Adiabatic Control of CARS in the Presence of Decoherence for Bioimaging

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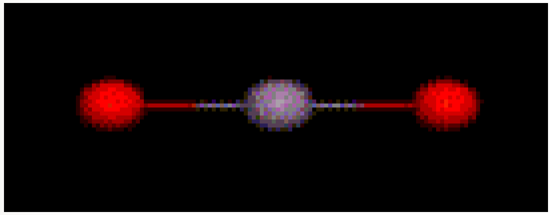
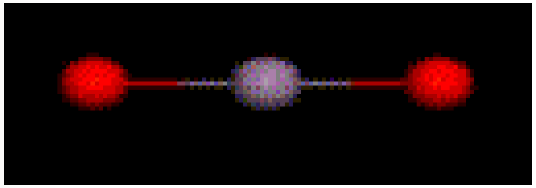
Normal vibrational modes in the CO_2 molecule

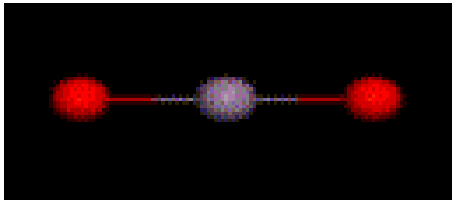
Totally-symmetric stretching mode

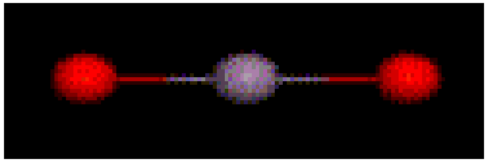


Asymmetric stretching mode

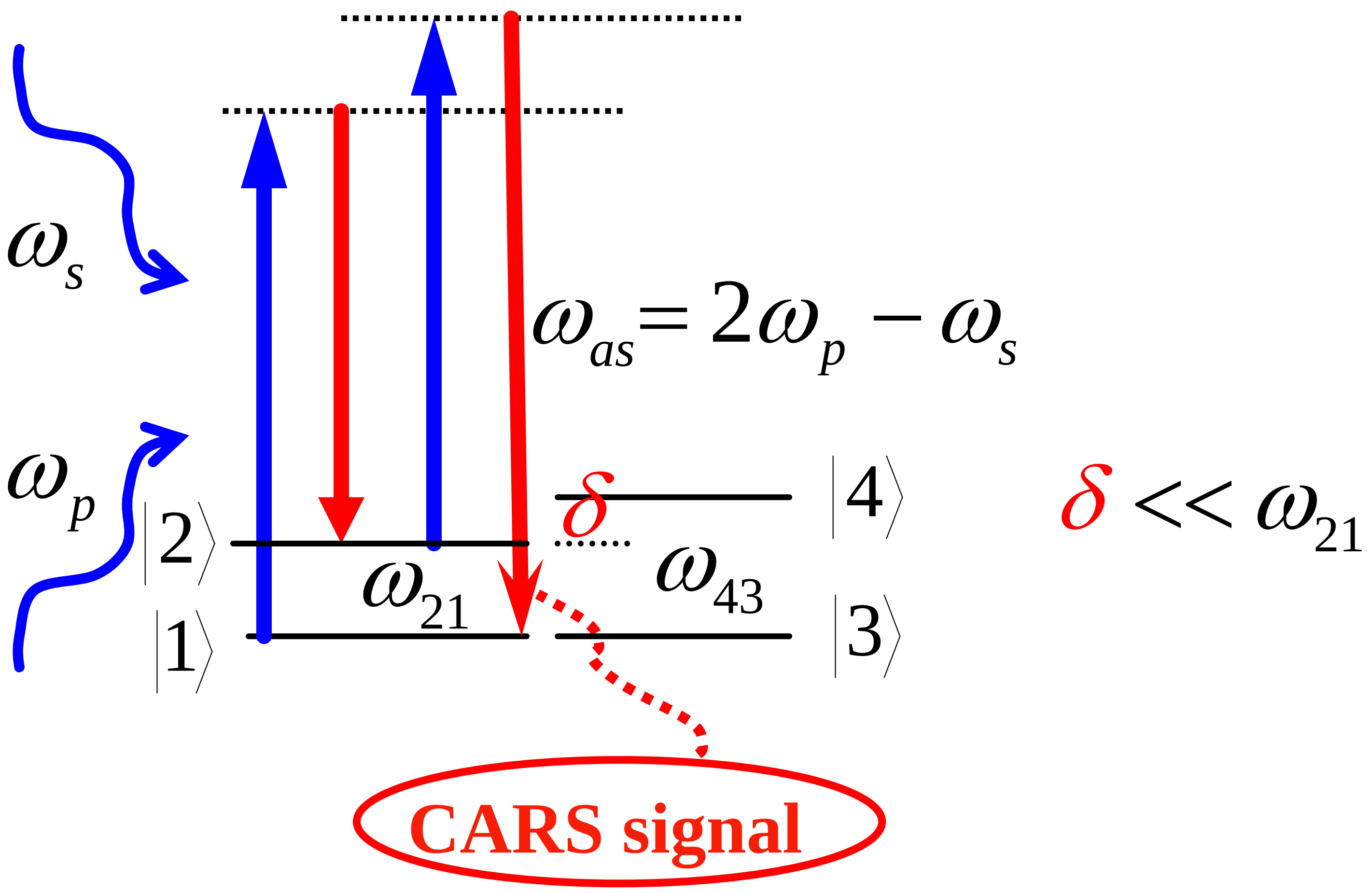


Bending mode (double degenerate)





Coherent Anti-Stokes Raman Scattering Spectroscopy



Noninvasive imaging of biological tissue

CARS microscopy image of mouse ear tissue ***in vivo***. Raman shift is at 2 845 cm^{-1} to address the lipid CH_2 symmetric stretch vibration

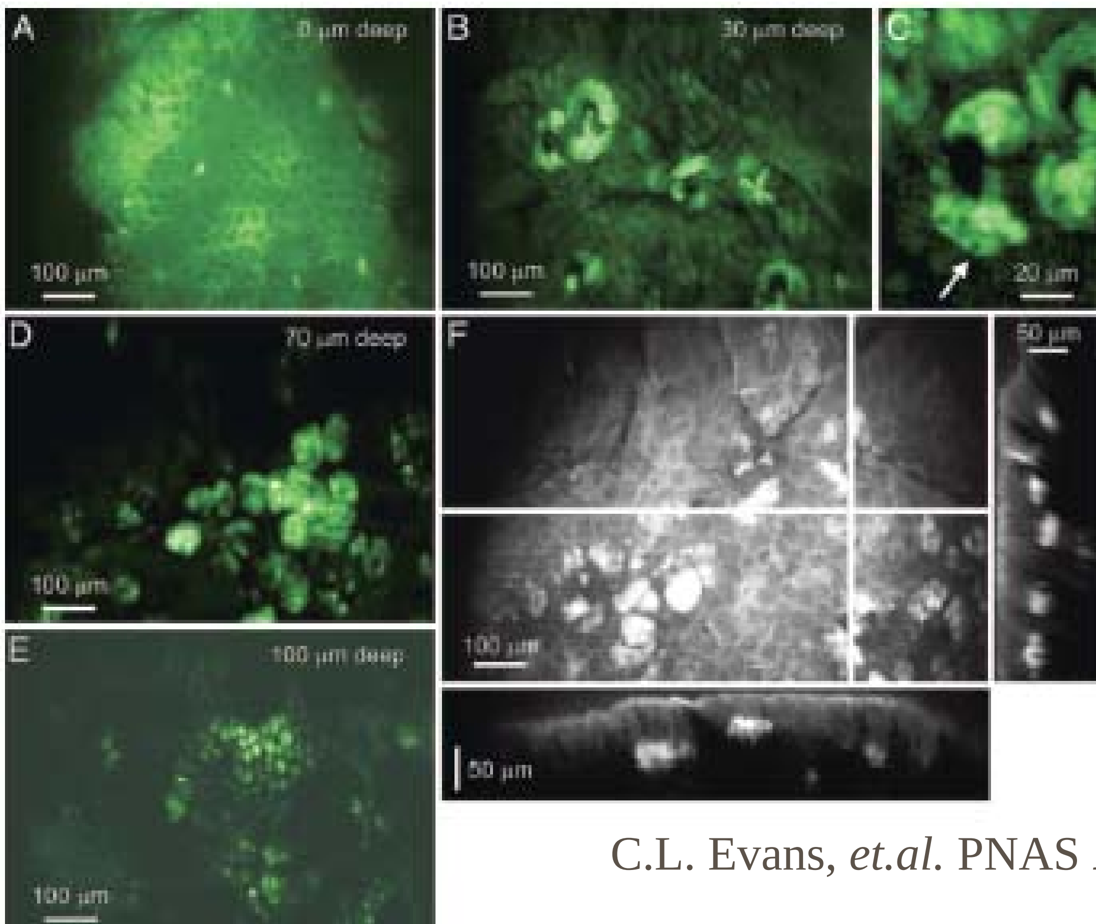
Skin surface, Corneocytes of the stratum corneum

20-40 µm Sebasteous glands

The nuclei of sebum-producing cells

60-80 µm Fat-producing adipocytes

100 µm Smaller adipocytes



3D tissue map:
A 2D overlap, composed of 60 depth-resolved slices, separated by 2 µm

C.L. Evans, *et.al.* PNAS ***102***, *16807 (2005)*

Diffusion of mineral oil through mouse epidermis

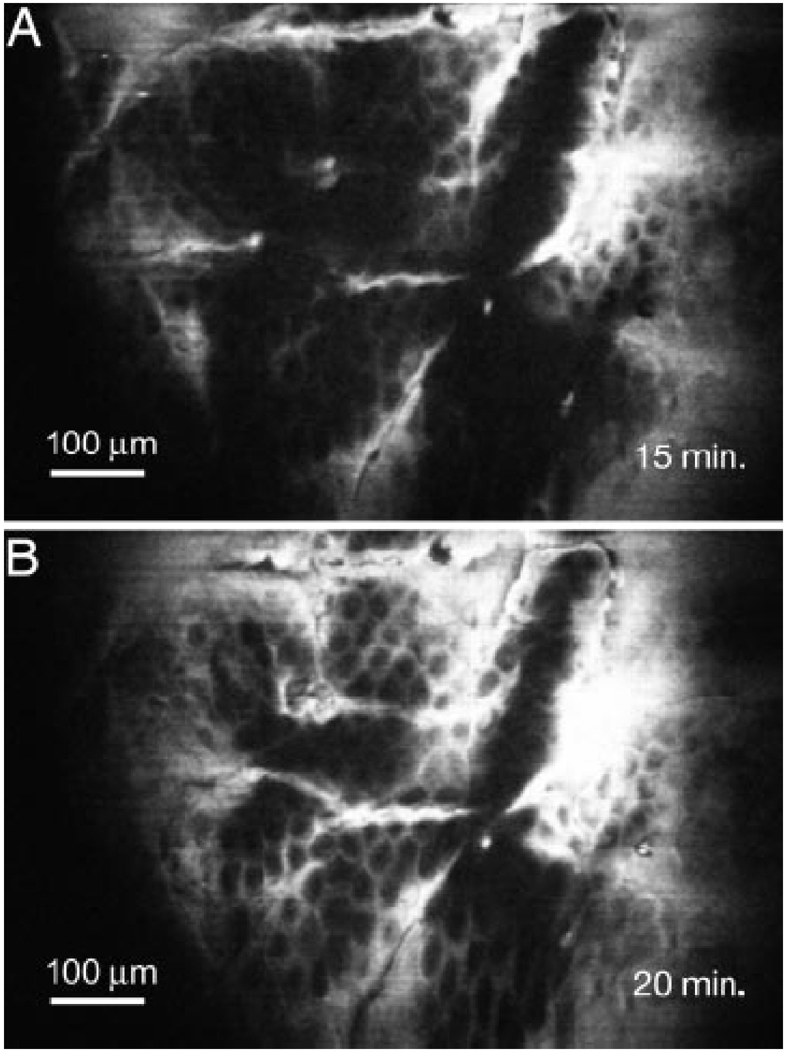
Raman shift equals CH2 stretching vibration

