2010)

Conclusion: fs-LID is an important method for PTM analysis:

Other methods: Electron capture dissoc., Electron Transfer dissoc., and VUV dissoc.

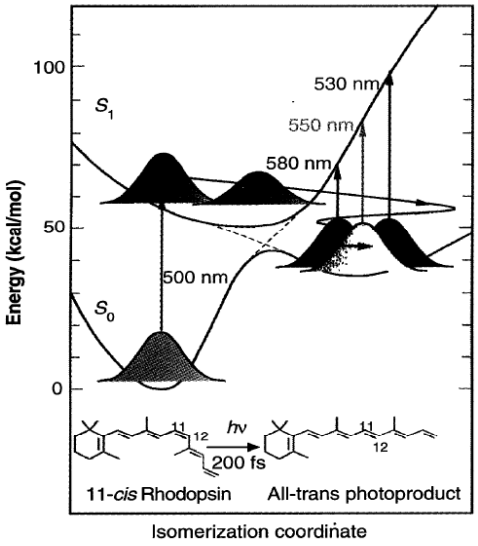
Perspective: Post-translational modifications: a challenge for proteomics and bioinformatics, Appel, Bairoch, *Proteomics* 6,1525-6 (2004)

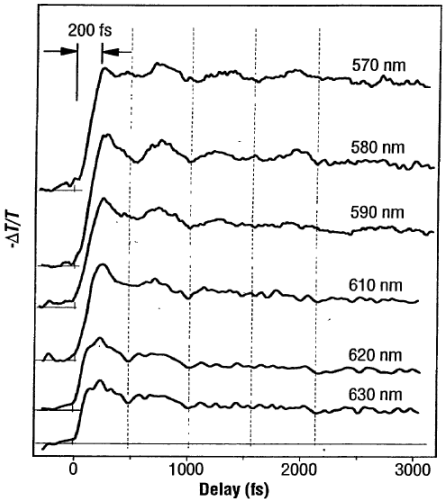
Cellular tumor antigen p53: (*Science* Molecule of the year 1993), has more than 18 phosphorylation sites.

DNA winds around Histones, gene regulation and DNA repair depend on histone PTM's.

3. Challenges in Coherent Control

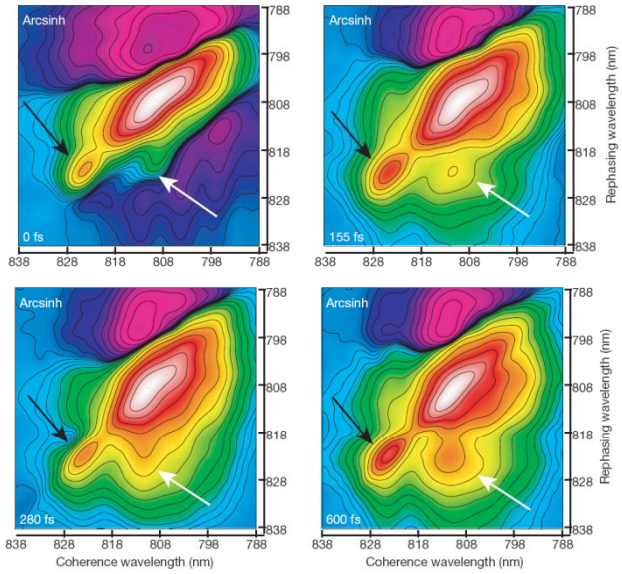
c. Determining the role of coherent quantum processes in nature





Vibrational Coherence in Vision

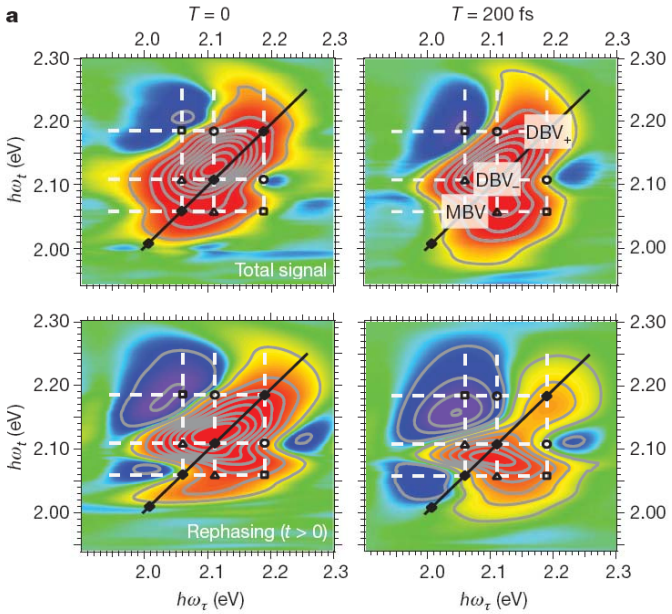
Wang et al. Science 266, 5184 (1994)

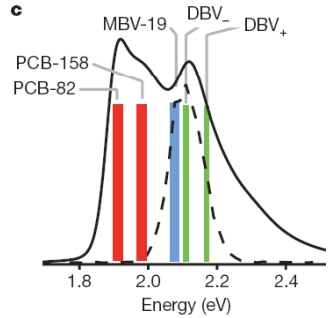


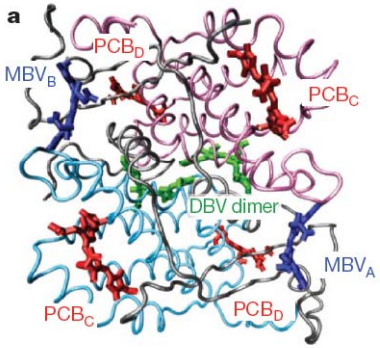
Coherence in Photosynthesis

Engel et al. Nature (2007)

"direct evidence for remarkably long-lived electronic quantum coherence playing an important part in energy transfer processes within this system."







Coherently wired light-harvesting in photosynthetic marine algae **at ambient temperature**

Collini et al. Nautre (2010)

4. Future Outlook and Applications

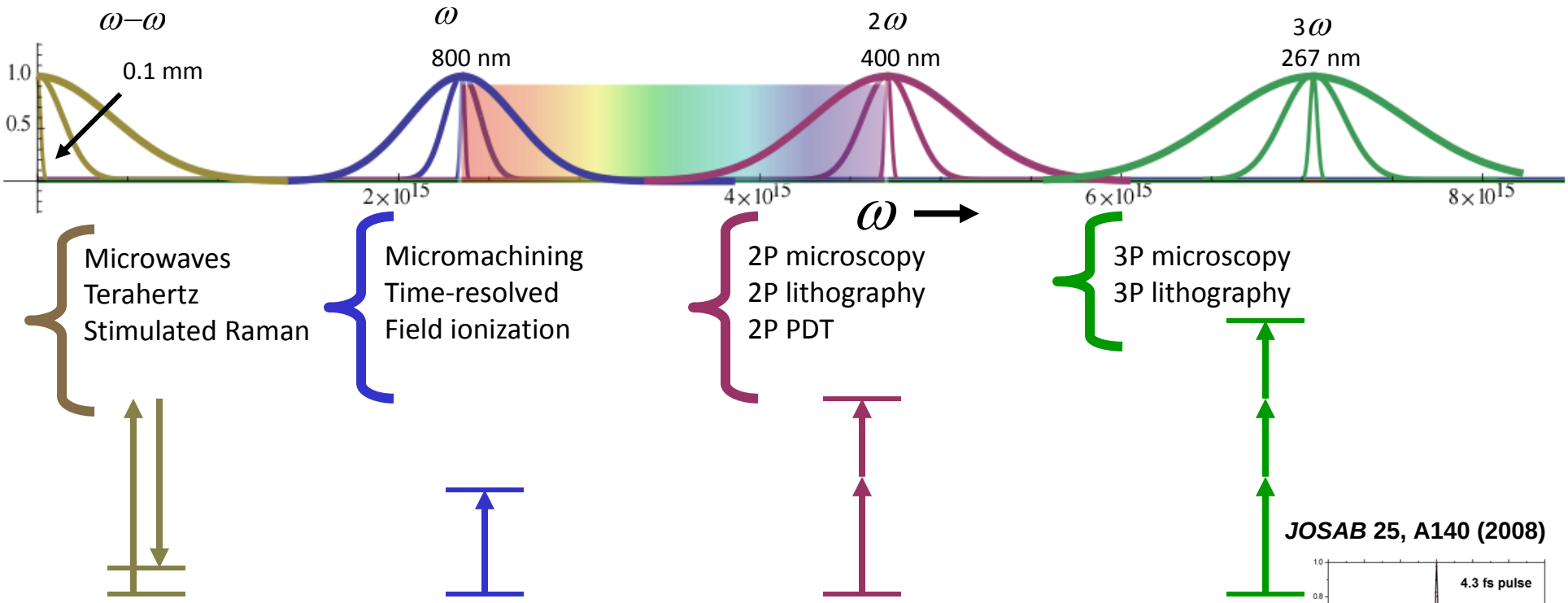
a. Phase Control of Advanced Sources – towards TL single-cycle pulses

Octave spanning (single-cycle) laser pulses can access any frequency in the spectrum

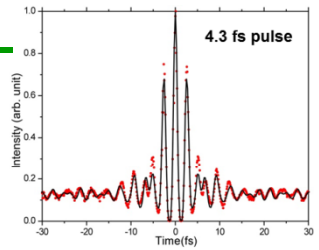
Typical 100 fs laser, centered at 800 nm

Ultrafast laser producing 12 fs pulses; 2P (x8.3), 3P (x69)

Ultrafast laser producing 4 fs pulses; 2P (x25), 3P (x625)



JOSAB 25, A140 (2008)



In 1997 Murnane and Kapteyn reported generation up to the 297th harmonic! =>Coherent X-rays with λ~2nm

A 3fs pulse doubles in pulse duration after propagating 12” in air.

Applications of single-cycle pulses require ultra-precise phase measurement and compression

MIIPS has the accuracy and precision to enable this research

4. Future Outlook and Applications

a. Phase Control of Advanced Sources

Femtosecond Free-Electron X-Ray Sources: Sub-10fs pulse duration

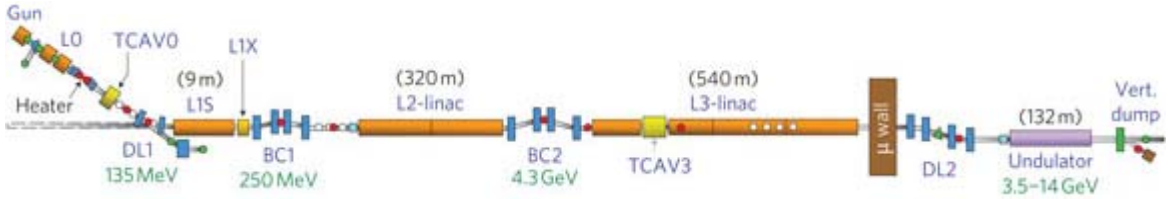


Table top x-ray sources:

Compact Coherent X-Ray Sources <10nm wavelength

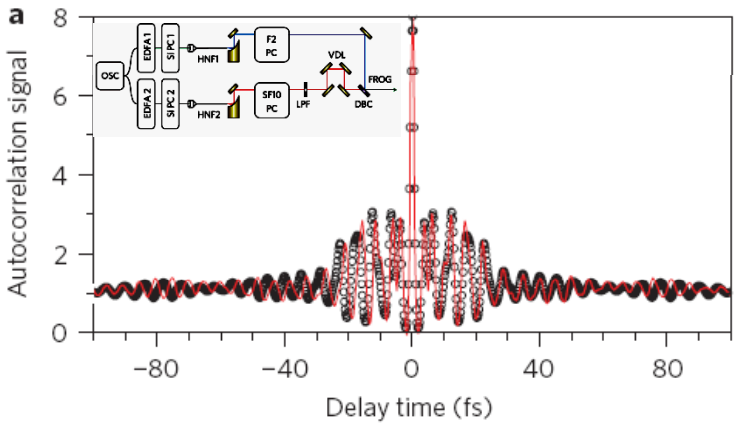
Table top attosecond laser sources:

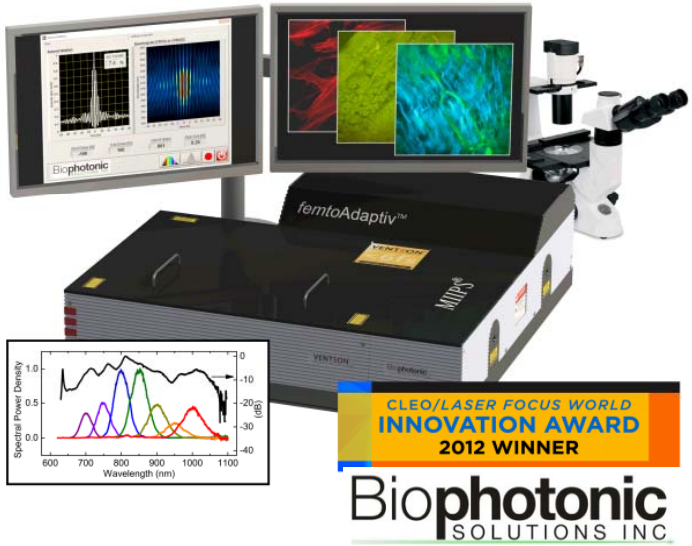
Sub-80as (sub-30as) isolated attosecond pulses in the x-ray regime

Ultrafast laser sources:

From single cycle pulses to self-compressing <5fs sources

It is all a matter of phase control.



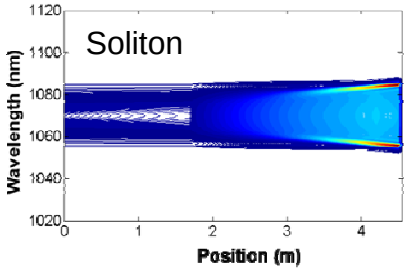


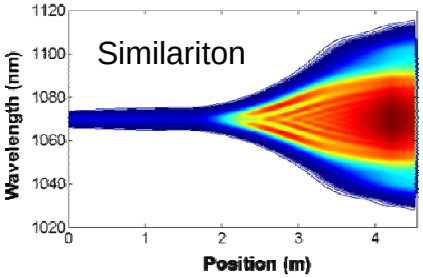
Single-cycle pulse from Er-doped fiber technology, Krauss et al. (2010)

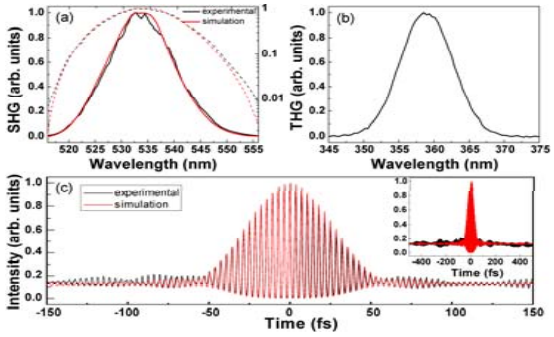
4. Future Outlook and Applications

a. Phase Control of Advanced Sources – new concepts

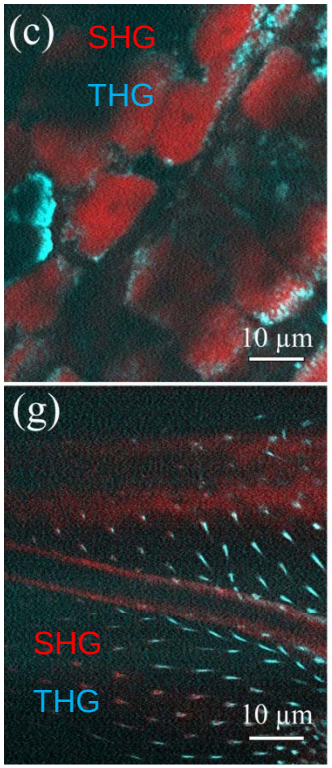
Fiber laser oscillators capable of sub-40fs 20nJ/pulse output. Dantus and Wise groups: **Opt. Express 19**, 12074 (2011)



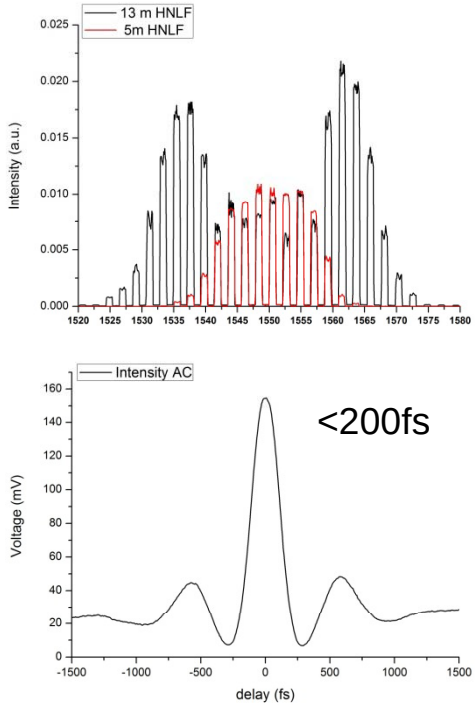




Sub-30fs Ytterbium oscillator

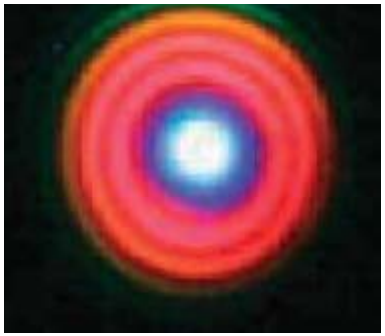


Synthetic comb sources



Compact <9fs fiber lasers

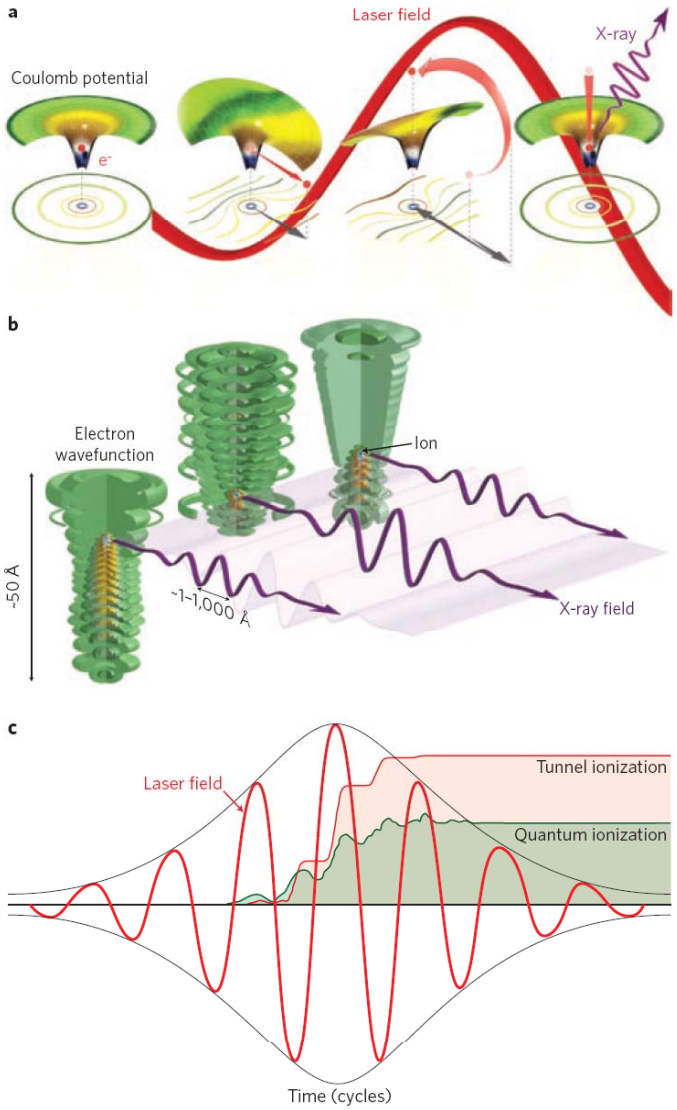
Filamentation phenomena

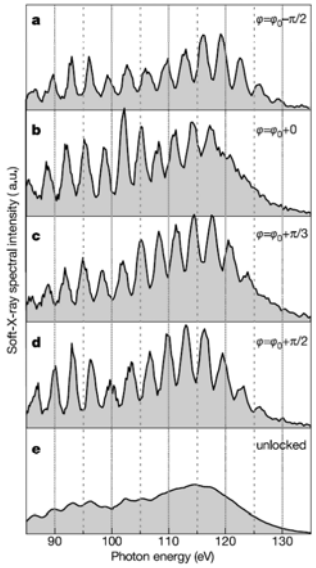


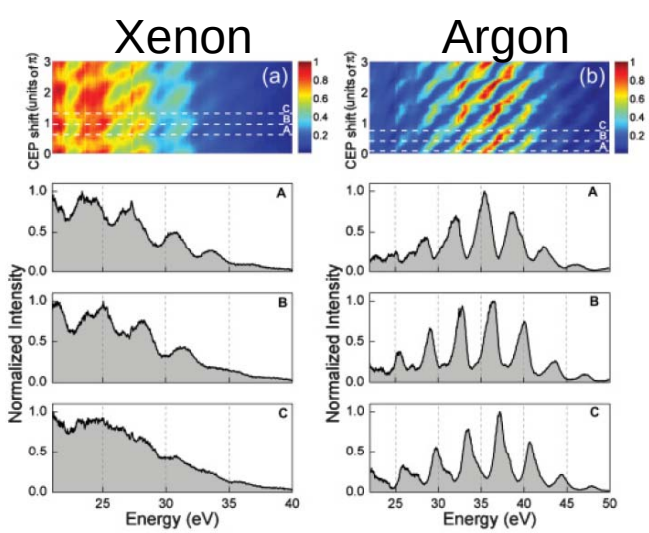
A. Braun et al., *Optics Lett. 20*, 73 (1995)

