



FT-CARS with dual-comb spectroscopy - System proposal -

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Purpose

- ✓ Ultra high spectral resolution
- ✓ High energy mol. vib. ($> 1500 \text{ cm}^{-1}$)
realized by Fourier Transform-CARS

Conventional

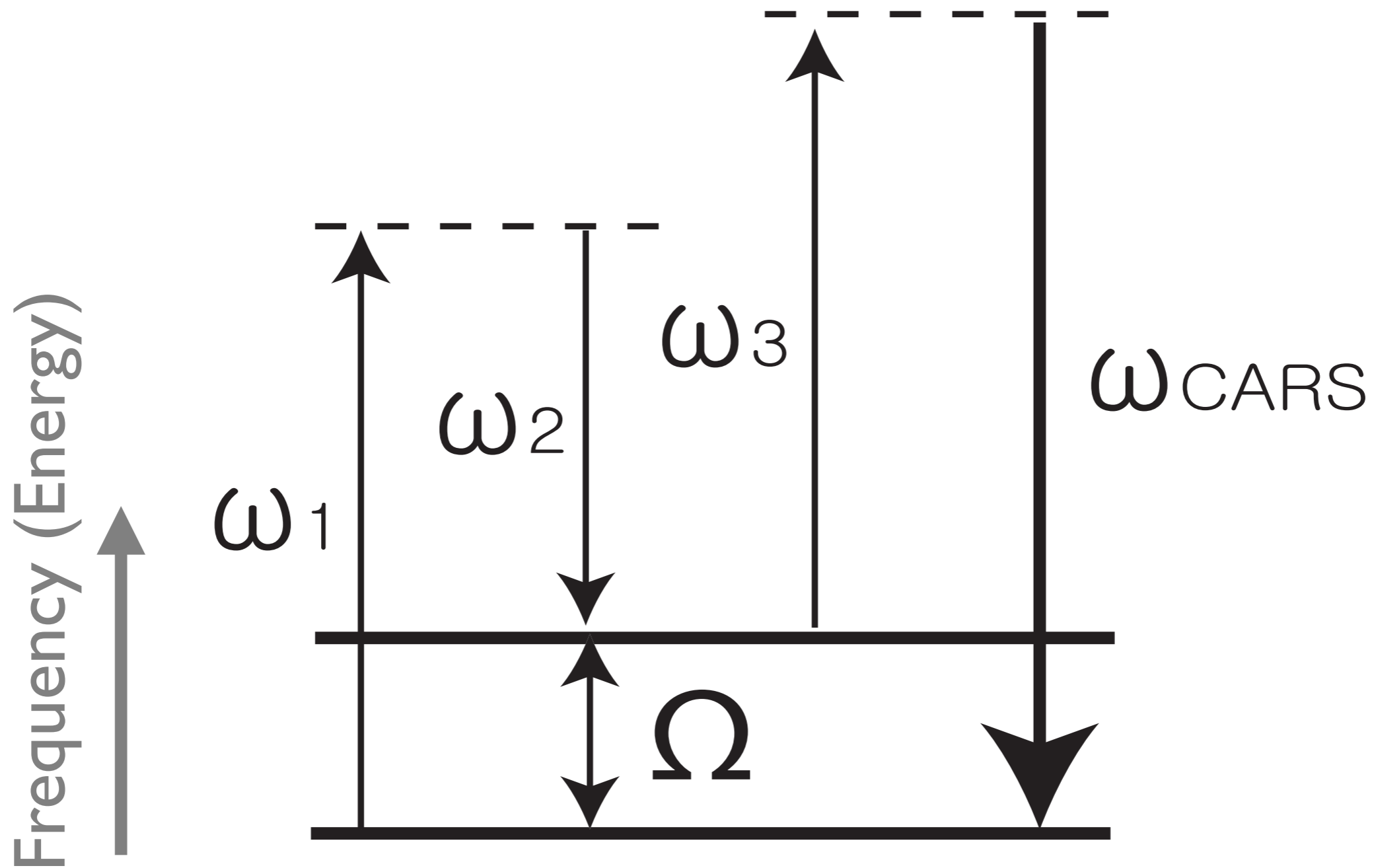
$> 3500 \text{ cm}^{-1}$ w/ $> 1 \text{ cm}^{-1}$ resolution

(depending on spectrometer)

$< 1500 \text{ cm}^{-1}$ w/ 10^{-7} cm^{-1} (expected)

(depending on pulse laser and dual-comb scheme)

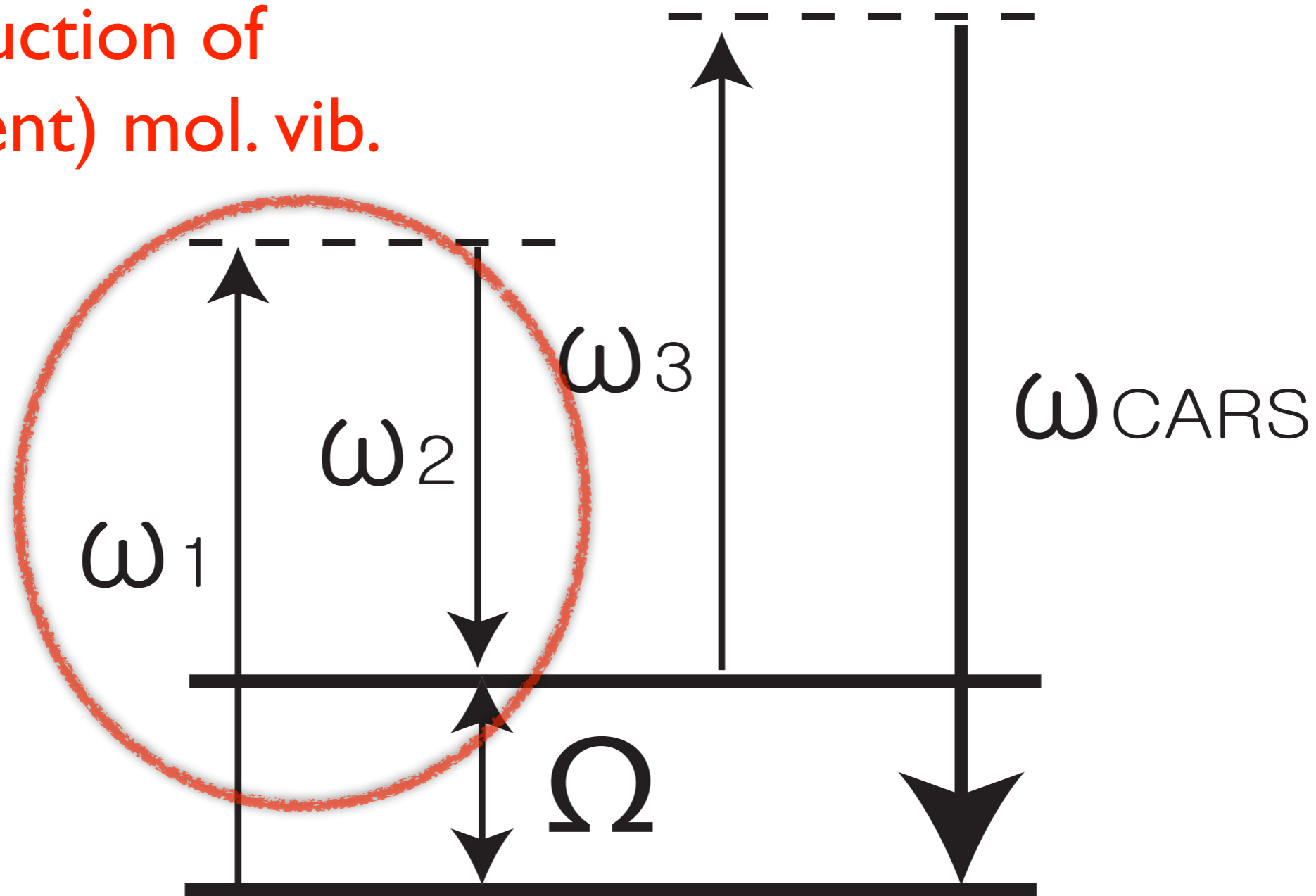
Principle



(a) Resonant process

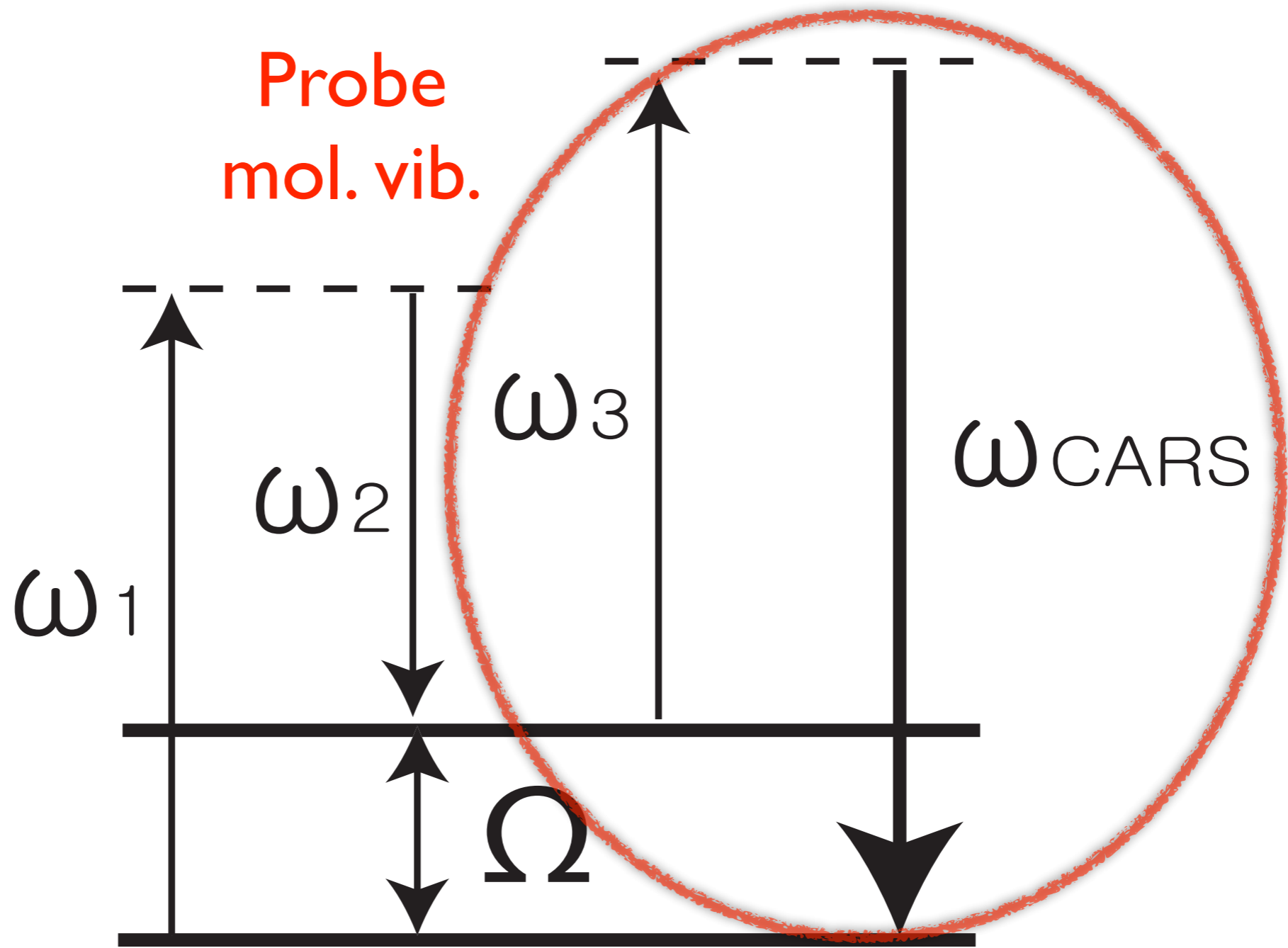
Principle

Induction of
(coherent) mol. vib.



(a) Resonant process

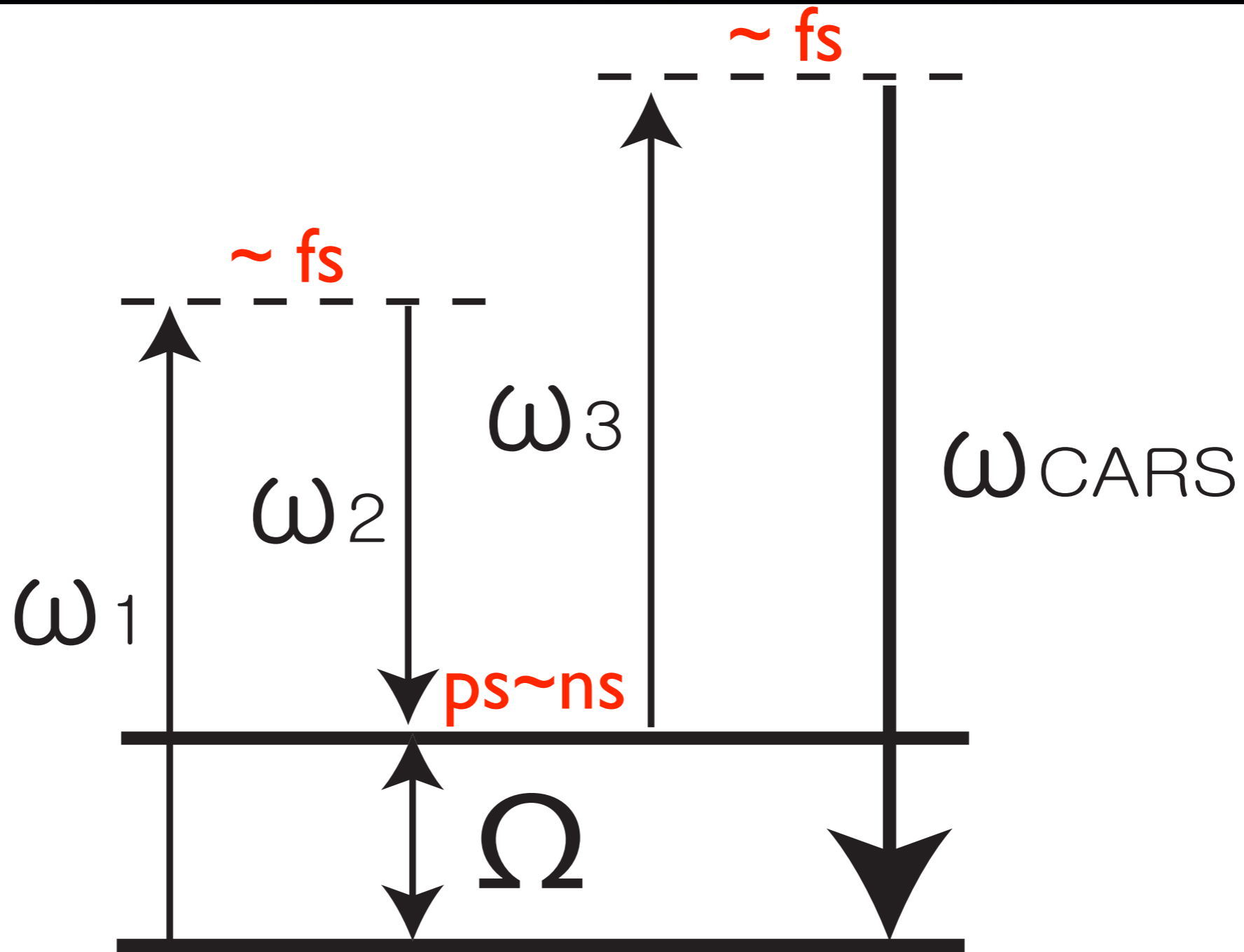
Principle



(a) Resonant process

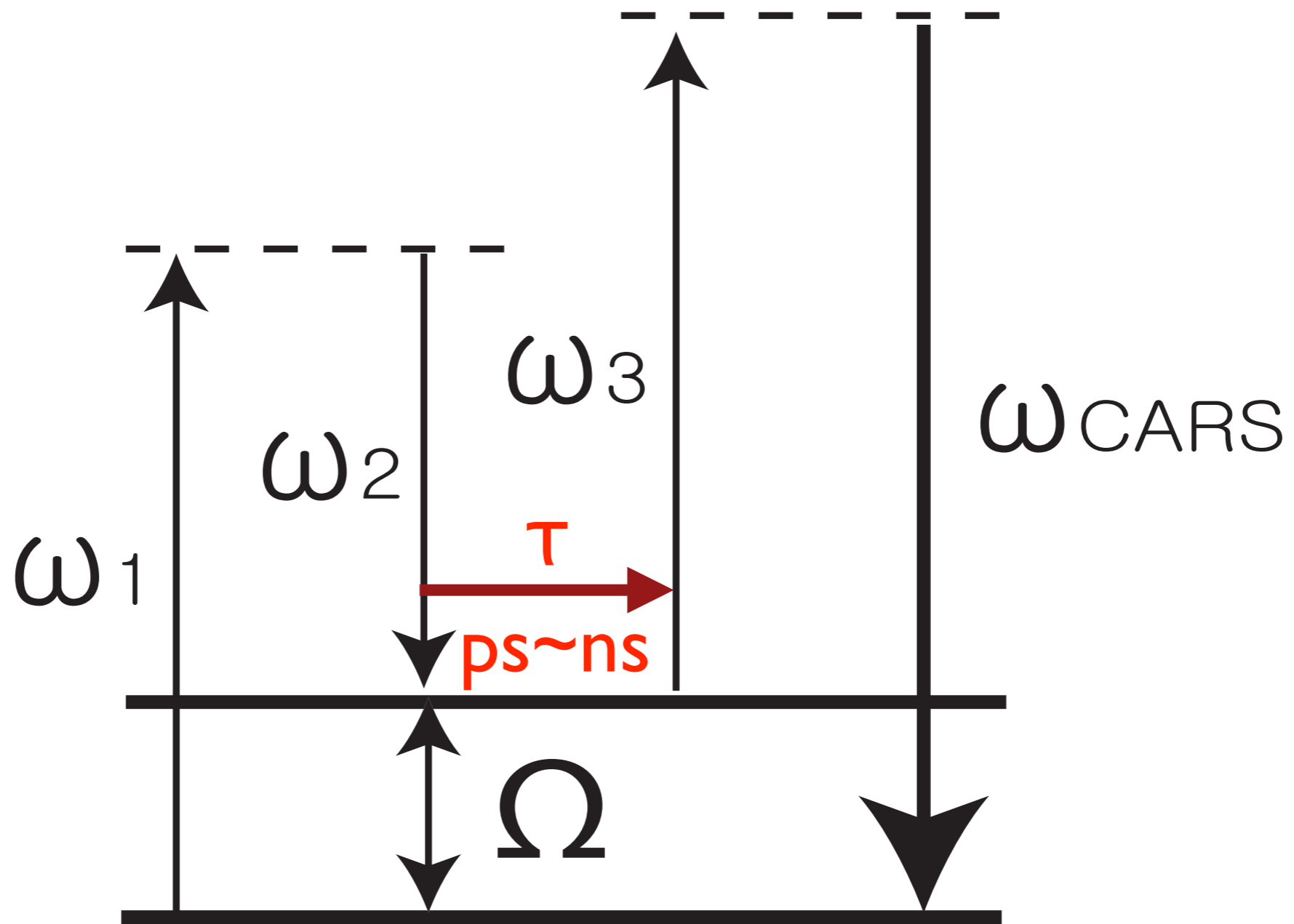
Principle

Lifetime



(a) Resonant process

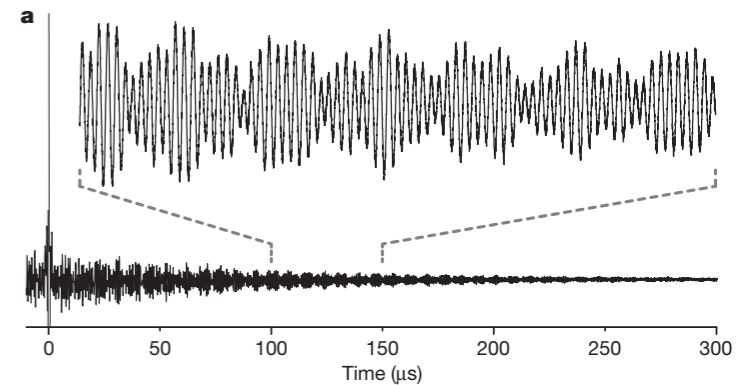
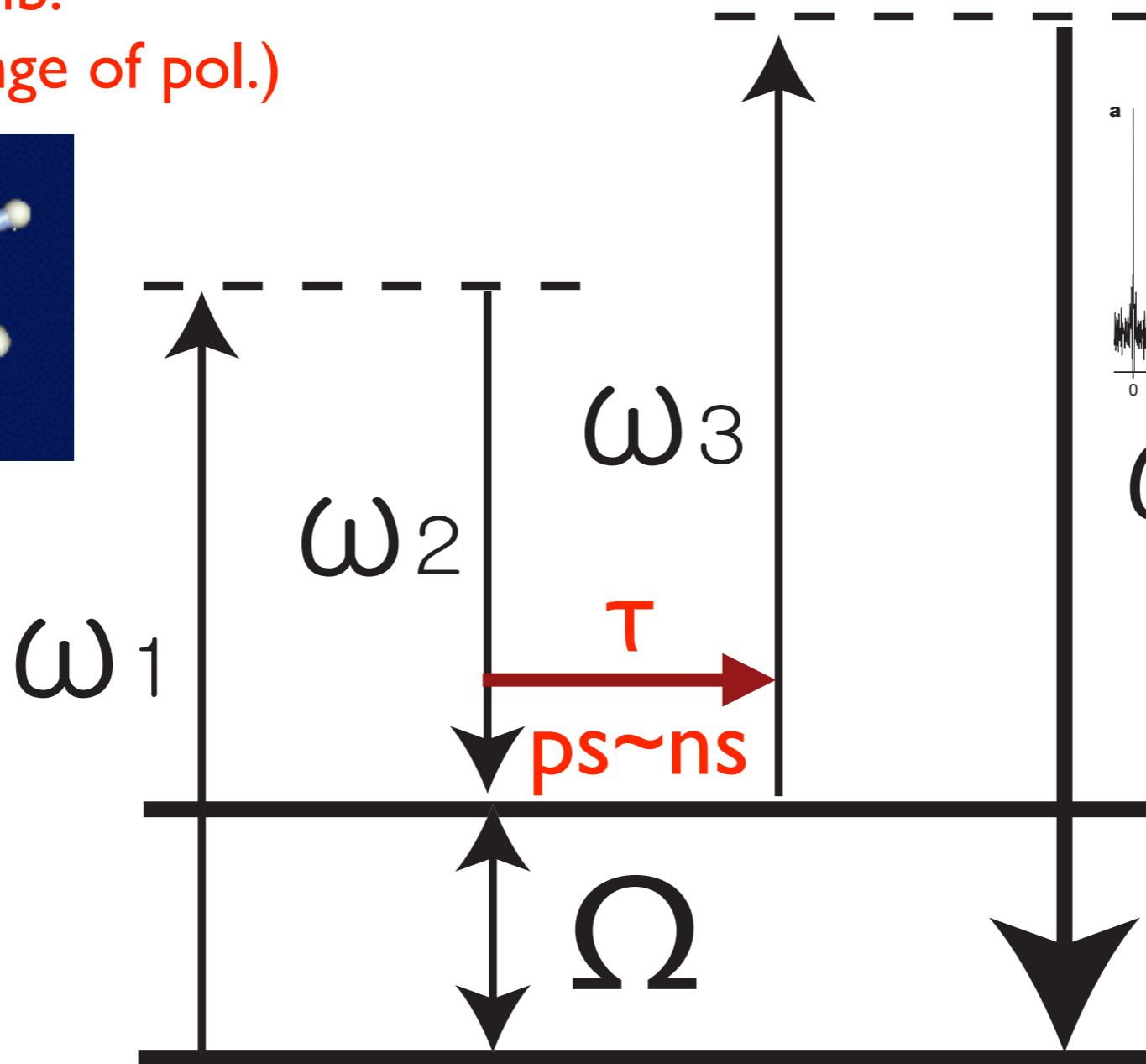
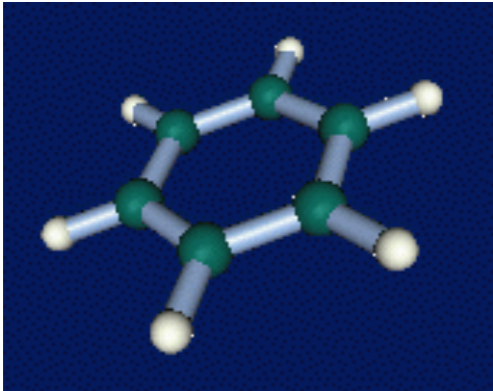
Principle