20μm below the surface

externally applied mineral oil penetrates the stratum corneum through the lipid clefts between corneocytes

The same area 5 min later.
Brighter signal indicates a higher oil concentration caused by time-dependent diffusion

C.L. Evans, *et.al.* PNAS ***102***, *16807 (2005)*

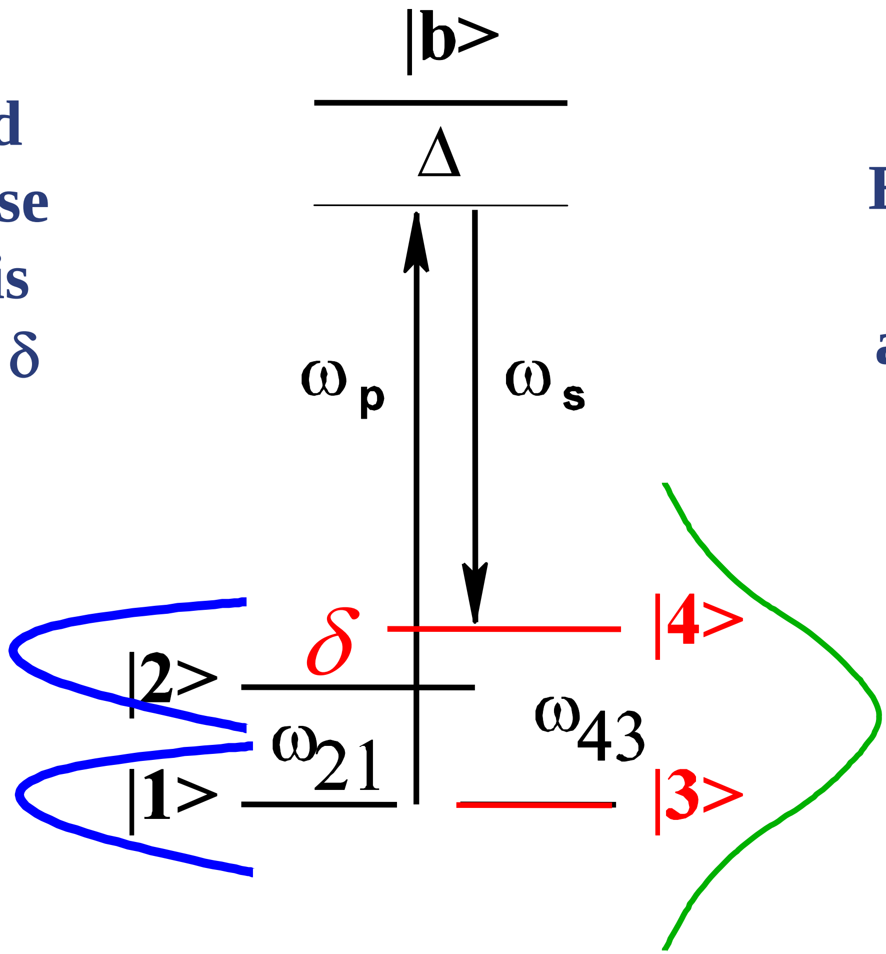


Impulsive and non-impulsive Raman scattering

Non-impulsive

Pump and Stokes pulse duration is about 1 / δ

CH in saturated and unsaturated fats, range 2800-3100 cm $^{-1}$



Impulsive

Exciting pulse duration is about 1 / ω_{43}

Heme proteins, Raman active vibrations range 600-2000 cm $^{-1}$

S.Malinovskaya, P.Bucksbaum, P.Berman, Phys. Rev. A 69, 013801 (2004)

Maxwell-Bloch equations for the field amplitudes

We couple quantum evolution equations for the states to Maxwell equations for the fields to see evolution of the fields.

$$E_{pr} = \frac{1}{2}\left(E_{pr}(z,t)e^{i(k_{pr}z-\omega_{pr}t)} + c.c.\right)$$

$$E_R = \frac{1}{2}\left(E_R(z,t)e^{i(k_R z-\omega_R t)} + c.c.\right)$$

$$\frac{\partial E_{pr}(z,t)}{\partial z} + \frac{1}{c}\frac{\partial E_{pr}(z,t)}{\partial t} = -\frac{k_{pr}}{2\varepsilon_0}\mu_{31}\mu_{23}\frac{E_R(z,t)}{2\hbar\Delta}\operatorname{Im}\{\rho_{12}\}$$

$$\frac{\partial E_R(z,t)}{\partial z} + \frac{1}{c}\frac{\partial E_R(z,t)}{\partial t} = -\frac{k_R}{2\varepsilon_0}\mu_{32}\mu_{13}\frac{E_{pr}(z,t)}{2\hbar\Delta}\operatorname{Im}\{\rho_{21}\}$$

$$\dot{\rho}_{12} = -i\Omega_{12}(\rho_{11}-\rho_{22}) - i(\Delta\omega_{12}-\delta)\rho_{12}$$

$$\dot{\rho}_{22} = -i\Omega_{12}\, 2\operatorname{Im}\{\rho_{12}\}$$

where

$$\boxed{\begin{aligned}&\delta = \omega_{12} - (\omega_{pr} - \omega_R),\ \Omega_{12} = \mu_{13}\mu_{32}\frac{E_R^*(t)E_{pr}(t)}{4\hbar^2\Delta}\\ &\Delta\omega_{12} = -\mu_{13}\mu_{31}\frac{|E_{pr}(t)|^2}{4\hbar^2\Delta} + \mu_{23}\mu_{32}\frac{|E_R(t)|^2}{4\hbar^2\Delta}\end{aligned}}$$

Two-photon transition using linearly chirped femtosecond pulses

$$E_p(t) = E_{p0}(t)\cos\left(\omega_p t + \frac{\alpha t^2}{2}\right)$$

$$E_s(t) = E_{s0}(t)\cos(\omega_s t + \frac{\beta t^2}{2})$$

$$\omega_p - \omega_s = \omega_{21}$$

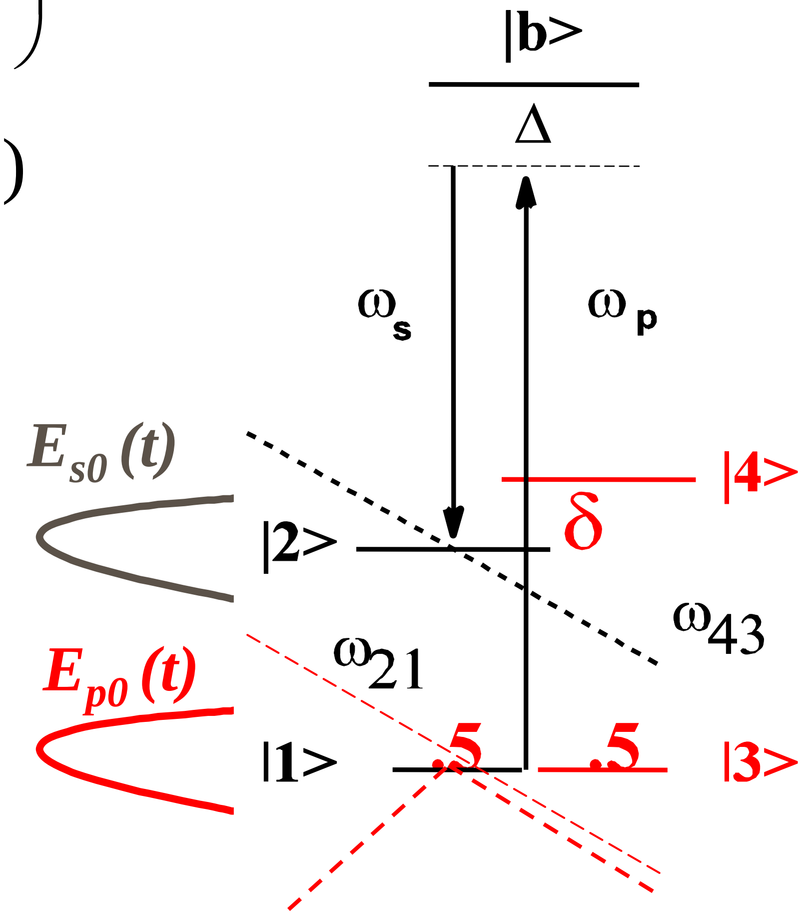
$$\text{for } \alpha = \beta = 0$$

$$I: \quad \alpha = 0, \quad \beta = const$$

$$II: \quad \beta = \alpha$$

$$III: \quad \beta = -\alpha \qquad t < t_c$$

$$\beta = \alpha \qquad t \geq t_c$$



Hamiltonian in the field interaction representation

$$\hat{H}_{int}=\begin{pmatrix} 1/2\left(\delta+(\beta-\alpha)t+\Omega_1(t)-\Omega_2(t)\right) & \Omega_3(t) \\ \Omega_3(t) & -1/2\left(\delta+(\beta-\alpha)t+\Omega_1(t)-\Omega_2(t)\right) \end{pmatrix}$$

Energy separation of the dressed states

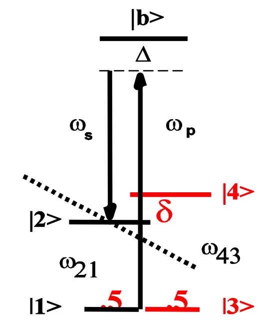
$$\Omega(t)=\sqrt{\left(\delta+(\beta-\alpha)t+\Omega_1(t)-\Omega_2(t)\right)^2+4\Omega_3(t)^2}$$