4. Future Outlook and Applications

a. Phase Control of Advanced Sources – pulse shaping attosecond pulses

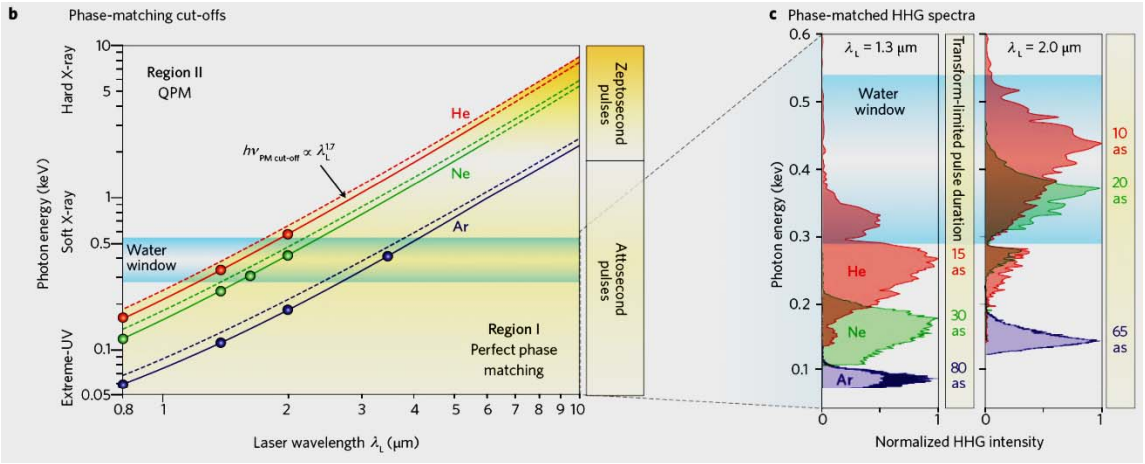






CEP phase effect on HHG
Baltuska et al. Nature (2003)

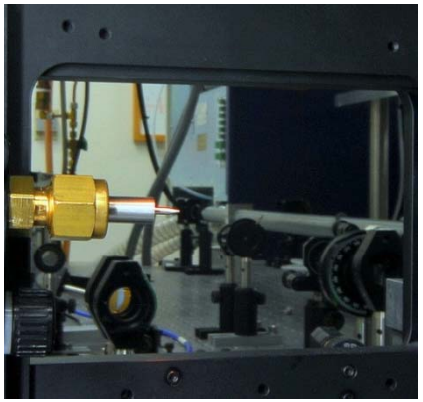
CEP phase effect on HHG
Calegari et al. PRA (2011)



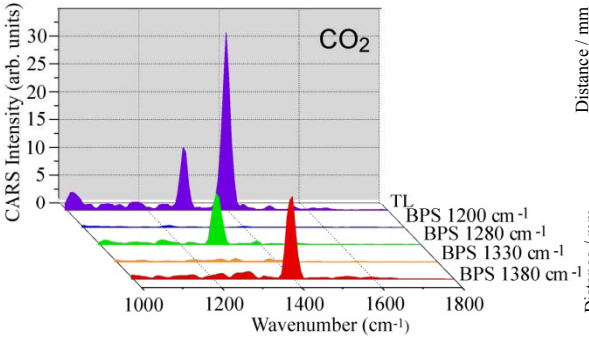
McPherson et al HHG JOSAB (1987); Classical model Corkum PRL (1993),
Popmintchev et al. Nature Photonics Review (2010); Chen et al. PRL (2010)

4. Future Outlook and Applications

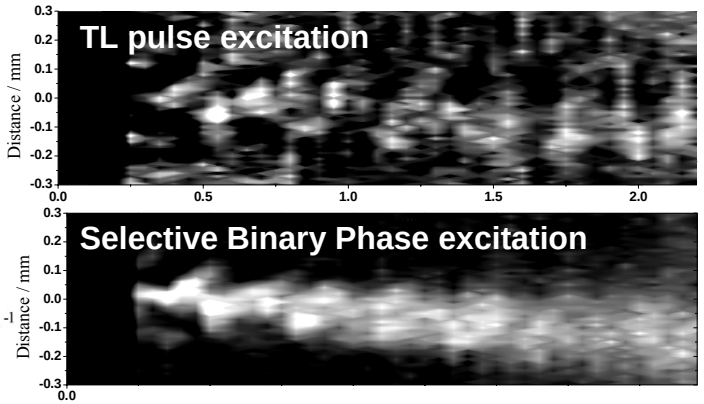
b. Quantum control-based molecular sensing and diagnostics



Selective Single Beam CARS



Images of turbulence in a jet of CO_2 in air

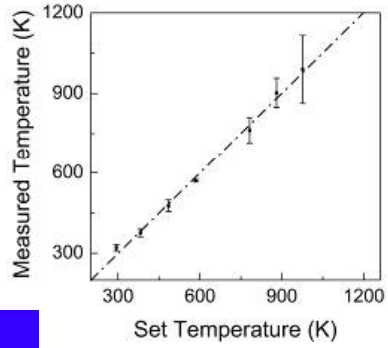


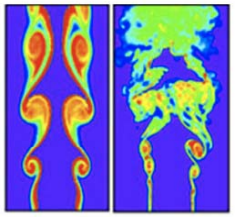
J. Raman Spec. **42**, 393-398 (2011)

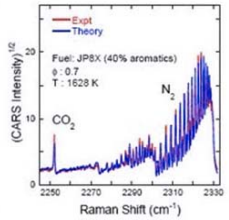
Collaboration with S. Roy Spectral Energies, and J. Gord (AFRL)

Single-Shot Gas Phase Thermometry via Multi-Pulse Time-to- Frequency Mapping of Coherence Dephasing

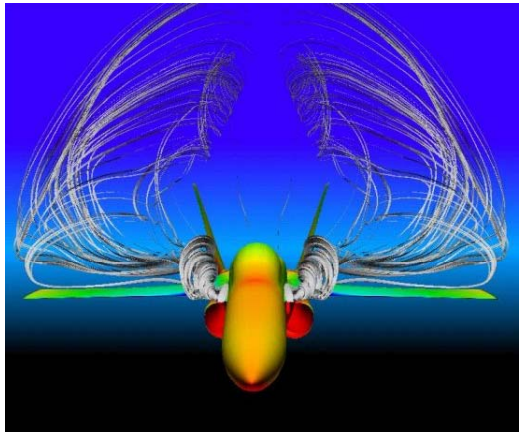
Yue et al., *J. Anal. Chem.* (2012)





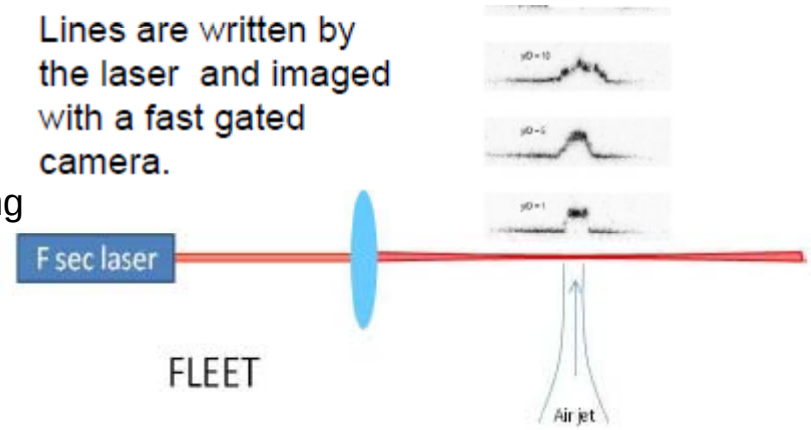


Spectral Energies, LLC



Femtosecond laser electronic excitation tagging
R. B. Miles

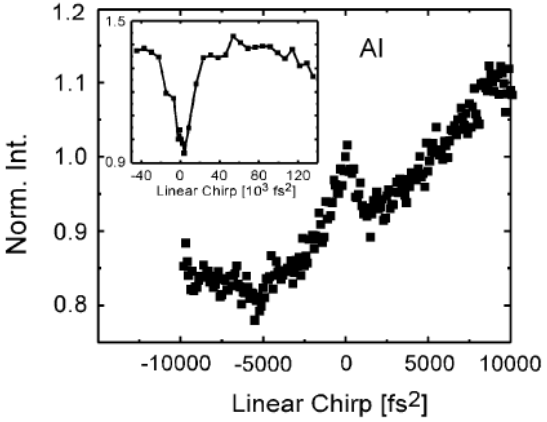
Appl. Opt. 50, 5158 (2011)

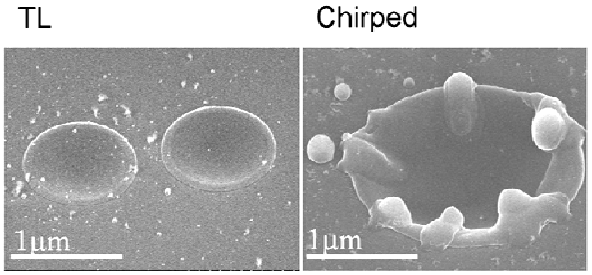


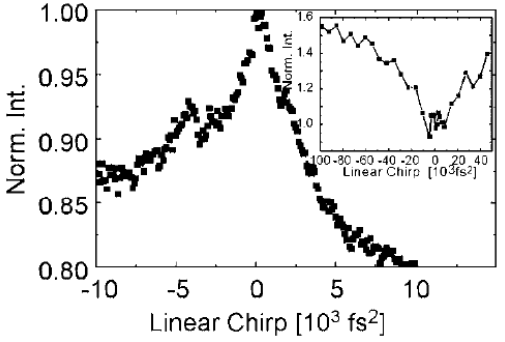
4. Future Outlook and Applications

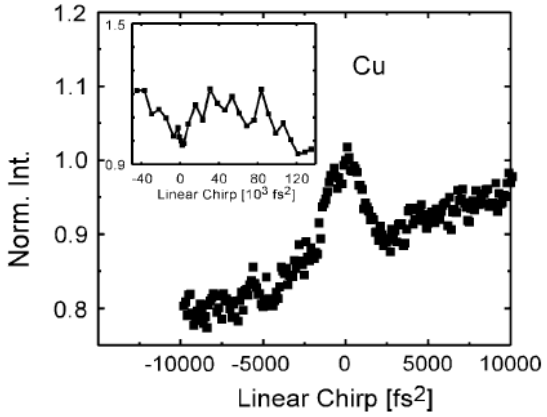
c. Material Synthesis and Material Processing

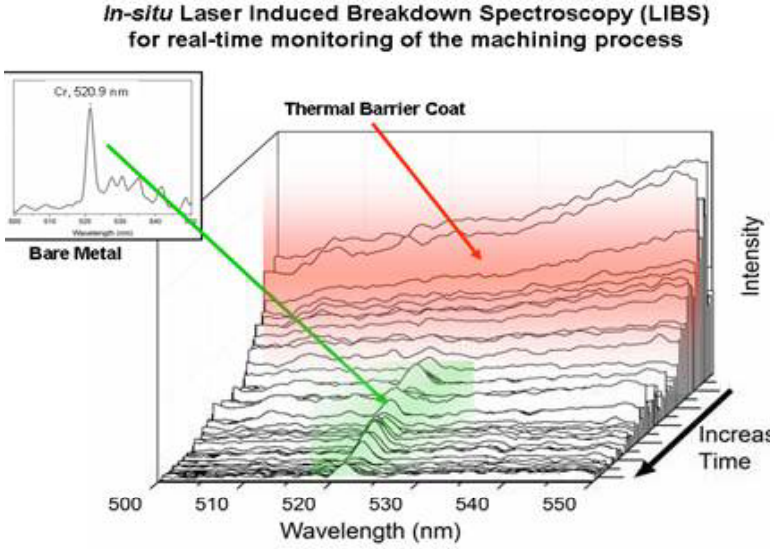
Single shot ablation of silicon

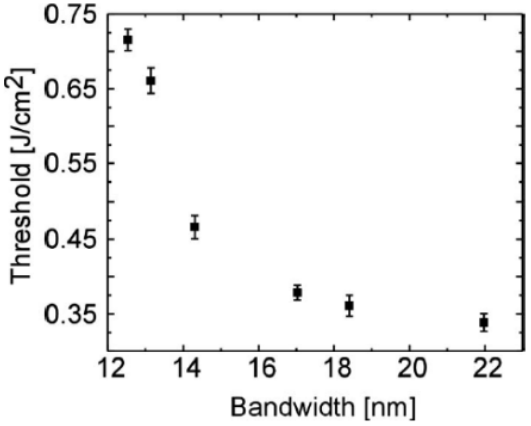












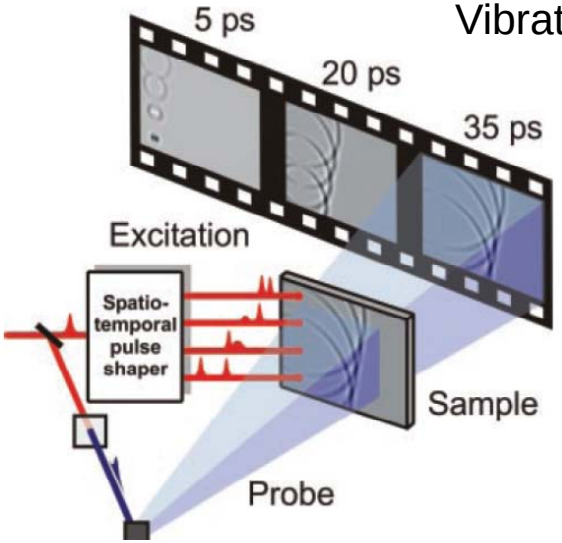
Gunaratne et al. J. App. Phys. 106, 123101(2009)

Gunaratne et al. Chem. Phys. Letts. 423, 197 (2006)

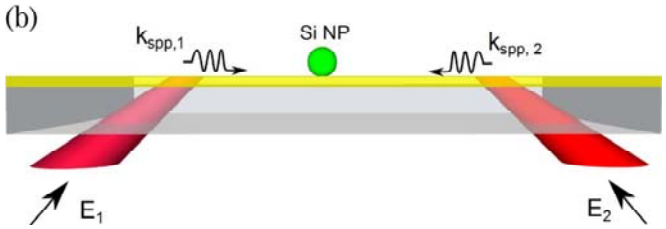
4. Future Outlook and Applications

d. Connecting the macro-micro-and-nano worlds

Spatiotemporal Coherent Control of Lattice Vibrational Waves



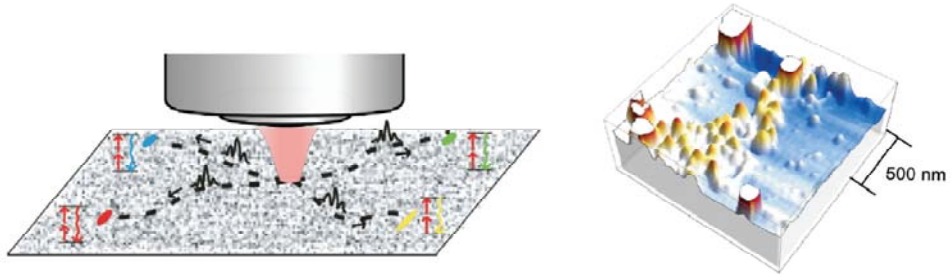
Feurer, Vaughan, Nelson, Science 299, 374 (2003)

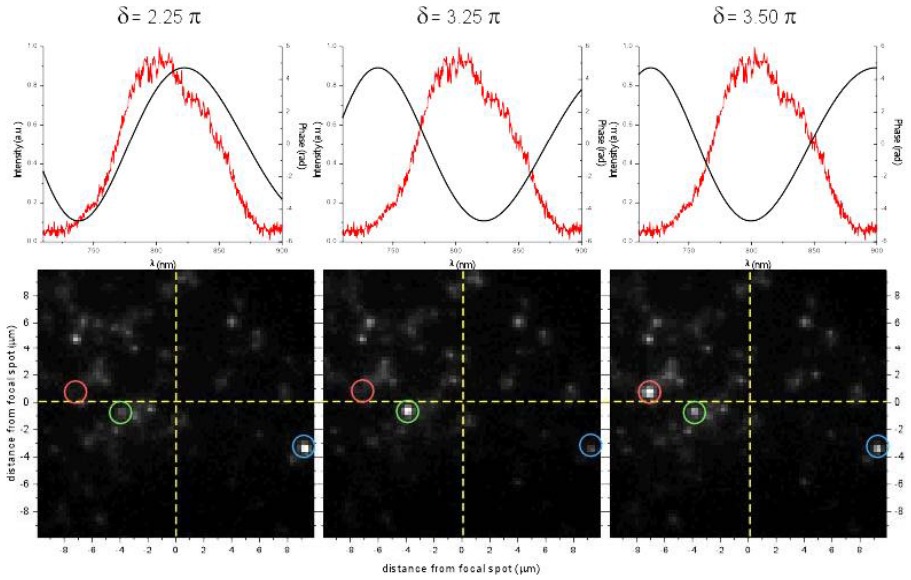


Surface-mediated four-wave mixing of nanostructures with counterpropagating surface plasmon polaritons

Liu, Wang, Potma, Opt Lett 36 2348 (2011)

Measurement and Control of Ultrashort Optical Pulse Propagation in Metal Nanoparticle-Covered Dielectric Surfaces





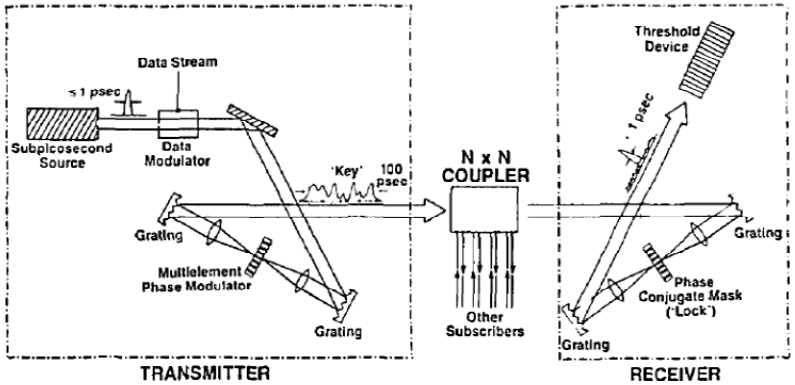
Gunn, Ewald, Dantus, *Nano Letters* 6, 2804 (2006).
Gunn, High, Lozovoy, Dantus J Phys Chem C 114 12375 (2010)

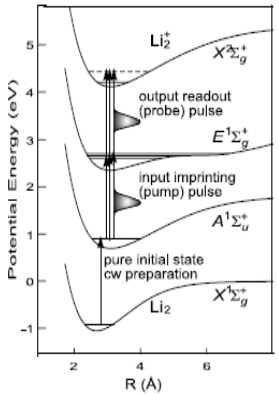
4. Future Outlook and Applications

e. Coherent control and communications and information processing

Encoding and decoding of femtosecond pulses

Weiner Heritage Salehi, Opt Lett 13 300 (1988)

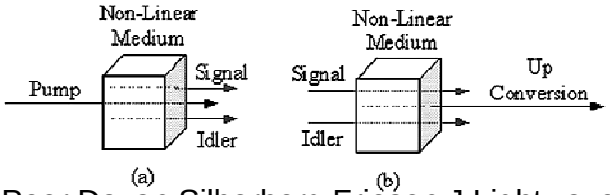




Experimental coherent computation of a multiple-input AND gate using pure molecular superpositions

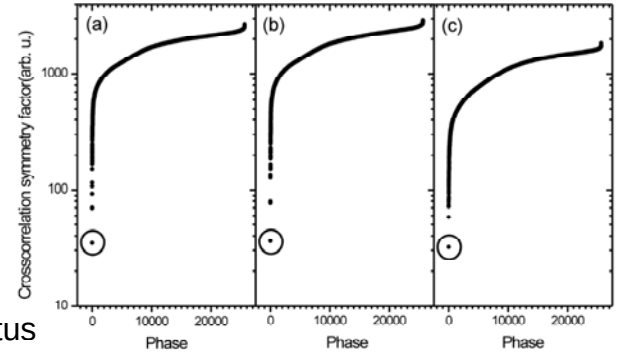
Amitay Kosloff Leone Chem Phys Lett 359 8 (2002)

Optical Code-Division Multiple Access Using Broad-Band Parametrically Generated Light