(a) Resonant process

Principle

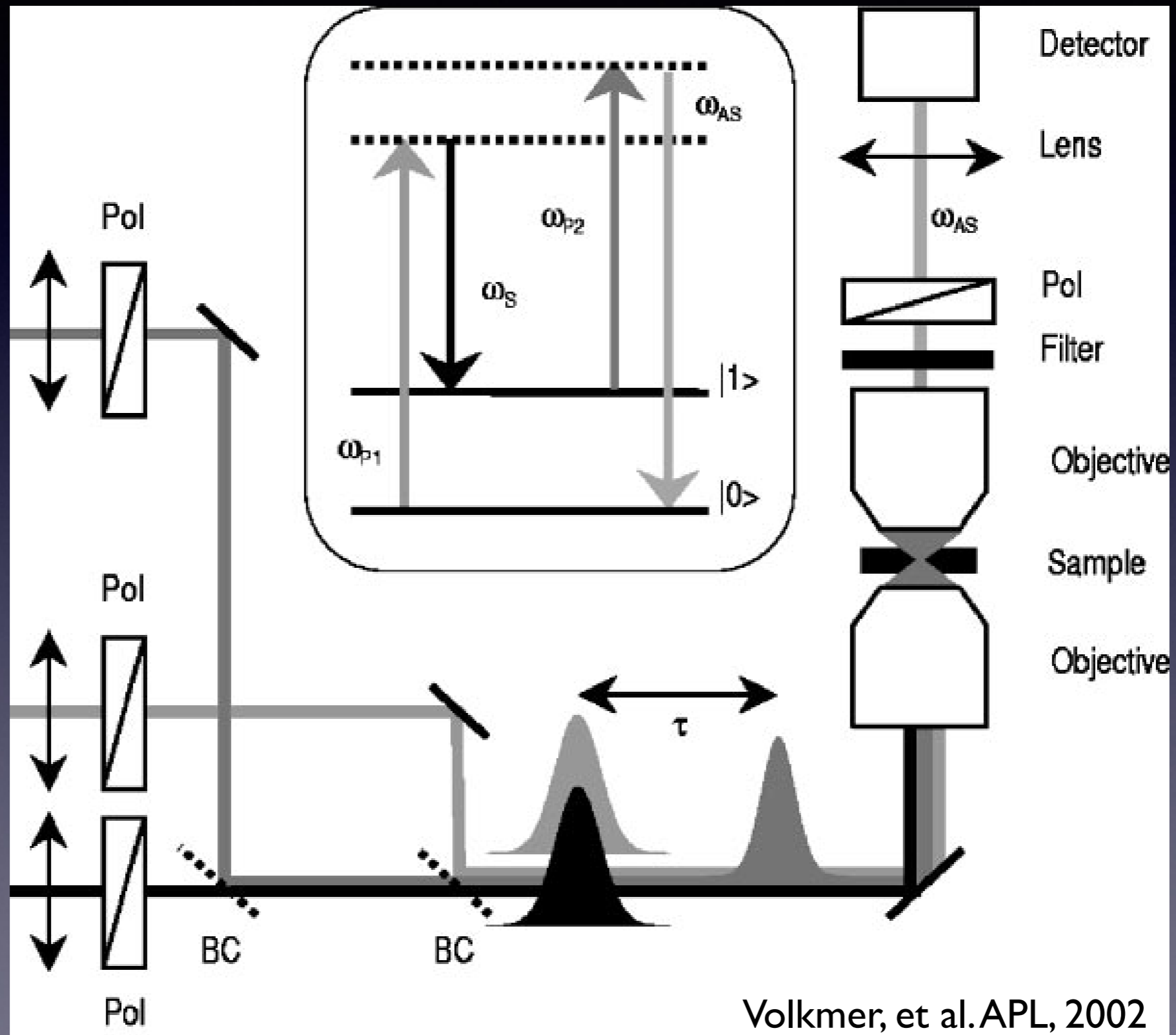
Mol.Vib.
(Periodic change of pol.)



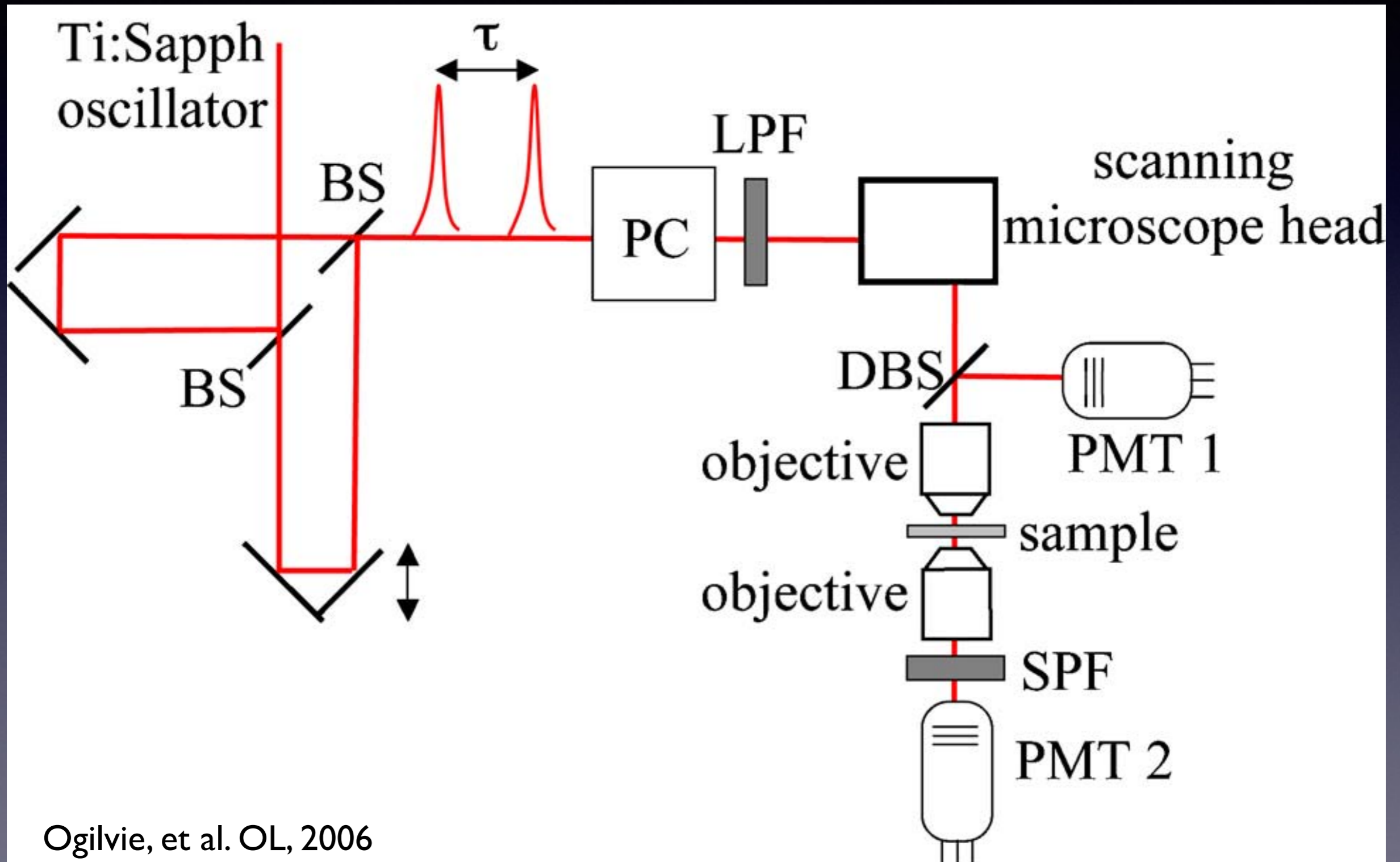
ω_{CARS}

(a) Resonant process

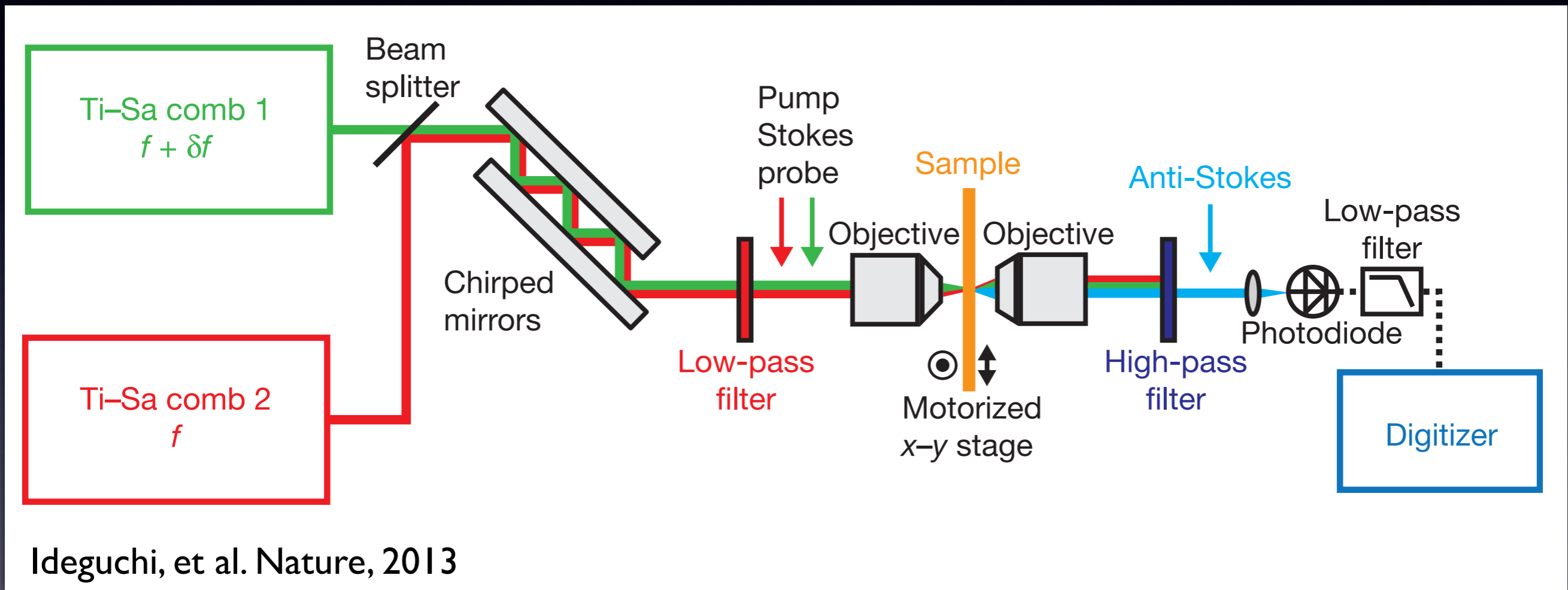
Setup (3 lasers)



Setup (1 laser)

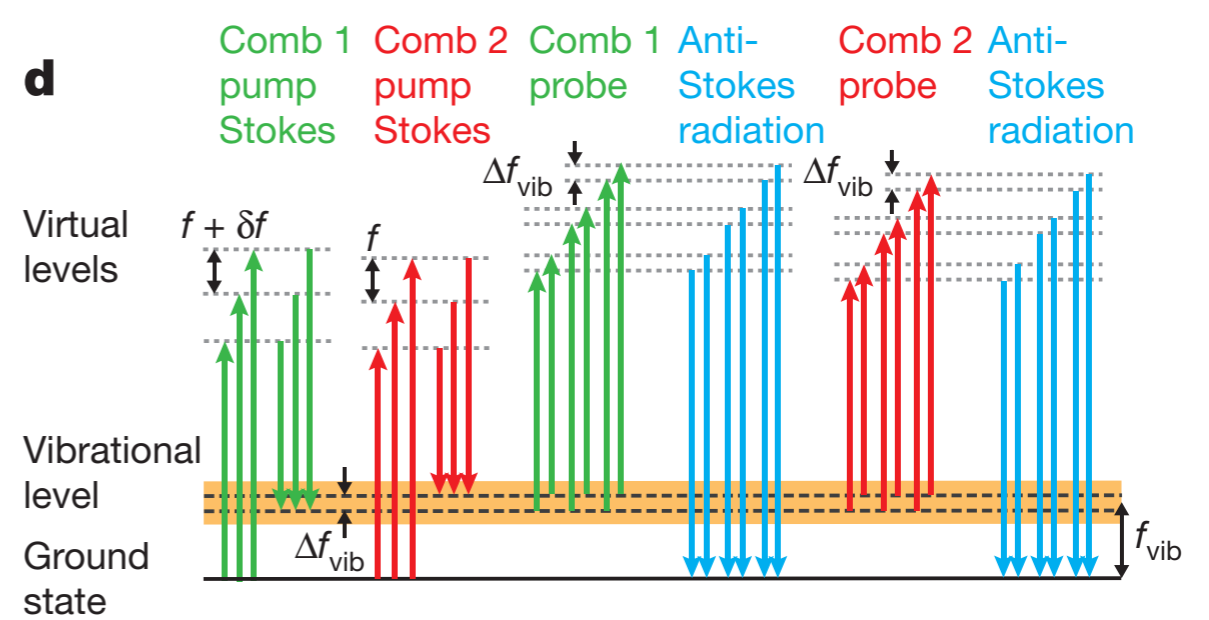
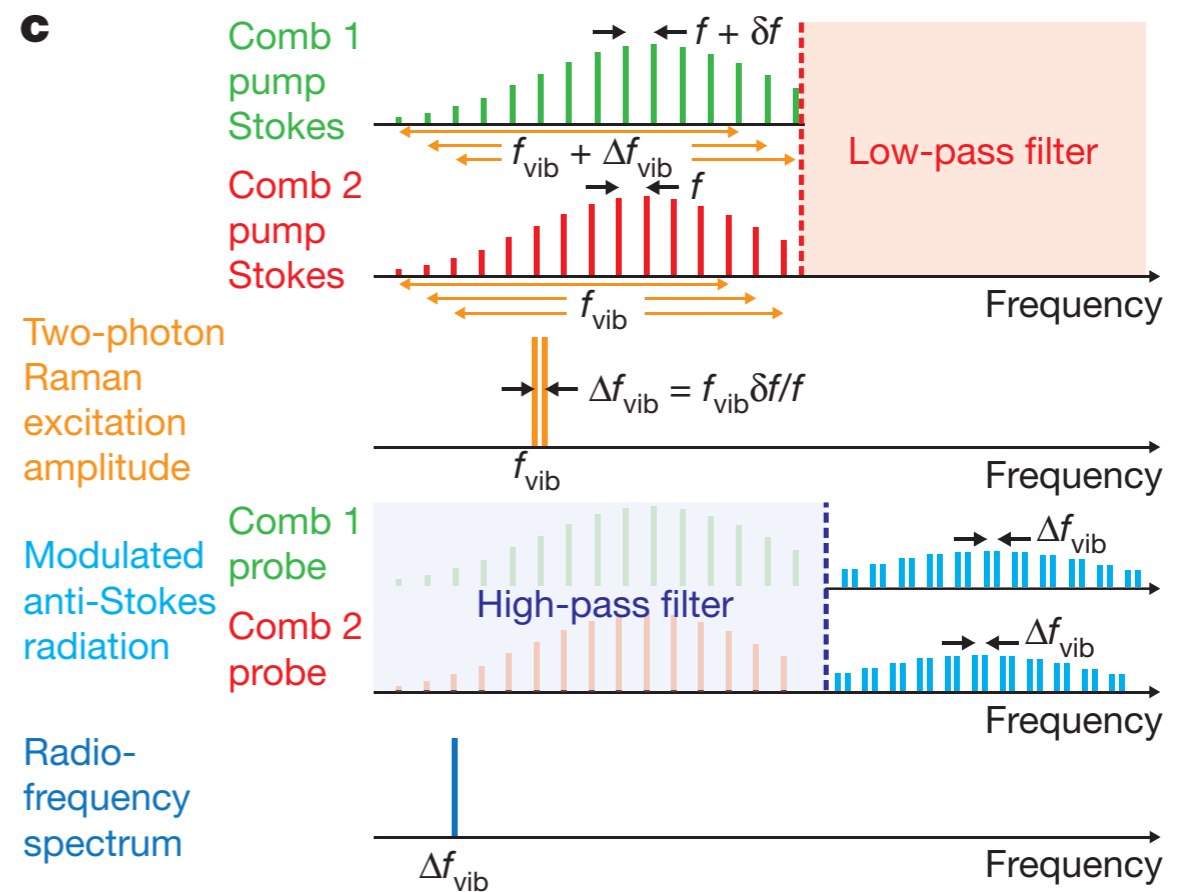
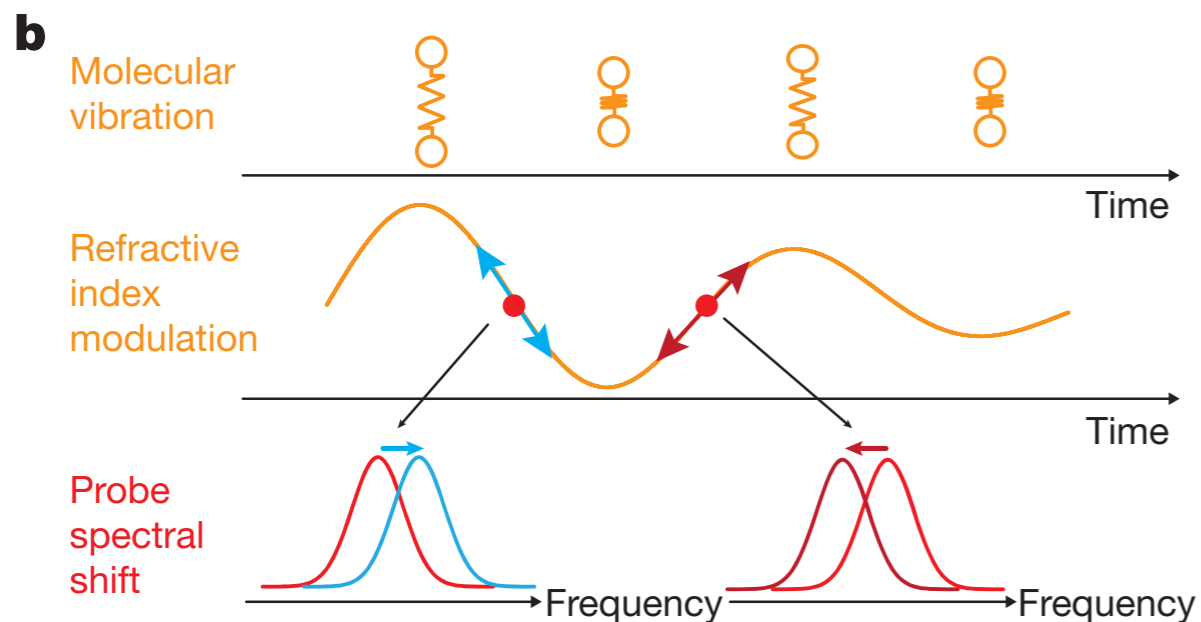
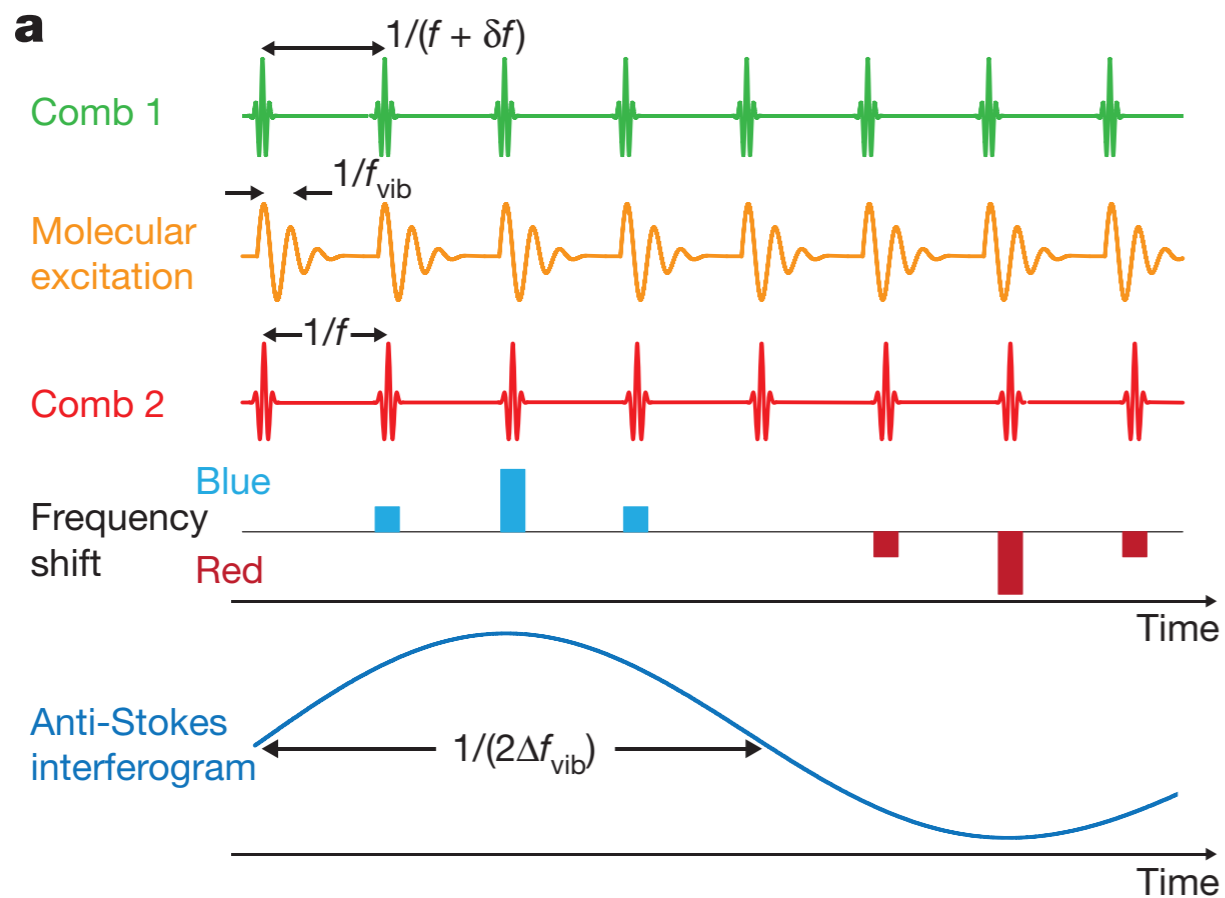