Probability amplitudes of dressed states

$$\dot{a}_{1(3)}=-{}^{i}\!/_{2}\,\Omega(t)\,a_{1(3)}+\dot{\Theta}(t)a_{2(4)}$$

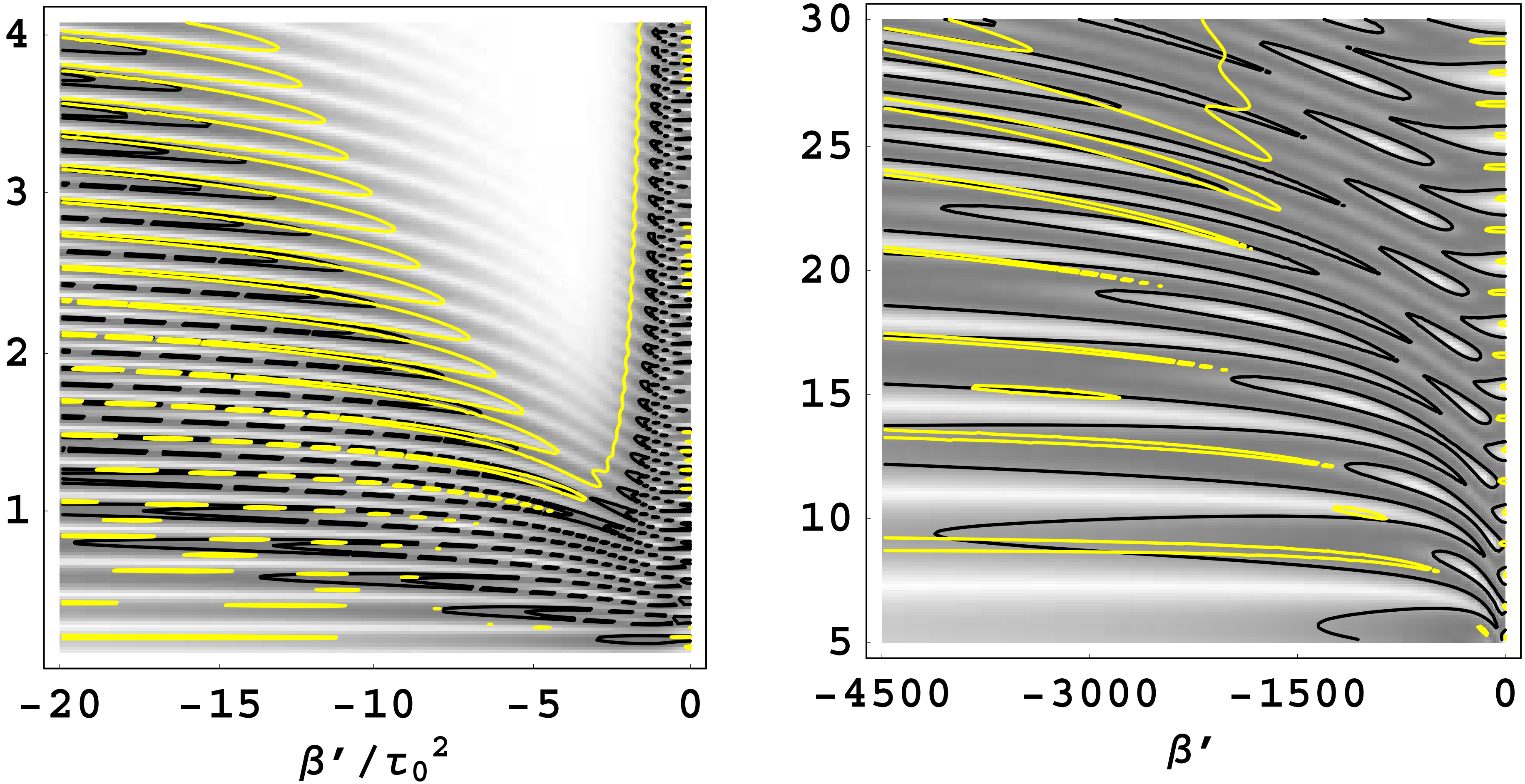
$$\dot{a}_{2(4)}={}^{i}\!/_{2}\,\Omega(t)\,a_{2(4)}-\dot{\Theta}(t)\,a_{1(3)}$$

Contour plots of coherences as a function of the peak effective Rabi frequency (a) and the duration of a transform-limited pulse (b) and a spectral chirp



Ω_3 5.10^7-3.10^8 V/cm (a)

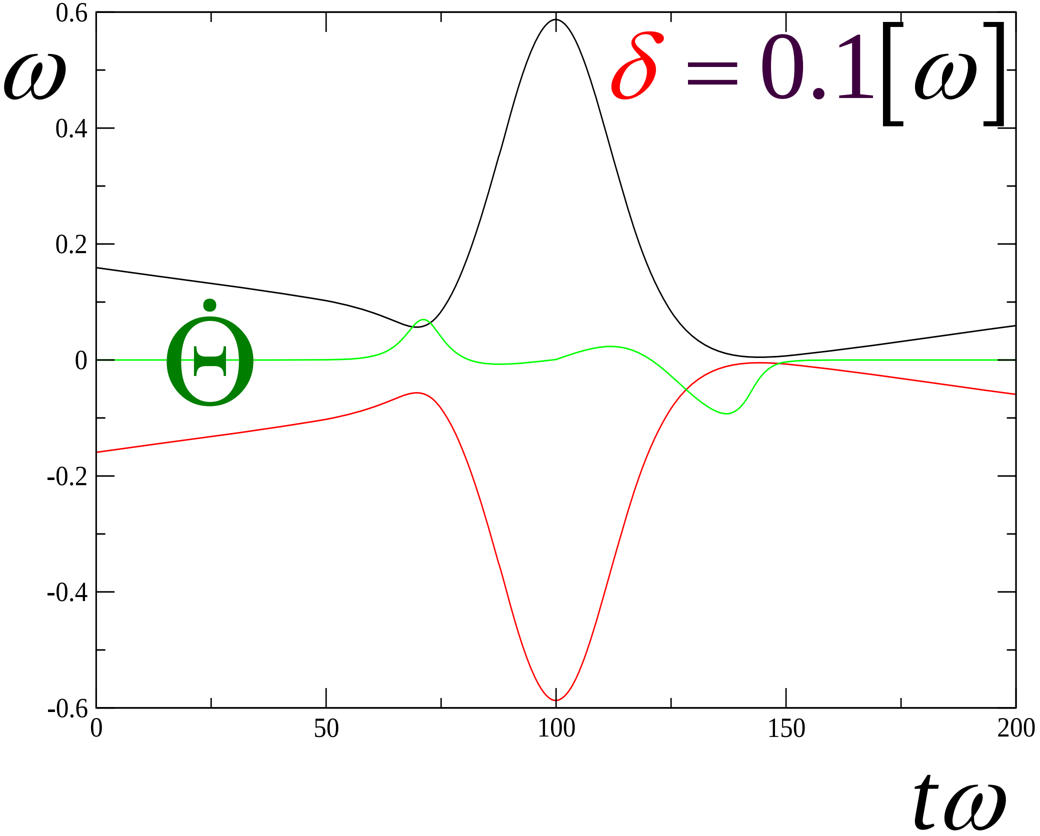
τ_0 (b)

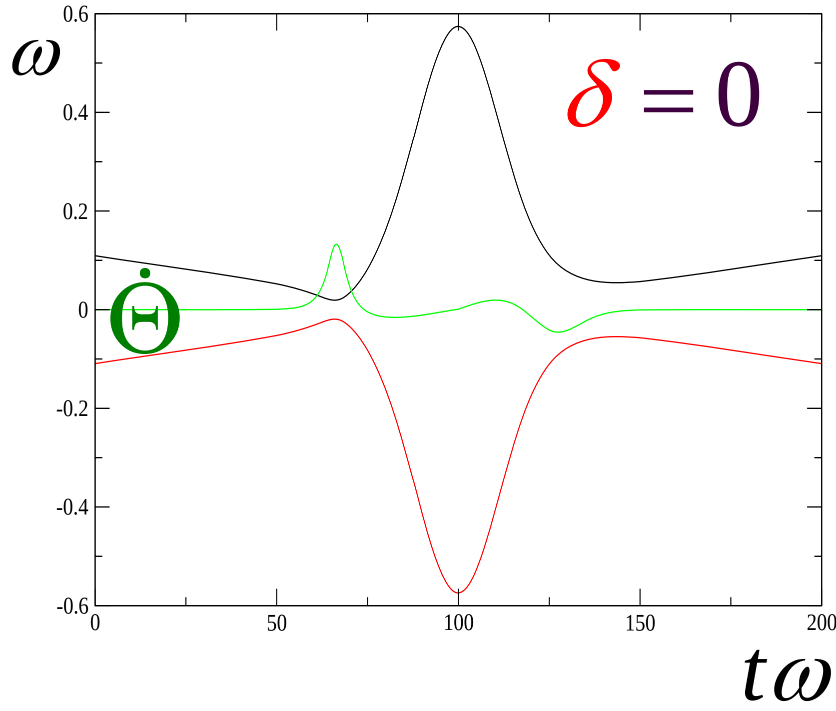


$|\rho_{12}|$=0.24, $|\rho_{34}|$=0.05 (yellow)

S.A. Malinovskaya, Phys. Rev. A **73**, 033416 (2006)

Energy separation of dressed states and coupling parameter $\dot{\Theta}$ as a function of time for Raman transition

