Fundamental Science and Applications of Ultra-cold Polar Molecules
KITP, Santa Barbara, CA

Raman transitions by chirped optical frequency combs: Prevention of decoherence

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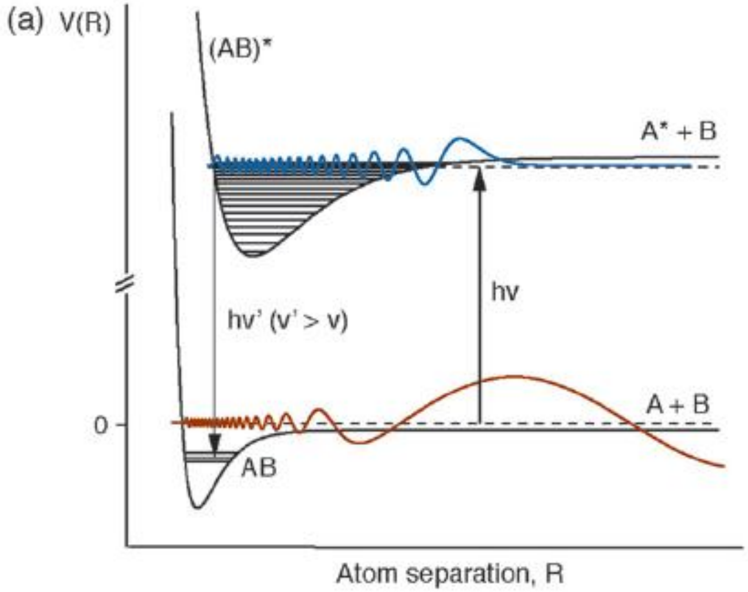
Quantum control at ultracold temperatures

- Originated on the base of latest developments in ultracold gases
- Possibilities to generate ultracold molecules
- Offer internal structure
- Exhibit long-range, dipole-dipole interactions



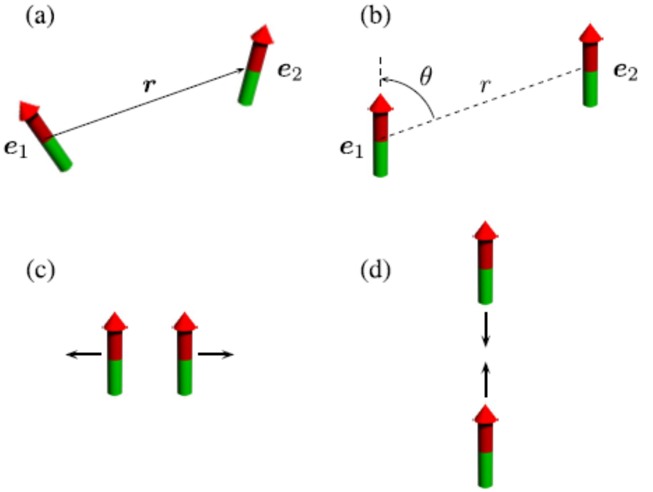
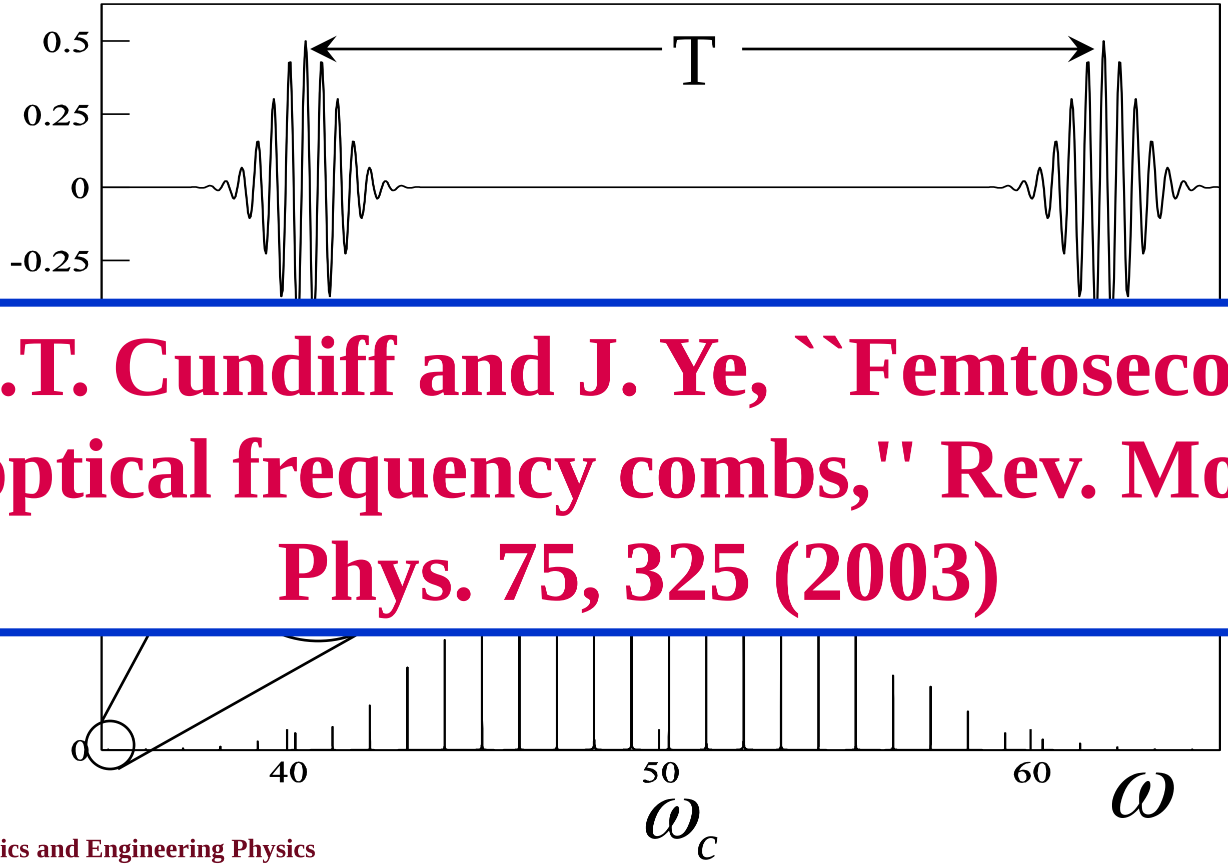


Figure 2. Two particles interacting via the dipole–dipole interaction. (*a*) Non-polarized case; (*b*) polarized case; (*c*) two polarized dipoles side by side repel each other (black arrows); (*d*) two polarized dipoles in a 'head-to-tail' configuration attract each other (black arrows).

