Narrow Transitions, Broad Light:

Quantum Control of Simple Systems with Shaped Femtosecond Pulses

Yaron Silberberg
www.weizmann.ac.il/~feyaron

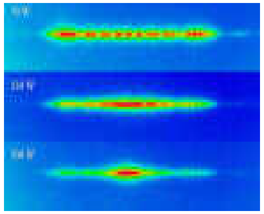
ULTRA FAST OPTICS

Cargese August 2008

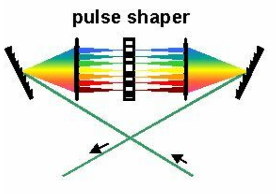
Ultrafast Optics Group

Yaron Silberberg
www.weizmann.ac.il/~feyaron

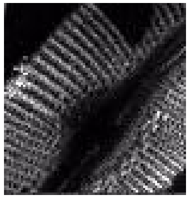
Physics of Complex Systems
Weizmann Institute of Science
Rehovot, Israel



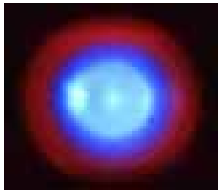
Optical
Solitons



Coherent
Quantum Control

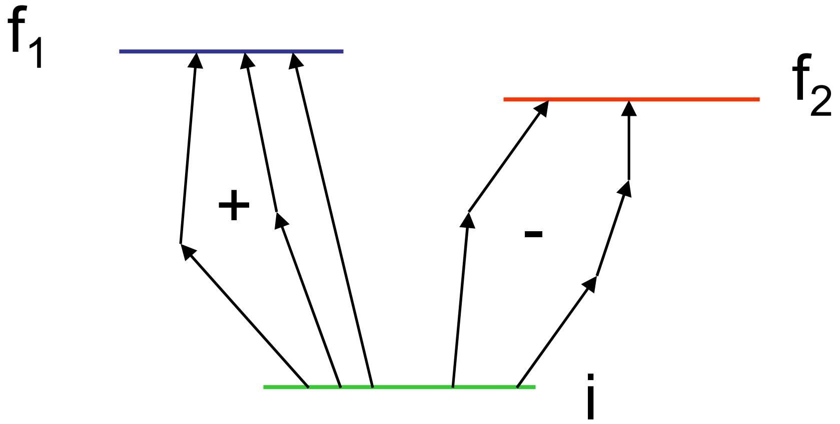


Nonlinear
Microscopy



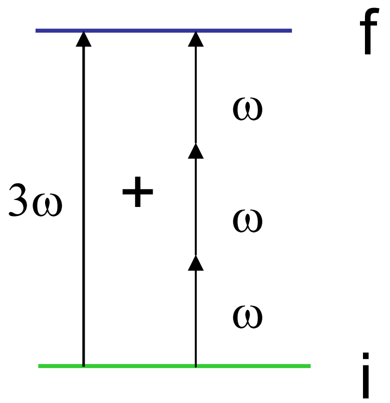
Quantum
Optics

Quantum Coherent Control



Use quantum interference to control the evolution of a system

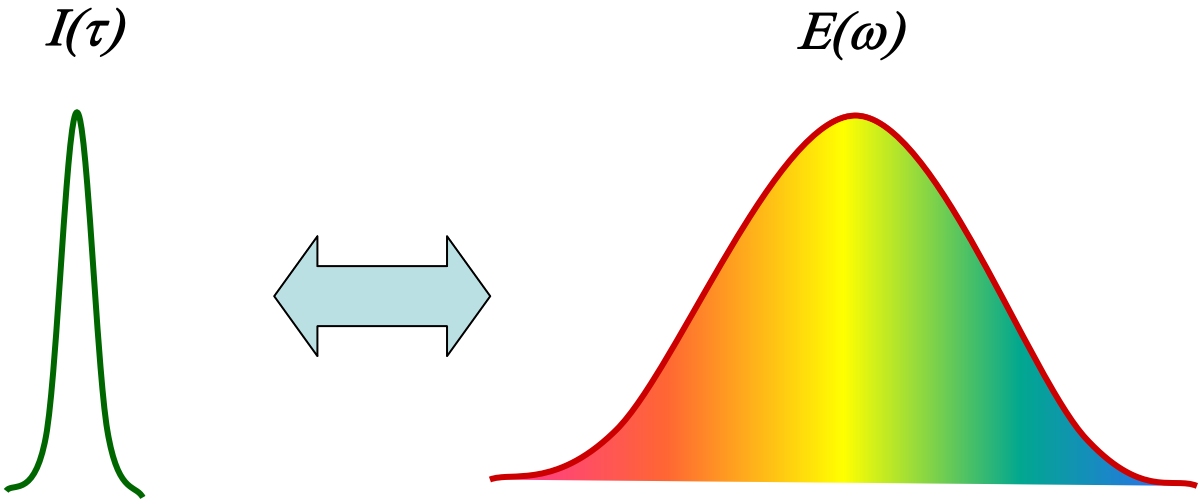
Quantum Coherent Control



…for example by interfering 1 and 3-photon transitions

Brumer & Shapiro
Tannor & Rice

Short Pulse = Broad Band

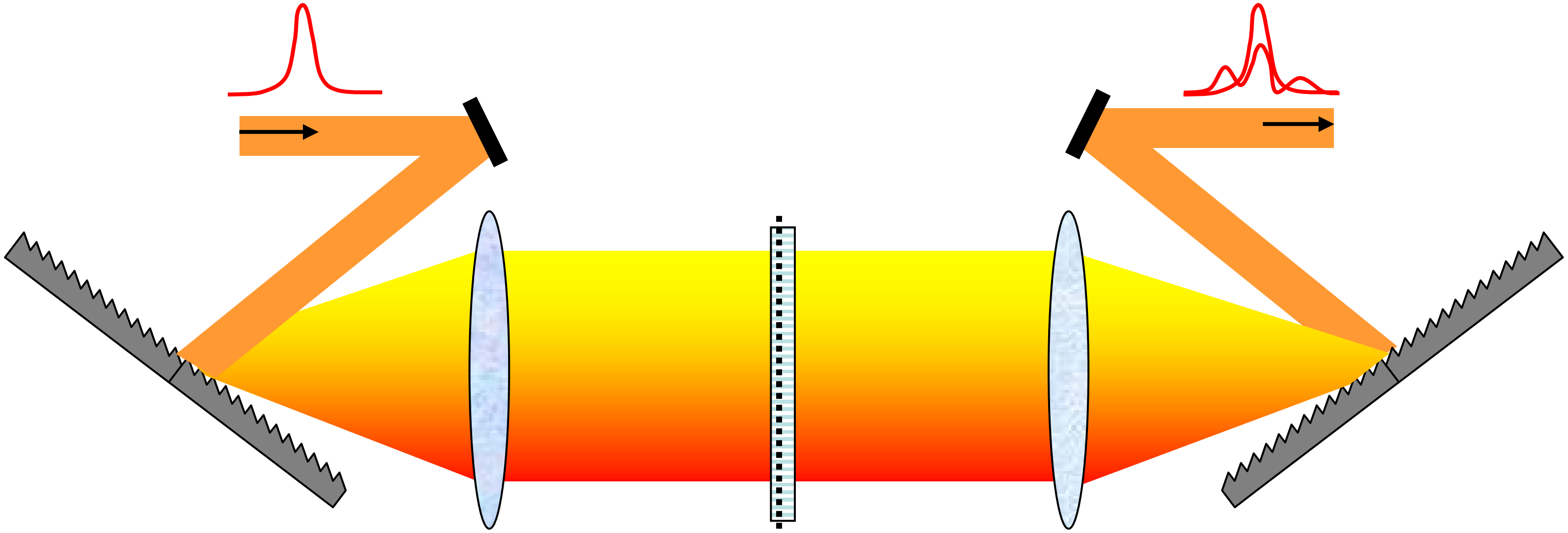


Broad, COHERENT Bandwidth

10 fs pulses @ 800 nm ~130 nm FWHM

Femtosecond Pulse Shaping

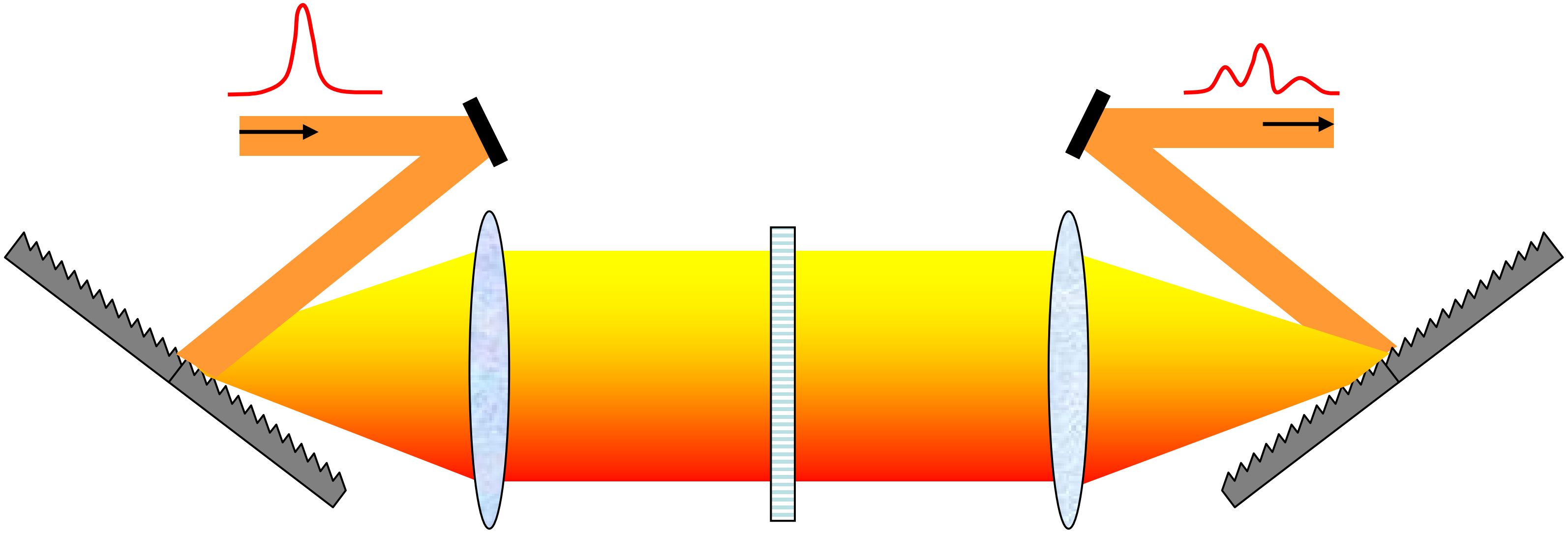
Phase, amplitude and polarization synthesizer



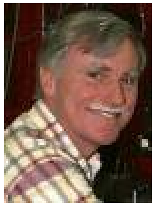
Heritage & Weiner Fourier Pulse-Shaping

Femtosecond Pulse Shaping

Phase, amplitude and polarization synthesizer



SLM

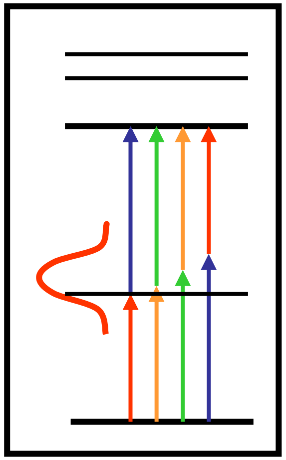


20 years anniversary
Heritage & Weiner Fourier Pulse-Shaping

Narrow Transitions, Broad Light

Atomic transitions ~ 1 GHz

10 fs pulse ~ 100,000 GHz



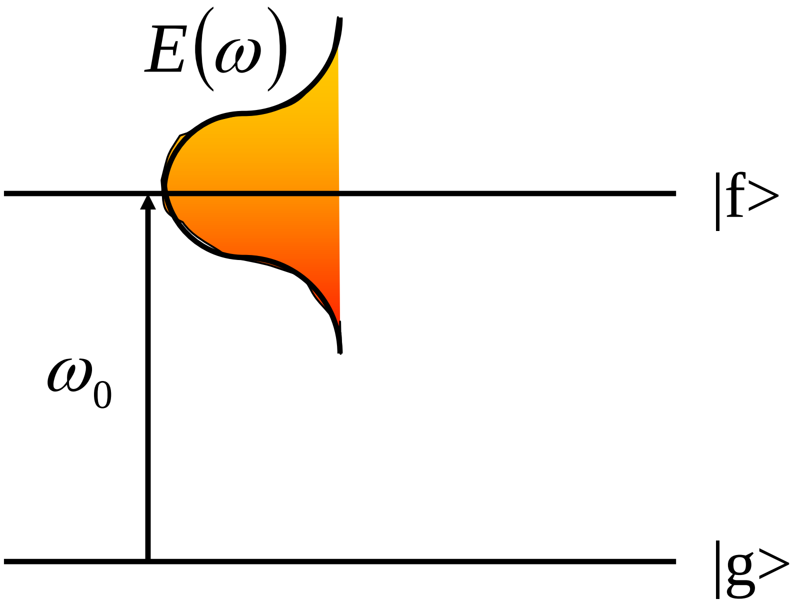
Crisp, Grichkowsky, Noordam, Girard, Chatel,…

Control of Multiphoton Transition

1. Two-Photon Absorption
2. Coherent Transients
3. Strong Fields
4. CARS spectroscopy

Is NLO always best with the shortest, most intense pulses?

Interaction of a broad band pulses and a narrow resonant transition



Resonant excitation of a two-level system

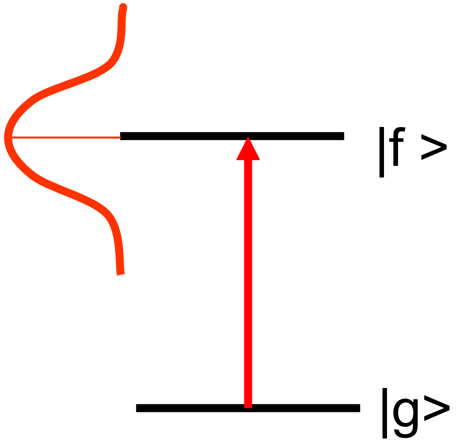
1st Order Time-Dependent Perturbation Analysis

$$a_f(t) = \frac{\mu_{fg}}{i\hbar} \int_{-\infty}^{t} dt_1\, \varepsilon(t_1)\, e^{i\omega_{fg} t_1}$$

$$\boxed{a_f(\infty) = \frac{\mu_{fg}}{i\hbar} E(\omega_{fg})}$$

Pulse Area

$$p_f(\infty) \propto \left| E(\omega_{fg}) \right|^2$$

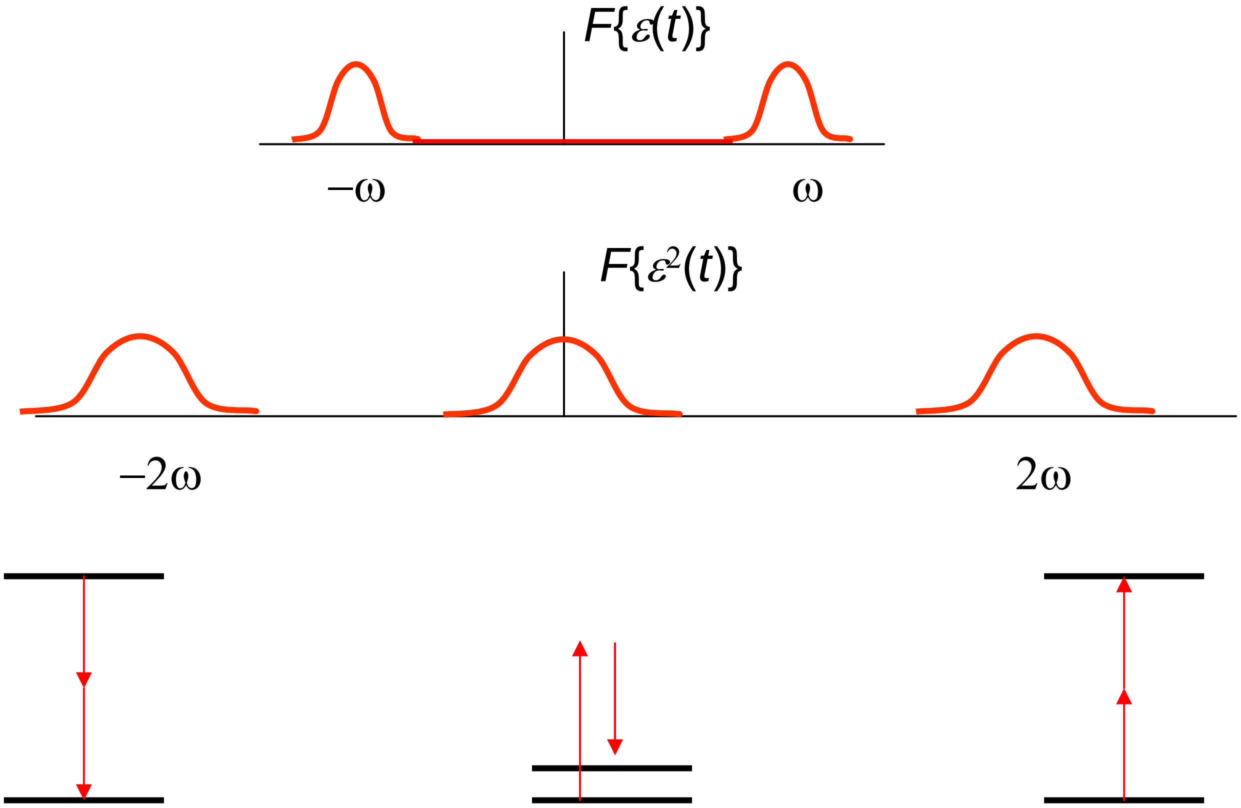


Transitions only by resonant light
Pulse shaping not effective (“Emperor’s new clothes?”)

Two-photon processes

2^{nd} Order Time-Dependent Perturbation Analysis

$$a_f(\infty) \propto \int \varepsilon^2(t)\exp(i\omega_{fg}t)dt$$

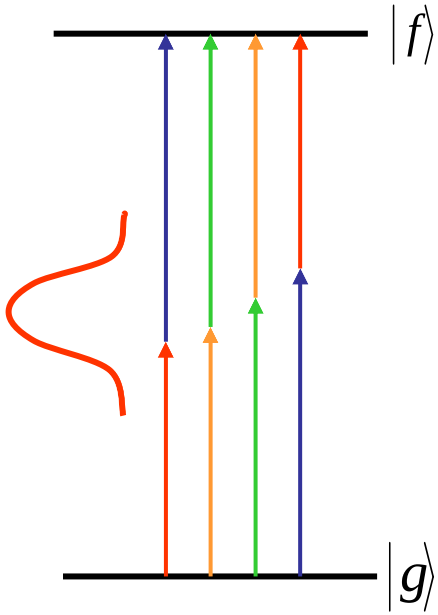


Nonresonant two-photon absorption

2^{nd} Order Time-Dependent Perturbation Analysis

$$a_f(\infty) \propto \int \varepsilon^2(t)\exp(i\omega_{fg}t)dt$$

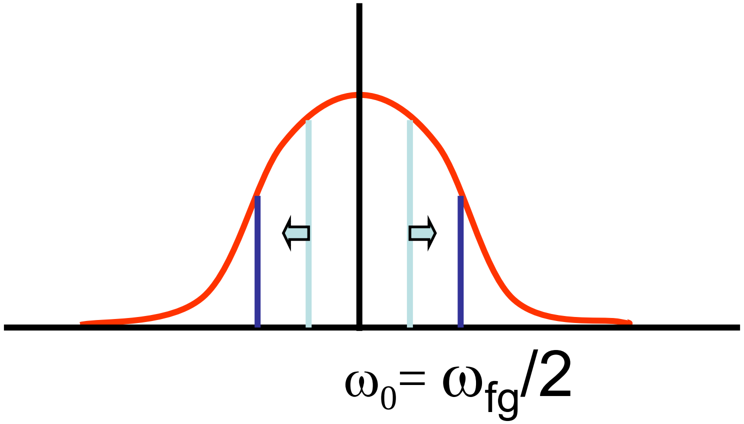
$$a_f(\infty) \propto \int E(\omega)E(\omega_{fg}-\omega)d\omega$$



Nonresonant TPA

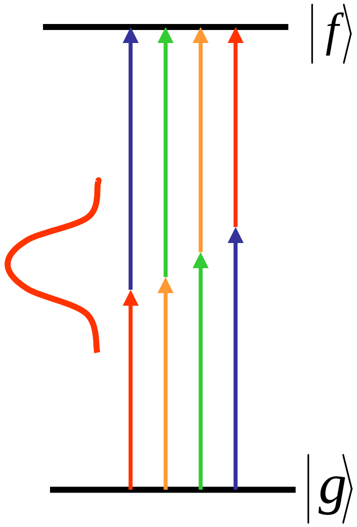
$$a_f(\infty) \propto \int d\delta\omega\, E(\omega_0+\delta\omega)E(\omega_0-\delta\omega) =$$

$$= \int d\delta\omega \left|E(\omega_0+\delta\omega)\right|\left|E(\omega_0-\delta\omega)\right| \cdot \underline{e^{i[\Phi(\omega_0+\delta\omega)+\Phi(\omega_0-\delta\omega)]}}$$