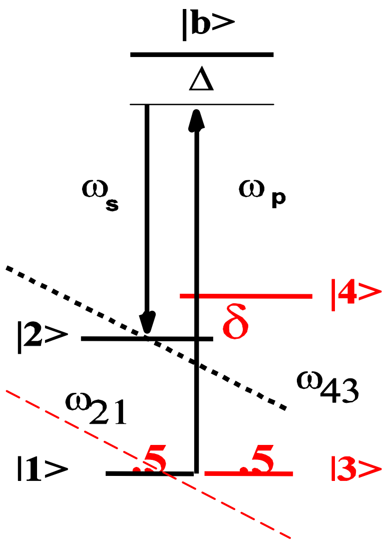


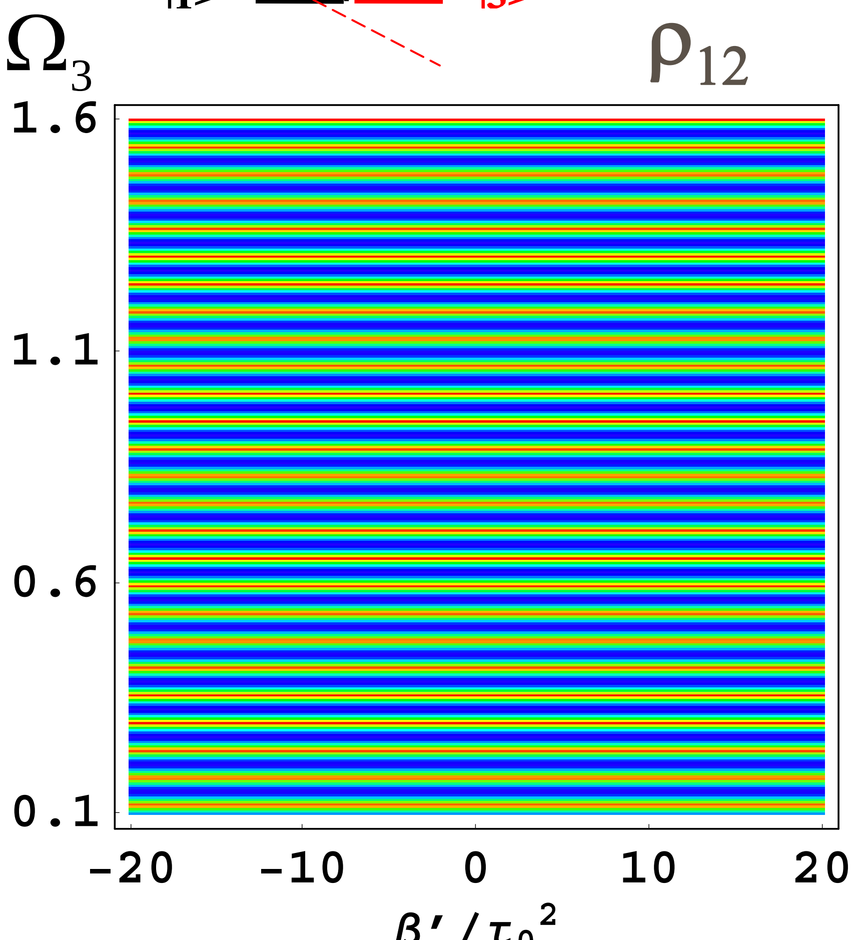


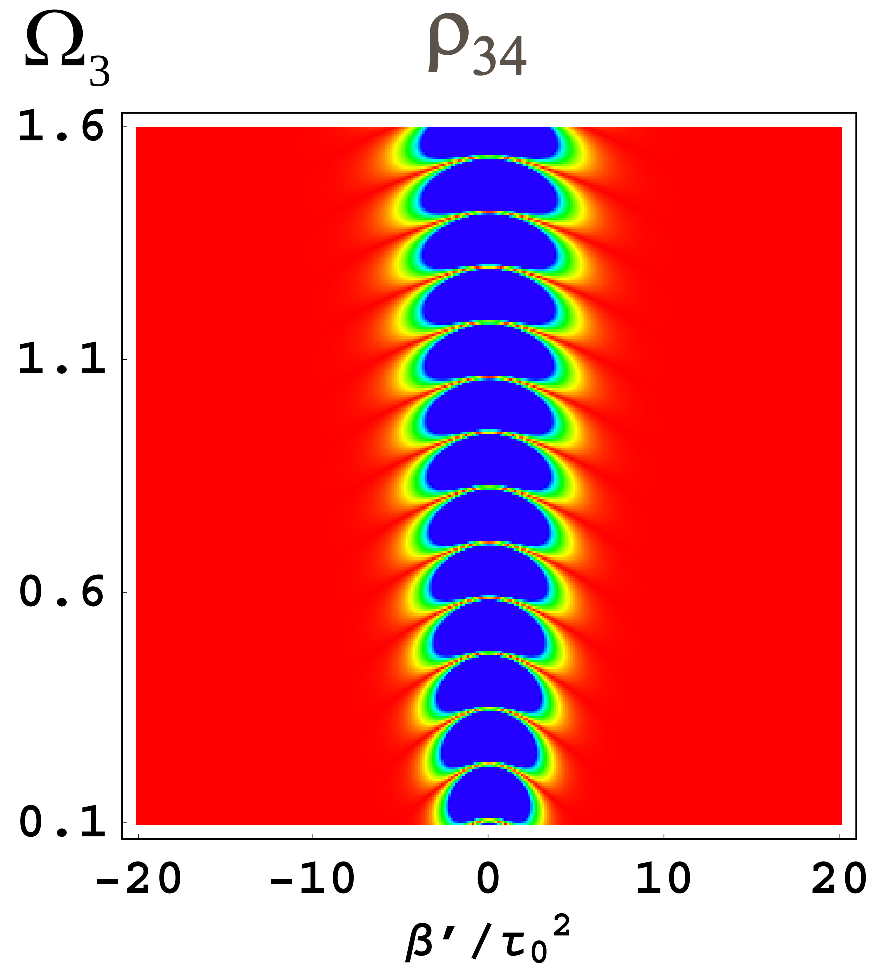
$$\Omega_0 = .7[\omega]\ \beta = .002\left[\omega^2\right]\ \tau_0 = 15.\left[\omega^{-1}\right]$$

Density plots of coherence as a function of the peak effective Rabi frequency and the spectral chirp,

(the same for pump and Stokes pulse)

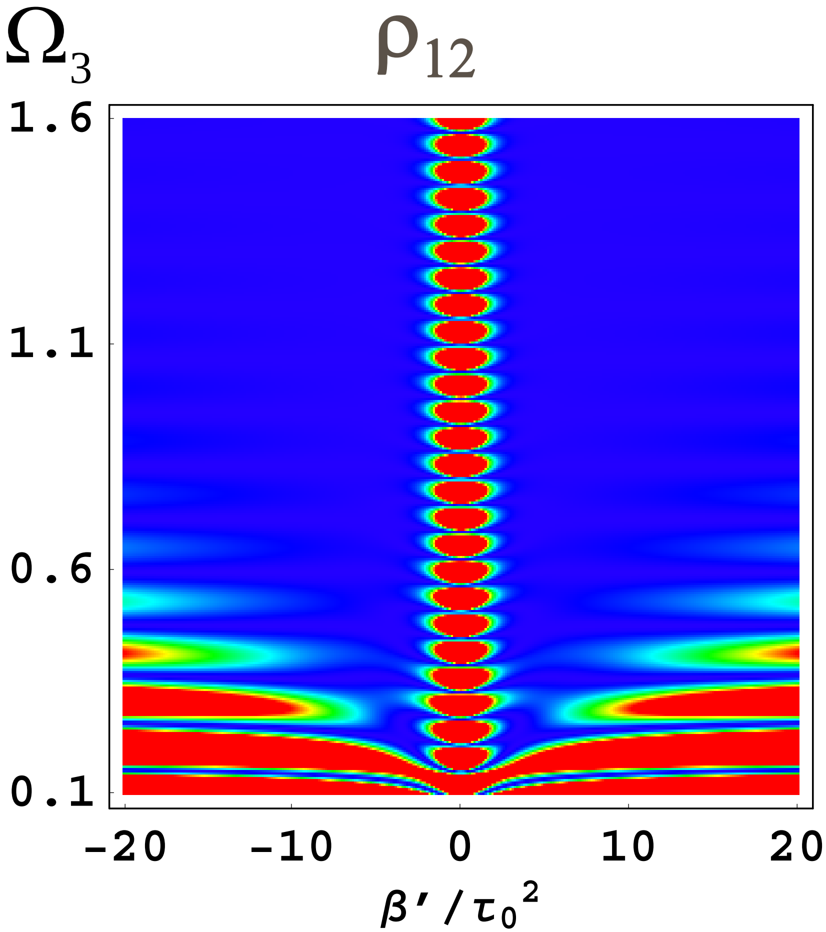


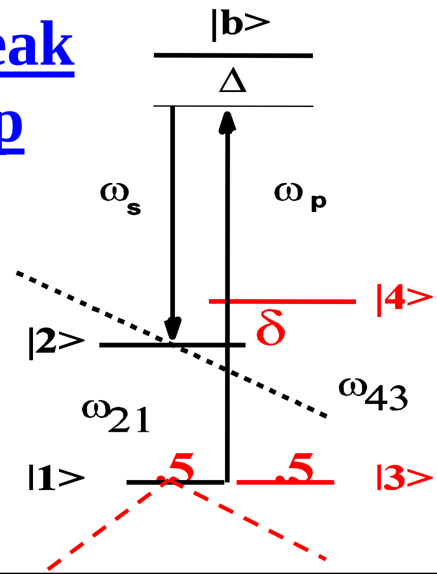


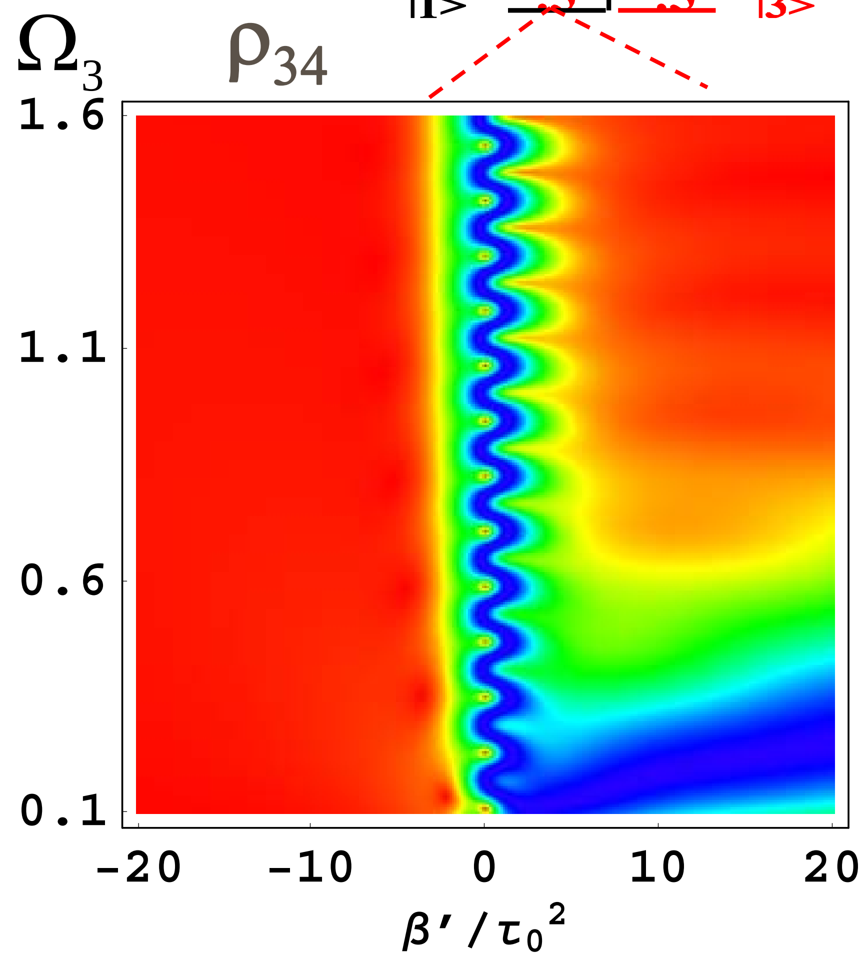


Blue $\rho = \rho_{\max}$ **, red** $\rho = 0.$

Density plots of coherence as a function of the peak effective Rabi frequency and the spectral chirp



