All-optical processing in coherent nonlinear spectroscopy

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Physics of Complex Systems

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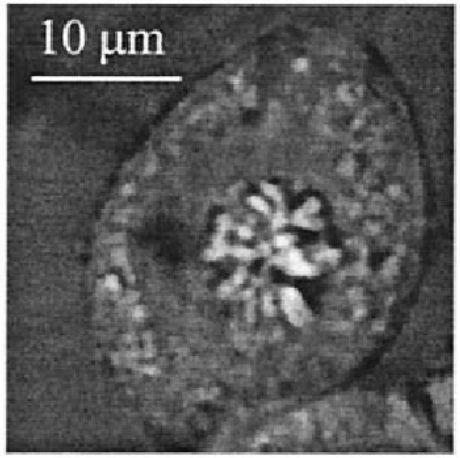
Outline

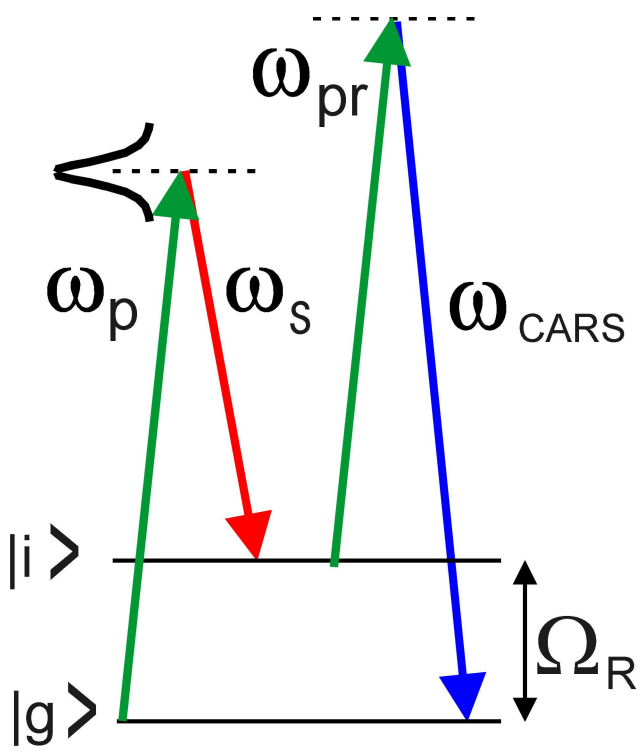
- CARS and multiphoton microscopy
- Single-pulse multiplexed CARS by coherent control
- All-optical processing of CARS spectra
- Single-pulse vibrational wavepacket generation and characterization
- Summary

Coherent Anti-Stokes Raman Scattering (CARS)

- Offers spectroscopic capability
- Typically $\omega_p=\omega_{pr}$,signal at $2\omega_p-\omega_s$ ($>\omega_p$)
- Requires two synchronized sources
- Spectral resolution limited by pulse bandwidth
- Lifetimes in picosecond range

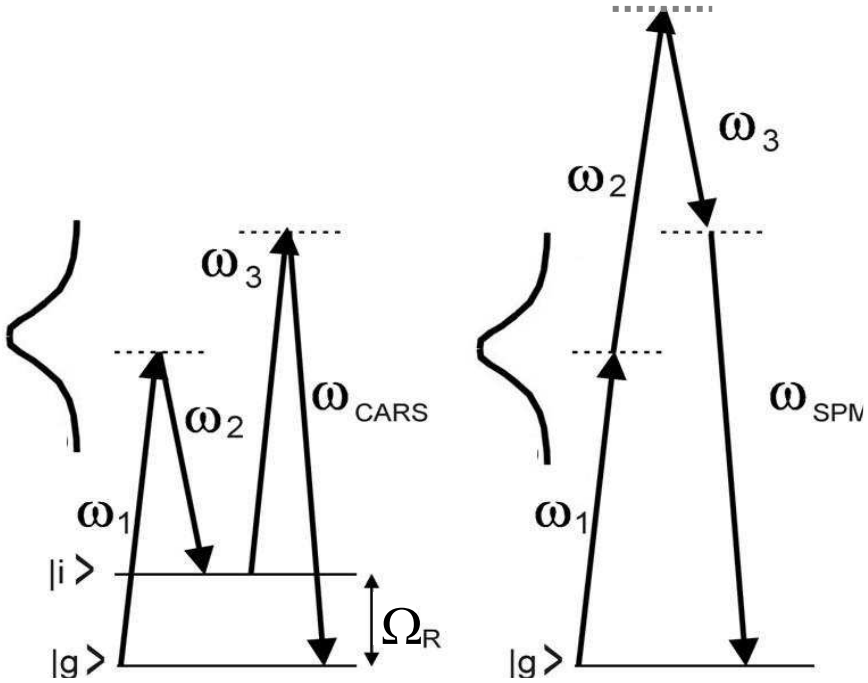
DNA backbone vibration ($1090\ cm^{-1}$) in mitosis





Cheng *et al*., Biophys. J. **83**, 502 (2002)
Volkmer *et al*., Phys. Rev. Lett. **87**, 23901 (2001)

