KITP Program: Frontiers of Intense Laser Physics
Jul 21 - Sep 19, 2014, Santa Barbara, CA

On quantum control in strong fields

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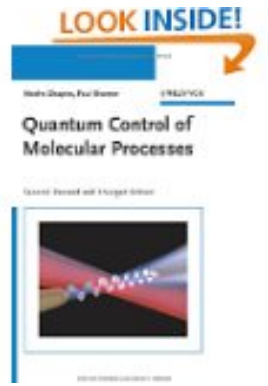
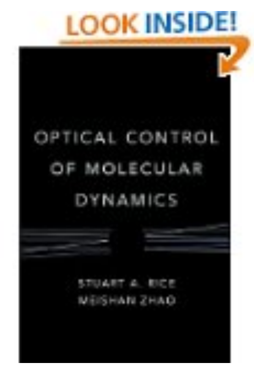
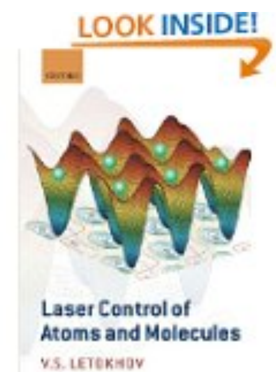
Quantum Control

- QC implies the development and implementation of quantum methods and techniques to manipulate ultrafast dynamics in atoms and molecules to stir them to a predetermined quantum yield by making use of ultrafast crafted laser pulses

- Books: Shapiro&Brumer, Rice&Shao,Tannor,Letokhov

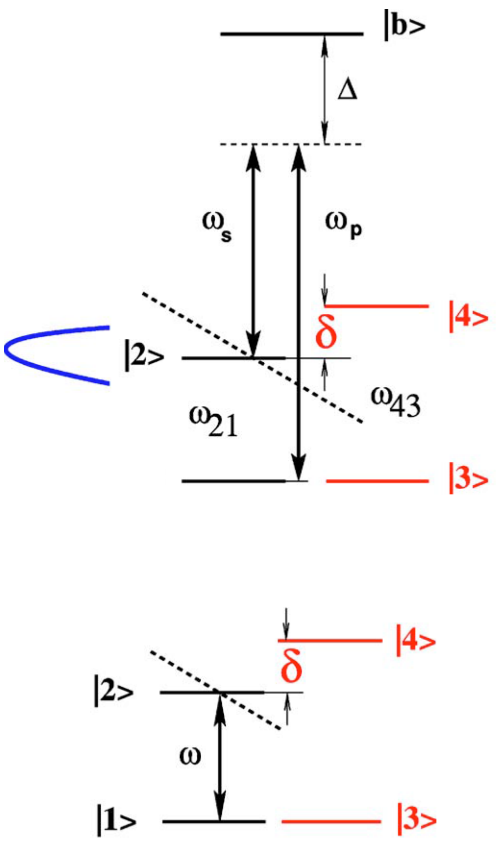






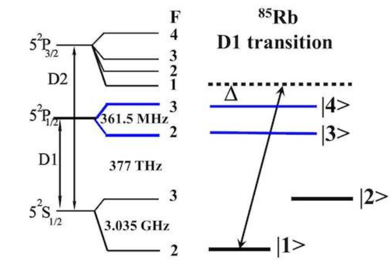
Strong field control

- Population is transferred from the ground state significantly or completely;
- Under the above conditions the perturbation theory is not applicable;
- The exact solution of the Schrödinger equation is required;
- Rabi frequency is on the order of the transition frequency;
- Strong field regime is determined in correlation with the the splitting of the energy levels in a system;



Few examples

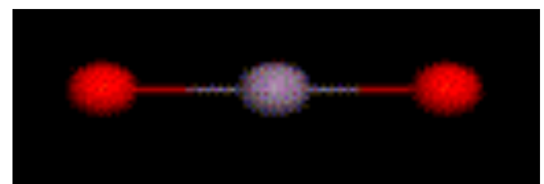
- For transitions between hyperfine structure in alkali atoms the field of kW/cm^2 is sufficient;



G. Liu, S. M. PRA 89, 041803® (2014)
T. Collins, S.M. Opt. Lett. 37, 2298 (2012)

- To address vibrational structure, the optical field intensities of10^{10} -10^{12} W/cm^2 are commonly used;

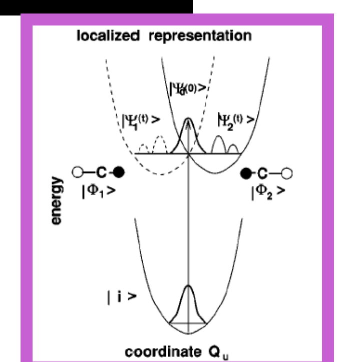
S. M. Opt. Lett. 33, 2245 (2008)
S.M.,V. Malinovsky Opt. Lett. 32, 707 (2007)



- Core electron dynamics – 10^{18} W/cm^2

S. M., L. Cederbaum PRA 61, 42706 (2000)

- Nuclear dynamics – 10^{24} W/cm^2



What it is for?

- Implementing the concept of quantum control means finding fields that stir the system to a desired quantum yield, besides, the solution would preferentially be robust in experimental realization.
- Robustness assumes some flexibility in the range of parameters of the field that work well and lead to a desired quantum yield.
- This flexibility is available within the adiabatic region of light-matter interaction.
- So, the goal is to find the adiabatic solution for the problem in hand. Adiabatic passage usually provides 100% efficiency.
- Adiabatic solution may be found numerically or analytically through the dressed state analysis.

Basic principle behind the dressed state analysis

We present a wave function as a linear superposition of bare states in the field interaction rprn

$$\left|\Psi(t)\right\rangle = \sum_i C_i \left|i\right\rangle$$

$$i\hbar\dot{\boldsymbol{C}} = \hat{H}_{\text{int}}\boldsymbol{C}$$

We apply unitary transformation T *– unitary eigenvectors matrix of* $\hat{H}_{\text{int}}$

We obtain the dressed state Hamiltonian $\hat{H}_d = T^+\hat{H}_{\text{int}}T$

Written in the basis of dressed states $|I>$

$$\left|\Psi(t)\right\rangle = \sum_i C_{di} \left|I\right\rangle, \quad \boldsymbol{C_d} = T\boldsymbol{C}$$

Putting $\boldsymbol{C} = T^+\boldsymbol{C_d}$ *into Schroedinger eqn we arrive*

$$i\hbar\dot{\boldsymbol{C}}_{\boldsymbol{d}} = \hat{H}_d\boldsymbol{C_d} - i\hbar T\dot{T}^+\boldsymbol{C_d}$$