Setup (Dual-comb)



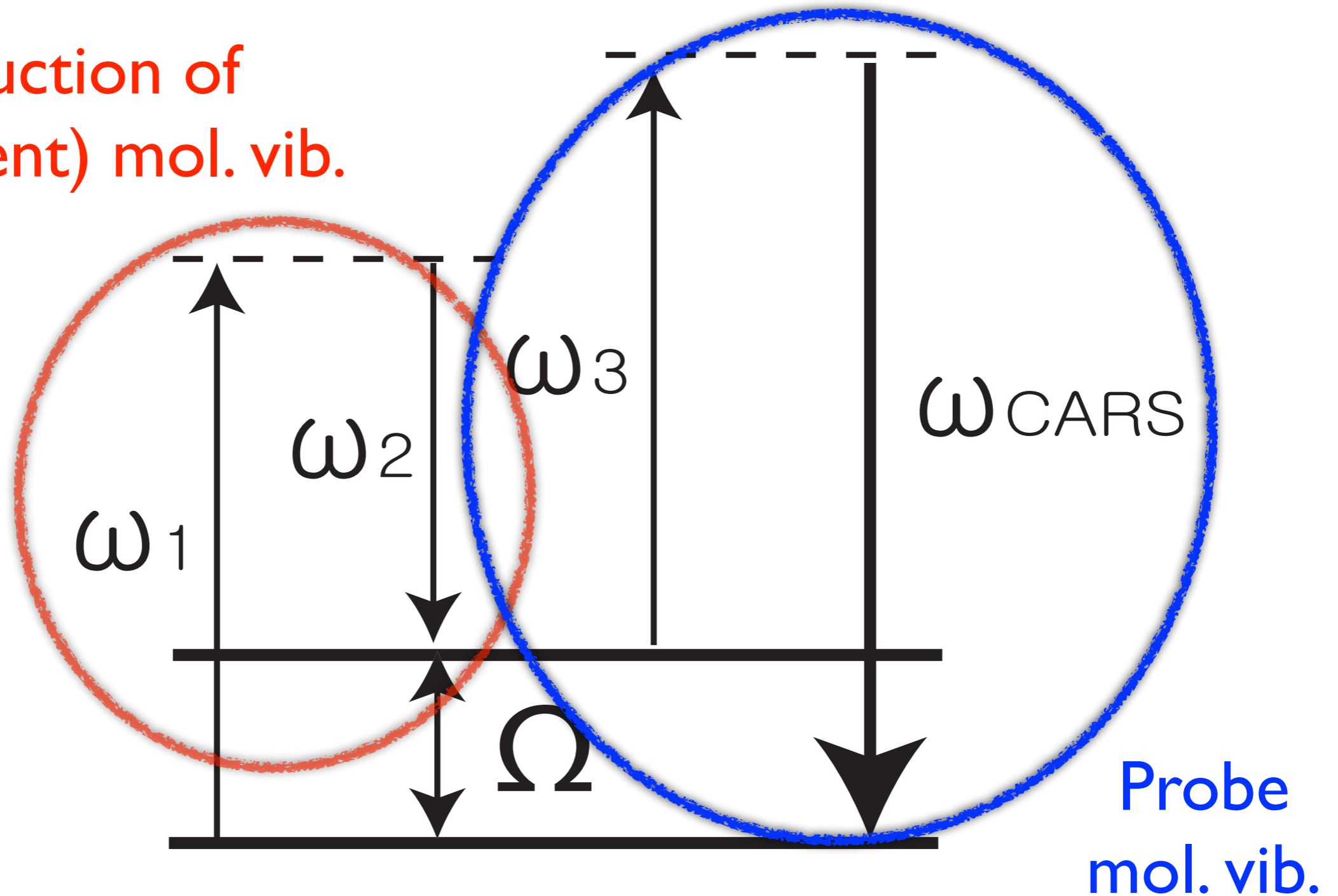
- ✓ Ultra high spectral resolution
- ✓ **Low** energy mol. vib. ($< 1500 \text{ cm}^{-1}$)

Setup (Dual-comb)



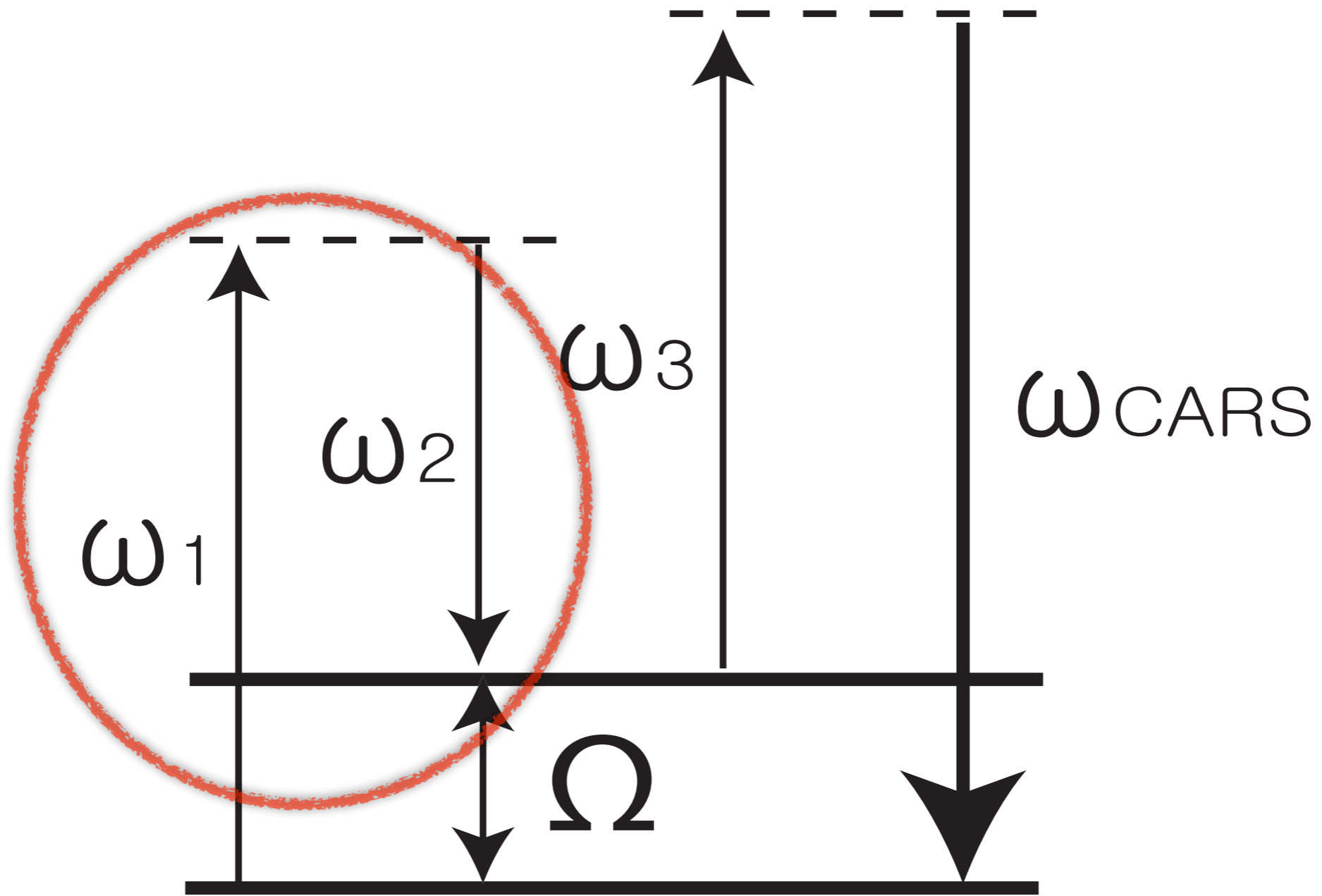
The essential point

Induction of
(coherent) mol. vib.



(a) Resonant process

How to excite mol. vib.?

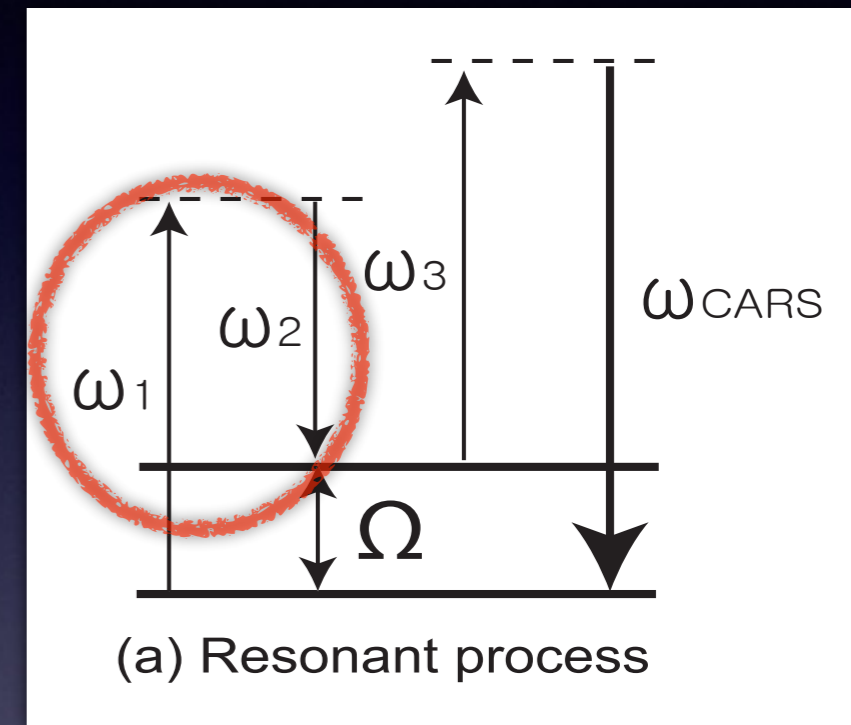


(a) Resonant process

How to excite mol. vib.?

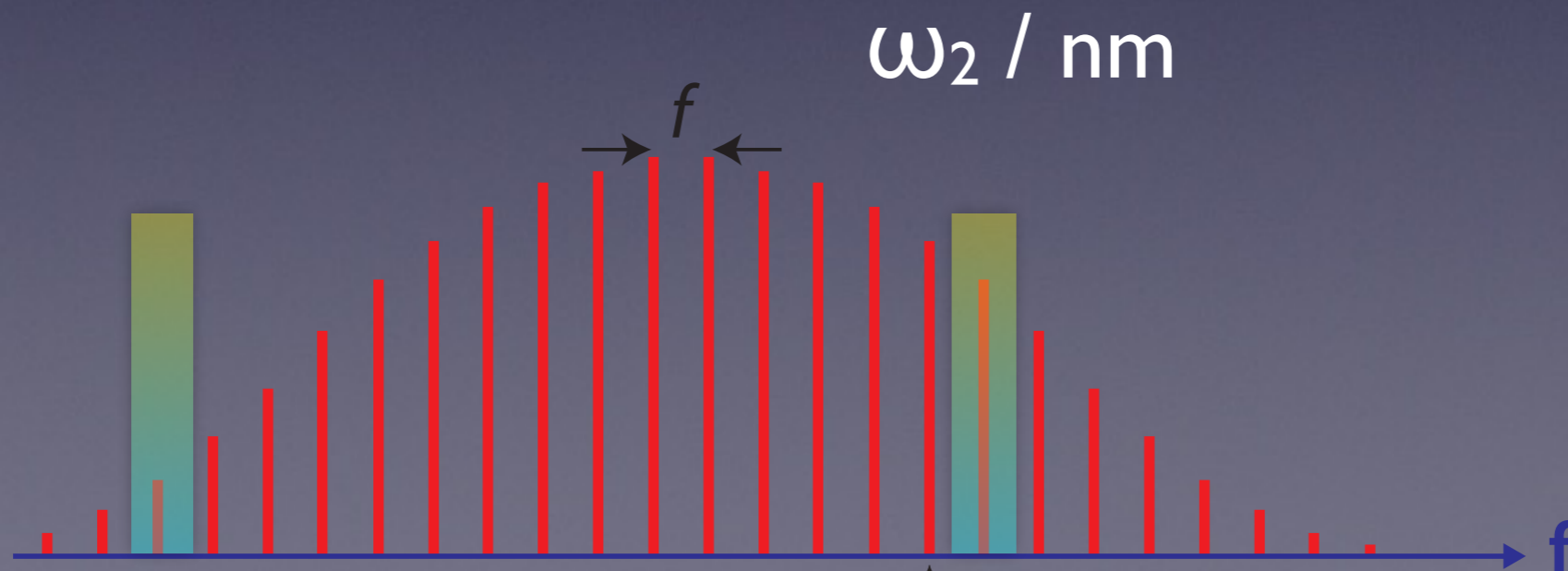
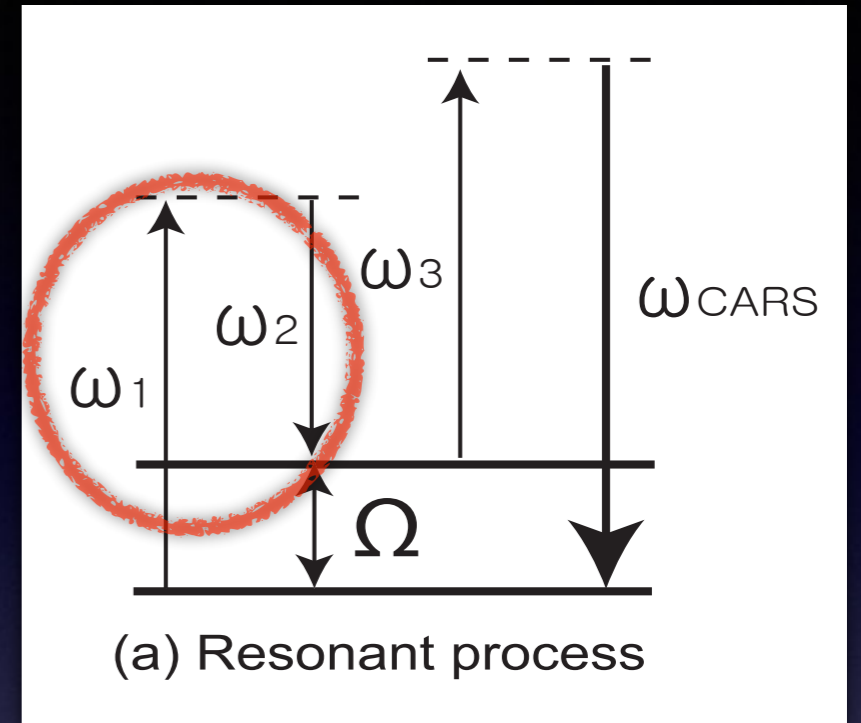
		ω_1 / nm		
		532	780	1064
Mol. Vib.	100 cm^{-1}	535	786	1075
	1000 cm^{-1}	562	846	1191
	3000 cm^{-1}	633	1018	1563

ω_2 / nm



How to excite mol. vib.?

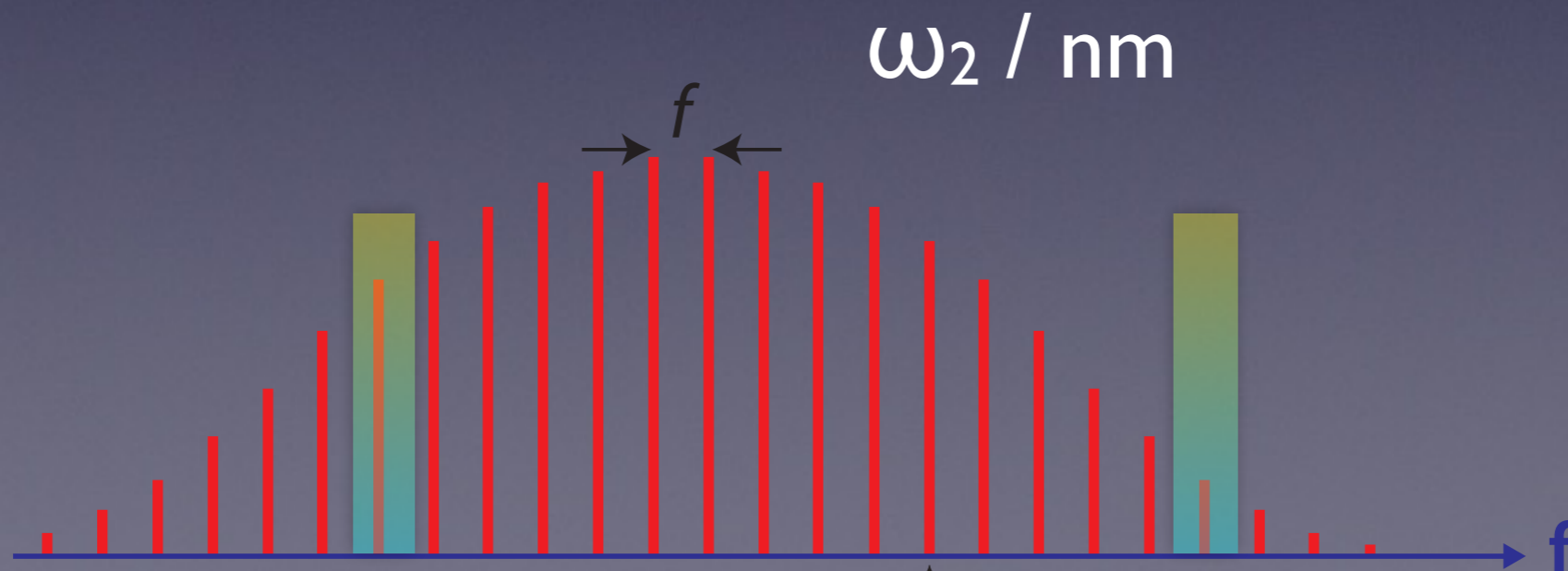
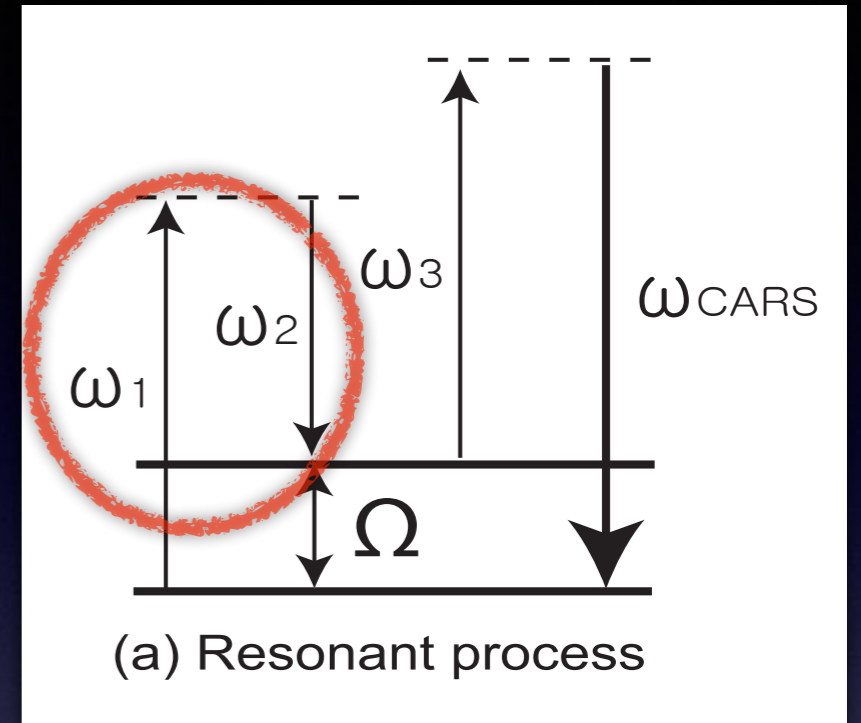
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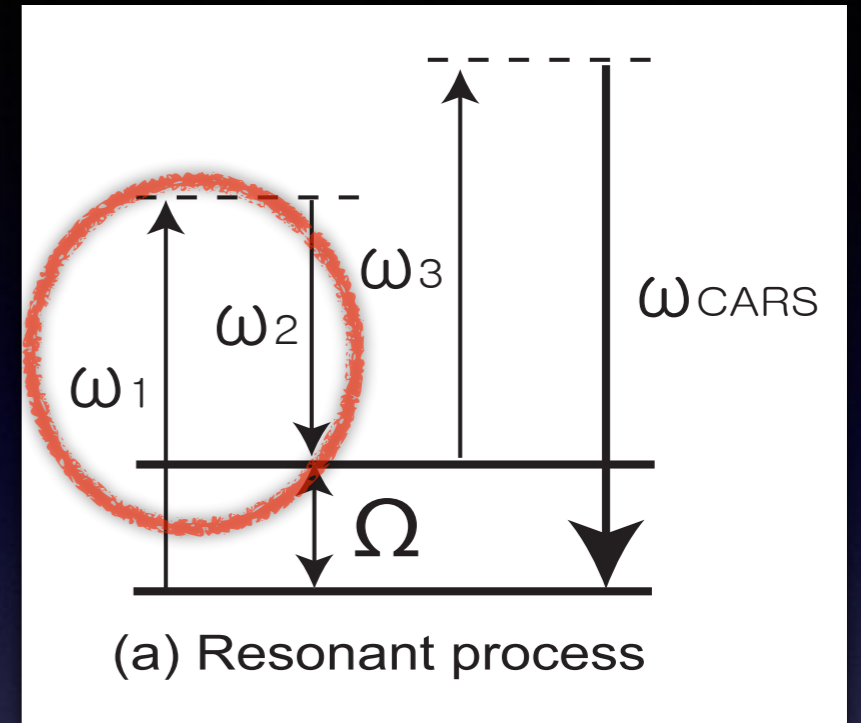


ω_2 / nm

How to excite mol. vib.?