Resonant vs. Nonresonant CARS

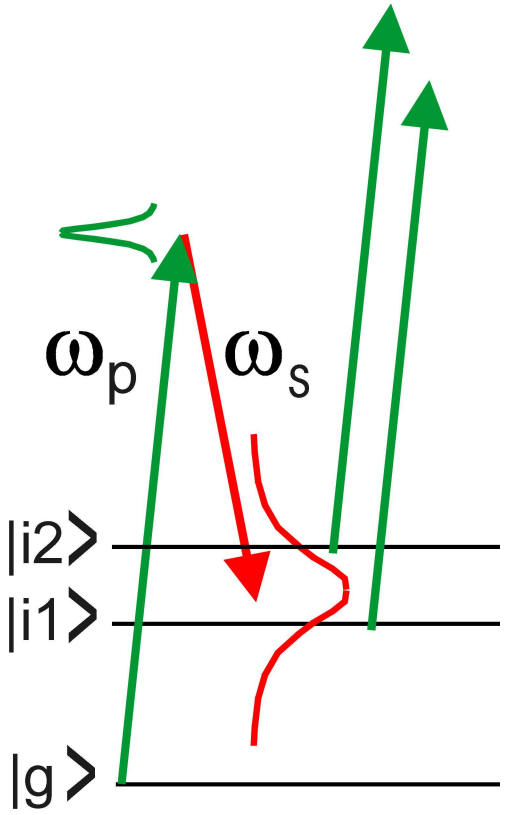


Resonant CARS is always accompanied by a nonresonant background

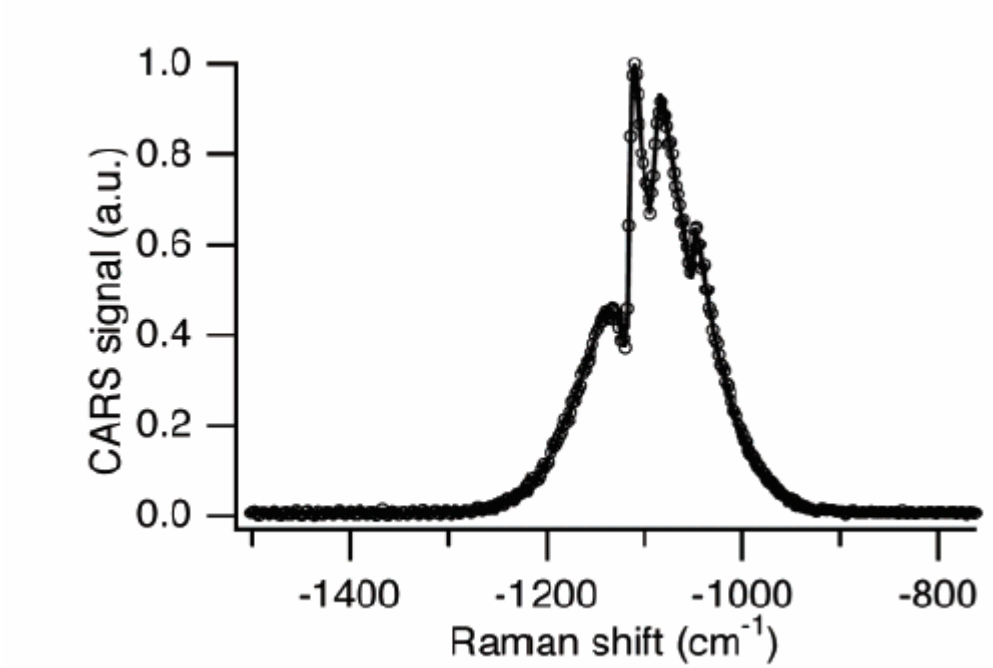
Nonresonant background is maximal for transform limited pulses (highest peak intensity)

Usually dealt with by using longer pulses and by polarization techniques

Multiplexed CARS



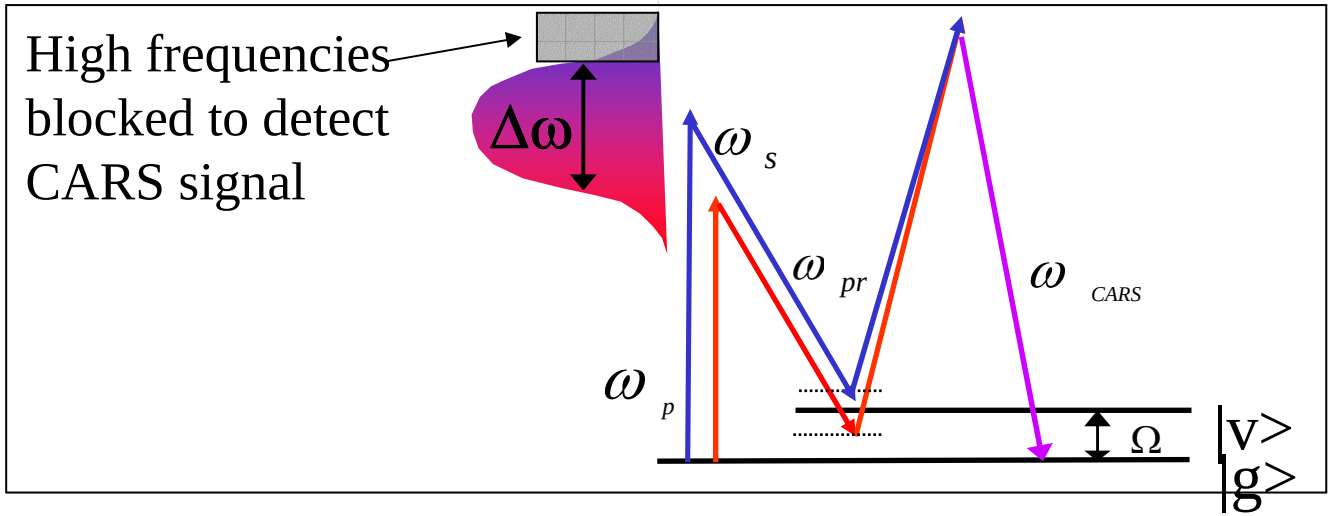
Measured signal is heterodyned with the nonresonant background (but $\pi/2$ phase at peak)



From Muller *et al.*, JPC B **106**, 3715 (2002)

Single-Pulse CARS Spectroscopy

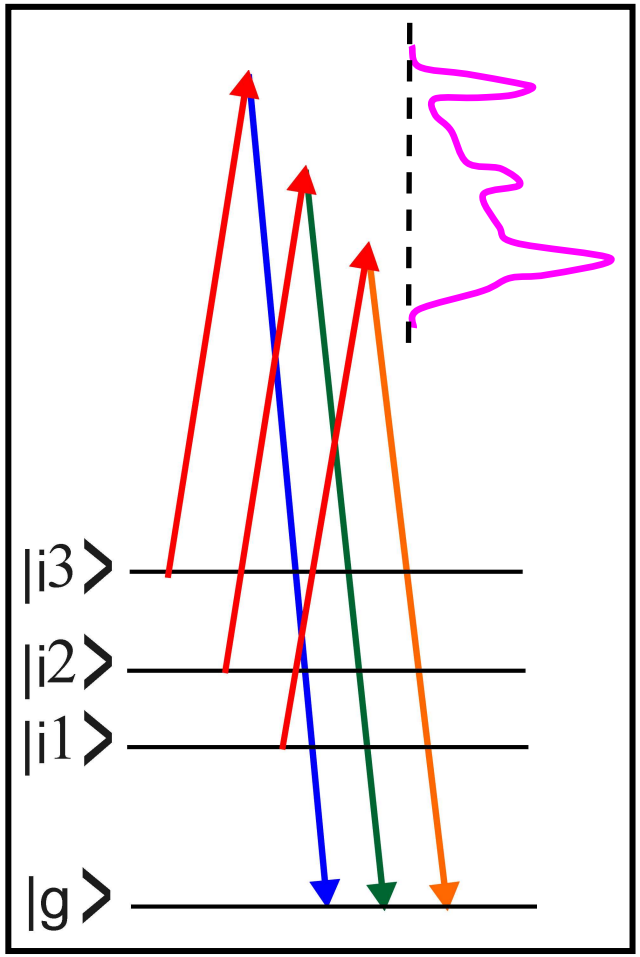
A single ultrashort, broadband pulse (shorter than the vibrational period) to provide all 3 photons:



But -

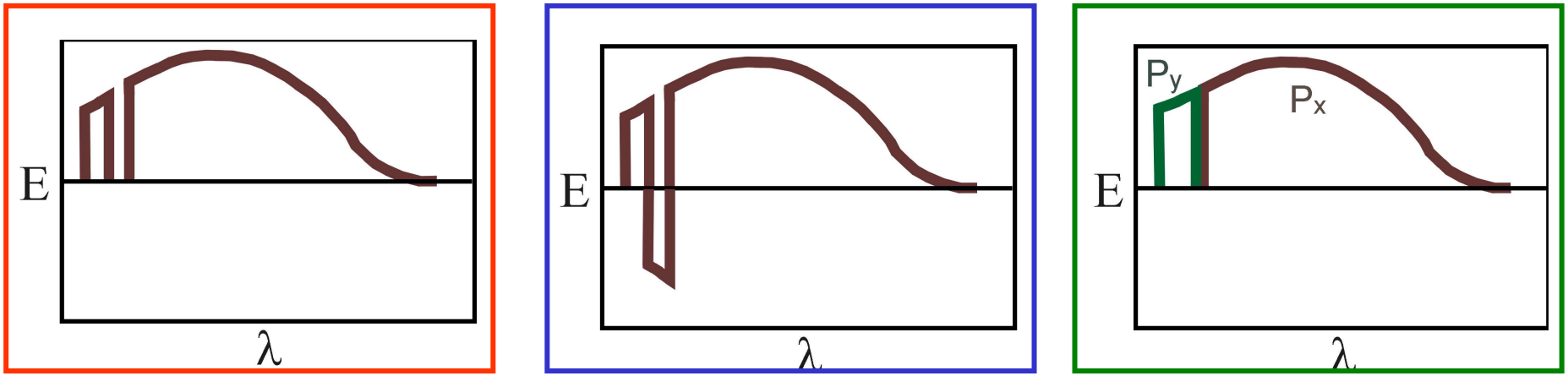
- Loss of resolution in broadband excitations
- Strong nonresonant background

Single-pulse multiplexed CARS



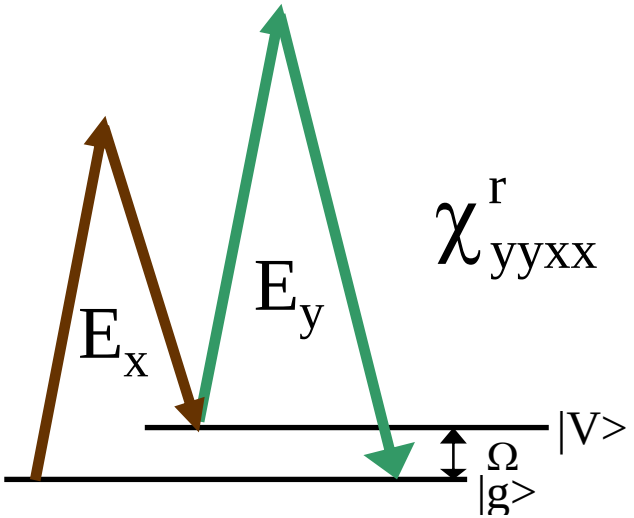
Simple schemes for separating a spectrally narrow probe within a broadband pulse

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

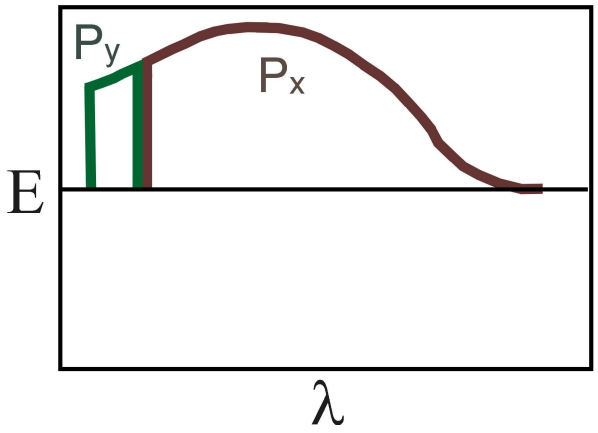


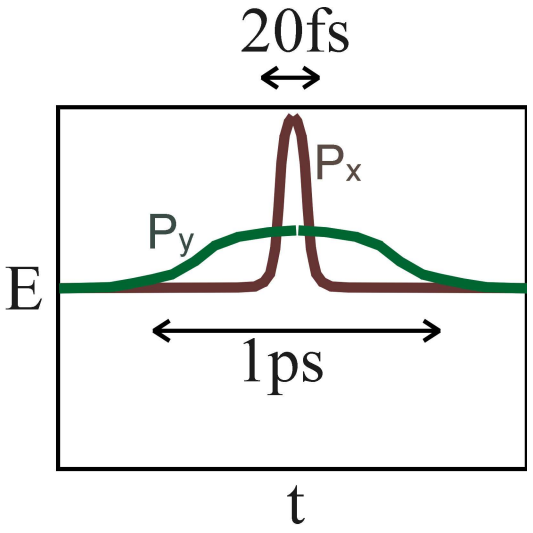
Oron *et al*., Phys. Rev. Lett. **89**, 273001 (2002)
Oron *et al*., Phys. Rev. Lett. **90**, 213902 (2003)

Narrow probing by an orthogonal polarization (naively)