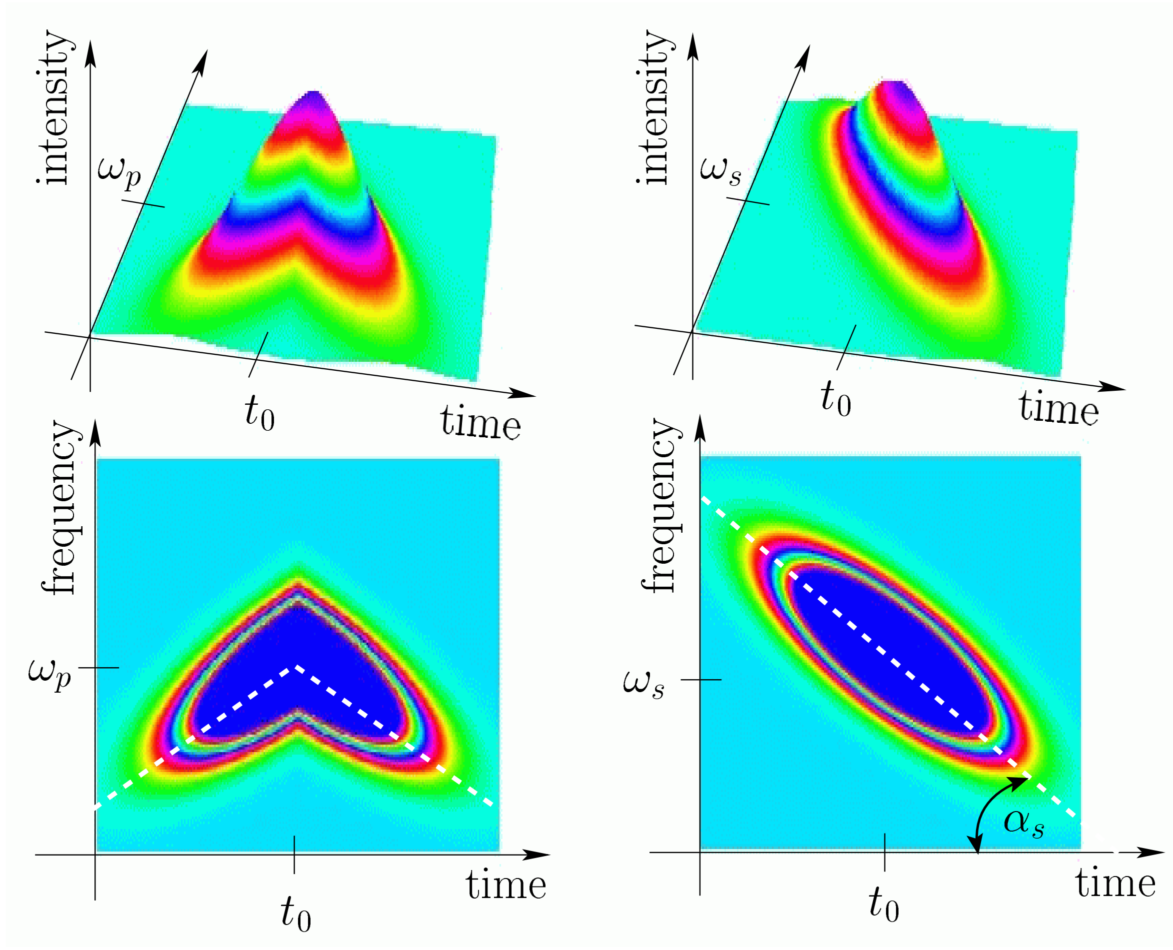




Blue $\rho = \rho_{max}$, **red** $\rho = 0.$

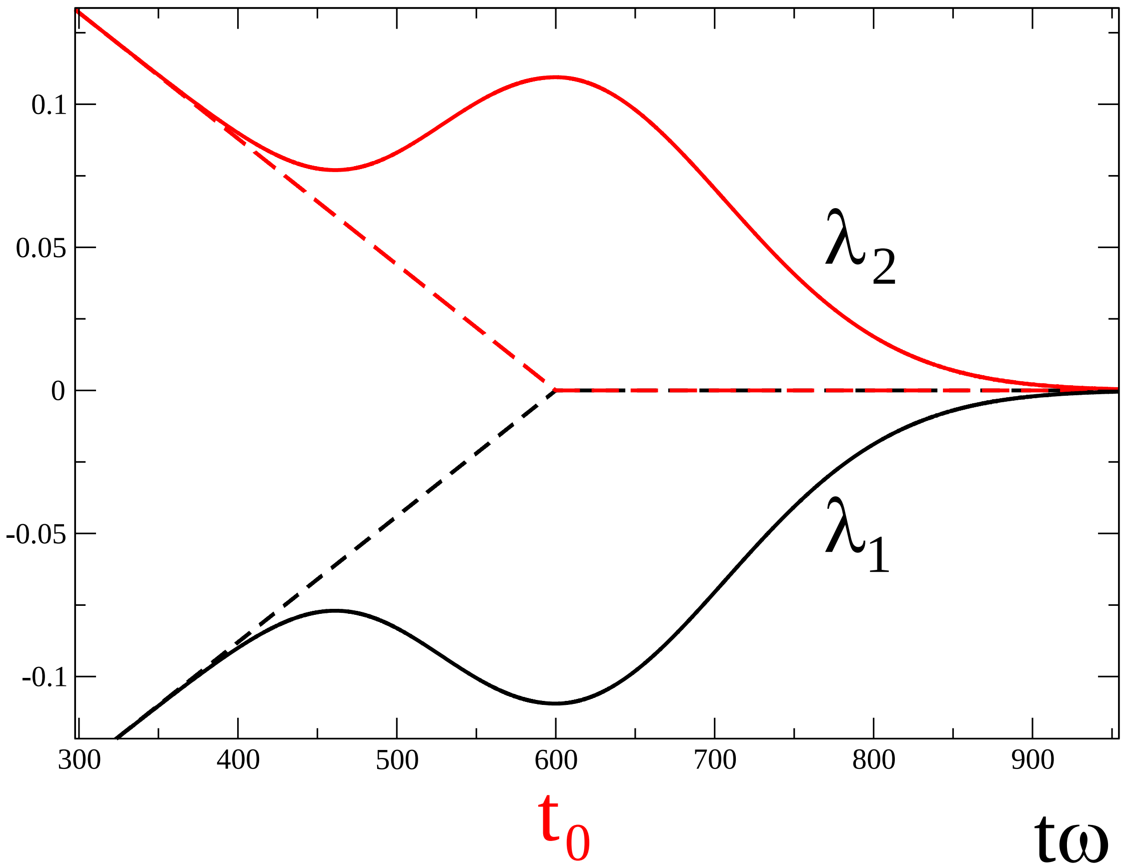
S.A. Malinovskaya, V.S. Malinovsky, Opt. Lett. 32, 76025 (2007)

The Wigner presentation of the pump (left panel) and Stokes (right panel) pulses.



The positive and negative slopes of white dashed lines on the density plots correspond to an upward and a downward frequency chirps.

Dressed-state picture: resonant two-level system, strong fields.



$t < t_0$

$$\hat{H}_{12} = \begin{pmatrix} \beta(t-t_0) & \Omega_3 \\ \Omega_3 & -\beta(t-t_0) \end{pmatrix}$$

$$\lambda_{1,2} = \pm\sqrt{\beta^2(t-t_0)^2 + \Omega_3^2}$$

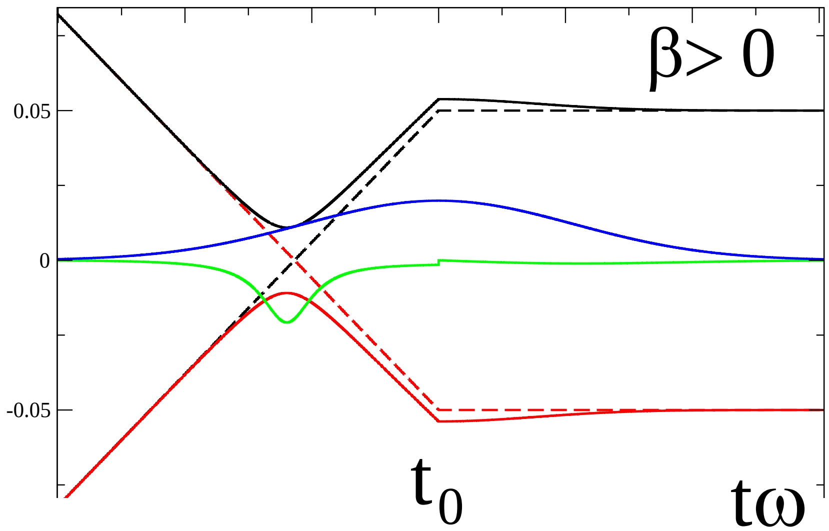
$t \geq t_0$

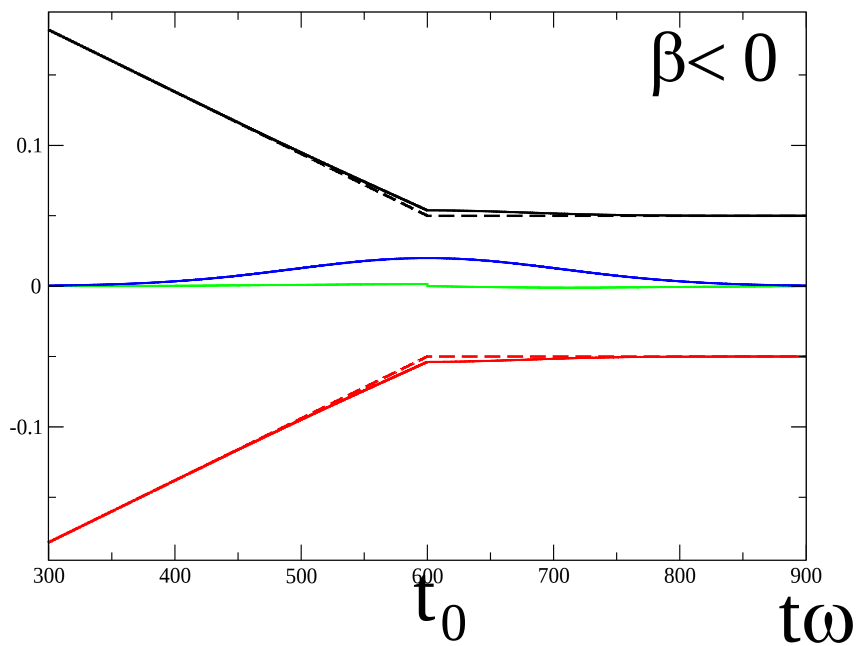
$$\hat{H}_{12} = \begin{pmatrix} 0 & \Omega_3 \\ \Omega_3 & 0 \end{pmatrix}$$

$$\lambda_{1,2} = \pm\Omega_3$$

$$\Omega_3 = 1.1, \tau_0 = 15, \ \beta'/\tau_0^2 = \pm 10$$

Dressed-state picture:
off-resonant two-level system, weak fields.





