Workshop on Time Domain Science Using X-ray Techniques
August 29 – September 1, 2004,
The Abbey, Fontana, Lake Geneva Area, Wisconsin

Coherent Laser Control of Physicochemical Processes

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The Goal: Control of nonlinear laser-molecule interactions

- Optimization of x-ray pulse sources
- Optimized laser excitation for PP exp.

- Principles of coherent control and GA optimization

- Phase characterization and compensation

Multiphoton microscopy

Photodynamic therapy

Photo microlithography

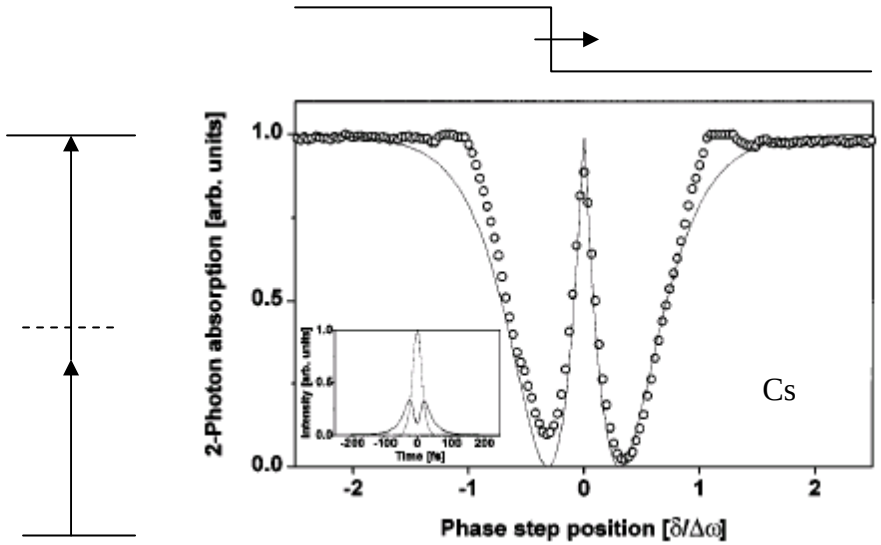
Optical switching

Functional Imaging

Optical distortion (suppression)

Chemical micro-environment probing

Coherent control of two-photon absorption: “An Atomic Switch”



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

B. Broers, L. D. Noordam, H. B. van Linden van den Heuvell, Phys. Rev. A **46**, 2749 (1992). Rb

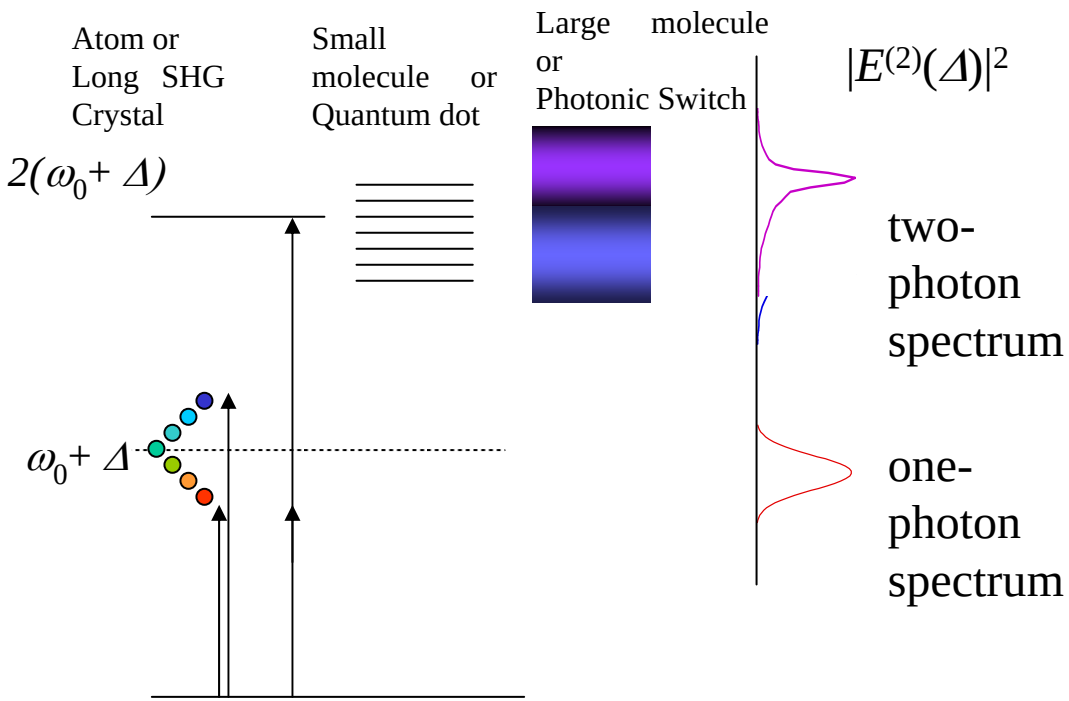
B. Broers, H. B. van Linden van den Heuvell, L. D. Noordam, Phys. Rev. Lett. **69**, 2062 (1992). Rb

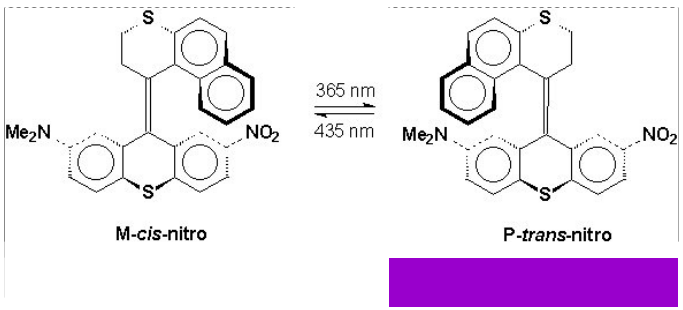
D. Meshulach, Y. Silberberg, Nature **396**, 239 (1998). Cs

D. Meshulach, Y. Silberberg, Phys. Rev. A **60**, 1287 (1999). Cs

T. Hornung, R. Meier, D. Zeidler, K. - L. Kompa, D. Proch, M. Motzkus, Appl. Phys. B **71**, 227 (2000) Na

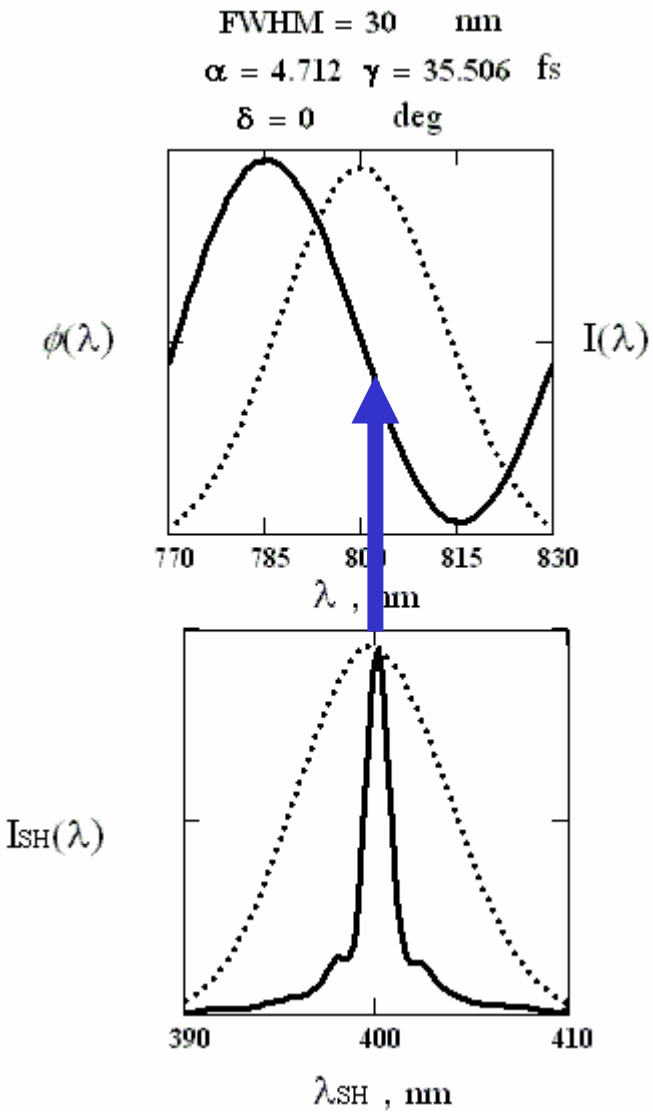
Intrapulse interference in nonlinear optics