Control using Frequency Combs

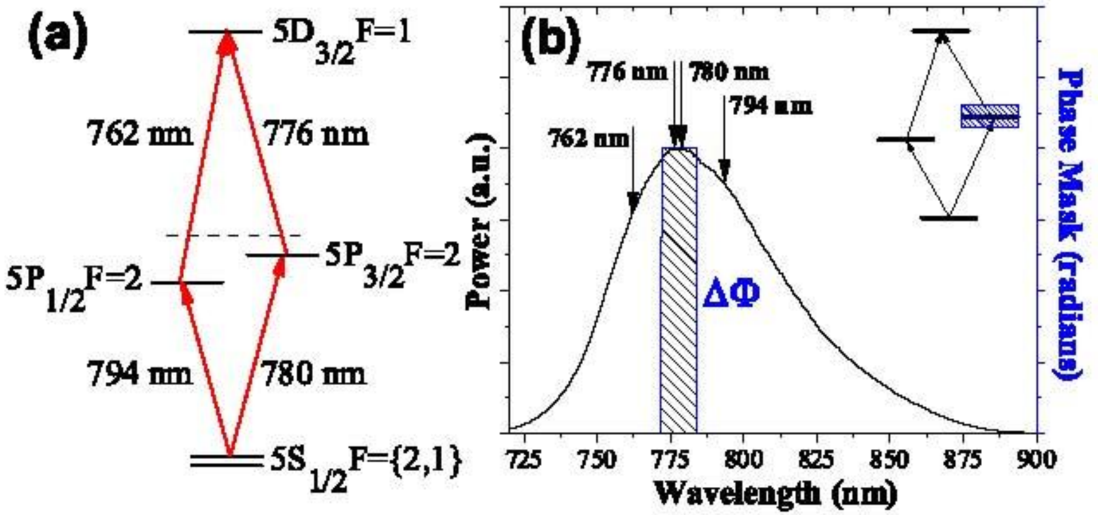
$$\omega_r = 1/\mathrm{T},\ \ f_0 = \Delta\varphi/\mathrm{T},\ \ \omega_n = n\omega_r + f_0,\ \ \ \tau,\ \ \ E_0$$



S.T. Cundiff and J. Ye, ``Femtosecond optical frequency combs,'' Rev. Mod. Phys. 75, 325 (2003)

Implementation of femtosecond optical frequency comb to ultrafast control of population dynamics

- Two-photon absorption in cold ^{87}Rb using a single phase-modulated OFC. M.C. Stowe, A. Peer, J. Ye, ``Control of Four-Level Quantum Coherence via Discrete Spectral Shaping of an Optical Frequency Comb,'' Phys. Rev. Lett., 100, 203001(4) (2008)



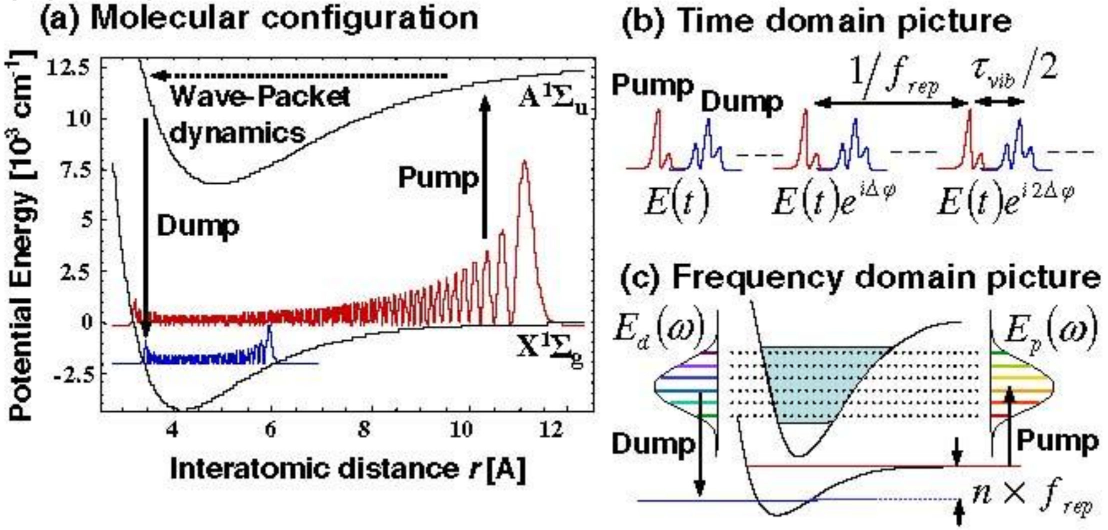
The excited state population is proportional to cos²(Ψ/2), here

$$\Psi = \varphi_{780} + \varphi_{776} - \varphi_{794} - \varphi_{762}$$

Implementation of femtosecond optical frequency comb to ultrafast control of population dynamics

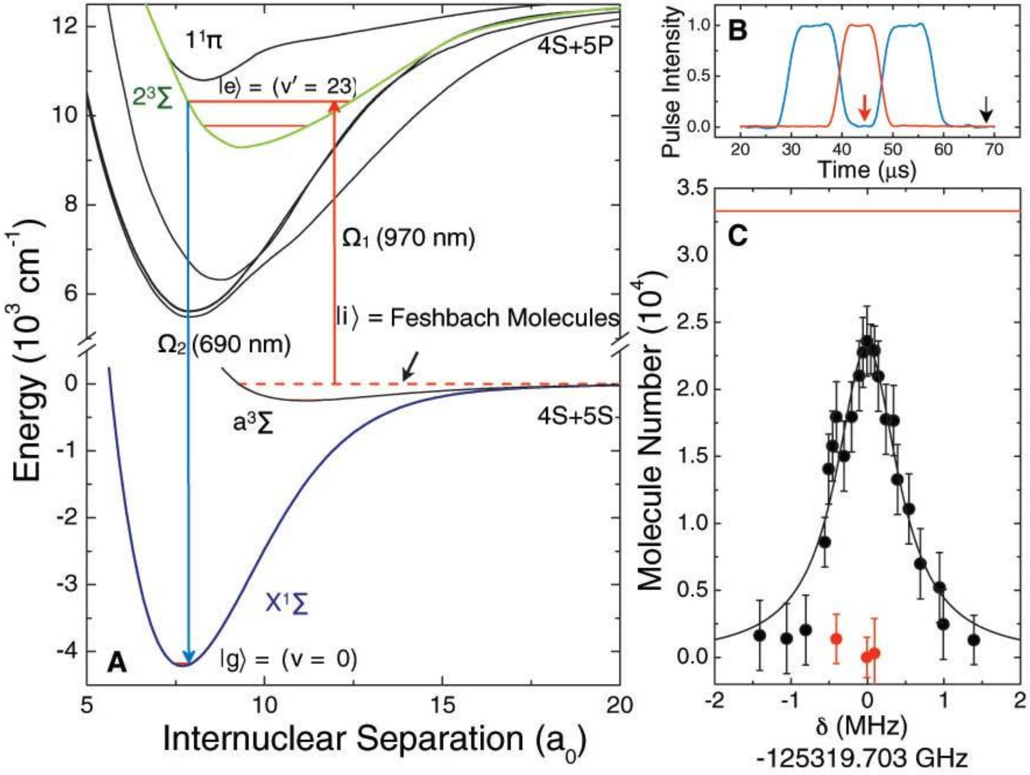
- **Piecewise adiabatic passage in multi-level system using amplitude and phase modulated two pulse trains; application to KRb molecular cooling from Feshbach states.**