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Adiabatic limit of light-matter interaction

$$i\hbar\dot{\mathbf{C}}_d = \hat{H}_d\mathbf{C}_d - i\hbar T\dot{T}^{+}\mathbf{C}_d$$

Adiabatic term **Non-adiabatic coupling**

If non-adiabatic term is naturally small or zero, the system is in the adiabatic regime. If this is not the case, our goal is to to find field

parameters that provide adiabatic nature of interaction

Variety of approaches to create control fields to get into adiabatic regime

~Pulse area solution

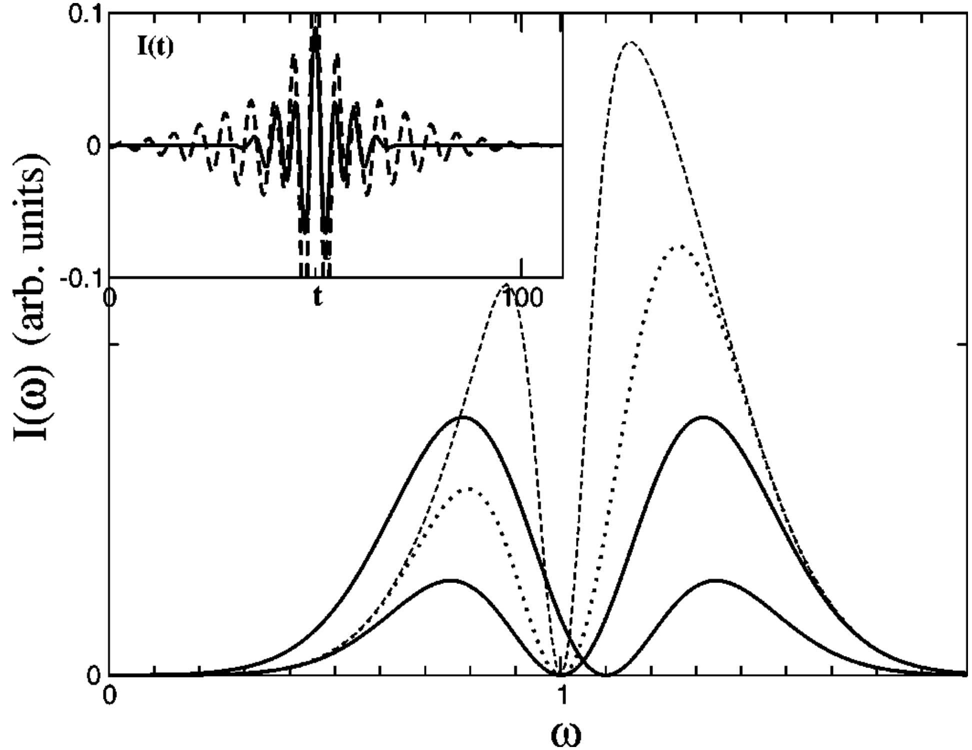
~Time delay between pulses (STIRAP)

~Femtosec pulse trains (OFC)

~Pulse shaping

- Amplitude modulation
- Phase modulation
- Both
- Analytically
- Numerically using OCT

Amplitude modulation



Intensity spectral profile as a function of frequency for T=10,5,3 (dashed, dotted, and solid lines). In the inset the intensity envelope as a function of time is presented for T=10,3. All frequencies are in units of ω_{21} and times in units of ω_{21}. S.A. Malinovskaya, P.H. Bucksbaum, P.R. Berman, PRA 69, 013801 (2004)

General expression for the frequency modulated (chirped) pulses

$$E(t) = E_0(t)\cos(\omega t + \phi(t)) = E_0(t)\cos((\omega + \dot{\phi}(t))t)$$

$$\phi(t) = b_0 + b_1 t + b_2 t^2 + b_3 t^3 + \ldots$$

$$\dot{\phi}(t) = b_1 + 2b_2 t + 3b_3 t^2 + \ldots$$

$$Linear \quad chirp: \dot{\phi}(t) = 2b_2 t$$

$$E(t) = E_0(t)\cos(\omega + 2b_2 t)t; \quad 2b_2 = \frac{\beta}{2}$$

$$E(t) = E_0(t)\cos(\omega t + \frac{\beta t^2}{2})$$

D. Goswami, PRL 88, 177901 (2002)

Chirped Laser Pulses

Time domain	**Frequency domain**
$E_0(t) = E_0 \exp\left[-\frac{t^2}{2\tau^2} - i\omega_0 t - i\alpha\frac{t^2}{2}\right]$	$E_0(\omega) = E'_0 \exp\left[-\frac{(\omega-\omega_0)^2}{2\Gamma^2} + i\alpha'\frac{(\omega-\omega_0)^2}{2}\right]$

E_0 is the peak amplitude;

$\tau\sqrt{\ln 16}$ is the pulse duration;

ω_0 is the center frequency;

α is the temporal chirp;

E'_0 is the peak amplitude;

$\Gamma\sqrt{\ln 16}$ is the frequency bandwidth;

ω_0 is the center frequency;

α' is the spectral chirp;

Transform-limited pulse $\rightarrow \alpha = \alpha' = 0,\ \Gamma = \frac{1}{\tau_0}$