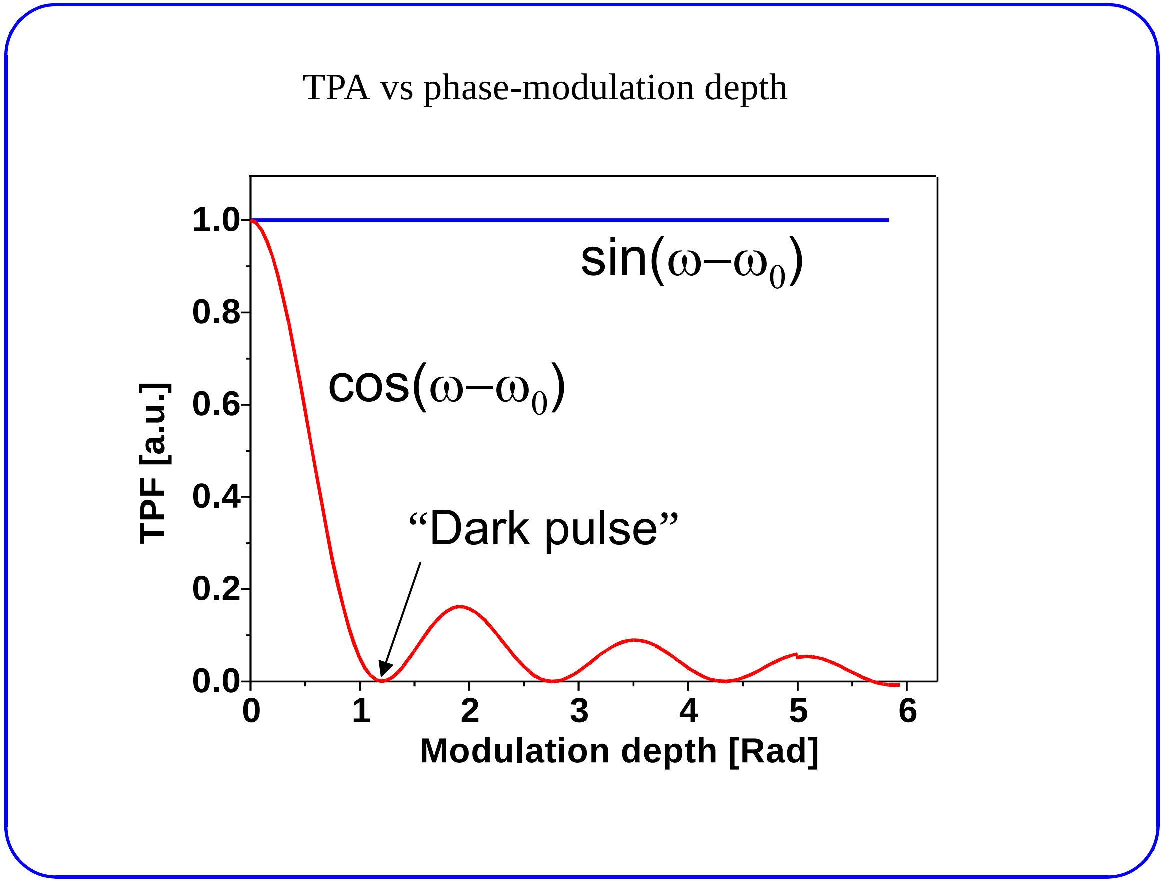
Transform limited pulses are most efficient, <u>but:</u>

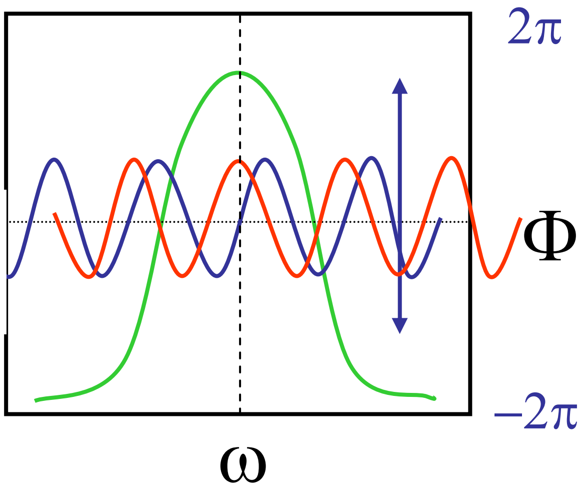
Antisymmetric phase has no effect on transition probability

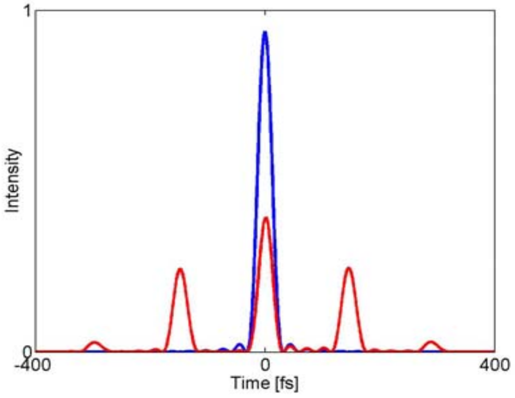


Nonresonant TPA

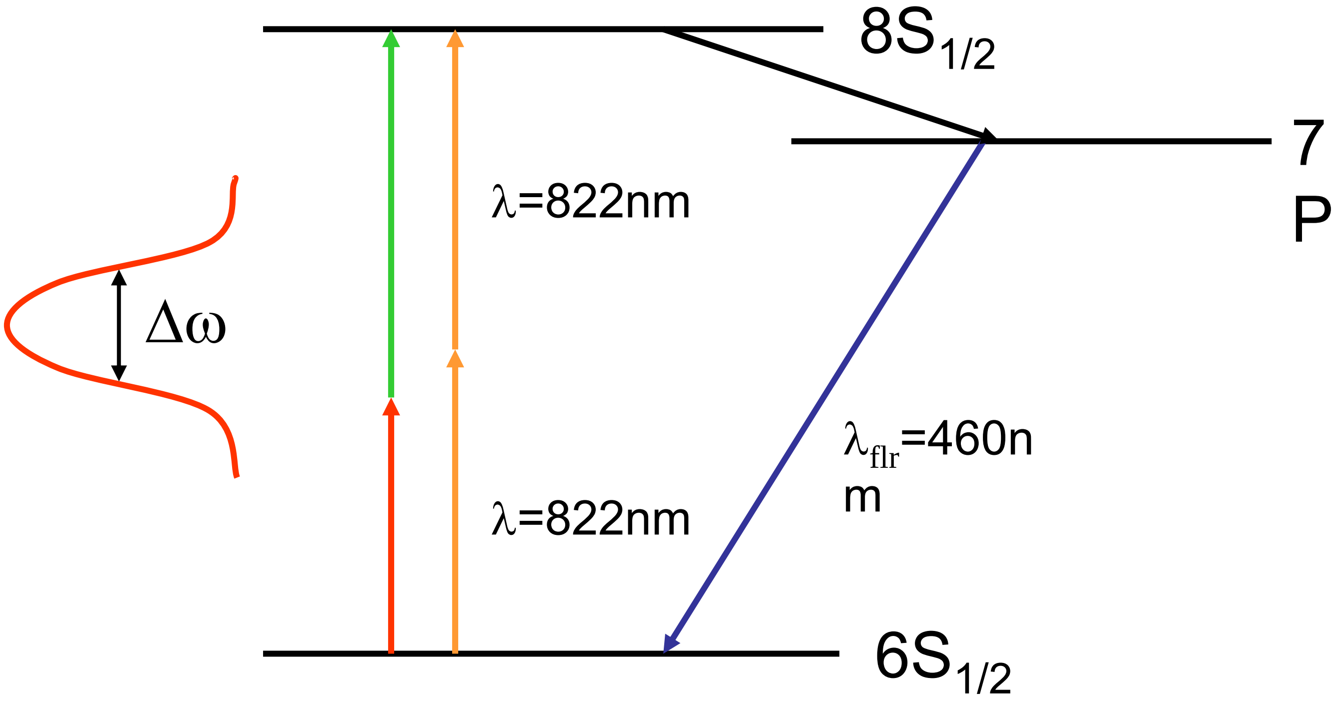
modulation with a periodic phase



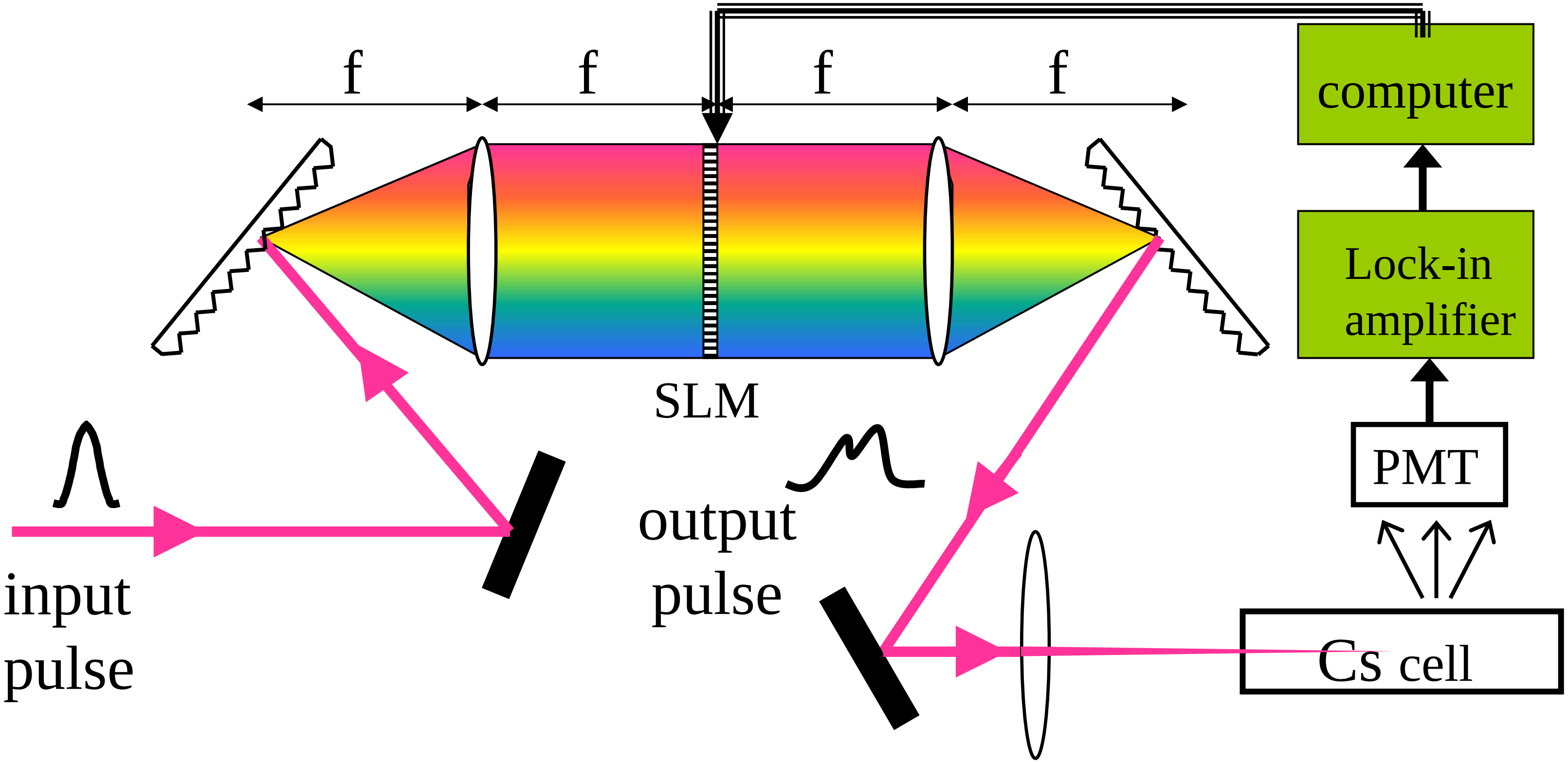




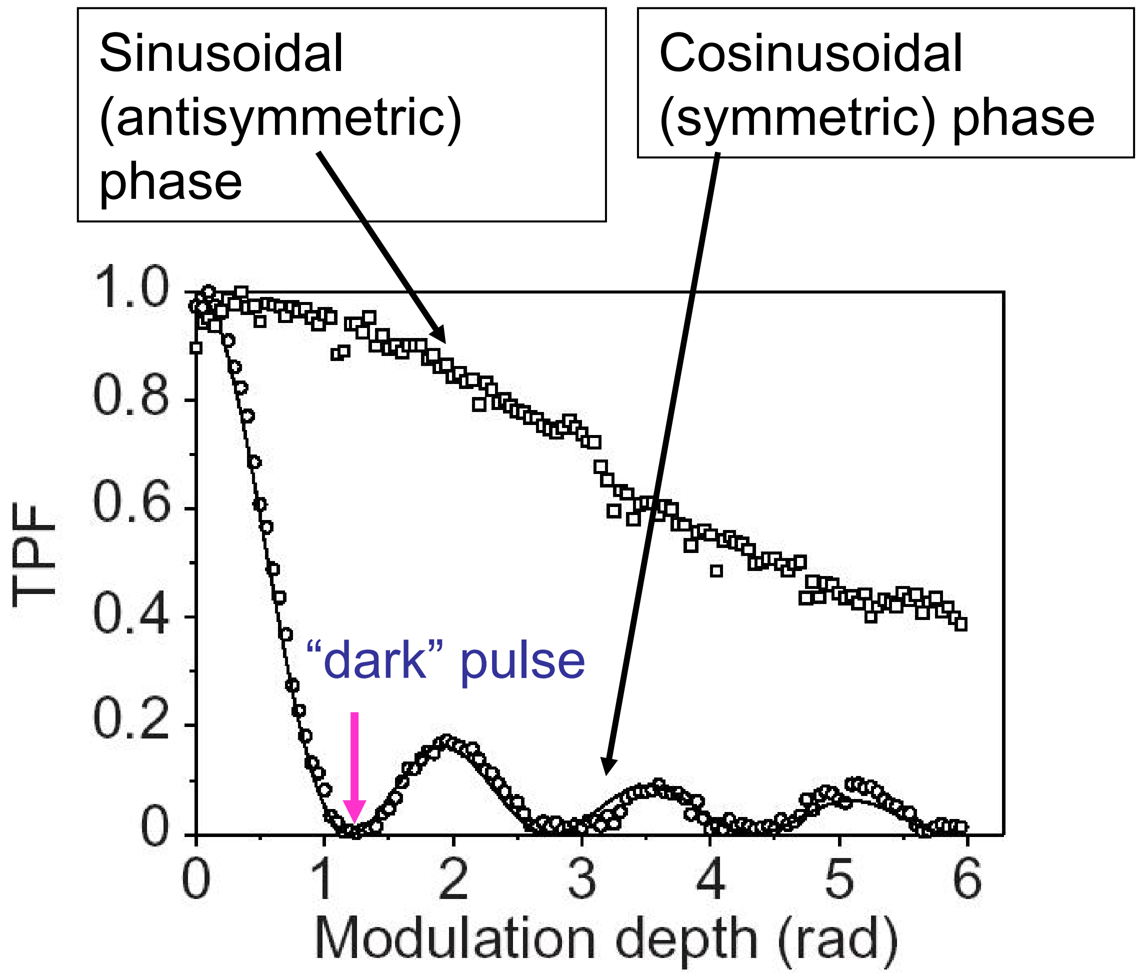
Energy level structure of Cesium



Nonresonant TPA-Experimental Set-Up

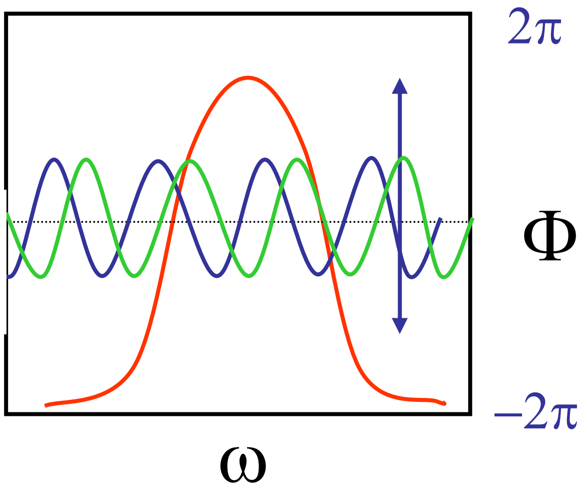


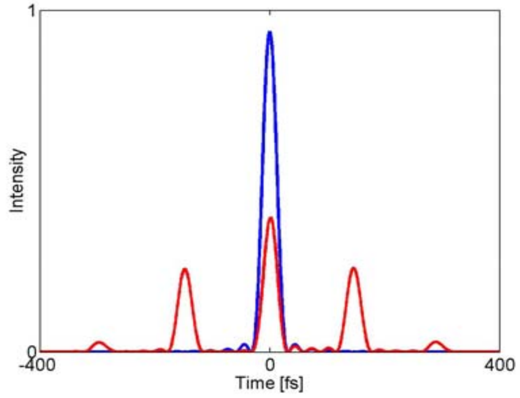
Nonresonant TPA:
modulation of a periodic phase mask



Sinusoidal (antisymmetric) phase

Cosinusoidal (symmetric) phase





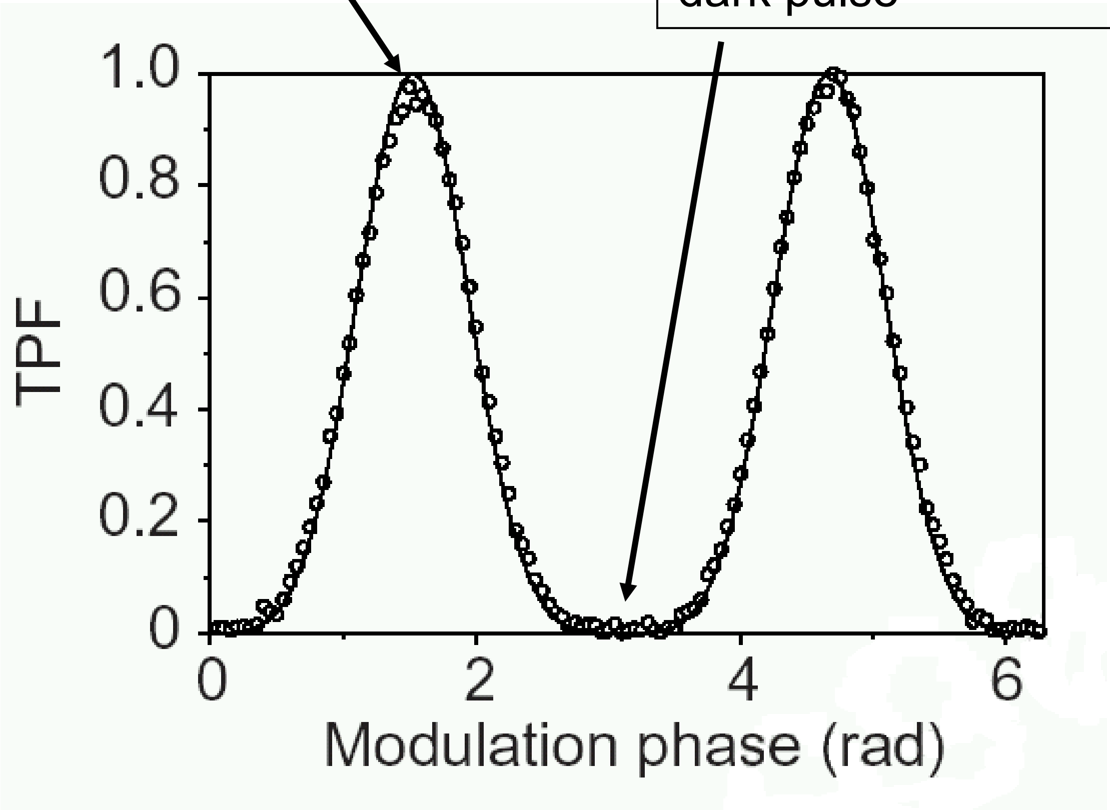
Meshulach & Silberberg, Nature, **396**, 239 (1998)

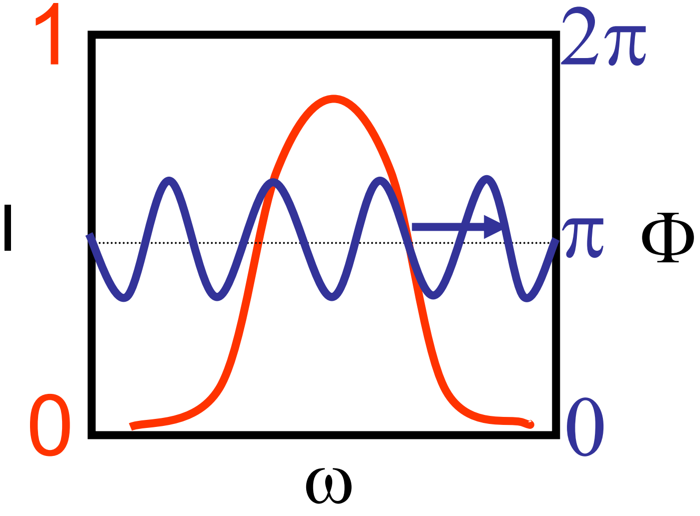
Nonresonant TPA

scan of a periodic phase mask

Sinusoidal (antisymmetric) phase

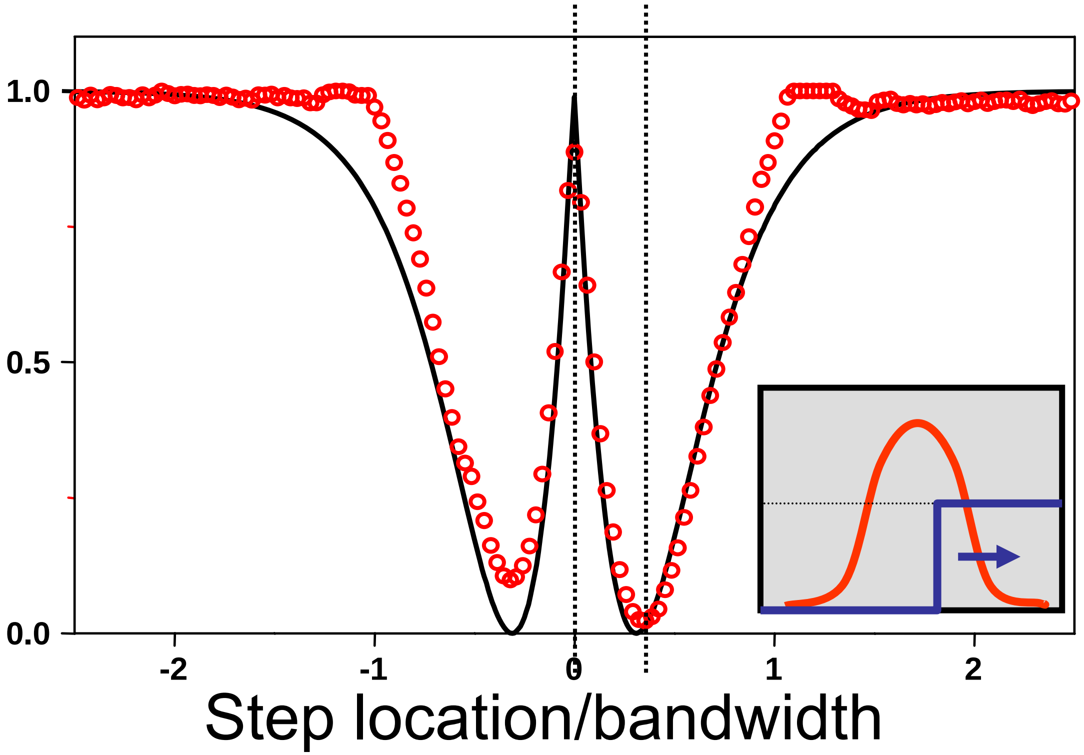
Cosinusoidal (symmetric) phase – dark pulse