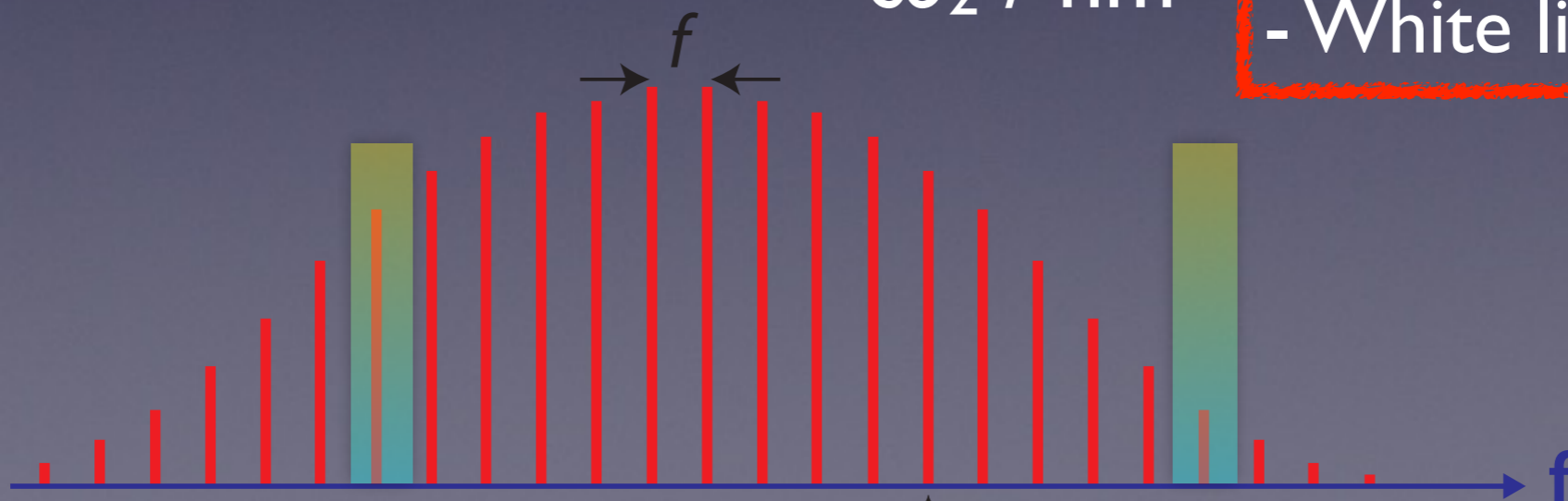
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ω_2 / nm

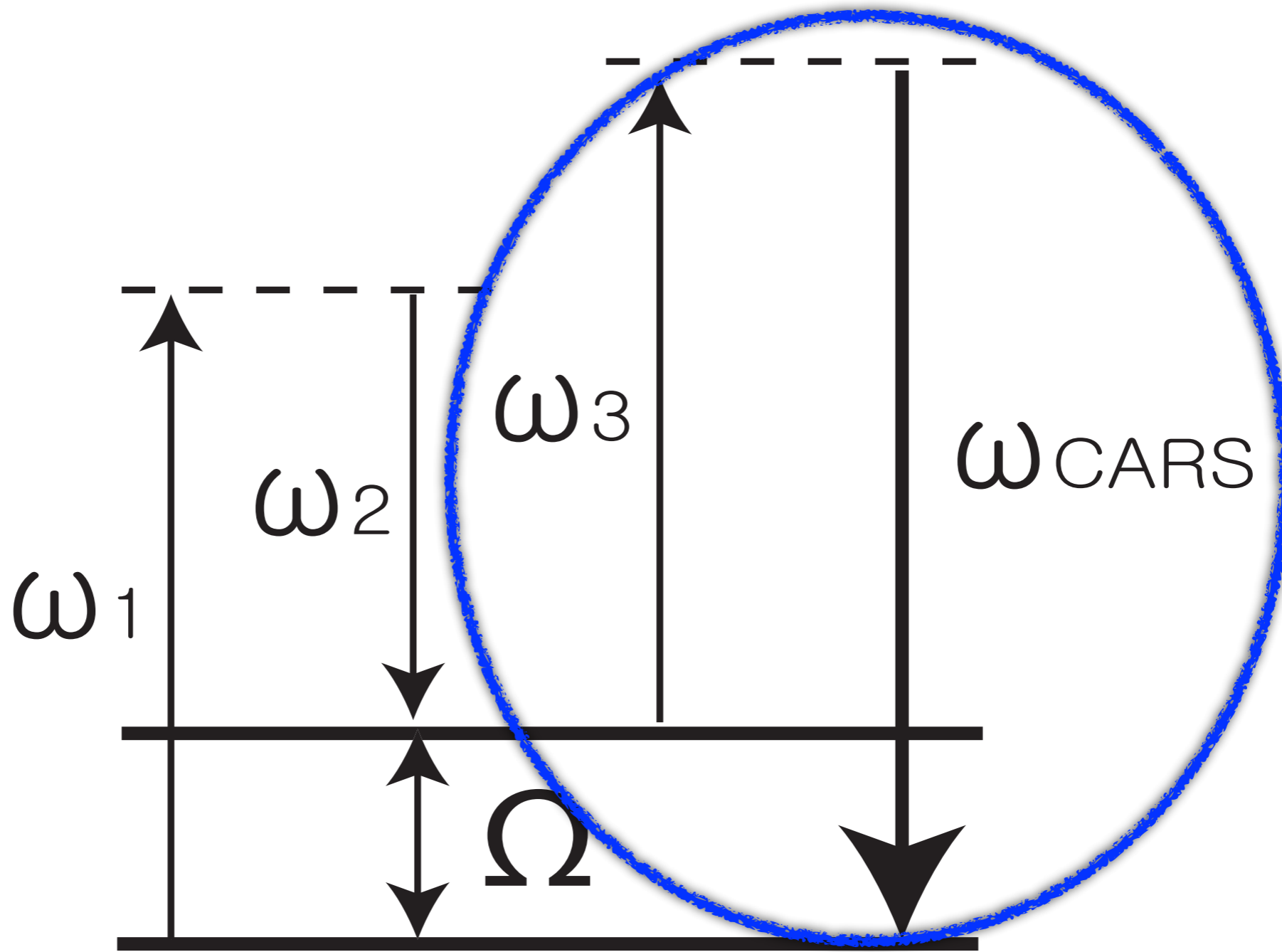


Solution

- Two lasers
- OPO, OPA
- White light continuum



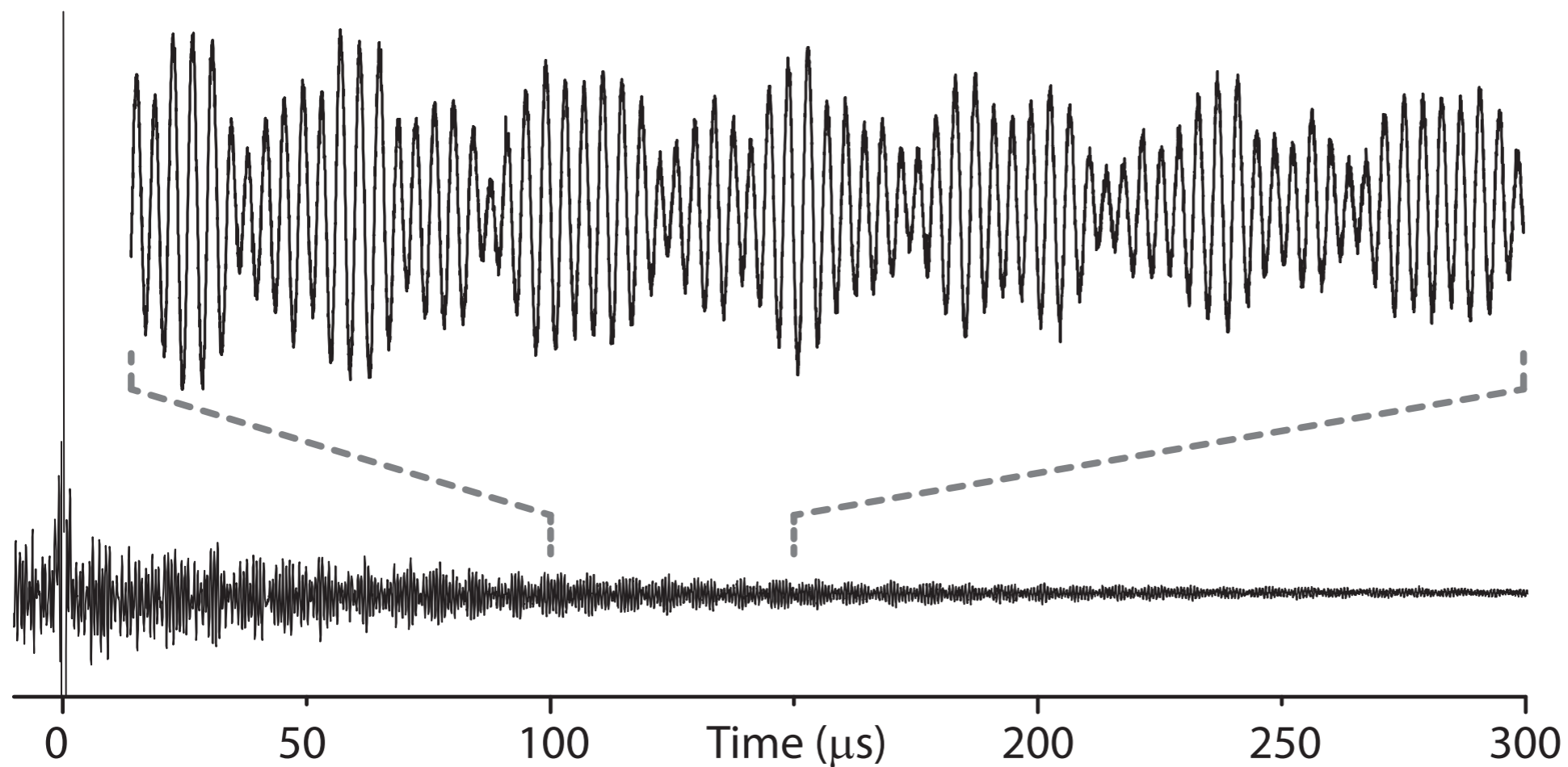
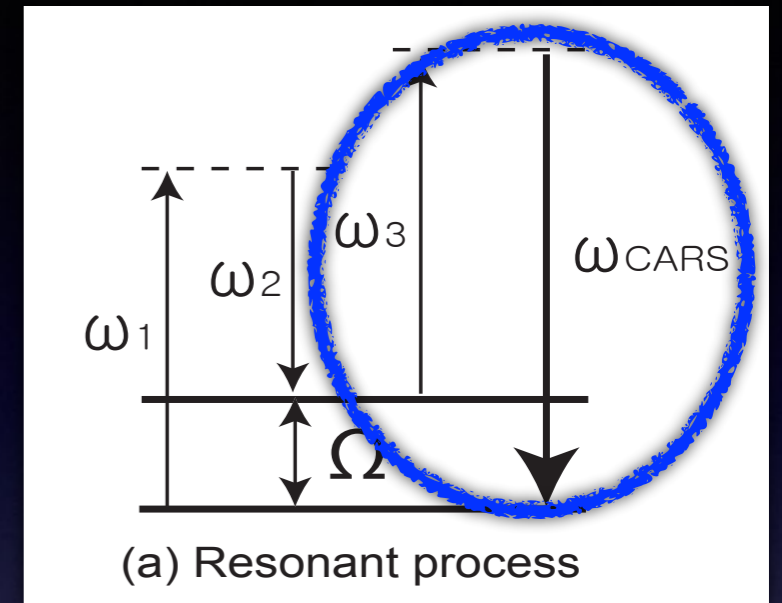
How to probe mol. vib.?



(a) Resonant process

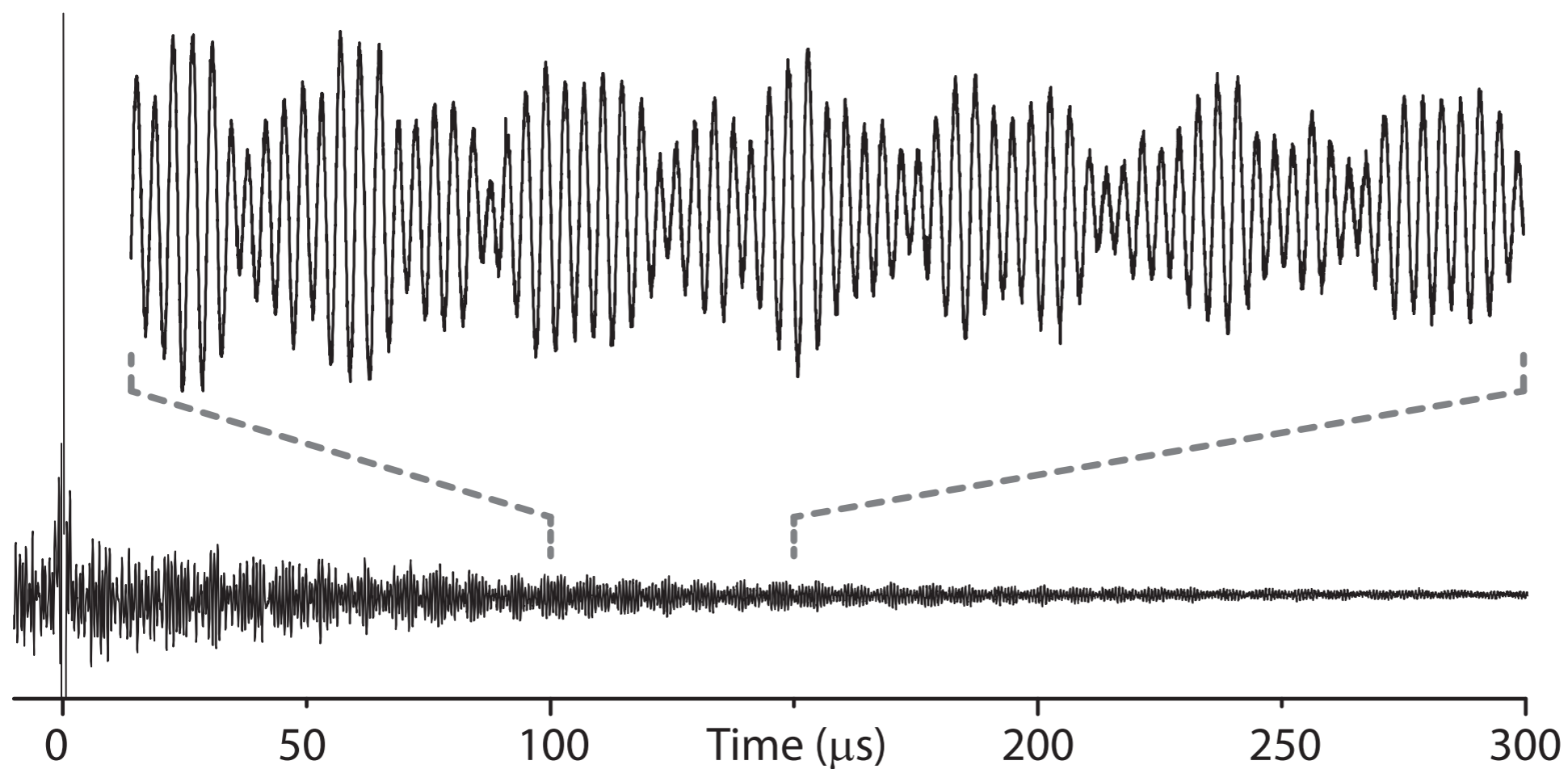
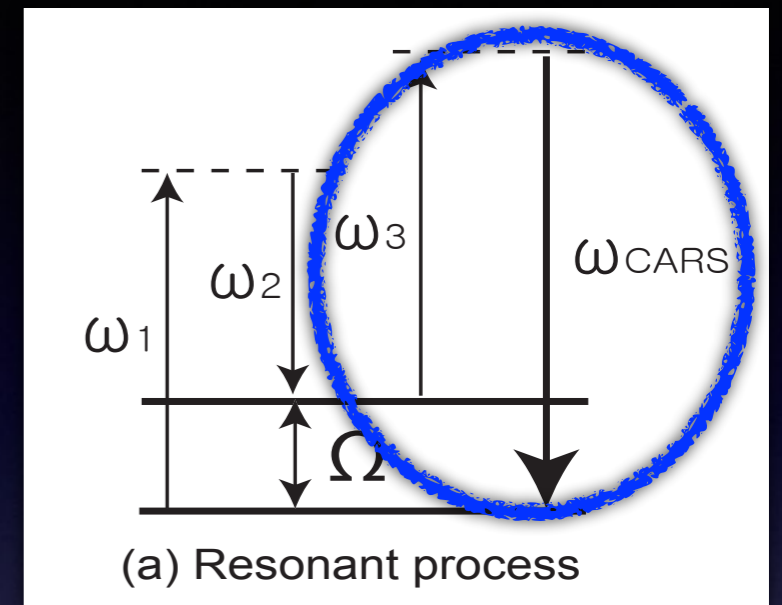
Solution I

Probe with a single laser
that modulated by mol vib.



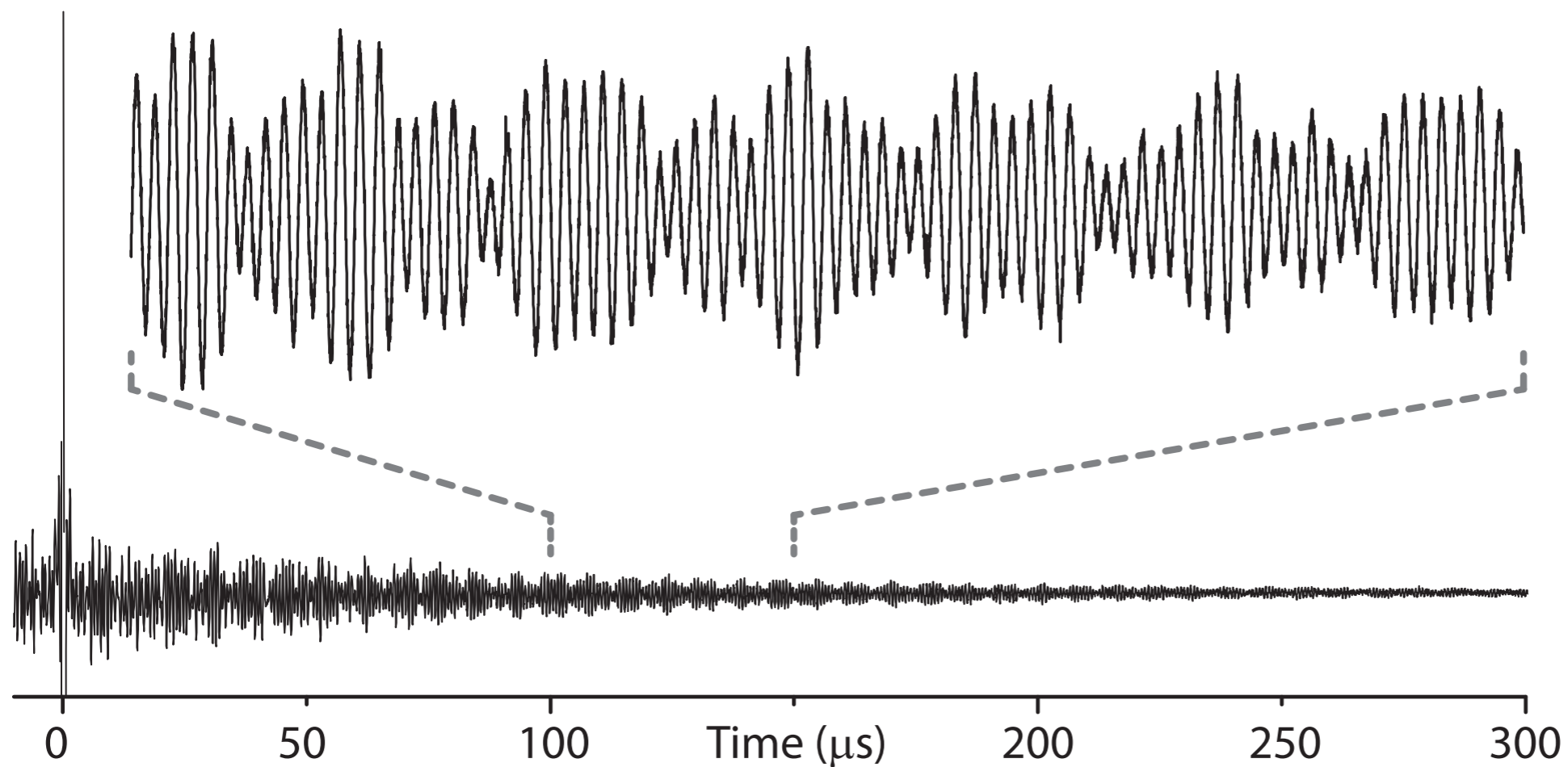
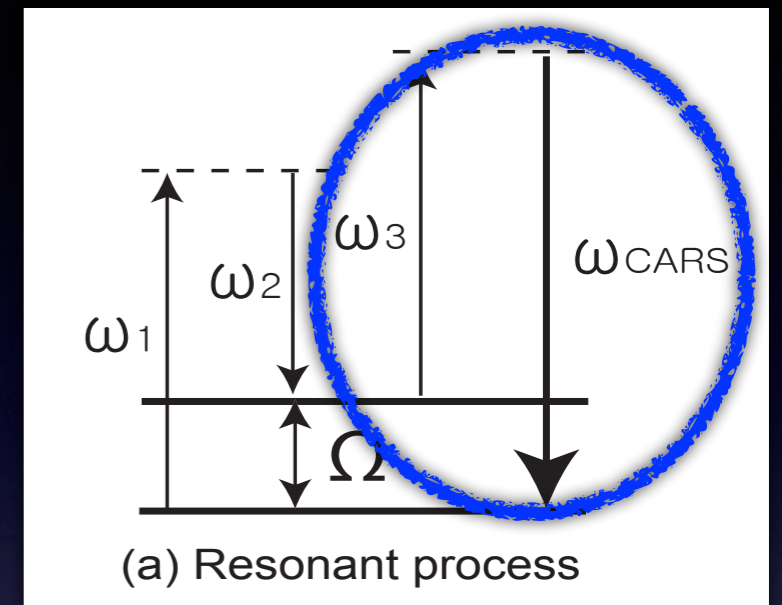
Solution 1

		Freq.	Wavelen.	Period
Mol. Vib.	100 cm^{-1}	3 THz	100 μm	333 fs
	1000 cm^{-1}	30 THz	10 μm	33 fs
	3000 cm^{-1}	90 THz	3.3 μm	11 fs



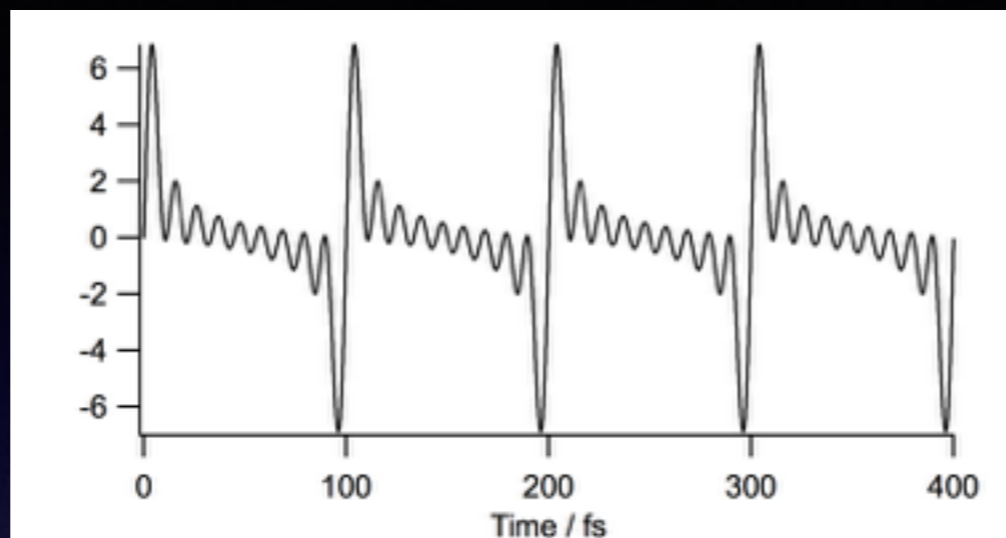
Solution 1

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Mol. Vib.	100 cm^{-1}	3 THz	100 μm	333 fs
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Solution 1