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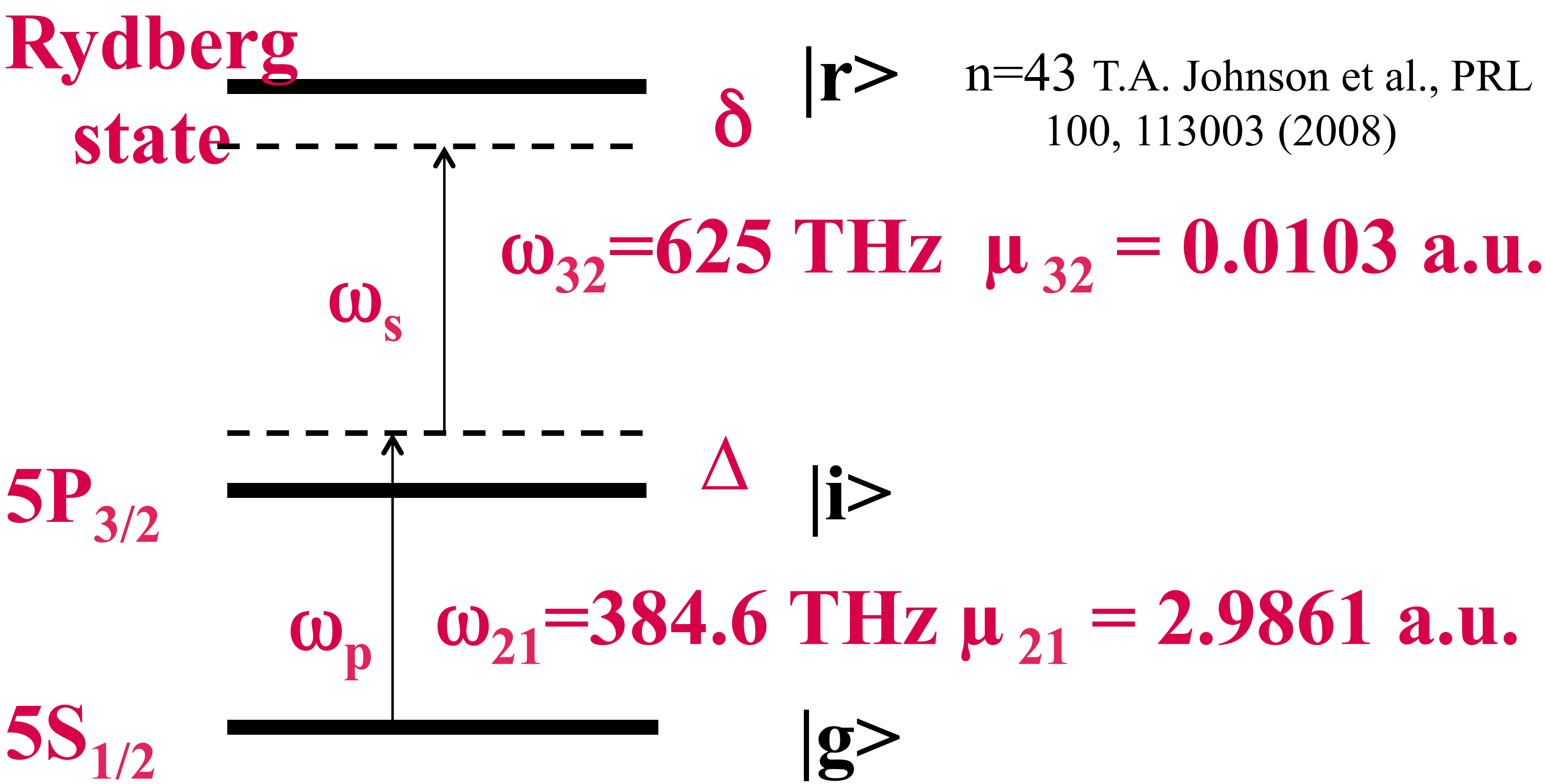
Let's look how a chirped pulse may be used to perform the adiabatic passage on an example of two-photon excitation to the Rydberg state in Rb.

Two-photon adiabatic passage to Rydberg state in Rb atom



$$E_{p,S}(t) = E_{p0,S0}e^{-t^2/\tau_0^2}\cos(\omega_{p,S}t + \alpha t^2/2)$$

Single Rydberg atom excitation

- Adiabatic rapid passage for a deterministic single Rydberg atom excitation within an ensemble of atoms.

- It is advantageous compared to π-pulse solution because it does not require explicit pulse duration and Rabi frequency [Rabi frequency of single excitation in collective state is collectively enhanced by $N^{1/2}$: $\Omega_N = \Omega_1 N^{1/2}$]

- One-photon passage may be inconveniently to implement to Rydberg excitations because it requires chirped pulse in the ultraviolet region; (Beterov *et al*, PRA **84** 023413 (2011))

Schrödinger equation in the field-interaction representation and RWA

$$i\begin{pmatrix}\dot{a}_g\\ \dot{a}_i\\ \dot{a}_r\end{pmatrix}=\begin{pmatrix}0 & \Omega_p/2 & 0\\ \Omega_p/2 & \Delta-\alpha t & \Omega_S/2\\ 0 & \Omega_S/2 & \delta-(\alpha+\beta)t\end{pmatrix}\begin{pmatrix}a_g\\ a_i\\ a_r\end{pmatrix}$$

$$\Omega_p=-\mu_{12}E_p(t)/\hbar,\ \Omega_S=-\mu_{23}E_S(t)/\hbar$$

$$\omega_2-\omega_p=\Delta-\text{one-photon detuning}\ (\omega_1=0)$$

$$\omega_3-\omega_p-\omega_S=\delta-\text{two-photon detuning}$$

Dressed state picture (1)

$$\Delta >> \Omega_p, \Omega_S$$

$$H = \begin{bmatrix} \frac{\Omega_2 - \Omega_1}{2} - \frac{\delta}{2} + \alpha t & -\Omega_3 \\ -\Omega_3 & -\left(\frac{\Omega_2 - \Omega_1}{2} - \frac{\delta}{2} + \alpha t \right) \end{bmatrix}$$

$$\Omega_1 = \frac{\Omega_p^2}{4(\Delta - \alpha t)}, \; \Omega_2 = \frac{\Omega_S^2}{4(\Delta - \alpha t)}, \; \Omega_3 = \frac{\Omega_p \Omega_S}{4(\Delta - \alpha t)}$$

Dressed state picture (2)

$$\lambda_{1,2} = \pm\sqrt{\left(\frac{\Omega_2 - \Omega_1}{2} - \frac{\delta}{2} + \alpha t\right)^2 + \Omega_3^2};\ \left|\Psi\right\rangle = \cos\Theta\left|1\right\rangle + \sin\Theta\left|3\right\rangle \textit{has lower energy}$$

We want that initially $\cos = 1$ *and* $\sin = 0$ *and at the end oppositly* $\cos = 0$ *and* $\sin = 1$

We analyze $\tan\Theta$ *and choose two – photon detuning to be blue – shifted* :

$$\tan\Theta = \frac{c_3}{c_1} = \frac{1}{\Omega_3}\left(\frac{\Omega_2 - \Omega_1}{2} - \frac{\delta}{2} + \alpha t + \sqrt{\left(\frac{\Omega_2 - \Omega_1}{2} - \frac{\delta}{2} + \alpha t\right)^2 + \Omega_3^2}\right)$$

$t \approx 0\ \ \delta > 0$ *indeed* $\tan\Theta \approx 0$ *means* $\sin\Theta = 0\ \cos\Theta = 1$

$t \approx \infty$, *to get* $\tan\Theta \to \infty$ *the condition* $(-\delta/2 + \alpha t) > 0$ *must be satisfied*

then, $\sin\Theta = 1\ \cos\Theta = 0$

means : *adiabaticity condition*: $\left|\alpha\right|\tau > \left|\frac{\delta}{2}\right|$