Meshulach & Silberberg, Nature, **396**, 239 (1998)

Nonresonant TPA Control by Phase Step



Antisymmetric phase has no effect on transition probability

Certain spectral phase functions can annihilate the absorption rate

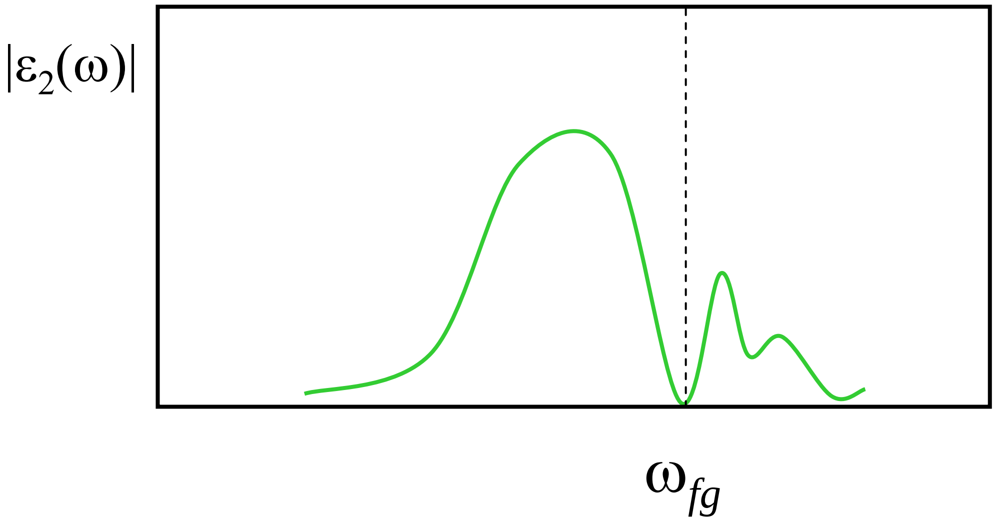
Meshulach & Silberberg, Nature, **396**, 239 (1998),Phys. Rev. A **60**, 1287 (1999)

Dark Pulses

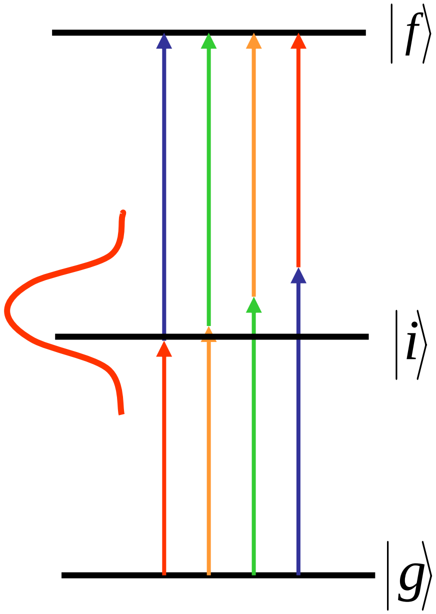
The spectrum of $\varepsilon^2(t)$ should have a zero at the resonant frequency

$$a_f(\infty) \propto \int \varepsilon^2(t)\exp(i\omega_{fg}t)dt = 0$$

$$\varepsilon_2(\omega) \equiv \int \varepsilon^2(t)\exp(i\omega\, t)dt$$



Resonant TPA



$$a_f(\infty) = \frac{-i}{(i\hbar)^2}\,\mu_{fi}\mu_{ig}\int d\omega\,\frac{E(\omega)E(\omega_{fg}-\omega)}{\omega_{ig}-\omega-i\Gamma_i}$$

Transition is dominated by
a single resonant level

For narrow levels ($\Gamma \ll \Delta\omega$):

$$a_f(\infty) \propto i\pi E(\omega_{ig})E(\omega_{fi}) + \wp\int d\omega\,\frac{E(\omega)E(\omega_{fg}-\omega)}{\omega_{ig}-\omega}$$

On-resonant term

Off-resonant term

Resonant Transitions

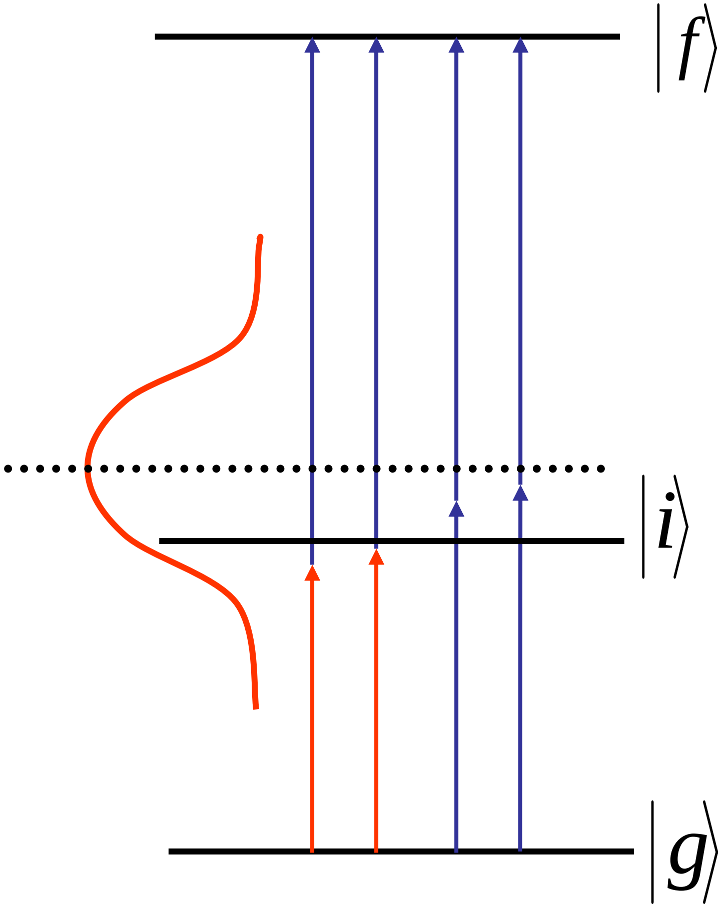
Looking at the off-resonant term:

$$\int d\omega \frac{E(\omega)E(\omega_{fg}-\omega)}{\omega_{ig}-\omega}$$

For transform limited pulses, frequencies around the resonance interfere destructively

Transform-limited pulses no longer maximize transition rates!

Pulses can be shaped to enhance TPA

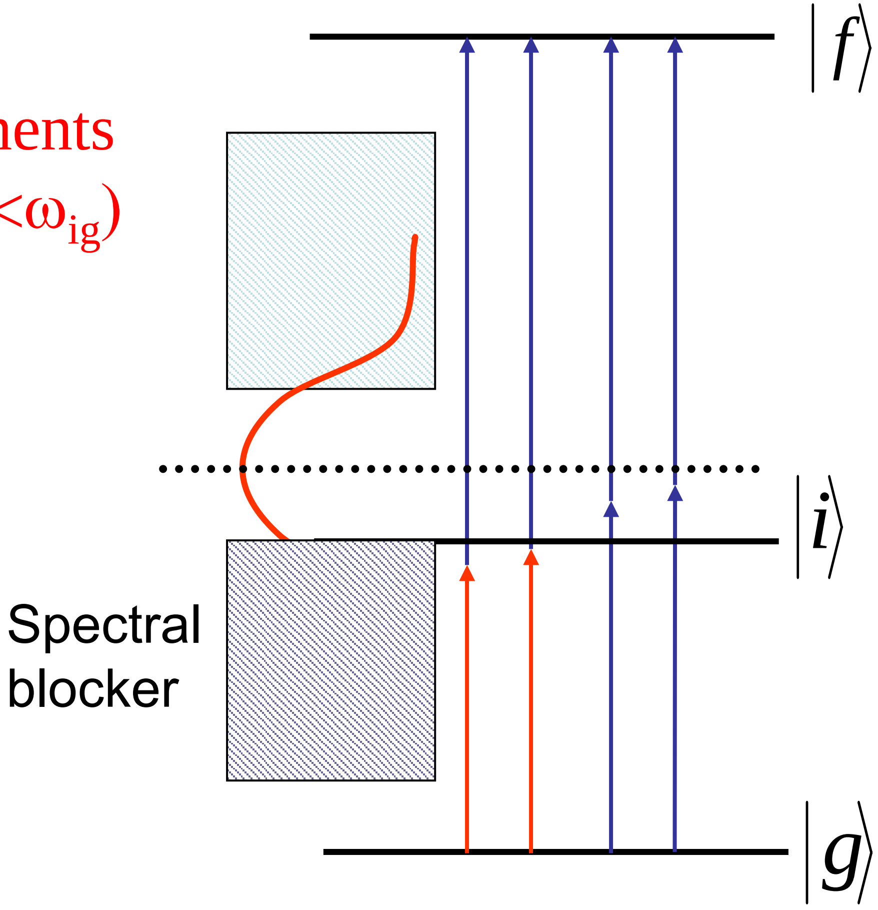


Enhancement of resonant TPA

Method 1 (amplitude shaping):

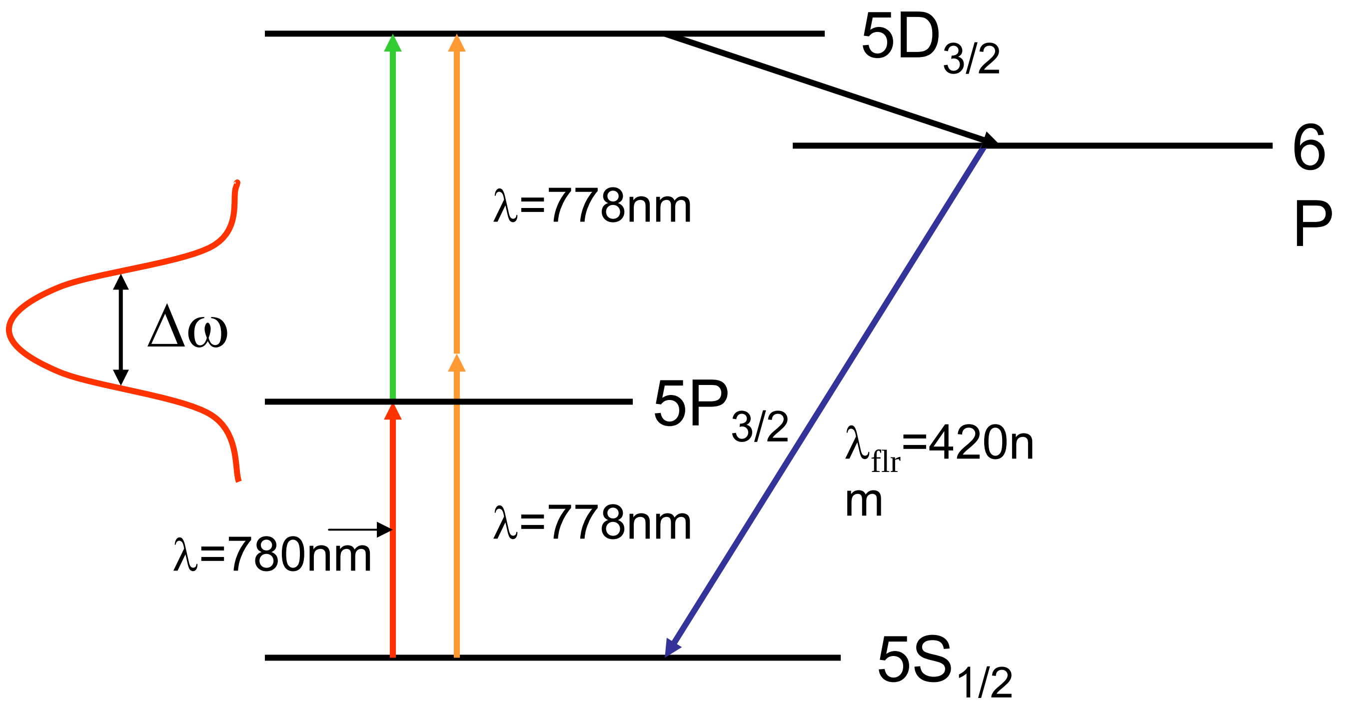
Eliminate all frequency components that contribute destructively ($\omega<\omega_{ig}$)

$$\int d\omega \frac{E(\omega)E(\omega_{fg}-\omega)}{\omega_{ig}-\omega}$$



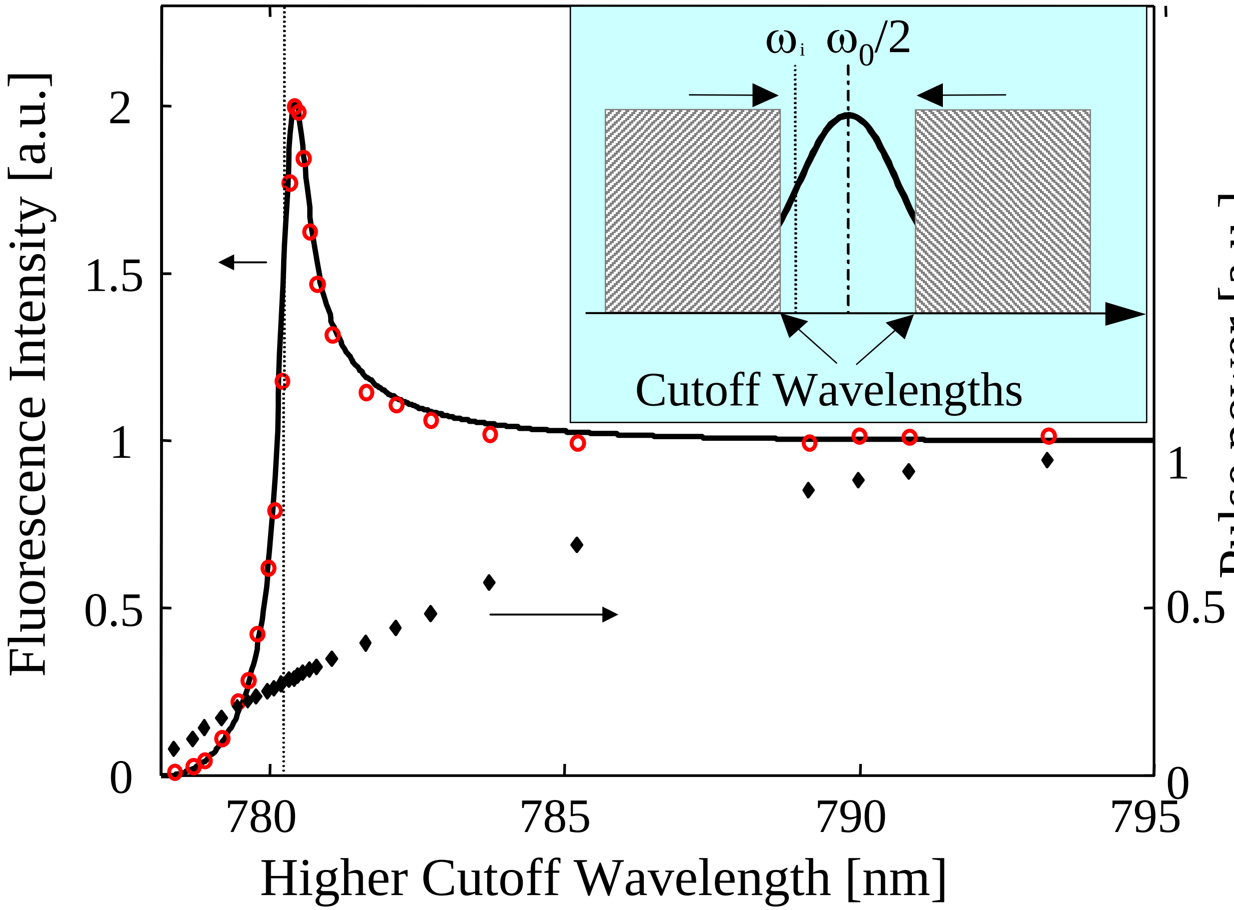
Dudovich *et al.*, Phys. Rev. Lett. **86**, 47 (2001)

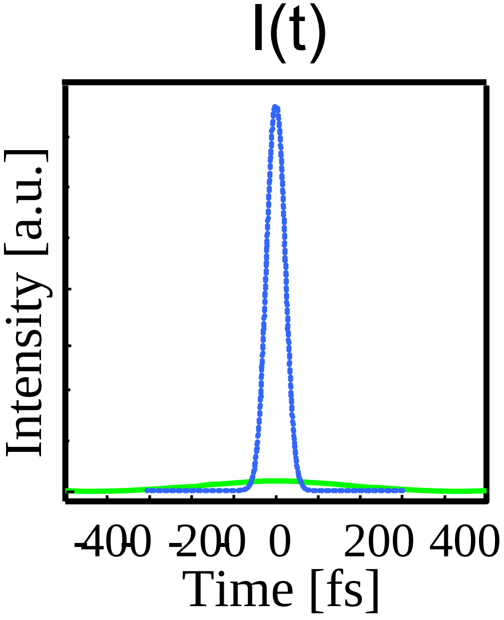
Energy level structure of Rubidium



Enhancement of resonant TPA

spectral blocking





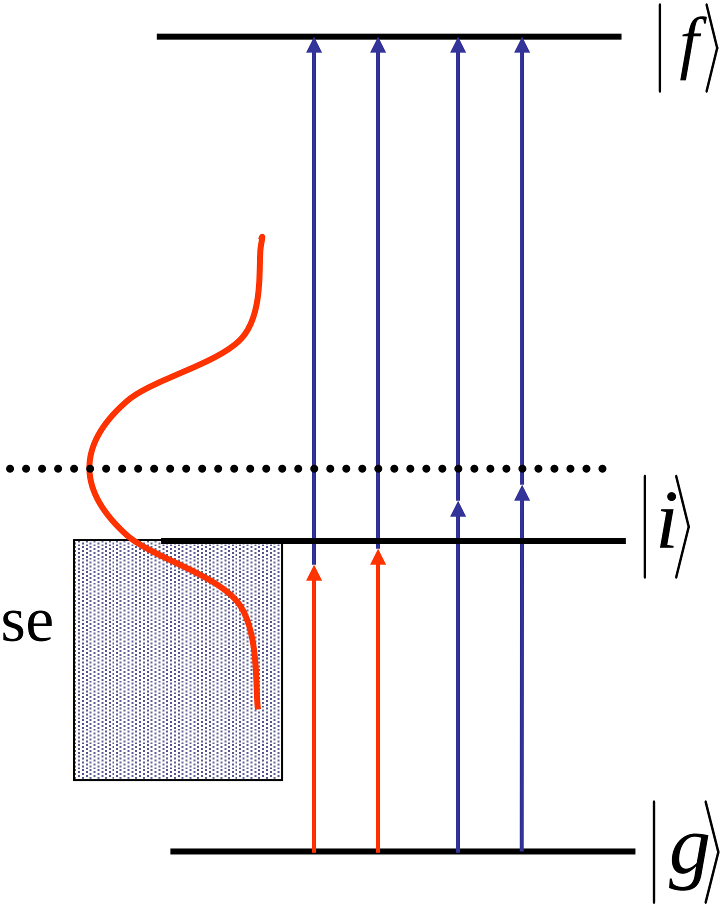
Dudovich *et al.*, Phys. Rev. Lett. **86**, 47 (2001)

Enhancement of resonant TPA

Method 2 (phase shaping):