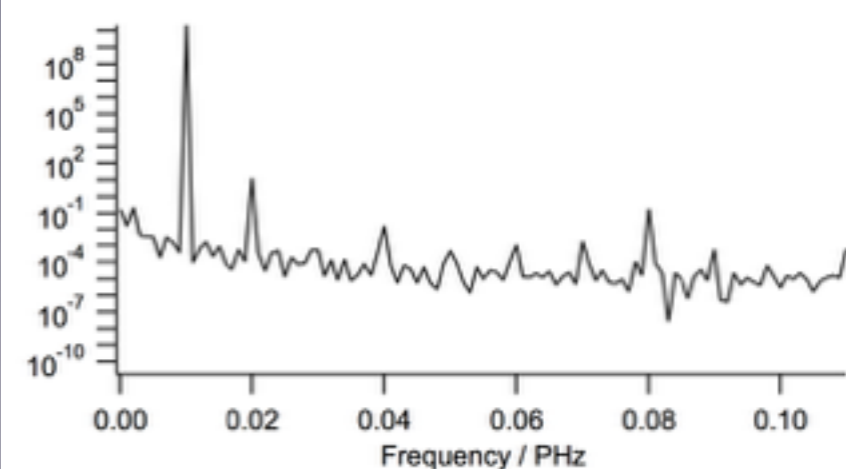
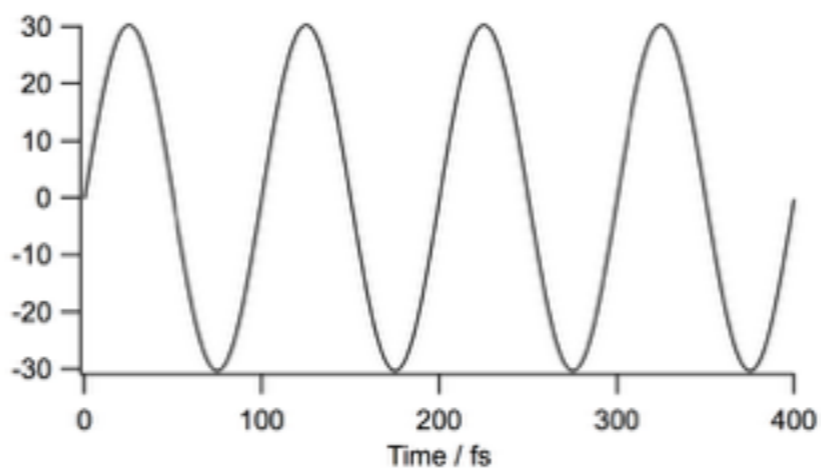
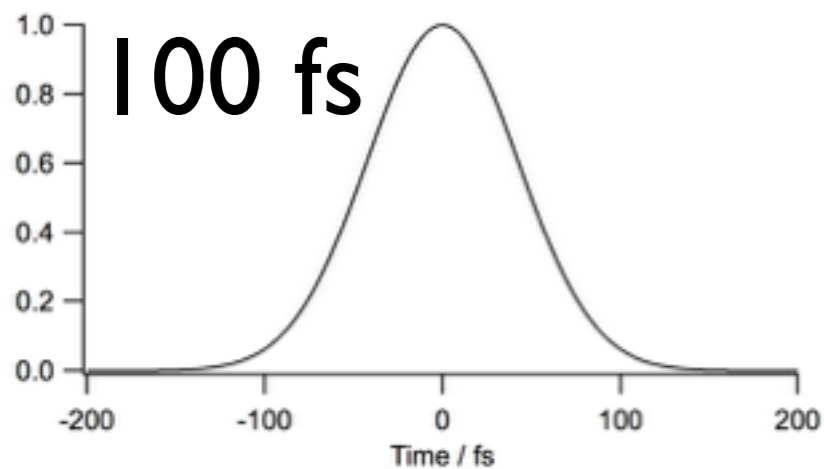
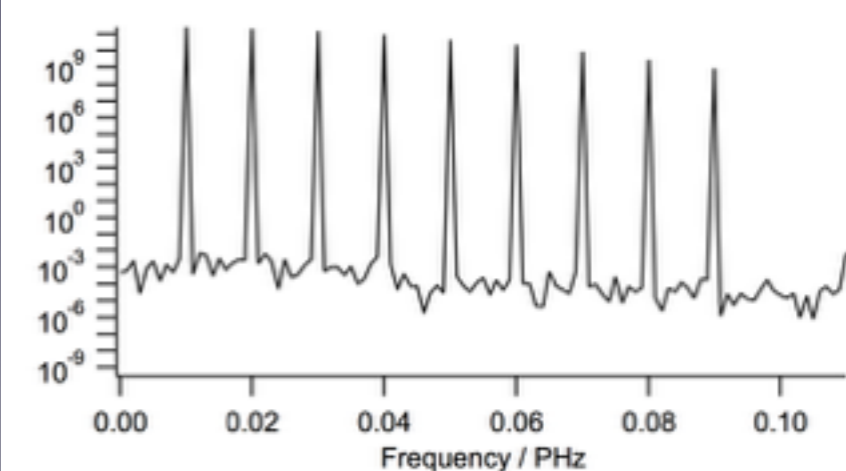
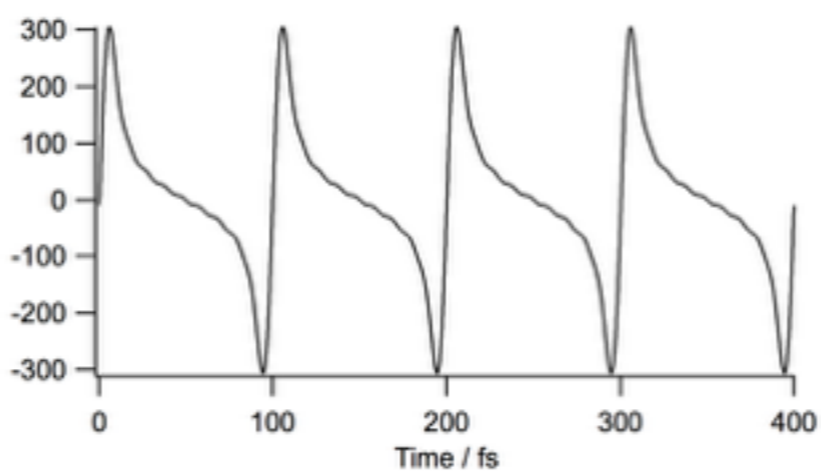
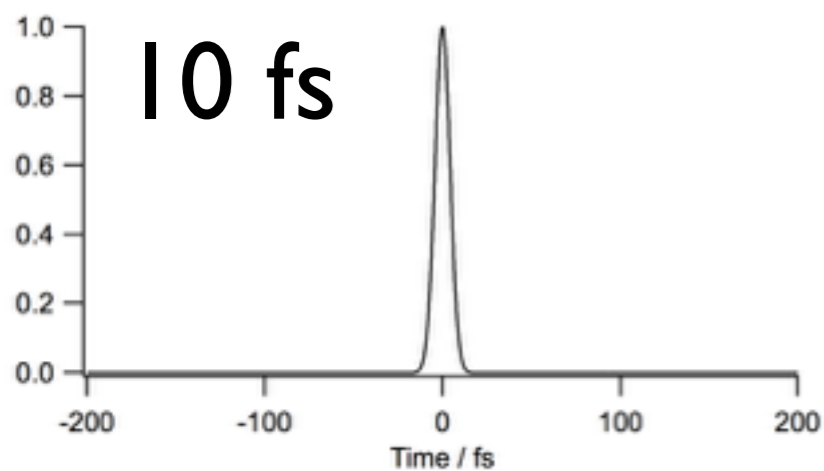
Mol. vib
10 THz - 90 THz



Probe pulse

Obs. mol. vib.

Fourier transform



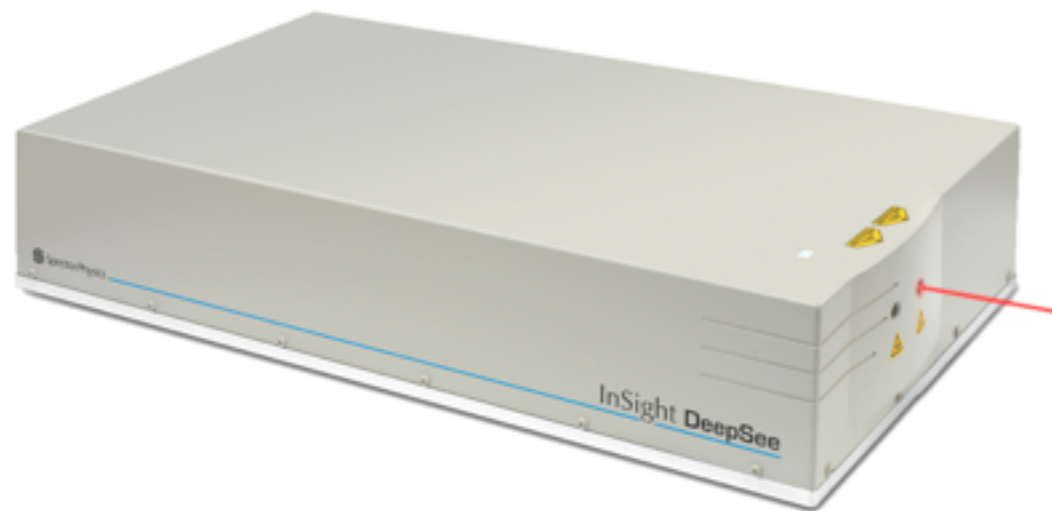
Solution 1

Short pulse duration with 10 fs or less
is required for probing 3000 cm^{-1}

Solution 1

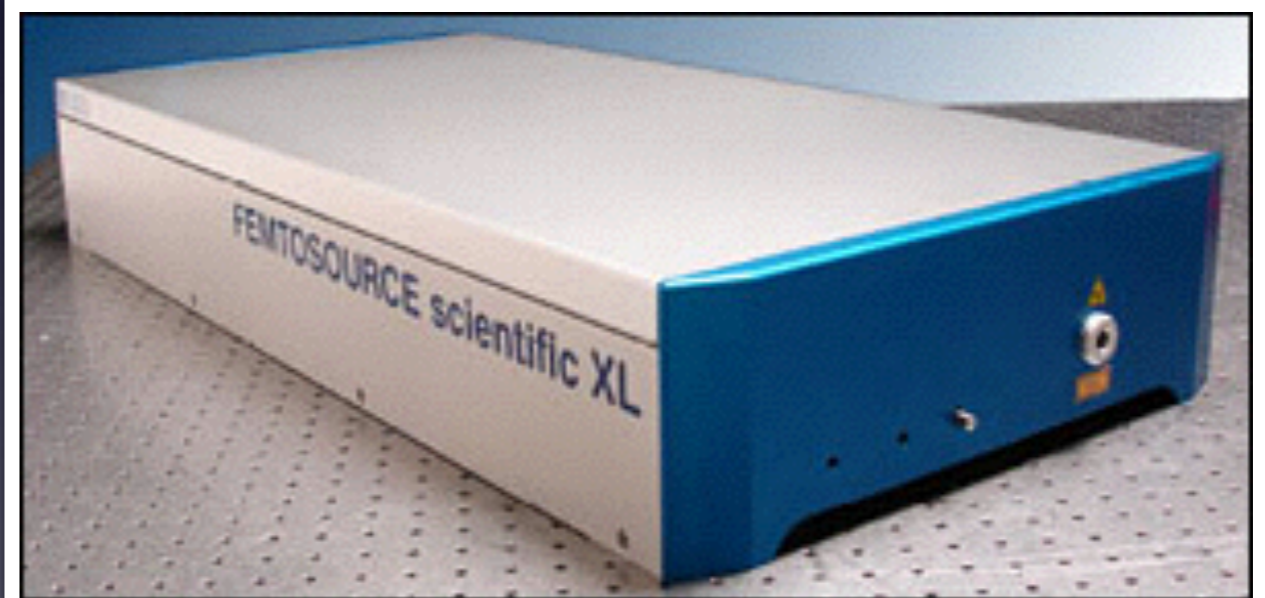
Short pulse duration with 10 fs or less is required for probing 3000 cm^{-1}

DeepSee



700-1250 nm, 100 fs, dual pulse

For mol. vib. excitation



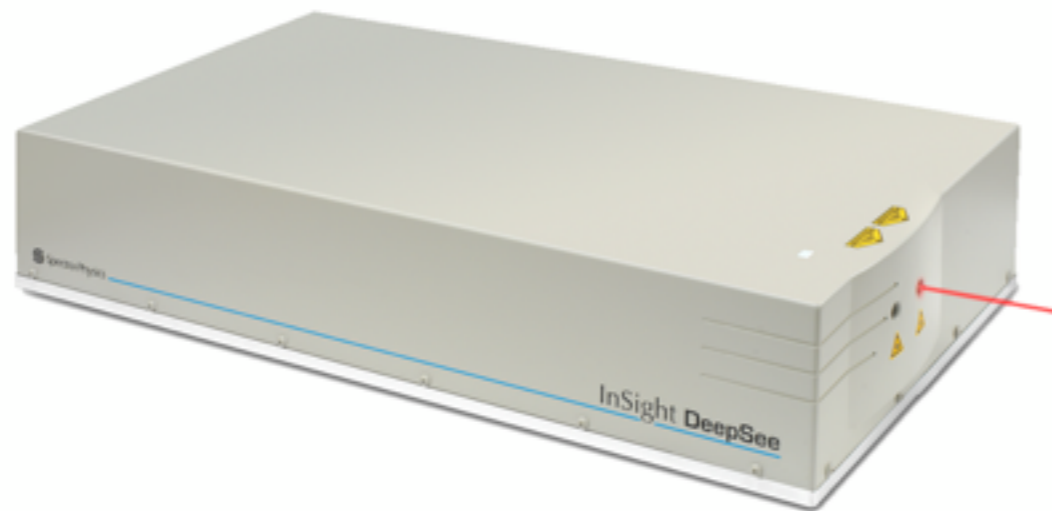
about 700-900 nm, 10 fs

For probe

Solution 1

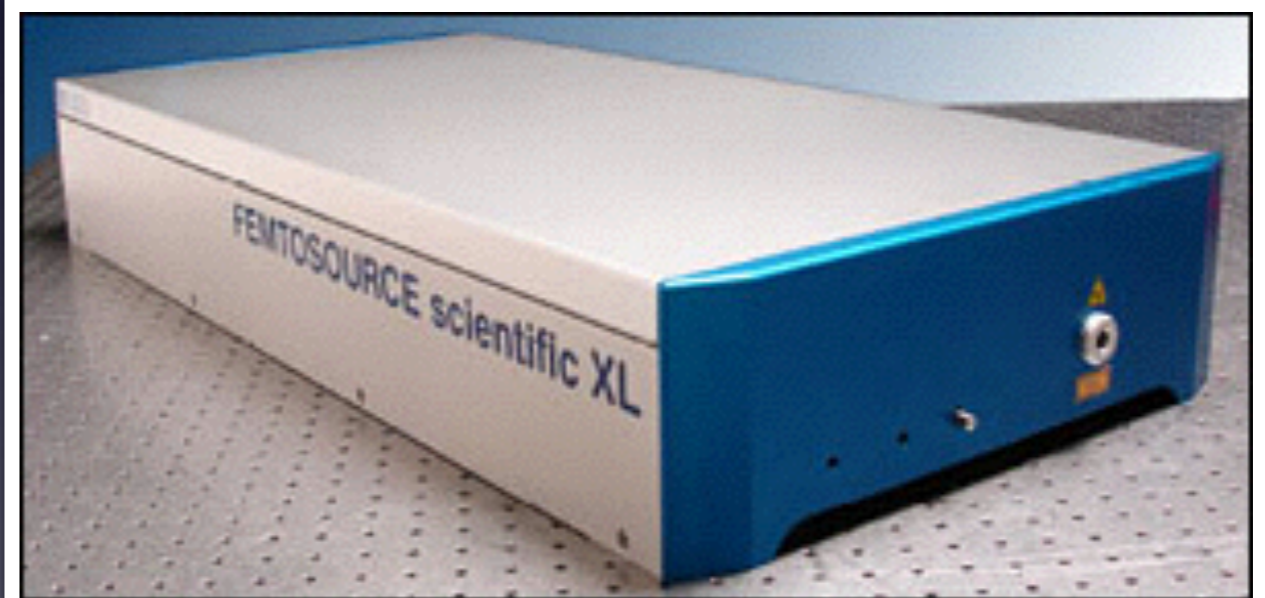
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DeepSee



700-1250 nm, 100 fs, dual pulse

For mol. vib. excitation

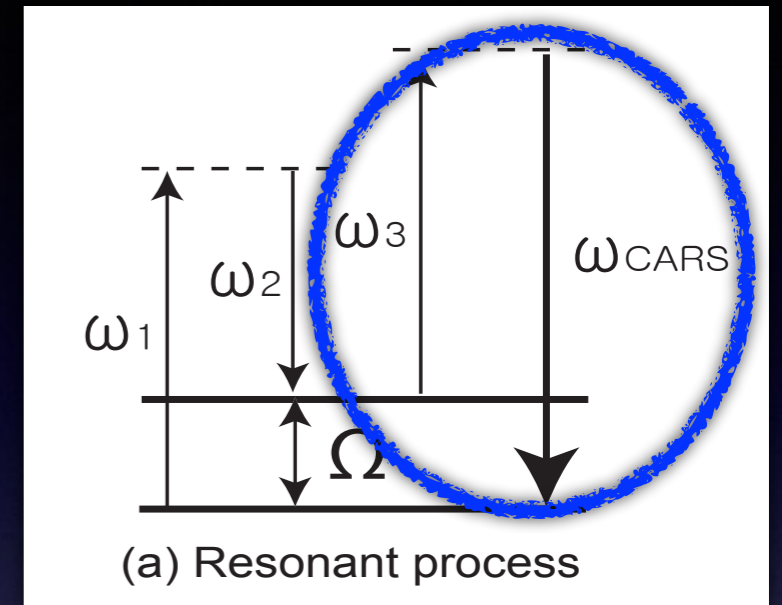


about 700-900 nm, 10 fs

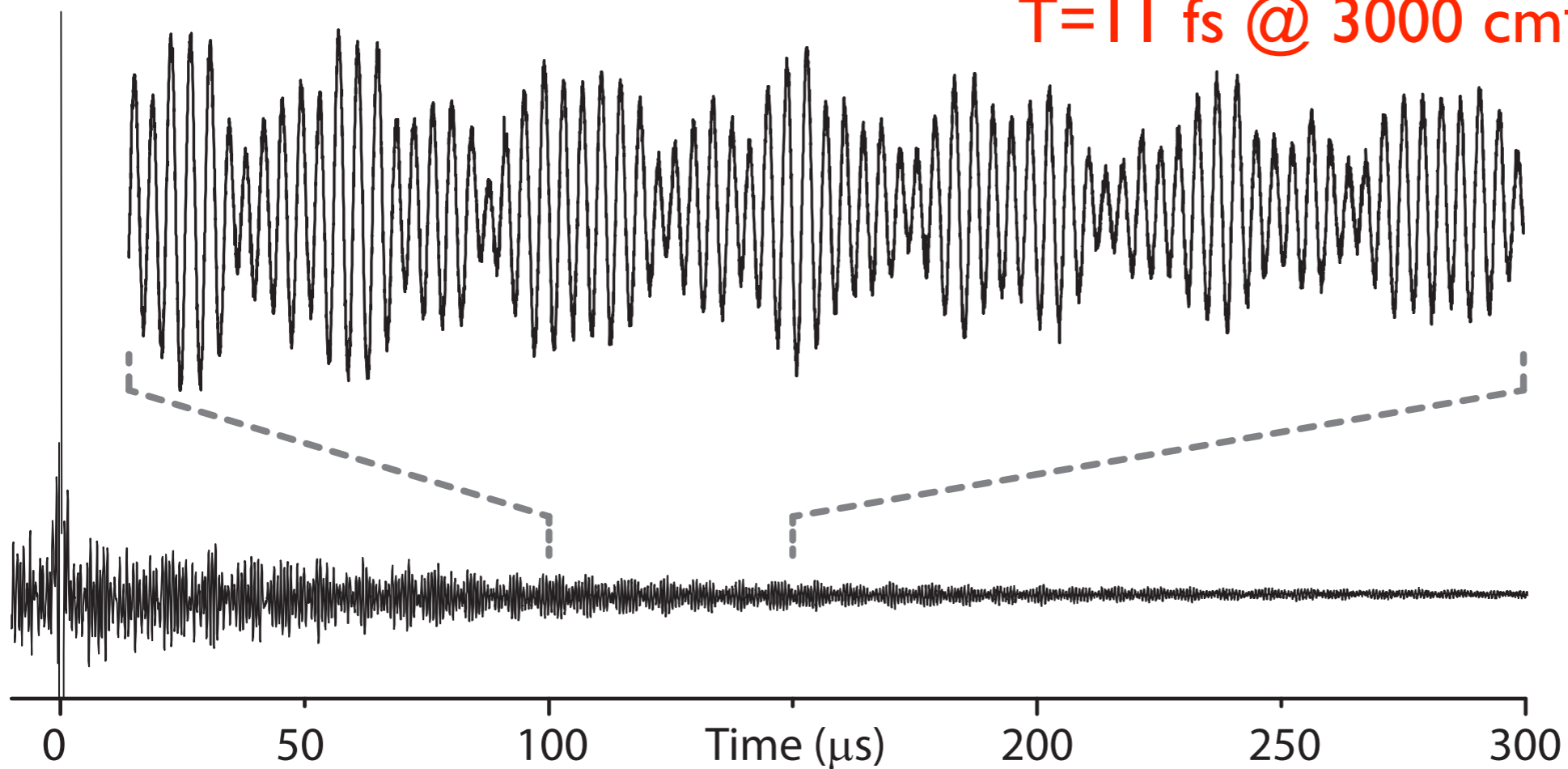
For probe

Solution 2

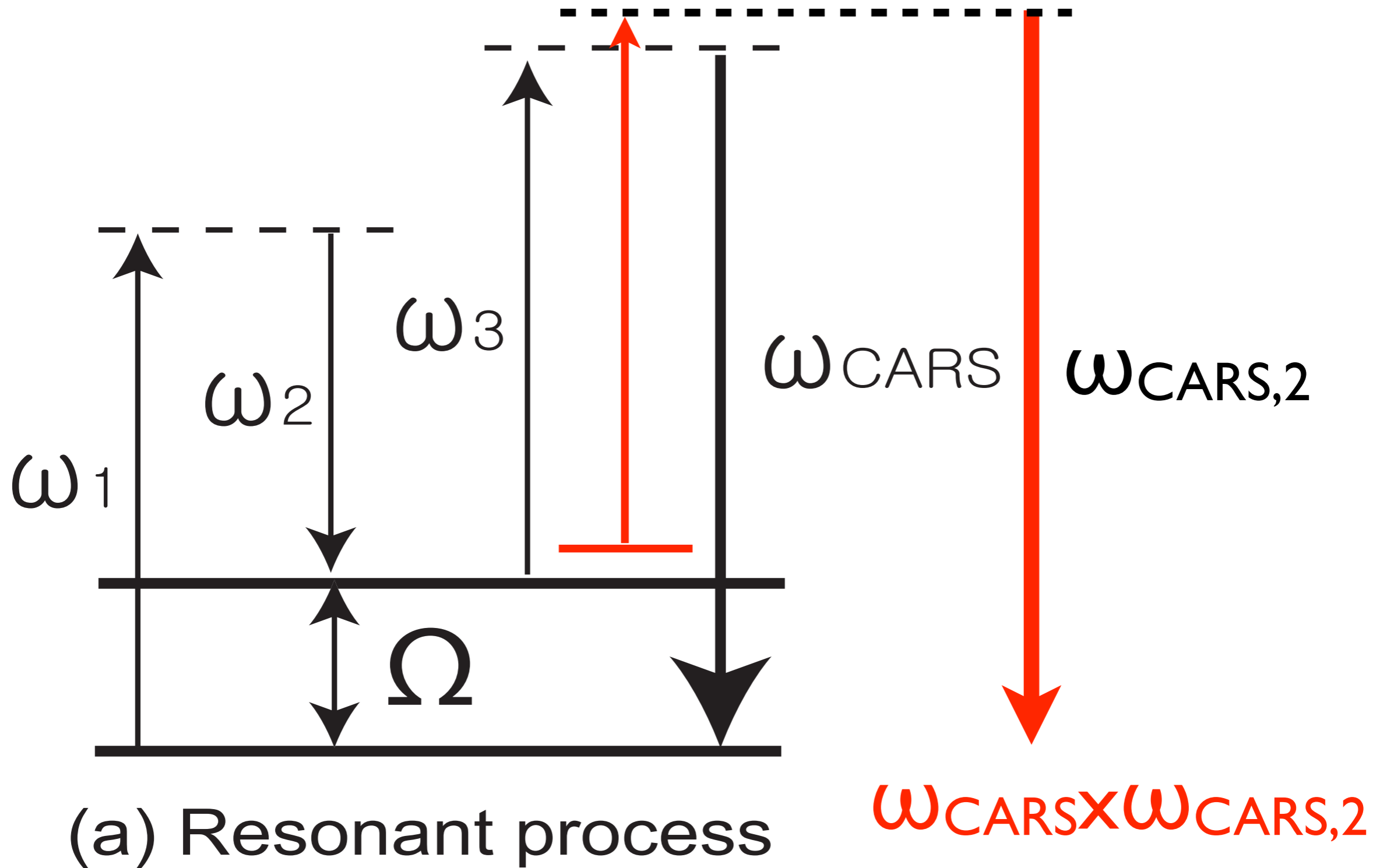
Ignore absolute frequency accuracy
but obtain relative frequency of mol vib