  A. Peer, E.A. Shapiro, M.C. Stowe, M. Shapiro, J.Ye, ``Precise control of molecular dynamics with a femtosecond frequency comb'', Phys. Rev. Lett., 98, 113004(4) (2007)



Creation of a High Phase-Space-Density Gas of Polar Molecules

- K.-K. Ni, S. Ospelkaus, M.H.G. de Miranda, A. Pe'er,B. Neyenhuis,J.J. Zirbel, S. Kotochigova, P.S. Juliene, D.S. Jin, J.Ye, ``A High Phase-Space-Density Gas of Polar Molecules", Science 322, 231 (2008)



Femtosecond optical frequency comb

- OFC generated by the pulse train with the *sin*-phase modulation

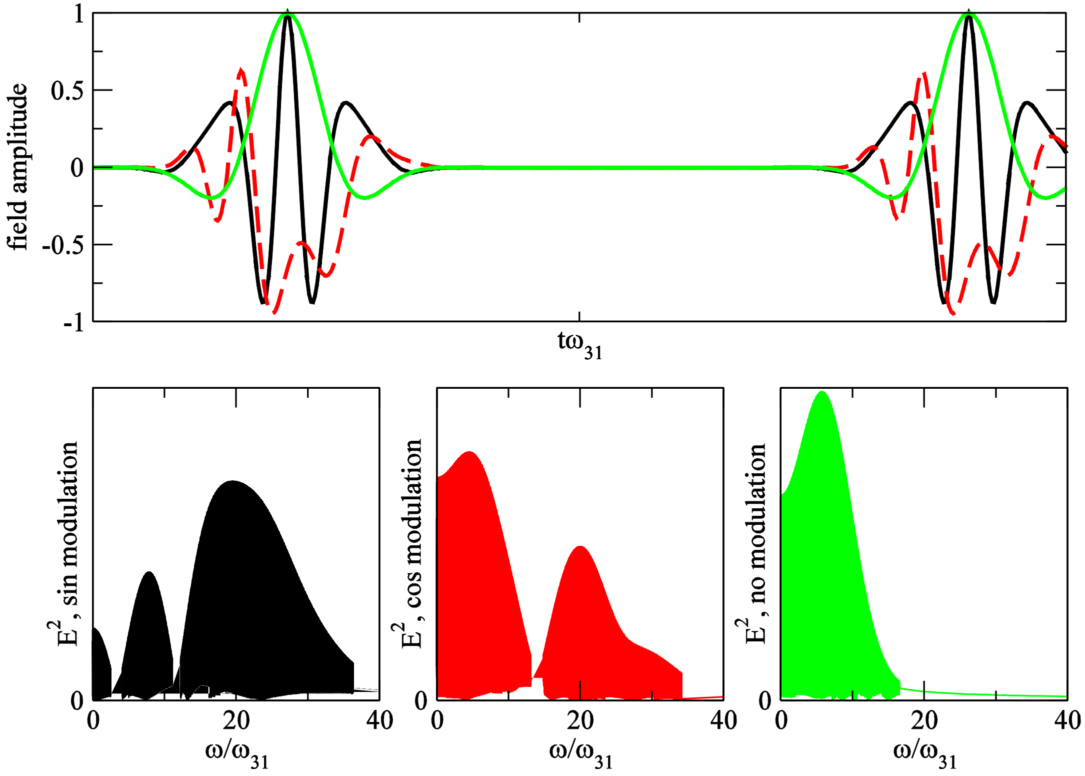
$$\mathrm{E(t)}=\sum_{\mathrm{k=0}}^{\mathrm{N}}\mathrm{E}_0 e^{-(t-kT)^2/(2\tau^2)}\cos(\omega_{\mathrm{L}}(\mathrm{t\text{-}kT})+A\sin\Omega(t-kT)+\varphi)$$

John L. Hall, L. Hollberg, T. Baer, H.G. Robinson, ``Optical heterodine saturation spectroscopy", Appl. Phys. Lett. 39, 680 (1981).

- Standard OFC

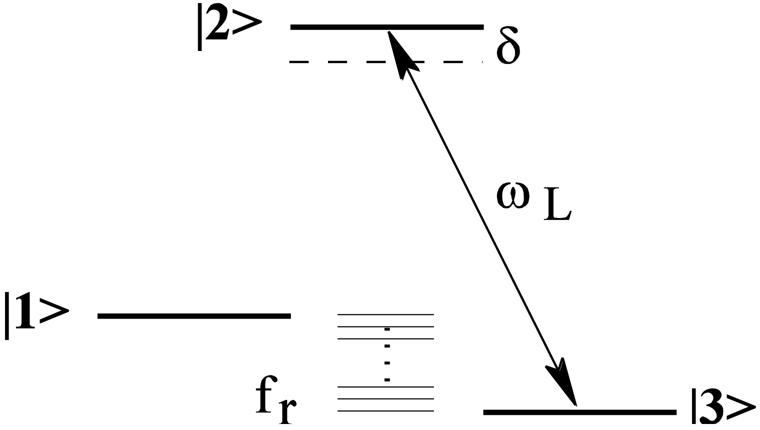
$$\mathrm{E}(t)=\sum_{\mathrm{k=0}}^{\mathrm{N}}\mathrm{E}_0 e^{-(t-kT)^2/(2\tau^2)}\cos(\omega_{\mathrm{L}}(\mathrm{t\text{-}kT})+\varphi)$$

Power spectrum for a standard, sin and cos modulated OFC



Pulse duration τ=0.25 [ω^{-1}], train period T=6400τ, the Rabi frequency is Ω_R= 1, carrier frequency ω_L=5.9 , modulation frequency Ω =4.9 [ω].

Semi-Classical Model of Raman Transitions in three-level Λ–system using an optical frequency comb



$$i\hbar\begin{pmatrix}\dot{a}_1\\ \dot{a}_2\\ \dot{a}_3\end{pmatrix}=\begin{pmatrix}\hbar\omega_1 & -\mu E(t) & 0\\ & \hbar\omega_2 & -\mu E(t)\\ c.c. & & \hbar\omega_3\end{pmatrix}\begin{pmatrix}a_1\\ a_2\\ a_3\end{pmatrix}$$

$$\mathrm{E(t)}=\sum_{\mathrm{k=0}}^{\mathrm{N}}\mathrm{E}_0 e^{-(t-kT)^2/(2\tau^2)}\cos(\omega_{\mathrm{L}}\,\mathrm{(t\text{-}kT)}+A\sin\Omega(t-kT)+\varphi)$$