Manipulate phases to induce constructive interference by all frequency pairs

$$\int d\omega \frac{E(\omega)E(\omega_{fg}-\omega)}{\omega_{ig}-\omega}$$

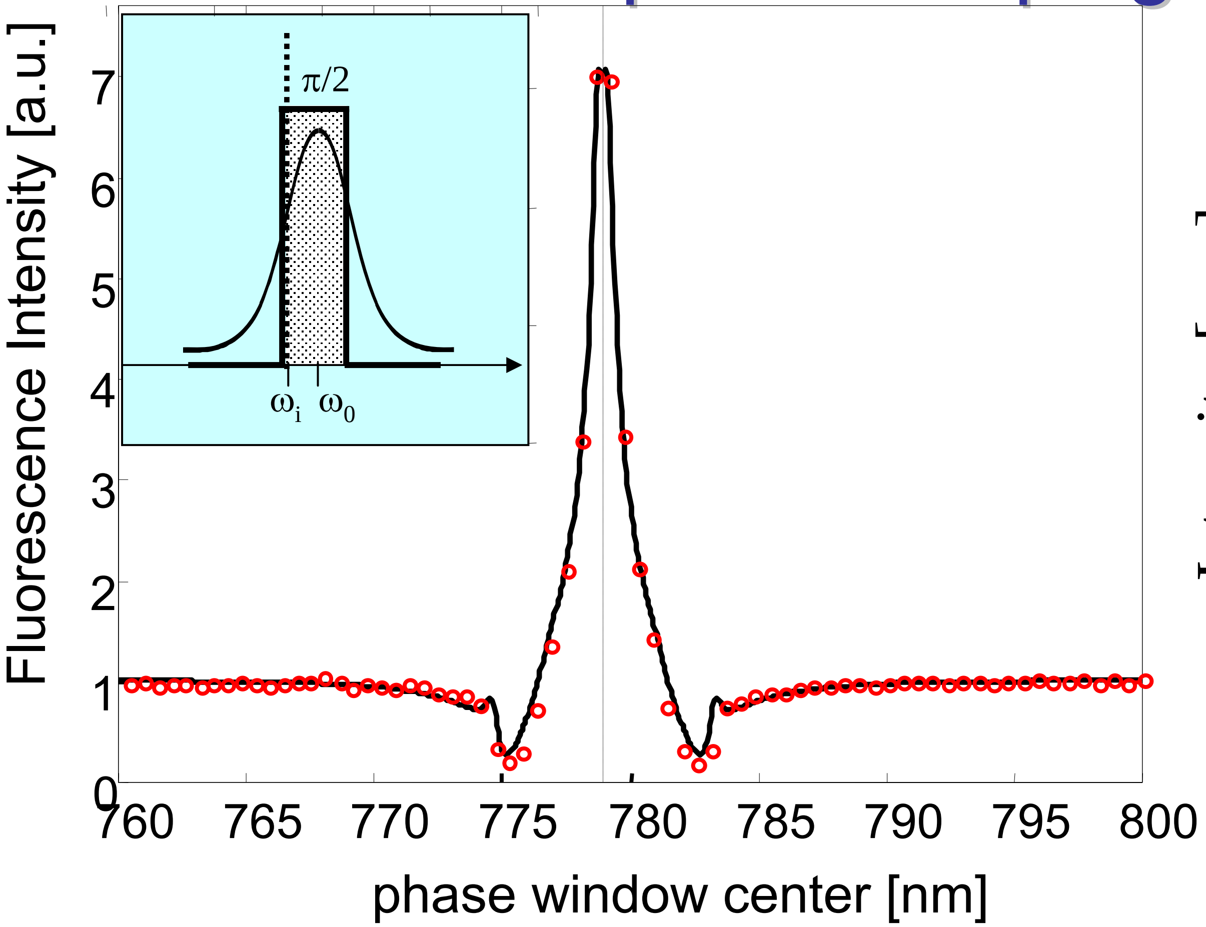


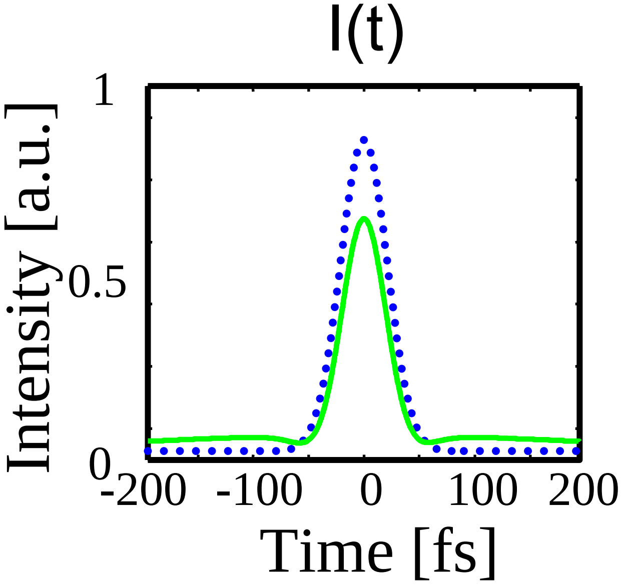
π Phase mask

Dudovich *et al*., Phys. Rev. Lett. **86**, 47 (2001)

Enhancement of resonant TPA

phase shaping



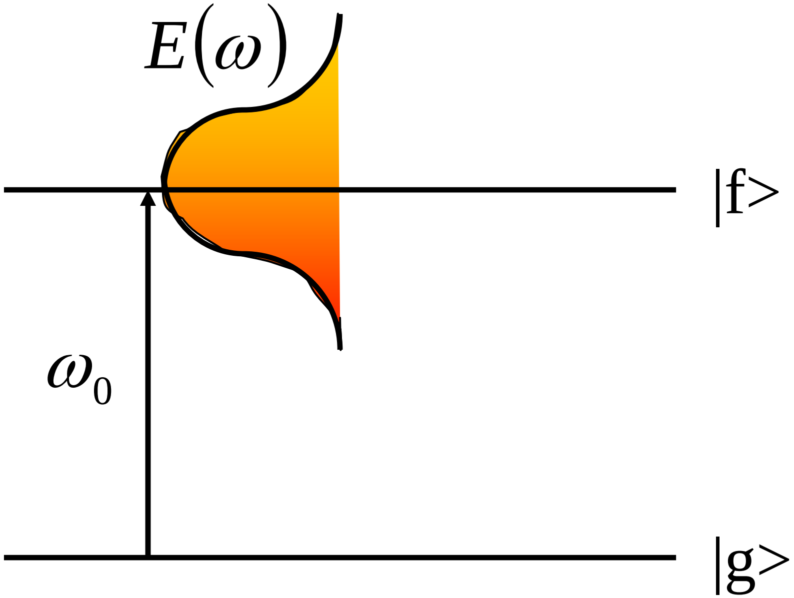


Dudovich *et al.*, Phys. Rev. Lett. **86**, 47 (2001)

Control of Multiphoton Transition

1. Two-Photon Absorption
2. Coherent Transients
3. Strong Fields
4. CARS spectroscopy

Interaction of a broad band pulses and a narrow resonant transition



Resonant excitation of a two-level system

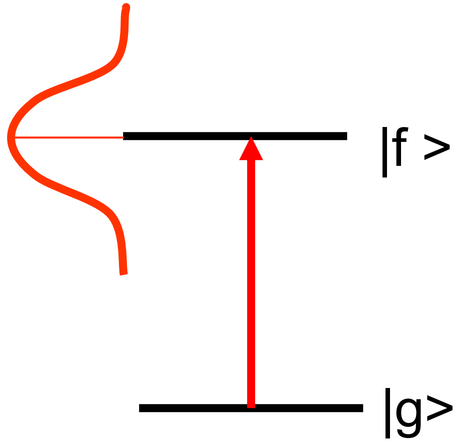
1^{st} Order Time-Dependent Perturbation Analysis

$$a_f(t) = \frac{\mu_{fg}}{i\hbar} \int_{-\infty}^{t} dt_1 \, \varepsilon(t_1) \, e^{i\omega_{fg} t_1}$$

$$\boxed{a_f(\infty) = \frac{\mu_{fg}}{i\hbar} E(\omega_{fg})}$$

Pulse Area

$$p_f(\infty) \propto \left| E(\omega_{fg}) \right|^2$$



Transitions only by resonant light
Pulse shaping not effective – control not possible
for times after the pulse (t=∞)

Interaction of a broad band pulse and a narrow resonant transition

Temporal area

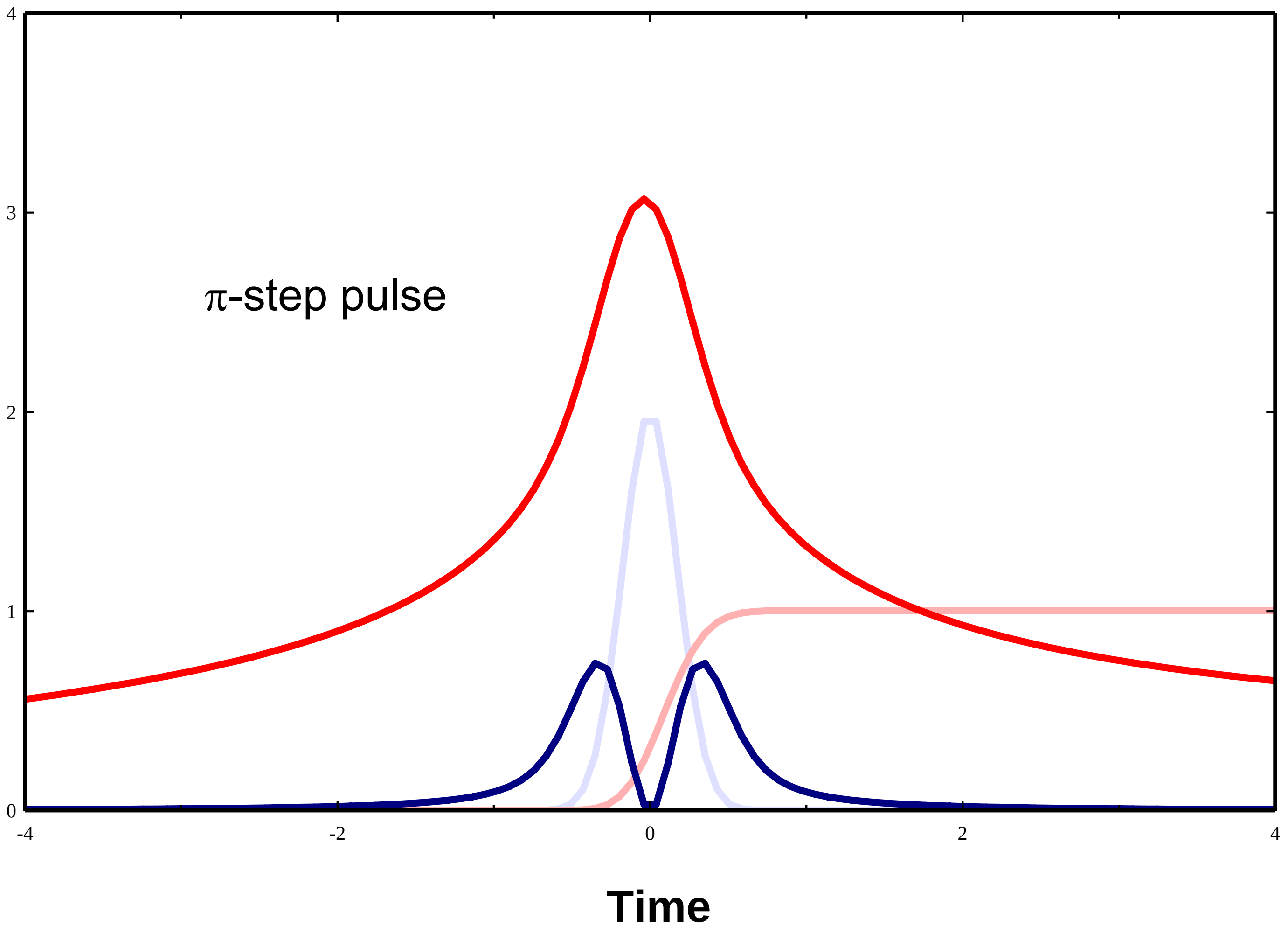
$$a^{(1)}(t) = \frac{\mu_{1g}}{i\hbar} \int_{-\infty}^{t} dt_1 e(t_1) \exp(i\omega_0 t_1)$$

During the pulse, all frequencies contribute:

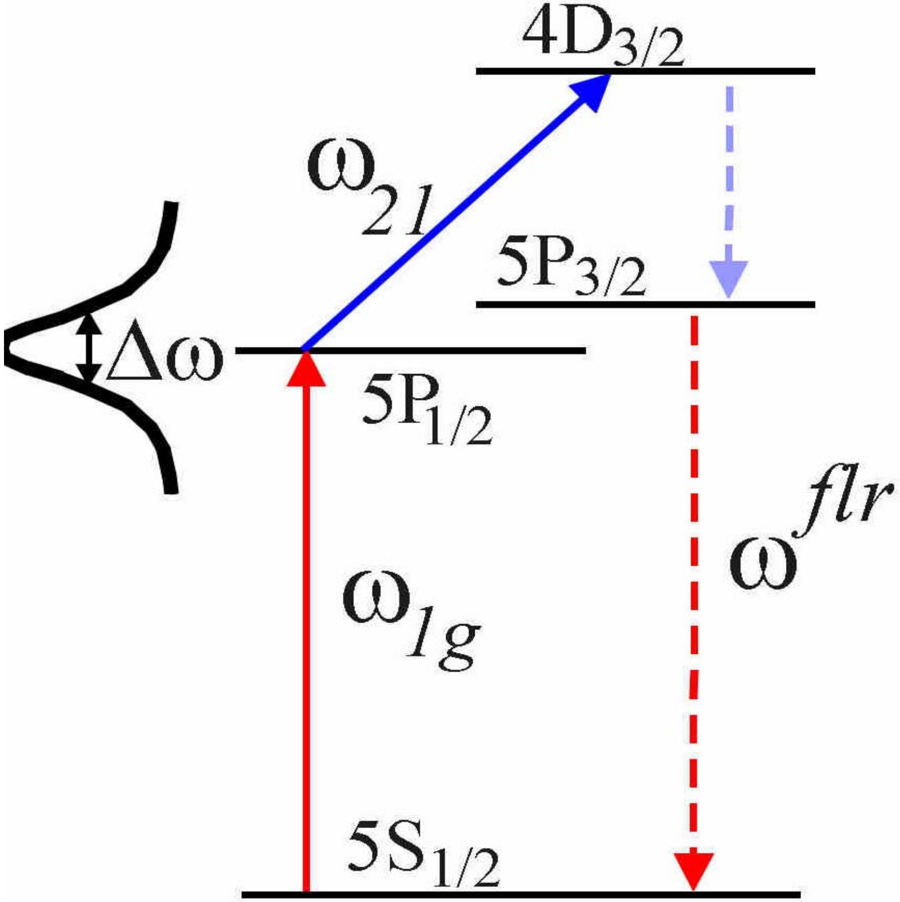
$$a^{(1)}(t=0) = -\frac{\mu_{1g}}{\hbar} \left[\underset{\uparrow}{i\pi E(\omega_0)} + \wp \int_{-\infty}^{\infty} d\omega_1 \underset{\uparrow}{\frac{E(\omega_1)}{\omega_0 - \omega_1}} \right]$$