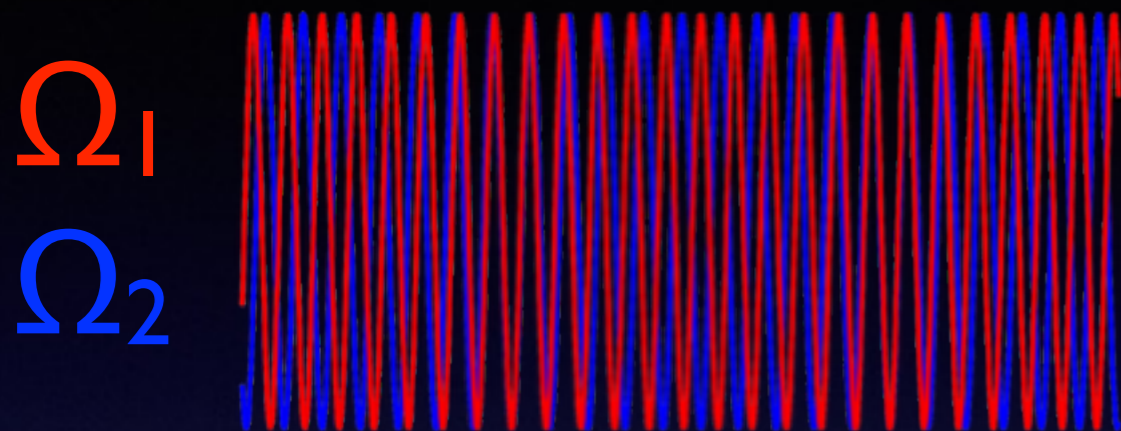
$T = 11 \text{ fs @ } 3000 \text{ cm}^{-1}$



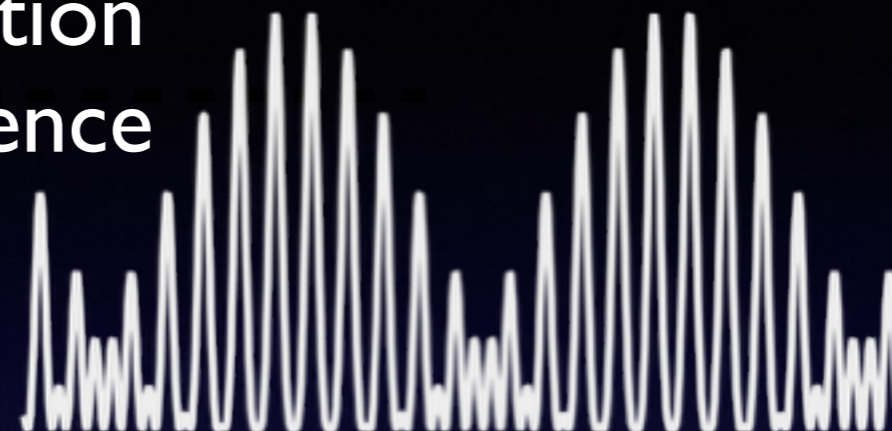
Solution 2



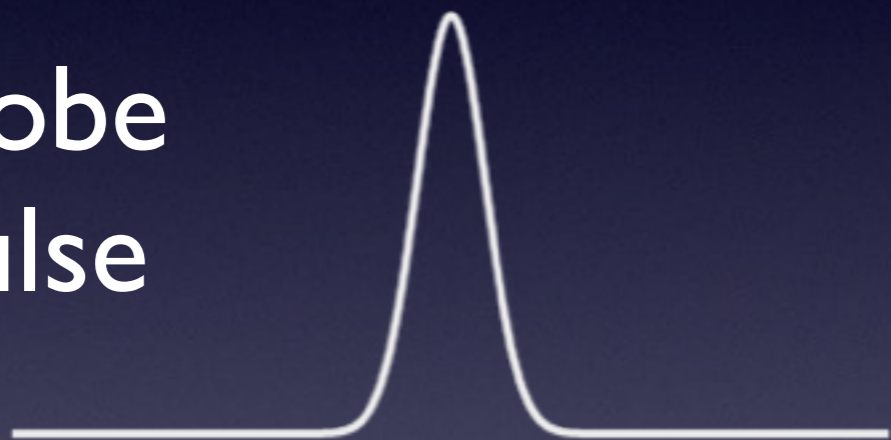
Solution 2



Polarization interference



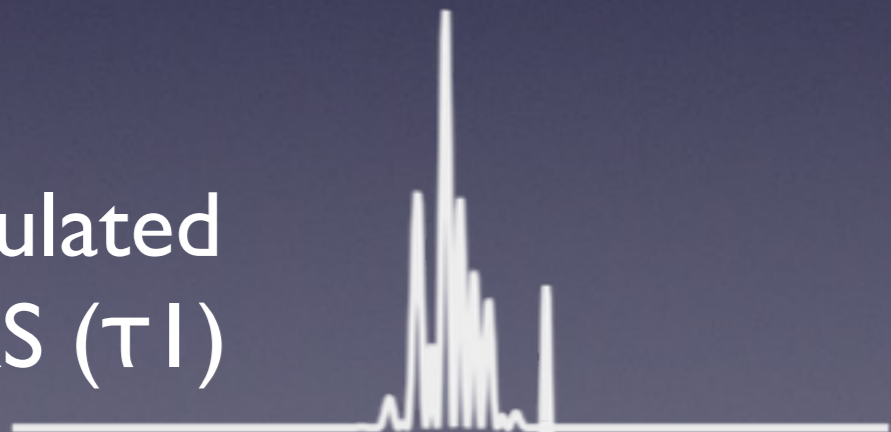
Probe pulse



Observed CARS



Modulated CARS (τ_1)



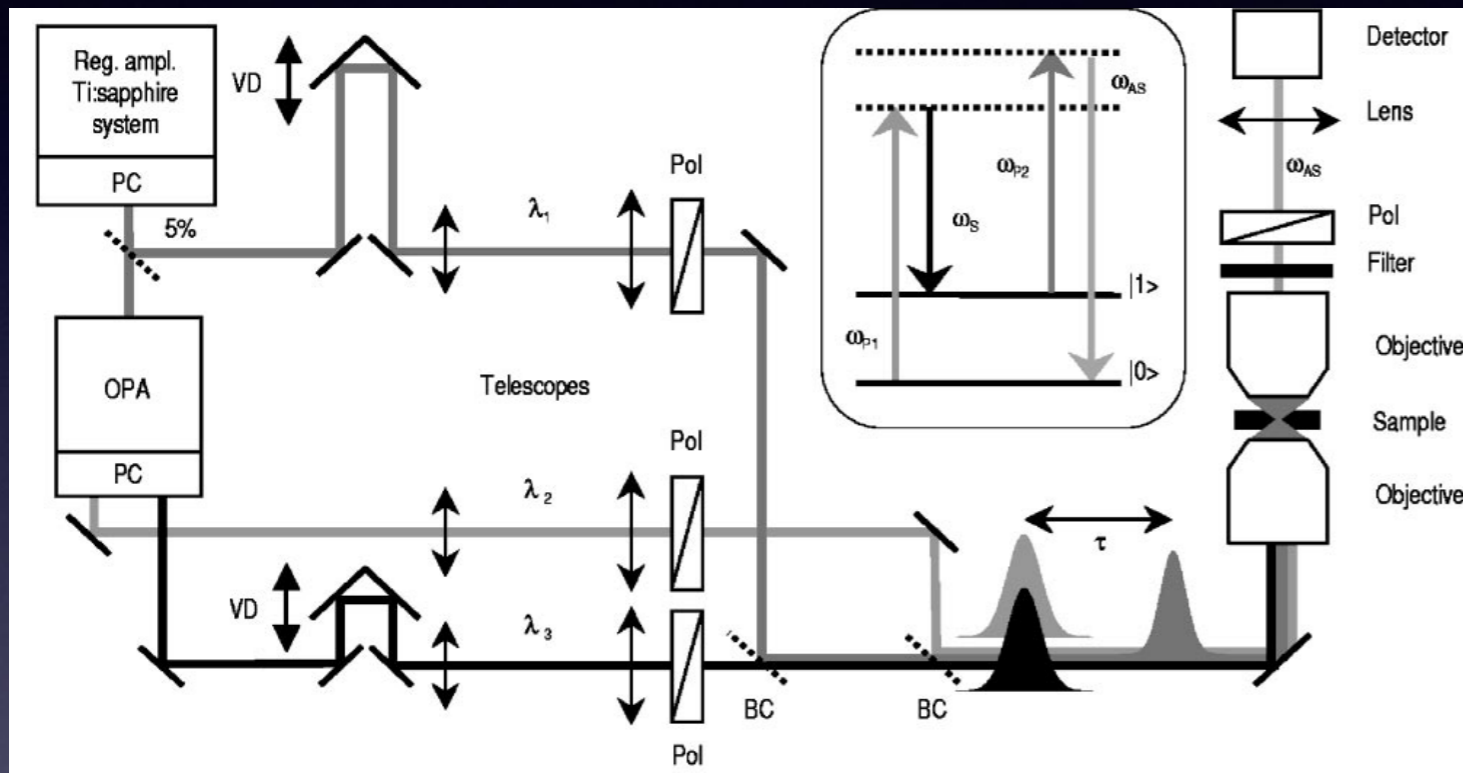
Modulated CARS (τ_2)



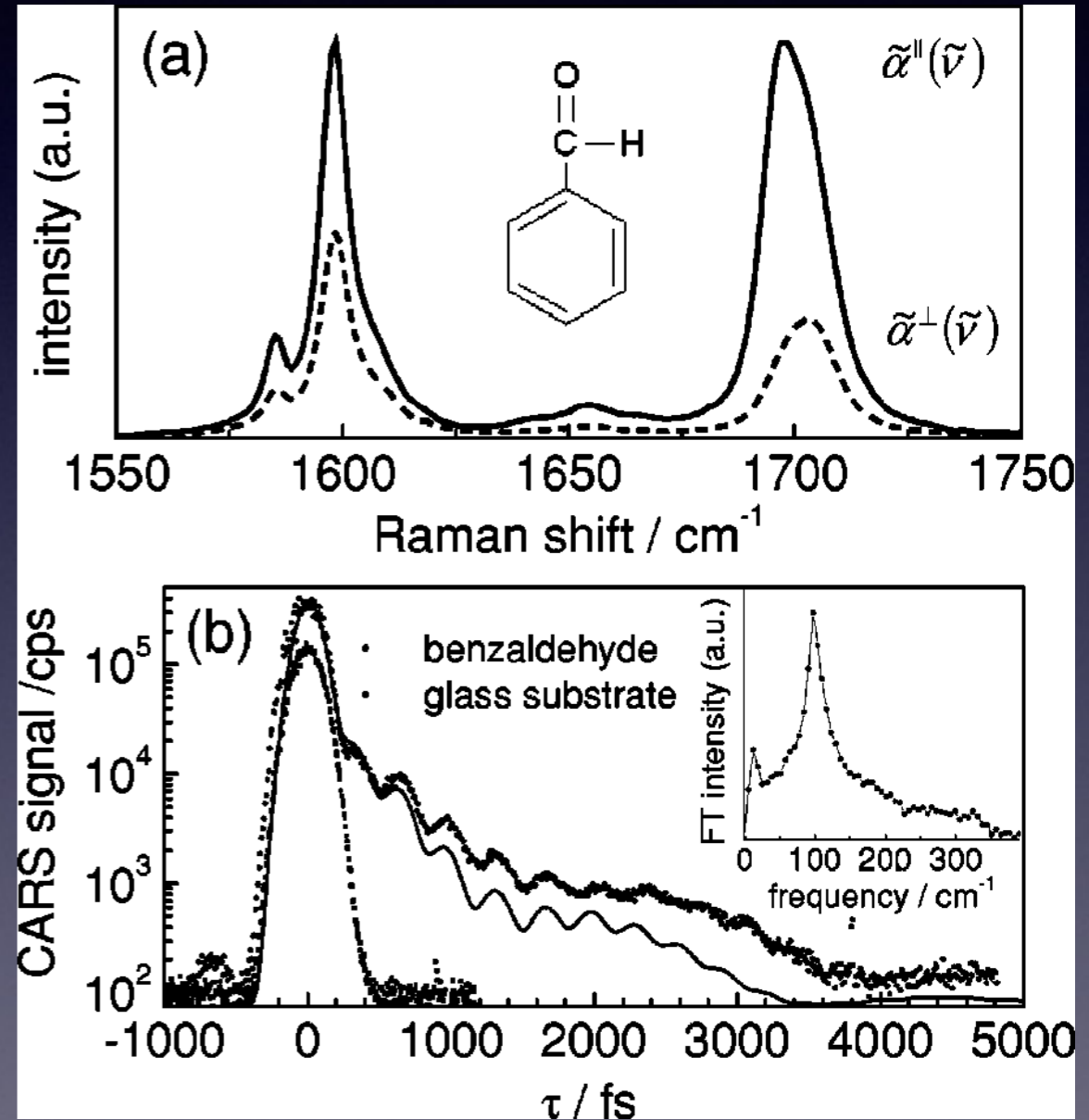
$$= \text{abs}(\Omega_1 - \Omega_2)$$

100 fs (333 cm^{-1})

Solution 2



Volkmer, et al. APL, 2002



Solution 2

For the determination of absolute frequency

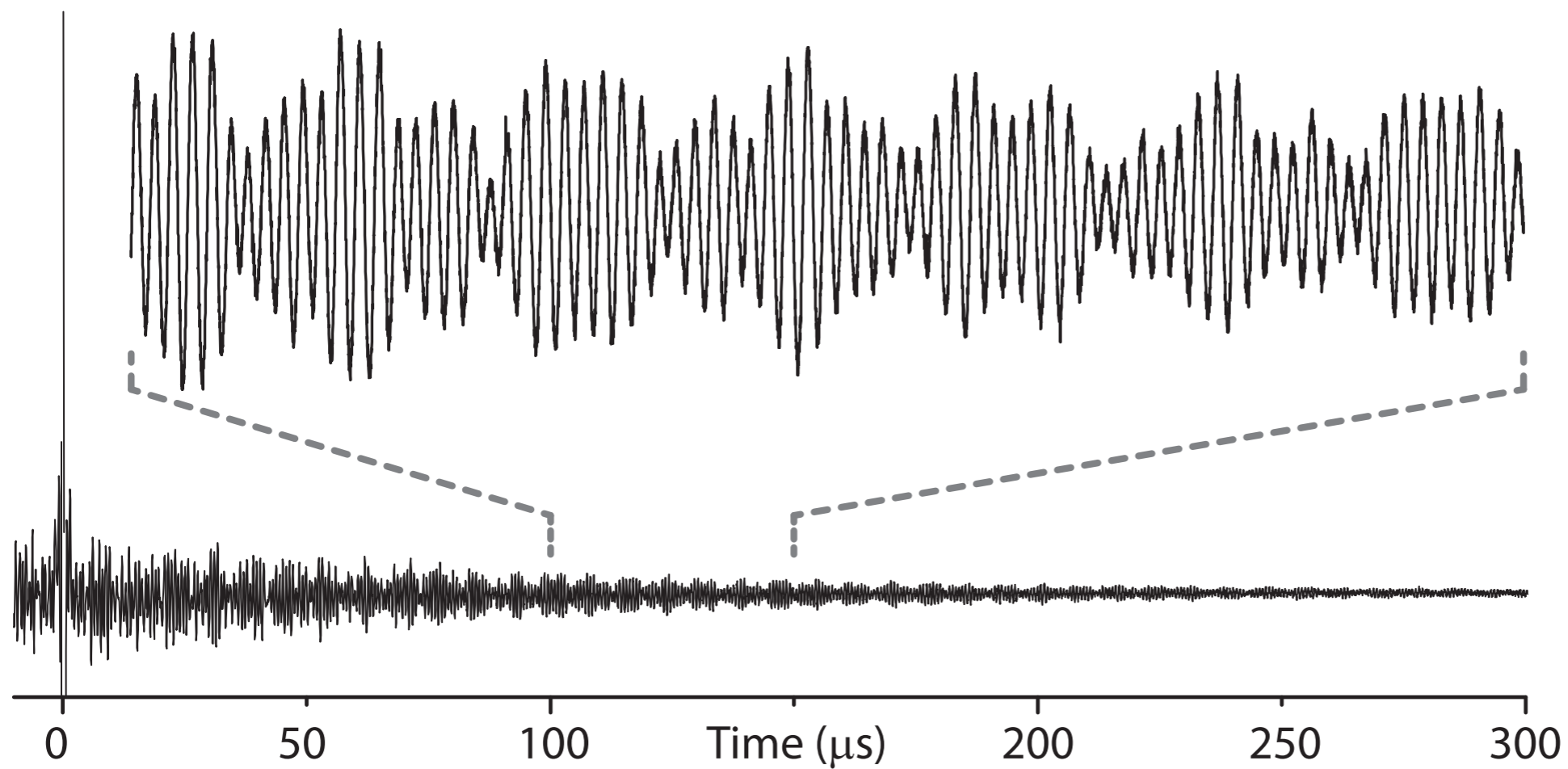
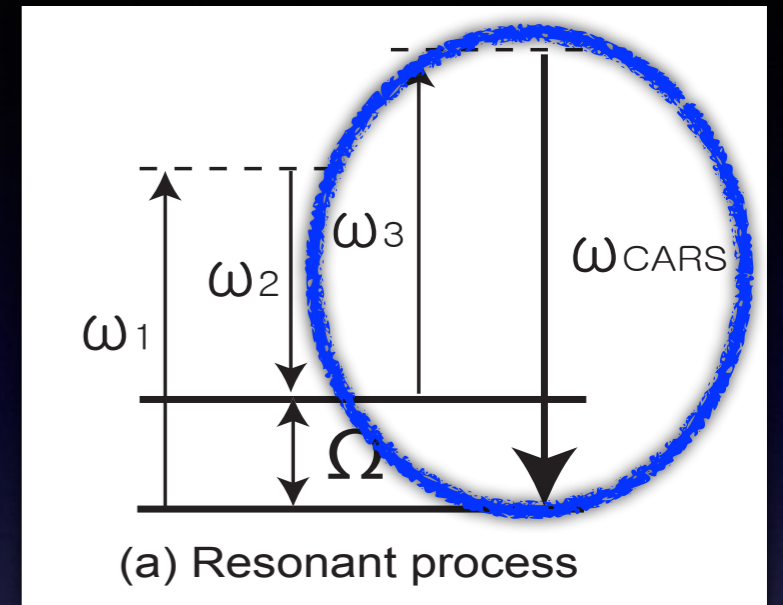
Interference w/ mid-infrared light and CARS beats