Phys. Scr. **T160** (2014) 014024

Adiabaticity conditions by means of using two linearly chirped pulses

$$E_{p,S}(t) = E_{p0,S0} e^{-t^2/\tau_0^2} \cos(\omega_{p,S} t + \alpha t^2 / 2)$$

$$|\alpha|\tau > |\delta| /2$$

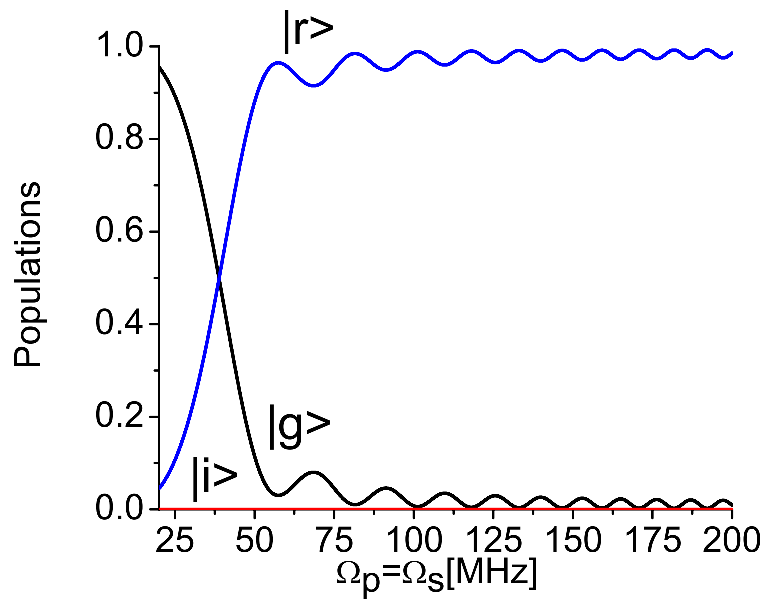
$$Landau - Zener\ adiabaticity\ param.\ \Omega^2_{p,S} / \alpha >> 1$$

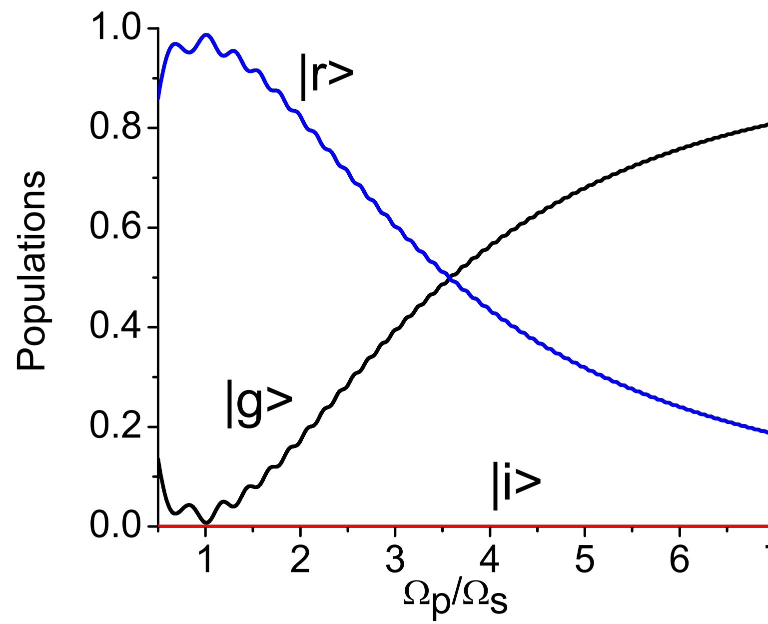
$$P_{i \to j} = P_i - \exp(-\pi \frac{\Omega_{ij}^2}{2\alpha})$$

$$L.D.Landau, Phys.Z.Sowjetunion\ 2, 46 (1932)$$

$$\Omega_p \simeq \Omega_s,\ E_{s0} \gg E_{p0},\ \frac{\Omega_p}{\Omega_s} = \frac{\mu_{21} E_{p0}}{\mu_{32} E_{s0}}.$$

Exact solution of the Schrödinger equation



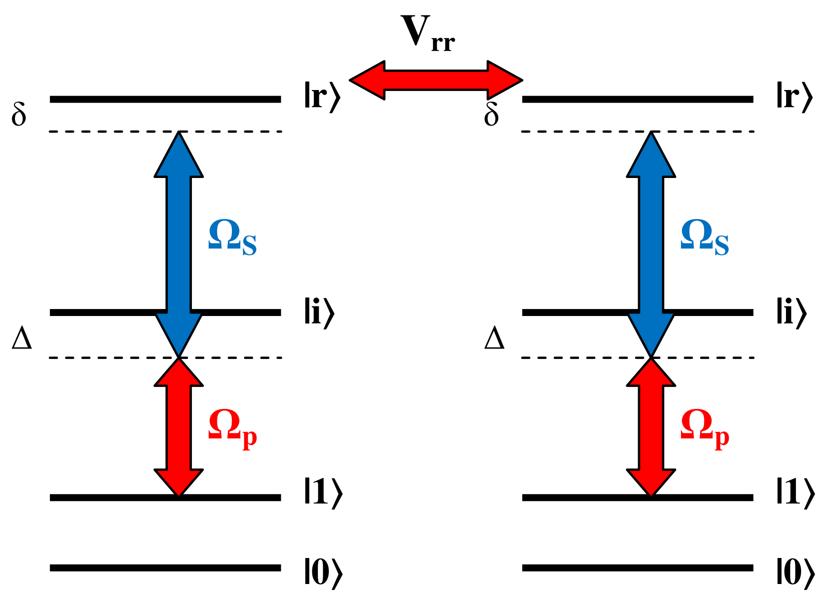


(left) Populations of the atomic states depending on the Rabi frequencies of the fields. Pulse duration 1μs, chirp rates 4.2 MHz/μs, one-photon detuning 1.5 GHz, two-photon detuning 1.5MHz;
(right) Populations of the atomic states depending on the ratio of the pump and Stokes Rabi frequencies.

Excitation of two atoms to the Rydberg states

- The idea is to realize a two-qubit phase gate based on Rydberg-Rydberg interaction and using two chirped pulses analogously to the previous case.
- The key ingredient is the ability to excite two atoms to the Rydberg states.
- Our work is an alternative to suggested implementation of STIRAP for two-atom excitation in the presence of dipole-dipole interaction.
- The goal is to improve performance of the gate, particularly, in our scheme there is no need to impose a restriction on one-photon detuning.

Two-atom excitation to the Rydberg states



$$E_{p,S}(t) = E_{p0,S0} e^{-t^2/\tau_0^2} \cos(\omega_{p,S} t + \alpha t^2 / 2)$$

Pulse interaction is much faster than all decays.