on-resonance off-resonance

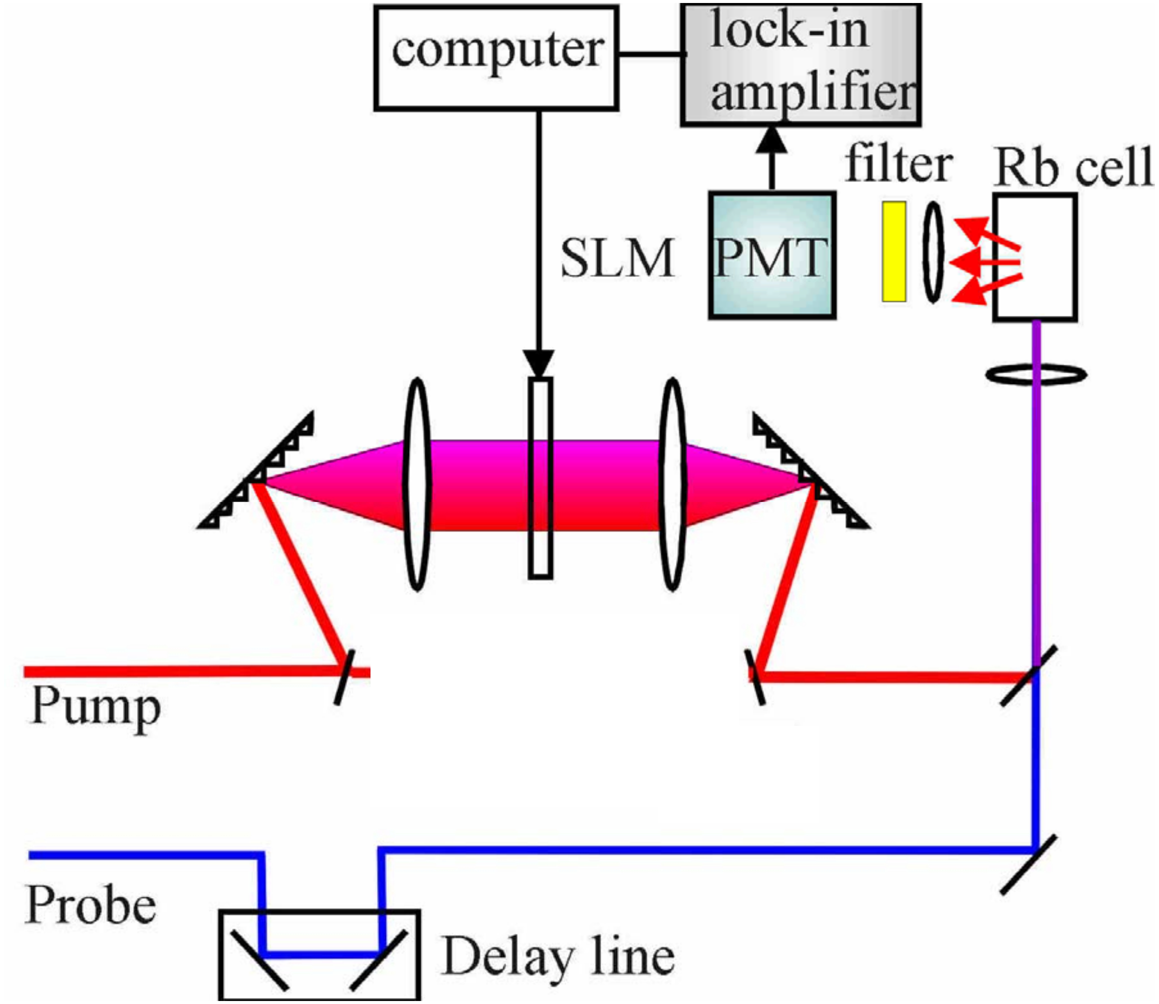
Transient population with shaped pulses



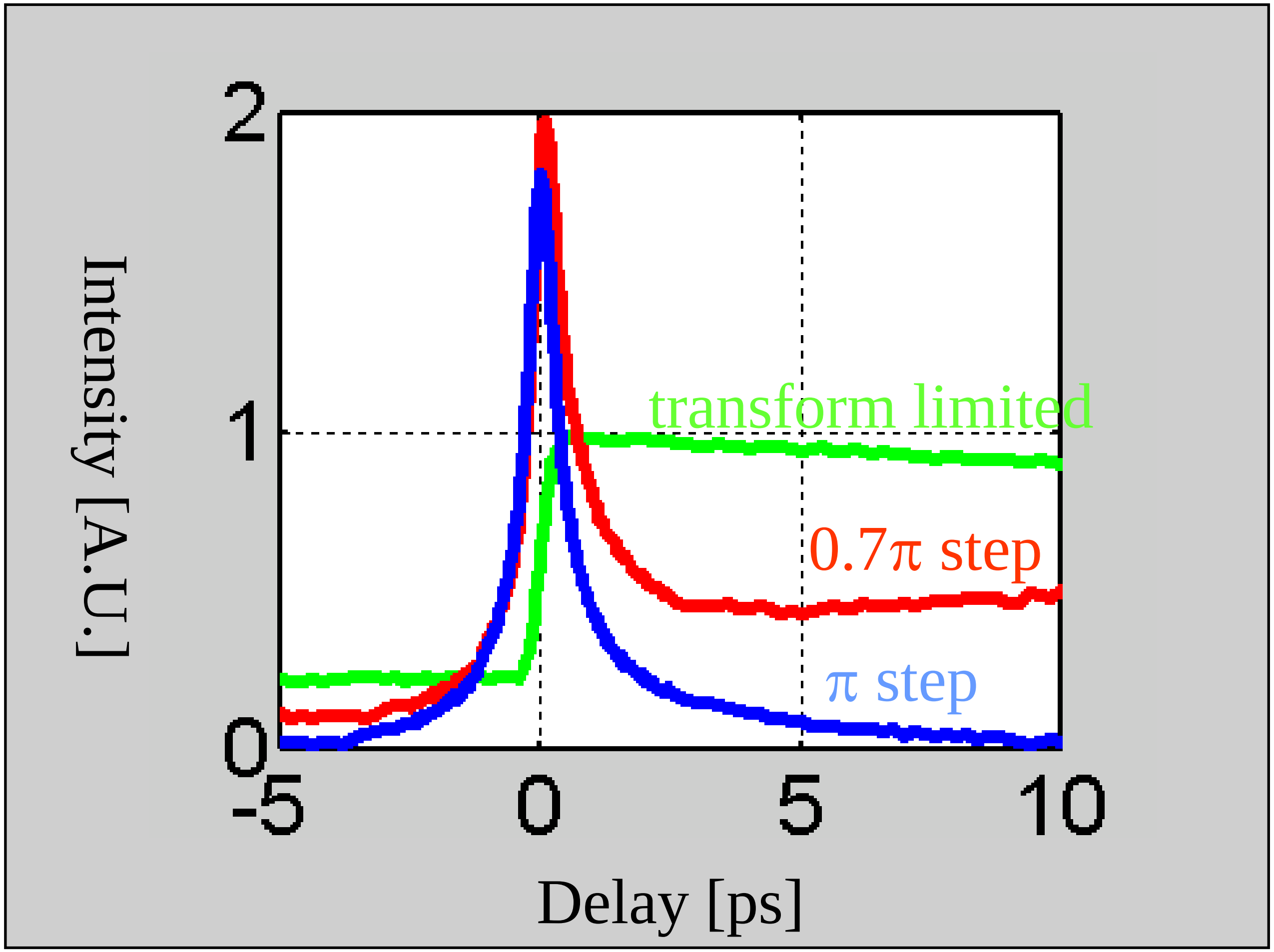
Pump probe experiment in Rb atoms



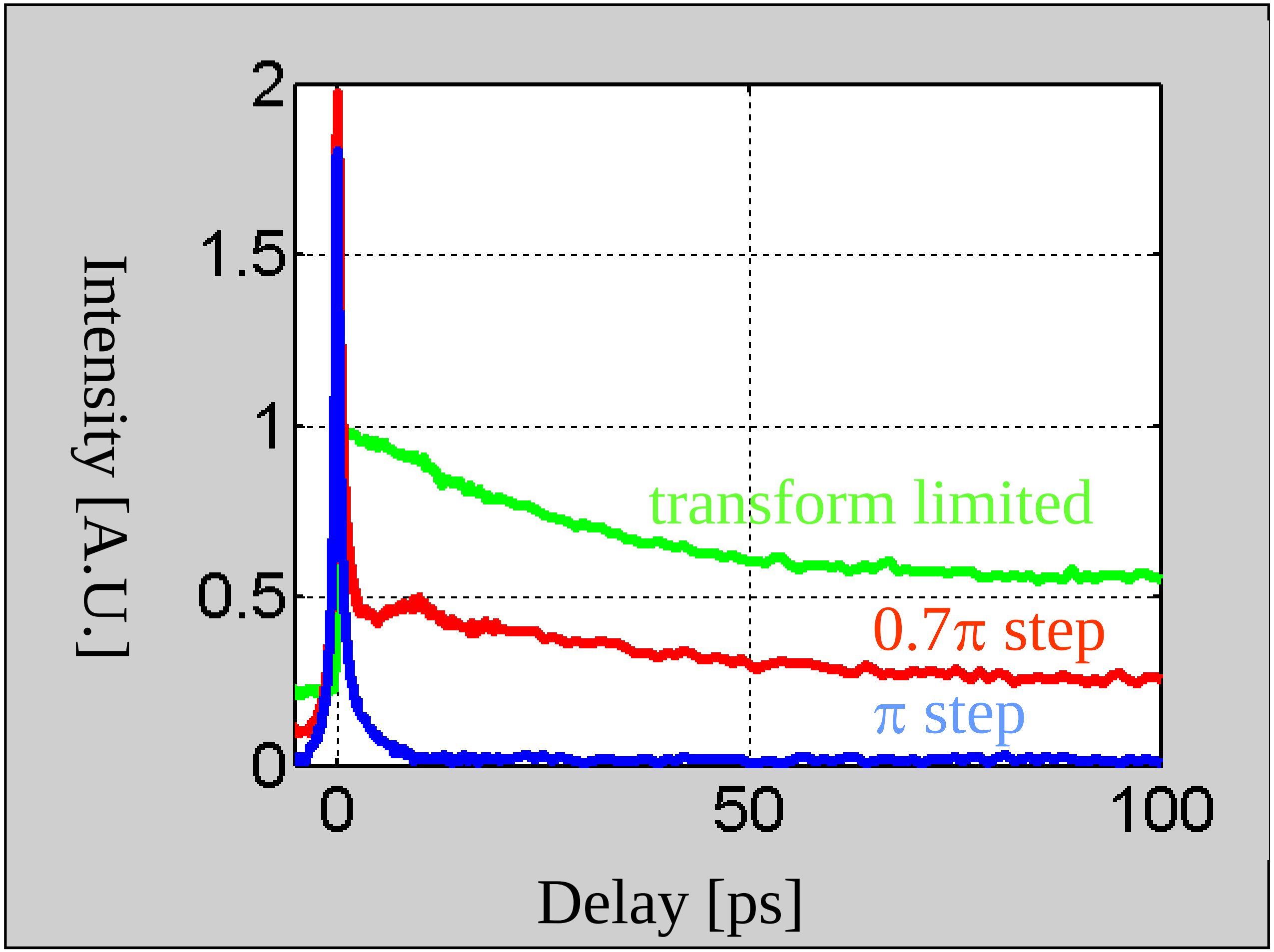
Experimental set-up



Transient population enhancement experimental results



Transient population enhancement experimental results



The theoretical limit

$$\frac{\max\left(a_{SH}^{(1)}\right)}{\max\left(a_{TL}^{(1)}\right)} \approx \frac{1}{\pi}\ln\left(\Delta\omega\cdot T_2\right)$$

Shaper
resolution

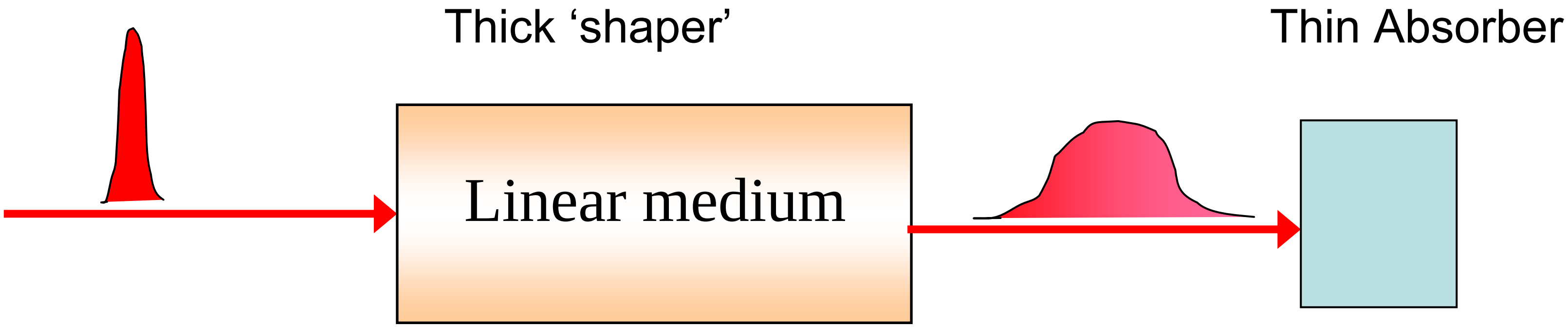
inhomogeneous
life time

With pulse shaper T2>> 1/dω

T_2 in atoms ≅ 10^{-9} sec

1/dω ≅ 3 psec

“Atomic shaper”



Thick ‘shaper’

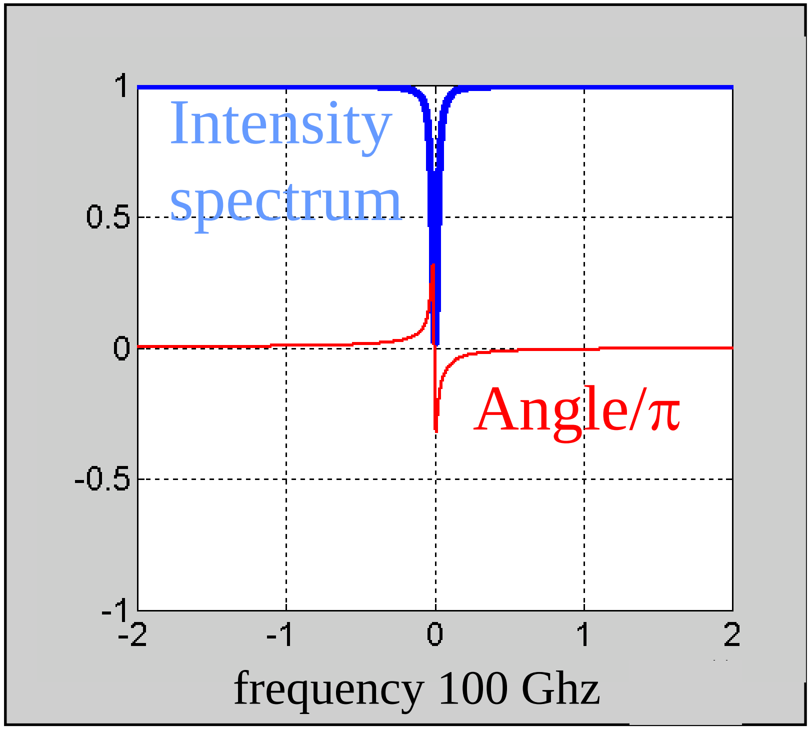
Thin Absorber

Linear medium

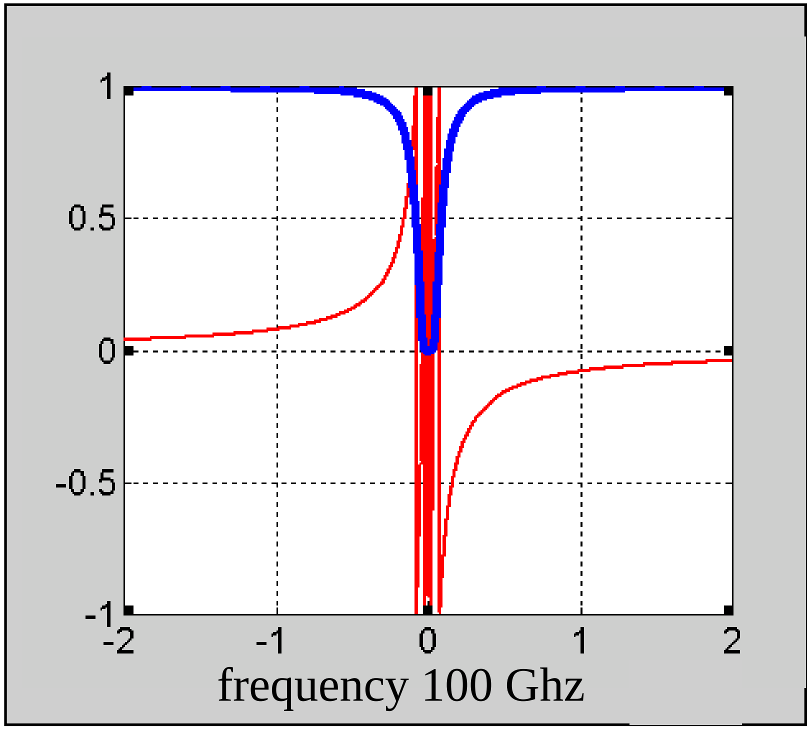
$$E(\omega,l) = E(\omega,0)\cdot\exp\left[\frac{-\alpha_0 l}{1-i(\omega-\omega_0)T_2}\right]$$

Phase inversion with a T_2 resolution!

“Atomic shaper”

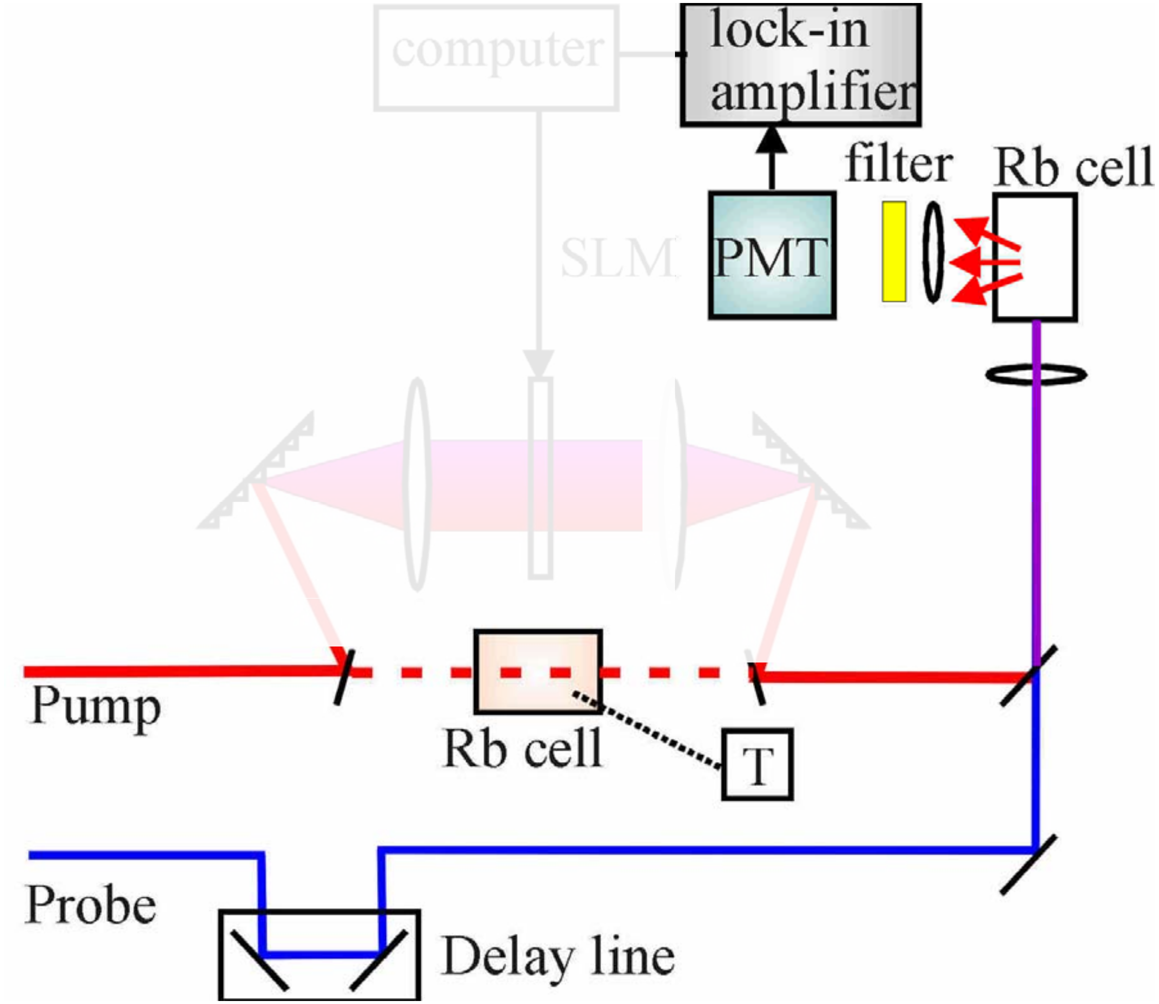


$$\alpha_0 l = 2$$

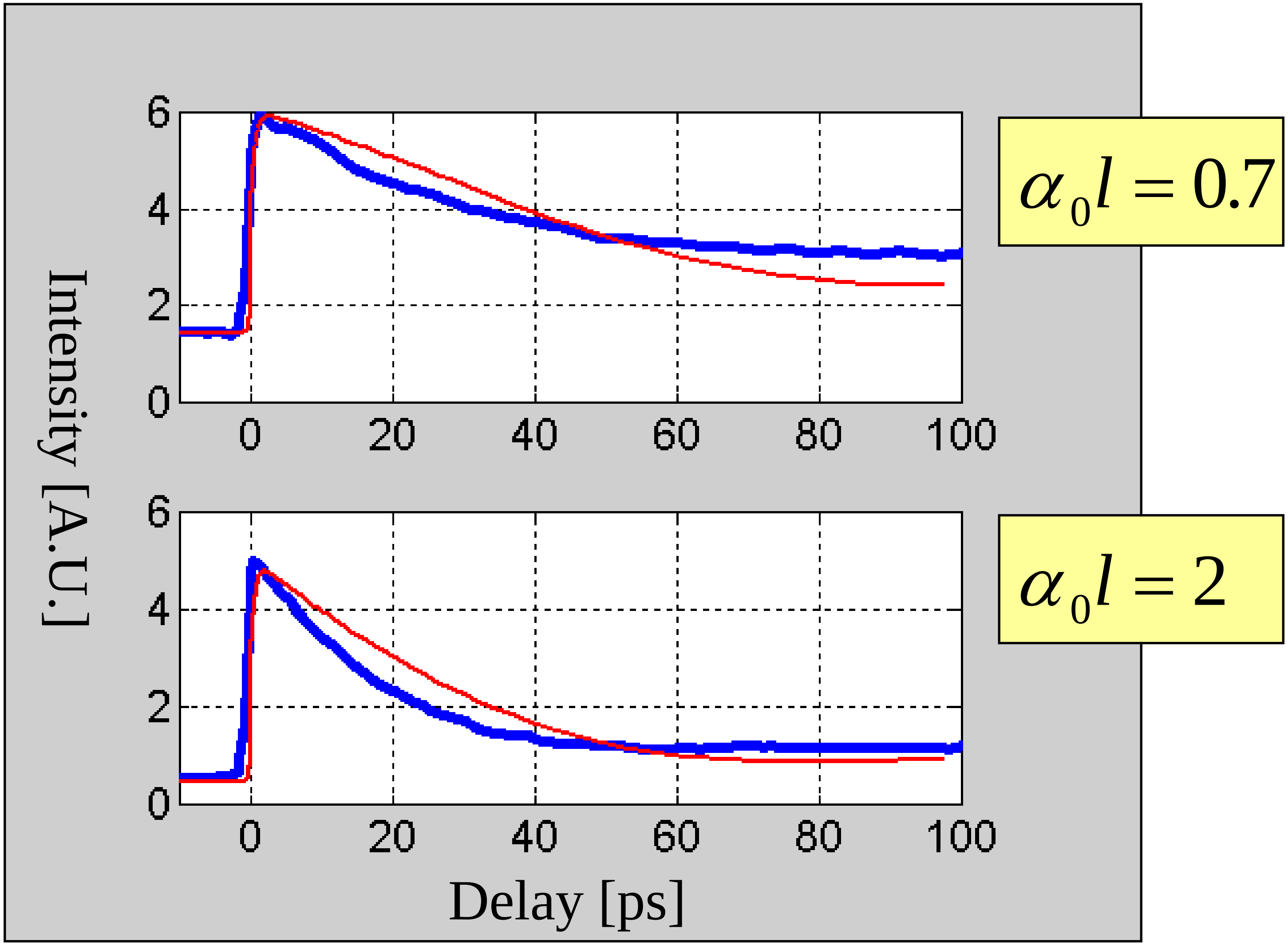


$$\alpha_0 l = 25$$

“Atomic shaper”



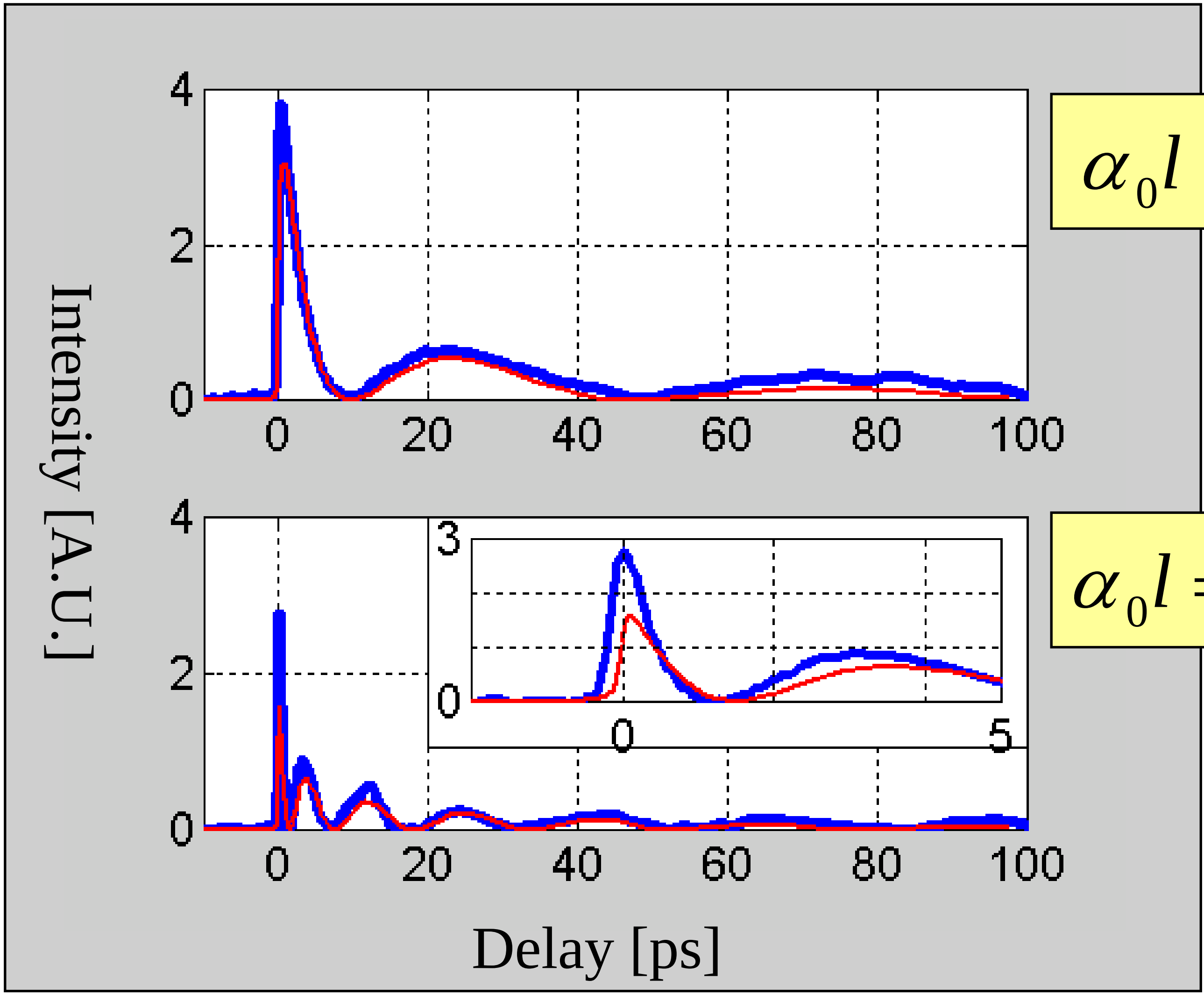
Transient population by propagation effects



$$\alpha_0 l = 0.7$$

$$\alpha_0 l = 2$$

Transient population by propagation effects



$$\alpha_0 l = 25$$

$$\alpha_0 l = 200$$

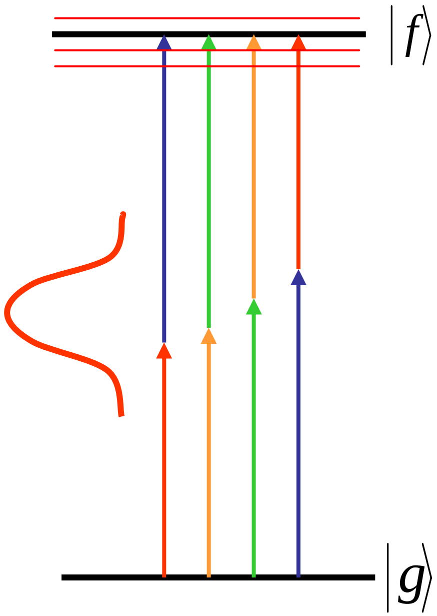
Control of Multiphoton Transition

1. Two-Photon Absorption
2. Coherent Transients
3. Strong Fields
4. CARS spectroscopy

The problem with strong-field control

$$\xcancel{a_f(\infty) \propto \int \varepsilon^2(t)\exp(i\omega_{fg}t)dt}$$

- Perturbation analysis is no longer valid
- Power broadening and AC shifts complicate response
- Transitions no longer depend on a single frequency-component of $\varepsilon^2(t)$



The Solution: Fields with a single quadrature

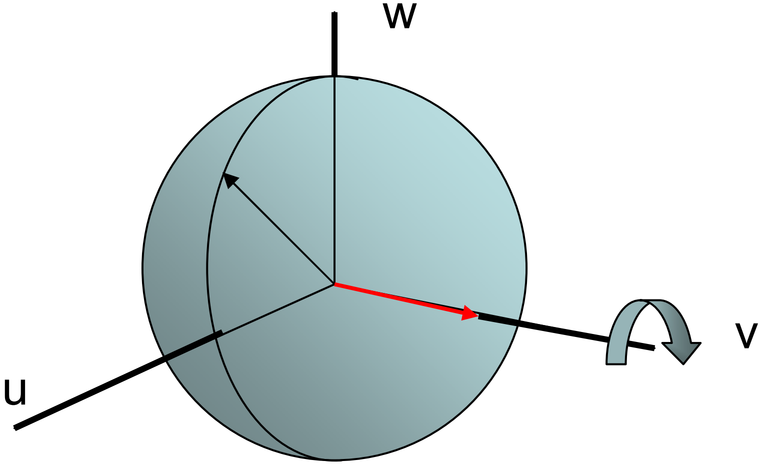
$$E(t) = A(t)\exp(i\omega t)$$

Carrier is modulated only in amplitude.