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

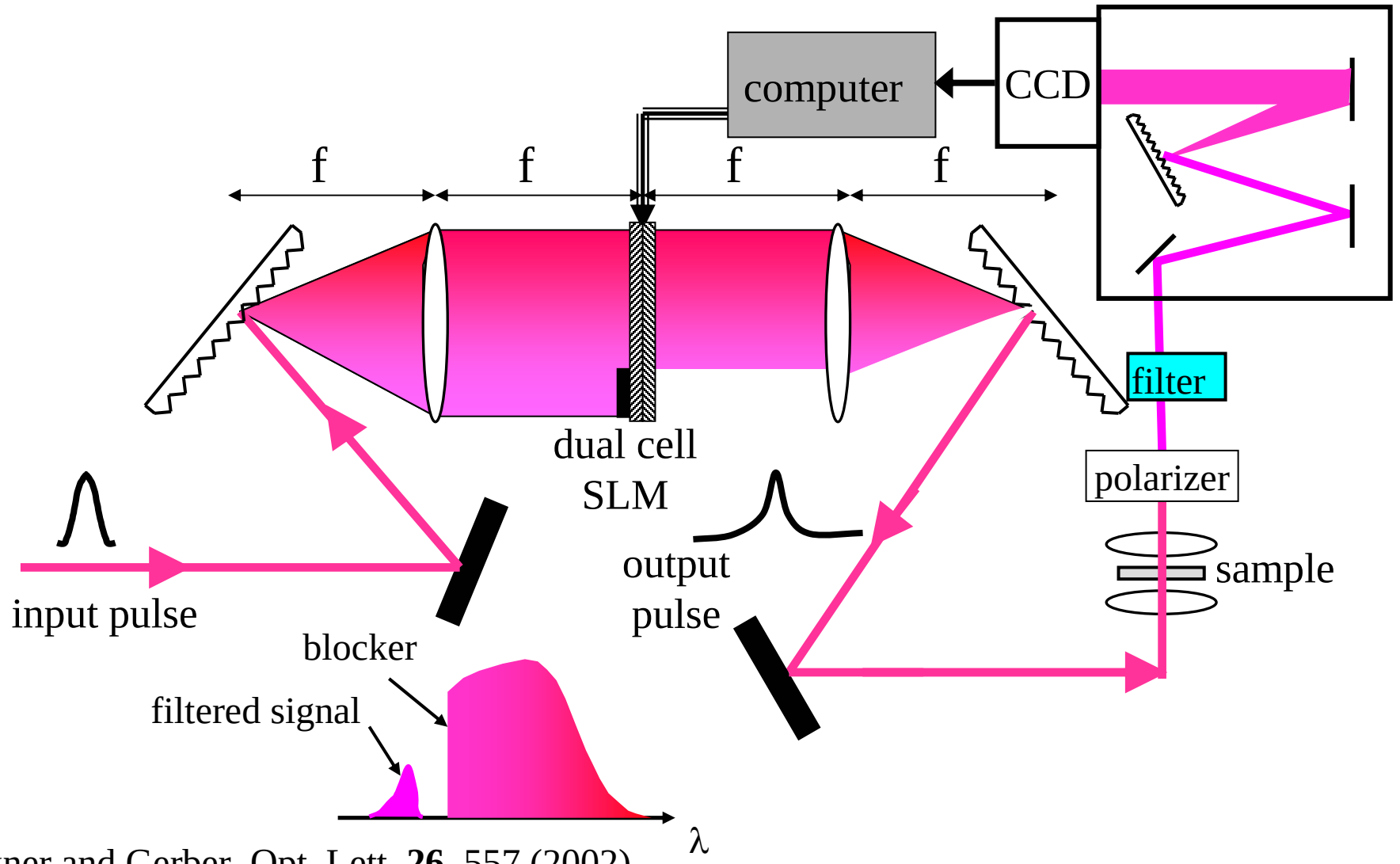




Oron *et al*., Phys. Rev. Lett. **90**, 213902 (2003)

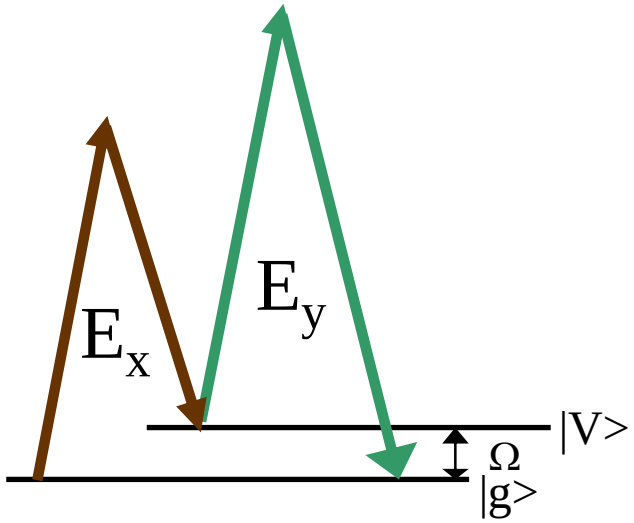
Experimental set-up

Polarization and phase shaping

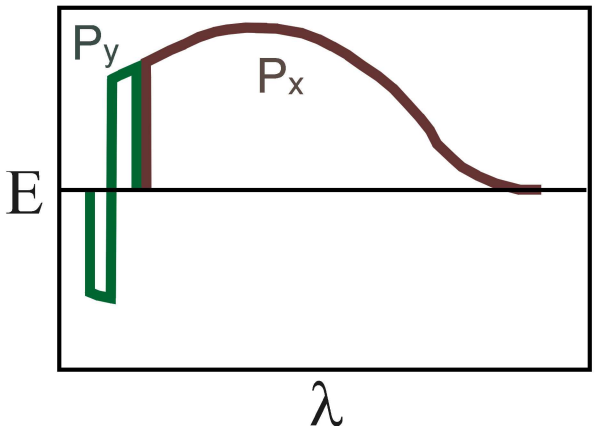


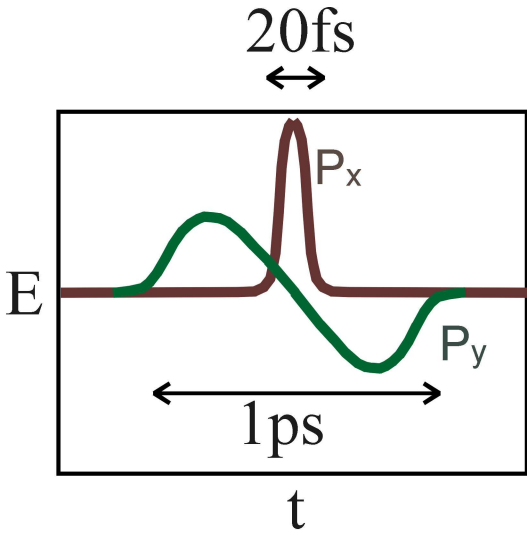
Brixner and Gerber, Opt. Lett. **26**, 557 (2002)