Interference w/ visible light beat and CARS beats

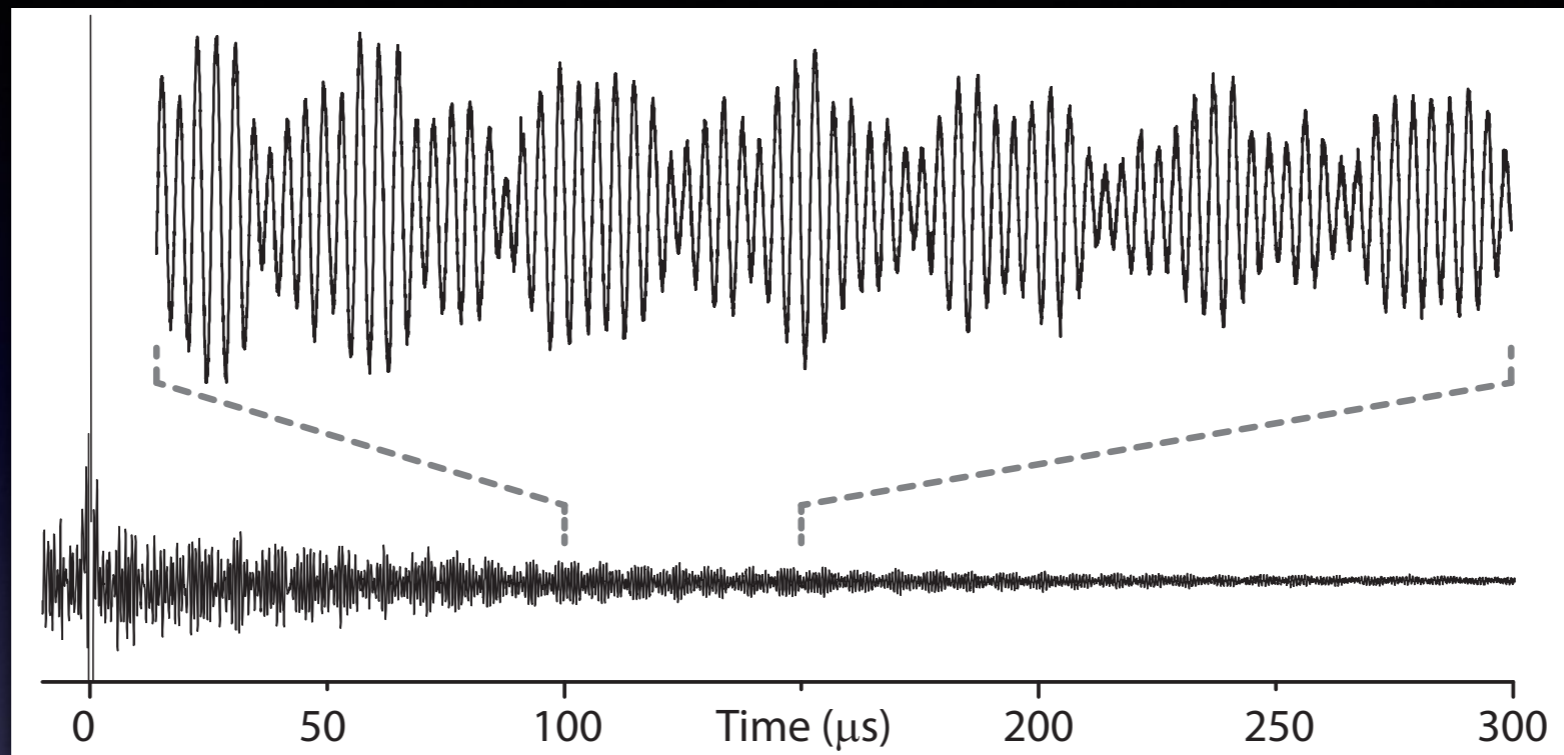
$$3000 \text{ cm}^{-1} = 3.3 \text{ } \mu\text{m} = 90 \text{ THz}$$