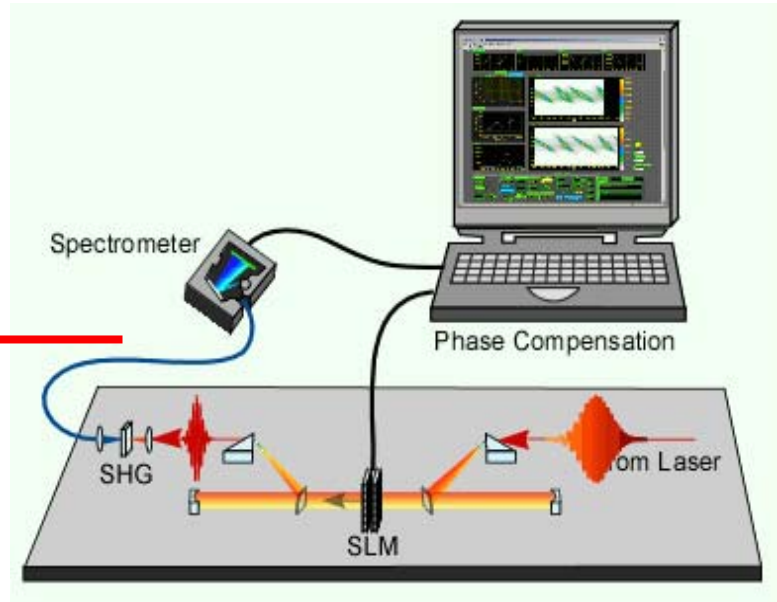




Multiphoton processes with spectral phase modulated pulses

Multiphoton intrapulse interference (MII) in the case of SHG





The effect of spectral phase on second harmonic generation and other nonlinear optical processes.

Experimental results

The Formula

$$S^{(n)} \propto \int_{-\infty}^{\infty} g^{(n)}(\Delta)\left|E^{(n)}(\Delta)\right|^2 d\Delta$$

Frequency integrated signal

Absorption spectrum or phase matching angle

MII second order spectrum

Assumes no dynamics during excitation

Robust control of multiphoton processes in molecules, proteins and nonlinear optical materials

Katherine A. Walowicz, Igor Pastirk, Vadim V. Lozovoy, and Marcos Dantus
J. Phys. Chem. A 106, 9369-9373 (2002)

V. V. Lozovoy, I. Pastirk, K. A. Walowicz, and M. Dantus,
J. Chem. Phys. 118, 3187, (2003)

One photon

Two photon

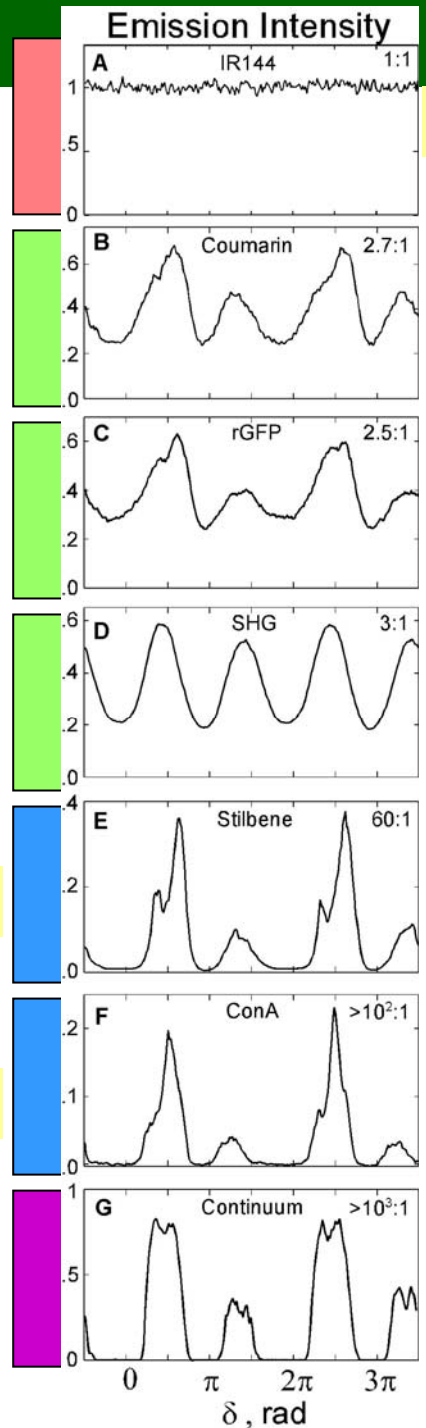
Two photon

Two photon

Three photon

Three photon

>3 photon



No effect

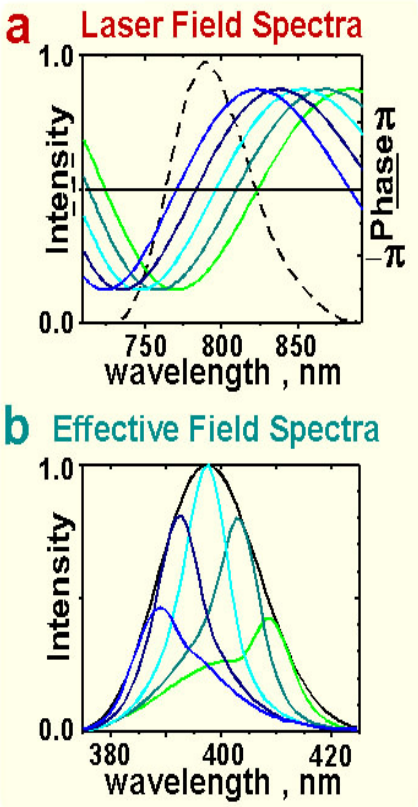
3 to 1 ratio

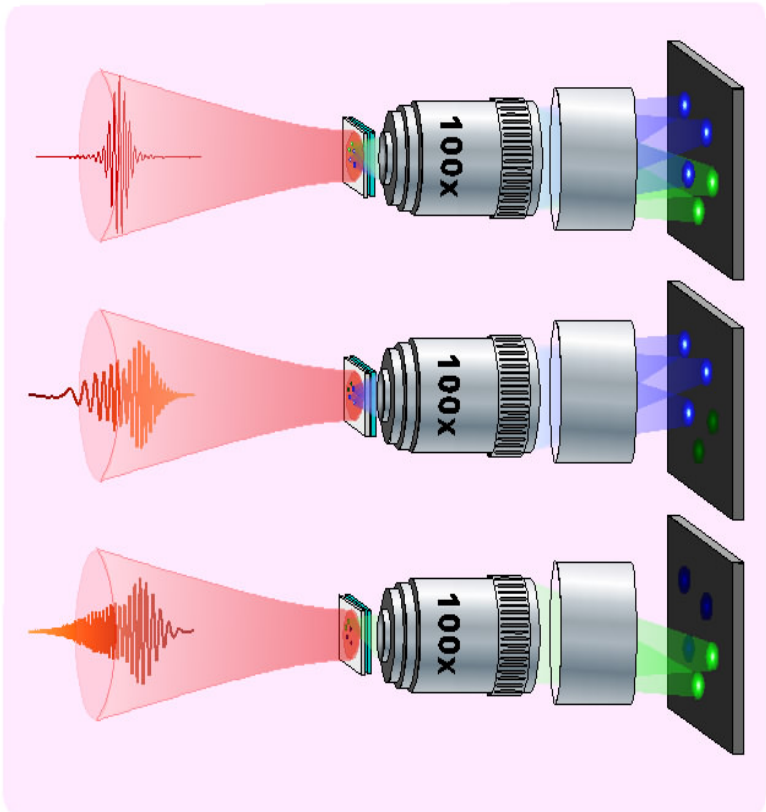
100 to 1 ratio

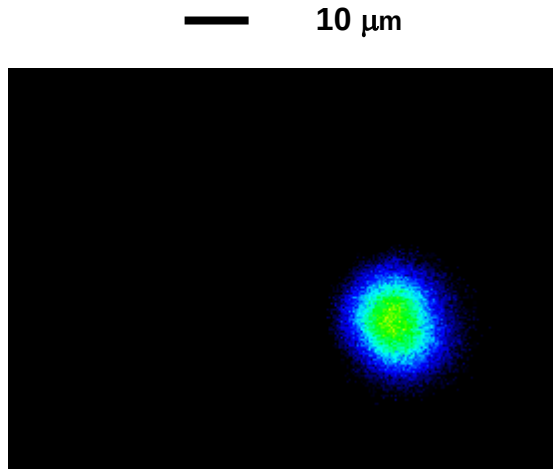
1000 to 1 ratio

Selective control of multiphoton microscopy

Two-photon excitation of microspheres (abs. max. blue 365 nm and green 450 nm) with sinusoidal phase modulated pulses