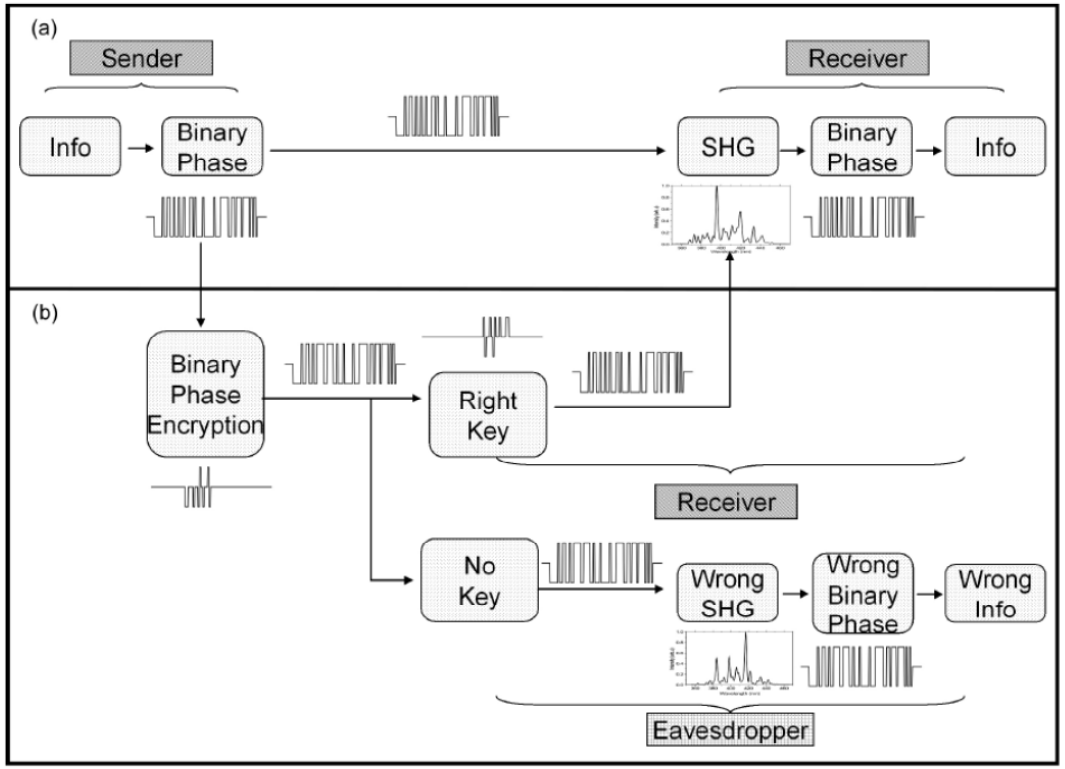


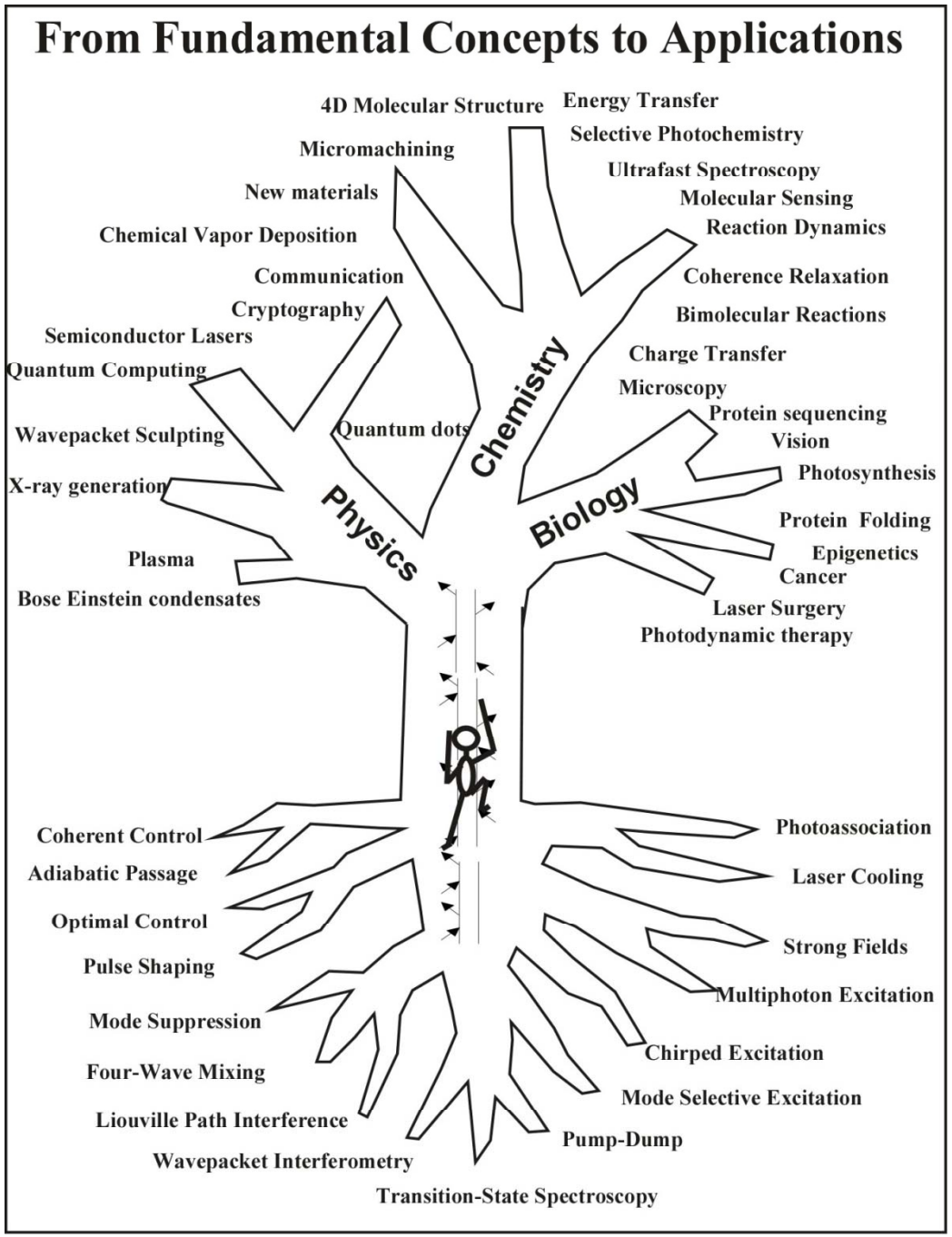
Peer Dayan Silberberg Friesen J Lightwave Tech 22 1463 (2004)

Asynchronous encrypted information transmission with sub-6 fs laser system at 2.12GHz

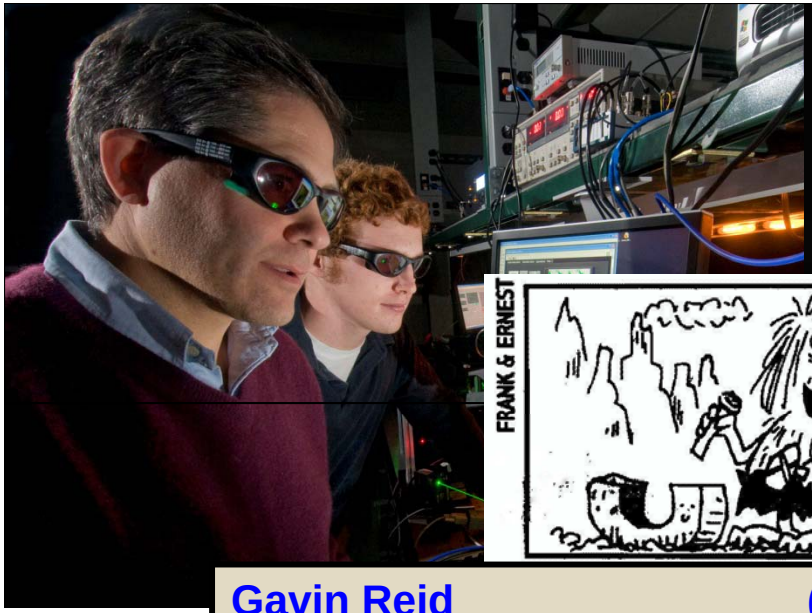


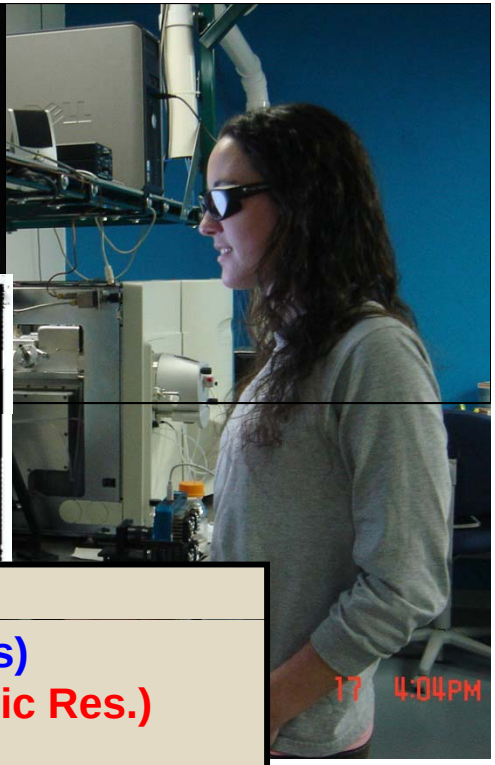
Xu, Coello, Nogueira, Cruz, Dantus Opt Express 16, 15109 (2008)





Dantus, Annual Review Phys. Chem. 2001

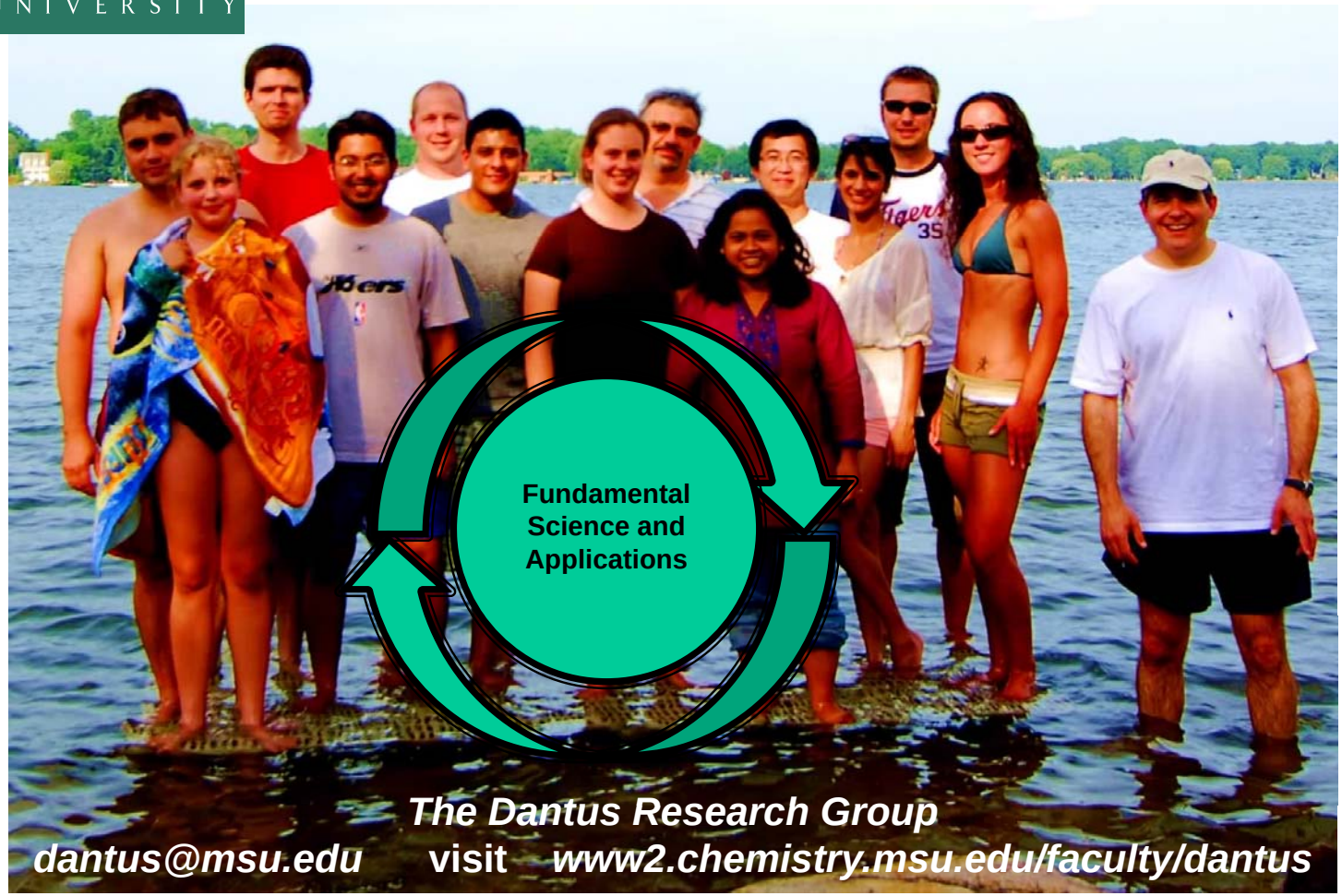




Gavin Reid	(MSU, Chemistry, Proteomics)
Daniel Jones	(MSU, Bio Chem, Metabolomics)
Steven P. Arnoczky	(MSU, Lab for Comp. Orthopedic Res.)
George Abela MD	(MSU, Cardiology)
Chong-Yu Ruan	(MSU, ultrafast electron diffraction)
James Resau MD	(Van Andel Institute, cancer)
Stephen Boppart	(UIUC, femtosecond endoscopy)
Jim Gord	(Air Force Research Lab, CARS)
Sukesh Roy	(Spectral Engines LLC, CARS, Machining)
Dominique Fourguette	(Michigan Aerospace, sensing)
Flavio Cruz	(UNICAMP, Brazil, Novel laser sources)
Sunney Xie	(Harvard, CARS microscopy)
Rich Mathis	(Berkeley, low frequency in biomolecules)
Frank Wise	(Cornell, fiber laser design)
Maurice Jansen	(Free Univ. Amsterdam, PEPICO)
Shaul Mukamel	(UC Irvine, NLO Theory)

Phase control enables fundamental research and applications

MICHIGAN STATE UNIVERSITY



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Office of Basic Energy Sciences
Serving the Present, Shaping the Future

NATIONAL INSTITUTES OF HEALTH

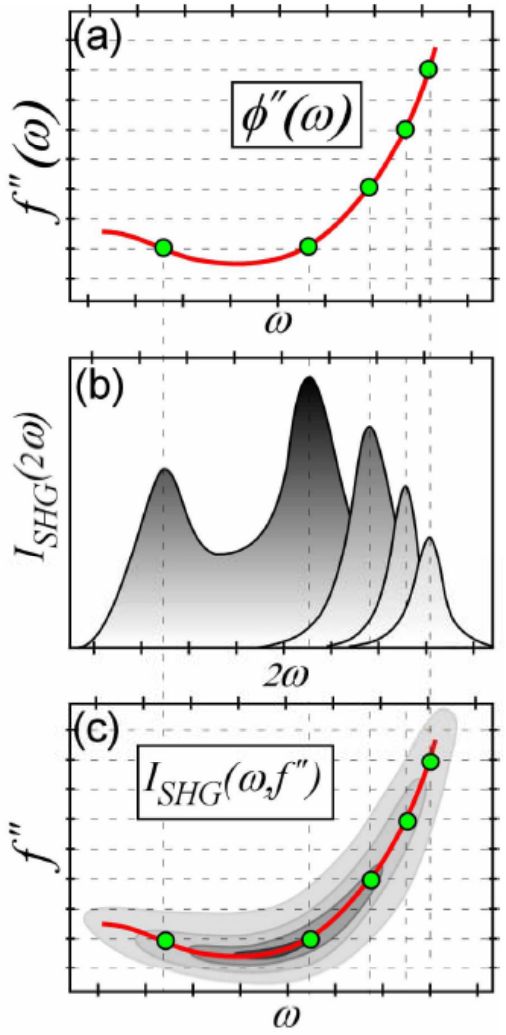
Biophotonic SOLUTIONS INC
ULTRAFAST PULSE SHAPING & COMPRESSION

Spectral Energies, LLC

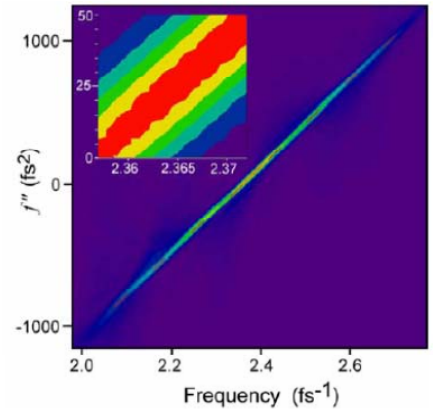
Backup Slides for Follow Up Discussion

Multiphoton Intrapulse Interference Phase Scan (MIIPS)

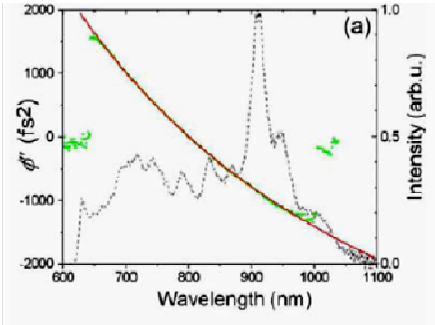
Theory

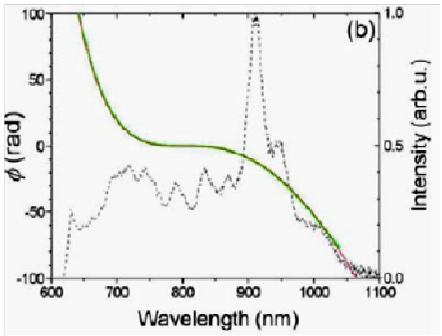


Experiment



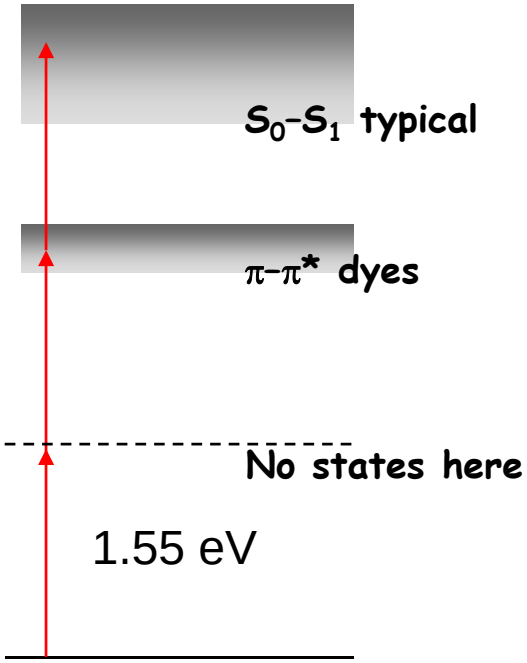
5 fs pulse with
500 fs^3 TOD

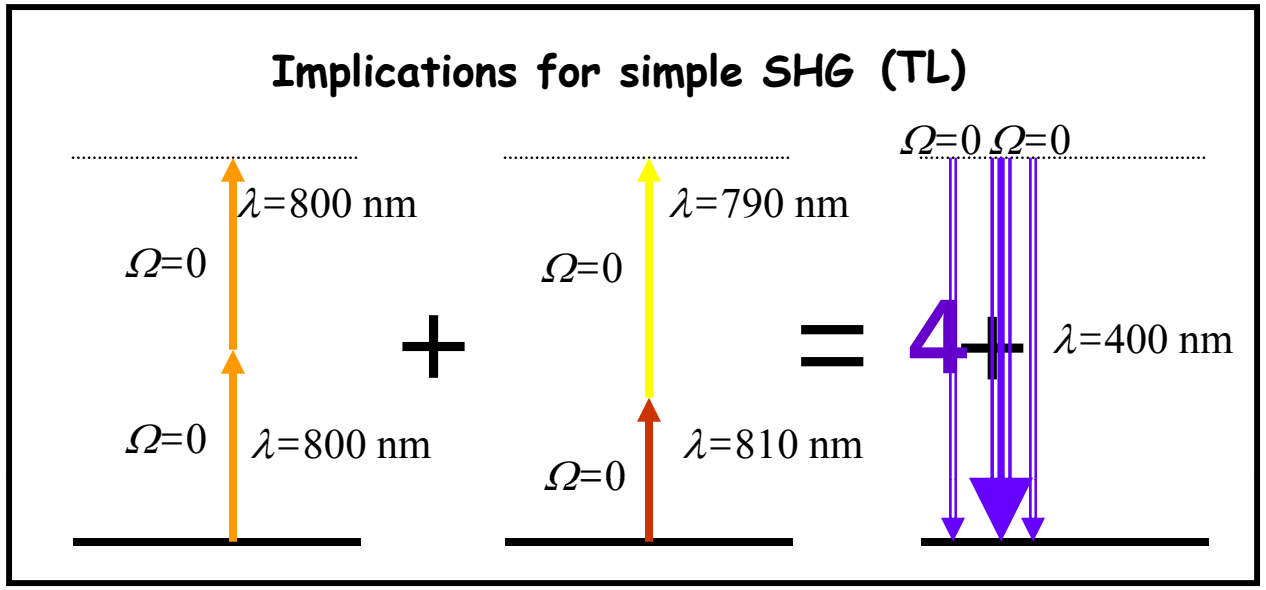


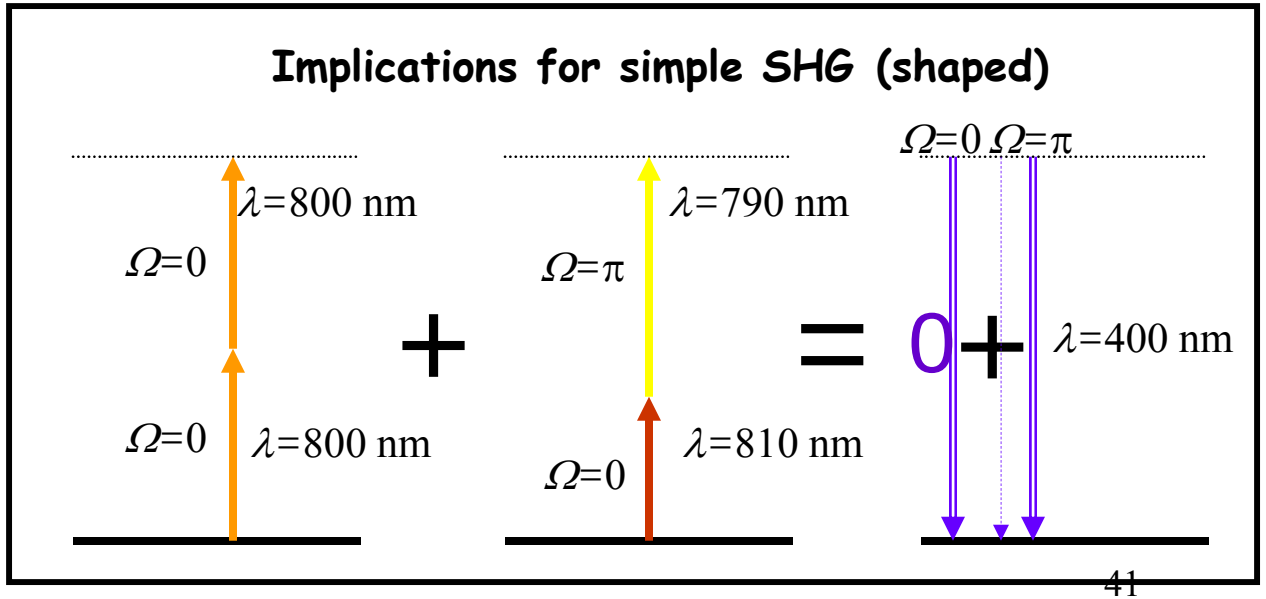


Optics Express **16**, 592 (2008)
JOSA B **25**, A140 (2008)

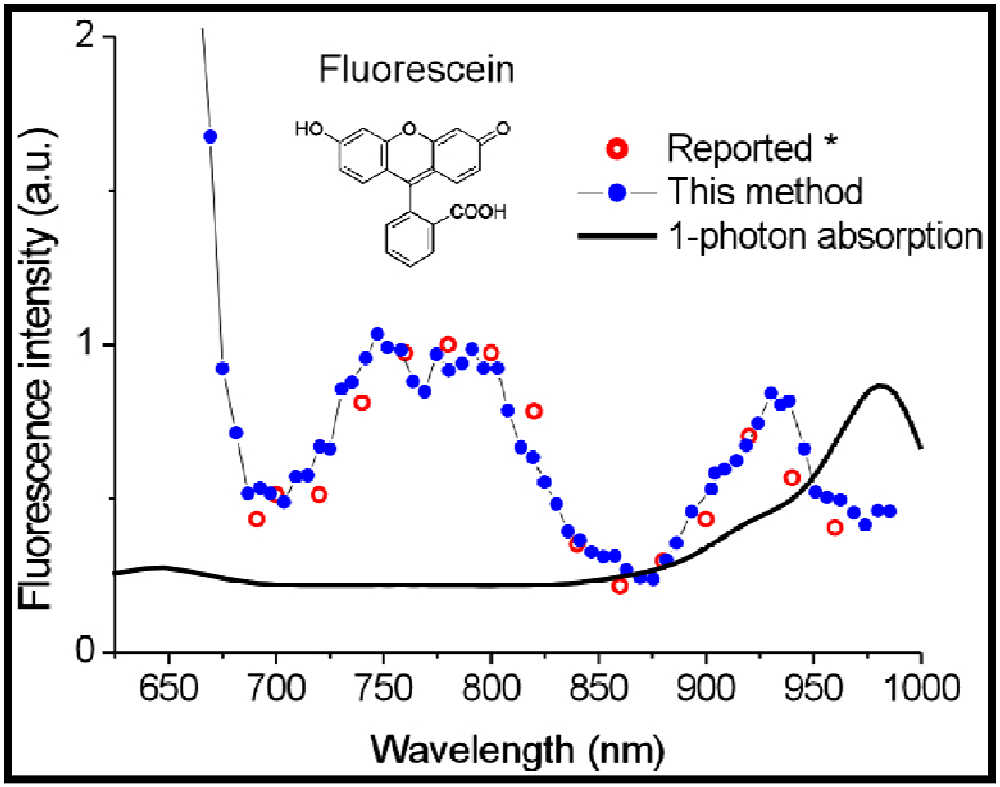
What is Multiphoton Intrapulse Interference?