With *A(t)* a REAL function, a two-level system evolves only with the pulse area

$$\theta = \int A(t)dt$$

The area is just the Fourier component of the resonant frequency



The Solution: Fields with a single quadrature

For a nonresonant N-photon transition, same is true if

$$E^{N}(t) = A(t)\exp(i\omega t)$$

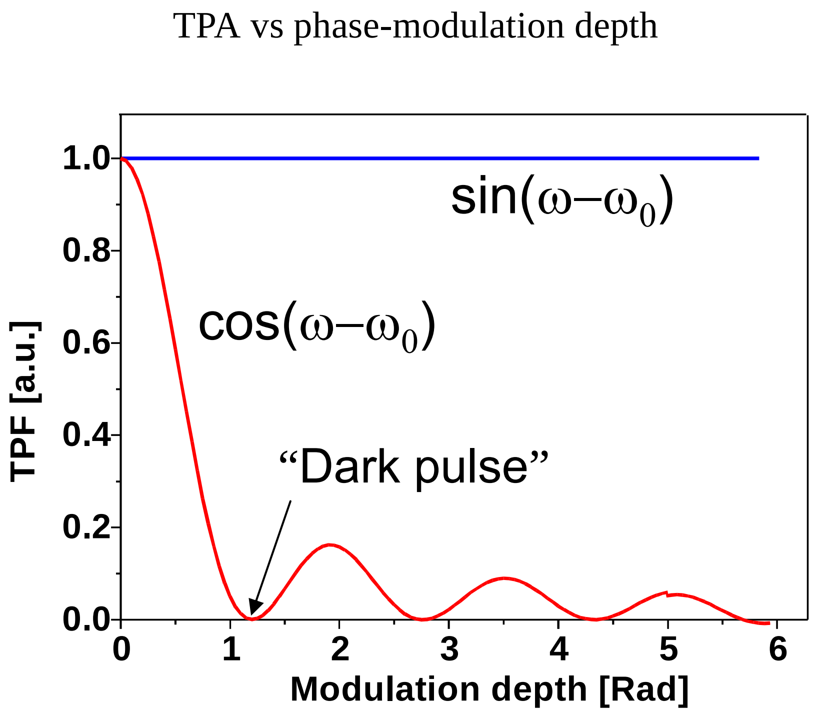
With *A(t)* a REAL function. The system is again driven by a single frequency

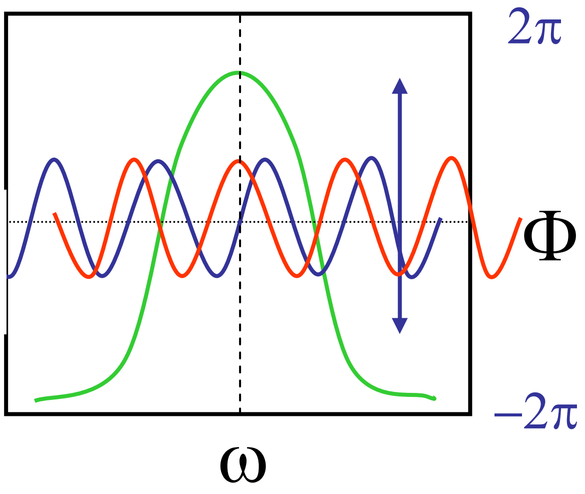
$$\theta_{N} = \int A_{N}(t)\,dt$$

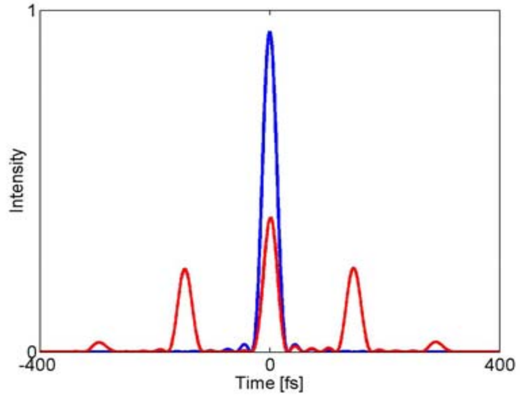
Phase modulation by *sin* or *cos* both yield single-quadrature $E^2(t)$ fields!

Nonresonant TPA

modulation with a periodic phase

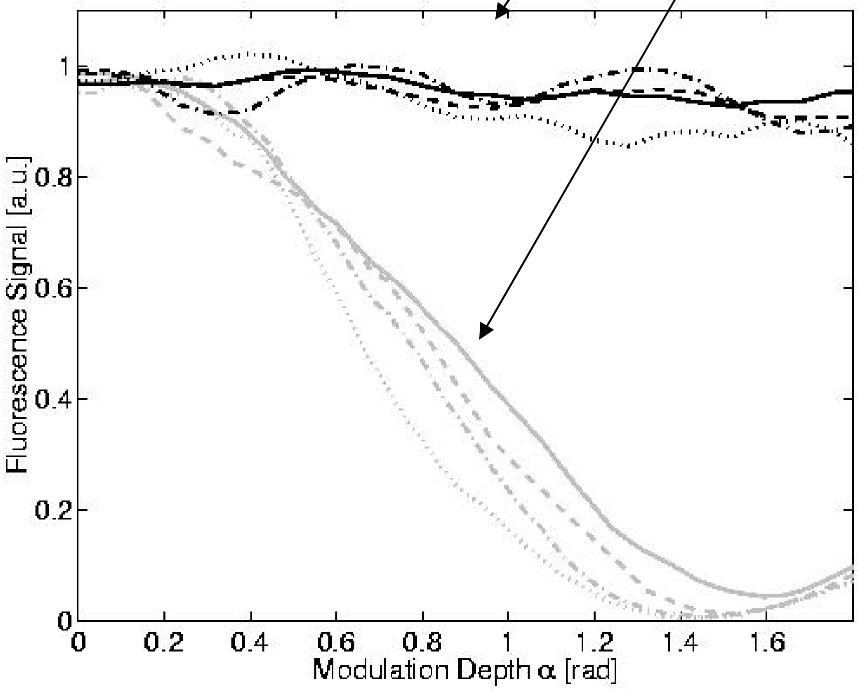


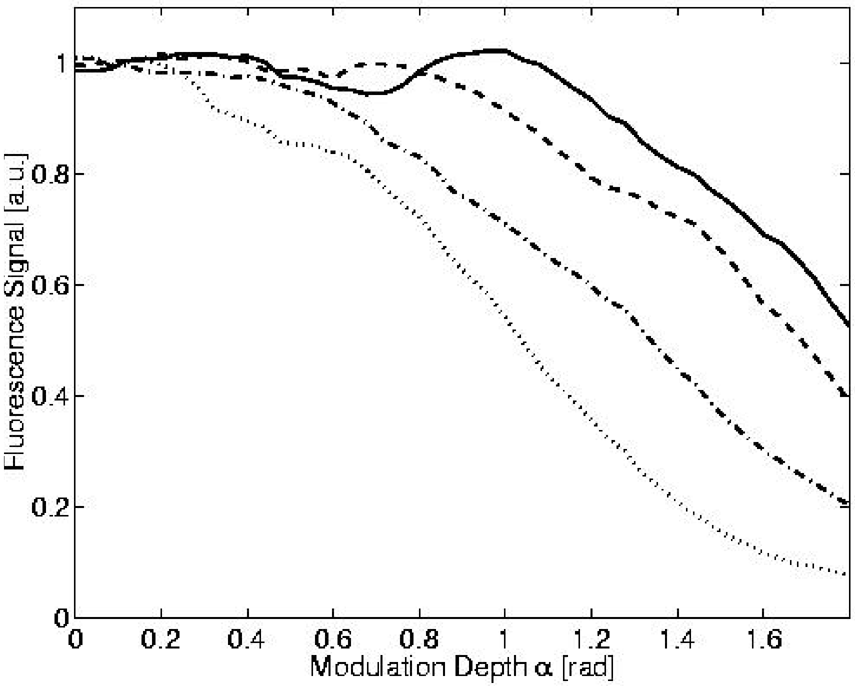




Control with a single quadrature

Phase modulation by sin or cos work also in the strong field regime





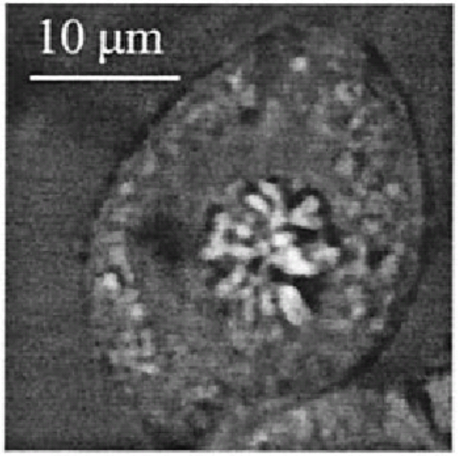
Dudovich et al. PRL 94, 083002 (2005)

Control of Multiphoton Transition

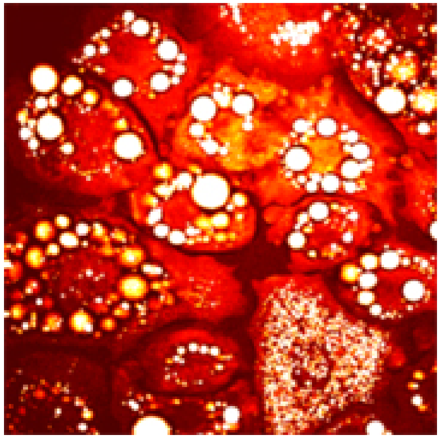
1. Two-Photon Absorption
2. Coherent Transients
3. Strong Fields
4. CARS spectroscopy

CARS Microscopy

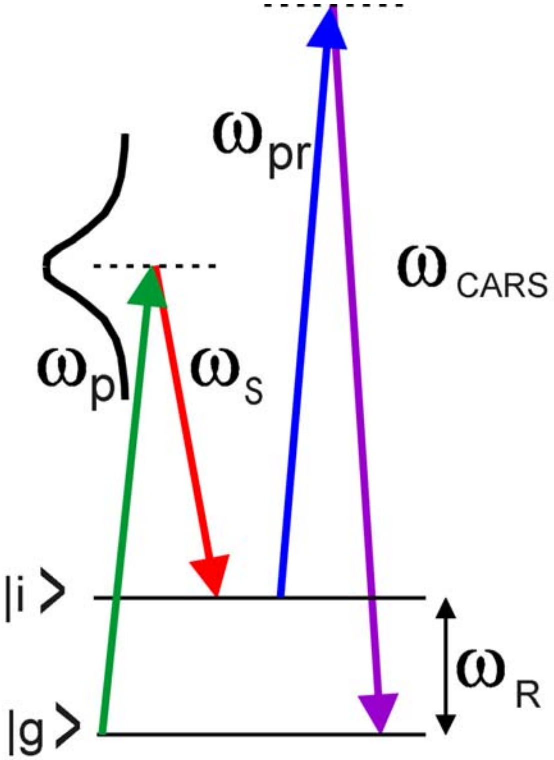
CARS Image tuned to DNA backbone vibration at 1090 cm^{-1} in mitosis



CARS image of **fibroblast cells** that are stimulated to synthesize lipids. The lipid droplets are visualized with CARS tuned to the C-H vibration at 2845 cm^{-1}.



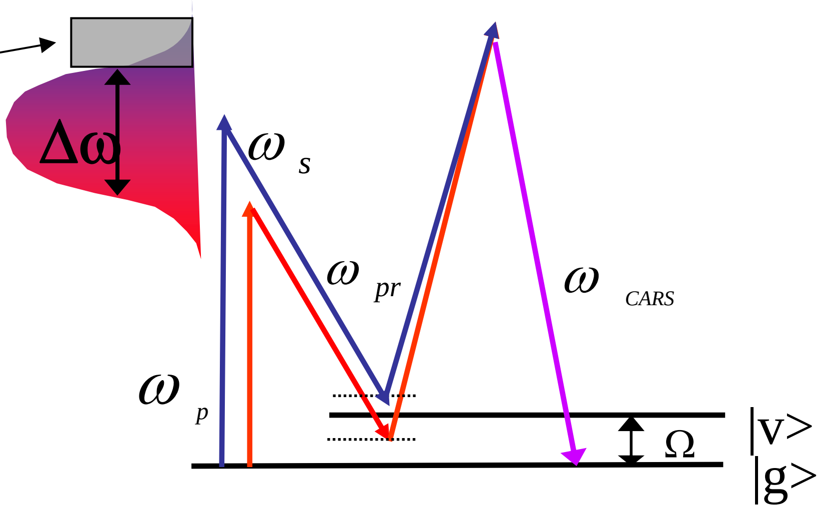
Xie's group, Harvard



Single-Pulse CARS spectroscopy

A single ultrashort, broadband pulse (shorter than the vibrational period) to provide all 3 frequencies

High frequencies blocked to detect CARS signal

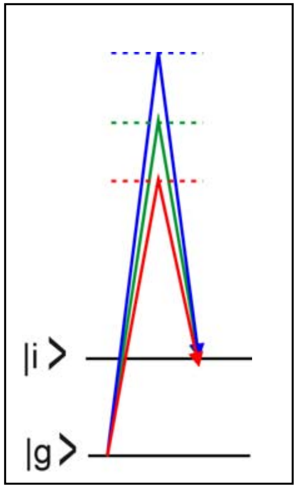


Issues: Resolution

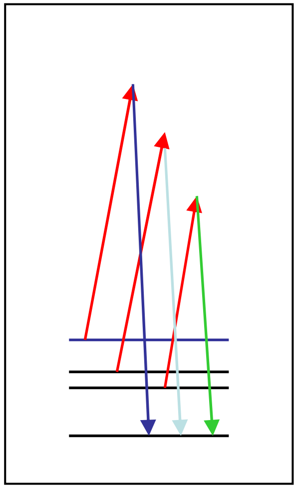
Nonresonant Background

CARS control schemes

- Goal: to achieve high-resolution (ps) CARS spectroscopy using a single broadband source through coherent control
- Methods:
  - Selective excitation
    Use quantum control to excite just a single Raman level



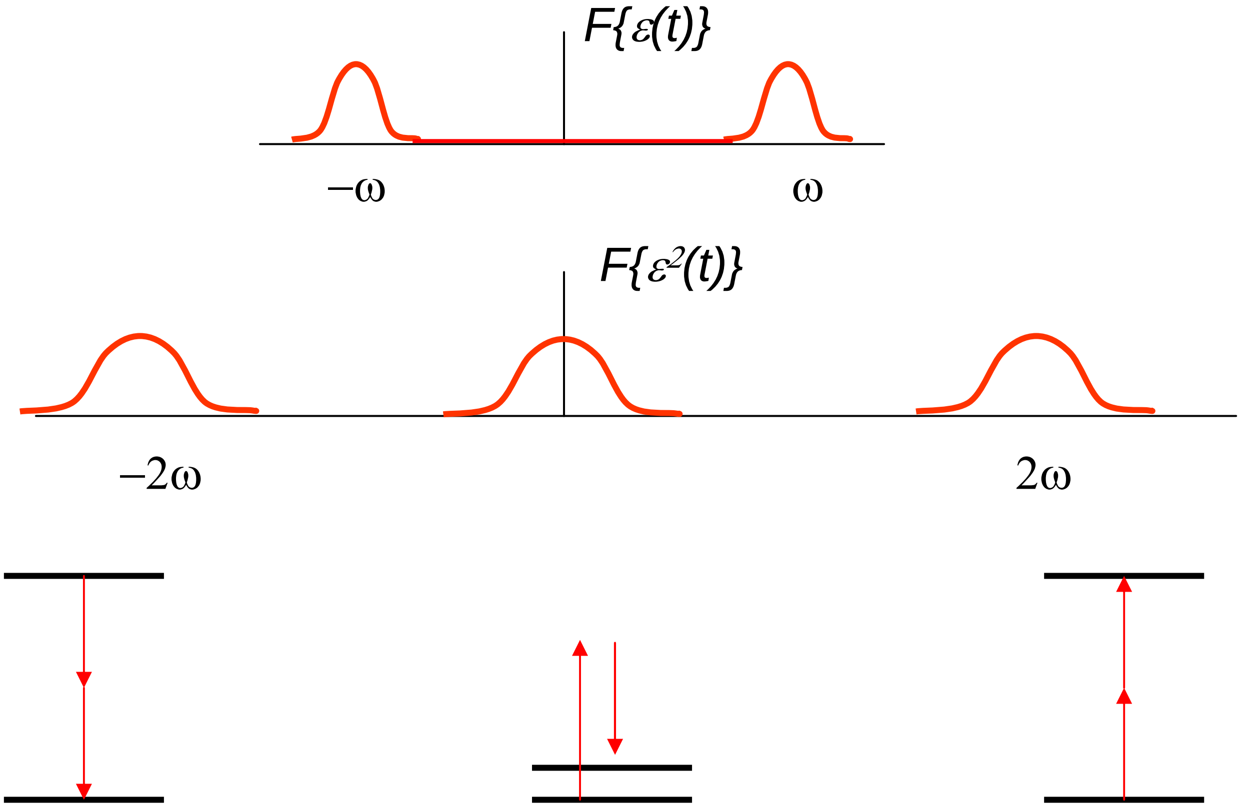
  - Multiplexed CARS
    Excite with wide band, read with an effective narrow probe to resolve spectrum



Two-photon processes

2^{nd} Order Time-Dependent Perturbation Analysis

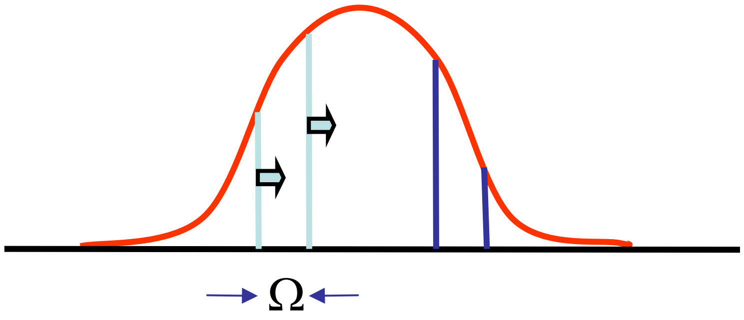
$$a_f(\infty) \propto \int \varepsilon^2(t) \exp(i\omega_{fg} t)\, dt$$

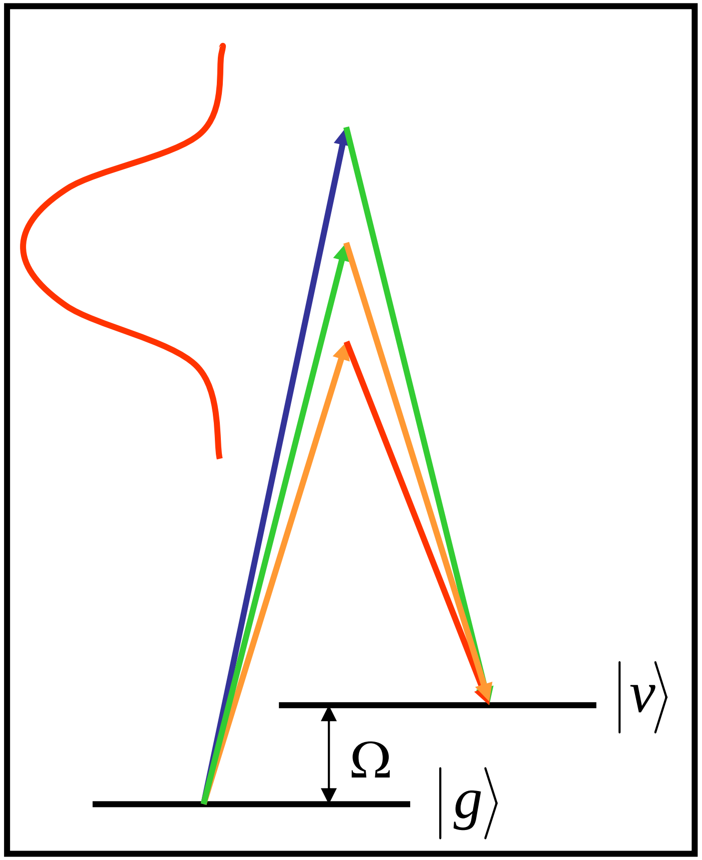


Broad-band excitation of a Raman transition

$$A_v(\Omega) \propto \int d\omega E(\omega) E^*(\omega - \Omega) =$$

$$= \int d\omega |E(\omega)| |E(\omega - \Omega)| \cdot \underline{e^{i[\Phi(\omega) - \Phi(\omega - \Omega)]}}$$