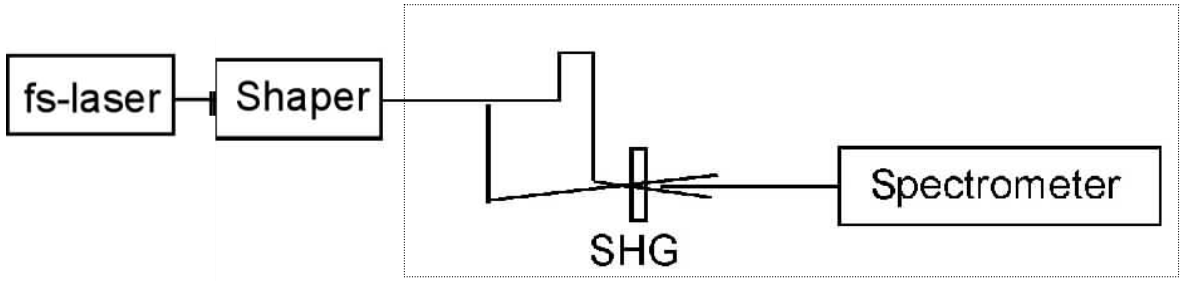






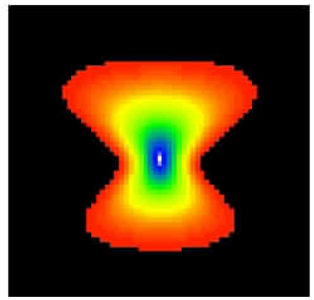
I. Pastirk, J. M. Dela Cruz, K.A. Walowicz, V.V. Lozovoy, M. Dantus *"Selective two-photon microscopy with shaped femtosecond pulses."* **Optics express *2003***, **11**(14), **1695**-1701.

Pulse characterization can be a problem, how can it be simplified?

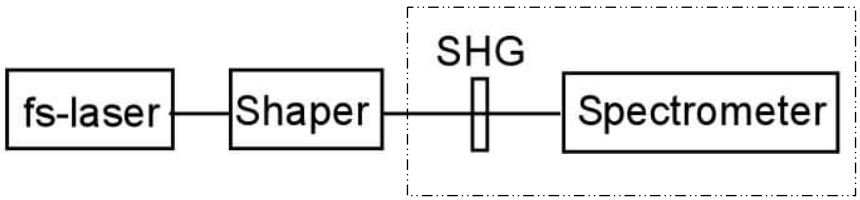


SHG-FROG from R. Trebino

Simulated
SHG-FROG



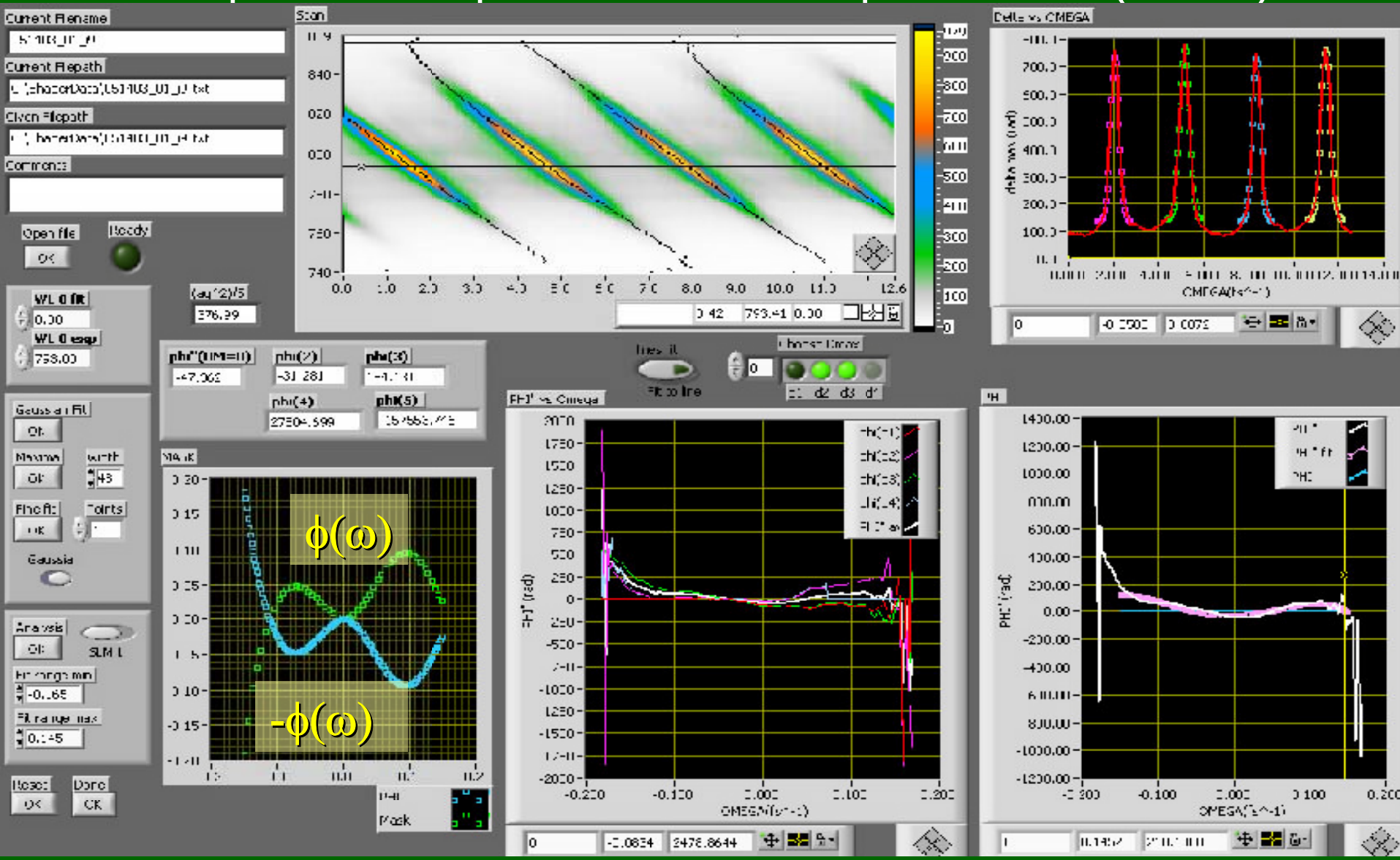
Spectral phase measurement methods, such as FROG have a resolution of ~ .1 radian.



MIIPS setup

I. Pastirk, K.A. Walowicz, V.V. Lozovoy, and M. Dantus, *Optics Letters* 29, 7, 775-777 (2004);
OE Magazine Dec.2002
J. Dela Cruz, I. Pastirk, V.V. Lozovoy, and M. Dantus, J. Phys. Chem. 108, 1, 53 (2004).

Multiphoton intrapulse interference phase scan (MIIPS)



V. V. Lozovoy, I. Pastirk, M. Dantus

"Multiphoton Intrapulse Interference 4. Characterization and compensation of the spectral phase of ultrashort laser pulses."