Set of equations used for exact numerical solution

$$\left|\Psi\right\rangle = c_{gg}\left|gg\right\rangle + c_{\pm,gi}\left|\pm\right\rangle_{gi} + c_{ii}\left|ii\right\rangle + c_{\pm,ir}\left|\pm\right\rangle_{ir} + c_{\pm,gr}\left|\pm\right\rangle_{gr} + c_{rr}\left|rr\right\rangle$$

$$e.g.\quad \left|\pm\right\rangle_{gi} = (\left|gi\right\rangle + \left|ig\right\rangle)/\sqrt{2};$$

$$i\frac{dc_{gg}}{dt} = \sqrt{2}\Omega_p c_{+,gi}$$

$$i\frac{dc_{+,gi}}{dt} = \Delta c_{+,gi} + \sqrt{2}\Omega_p c_{gg} + \sqrt{2}\Omega_p c_{ii} + \Omega_S c_{+,gr}$$

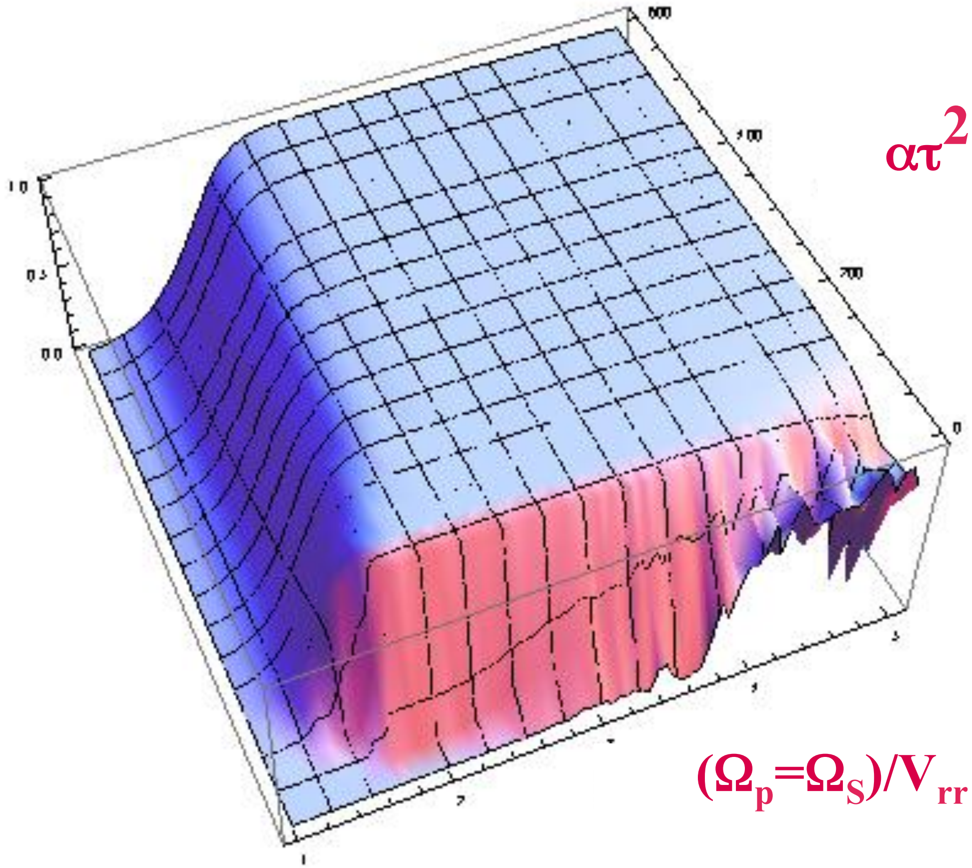
$$i\frac{dc_{ii}}{dt} = 2\Delta c_{ii} + \sqrt{2}\Omega_p c_{+,gi} + \sqrt{2}\Omega_p c_{ir}$$

$$i\frac{dc_{+,gr}}{dt} = \delta c_{+,gr} + \Omega_S c_{+,gi} + \Omega_p c_{+,ir}$$

$$i\frac{dc_{+,ir}}{dt} = (\delta+\Delta)c_{+,ir} + \sqrt{2}\Omega_S c_{ii} + \sqrt{2}\Omega_S c_{rr} + \Omega_p c_{+,gr}$$

$$i\frac{dc_{rr}}{dt} = (2\delta + V_{rr})c_{rr} + \sqrt{2}\Omega_S c_{ir} \qquad \Delta = \omega_{ig} - \omega_p;\ \ \delta = \omega_{rg} - \omega_p - \omega_S$$

Exact solution:
population of two-atomic excited Rydberg state at the end of the pulse as a function of normalized peak Rabi frequencies and chirp rate



V_{rr} =60 MHz , τ=1.5μs

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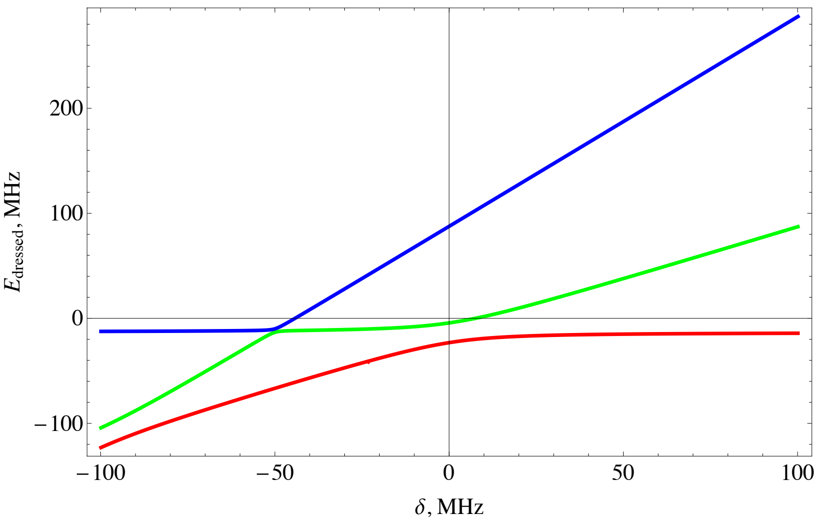
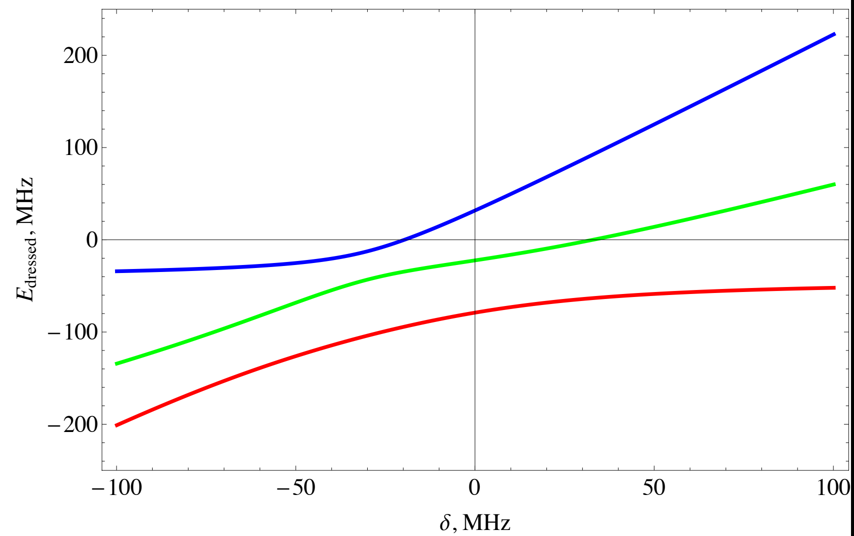
Dressed state picture for two-atomic Rydberg excitation