Hamiltonian matrix elements in the interaction representation

$$H_{12} = c\left[e^{-i(\omega_L+\omega_2-\omega_1)t-iM} + e^{i(\omega_L-\omega_2+\omega_1)t+iM} \right]$$

$$H_{23} = c\left[e^{-i(\omega_L+\omega_3-\omega_2)t-iM} + e^{i(\omega_L-\omega_3+\omega_2)t+iM} \right]$$

$$H_{11} = H_{22} = H_{33} = H_{13} = 0$$

$$c = \mu E_0(t)/\hbar \qquad M = A\sin\Omega(t-kT)+\varphi$$

Taking into account decoherence

$$i\hbar\dot{\rho} = [H, \rho] + relaxation\ terms$$

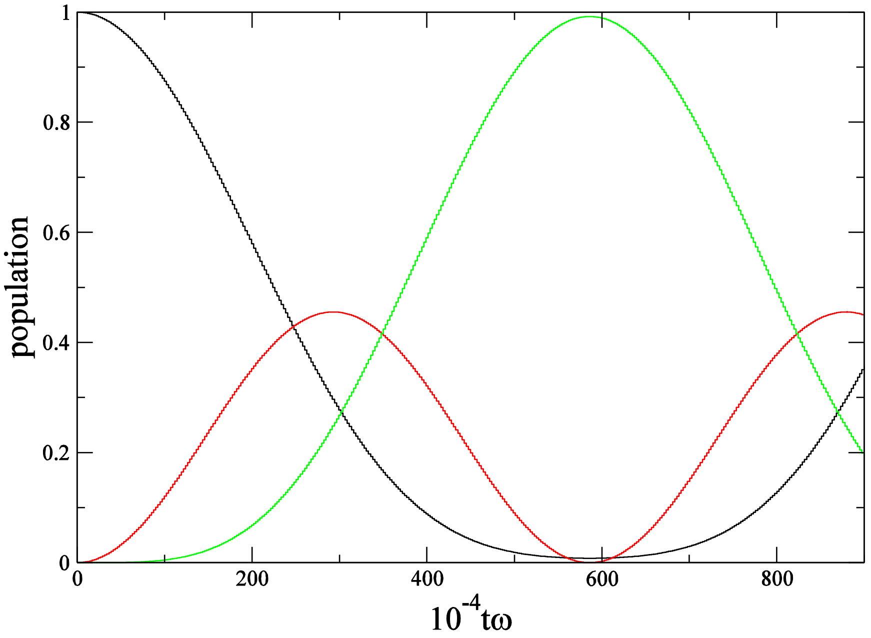
Reduced density matrix elements are

$$\left.\dot{\rho}_{11}\right)_{sp} = \gamma_{2,1}\rho_{22} - \gamma_{3,1}\rho_{11} \qquad \left.\dot{\rho}_{12}\right)_{sp,col} = -\left(\frac{\gamma_{2,1}}{2} + \frac{\gamma_{2,3}}{2} + \Gamma_{21}\right)\rho_{12}$$

$$\left.\dot{\rho}_{22}\right)_{sp} = -\gamma_{2,1}\rho_{22} - \gamma_{2,3}\rho_{22} \qquad \left.\dot{\rho}_{13}\right)_{sp,col} = -\left(\frac{\gamma_{3,1}}{2} + \Gamma_{31}\right)\rho_{13}$$

$$\left.\dot{\rho}_{33}\right)_{sp} = \gamma_{2,3}\rho_{22} + \gamma_{3,1}\rho_{11} \qquad \left.\dot{\rho}_{23}\right)_{sp,col} = -\left(\frac{\gamma_{2,1}}{2} + \frac{\gamma_{2,3}}{2} + \Gamma_{23}\right)\rho_{23}$$

Resonant population transfer in the λ-system using a standard optical frequency comb with period T=0.2 ns



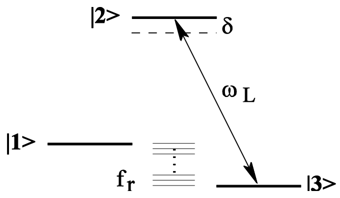
Black – state |1>

Red – state |2>

Green – state |3>

T=0.2 ns

f_r=5 GHz

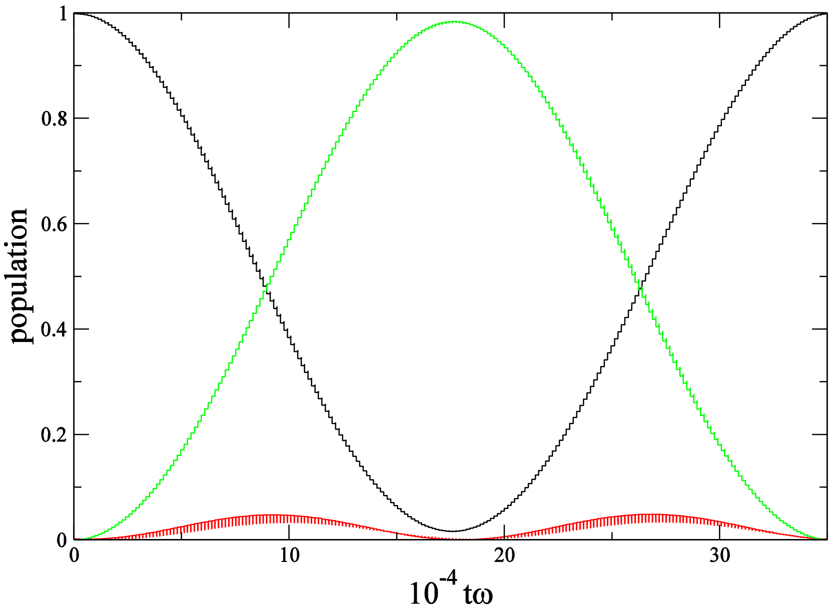


$$\omega_{21} = 309.3\,\mathrm{THz} \quad \omega_{32} = 434.8\,\mathrm{THz} \quad \omega_{31} = 125.5\,\mathrm{THz}$$

$$\omega_L = \omega_{32} = 434.8\,\mathrm{THz} \qquad I = (10^8 \div 10^{12}\,\frac{\mathrm{W}}{\mathrm{cm}^2})$$

Resonant case: Population transfer in the λ–system using a sine-phase modulated OFC