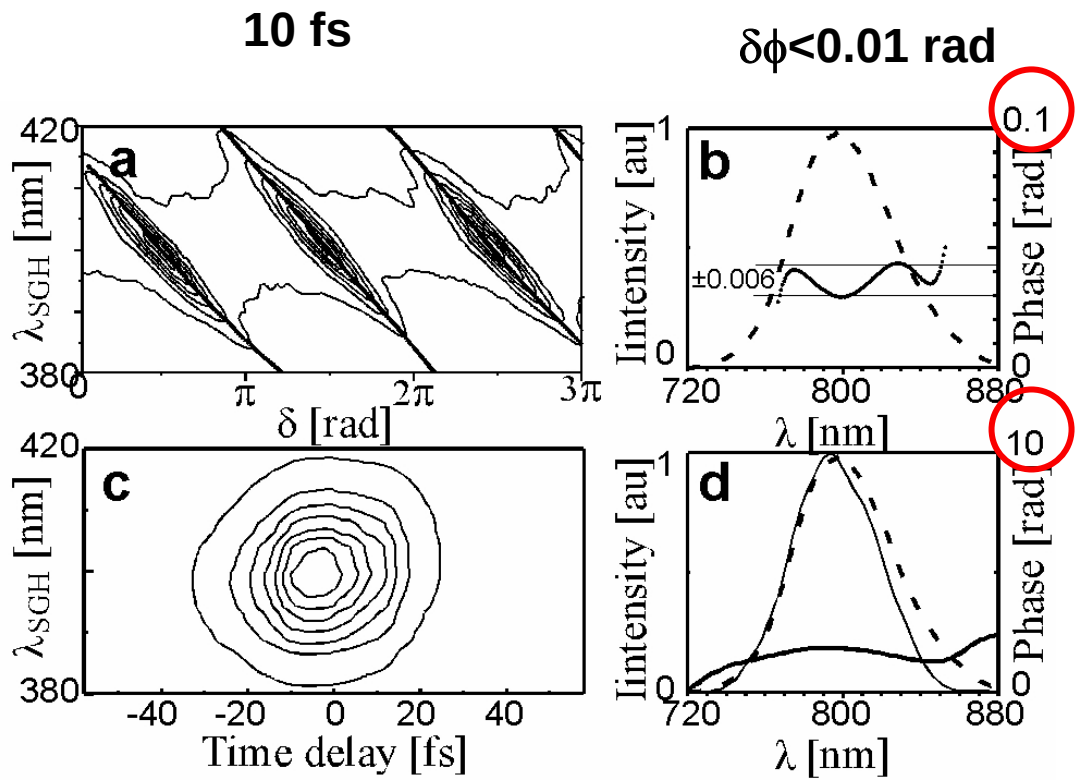
Optics Letters *2004*, 29(7), **775**-777.

Phase compensation

MIIPS of TL pulse

MIIPS

SHG-FROG

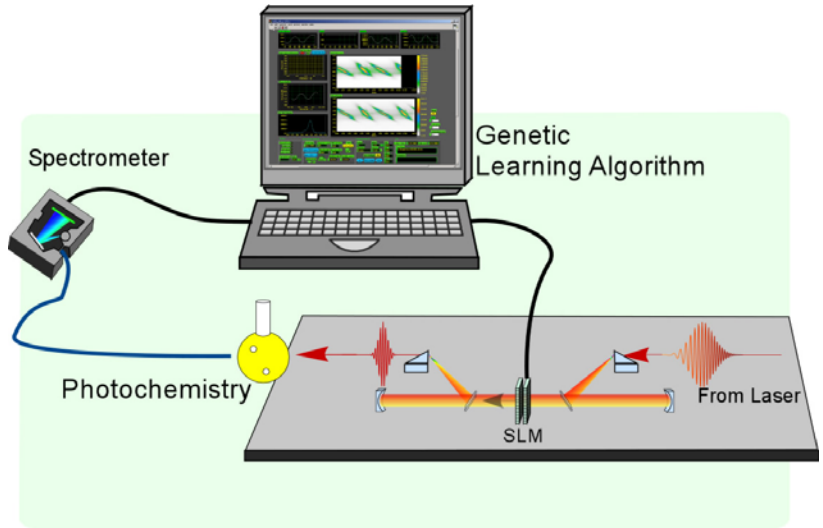


A complete MIIPS setup is available from
Biophotonic Solutions Inc.
info@biophotonicsolutions.com

M. Dantus, V. V. Lozovoy, I. Pastirk
*"Measurement and repair. **The** femtosecond Wheatstone bridge."*
OE magazine *2003*, **3**(9), **15**-17

Optimal control with shaped pluses

"The molecule solves its own Hamiltonian" Hershel Rabitz



Some successes of GA based laser control

Kent Wilson, control of two photon induced fluorescence
David Ritze on second harmonic generation
Gustav Gerber on large molecules (gas and solution)
Phil Bucksbaum on vibrational excitation of methanol
Ludger Wöste on large organometallic molecules (gas)
Robert Levis on large organic molecules (gas)
Kapteyn&Murnane optimization of the 27^{th} Harmonic
Marcus Motzkus on large organic molecules in solution

Evolutionary learning algorithms (ELA)

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

What are the limitations?

- Size of parameter space vs. sampling (10^8 per day)
- Periodicity of the field (2π) leads to redundancies
- Early convergence on local minima
- Low repeatability and portability of results
- Results difficult to understand and simulate

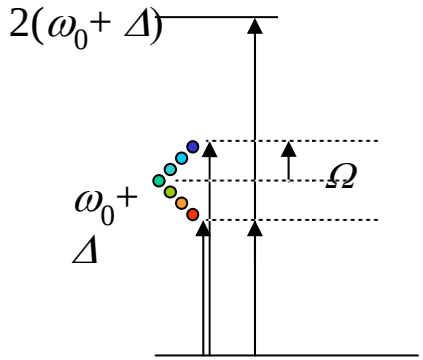
Goals and optimization

The goal: Optimum pulse
Enhances desired process
Suppresses undesired process

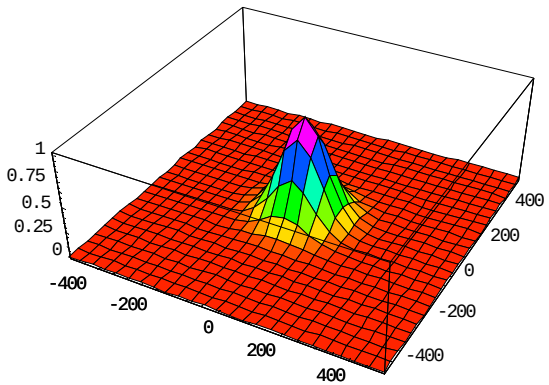
For two-photon transitions

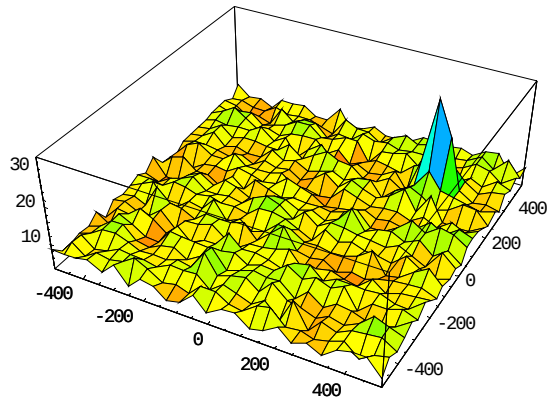
$P(2(w_0+\Delta)) \sim \exp[i\{\varphi(\Delta+\Omega)+\varphi(\Delta-\Omega)\}]$

Therefore $\varphi = \pi$ or $\varphi = 0$.
Binary phase shaping is ideal!



Visualizing the Search Space
Convex or needle in the haystack?