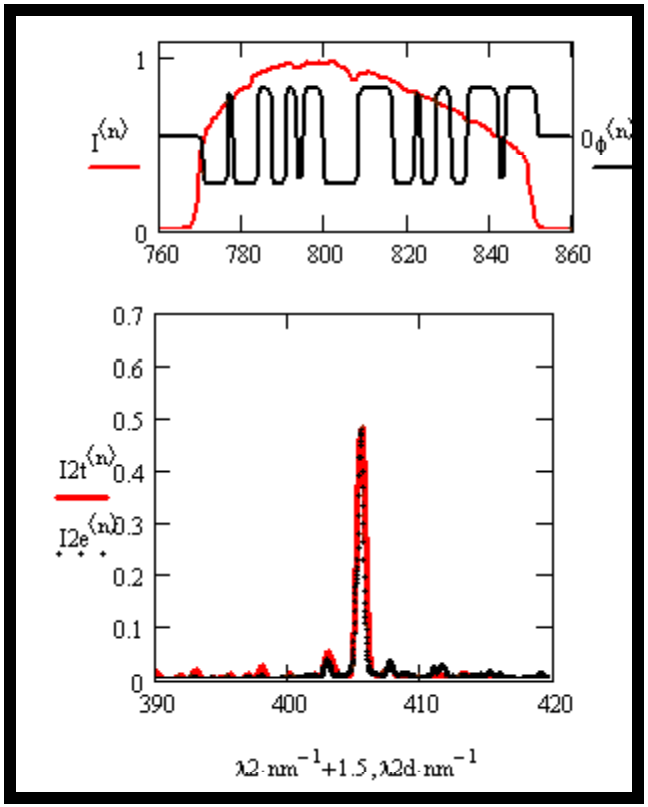






Selective nonlinear excitation is useful!

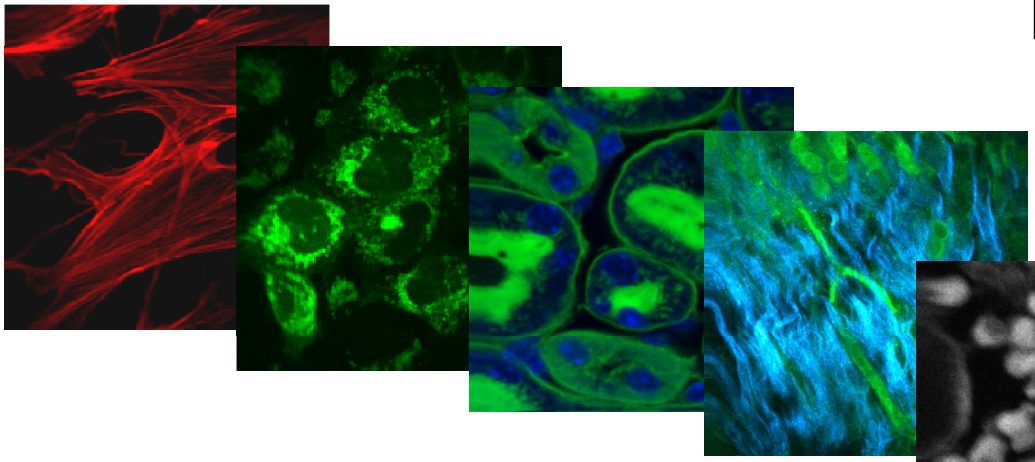




* Albota, M.A., C. Xu, and W.W. Webb. Applied Optics 37,7352 (1998)

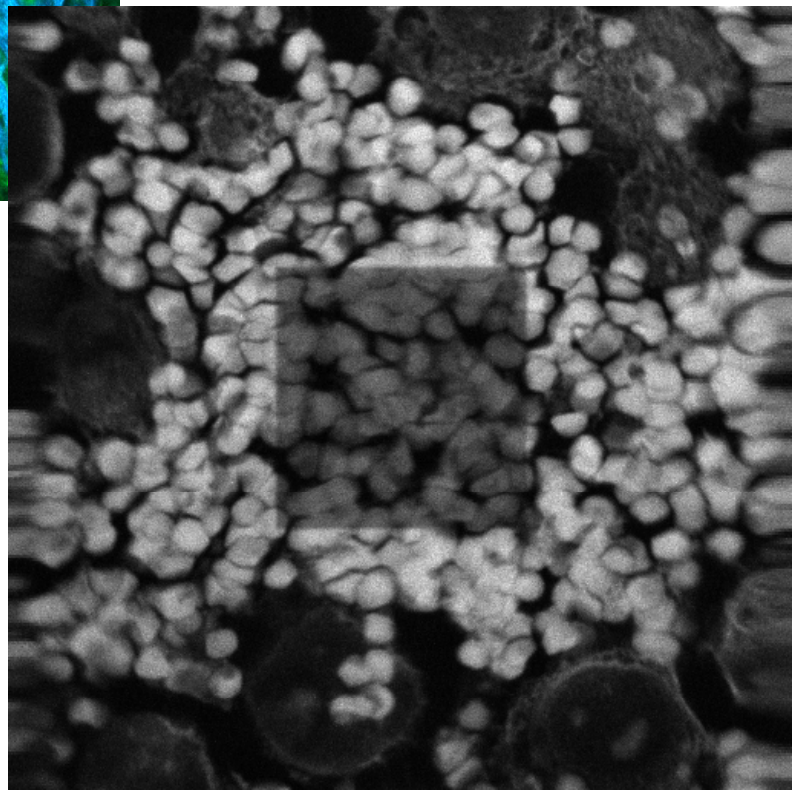
4.6 fs laser Bingwei Xu, Yves Coello Optics Express *14, 10939* (2006)

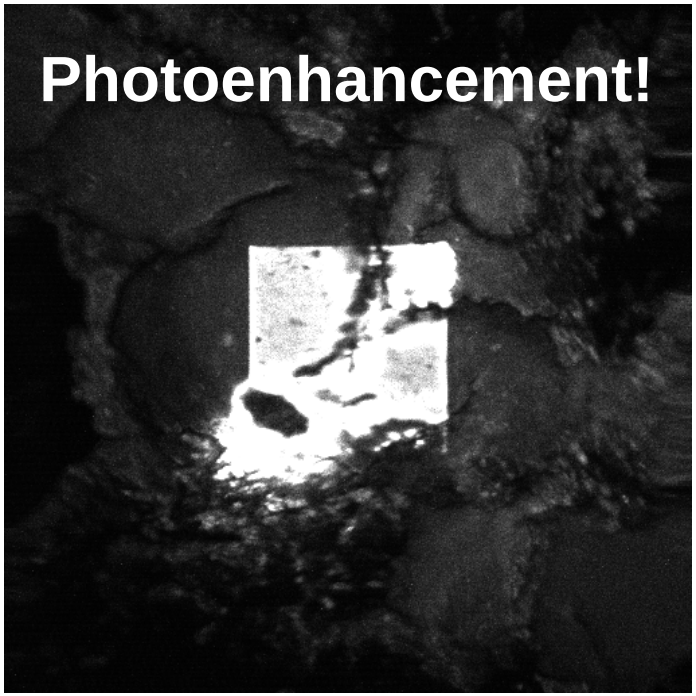
Coello, Xu, and Dantus, Appl. Optics In press (2010)



Pulse shaping for:

Greater signal
Reduced photodamage
Selective excitation





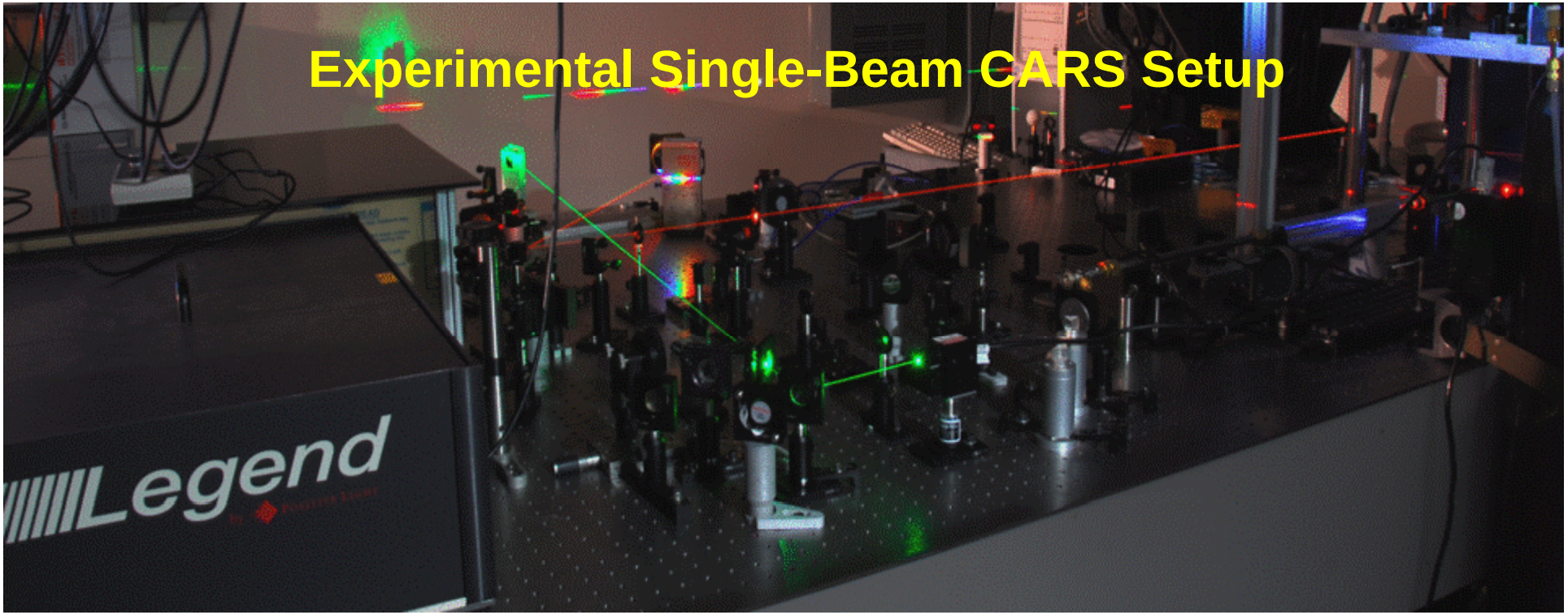
***J.Phys.Chem.*A 106, (2002)**

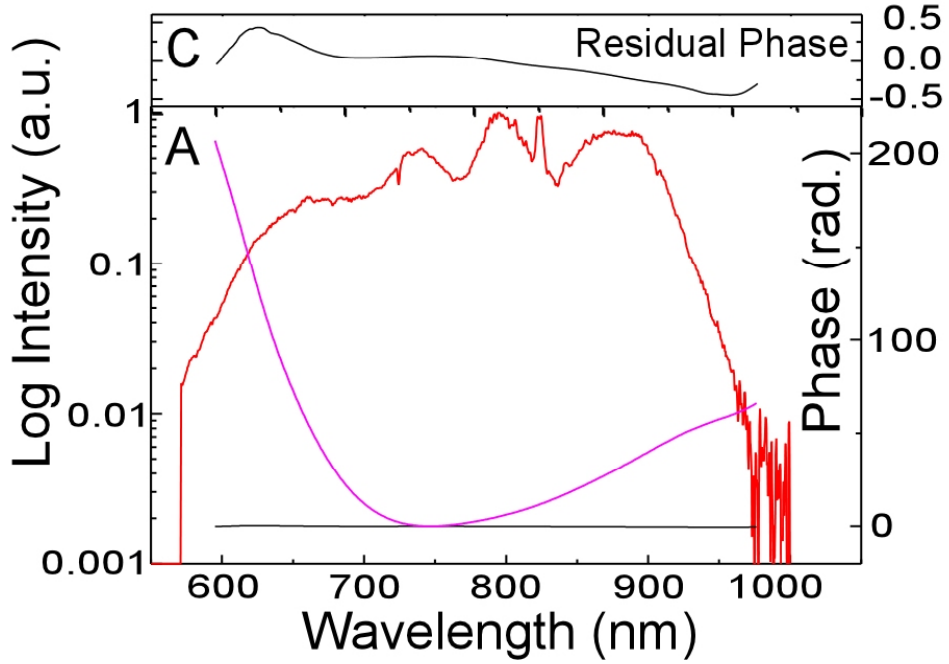
Optics Express 11, 1695 (2003)

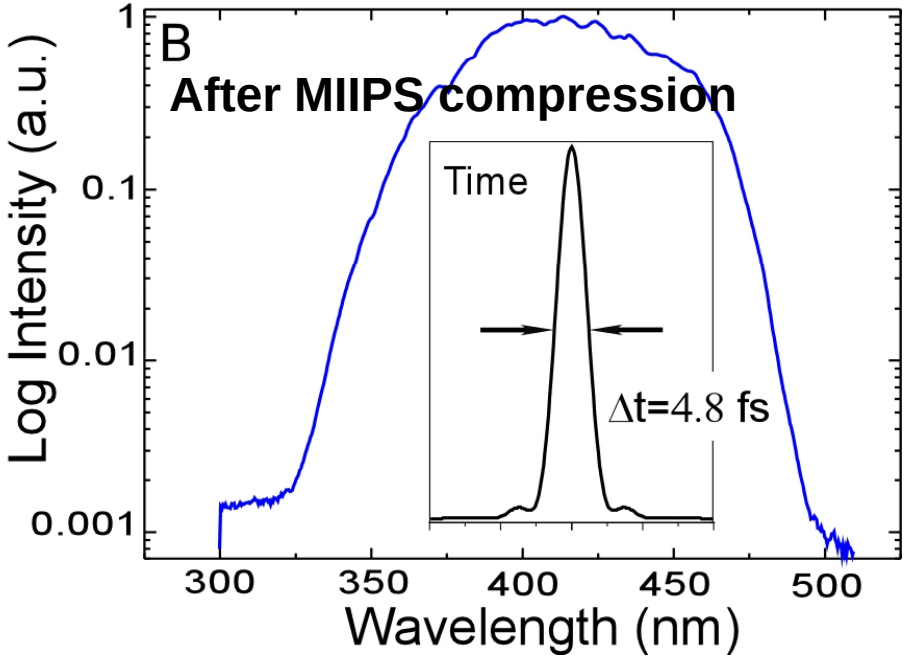
J. Phys. Chem. 108, 53 2004 Opt. Commun. 241, 1841 (2008)

PNAS, 101, 16996 (2004)

Experimental Single-Beam CARS Setup







2010 Pulse Shaping Workshop
August 20-22, 2010

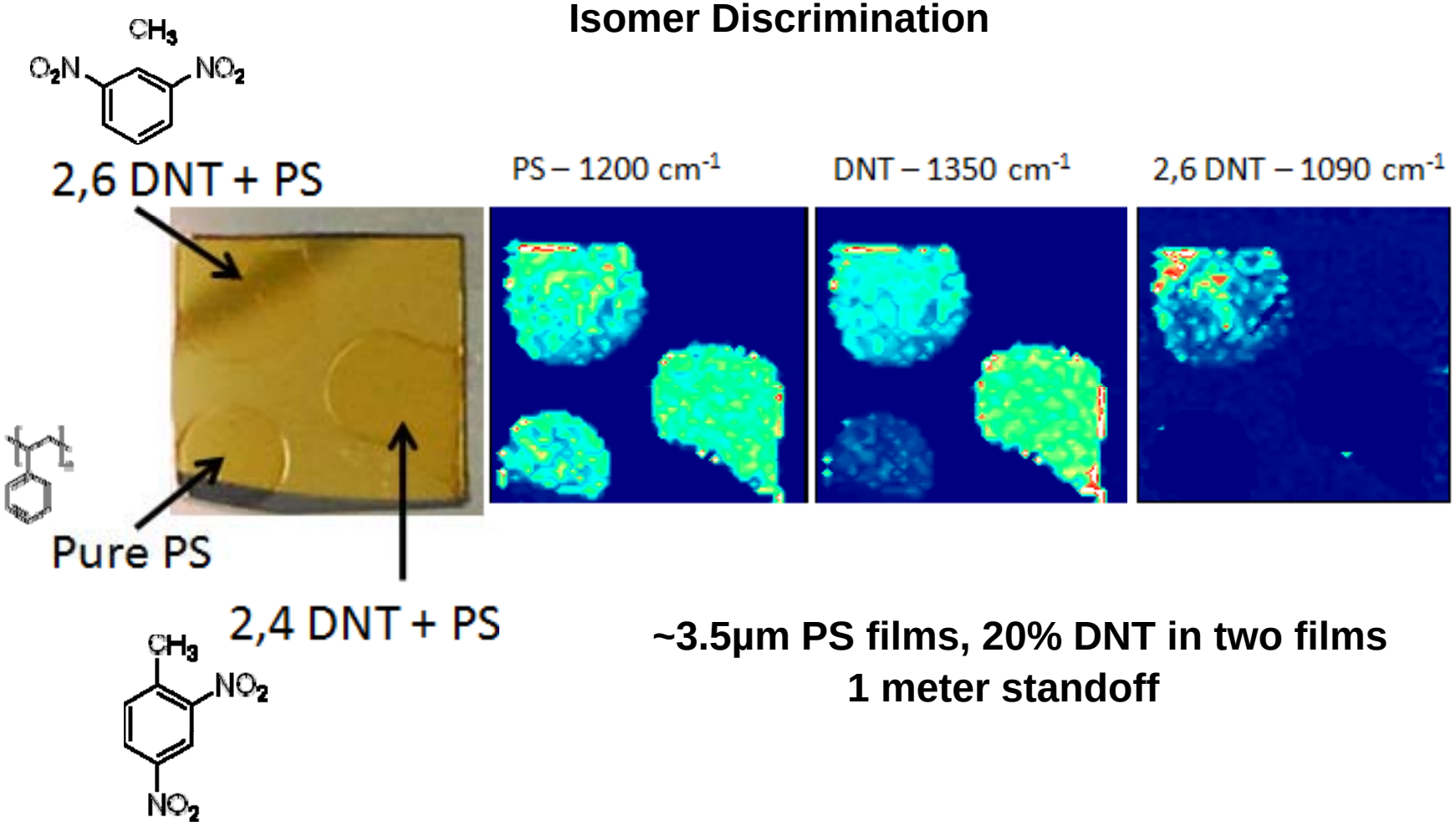
Teaching scientists worldwide how to use sub-5fs pulses routinely

2011 Pulse Shaping Workshop @ MSU Aug 19-21. Keynote by Andrew Weiner

High Sensitivity and Selectivity

SUPER-CARS Imaging

Isomer Discrimination



~3.5μm PS films, 20% DNT in two films
1 meter standoff

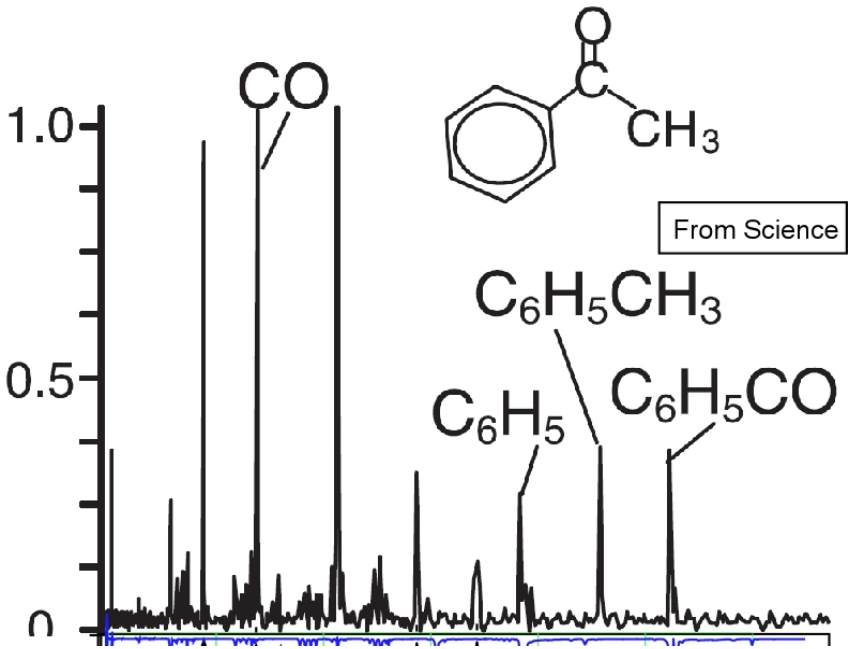
Scientific Goals Achieved: Sensitivity, Selectivity, Imaging