$$H = \begin{bmatrix} -\frac{2\Omega_p^2}{\Delta} & -\frac{\sqrt{2}\Omega_p\Omega_S}{\Delta} & 0 \\ -\frac{\sqrt{2}\Omega_p\Omega_S}{\Delta} & \delta - \frac{\Omega_p^2 + \Omega_S^2}{\Delta} & -\frac{\sqrt{2}\Omega_p\Omega_S}{\Delta} \\ 0 & -\frac{\sqrt{2}\Omega_p\Omega_S}{\Delta} & 2\delta + V_{rr} - \frac{2\Omega_S^2}{\Delta} \end{bmatrix}$$

Energies of the dressed states as a function of two-photon detuning

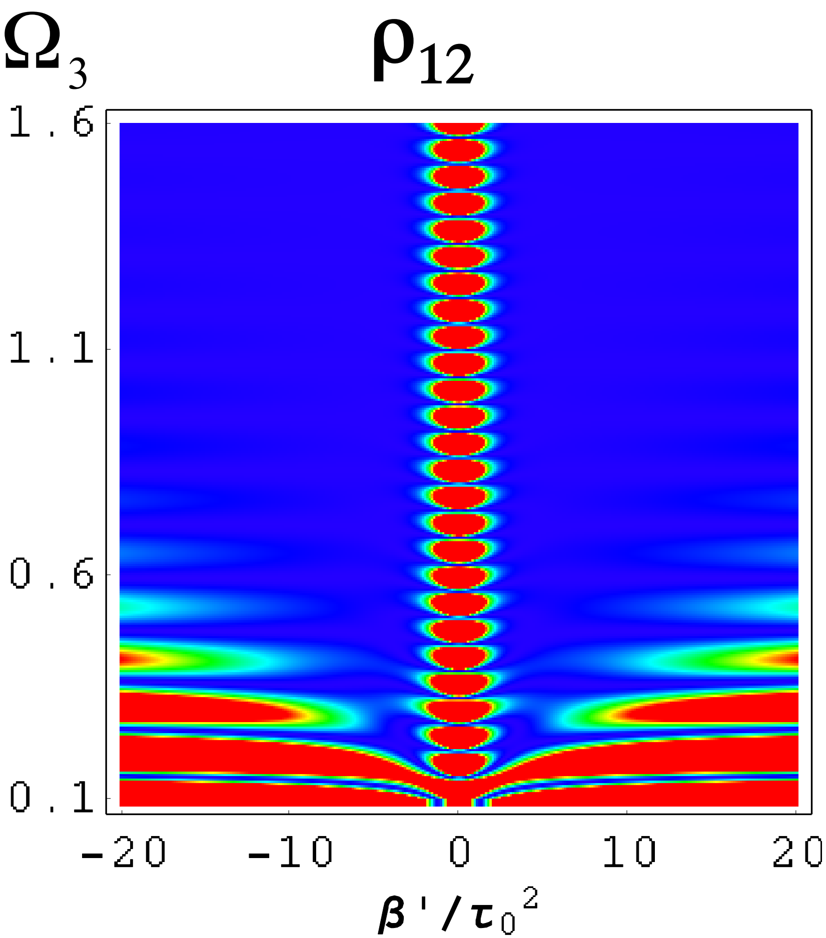


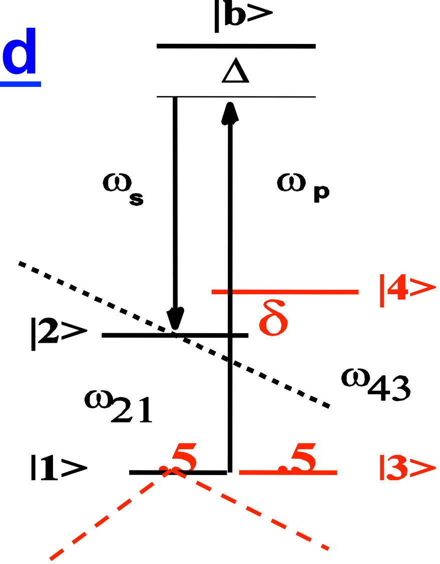


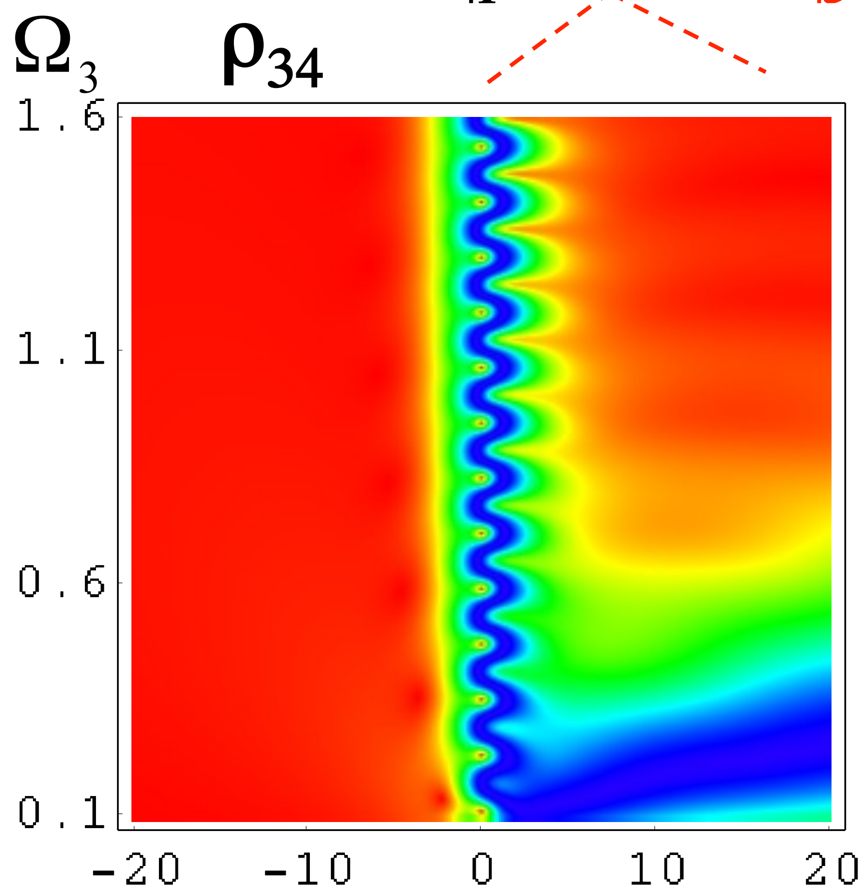
$V_{rr}=\Omega_{p,S}=60MHz$
$\Delta=1.5GHz$