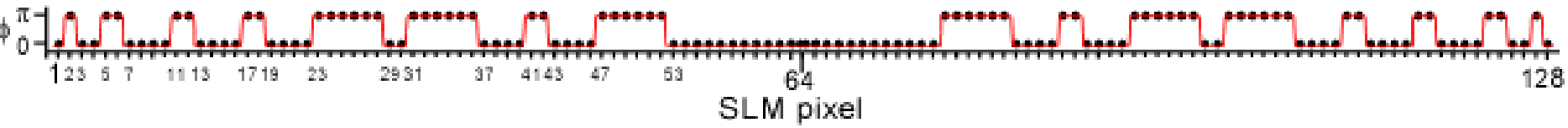


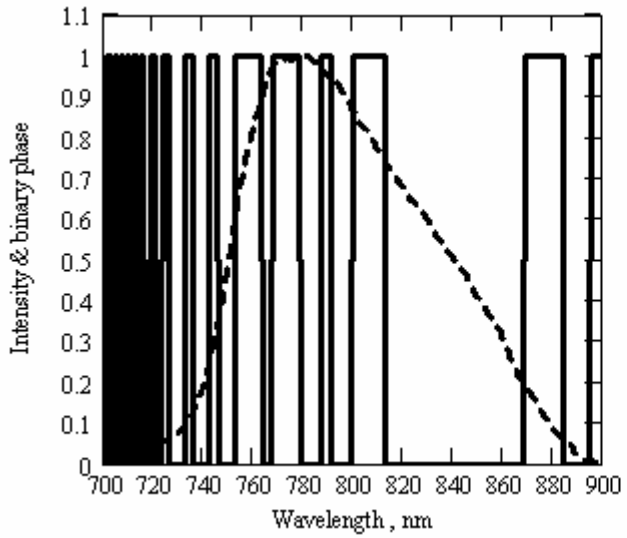
Possible only with BPS

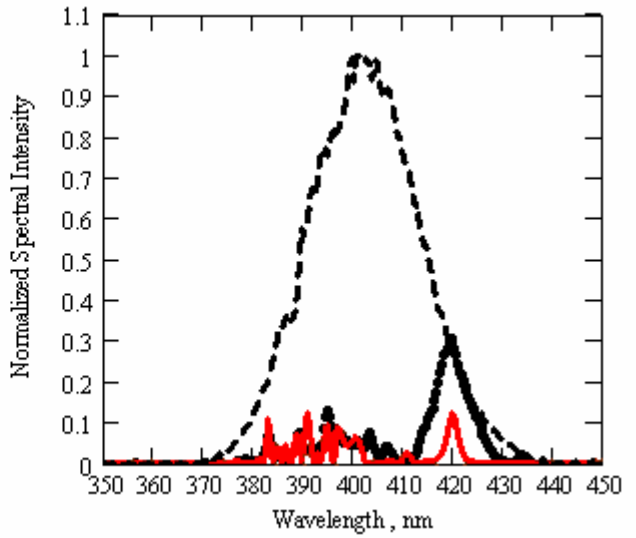
V. V. Lozovoy, M. Dantus, "*Systematic control of nonlinear optical processes using optimally shaped femtosecond pulses. Multiphoton Intrapulse Interference 7*" **Chem. Phys. Chem.,** (invited review submitted ***2004***)

Binary phase shaping (BPS)

Prime number inspired BPS mask 6x greater contrast than sinusoidal mask. BPS mask is further improved (2.5x) using an evolutionary learning algorithm.







Binary phase mask and spectrum of the fundamental laser pulse ~ 11 fs (dashed).

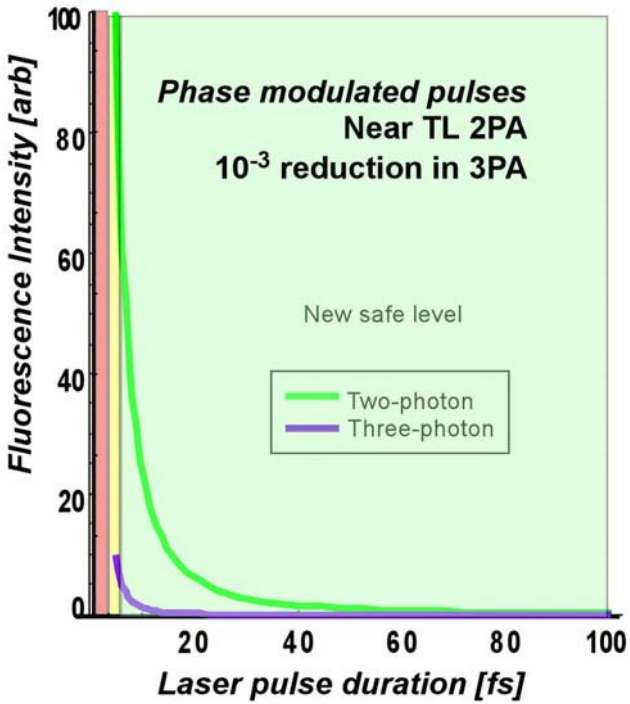
SHG output for TL pulses (dashed) and for shaped pulses (dots). The red trace is a simulation of the data.

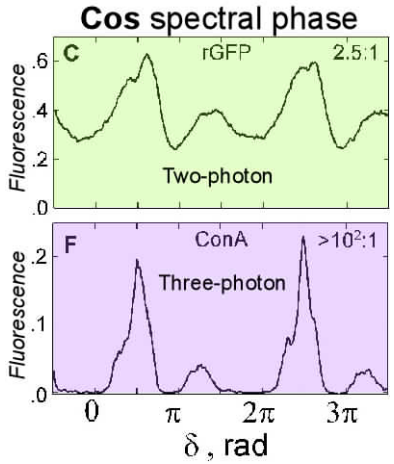
M. Comstock, V. V. Lozovoy, I. Pastirk, M. Dantus
"Multiphoton Intrapulse Interference 6;Binary Phase Shaping"
Optics Express *2004*, **12**(6), **1061**-1066.

What about sample damage caused by the pump laser? Optimized sample pumping, reducing three-photon damage