Next step: making it practical

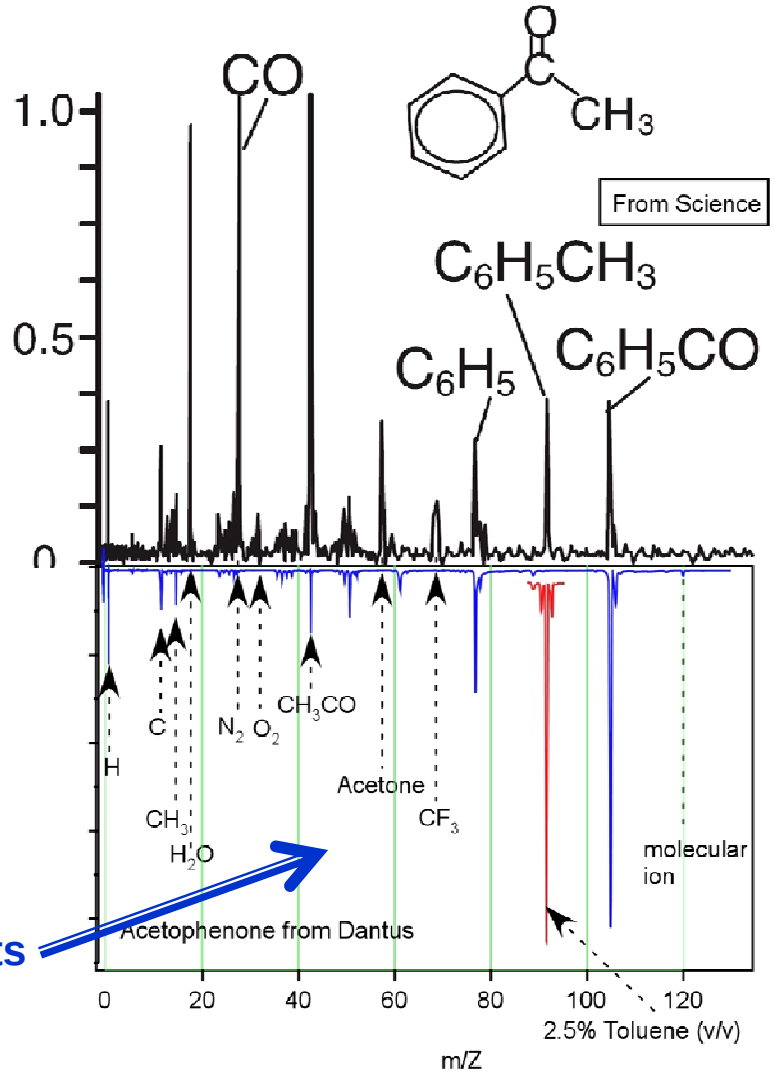
M. Bremer, P. Wrzesinski, N. Butcher, V. Lozovoy and M. Dantus, App. Phys. Letters **99**, 101109 (2011)

Regarding the Reported Selective Synthesis of Toluene from Acetophenone

Original Figure:



Mass spec. obtained for ~100 fs TL pulses



Dashed lines and red peak indicate contaminants

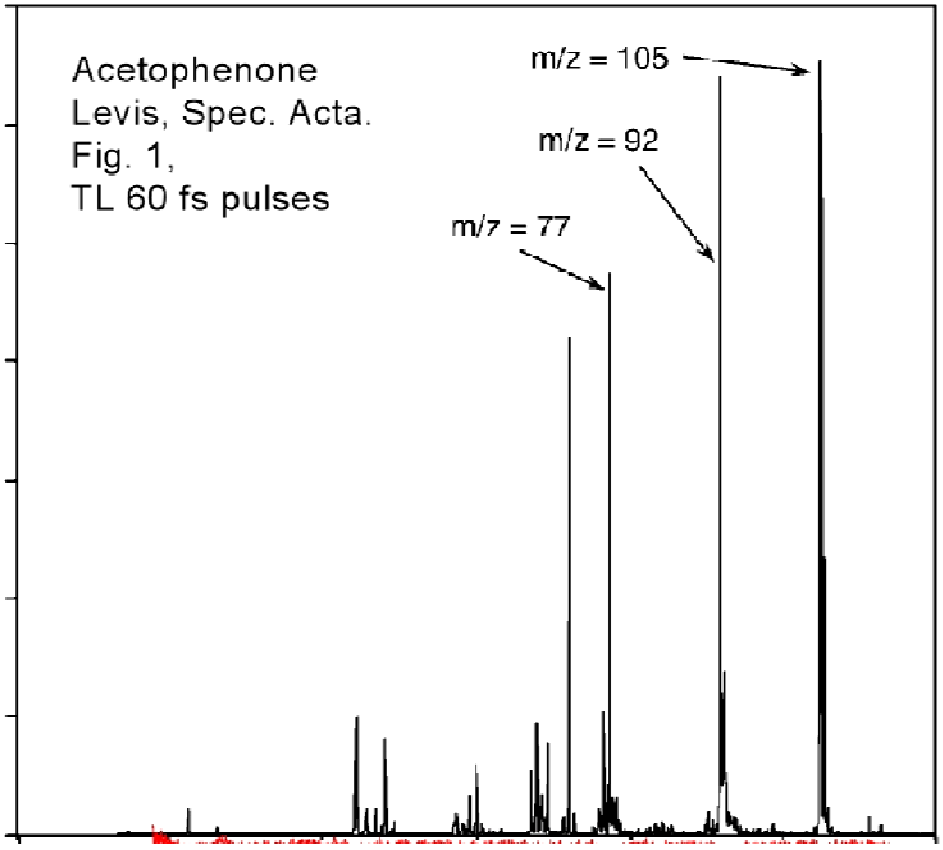
[1] Levis et al., Science 293, 709 (2001).

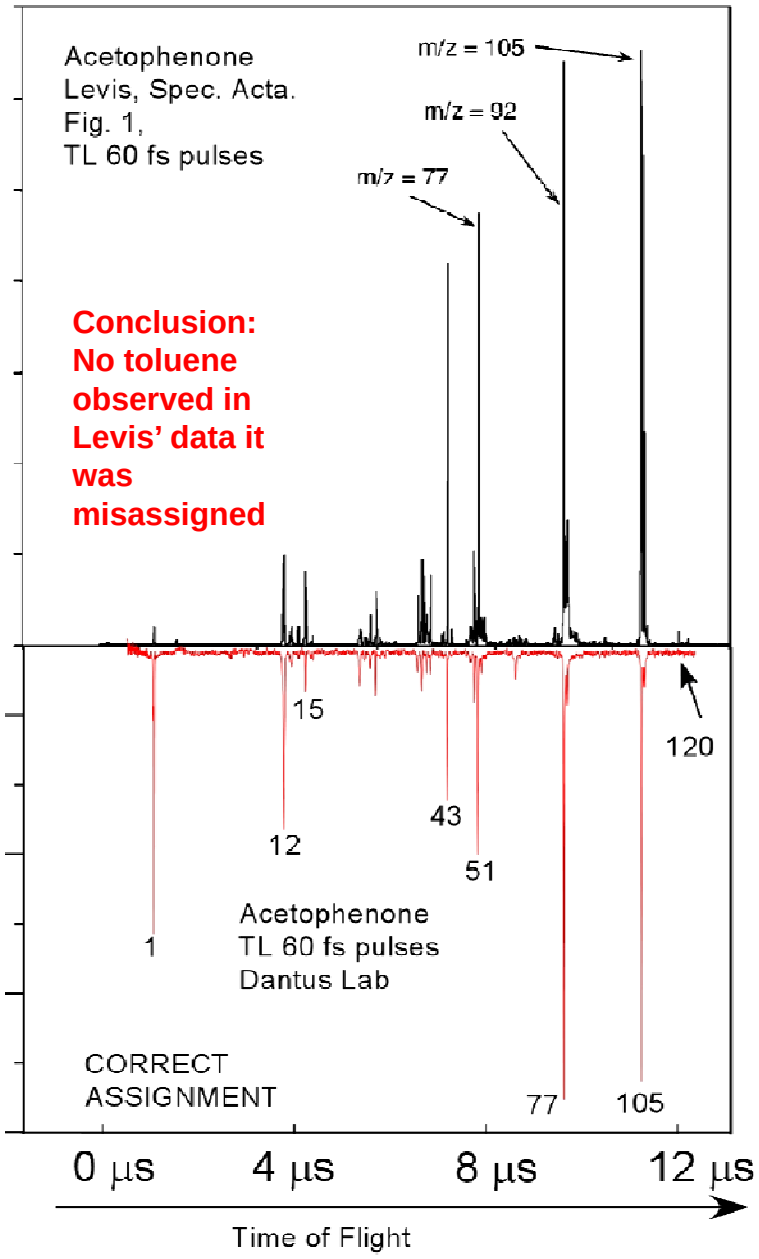
[2] R. J. Levis and H. A. Rabitz, J. Phys. Chem. A, 106, 6427 (2002)

Comment to J. Phys. Chem. A **113**, 5264 (2009)

Regarding Toluene from Acetophenone Reported in 2004

Original Figure from Graham et al.:





[1] Levis et al., Science 293, 709 (2001).

[2] R. J. Levis and H. A. Rabitz, J. Phys. Chem. A, 106, 6427 (2002)

[3] Graham, Menkir and R. J. Levis, Spectrochemica Acta B 58, 1097 (2004)

Comment to J. Phys. Chem. A **113**, 5264 (2009)

Project from DHS Phase I (cleared)

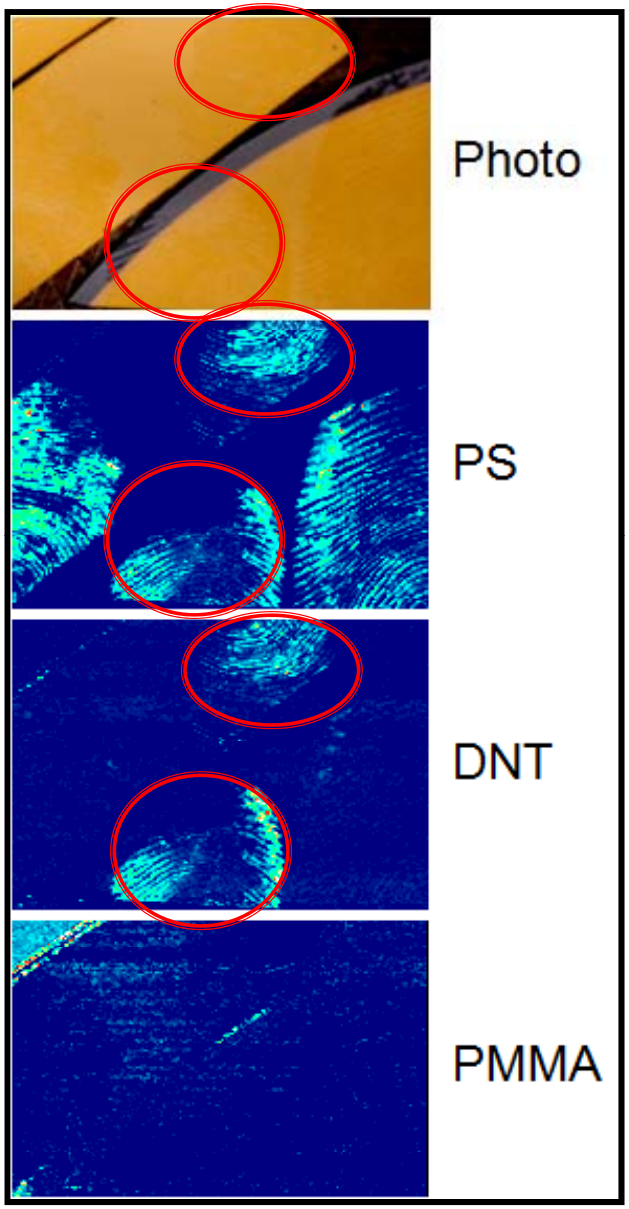
Determine sensitivity and selectivity in a complex background at standoff distances and evaluate imaging capability.

Looking at the fingerprint region!

Complex background

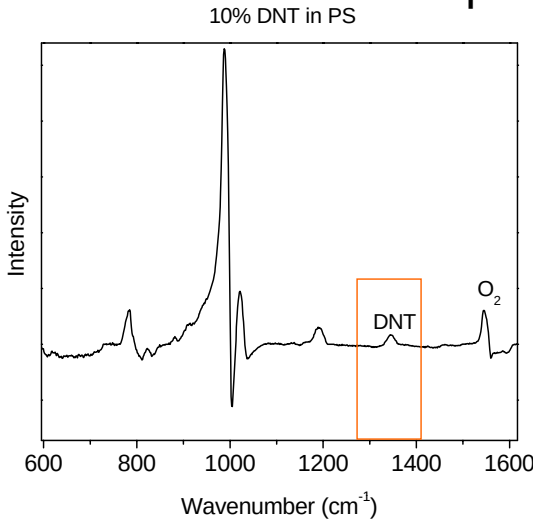
↘

Samples: Polymer solution with trace amount of DNT spin-coated on substrate.

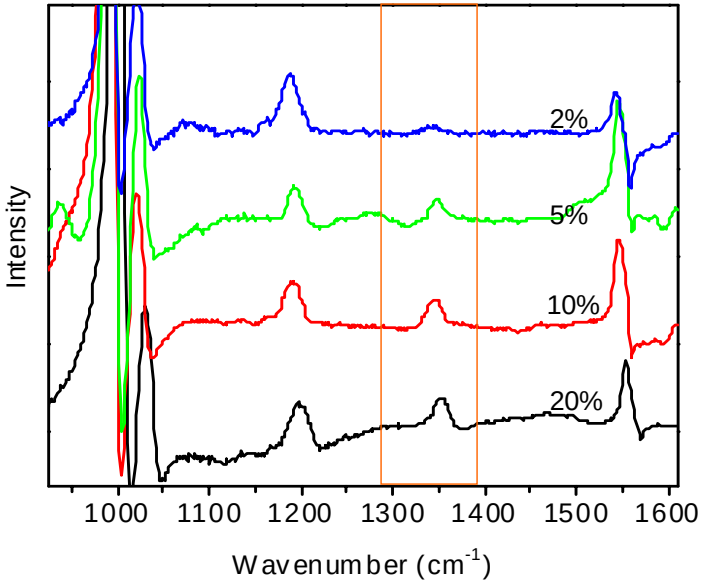


SUPER-CARS Sensitivity at 1 meter standoff

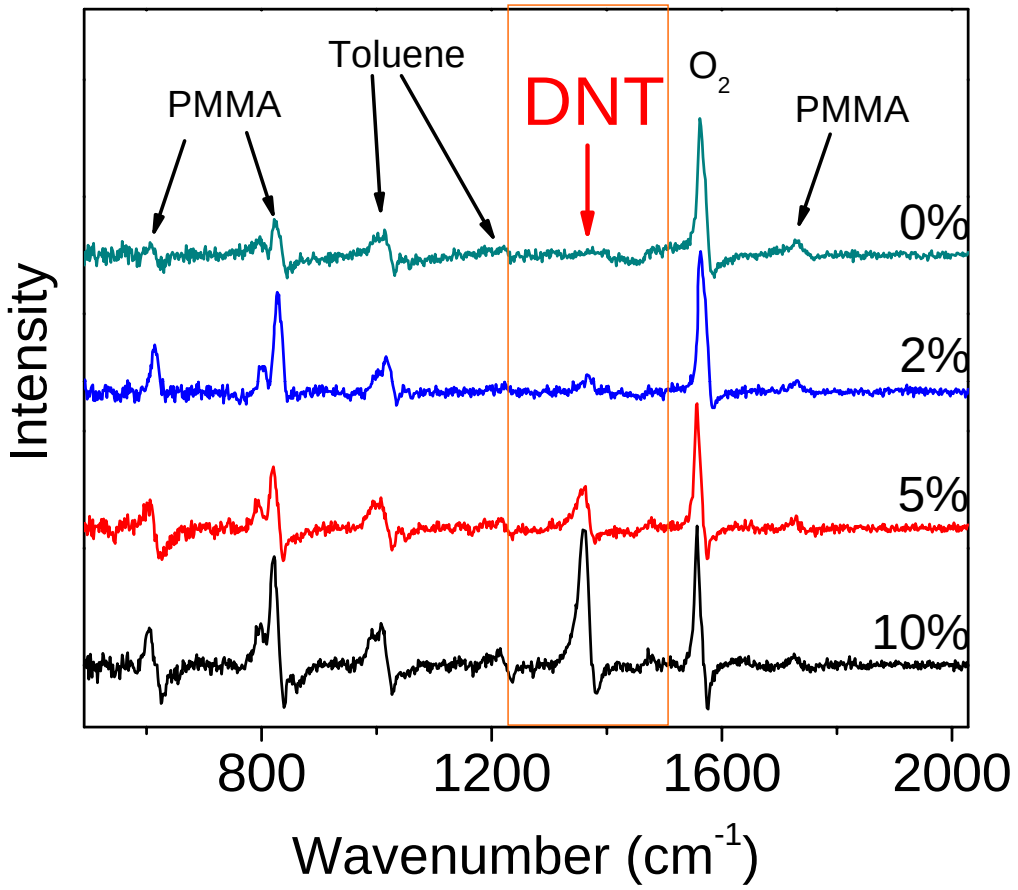
1s exposure, <4mW average laser power (Raw Data)



< 5µm films



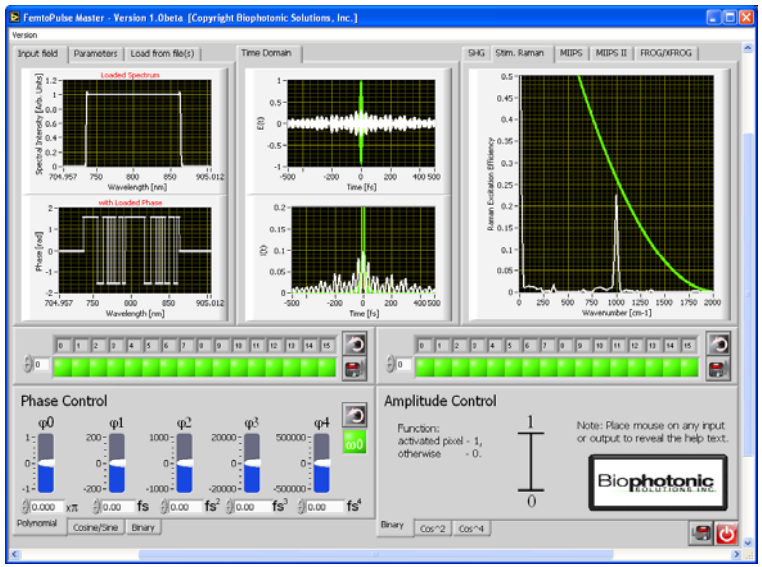
2.8µm films of PMMA

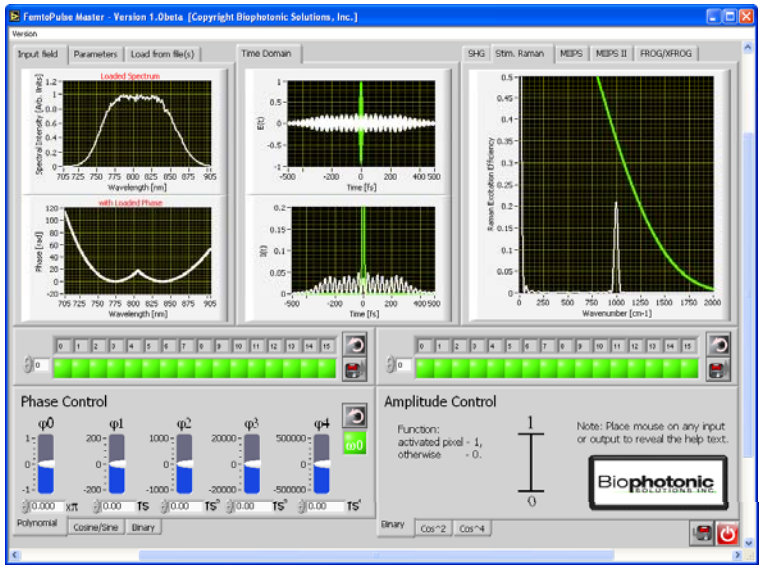


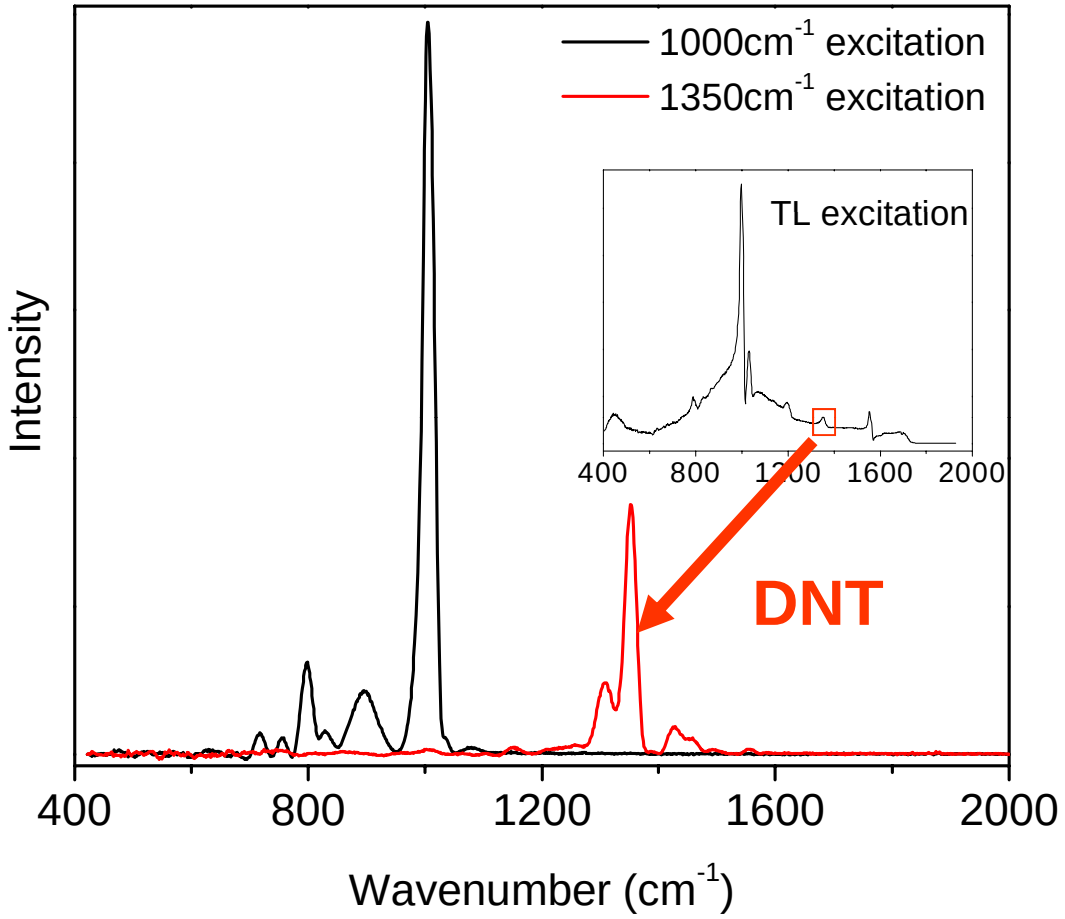
<3 µg/cm² DNT detection at 1 meter, in a complex background