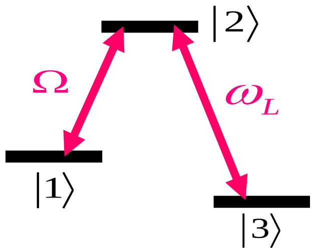
population



time

Black – state |1>
Red – state |2>
Green – state |3>

f_r=50 GHz

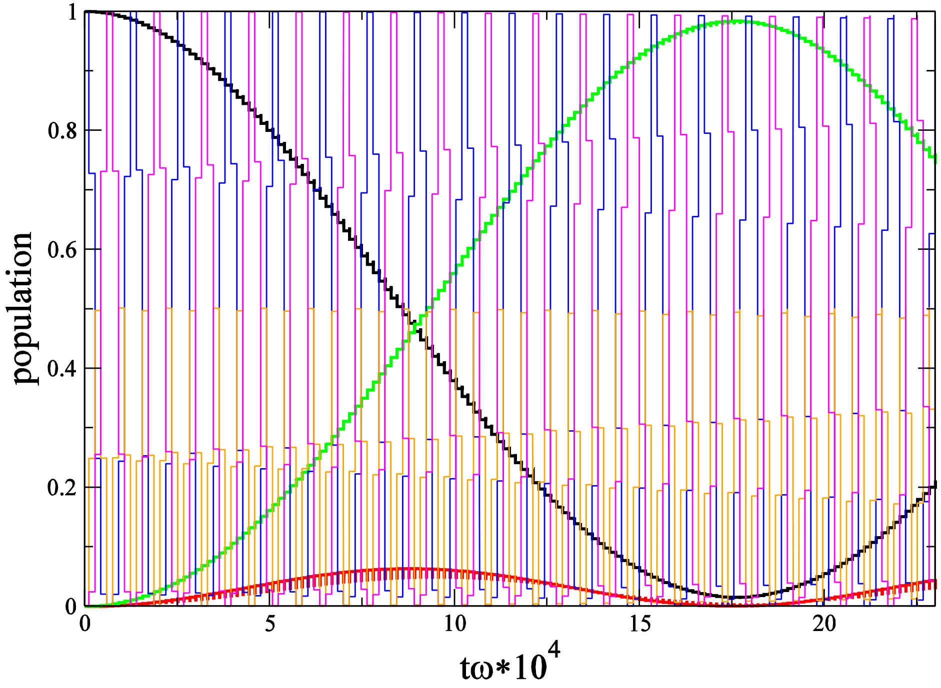


$$\omega_{21} = 340.7THz \qquad \omega_{32} = 410.7THz \qquad \omega_{31} = 70THz$$

$$\omega_L = \omega_{32} = 410.7THz \qquad \Omega = \omega_{21} = 340.7THz$$

$$\Omega_R = 70THz \ \ (I = 5.85 \cdot 10^{12} \ \tfrac{W}{cm^2})$$

Population transfer in the three-level system using optical frequency comb with the cos modulation

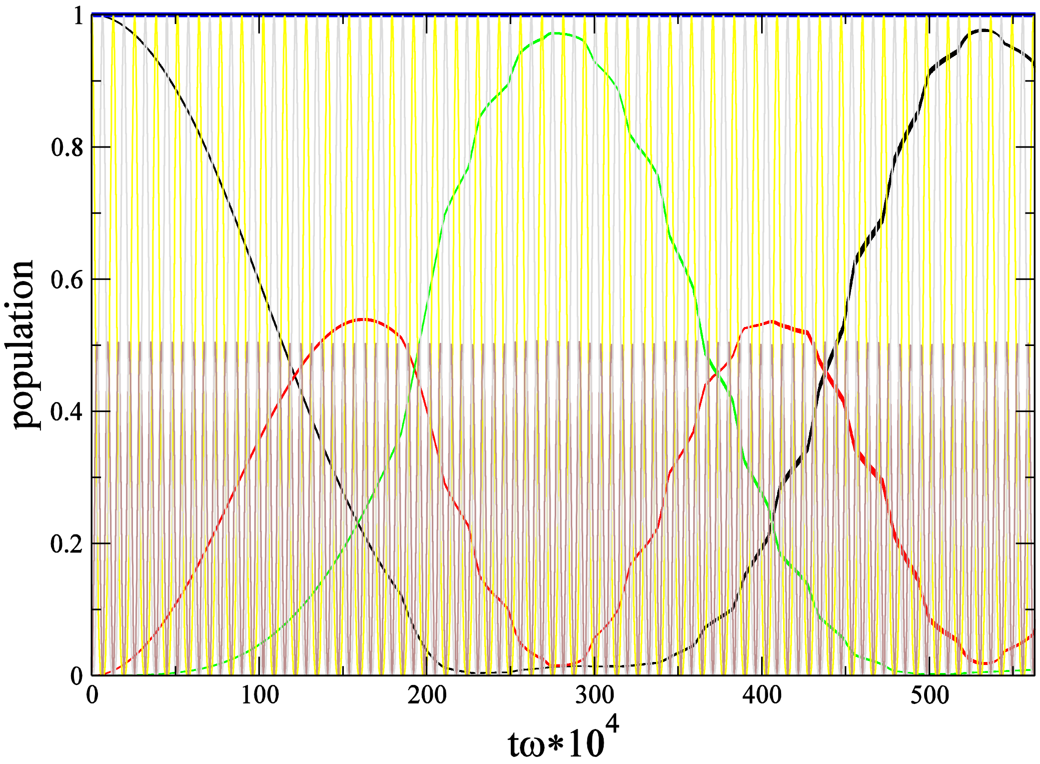


$$\omega_{21} = 340.7THz \quad \omega_{32} = 410.7THz \quad \omega_{31} = 70THz$$

$$\omega_L = \omega_{32} = 410.7THz \quad \Omega = \omega_{21} = 340.7THz$$

$$\Omega_R = 70THz \;\; (I = 5.85 \cdot 10^{12} \tfrac{W}{cm^2})$$

Weak field regime

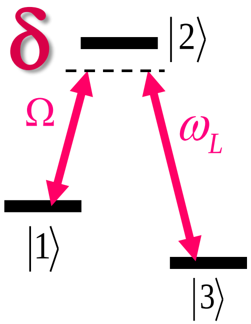


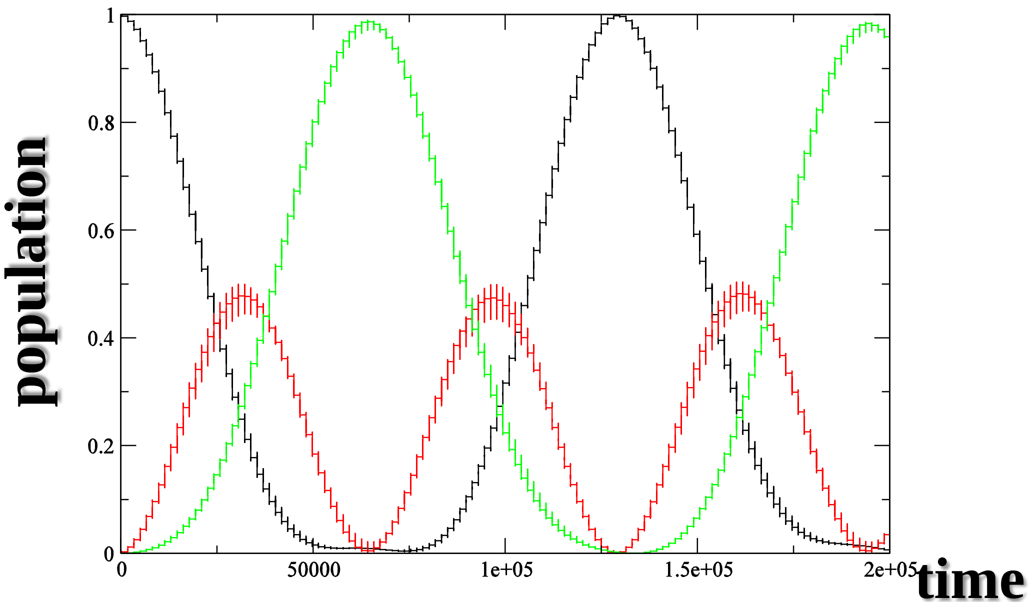
$$\omega_{21} = 340.7THz \qquad \omega_{32} = 410.7THz \qquad \omega_{31} = 70THz$$