Manipulate the multiphoton intrapulse interference

Constructive for 2PA, Destructive for 3PA

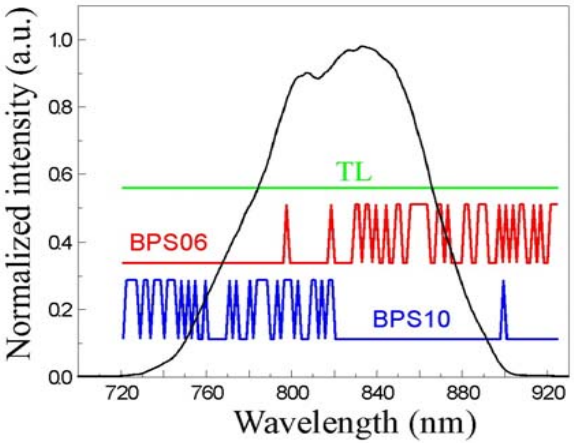


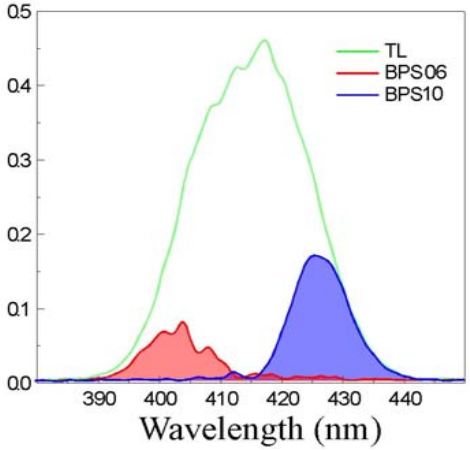


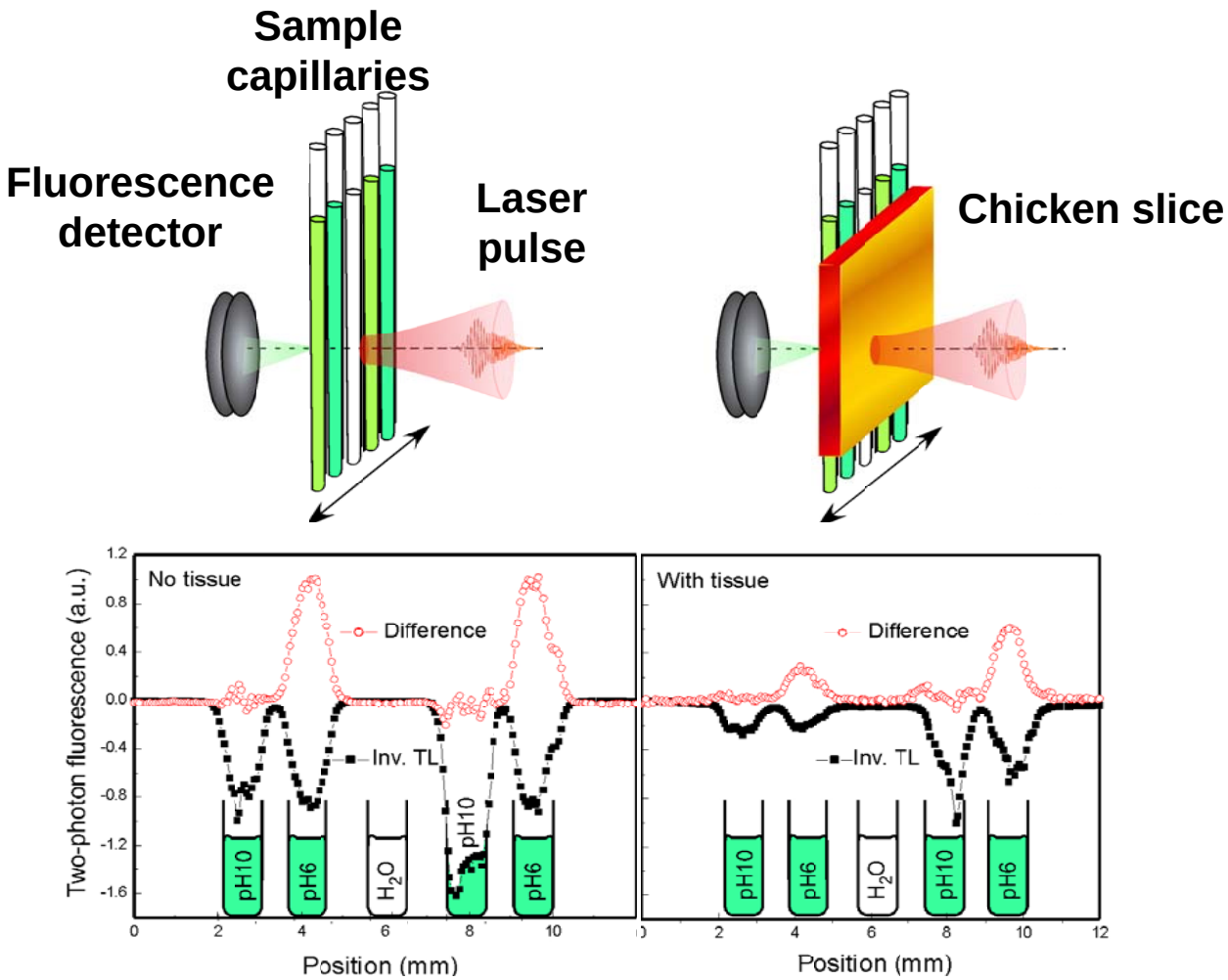
V.V. Lozovoy, I. Pastirk, K.A. Walowicz, and M. Dantus *J. Chem. Phys*. 118, 3187 (2003)

Selective excitation through scattering tissue

Two-photon excitation through scattering tissue with binary phase shaped pulses





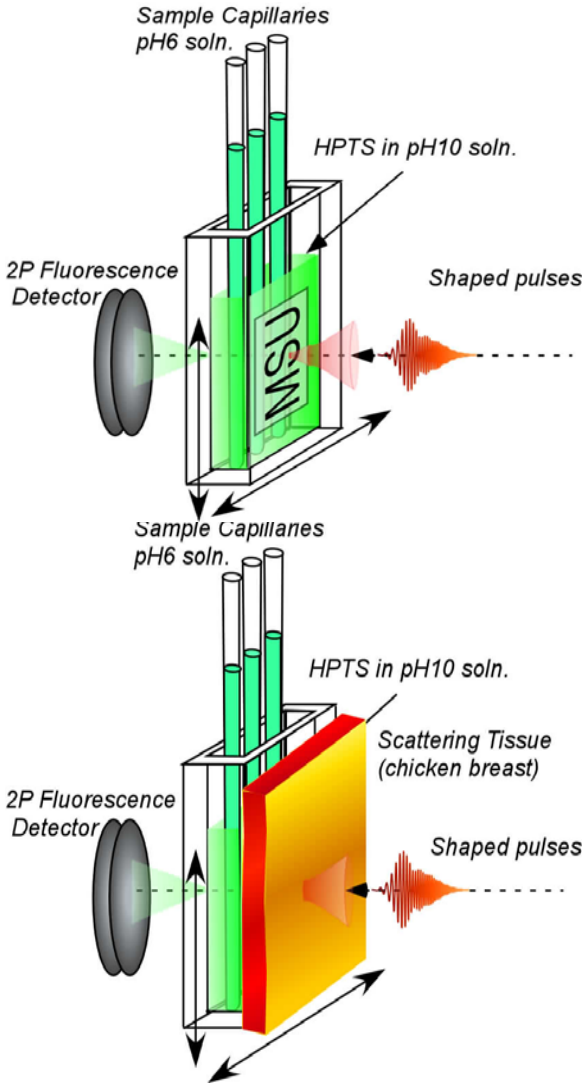


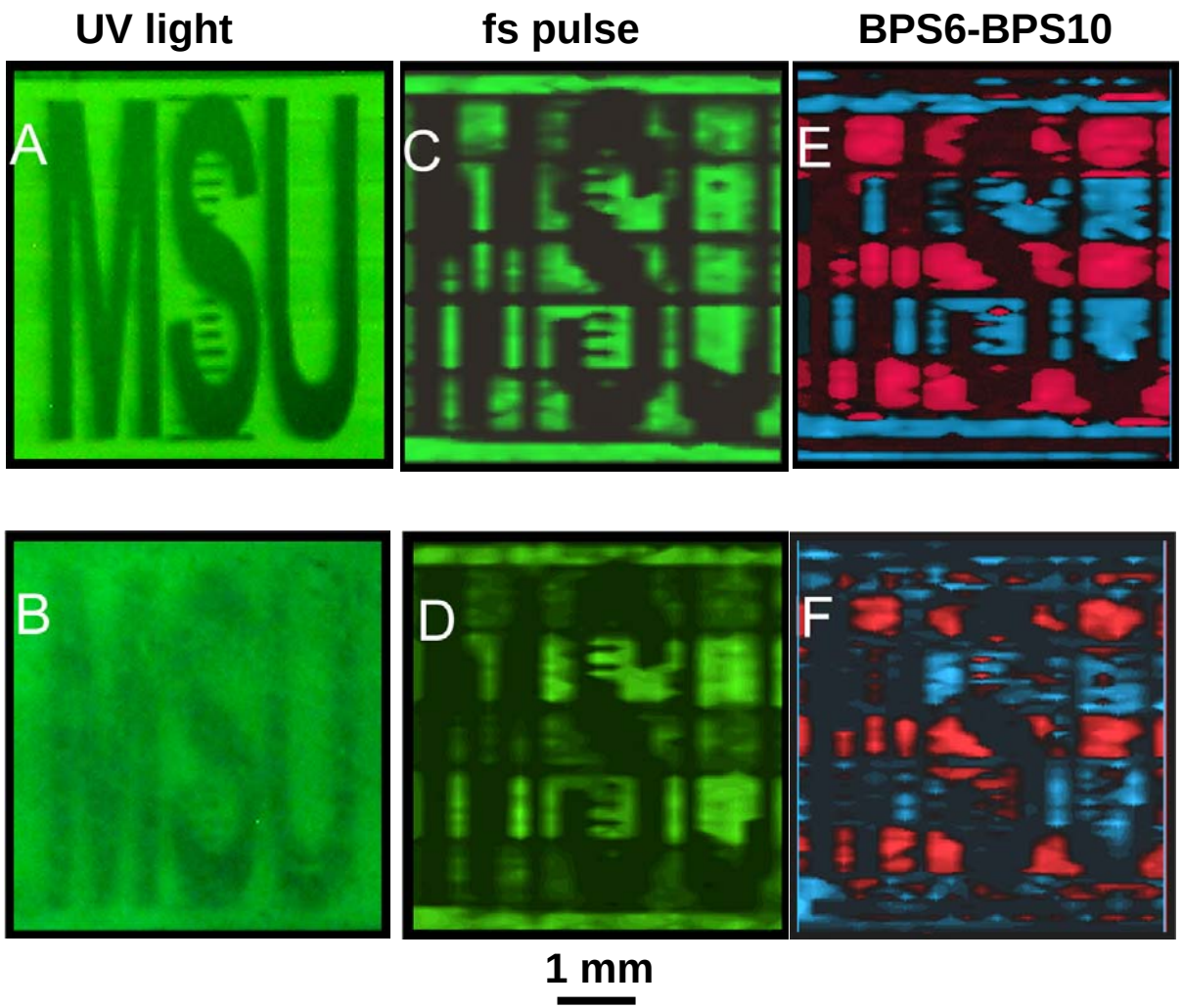
J. M. Dela Cruz, I. Pastirk, M. Comstock, M. Dantus

"Coherent control through scattering tissue, Multiphoton Intrapulse Interference 8,

Optics Express *2004*, submitted

Imaging through scattering tissue





J.M. Dela Cruz, I. Pastirk, M. Comstock, V. V. Lozovoy, M. Dantus

"Can coherent control methods be used through scattering tissue to achieve functional imaging?"