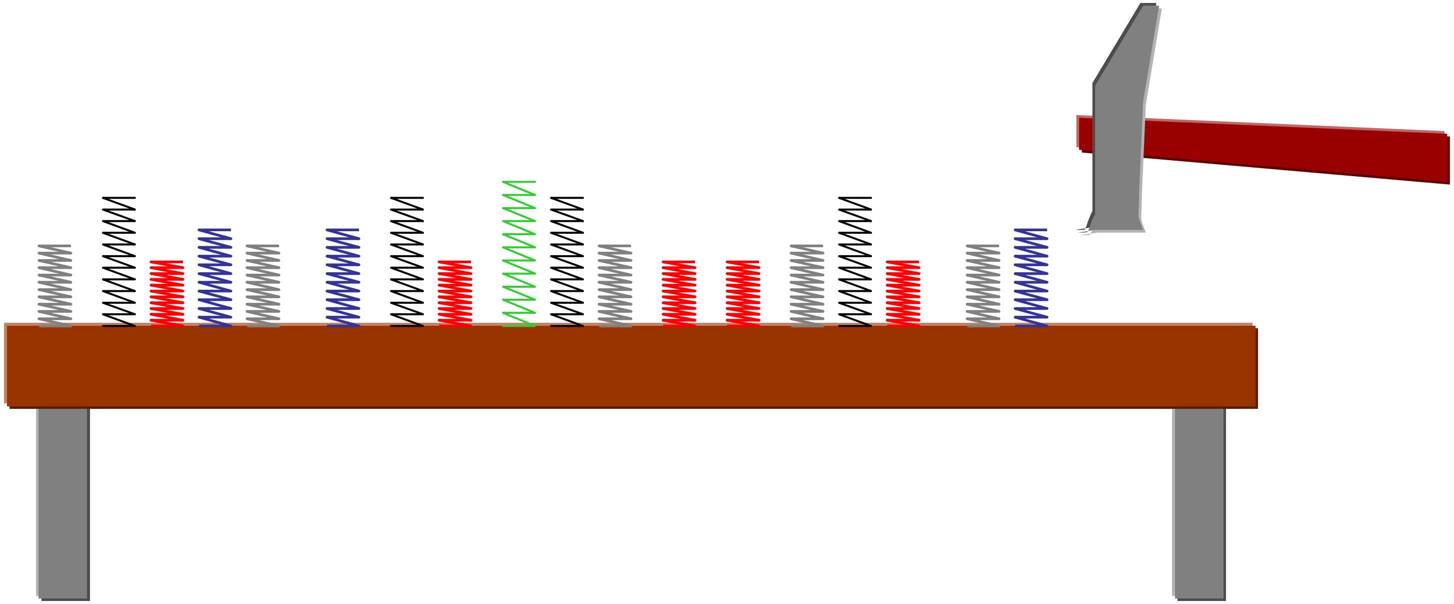


Transform-limited pulses maximize transition rates

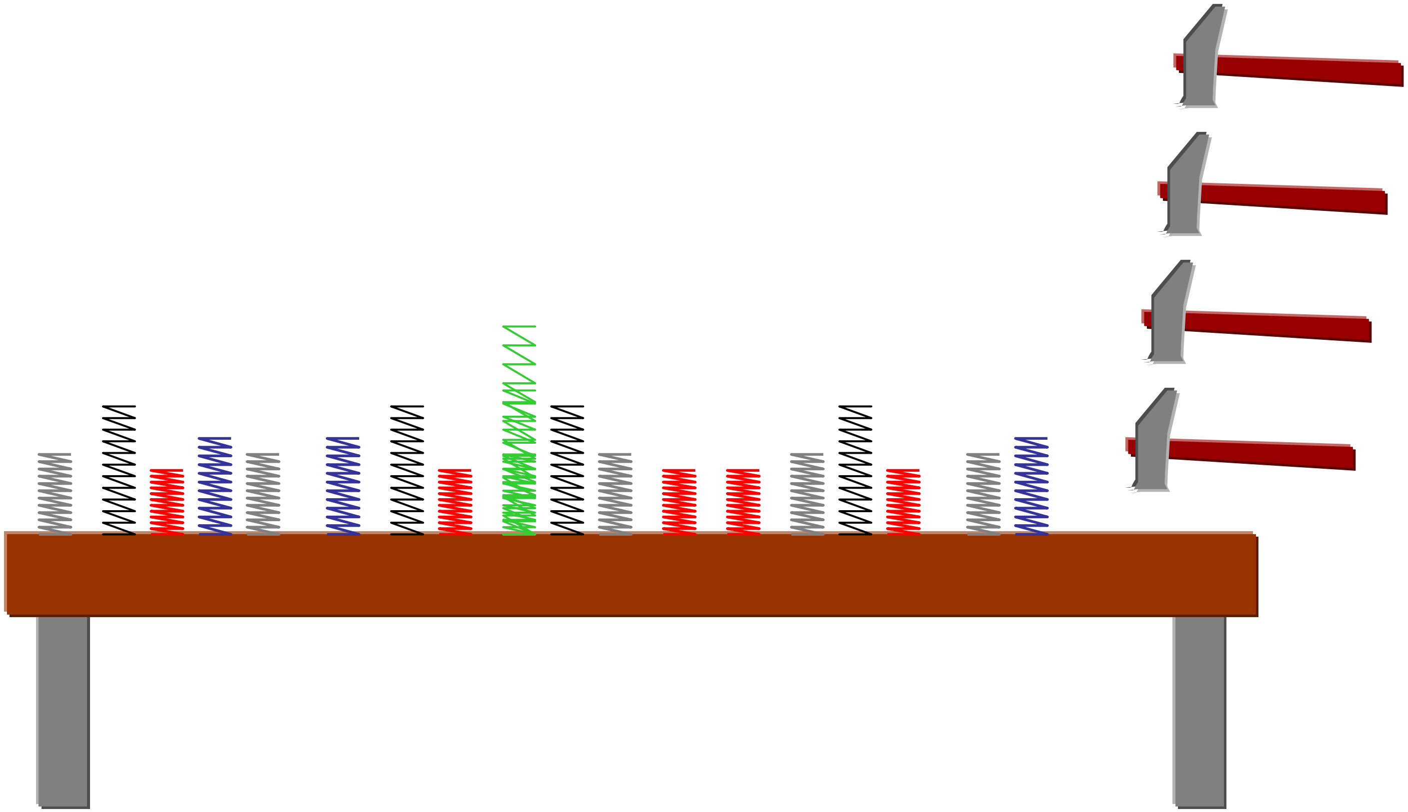
Periodic phase functions maintain efficiency

Oron *et al*., Phys. Rev. A **65**, 043408 (2002)

Impulsive excitation

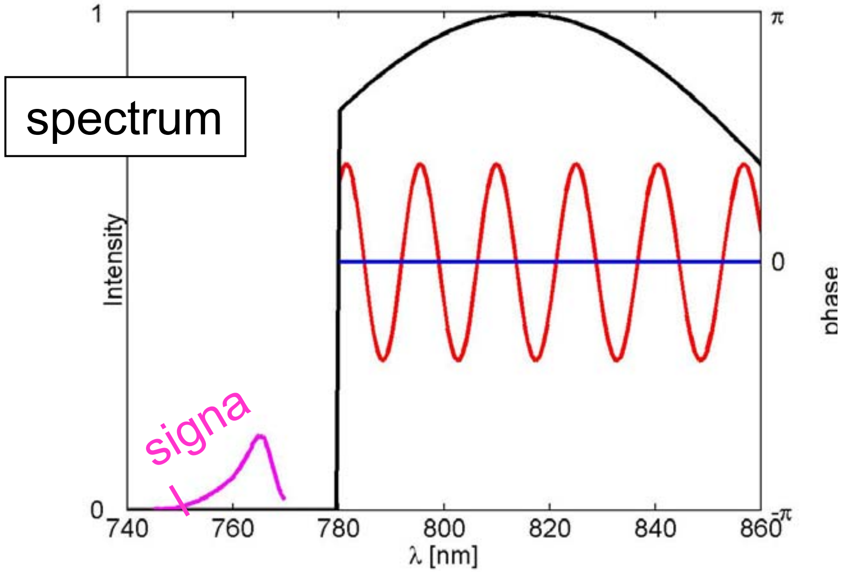


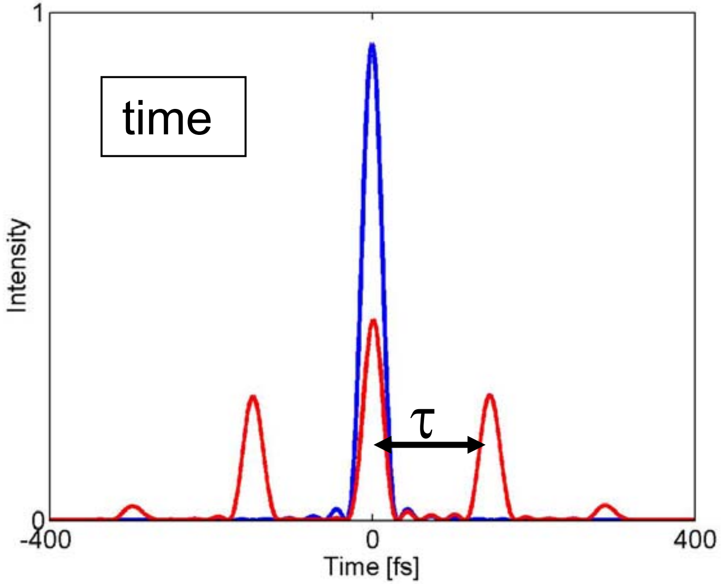
Selective excitation



Weiner *et al.*, Science **273**, 1317 (1990)

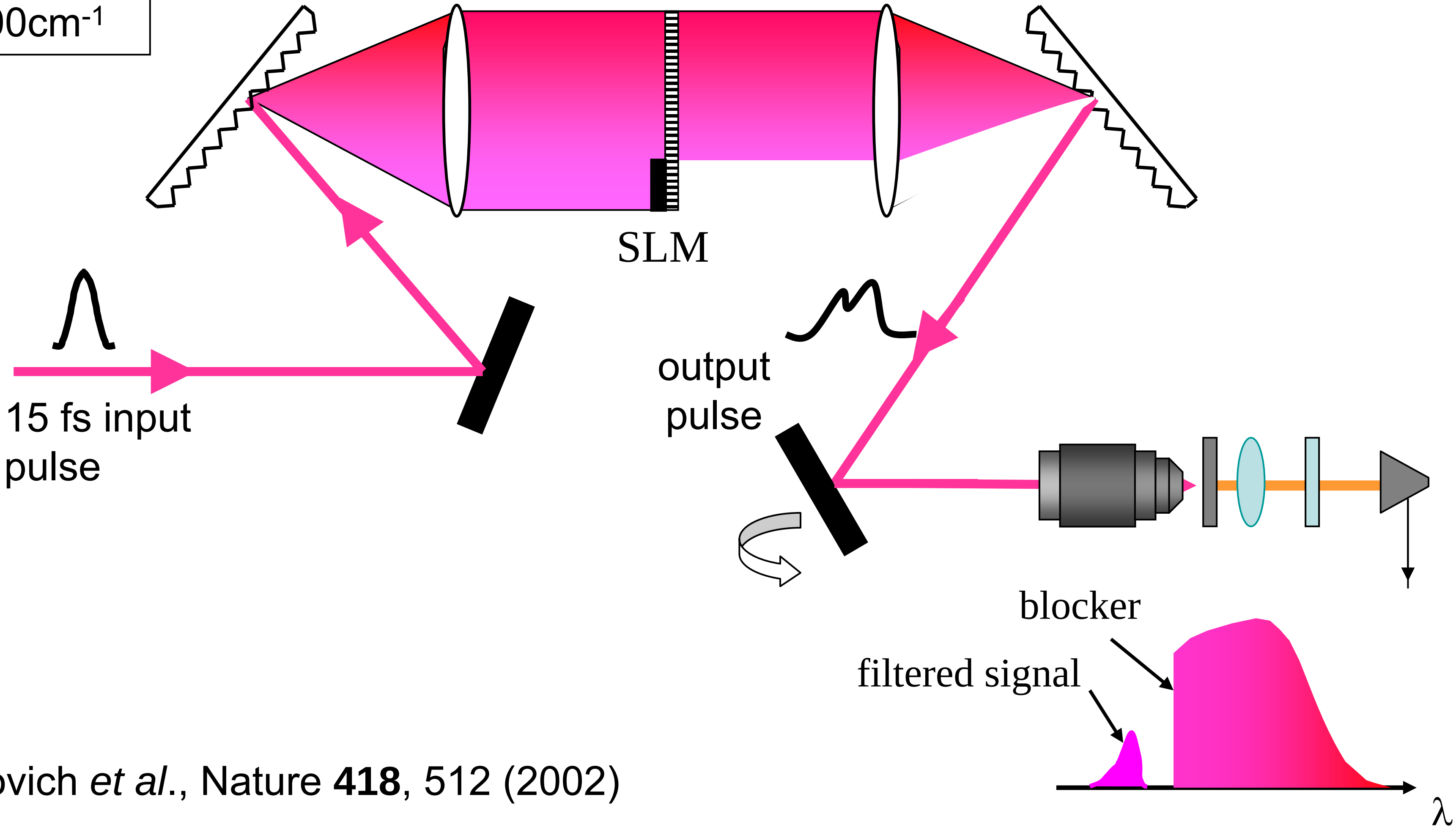
Single-pulse CARS with periodic phase





Single-pulse CARS microscopy

Pulse bandwidth $1500 cm^{-1}$



SLM

15 fs input pulse

output pulse

blocker

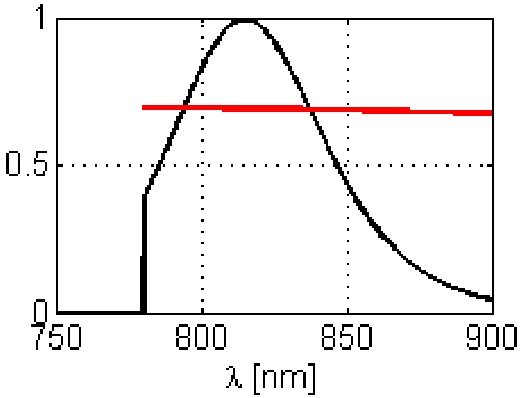
filtered signal

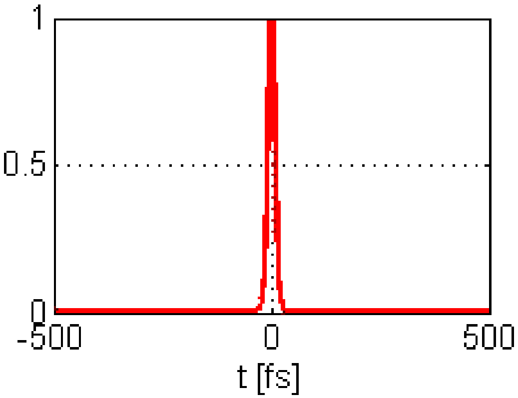
λ

Dudovich *et al*., Nature **418**, 512 (2002)

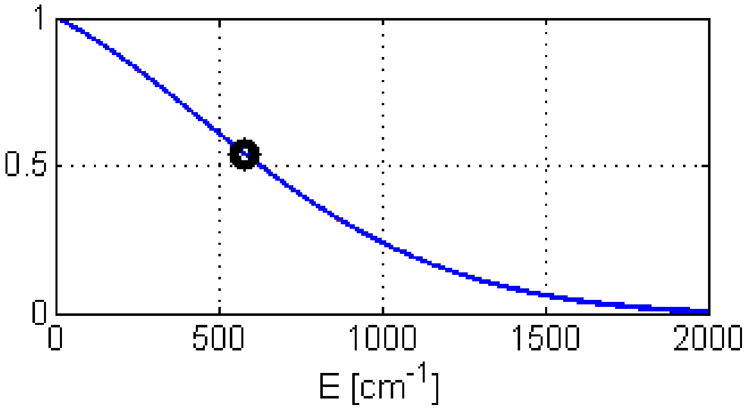
Single-pulse CARS with periodic phase - Spectroscopy by selective excitation

Spectral phase





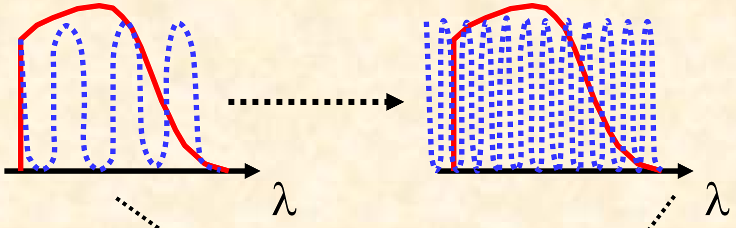
Temporal profile



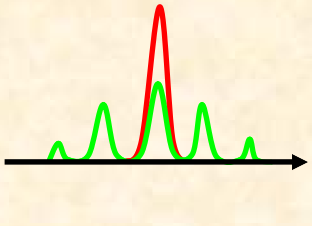
Population amplitude (monitor 577cm^{-1} level)

CARS spectroscopy

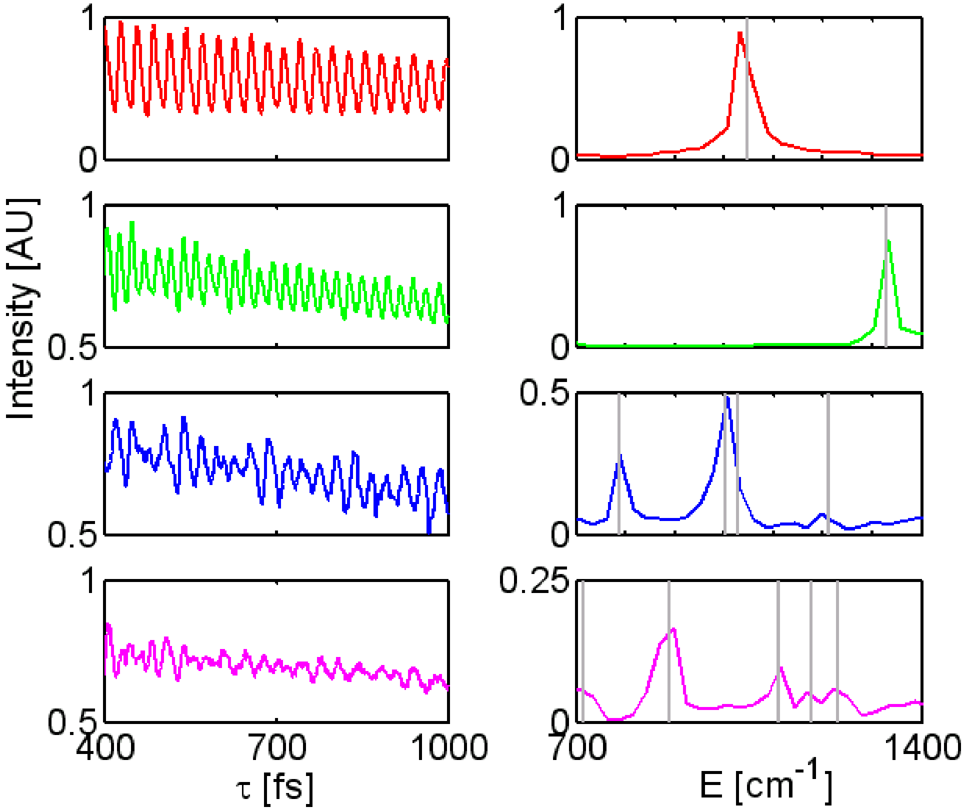
Modulated spectral phase function



$$\Phi = 1.25\cos\left(c\omega\right)$$



Fourier transform



$Ba(NO_3)_2$ (1048 cm^{-1})

Diamond (1333 cm^{-1})

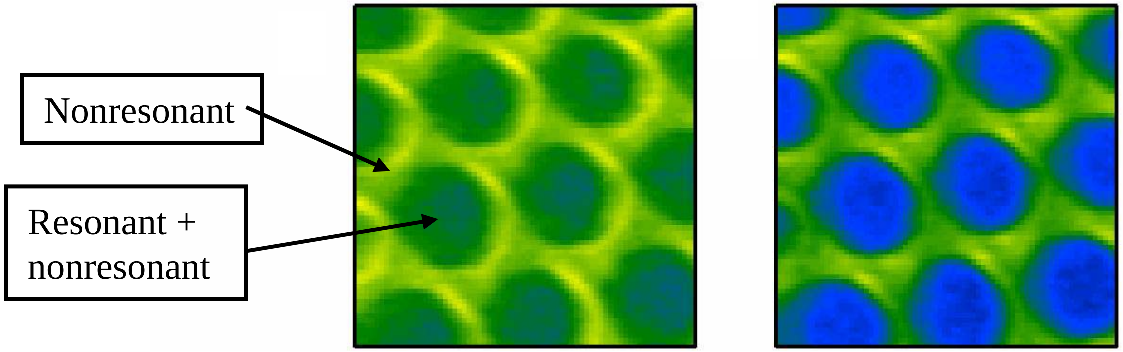
Toluene (788, 1001 cm^{-1})

lexan

Single-pulse CARS microscopy

Maximal resonant contribution

Minimal resonant contribution



Nonresonant

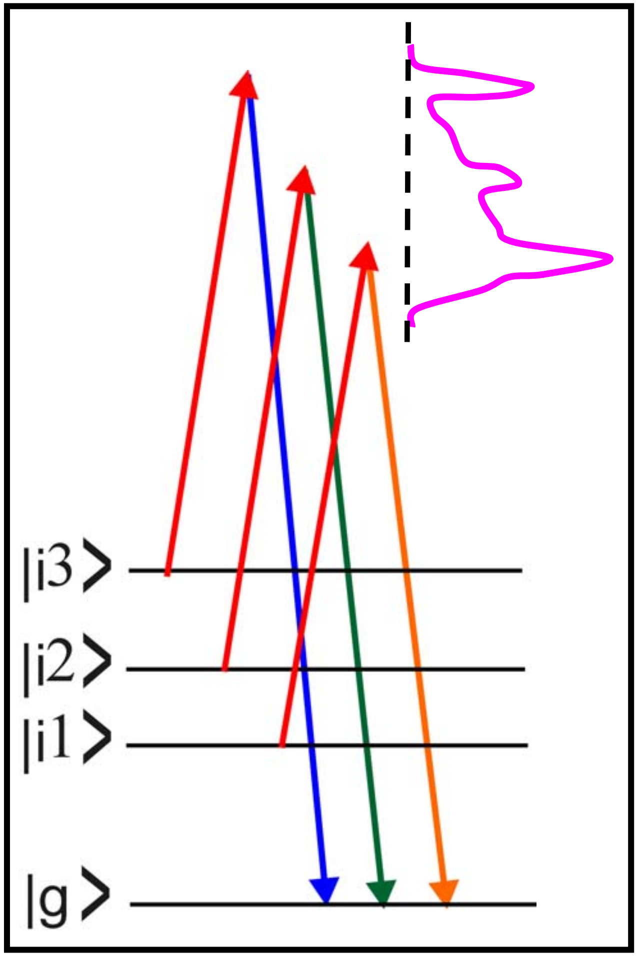
Resonant + nonresonant

Dudovich *et al*., Nature **418**, 512 (2002)