$V_{rr}=60MHz;\ \Omega_{p,S}=180MHz$
$\Delta=1.5GHz$

Selective excitation of vibrational modes and creation of a maximum coherence for noninvasive molecular specific imaging



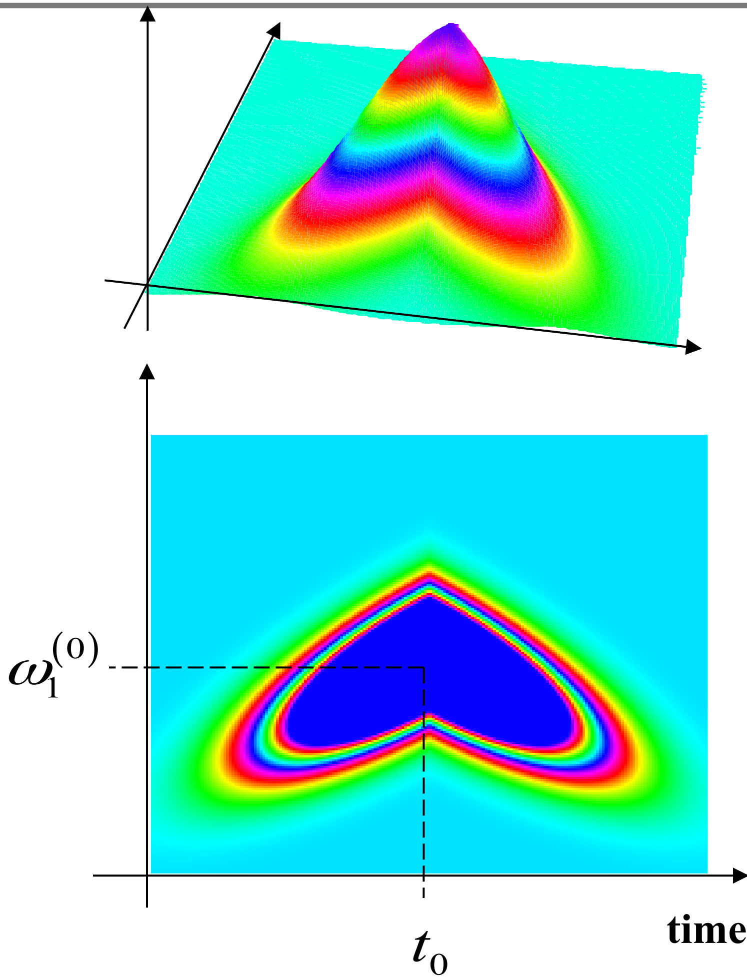


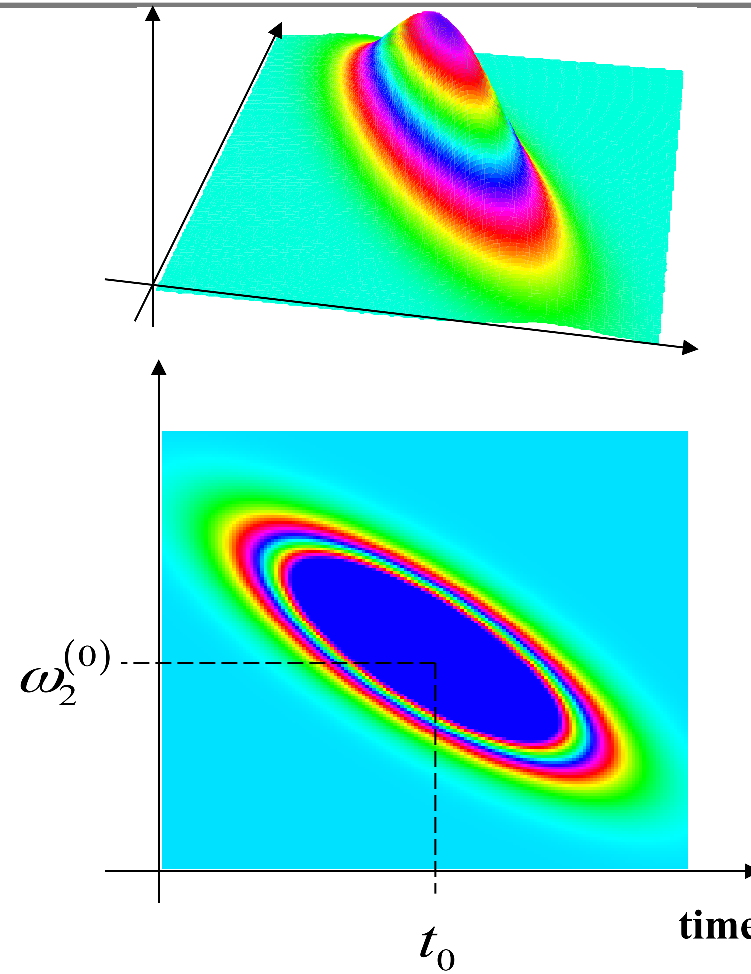


Blue $\rho = \rho_{max}$ **, red** $\rho = 0.$

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

Wigner plots of the pump (left) and Stokes (right) pulses





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Taking into account decoherence

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

Reduced density matrix elements are

$$\left.\dot{\rho}_{11}\right)_{cp} = \gamma_{2,1}\rho_{22} - \gamma_{3,1}\rho_{11}$$

$$\left.\dot{\rho}_{22}\right)_{cp} = -\gamma_{2,1}\rho_{22} - \gamma_{2,3}\rho_{22}$$