SUPER-CARS Selectivity for standoff imaging

<4mW average laser power







SUPER-CARS Selective excitation ideal for Standoff Imaging Applications

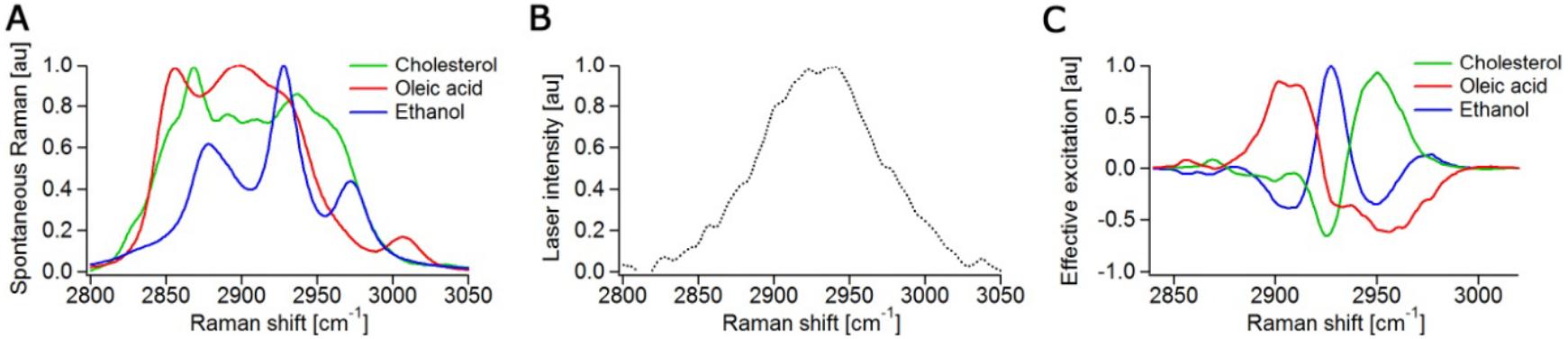
Used by Bartels, *et al* for CARS

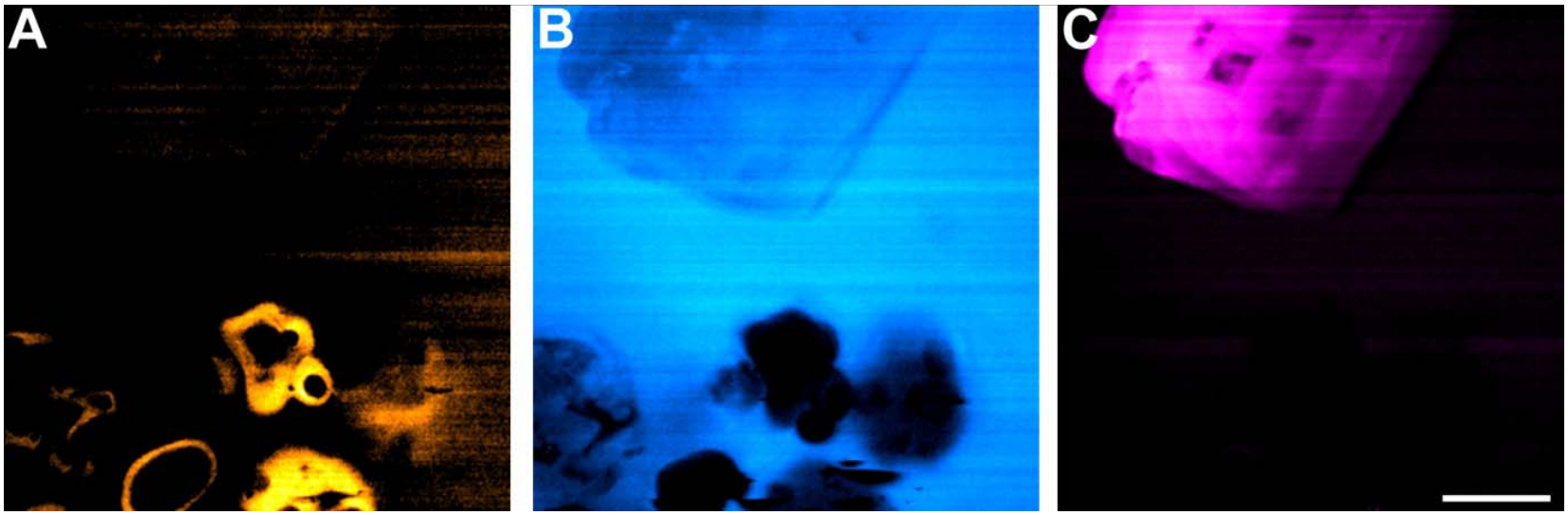
Phys. Rev. Lett. **88**, (2002) in multi-beam geometry. GA selective vib. excitation by Bucksbaum.

Spectral imaging by stimulated Raman scattering

Christian Freudiger, Sunney Xie, Nature Photonics (submitted 2010)

Collaboration with Bingwei Xu, and Marcos Dantus





Milestones in Quantum Control

- Young's double-slit experiment (1803), although it is just linear interference

$$P \propto \left| e^{i\phi_1} + e^{i\phi_2} \right|^2 \propto \cos\left(\phi_1 - \phi_2\right)$$

- Zewail proposes femtosecond lasers will be able to beat IVR (1980)

- Tannor Kosloff and Rice propose selectivity via control of wave packet evolution (1985-6)

Re Optimization: This result has analogs in electrical engineering and information theory, where it is part of matched filter theory.

Variational optimization of the shape, duration, and separation of the pulses used to generate the selectivity of reactivity, and analysis of the changes induced by Inclusion of all degrees of freedom of the molecule (say in the sense of a reaction path Hamiltonian).

- Brumer and Shapiro propose one vs. three-photon quantum control (1986)

$$P \propto \left| e^{i\phi_3} + e^{i3\phi_1} \right|^2 \propto \cos\left(\phi_3 - 3\phi_1\right)$$

- Dantus, Rosker and Zewail Femtosecond Transitions State Spectroscopy (1987)

- Kosloff, Rice, Gaspard, Tersigni, and Tannor propose achieving chemical selectivity by shaping of intense femtosecond pulses, and introduce an iteration procedure for optimization of the pulse shapes (1989).

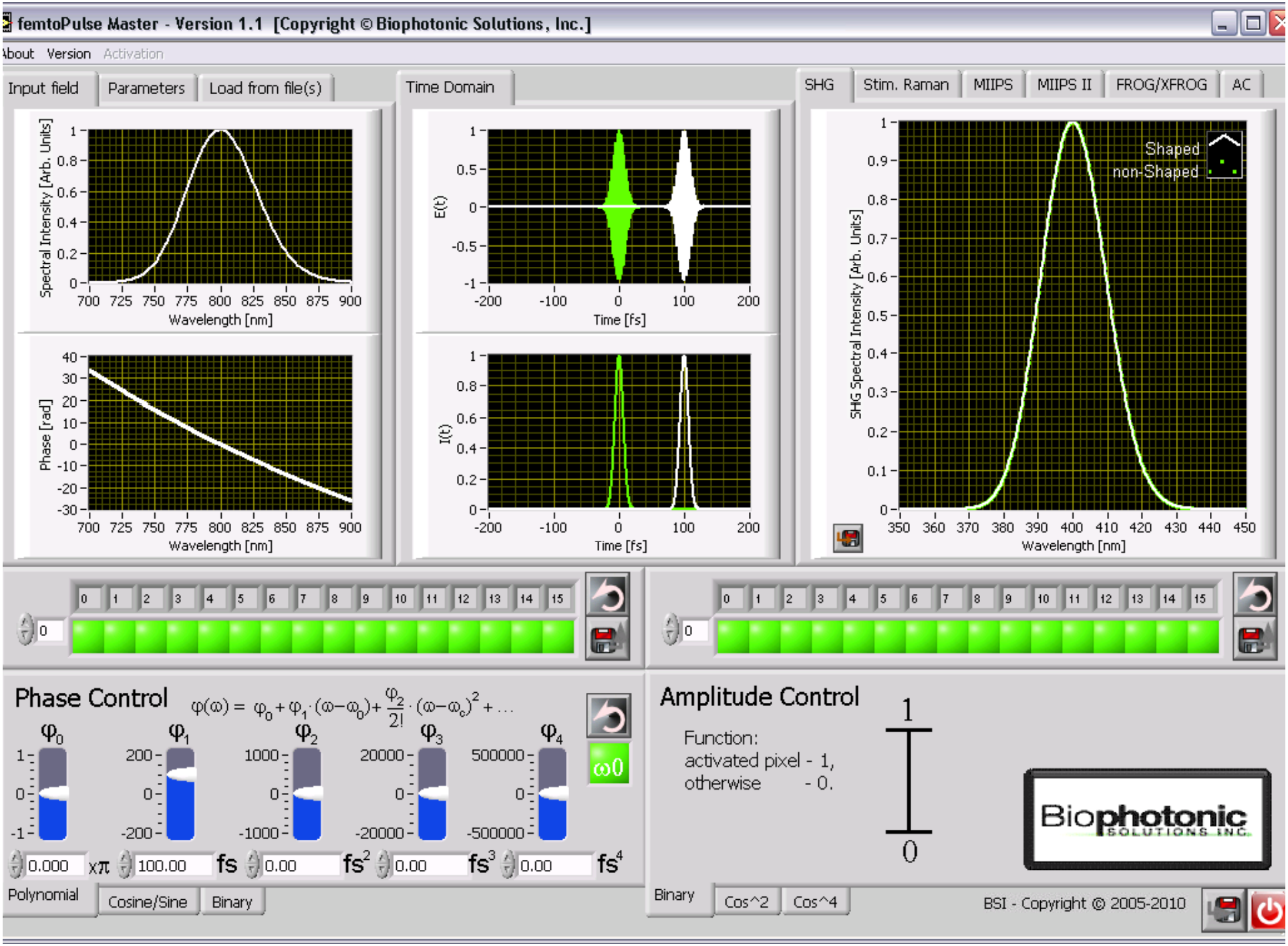
- Bergmann et al. Adiabatic population transfer driven by delayed pulses proposed (1989)

Milestones in Quantum Control

- Chelkowski, Bandrauk Corkum propose and Boers, van Linden van der Heuvell and Noordam demonstrate efficient population transfer in a three-level ladder system by using chirped pulses ChiRAP (1990)
- Judson and Rabitz propose using a learning algorithm to control molecules (1992)
- Bergmann et al. STIRAP experimentally demonstrated (1993)
- Cerullo, Bardeen, Wang and Shank chirped pulse excitation of molecules in solution (1996)
- Gordon Experimental demonstration of one vs. three-photon control (1997)
- Weinacht, Ahn and Bucksbaum “read-write” on sculpted Rydberg wave packet (1998)
- Pastirk, Brown, Zhang and Dantus chirp control of a chemical reaction (1998)
- Silberberg demonstrates multi-chromatic two photon control (1998)

$$P(2\omega) \propto \left| 1 + e^{i\phi(\omega-\Delta)+i\phi(\omega+\Delta)} \right|^2$$