Narrow probing by polarization and phase shaping



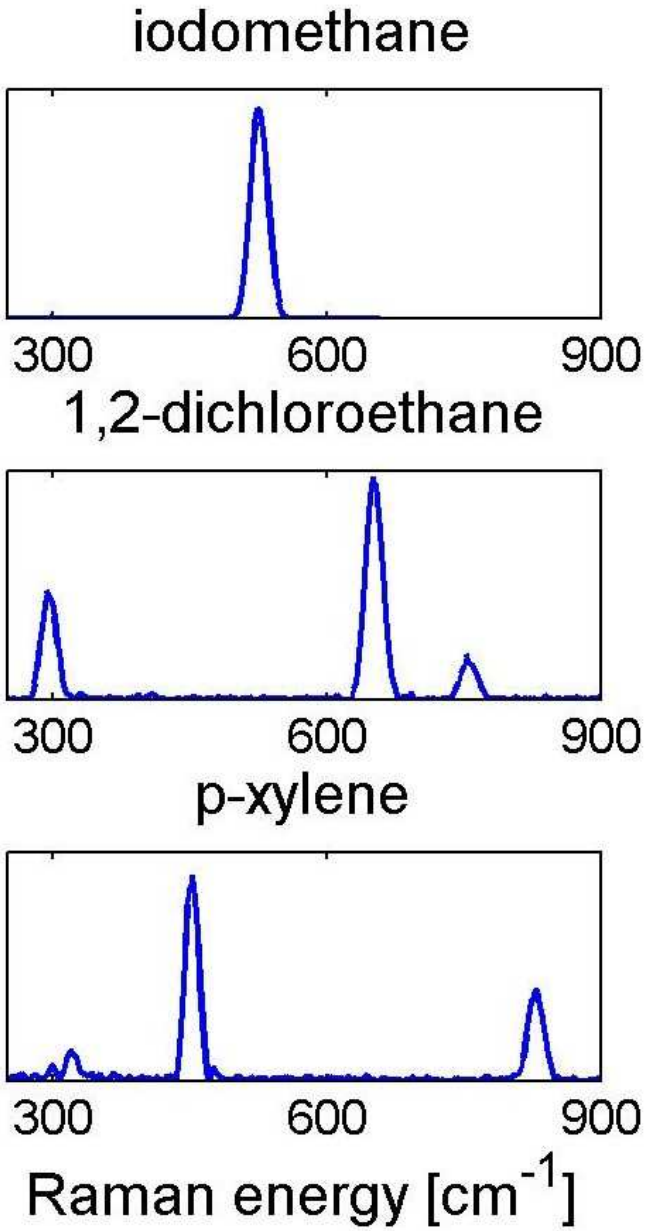
Contribution only by χ^{r}_{yyxx}



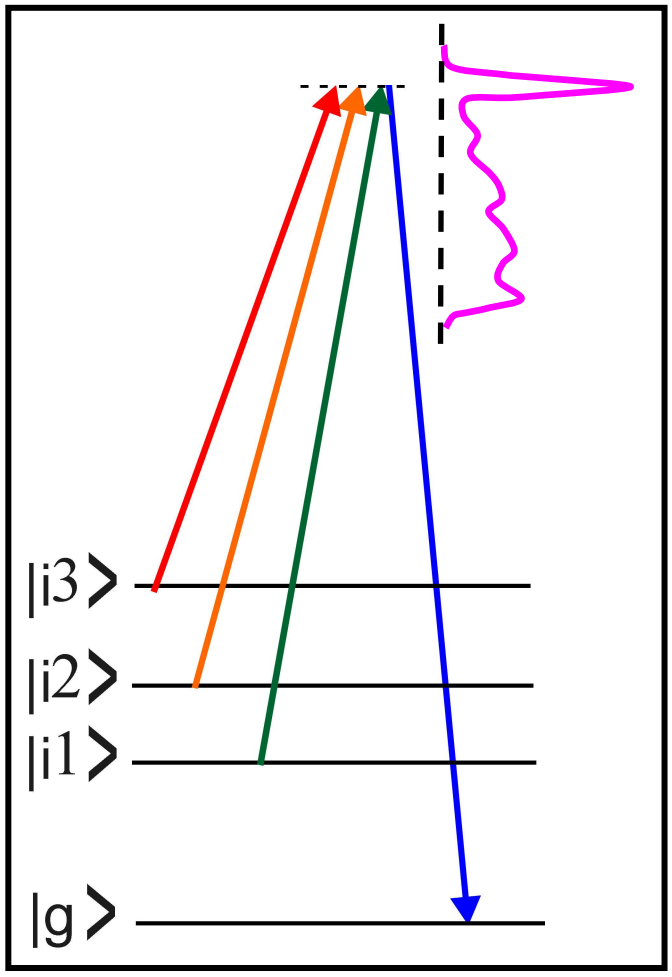


Multiplexed CARS spectra

Spectral resolution currently limited by SLM pixellization