Proceedings of the National Academy of Sciences USA, submitted ***2004***

Conclusions

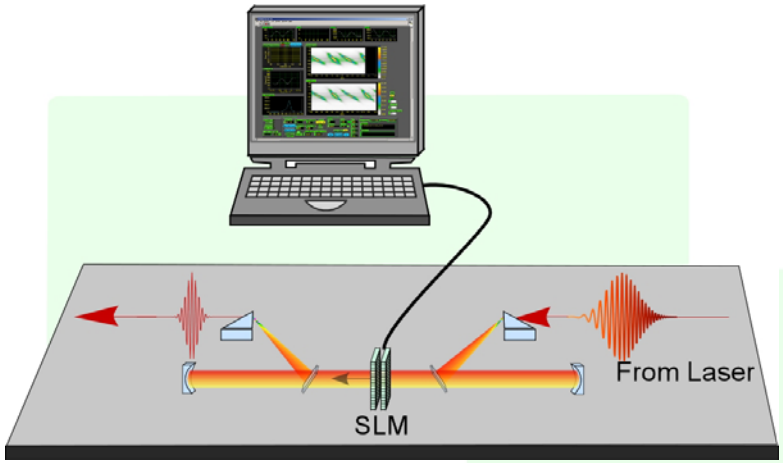
- Optimization of femtosecond laser pulses for x-ray pulse generation
- Optimized femtosecond laser excitation for pump/probe experiments

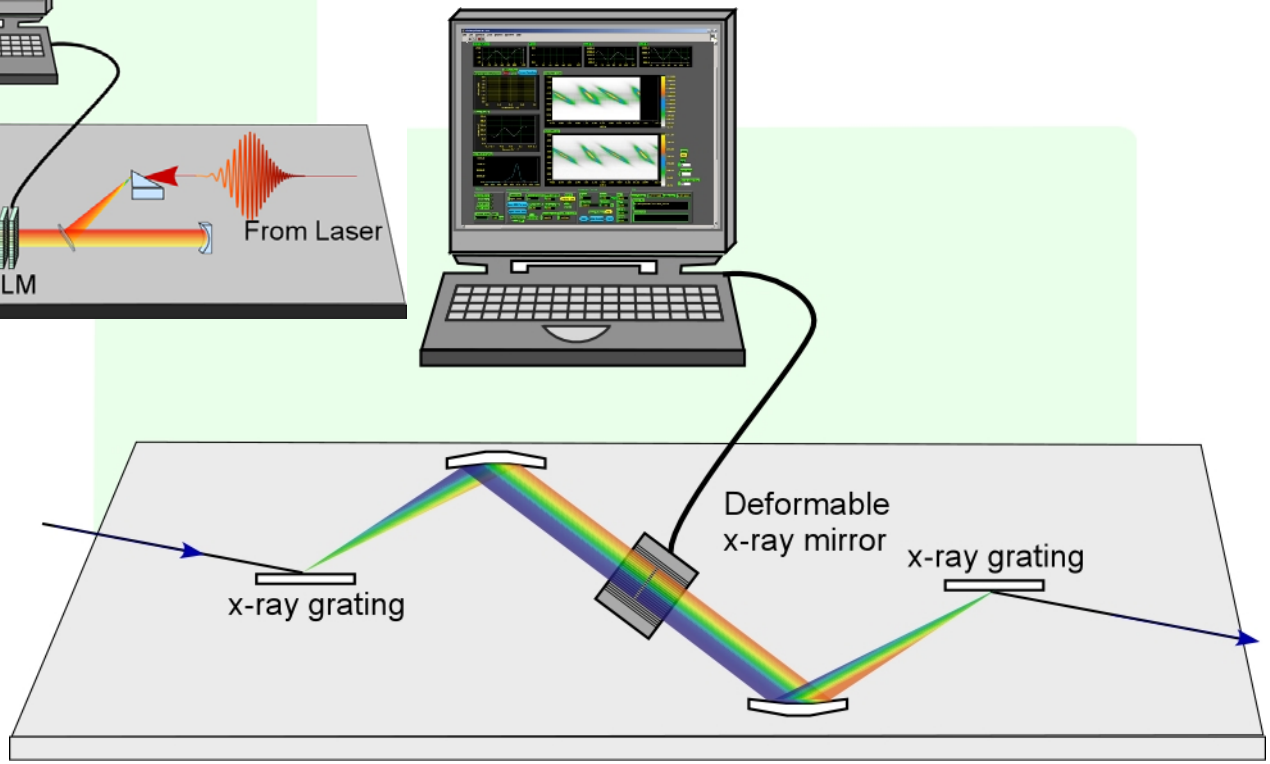
- Principles of coherent control and efficient GA optimization

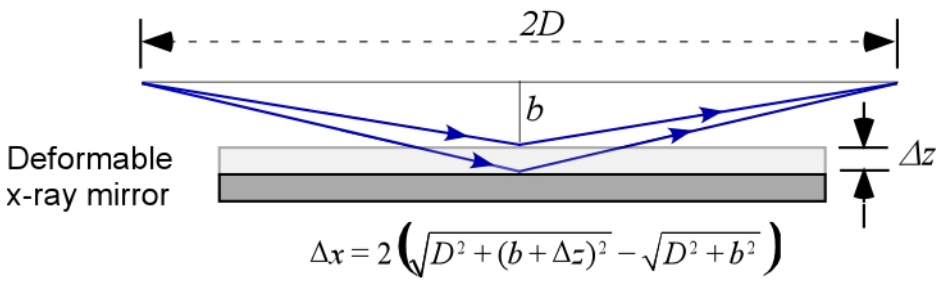
- Phase characterization and compensation for x-ray pulses?

- Use MIIPS!