- GA assisted pulse compression (1997)
- Assion, Baumert, Bergt, Brixner, Kiefer, Seyfried, Strehle, Gerber Control of chemical reactions by feedback-optimized phase shaped laser pulses (1998)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Input field
Parameters
Load from file(s)
Spectral Intensity [Arb. Units]
Wavelength [nm]
Phase [rad]
Time Domain
E(t)
I(t)
Time [fs]
SHG
Stim. Raman
MIIPS
MIIPS II
FROG/XFROG
AC
Shaped
non-Shaped
SHG Spectral Intensity [Arb. Units]
Phase Control
$\varphi(\omega) = \varphi_0 + \varphi_1 \cdot (\omega-\omega_0) + \frac{\varphi_2}{2!} \cdot (\omega-\omega_c)^2 + \ldots$
φ_0
φ_1
φ_2
φ_3
φ_4
0.000 xπ
100.00 fs
0.00 fs²
0.00 fs³
0.00 fs⁴
ω0
Polynomial
Cosine/Sine
Binary
Amplitude Control
Function:
activated pixel - 1,
otherwise - 0.
Binary
Cos^2
Cos^4
Biophotonic SOLUTIONS INC.
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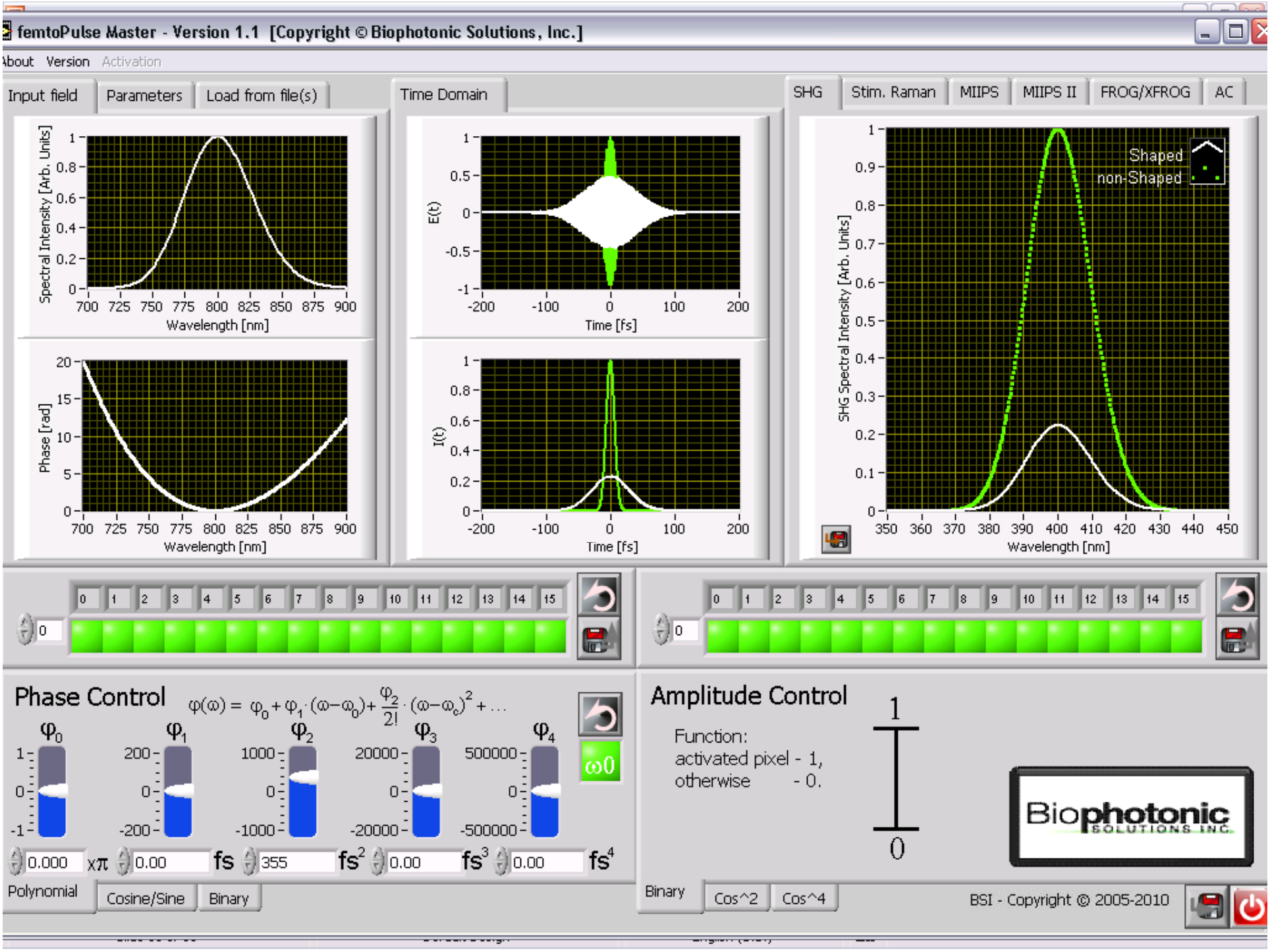
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About Version Activation
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Phase [rad]
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Phase Control
$\varphi(\omega) = \varphi_0 + \varphi_1 \cdot (\omega-\omega_0) + \frac{\varphi_2}{2!} \cdot (\omega-\omega_c)^2 + \ldots$
φ_0
φ_1
φ_2
φ_3
φ_4
0.000 xπ
0.00 fs
355 fs²
0.00 fs³
0.00 fs⁴
ω0
Polynomial
Cosine/Sine
Binary
Amplitude Control
Function:
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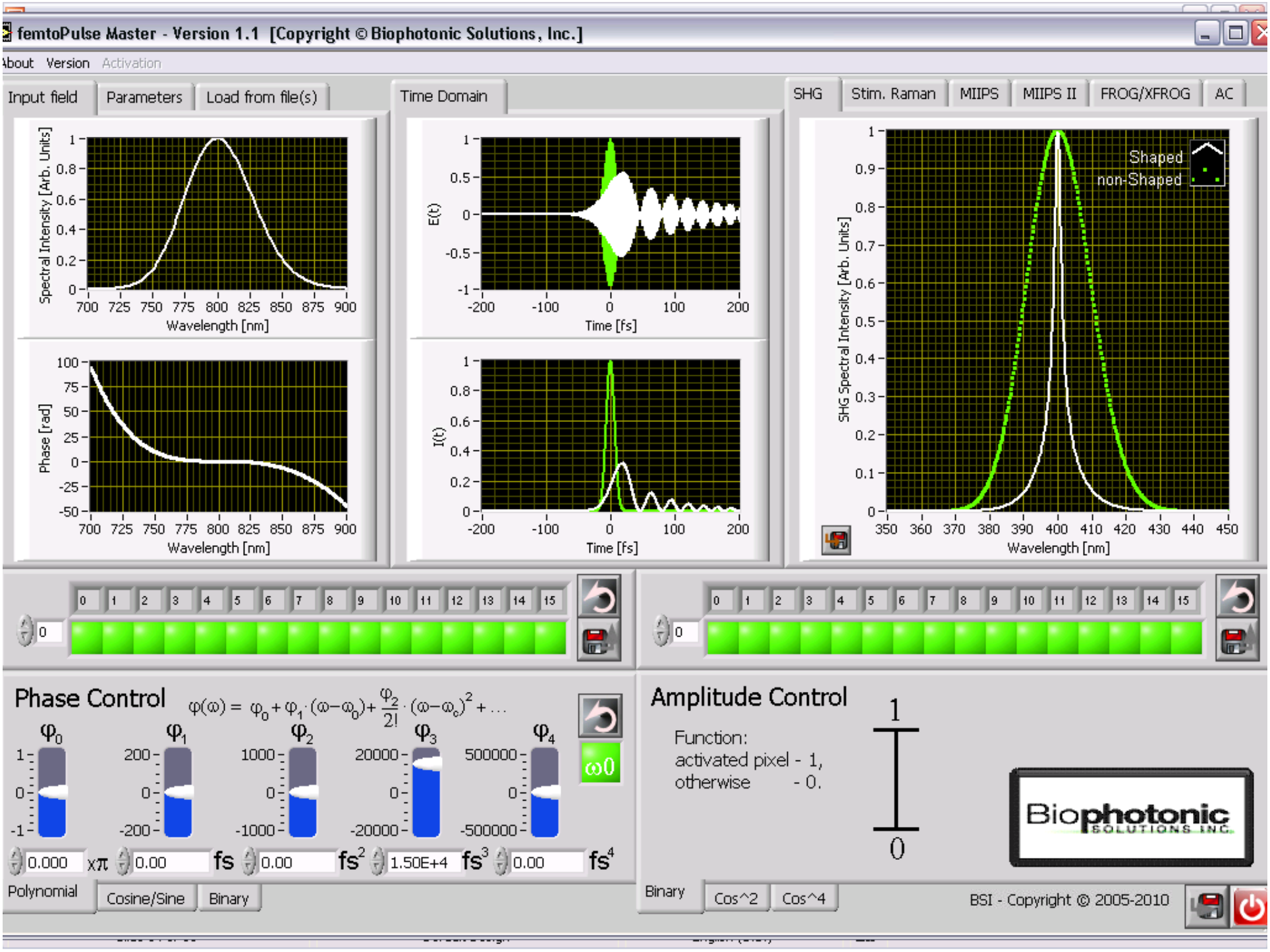
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AC
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non-Shaped
SHG Spectral Intensity [Arb. Units]
Phase Control
$\varphi(\omega) = \varphi_0 + \varphi_1 \cdot (\omega-\omega_0) + \frac{\varphi_2}{2!} \cdot (\omega-\omega_0)^2 + \ldots$
φ_0
φ_1
φ_2
φ_3
φ_4
ω0
0.000 xπ
0.00 fs
0.00 fs²
1.50E+4 fs³
0.00 fs⁴
Polynomial
Cosine/Sine
Binary
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otherwise - 0.
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Cos^4
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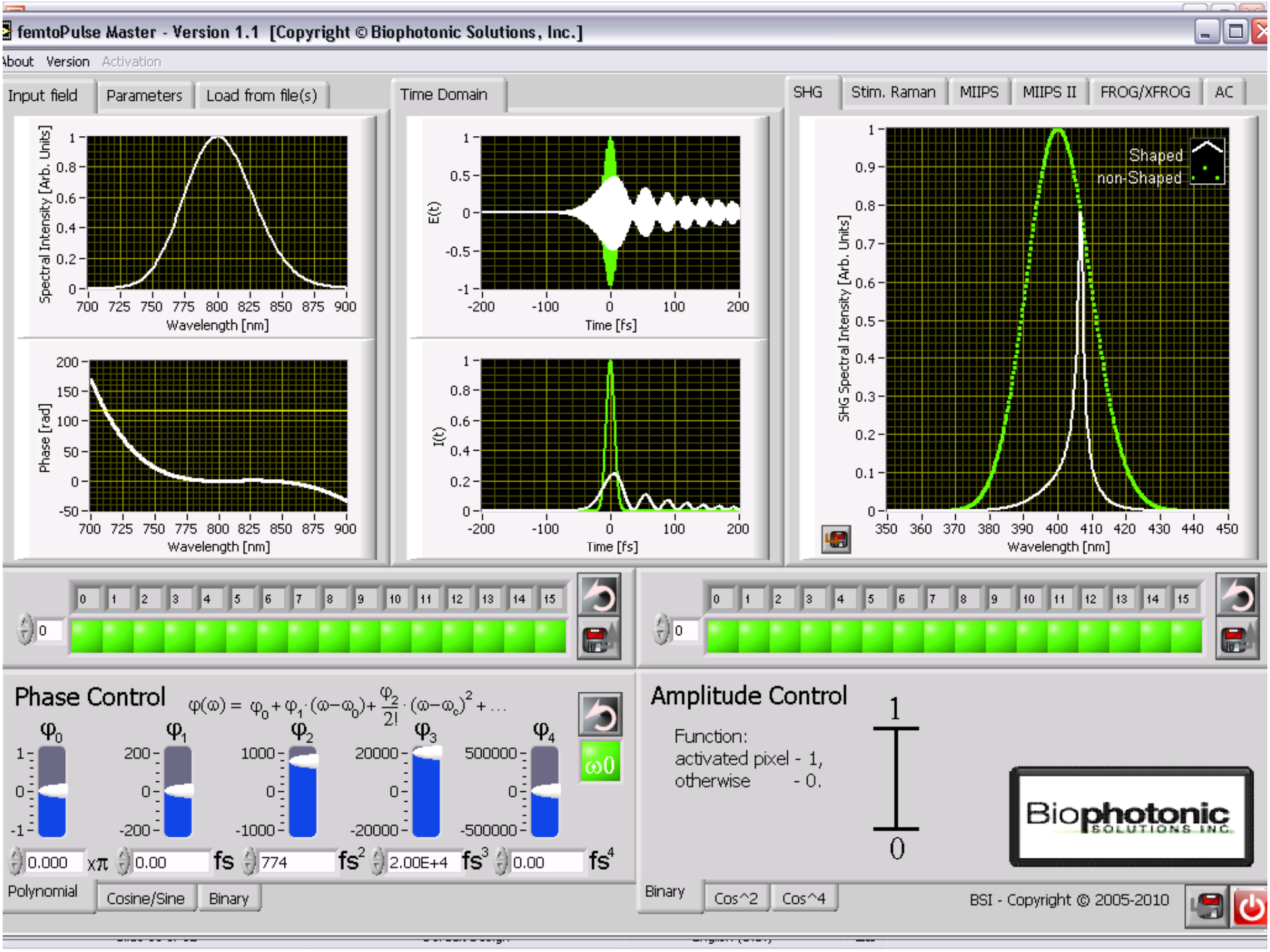
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E(t)
Time [fs]
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Shaped
non-Shaped
SHG Spectral Intensity [Arb. Units]
Phase Control
$\varphi(\omega) = \varphi_0 + \varphi_1 \cdot (\omega-\omega_0) + \frac{\varphi_2}{2!} \cdot (\omega-\omega_c)^2 + \ldots$
φ_0
φ_1
φ_2
φ_3
φ_4
0.000 xπ
0.00 fs
774 fs²
2.00E+4 fs³
0.00 fs⁴
ω0
Polynomial
Cosine/Sine
Binary
Amplitude Control
Function:
activated pixel - 1,
otherwise - 0.
Binary
Cos^2
Cos^4
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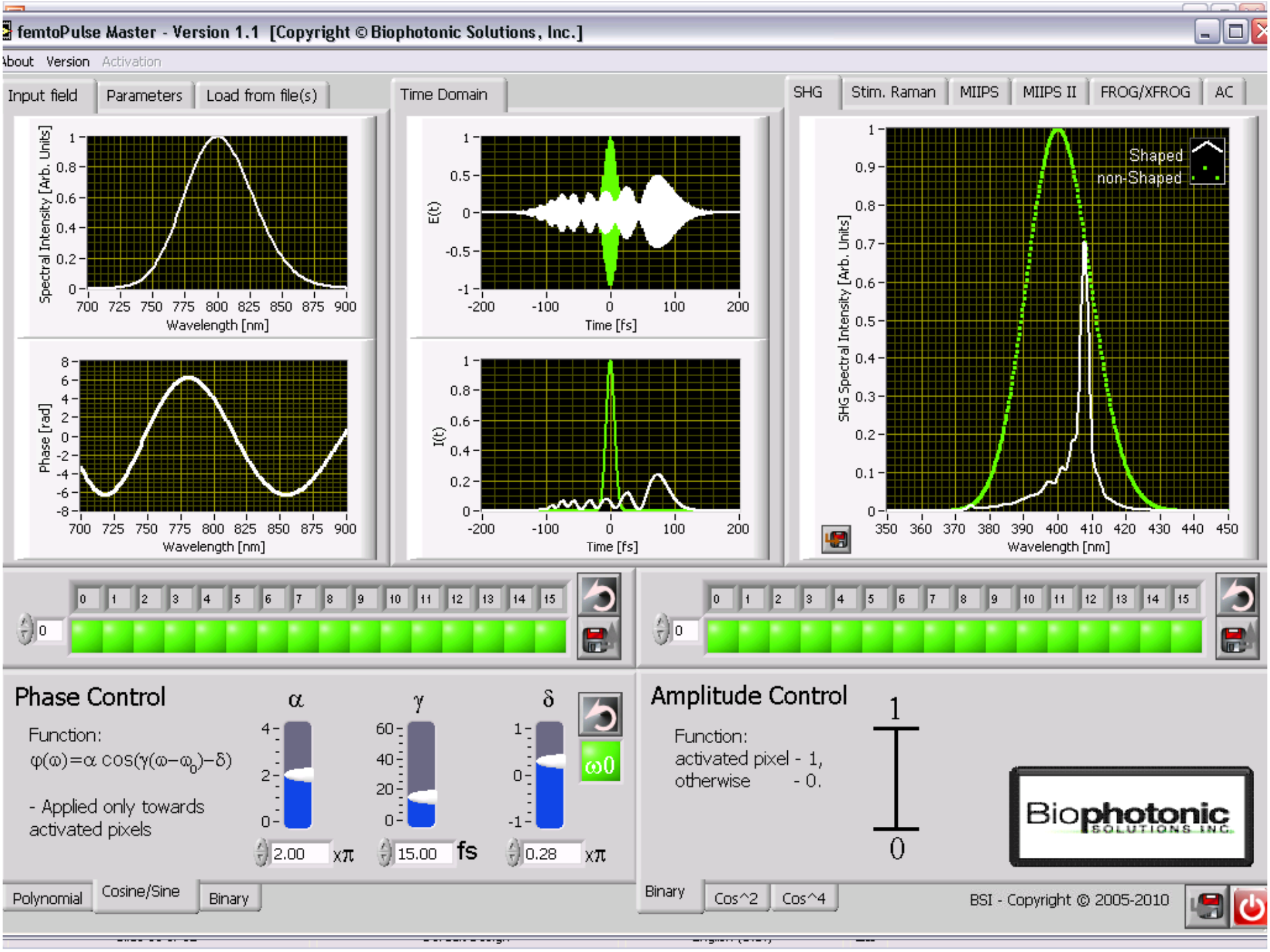
femtoPulse Master - Version 1.1 [Copyright © Biophotonic Solutions, Inc.]
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Load from file(s)
Time Domain
SHG
Stim. Raman
MIIPS
MIIPS II
FROG/XFROG
AC
Spectral Intensity [Arb. Units]
Wavelength [nm]
Phase [rad]
E(t)
I(t)
Time [fs]
Shaped
non-Shaped
SHG Spectral Intensity [Arb. Units]
Phase Control
Function:
φ(ω)=α cos(γ(ω−ω0)−δ)
- Applied only towards activated pixels
α
γ
δ
2.00 xπ
15.00 fs
0.28 xπ
ω0
Polynomial
Cosine/Sine
Binary
Amplitude Control
Function:
activated pixel - 1,
otherwise - 0.
1
0
Binary
Cos^2
Cos^4
Biophotonic SOLUTIONS INC.
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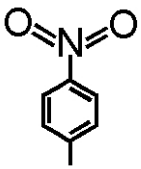
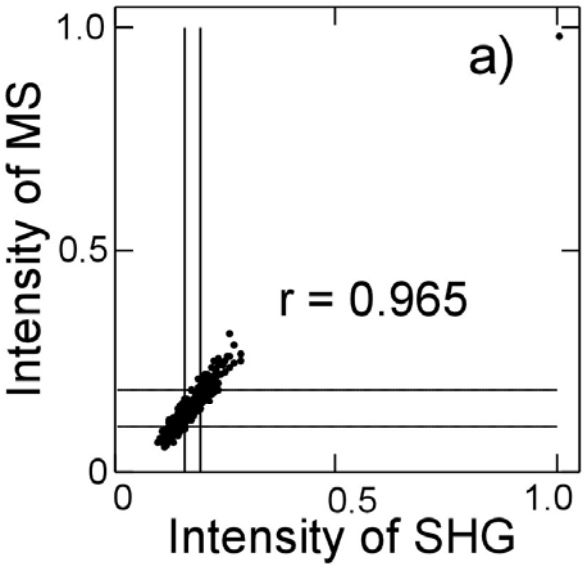
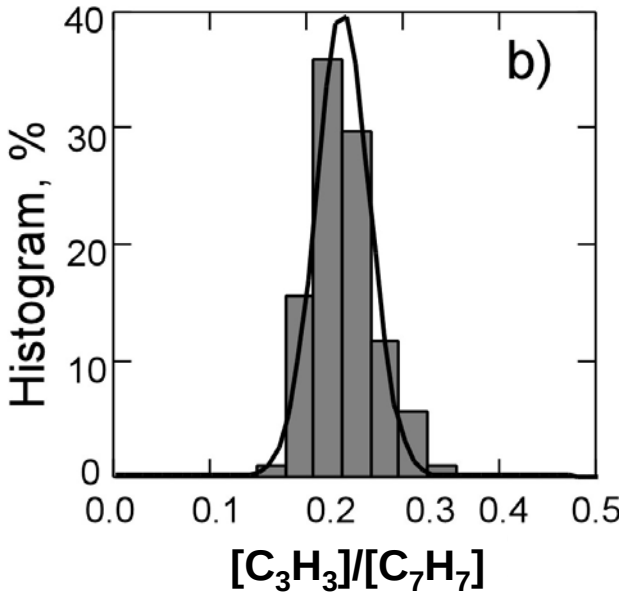
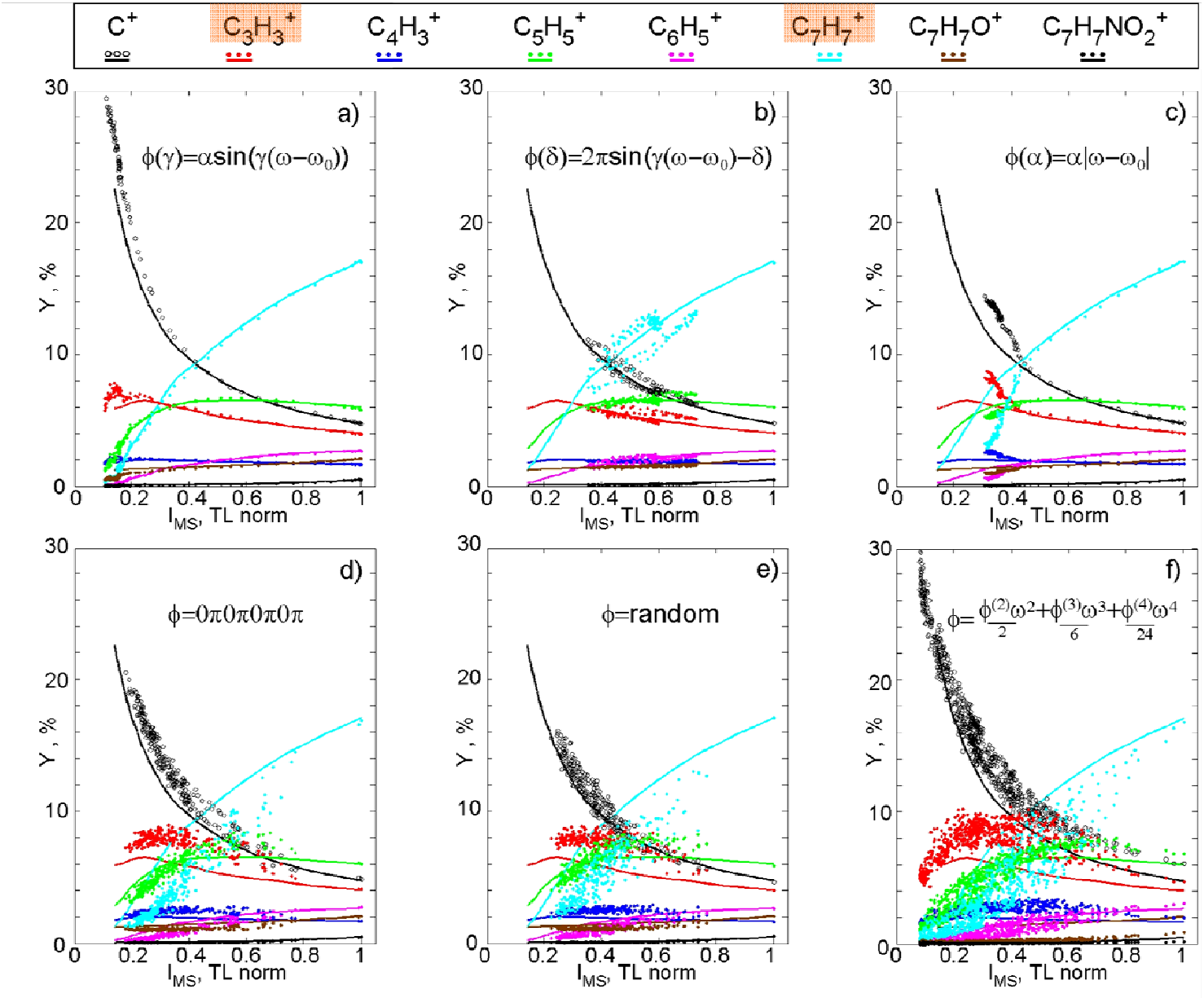
Sample of random 16-bit binary phases



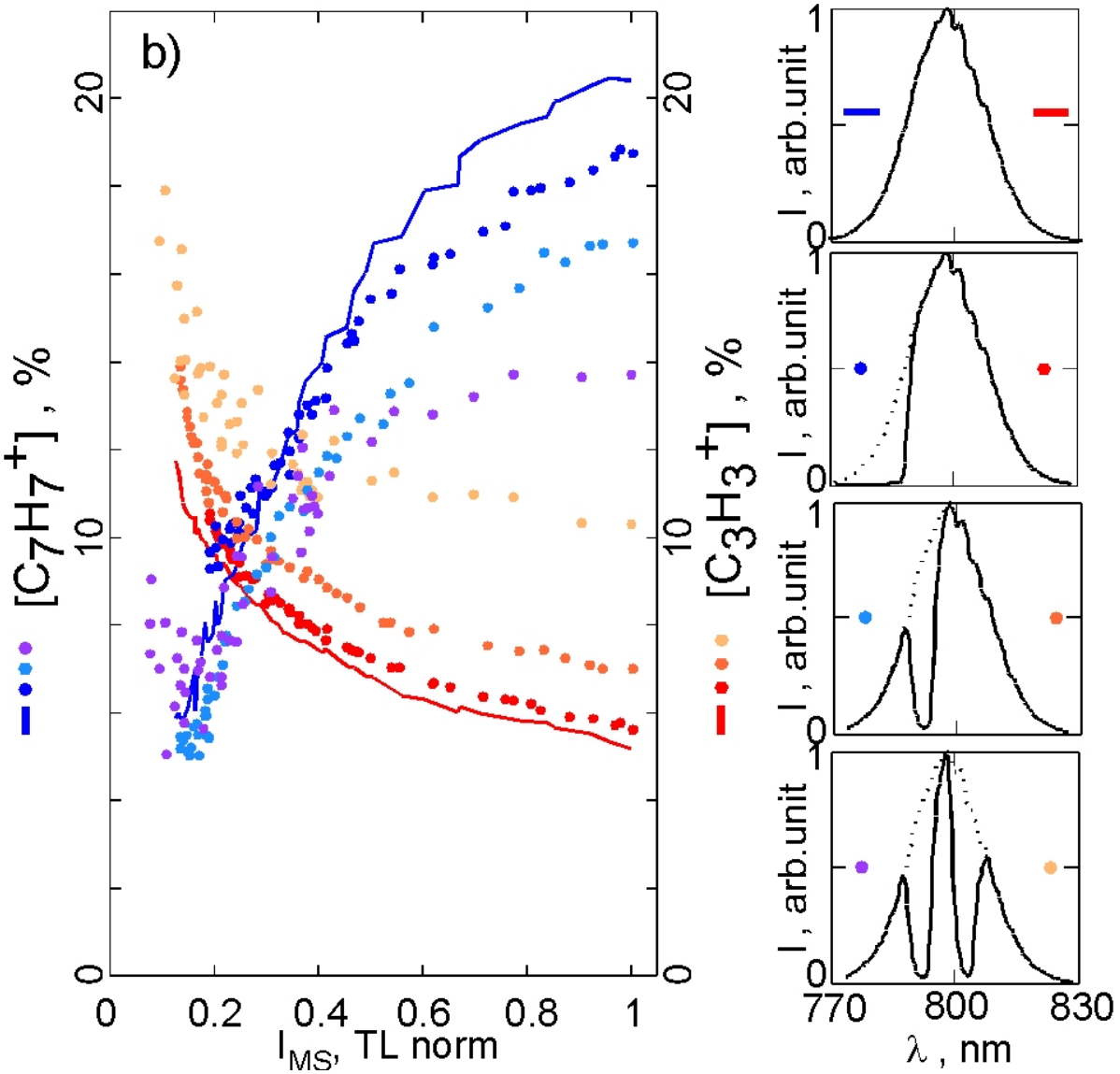




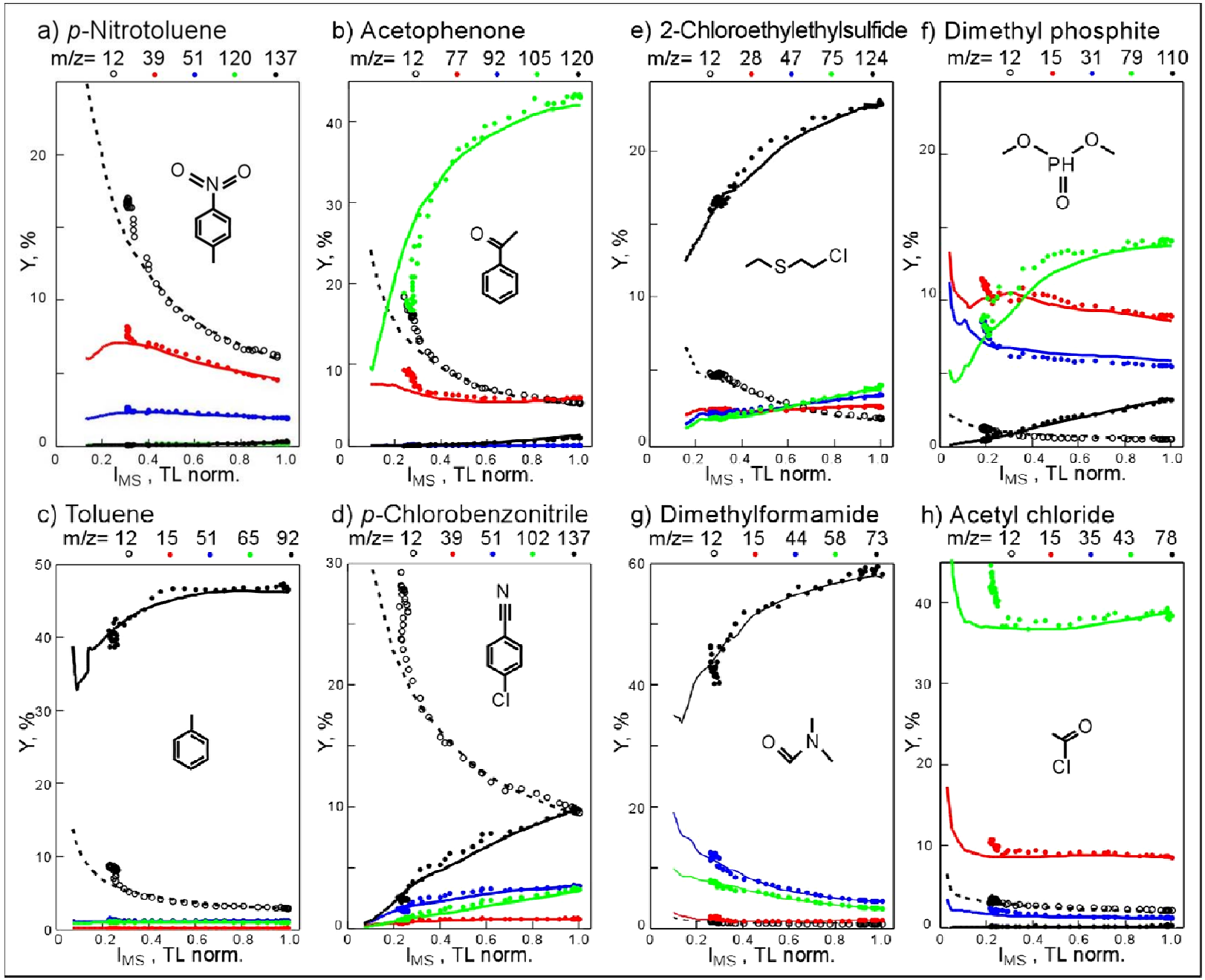
Is molecular response predictable?