$t < t_0$

$$\hat{H}_{12} = \begin{pmatrix} \delta/2 + \beta(t - t_0) & \Omega_3 \\ \Omega_3 & -\delta/2 - \beta(t - t_0) \end{pmatrix}$$

$$\lambda_{1,2} = \pm\sqrt{(\delta/2 + \beta(t - t_0))^2 + \Omega_3^2}$$

$t \geq t_0$

$$\hat{H}_{12} = \begin{pmatrix} \delta/2 & \Omega_3 \\ \Omega_3 & -\delta/2 \end{pmatrix}$$

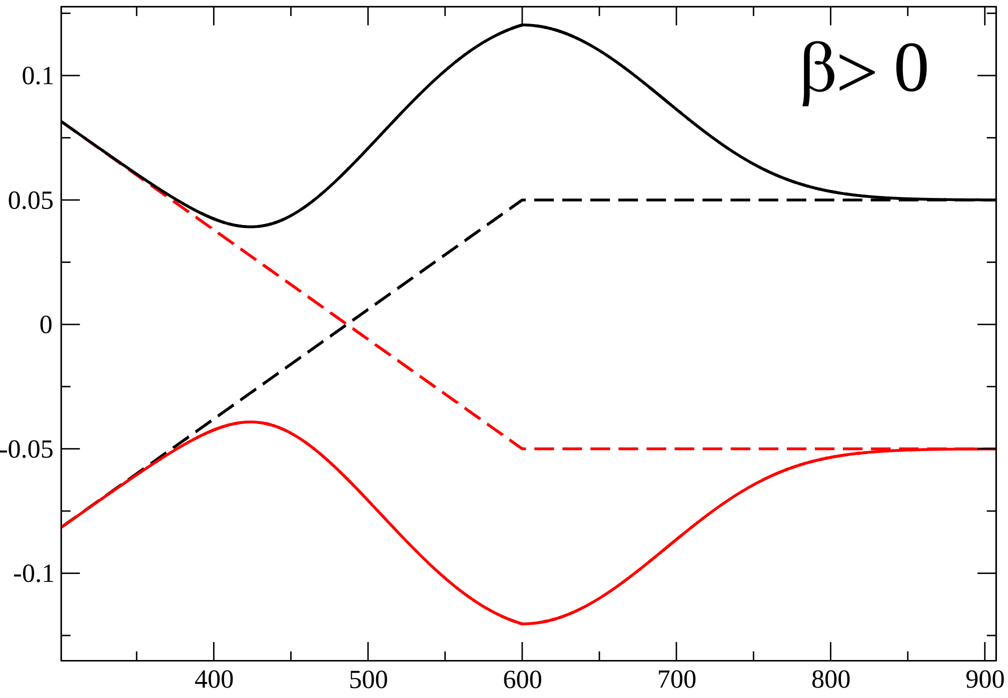
$$\lambda_{1,2} = \pm\sqrt{\delta^2/4 + \Omega_3^2}$$

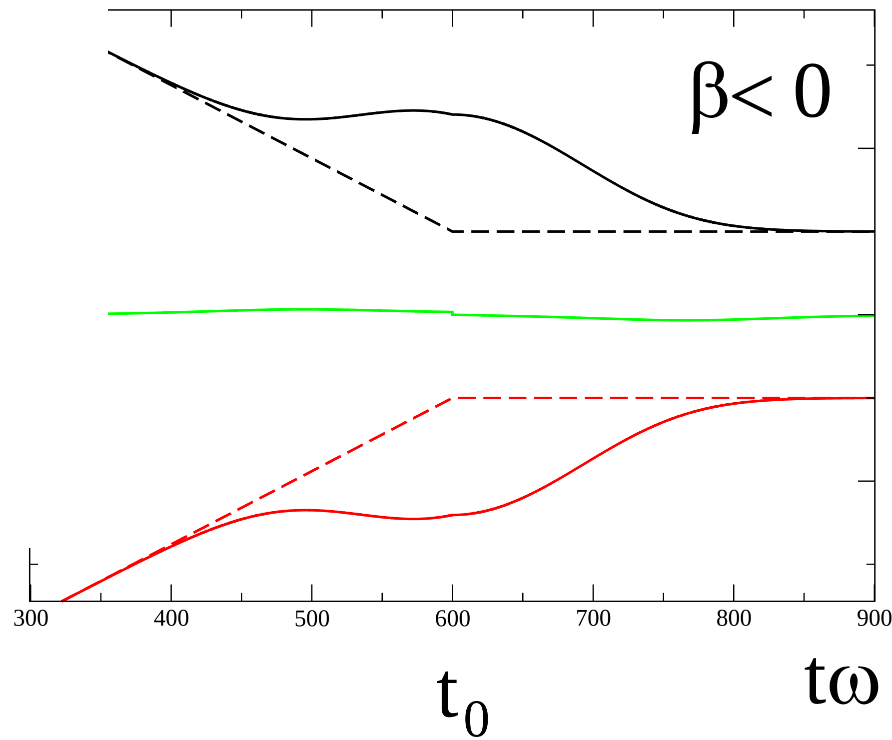
$$\Omega_3 = 0.2,\ \tau_0 = 15,\ \beta'/\tau_0^2 = \pm 10$$

Dressed-state picture: off-resonant two-level system, strong fields.

$$\Omega_3 = 1.1, \tau_0 = 15$$

$$\beta' / \tau_0^2 = \pm 10$$





Dressed State Analysis

$$\left|I\right\rangle = \cos\Theta\left|1\right\rangle + \sin\Theta\left|2\right\rangle$$

$$\cos\Theta = \left(\frac{1}{2}\left(1 + \frac{\delta + (\alpha - \beta)t}{(4\Omega_3^2 + (\delta + (\alpha - \beta)t)^2)^{1/2}}\right)\right)^{1/2}$$

$$\sin\Theta = \left(\frac{1}{2}\left(1 - \frac{\delta + (\alpha - \beta)t}{(4\Omega_3^2 + (\delta + (\alpha - \beta)t)^2)^{1/2}}\right)\right)^{1/2}$$

$$t \approx 0, \alpha = -\beta, \delta = 0, \cos\Theta = 1, \sin\Theta = 0$$

$$t \to \infty, \alpha = \beta, \delta = 0, \cos\Theta = \frac{1}{\sqrt{2}}, \sin\Theta = \frac{1}{\sqrt{2}}$$

$$t \approx 0, \alpha = -\beta, \delta \neq 0, \cos\Theta = 1, \sin\Theta = 0$$

$$t \to \infty, \alpha = \beta, \delta \neq 0, \cos\Theta = 1, \sin\Theta = 0$$

Method Advantages:

- Our proposed method magnifies CARS signal by *three orders of magnitude* as compared to current state of art methods
- Molecular selectivity without need for labeling
- Signal directionality
- Increased signal-to-noise ratio
- Low excitation power
- 3D resolution

Vibrational energy relaxation and collisional dephasing in the chirped pulse adiabatic passage in CARS

- Using Liouvile von Neuman equation for the time evolution of the density matrix and adding reduced matrix elements to account for the decoherence, we get

$$\dot{\rho}_{11} = -2\Omega_3 \operatorname{Im}\{\rho_{21}\} + \gamma_2 \rho_{22}$$

$$\dot{\rho}_{22} = 2\Omega_3 \operatorname{Im}\{\rho_{21}\} - \gamma_2 \rho_{22}$$

$$\dot{\rho}_{12} = i(\delta + (\beta - \alpha)t + \Omega_1 - \Omega_2)\rho_{12} + i\Omega_3(\rho_{22} - \rho_{11}) - (\frac{\gamma_2}{2} + \Gamma)\rho_{12}$$

$$\dot{\rho}_{21} = i(\delta + (\beta - \alpha)t + \Omega_1 - \Omega_2)\rho_{21} + i\Omega_3(\rho_{22} - \rho_{11}) - (\frac{\gamma_2}{2} + \Gamma)\rho_{21}$$

Γ, γ_2 Dependence of Coherence ρ_{12}

- **For γ_2=0, increasing Γ (decreasing collisional dephasing time from ∞**

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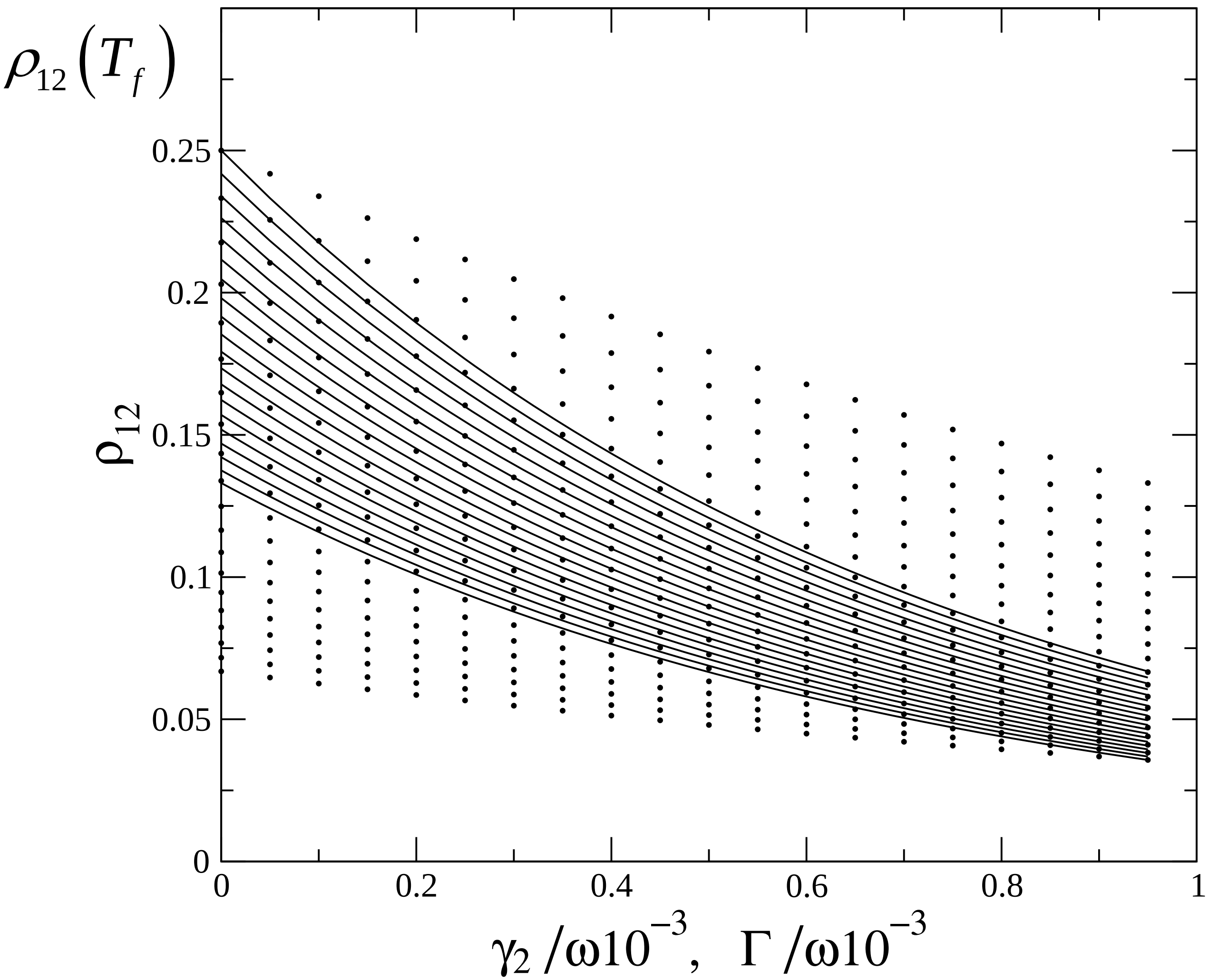