$$\omega_L = \omega_{32} = 410.7THz \qquad \Omega = \omega_{21} = 340.7THz$$

$$\Omega_R = 7THz$$

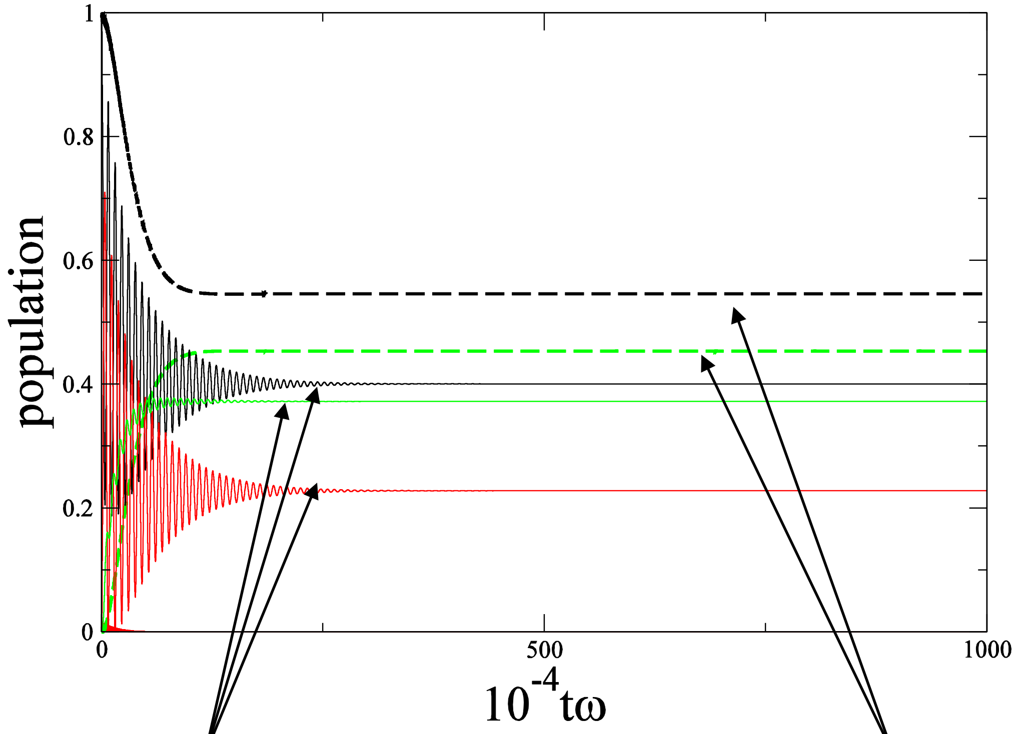
Detuned case: Population transfer in the λ–system using an OFC with carrier frequency ω_L and modulation frequency Ω detuned off resonance by $\delta=\omega_{31}/2$

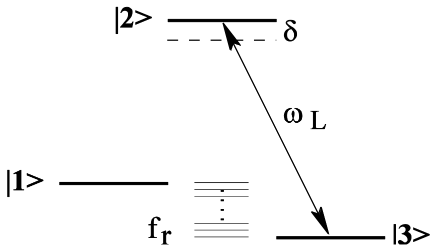




- Total population transfer occurs after 42 applied pulses.
- The individual pulse duration is 3 fs.
- The pulse train period is 23 ps, thus, total population transfer is accomplished within 1 ns
- However, the population of the electronically excited state is substantial during the dynamics, reaching almost 50%.

Population dynamics induced by a standard optical frequency comb in the presence of spontaneous decay and collisions





collisions, no decay

$\gamma_{21} = 0.,\ \Gamma_{32} = \Gamma_{12} = 10^{-3}$

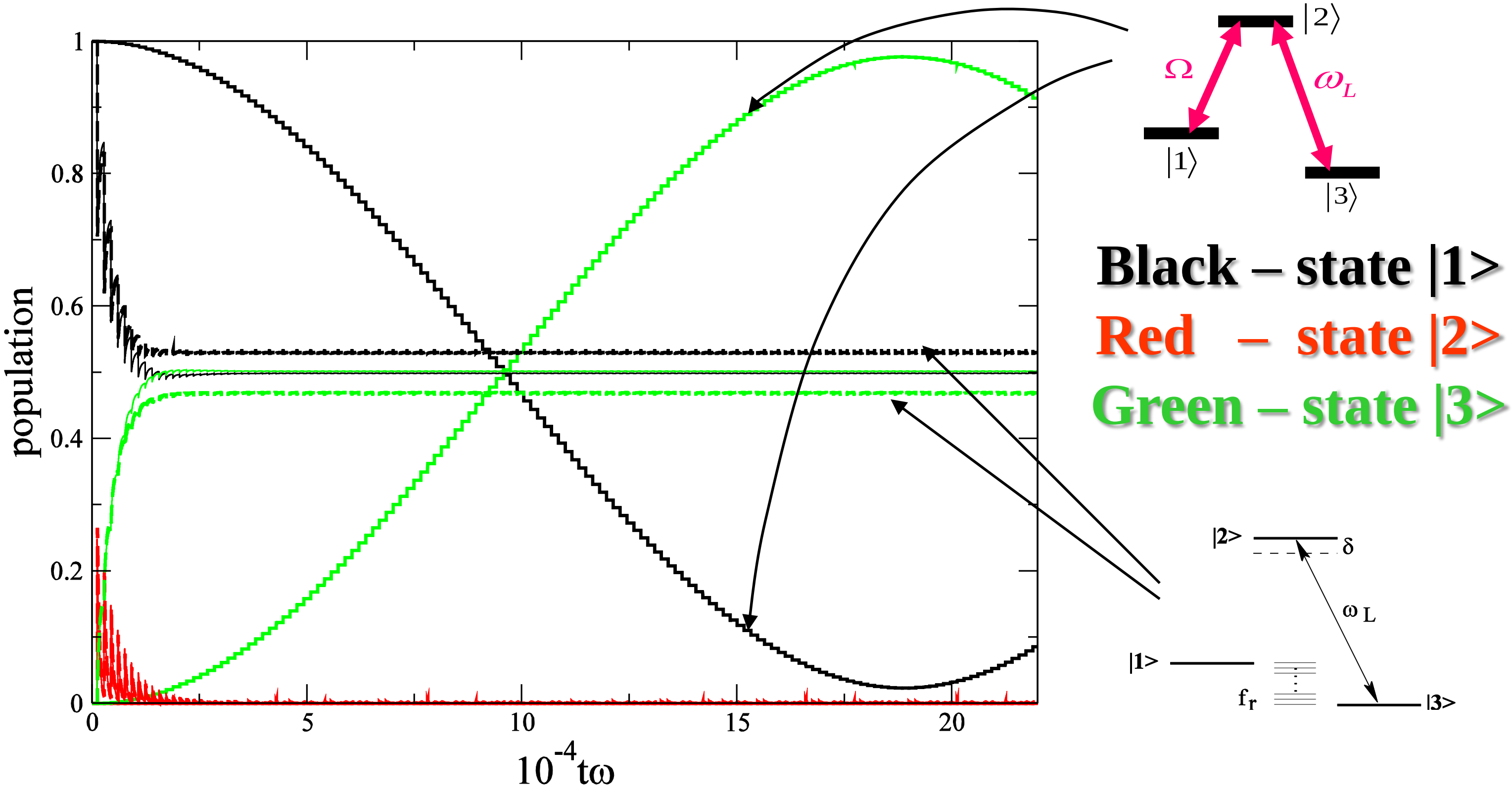
$\Omega_{Rabi} = 1.\ [\omega_{31} = 70THz]$

collisions + decay

$\gamma_{21} = \gamma_{23} = 10^{-3},\ \Gamma_{32} = \Gamma_{12} = 10^{-3}$

$\Omega_{Rabi} = 1.\ [\omega_{31} = 70THz]$

Population dynamics induced by the phase modulated optical frequency comb (solid lines) vs the standard one (dashed lines)