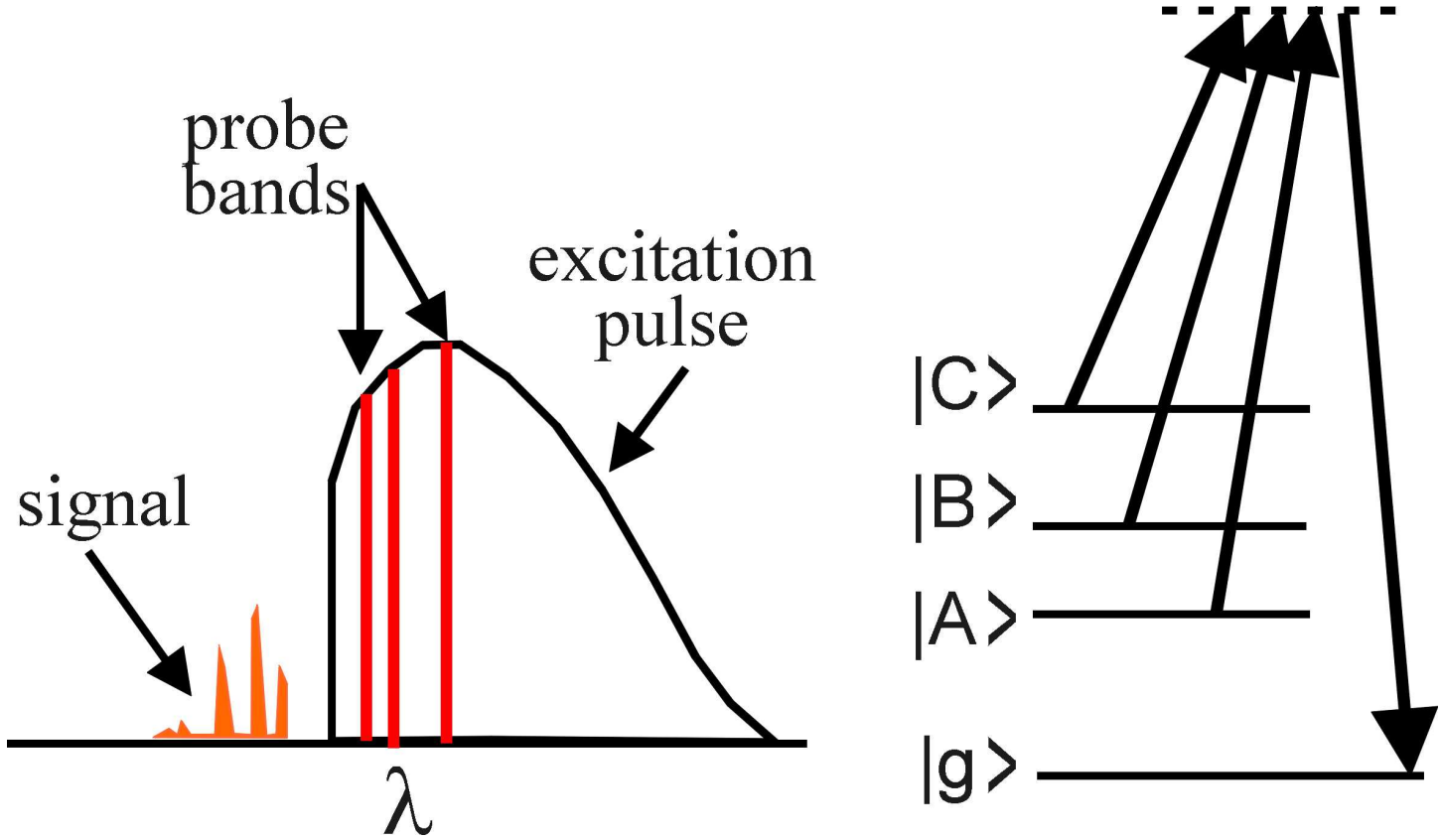


All optical processing



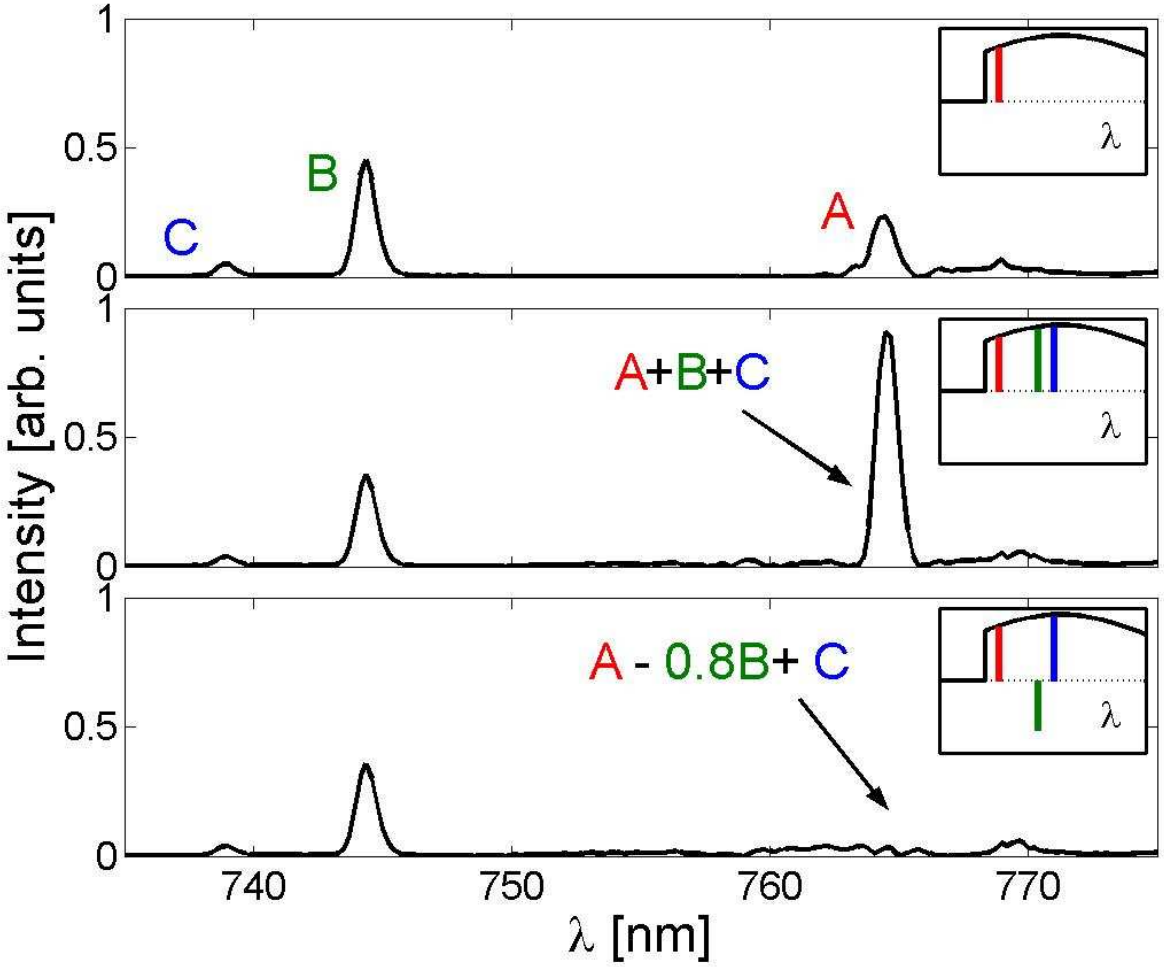
All-optical analysis of Raman spectra

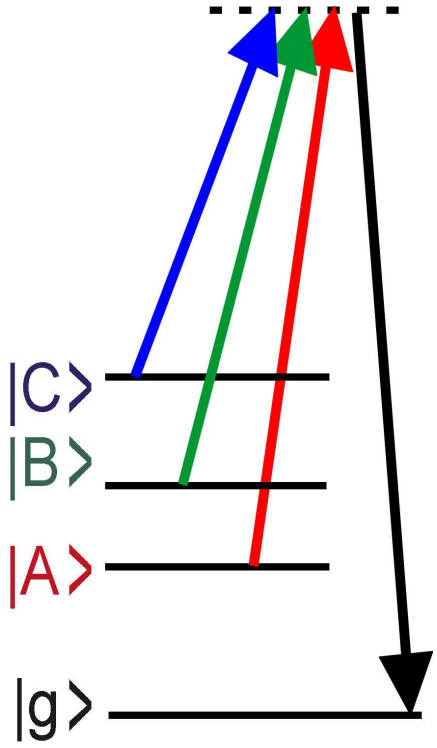
Use broadband probing to induce interferences between contributions from several Raman levels