Solution 3

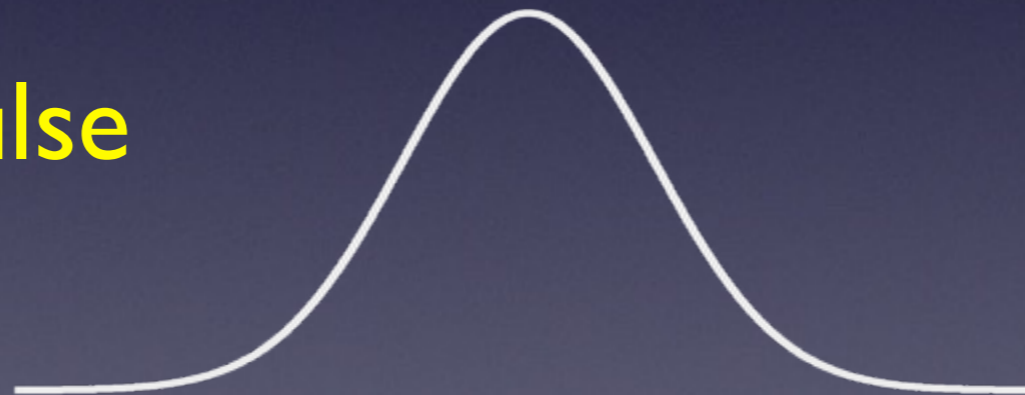
Probe by
auto- or cross-correlation



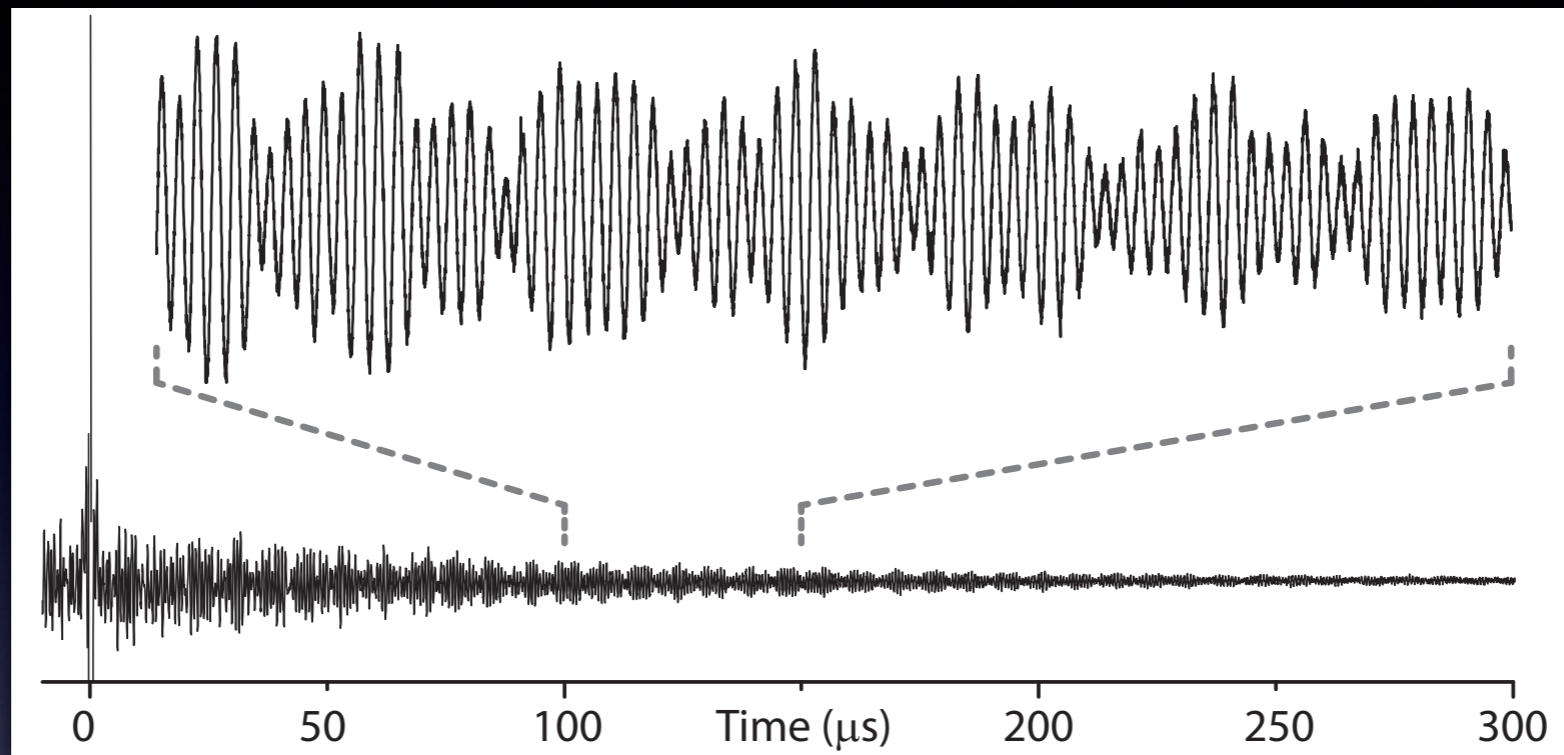
Solution 3



Probe pulse



Solution 3



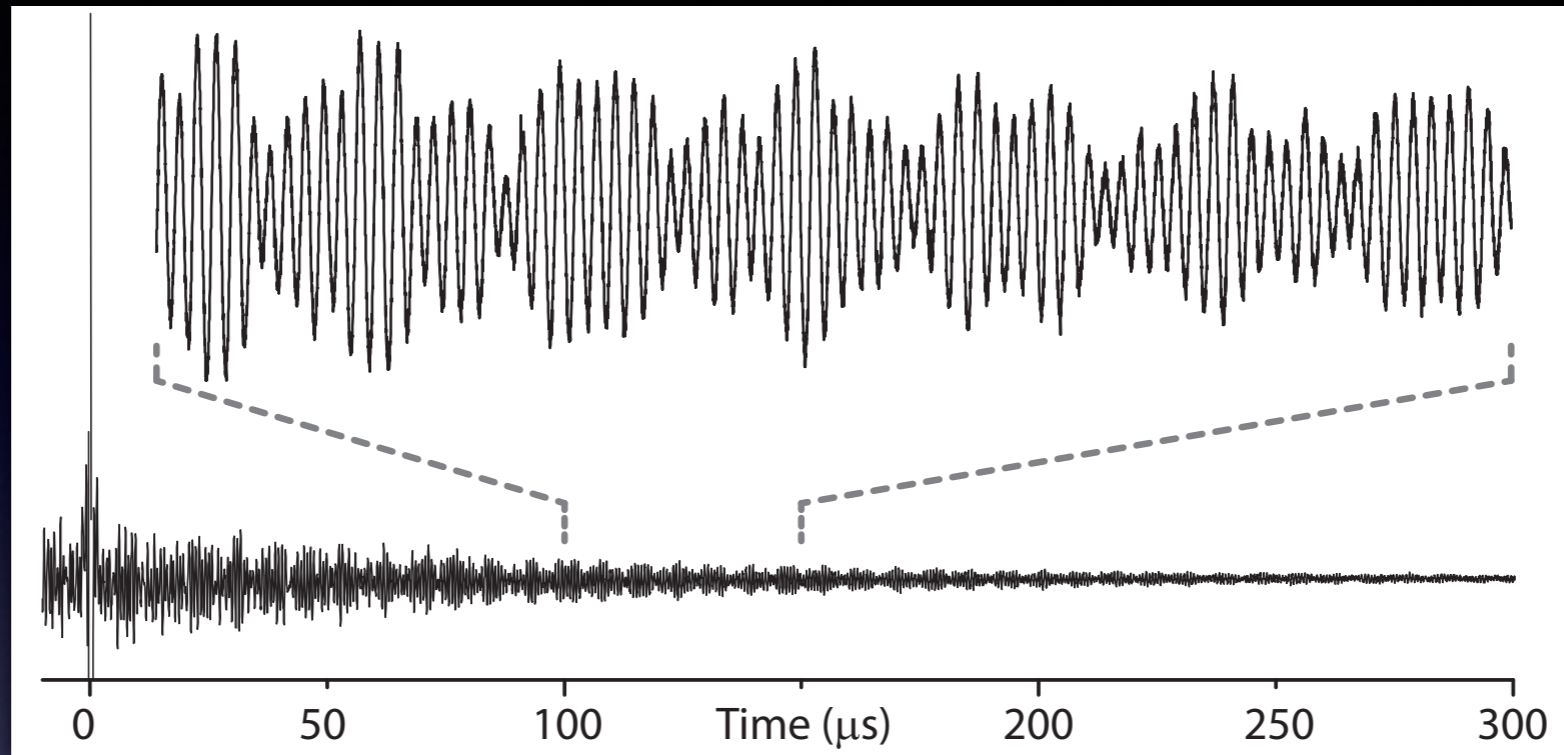
Probe pulse



CARS



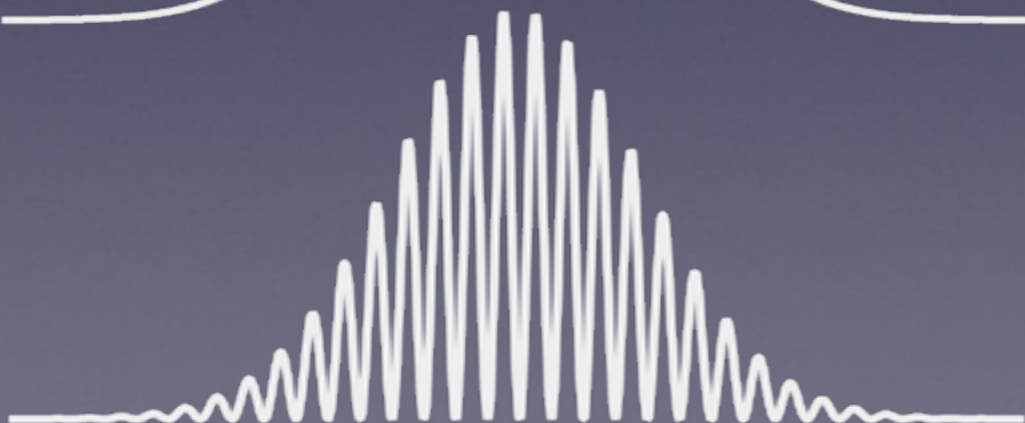
Solution 3



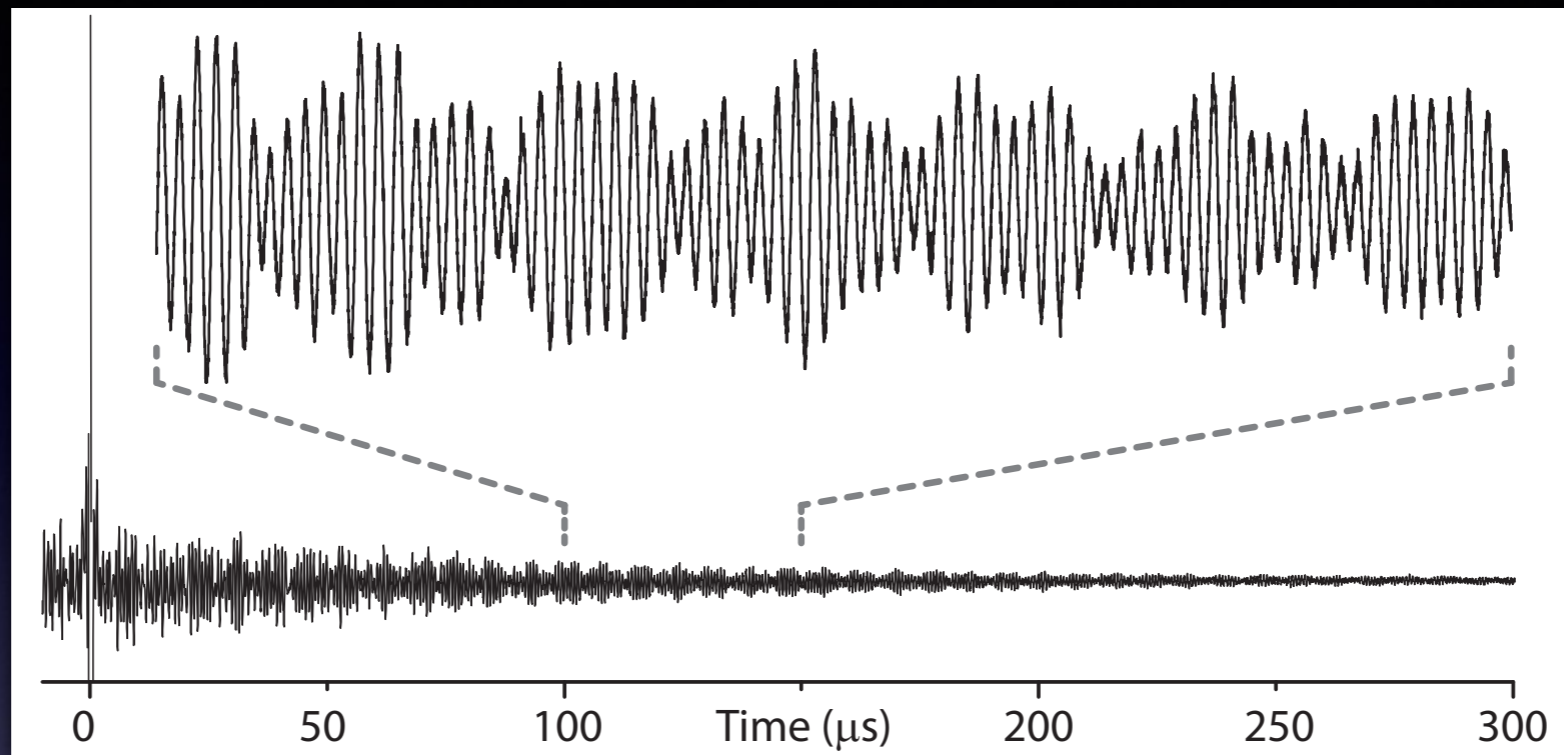
the pulse



ARS



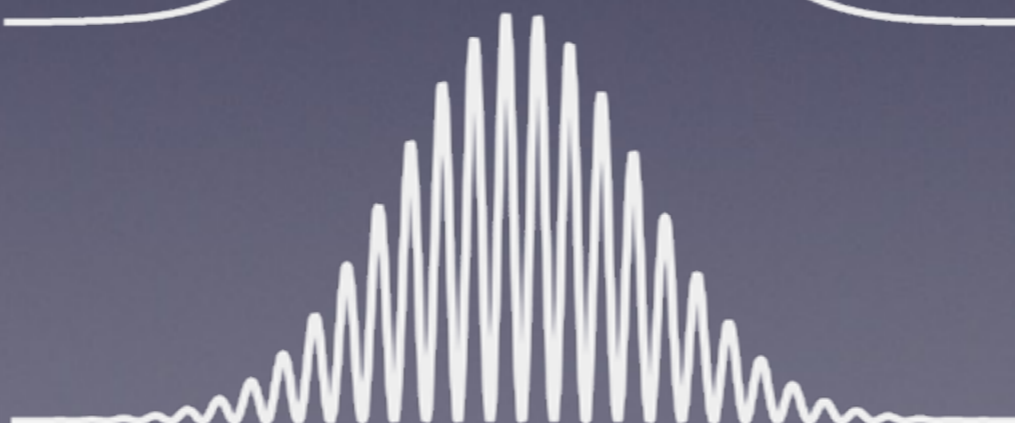
Solution 3



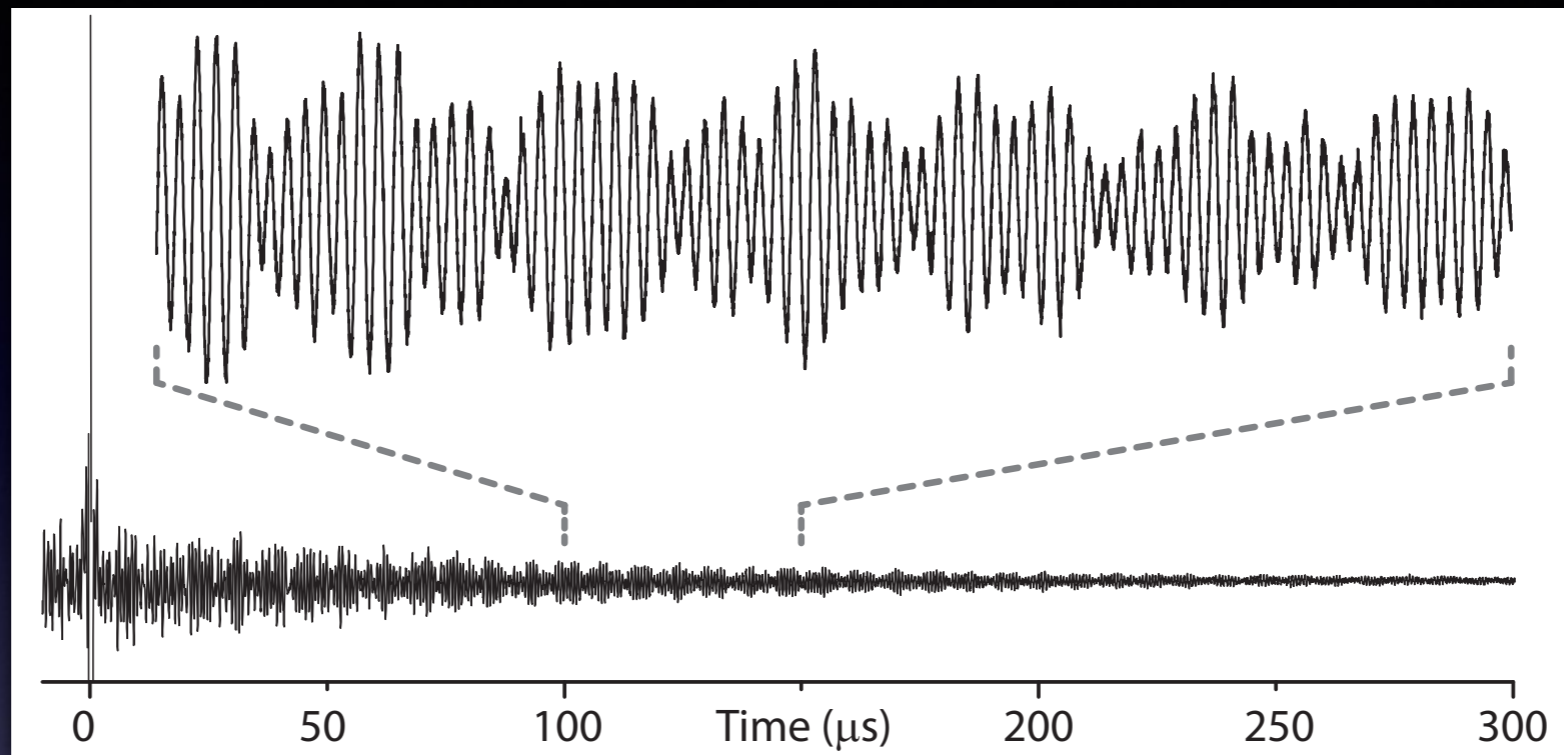
Probe pulse



CARS



Solution 3



Probe pulse

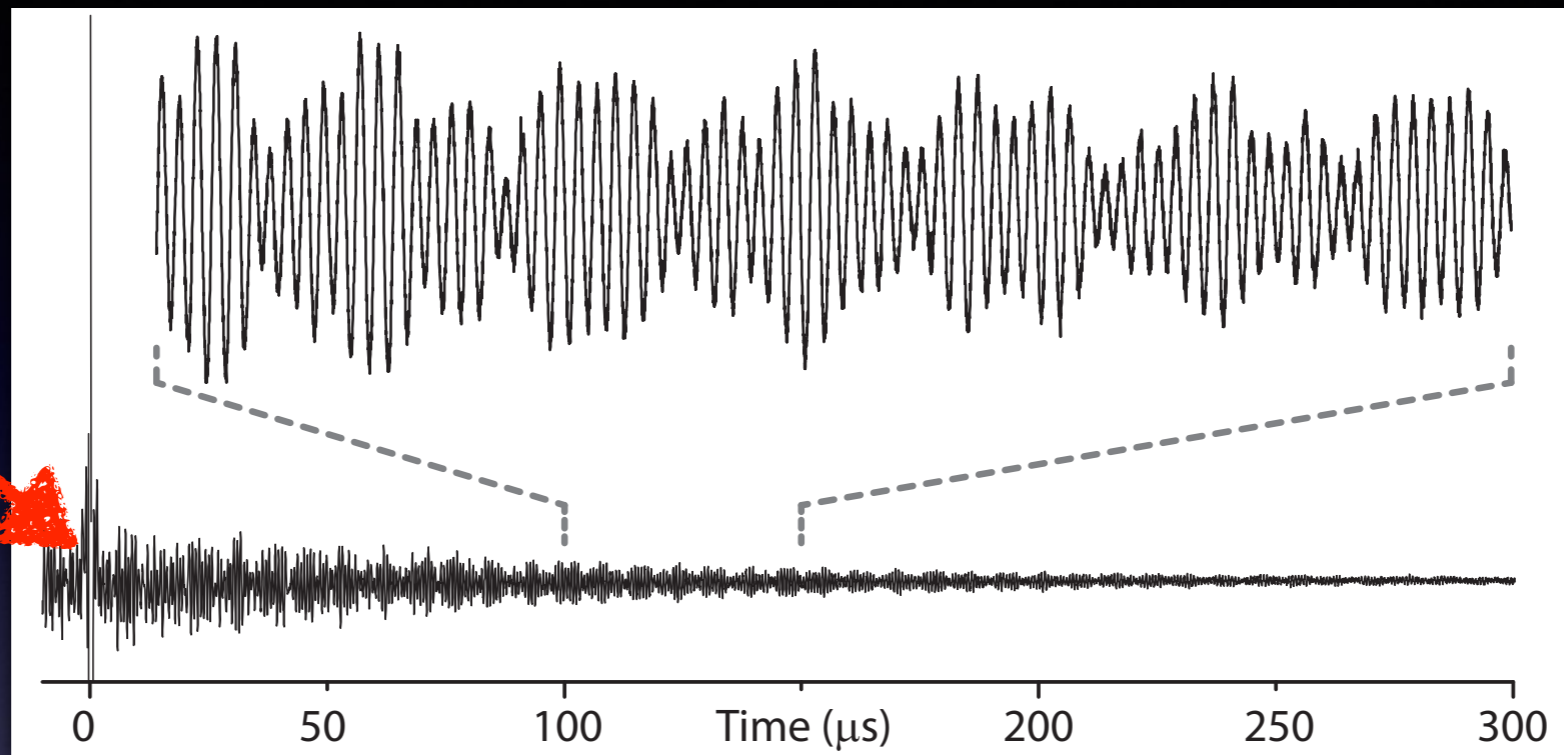


CARS



Solution 3

Pump
Stokes



Probe pulse

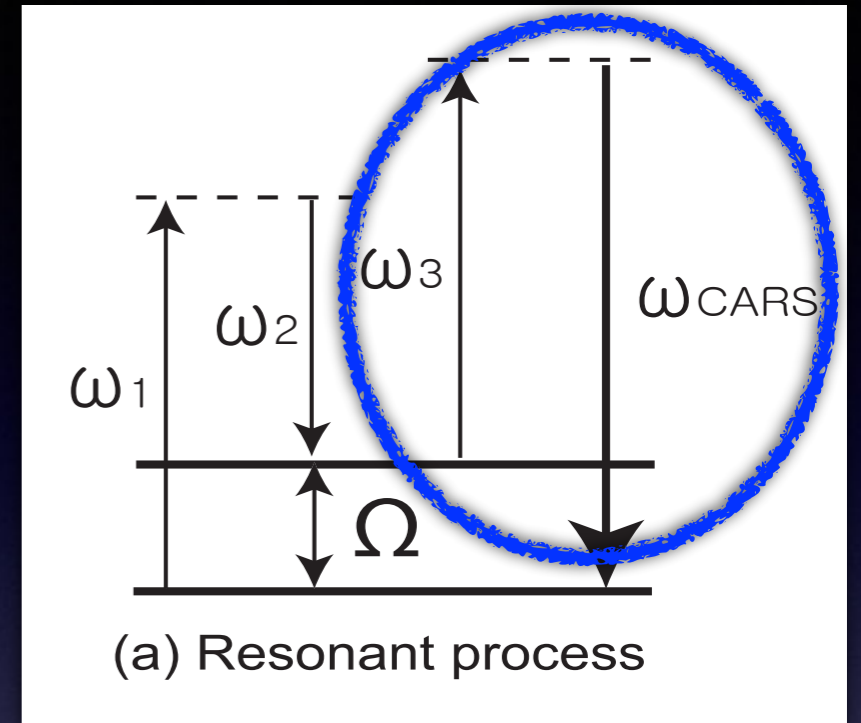


pulse for
modulated CARS
detection

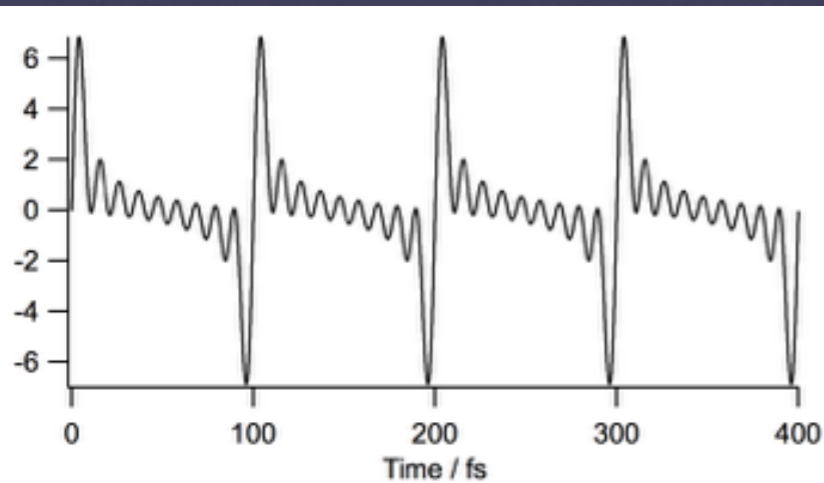


Solution 4

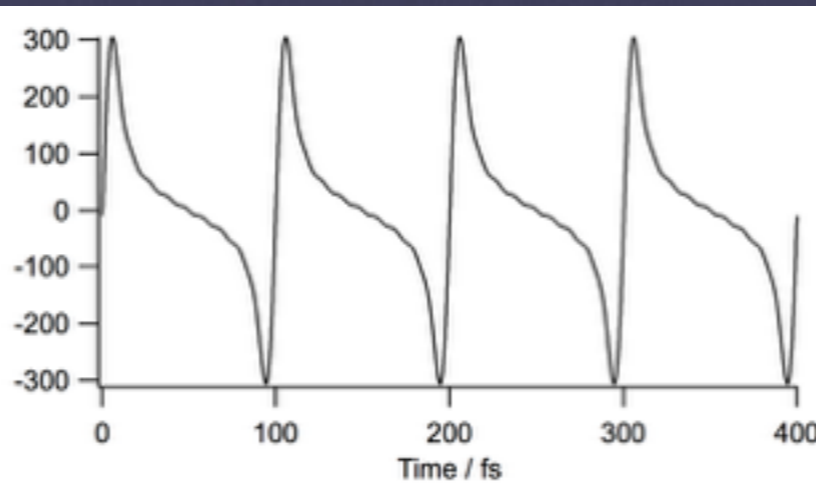
Dual-comb spectroscopy w/ TiS laser
but SHG for excitation



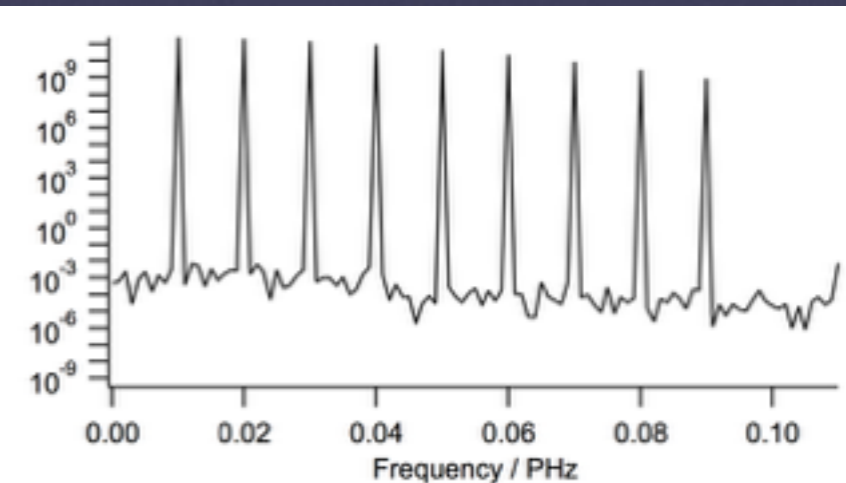
Mol. vib.



Obs. mol. vib.



Fourier transform



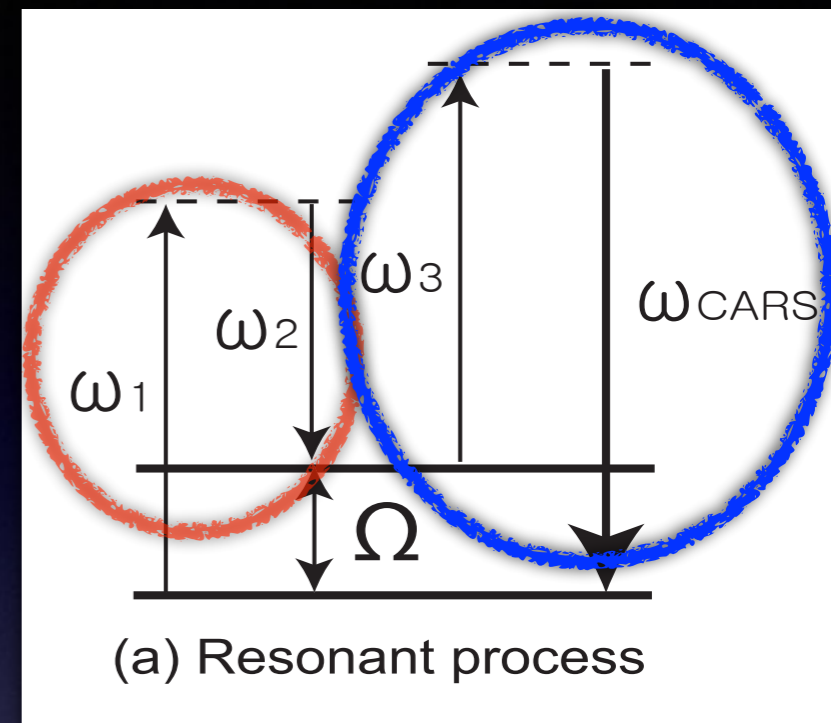
10 THz - 90 THz

Observing with 10 fs laser

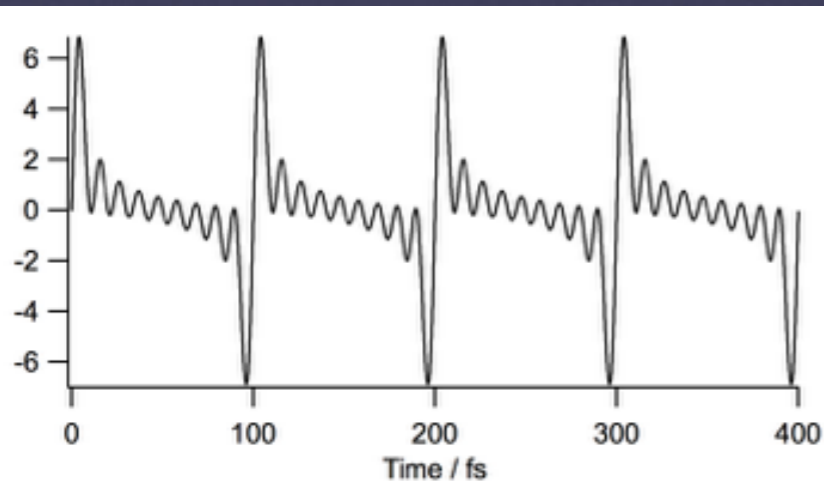
Solution 4

Dual-comb spectroscopy w/ TiS laser
but SHG for excitation

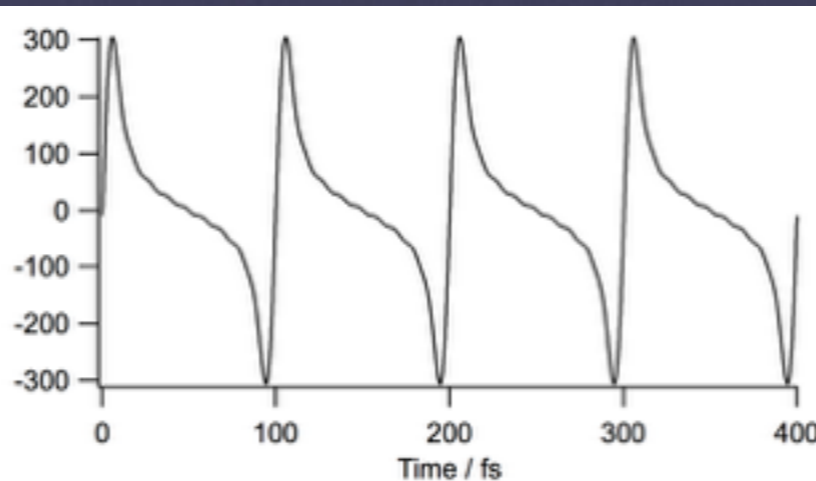
Limit: $< 1500 \text{ cm}^{-1}$



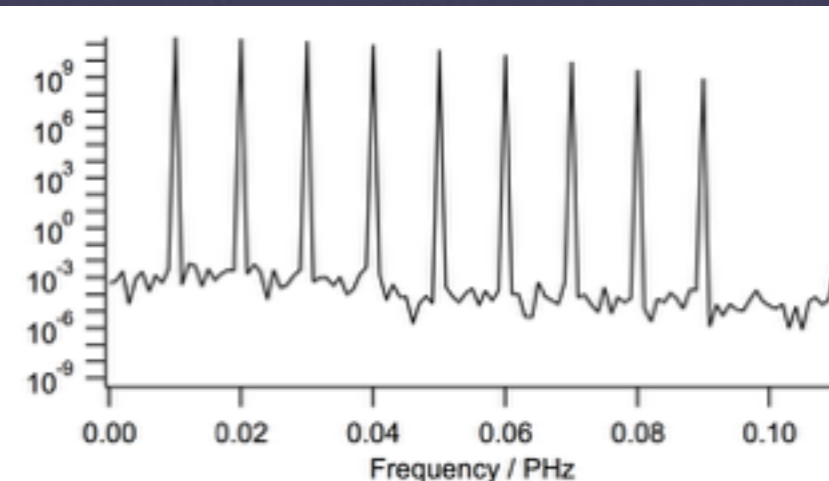
Mol. vib.



Obs. mol. vib.



Fourier transform

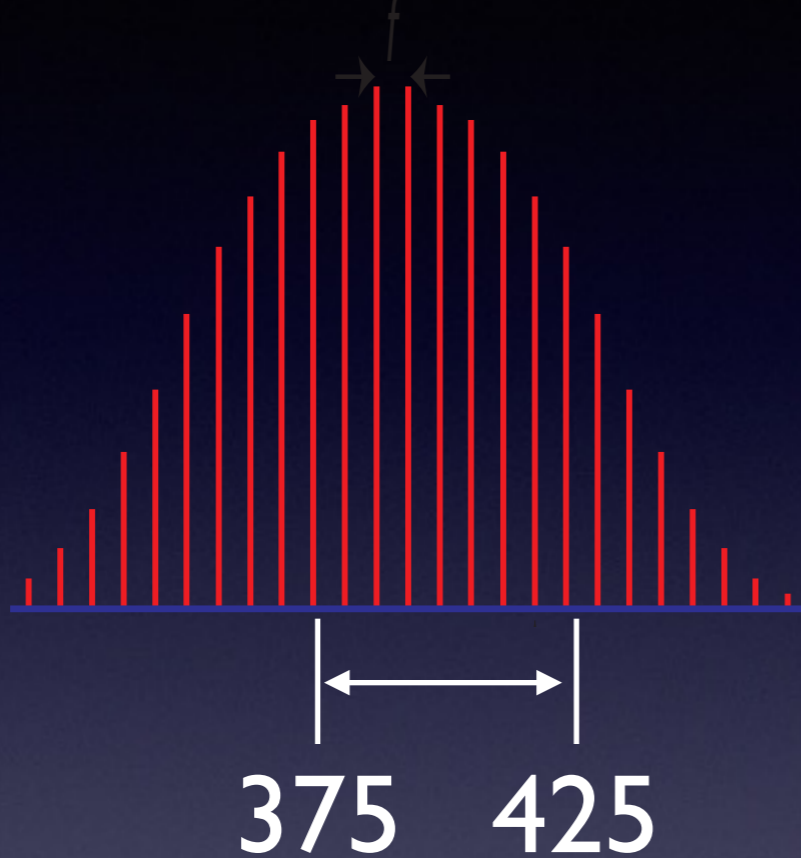


10 THz - 90 THz

Observing with 10 fs laser

Solution 4

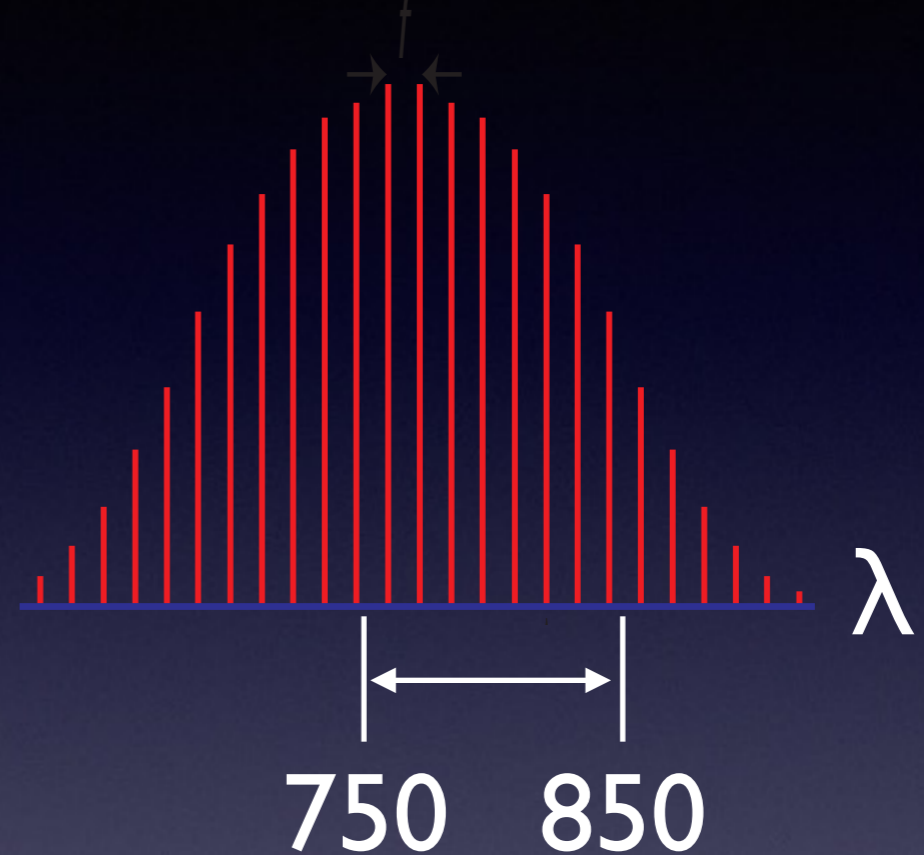
SHG



$$3137 \text{ cm}^{-1}$$

$$\Omega = \frac{1}{\frac{\lambda_1}{2}} - \frac{1}{\frac{\lambda_2}{2}} = 2 \left(\frac{1}{\lambda_1} - \frac{1}{\lambda_2} \right)$$

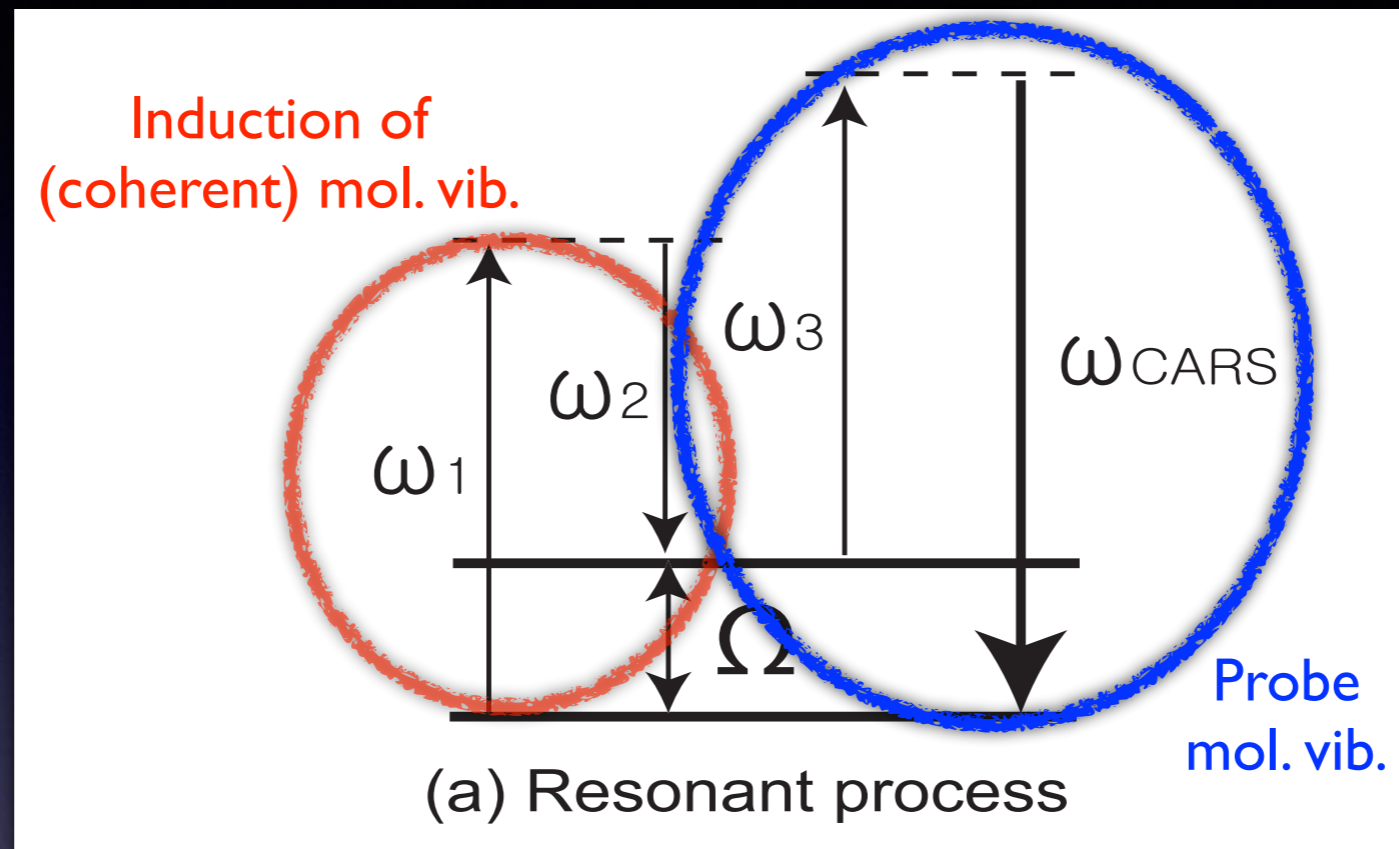
Fundamental



$$1567 \text{ cm}^{-1}$$

$$\Omega = \frac{1}{\lambda_1} - \frac{1}{\lambda_2}$$

Summary



- Solution 1: Two lasers (OPO and femtosource)
- Solution 2: Beats of two vibrations
- Solution 3: Detection from modulated CARS
- Solution 4: Dual-comb w/ Fund. and SHG