Effect of phase & amplitude shaping



Generalizing to other molecules



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VOLUME 112
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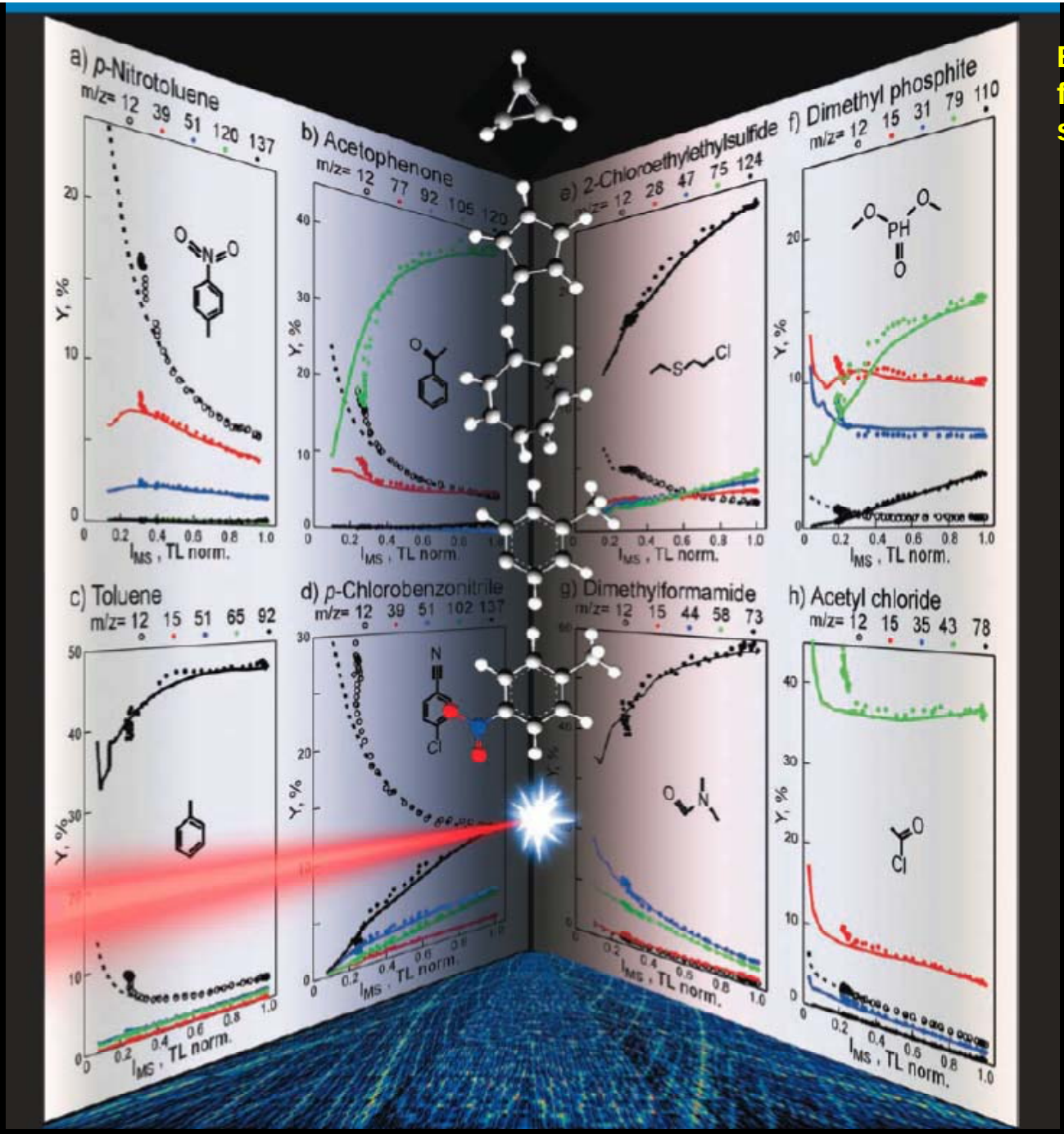
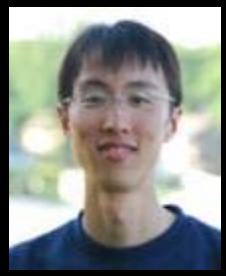
http://pubs.acs.org/JPCA



Back to MS fragmentation in strong fields



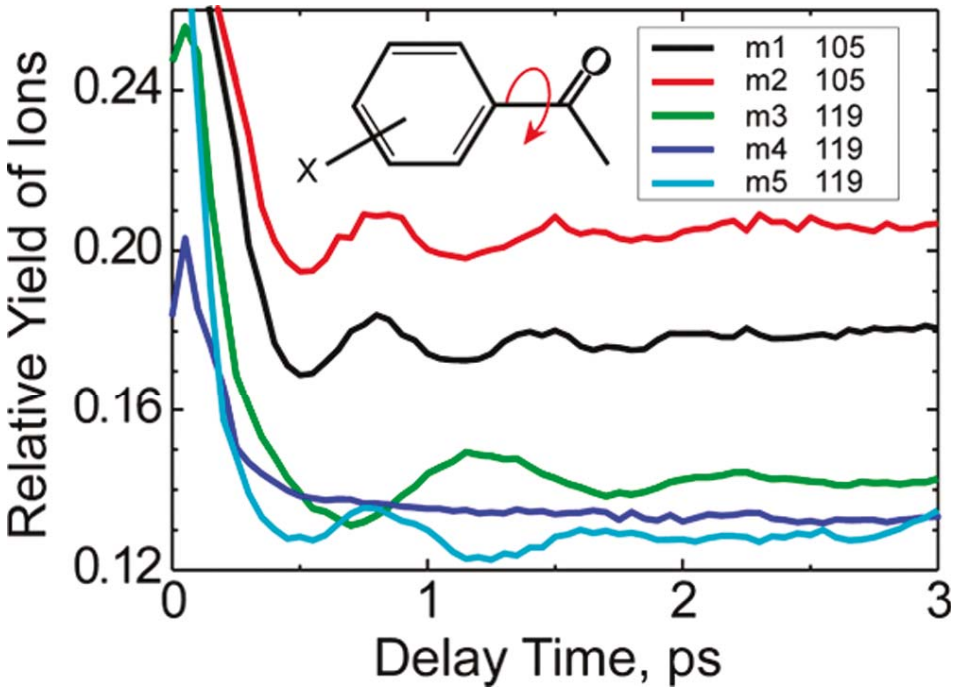
Xin Zhu

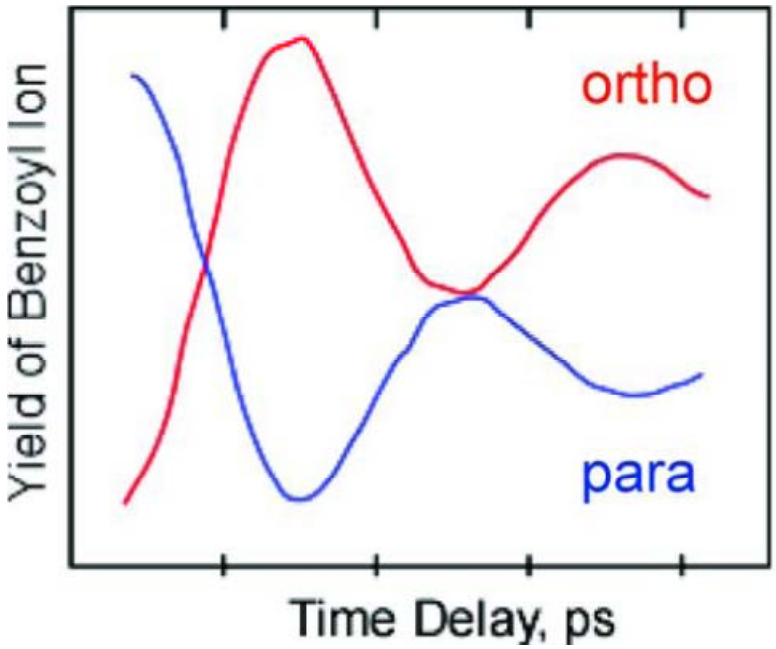
Either no e-coherence or very short lived.

J. Phys. Chem. A. 112, 3789 (2008)

Searching for coherence

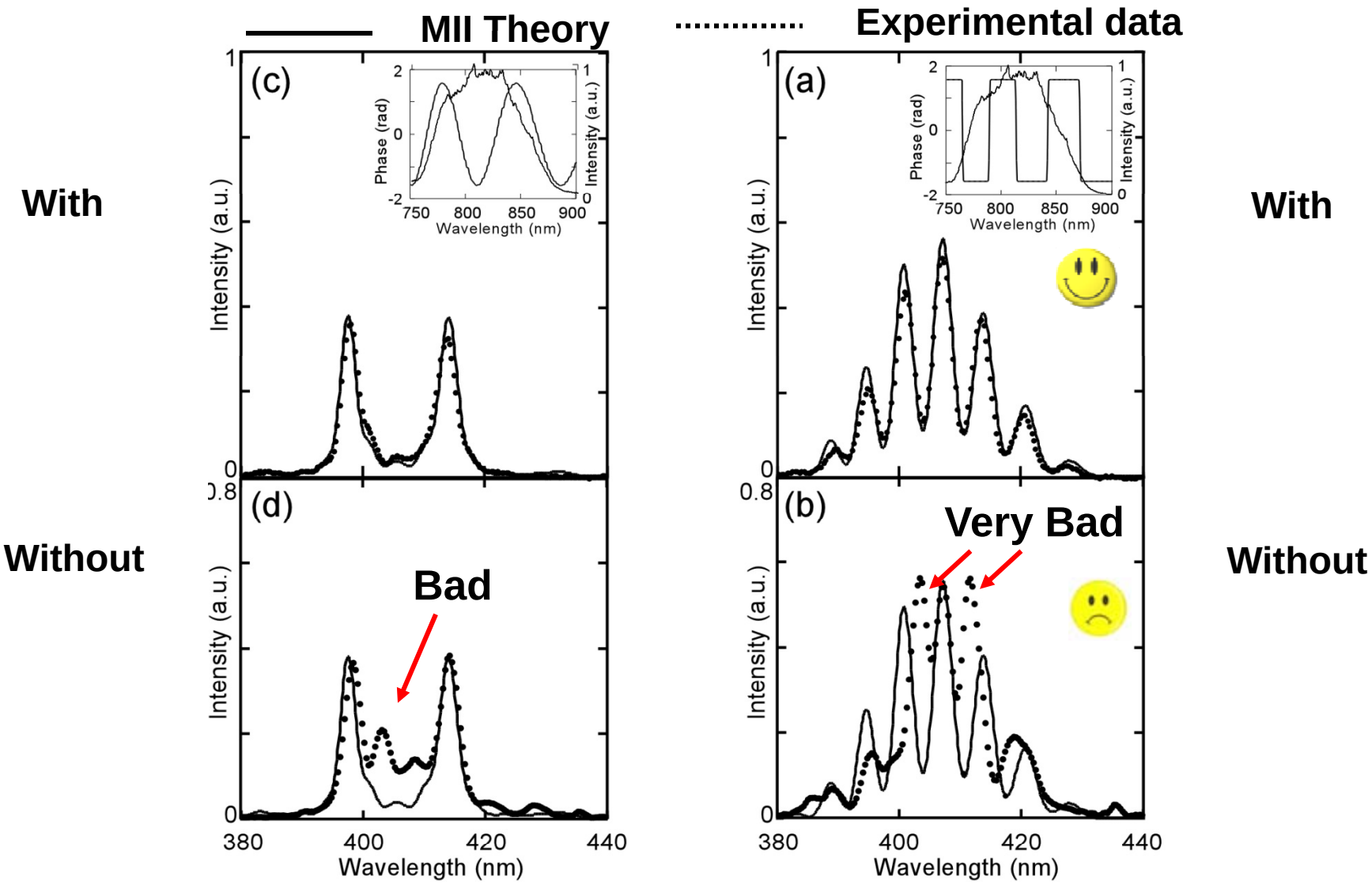
Coherence found on acetophenone





Zhu, X., Lozovoy, V. V., Shah, J. D. & Dantus, M. "Photodissociation Dynamics of Acetophenone and Its Derivatives with Intense Nonresonant Femtosecond Pulses." *J. Phys. Chem. A* **115**, 1305-1312 (2011).

Part III. 6. The need for pulse compression

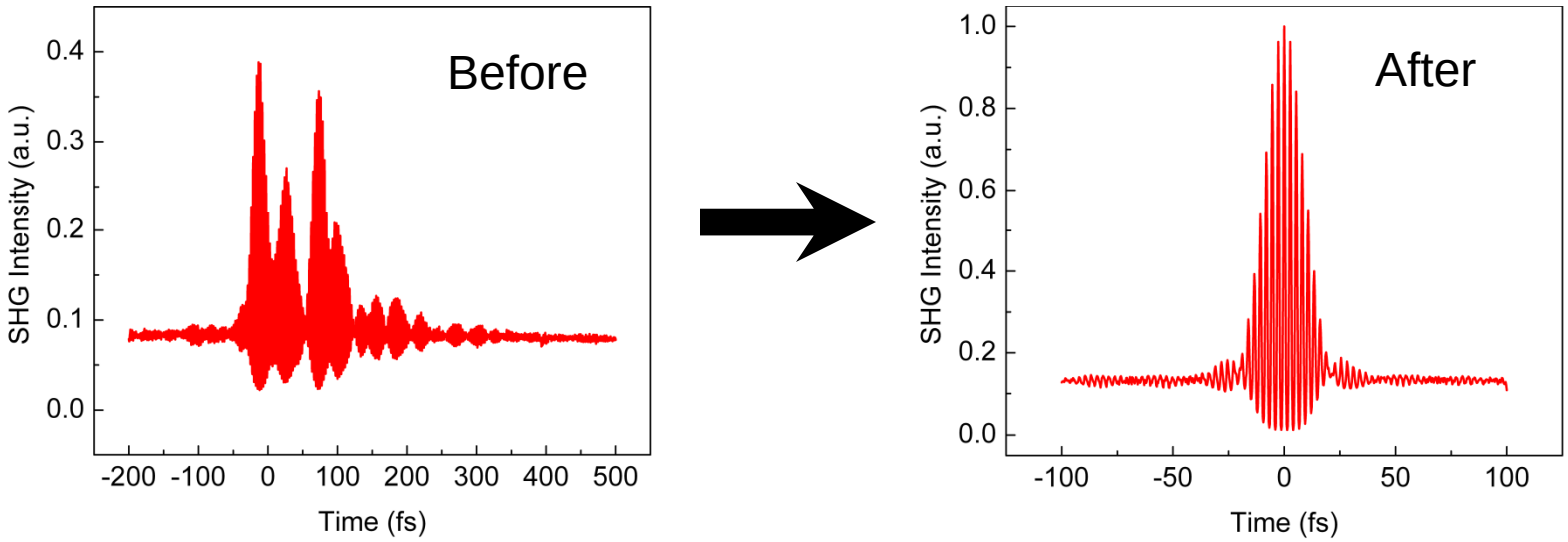


JOSA B **23**, 750 (2006)

Pulse Compression

The goal is to take a broad-bandwidth pulse with highly nonlinear dispersion (quadratic, cubic, etc…) and to compensate the phase distortions to recover a TL pulse.

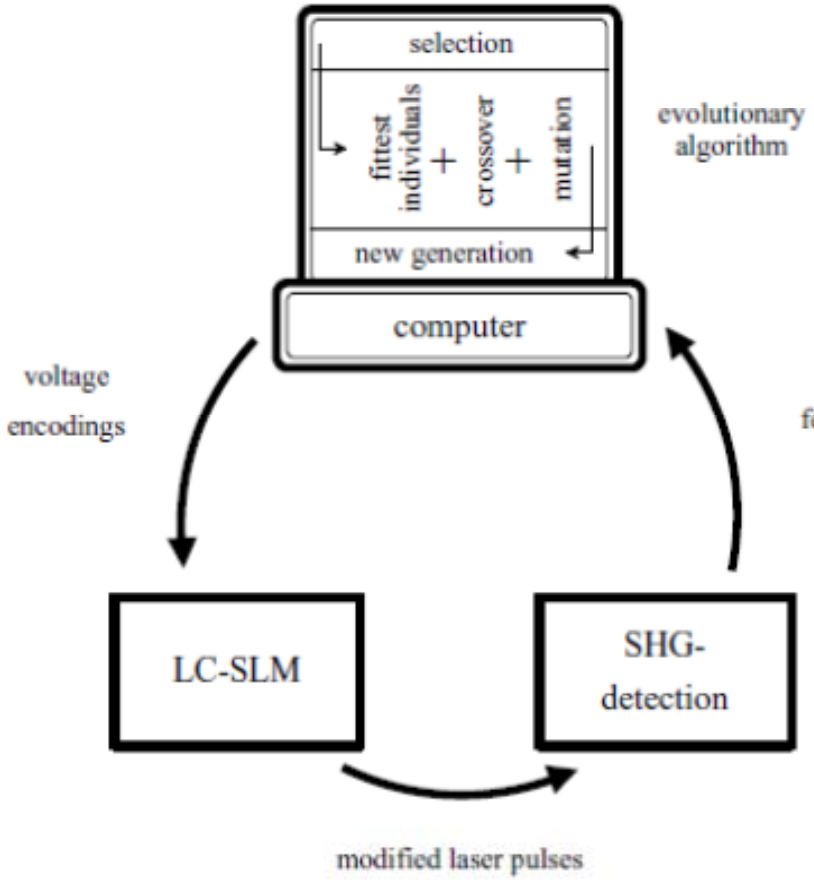
Pulse after single bounce from dielectric coated mirror

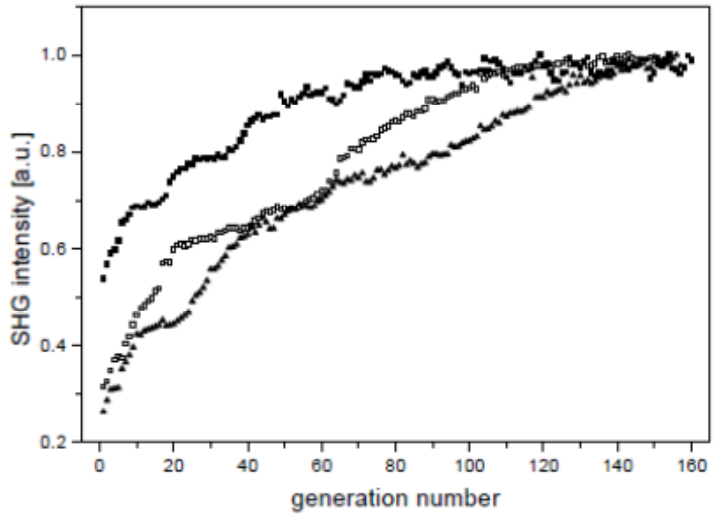


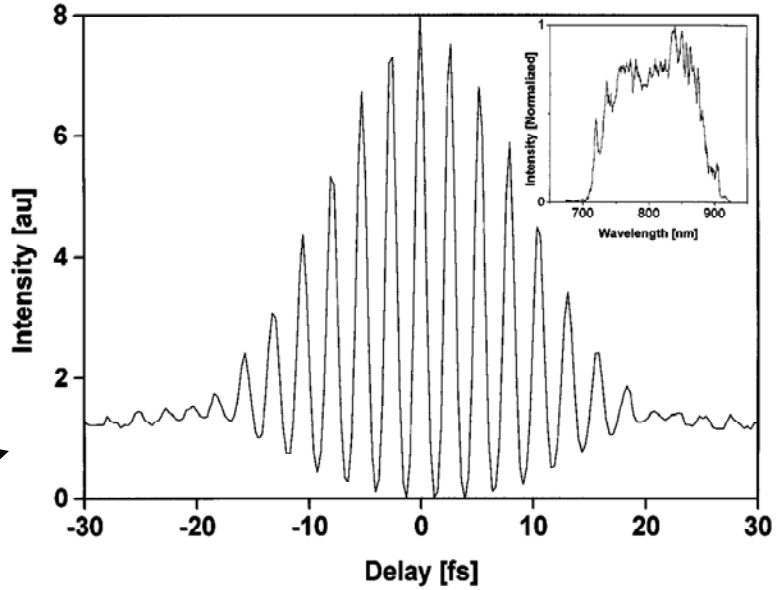
D. Pestov, V. V. Lozovoy, and M. Dantus “Single-beam shaper-based pulse characterization and compression using MIIPS sonogram”, Opt. Letters **35**, 1422-1424 (2010)

GA. Pulse Compression

Works but it is slow and not very accurate because search space near TL is flat







Optics Letters **22**, 1793 (1997)

Appl. Phys. B. **65**, 779 (1997)

About evolutionary learning algorithms

The search space = $(PxA)^N$

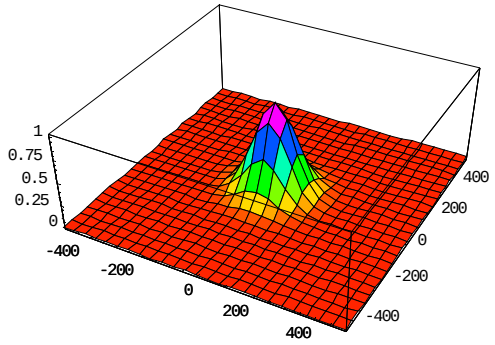
N = # of pixels
P = # of phases
A = # Amplitudes

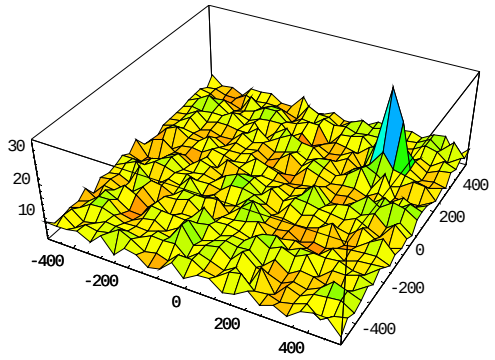
For:
N = 100
P = 100
A = 10
Search space = 10^{300}

10^{200} for phase only

Limited application in the field!

- Size of parameter space vs. sampling (10^8 per day, 10^{12} per life)
- Low repeatability and portability of results
- The search space: Is it convex or needle in the haystack?





Conclusion:
We need a systematic approach