Pulses are shaped to maximize CARS signals from specific molecules

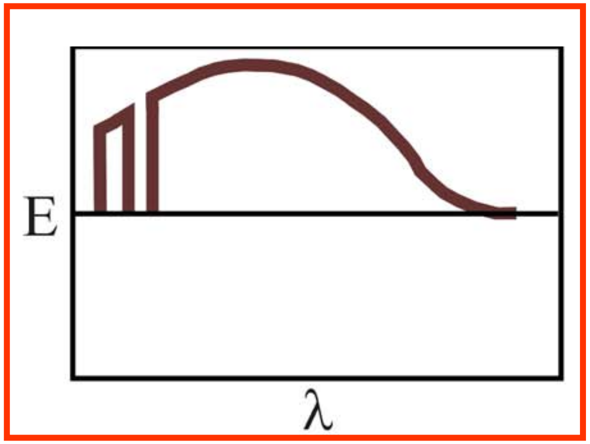
New fast pulse-shape modulation techniques are useful for Lock-in detection on pulse shapes

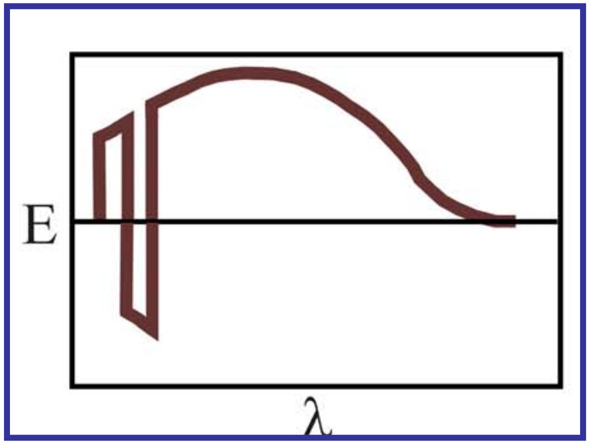
Single-pulse analog of multiplexed two-color CARS

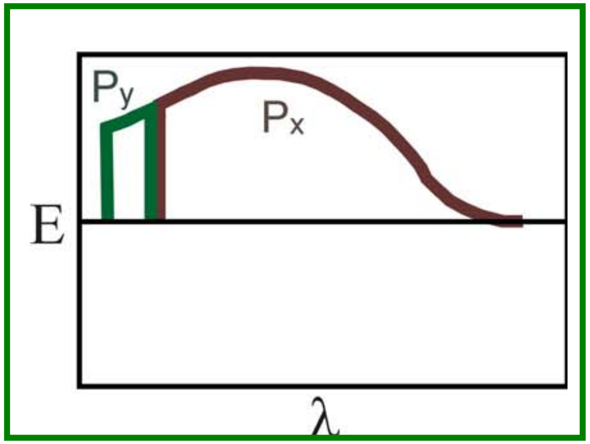


Simple schemes for separating a spectrally narrow probe within a broadband pulse

- Modulation of spectral amplitude
- Modulation of spectral phase
- Modulation of spectral polarization



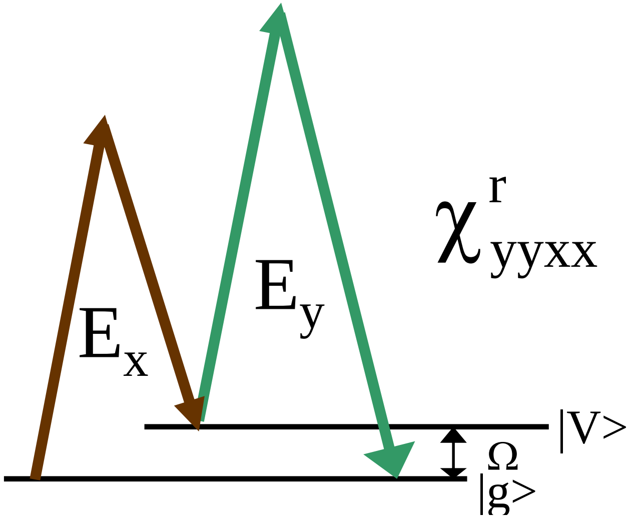




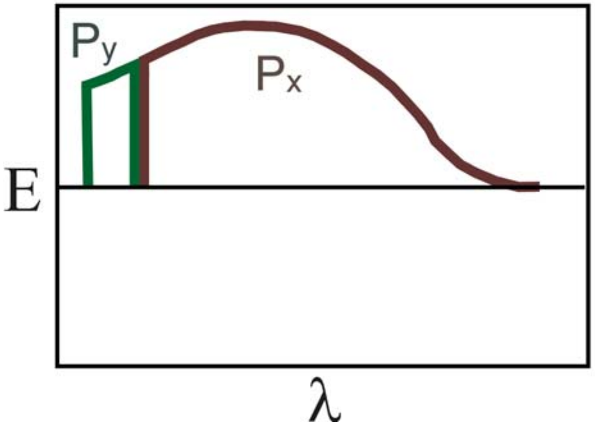
Oron *et al*., Phys. Rev. Lett. **89**, 273001 (2002)
Oron *et al*., Phys. Rev. Lett. **90**, 213902 (2003)

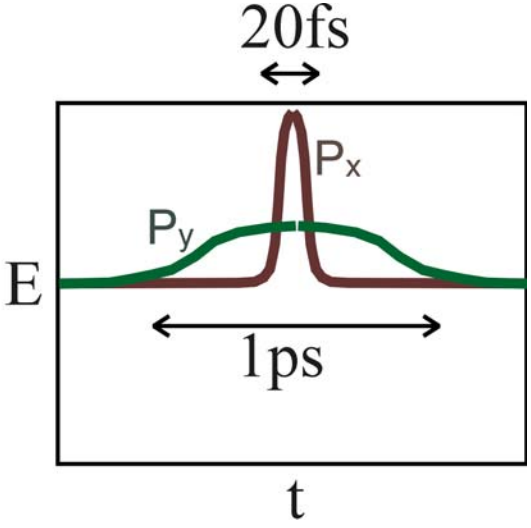
Steve Leone, Berkeley

Narrow probing by an orthogonal polarization



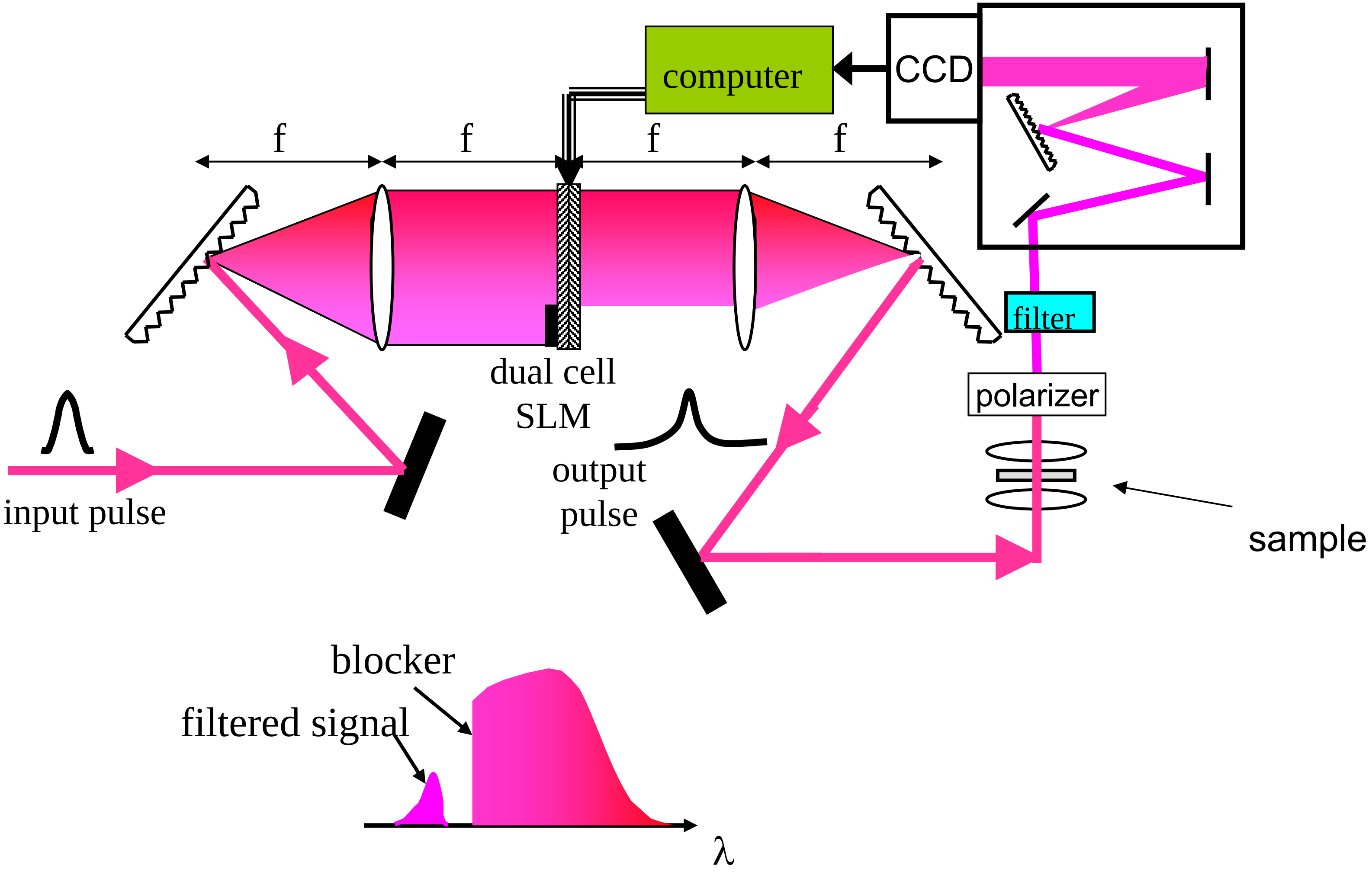
$$\chi^{r}_{yyxx} \; \ldots \; \chi^{nr}_{yyxx}, \chi^{nr}_{yxxy}, \chi^{nr}_{yxyx}$$





Oron *et al.*, Phys. Rev. Lett. **90**, 213902 (2003)

Polarization and phase shaping

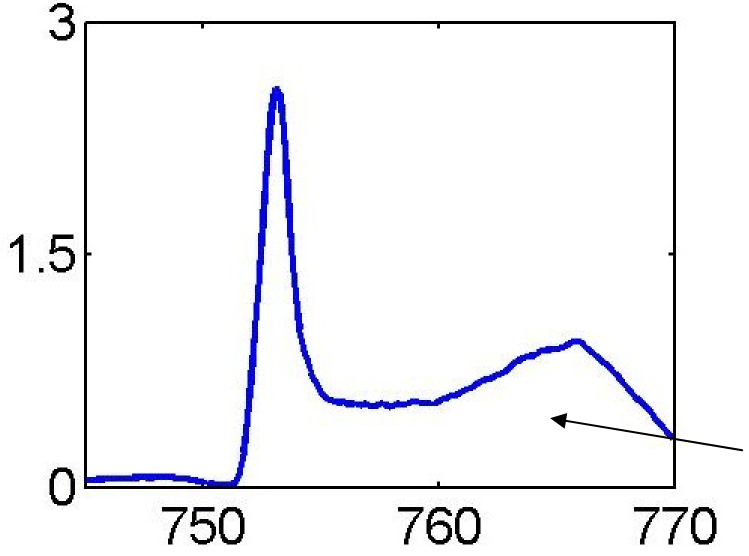


Brixner and Gerber, Opt. Lett. **26**, 557 (2002)

CARS spectrum - Narrow probing

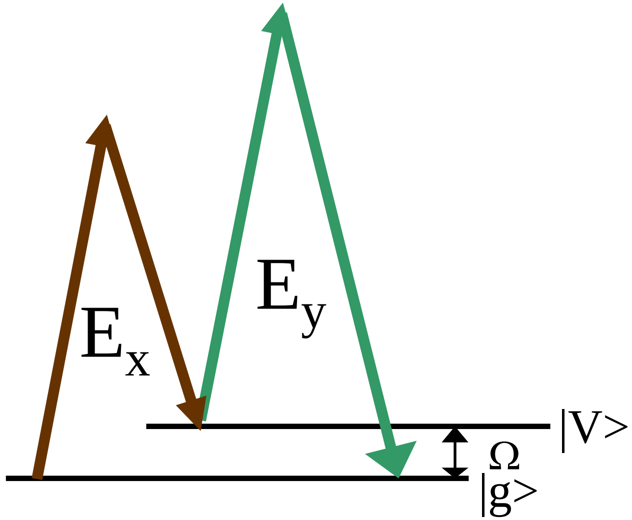
Measured CARS spectrum from iodomethane (523cm^{-1})

1.2nm (20cm^{-1}) wide y polarized probe

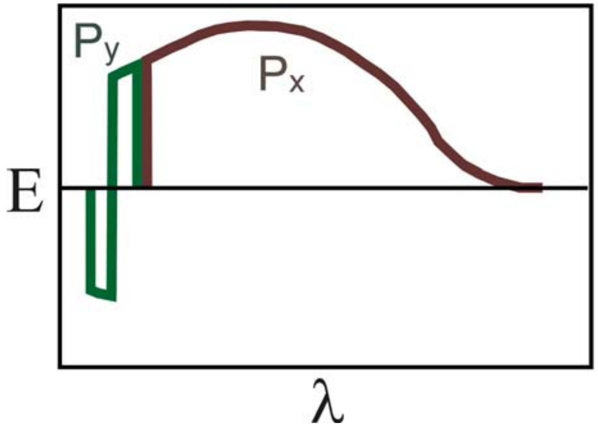


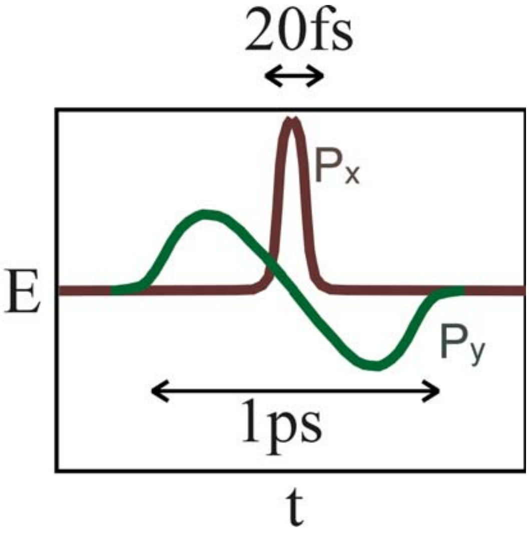
Nonresonant Background from overlap of the pump and probe

Narrow probing by polarization and phase shaping



Contribution only by χ^{r}_{yyxx}



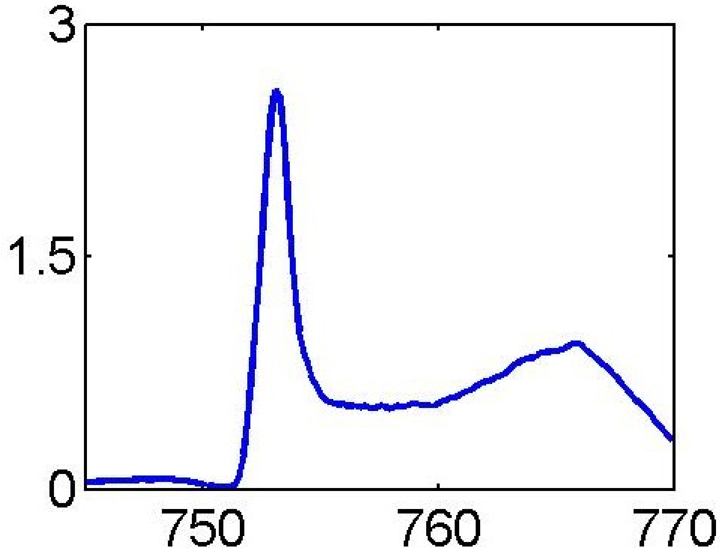


CARS spectrum - Narrow probing

Measured CARS spectrum from iodomethane (523cm^{-1})

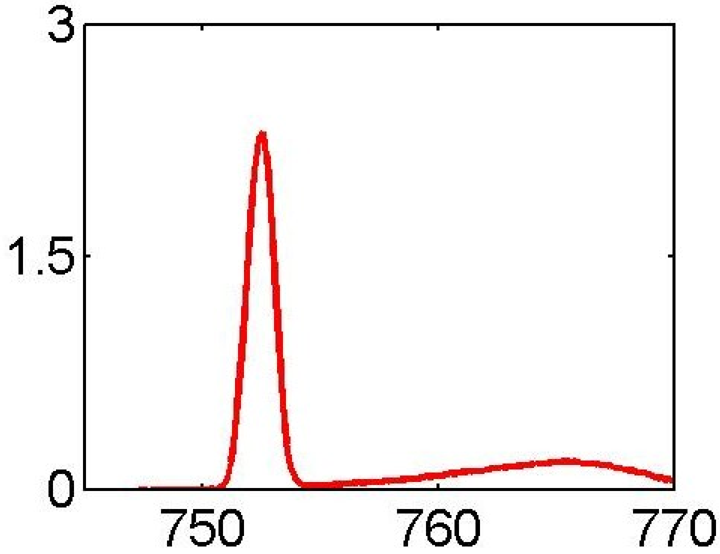
1.2nm (20cm^{-1}) wide y polarized probe

Polarization only



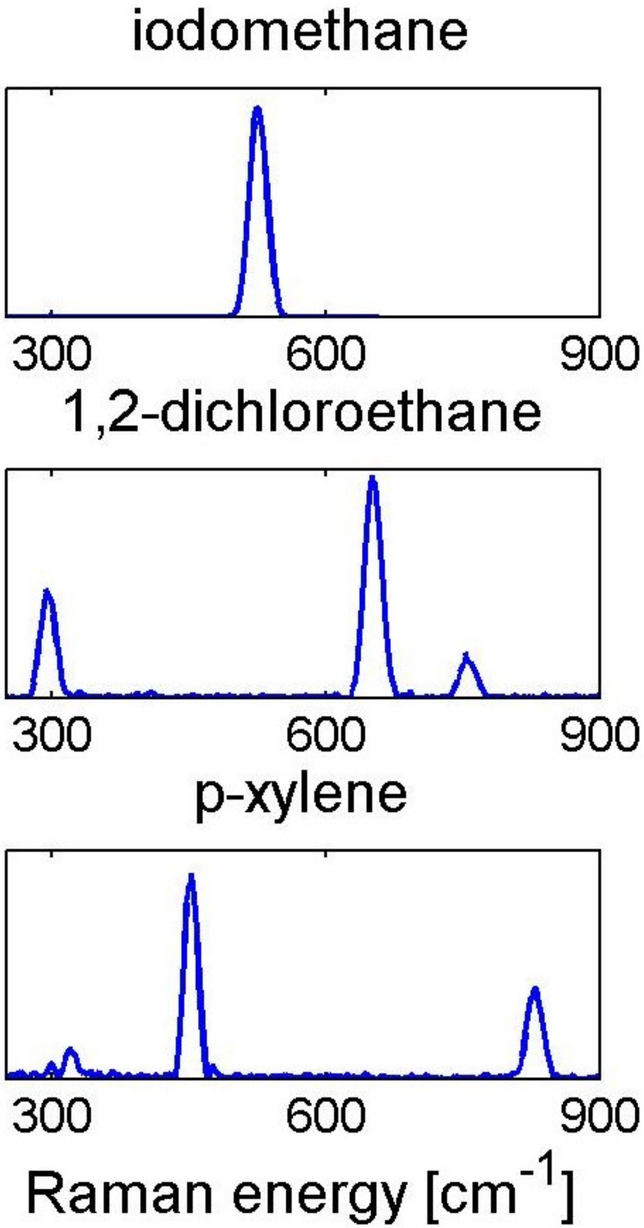
Polarization and phase:

π step at the center of the probe



Multiplexed CARS spectra

Spectral resolution currently limited by SLM pixellization



Thanks…

Coherent Control:

Doron Meshulach
Nirit Dudovich
Dan Oron
Thomas Polack
Evgeny Frumker
Adi Natan
Haim Suchowski
Barry Bruner
V Prabhudesai

Nonclassical Light:

Barak Dayan
Avi Pe'er
Itay Afek
Yaron Bromberg

Microscopy:

Dvir Yelin
Eran Tal
Ori Katz

Solitons:

Hagai Eisenberg
Yaniv Barad
Roberto Morandotti
Daniel Mandelik
Asaf Avidan
Yoav Lahini

Next :
Can you shape a single photon?

www.weizmann.ac.il/~feyaron

We have seen…

- Precise control of multiphoton transitions
- Transform limited pulses are not necessarily optimal
- Single-photon absorption is not necessarily boring
- CARS with single source via coherent control