All-optical analysis of Raman spectra –

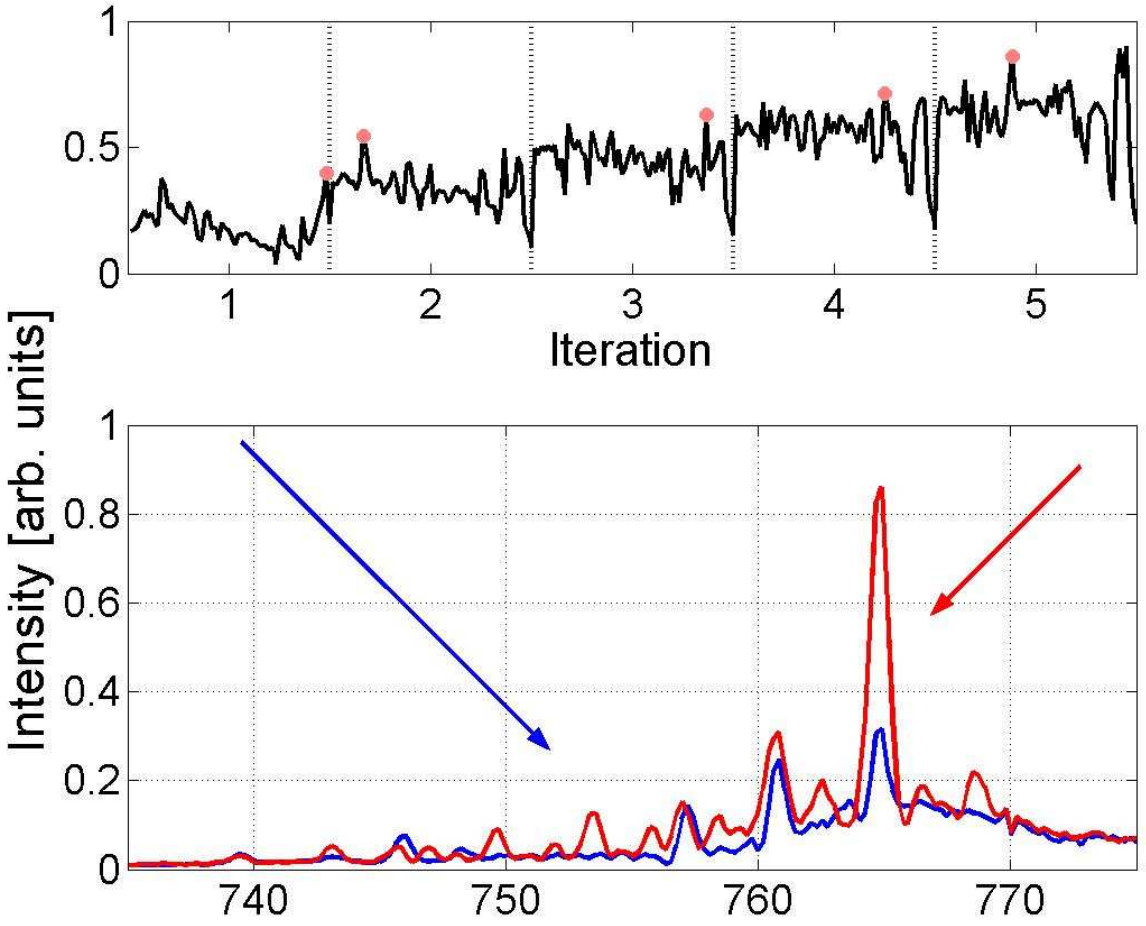
application to spectroscopy of 1,2 dichloroethane



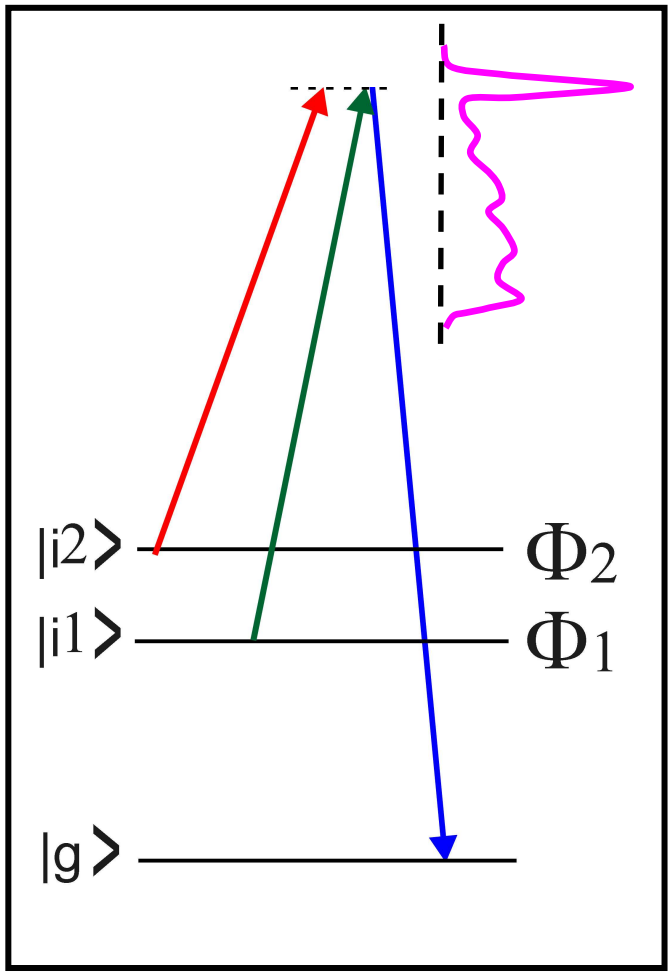


All-optical analysis of Raman spectra –

application of adaptive techniques for spectroscopy of 1,2 dichloropropane



Characterization of vibrational wavepackets

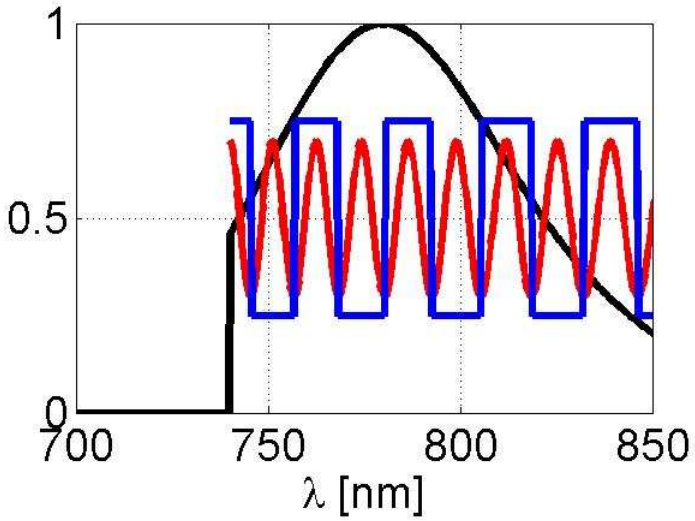


Characterization of vibrational wavepackets

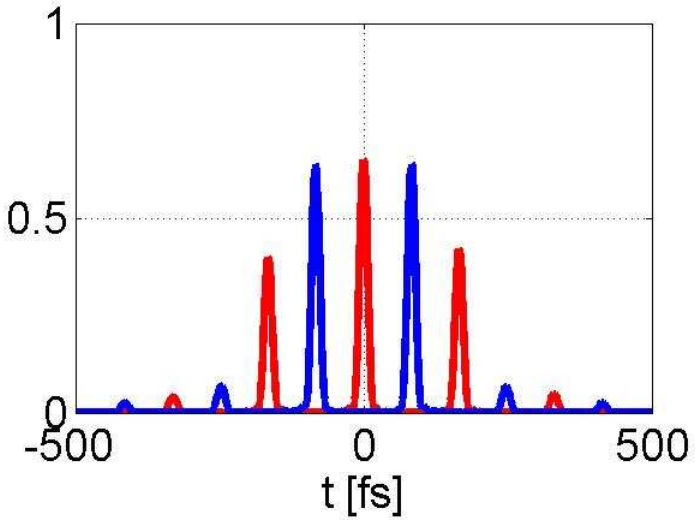
When relative phases of excited Raman bands are unknown, use interference in CARS spectrum to probe them