om
THz

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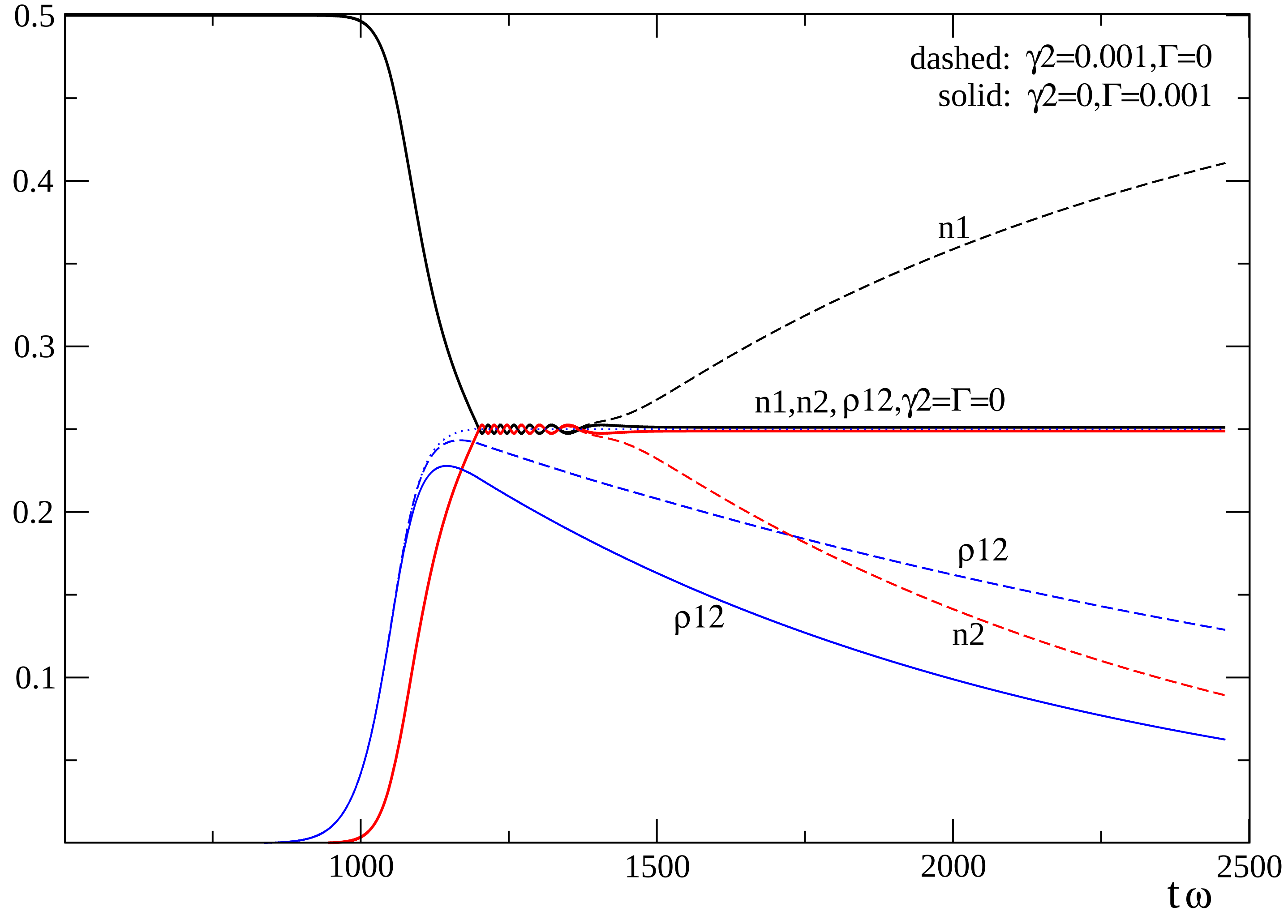
THz

Time-dependent picture of coherence in the presence of



energy relaxation and does not change population distribution

Dressed State Equations in Adiabatic Approximation

- Adiabatic approximation is valid in case γ_2, Γ are much less than Ω_3. Then ρ_{12}^d and ρ_{21}^d are approximately zero.

$$\dot{\rho}_{11}^d = -(\gamma_2 \sin^2\theta + \tfrac{1}{2}(\Gamma - \frac{\gamma_2}{2})\sin^2 2\theta)\rho_{11}^d + (\gamma_2 \cos^4\theta + \tfrac{1}{2}\Gamma \sin^2 2\theta)\rho_{22}^d$$

$$\dot{\rho}_{22}^d = (\gamma_2 \sin^4\theta + \tfrac{1}{2}\Gamma \sin^2 2\theta)\rho_{11}^d - (\gamma_2 \cos^2\theta + \tfrac{1}{2}(\Gamma - \frac{\gamma_2}{2})\sin^2 2\theta)\rho_{22}^d$$

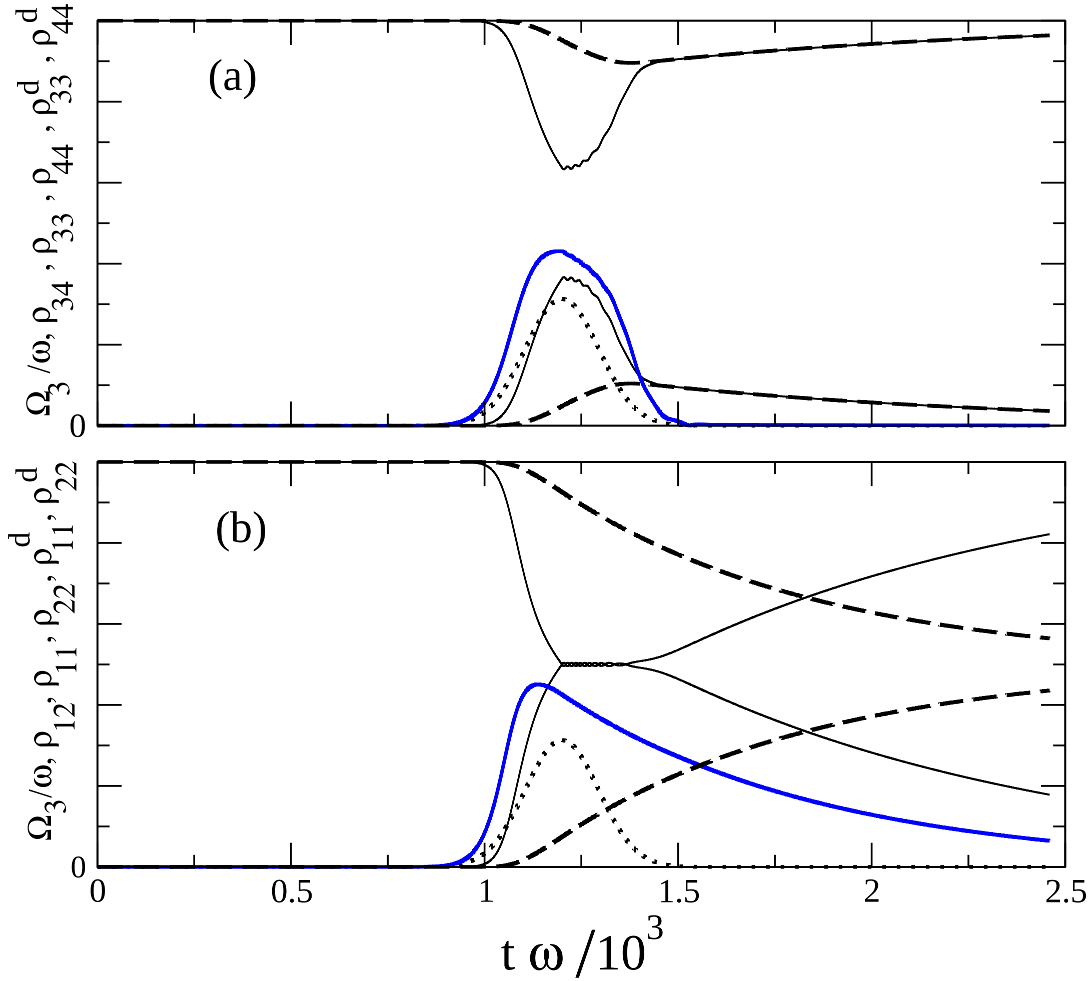
$$\sin\theta = \sqrt{\frac{1}{2}\left(1 - \frac{R_0}{R}\right)} \quad \cos\theta = \sqrt{\frac{1}{2}\left(1 + \frac{R_0}{R}\right)}$$

$$R_0 = \delta + (\beta - \alpha)t + \Omega_1(t) - \Omega_2(t)$$

$$R = \sqrt{\left(\delta + (\beta - \alpha)t + \Omega_1(t) - \Omega_2(t)\right)^2 + 4\Omega_3(t)^2}$$

$\gamma_2 = \Gamma$

$= 85 \cdot 10^{-3} T$



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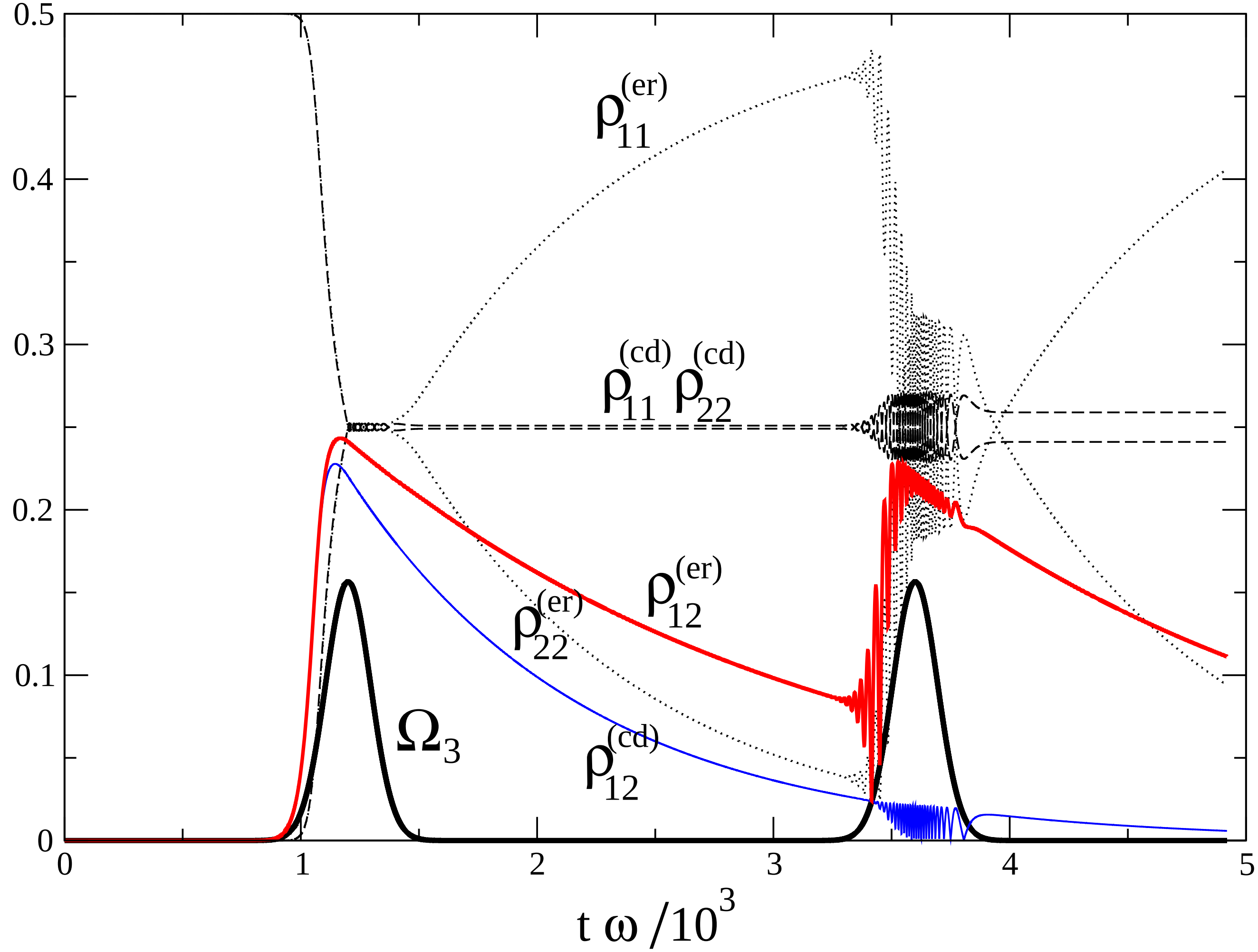
is nonzero and the dressed states are a sum of population of the bare states and nonzero coherence.