Shaping x-ray pulses



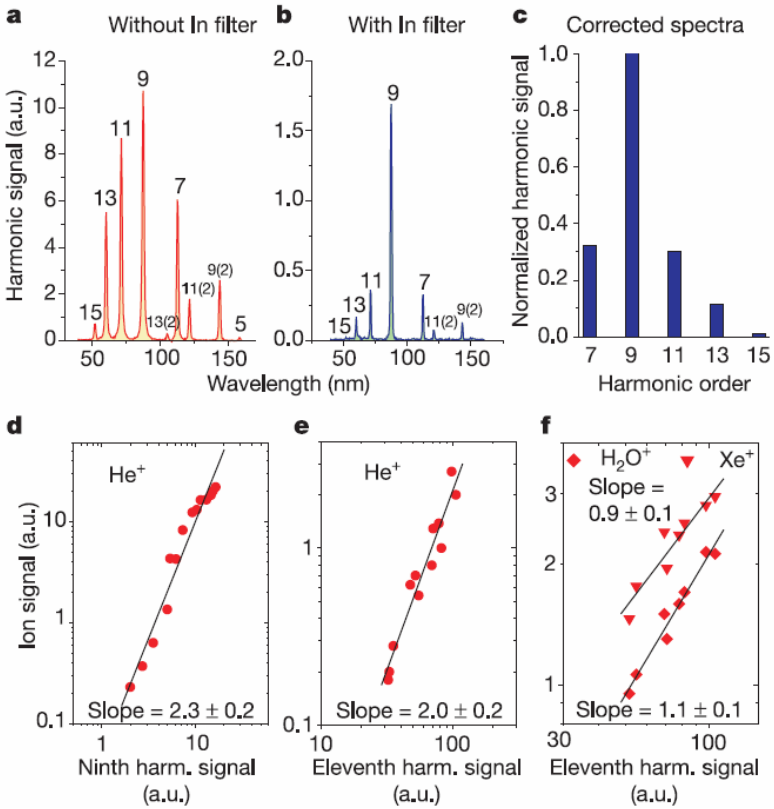


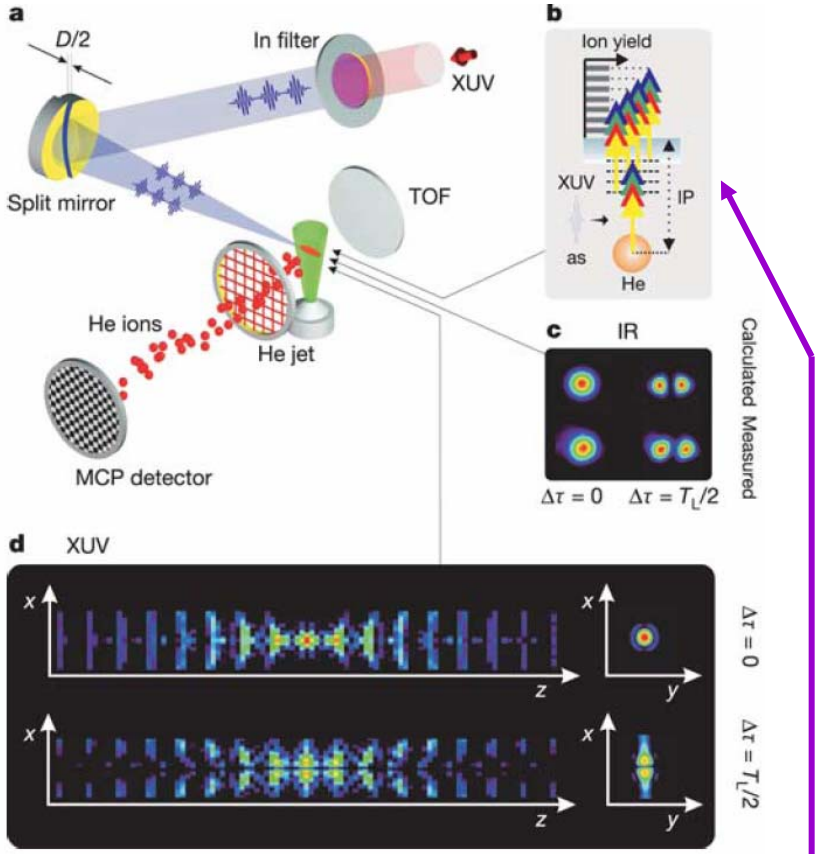


$$\Delta x = 2\left(\sqrt{D^2+(b+\Delta z)^2}-\sqrt{D^2+b^2}\right)$$

Order of Magnitude
λxray=λoptical/100
For D =1m and b=1cm
Δx=10nm when Δz=0.5μm

Nonlinear optics with x-ray pulses





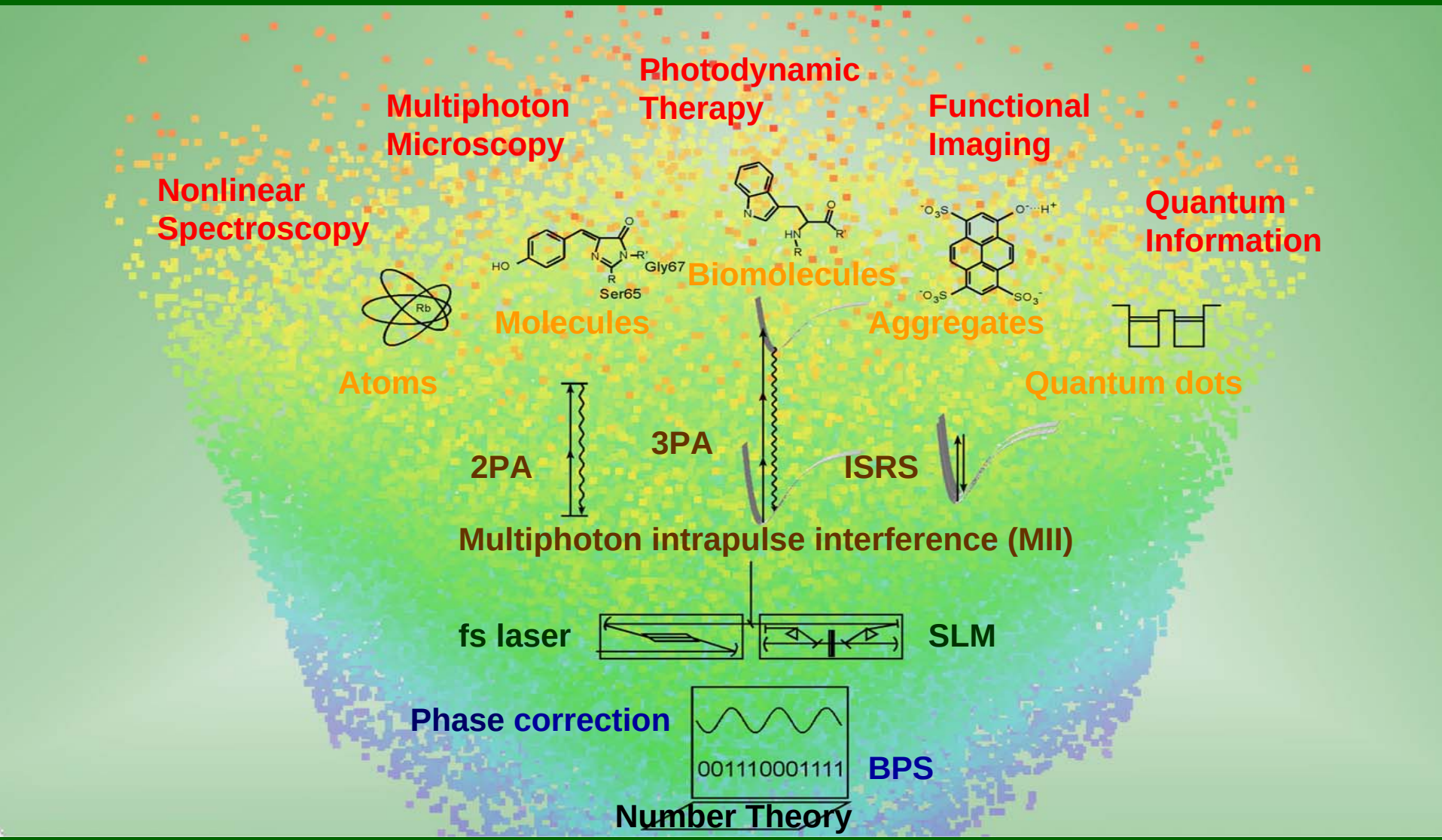
Two-photon XUV ionization of He, IP. 24.6 eV
One-photon XUV ionization of H2O and Xe, IP 12.6 and 12.1 eV

Volume autocorrelator

By recording the photoelectron energy one could carry out FROG
Without the split mirror one could carry out MIIPS

P. Tzallas, D. Charalambidis, N. A. Papadogiannis, K. Witte, G. D. Tsakiris," Autocorrelation of attosecond x-ray pulses,"
2003 *Nature* **426** 267

Future prospects



V. V. Lozovoy, M. Dantus,

"*Systematic control of nonlinear optical processes using optimally shaped femtosecond pulses. Multiphoton Intrapulse Interference 7*"

ChemPhysChem, (invited review submitted ***2004***)

Coherent Laser Control of Physicochemical Processes

Dr. Igor Pastirk

Johanna Dela Cruz

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Prof. Vadim Lozovoy

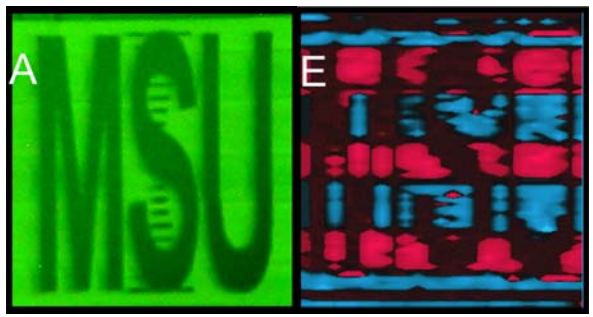
Prof. Marcos Dantus

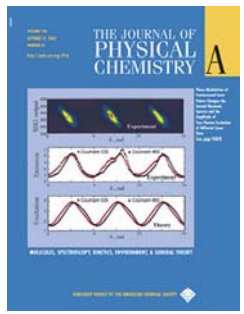
Collaborators, Students and post-docs are welcome to join the team

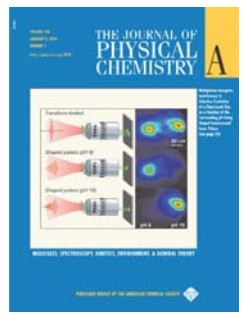
Tech transfer through Biophotonic Solutions Inc.

The Dantus Research Group

dantus@msu.edu **visit** ***www.cem.msu.edu/~dantus***







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