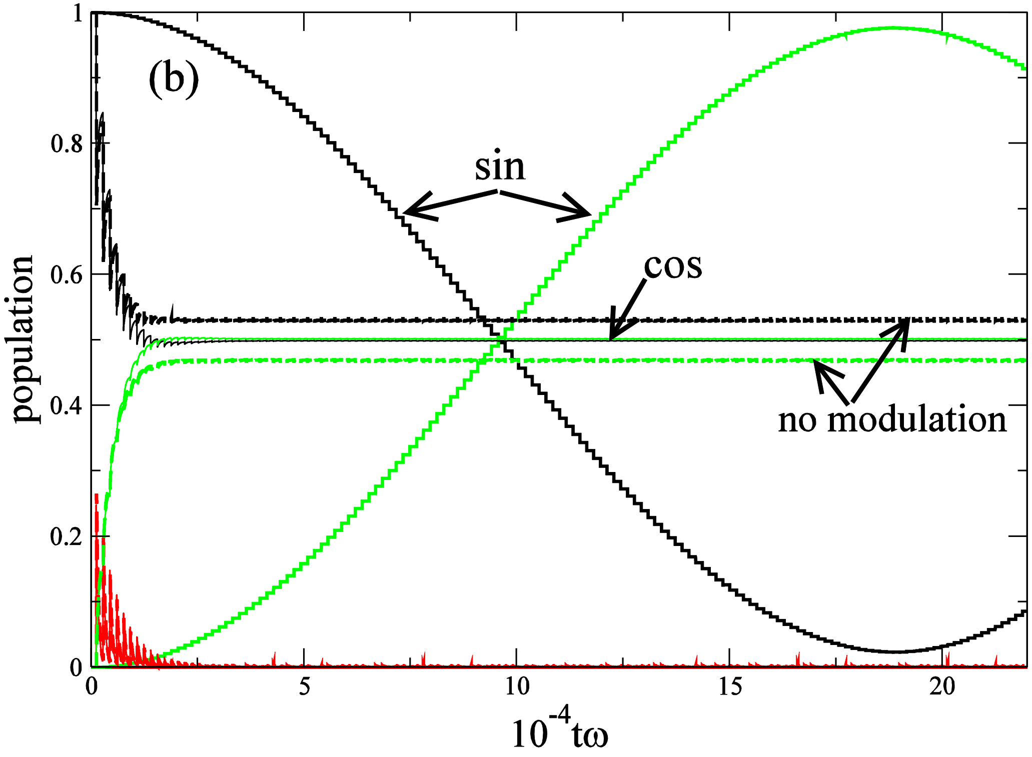
Black – state |1>

Red – state |2>

Green – state |3>

$$\gamma_{21}=\gamma_{23}=10^{-3},\ \ \Gamma_{32}=\Gamma_{12}=10^{-3},\ \ \Omega_{Rabi}=1.\ [\omega_{31}=70THz]$$

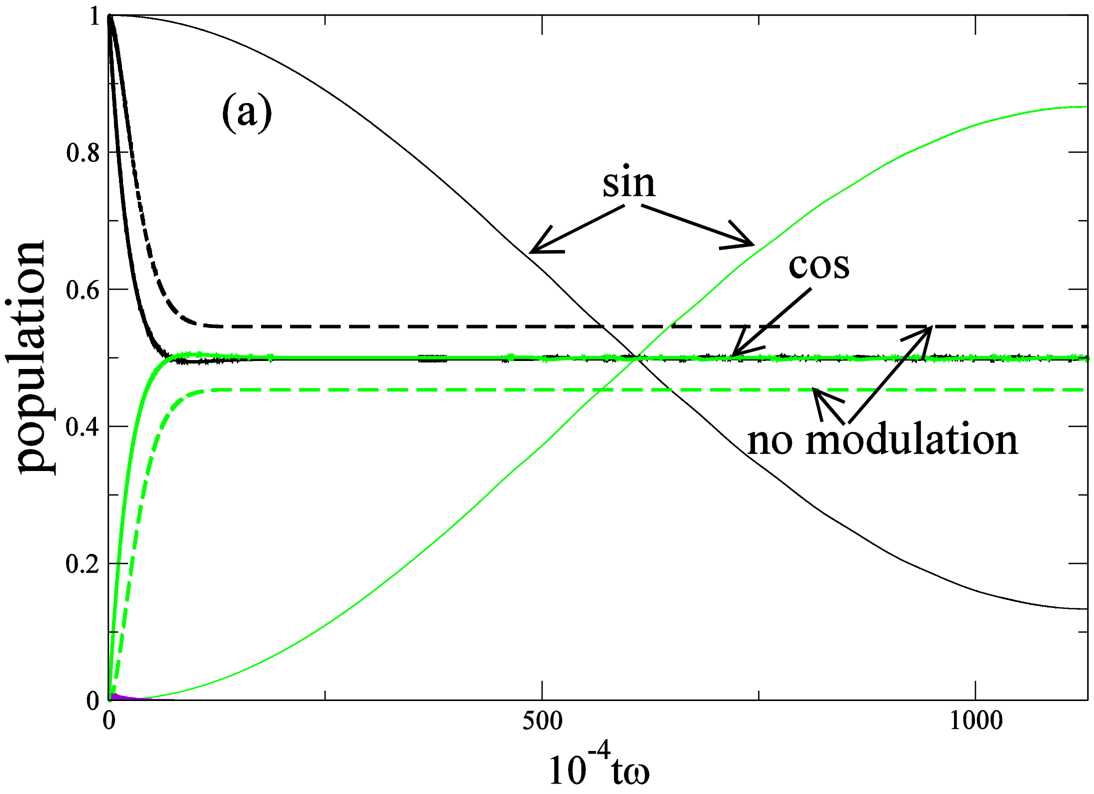
Population dynamics induced by sine/cosine and standard OFCs



$$M.Shapiro: \omega_{21} = 340.7THz \quad \omega_{32} = 410.7THz \quad \omega_{31} = 70THz$$

$$\omega_L = \omega_{32} \quad \Omega = \omega_{21} \quad \Omega_R = 70THz \ \ (I = 5.85 \cdot 10^{12} \tfrac{W}{cm^2})$$