$$t \to \infty \quad \sin^2\theta = \cos^2\theta = \frac{1}{2} \Rightarrow$$

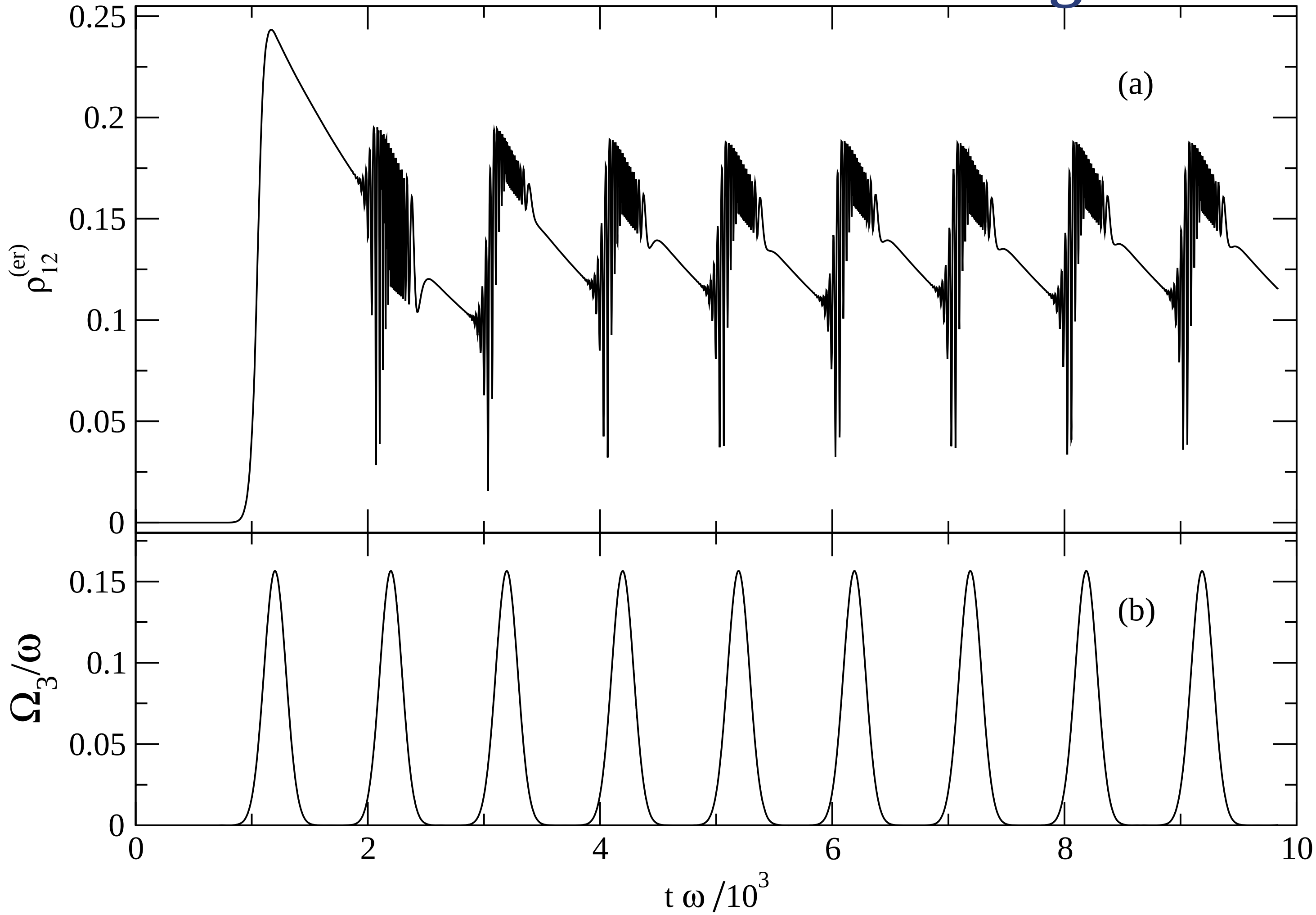
$$\rho^d_{11} = \frac{1}{2}(\rho_{11} + \rho_{22}) - \mathrm{Re}\{\rho_{12}\}, \qquad \rho^d_{22} = \frac{1}{2}(\rho_{11} + \rho_{22}) + \mathrm{Re}\{\rho_{12}\}$$

Rest ns



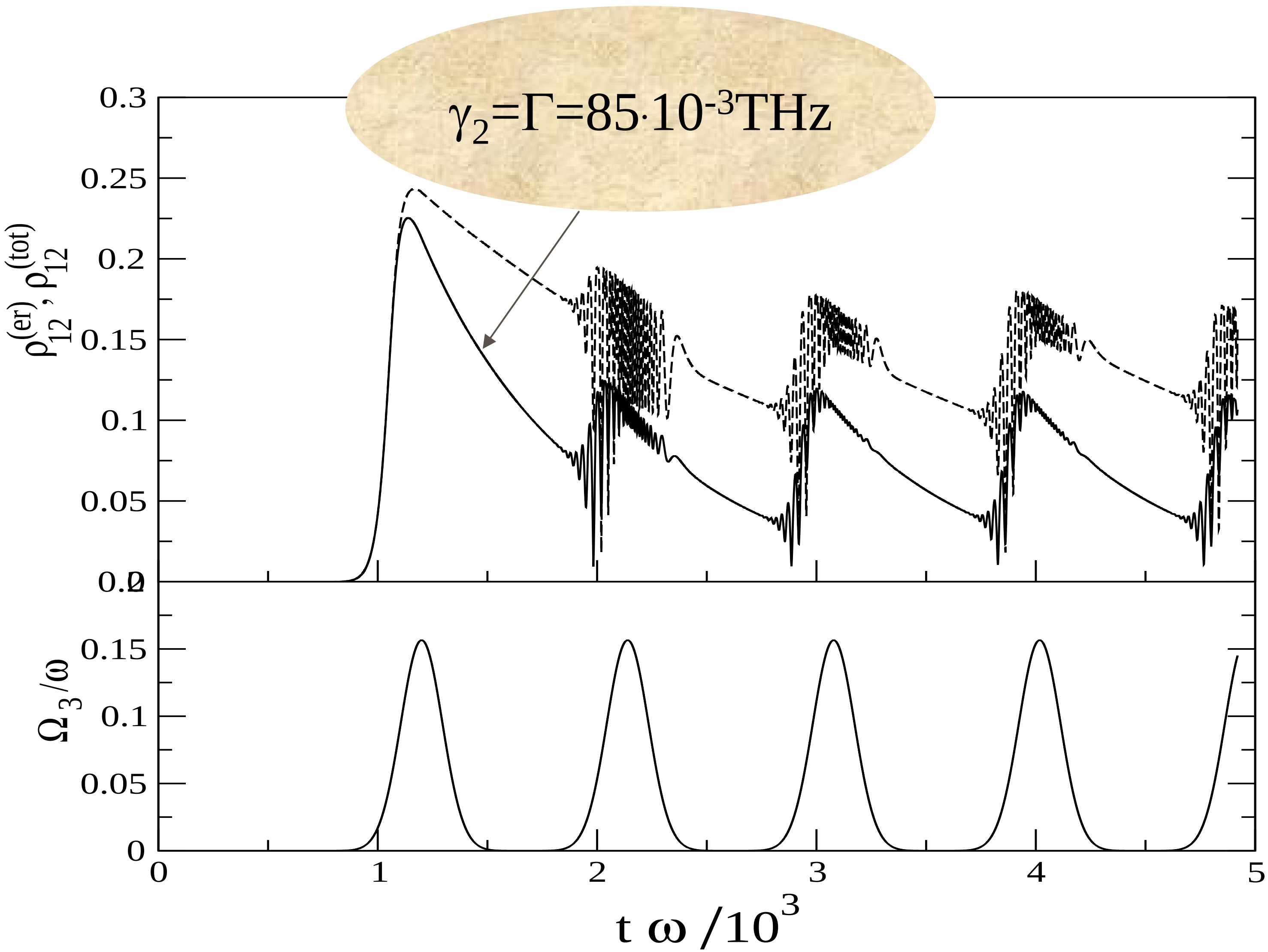
- **Fig. ieves maximum during interaction with the first pulse pair and undergoes decay up to the instant when the second pulse pair arrives and restores population of the upper level and the coherence.**

Effect of two pulse trains having same period as vibrational energy relaxation time: coherence drops to 0.1 and varies within 0.1-0.15 region.



S.A. Malinovskaya, ``Prevention of decoherence by two femtosecond chirped pulse trains'', Opt. Lett. 33, 2245-2247(2008)

Strong Collisions



Conclusions

❖We propose to use quantum control methods for mode selective excitation to advance CARS microscopy techniques.

❖A new method based on the chirped pulse adiabatic passage is developed for the mode selective excitation.

❖The feasibility of this method is investigated in the presence of the vibrational energy relaxation and collisional dephasing as factors of decoherence.

❖It is shown that the use of two chirped pulse trains with the same period as decoherence time allows one to periodically restore population to the upper level in the selected vibrational mode and to preserve high level of the coherence.

Experimental groups

•C.L. Evans, E.O. Potma, M. Puoris'haag, D. Cote, C.P. Lin, X.S. Xie, Proc. Natl. Acad. Sci. 102, 16807 (2005).

•K. R. Wilson, D. S. Peterka, M. Jimenez-Cruz, S. R. Leone, and M. Ahmed, Phys. Chem. Chem. Phys. (2006)

•B. Yellampalle, R.D. Averitt, A. Efimov, and A.J. Taylor, Optics Express 13, 7672, (2005).

•N. Dudovich, D. Oron, and Y. Silberberg, Nature 418, 512 (2002).

•J. Ogilvie, D. Debarre, X. Solinas, J. Martin, E. Beaurepaire, and M. Joffre, Optics Express 14, 759, (2006).

•Thomas Hellerer, Annika Enejder, Ondrej Burkacky, Andreas Zumbusch, Multiphoton Microscopy in the Biomedical Sciences, Proceedings of SPIE, vol. 5323, 1605 (2004).