Toluene: 788, 1001 cm^{-1} bands excited in phase or out of phase

Spectral phase

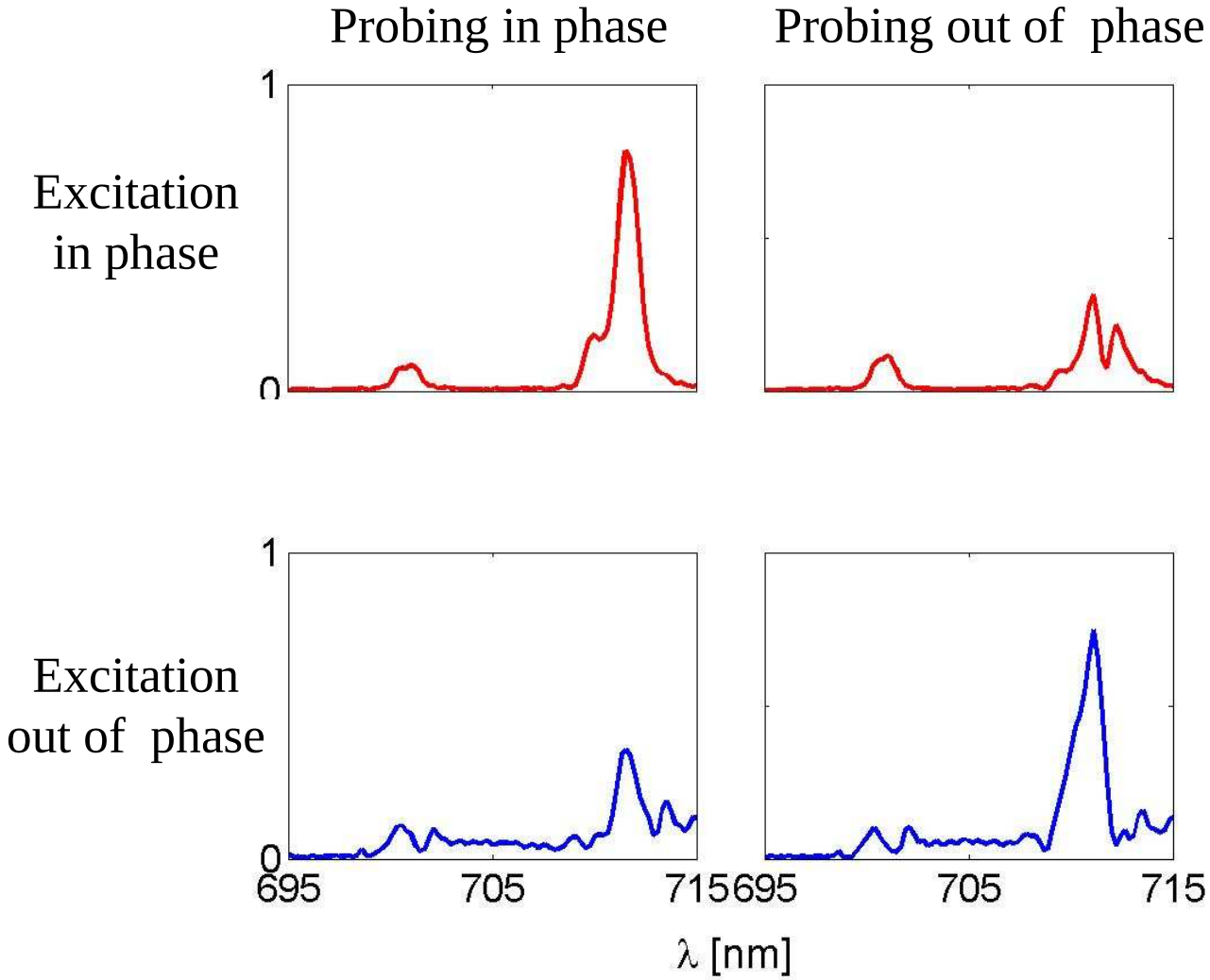


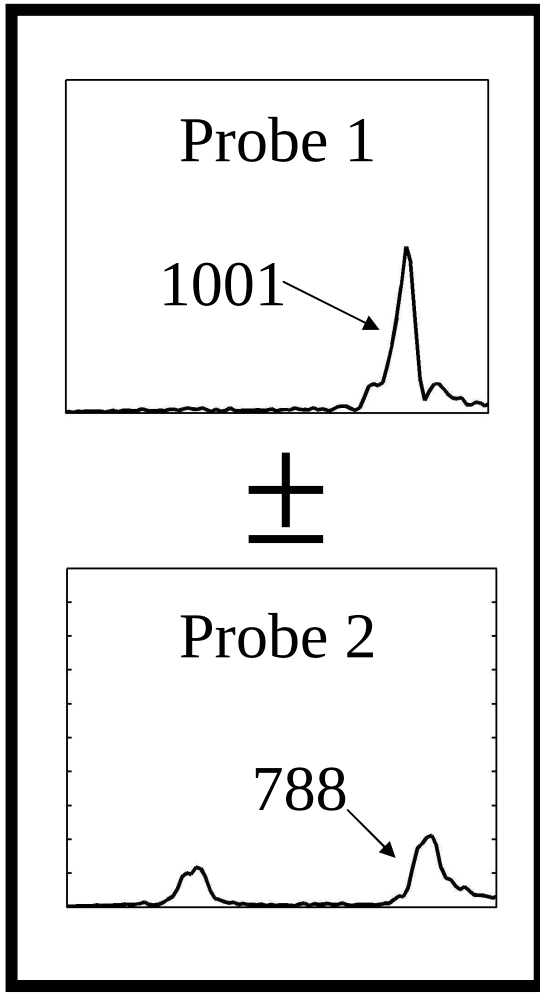
Temporal intensity



Characterization of vibrational wavepackets

Larger probe-free time window necessary achieved by more delicate phase shaping of probe pulse





Summary

- Single-pulse multiplex CARS provides the ability to perform simple interference-based optical analysis of observed vibrational spectra
- Broadband probing allows frequency domain characterization of vibrational wavepackets
- Possibility of single-pulse multidimensional vibrational spectroscopy
- Applicability to any multiphoton spectroscopy technique.

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