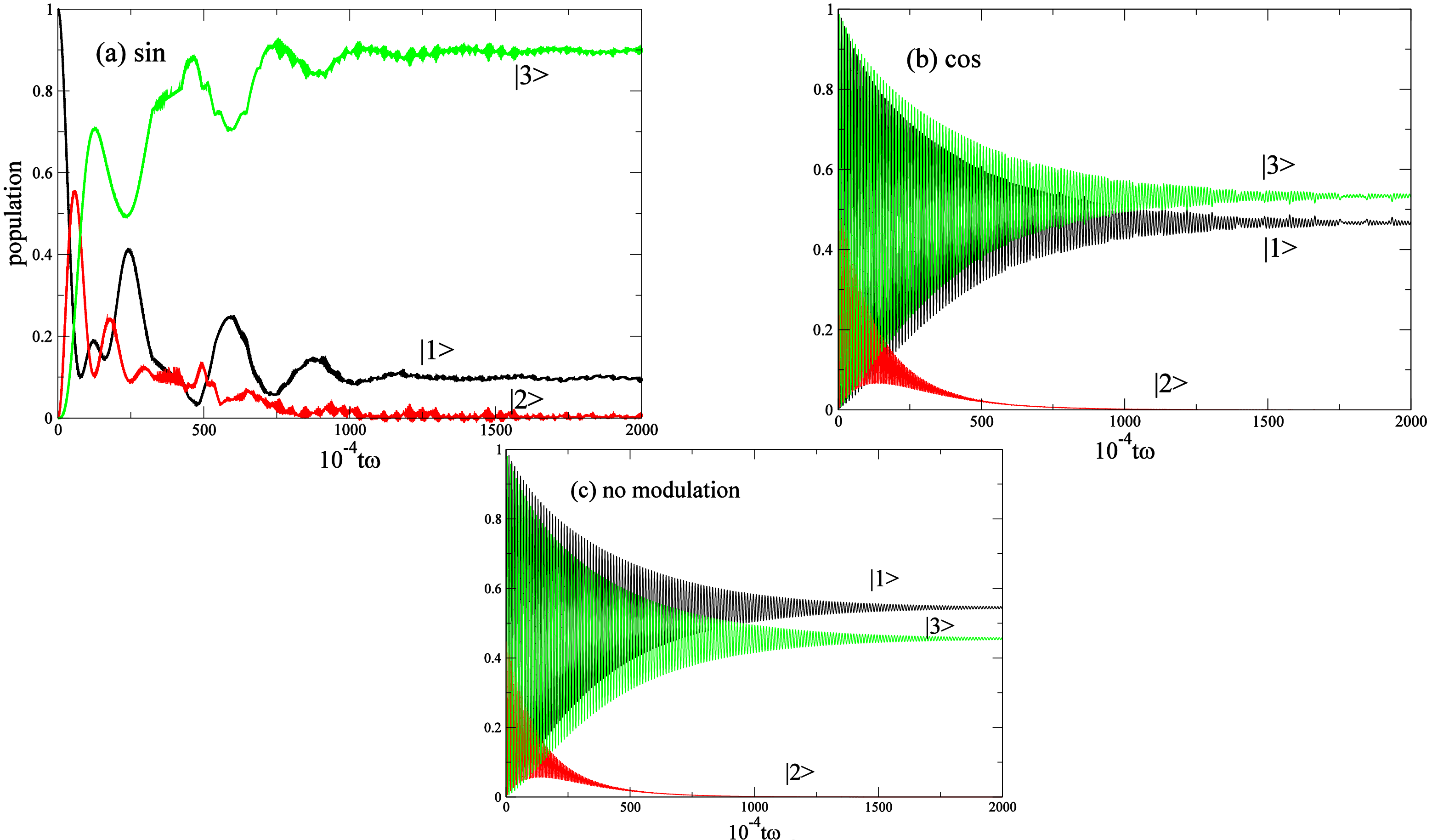
Population dynamics induced by sine/cosine and standard OFCs



$$J.Ye: \omega_{21} = 309.3THz \quad \omega_{32} = 434.8THz \quad \omega_{31} = 125.5THz$$

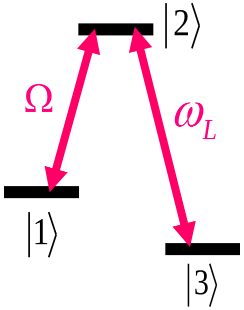
$$\omega_L = \omega_{32} \quad \Omega = \omega_{21} \quad \Omega_R = 12.5THz$$

Population dynamics in the presence of experimental decoherence

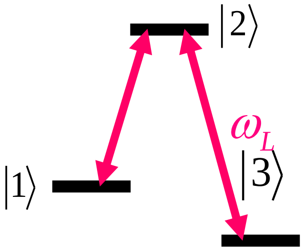


$$\gamma_{21} = \gamma_{32} = 0.1GHz,\ \ \Gamma_{21} = \Gamma_{32} = 10^3 Hz,\ \ \Omega_{Rabi} = 12.5THz$$

Raman resonances



- For phase-modulated OFC, the condition for the Raman resonance is satisfied by making the difference $(\omega_L - \Omega)$ equal to the two-photon transition frequency ω_{31}.
- Additionally, the modes that are multiples of f_r provide pairs of optical frequencies that differ by exactly the transition frequency ω_{31} .



- For the standard OFC, the two-photon resonance condition is satisfied by optical frequencies that are multiples of f_r and $mf_r - nf_r = \omega_L - n'f_r = \omega_{31}$, here m,n,n' are integer numbers.

Summary

- Optical frequency comb may be efficiently used to perform internal state cooling from the Feshbach state.
- Parity of the chirp of the comb is of key importance in achieving a desired quantum yield.
- Quantum Control techniques that implement specific pulse shapes and schemes may be successfully adapted to the presence of decoherence and mitigate its effects.

Thank you!

- Tomas Collins (PhD in Physics, 2013)
- Vladislav Zakharov (PhD student, Stony Brook)
- Wufu Shi (grad. student)
- Spencer Horton (PhD student, Stony Brook)
- Vishesha Patel (PhD in Physics, 2011)
- Praveen Kumar (Postdoc at Texas Tech U.)
- Vladimir Malinovsky (Stevens and ARL)

National Science Foundation
WHERE DISCOVERIES BEGIN

Recent publications

- S. A. Malinovskaya, S. Horton, ``Rovibrational cooling using optical frequency combs in the presence of decoherence,'' J. Opt. Soc. Am. B **30**, 482 (2013).
- S. A. Malinovskaya, T. Collins, V. Patel, ``Ultrafast manipulation of Raman transitions and prevention of decoherence using chirped pulses and optical frequency combs," Advanc. Quant. Chem., **64,** 211 (2012).
- W. Shi, S. Malinovskaya, ``Implementation of a single femtosecond optical frequency comb for molecular cooling", Phys. Rev. A **82**, 013407 (2010).
- S. Malinovskaya, W. Shi, ``Feshbach-to-ultracold molecular state Raman transitions via a femtosecond optical frequency comb", J. Mod. Opt. **57**, 1871 (2010).