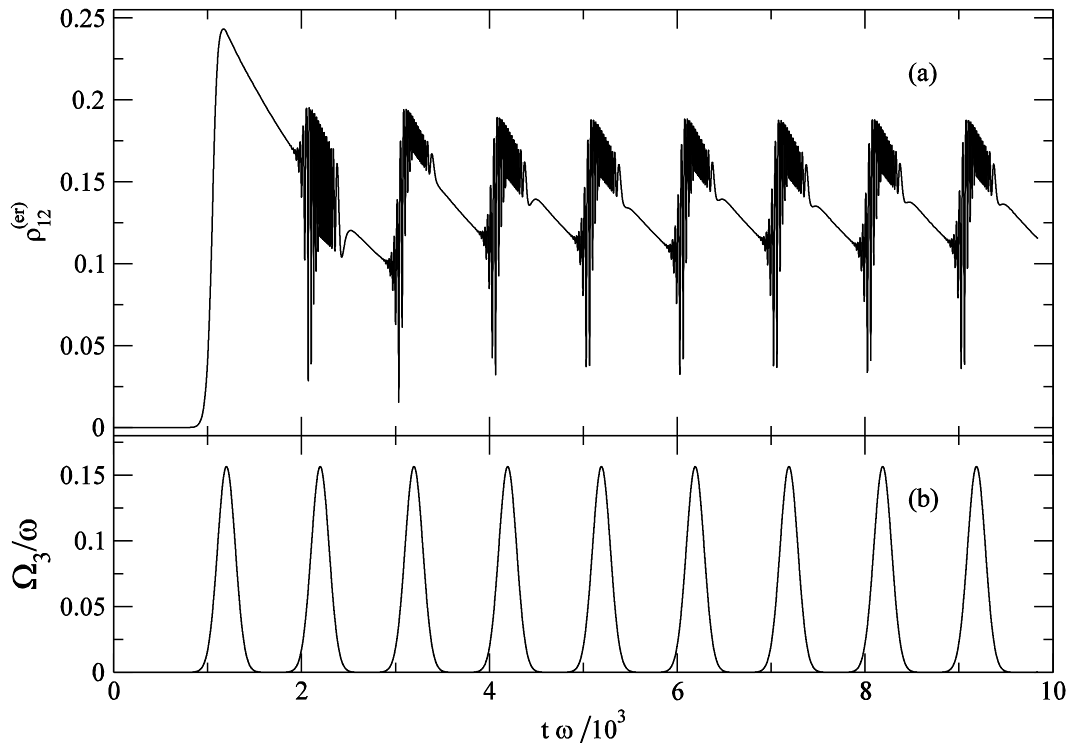
$$\left.\dot{\rho}_{33}\right)_{cp} = \gamma_{2,3}\rho_{22} + \gamma_{3,1}\rho_{11}$$

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

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

$$\left.\dot{\rho}_{23}\right)_{cp,col} = -\left(\frac{\gamma_{2,3}}{2} + \Gamma_{23}\right)\rho_{23}$$

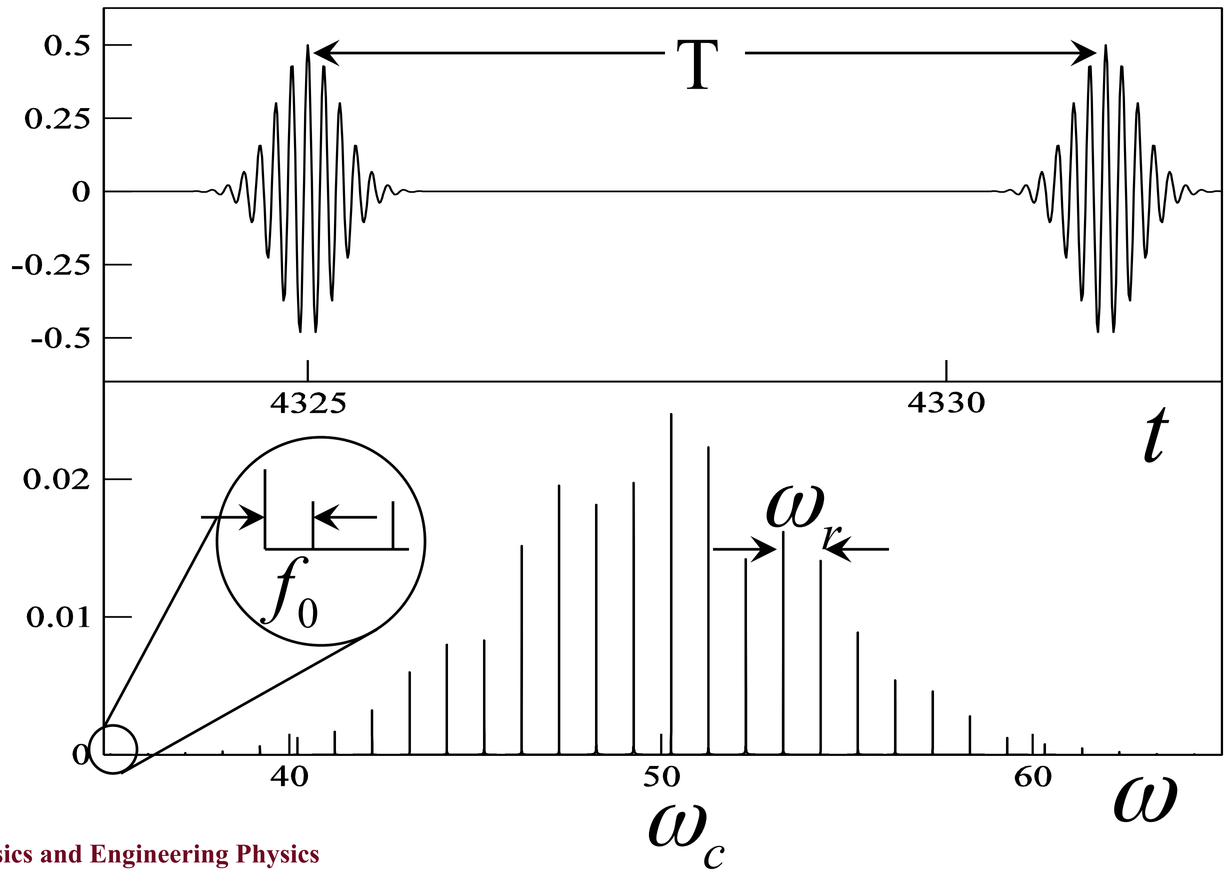
Effect of two pulse trains having same period as vibrational energy relaxation time: coherence drops to 0.1 and varies within 0.1-0.15 region



S. A. Malinovskaya, \Prevention of decoherence by two femtosecond chirped pulse trains", Opt. Lett. 33, 2245-2247 (2008);
S.A. Malinovskaya, "Robust control by two chirped pulse trains in the presence of decoherence", J. Mod. Opt. **56**, 784 (2009)

Control using Frequency Combs

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



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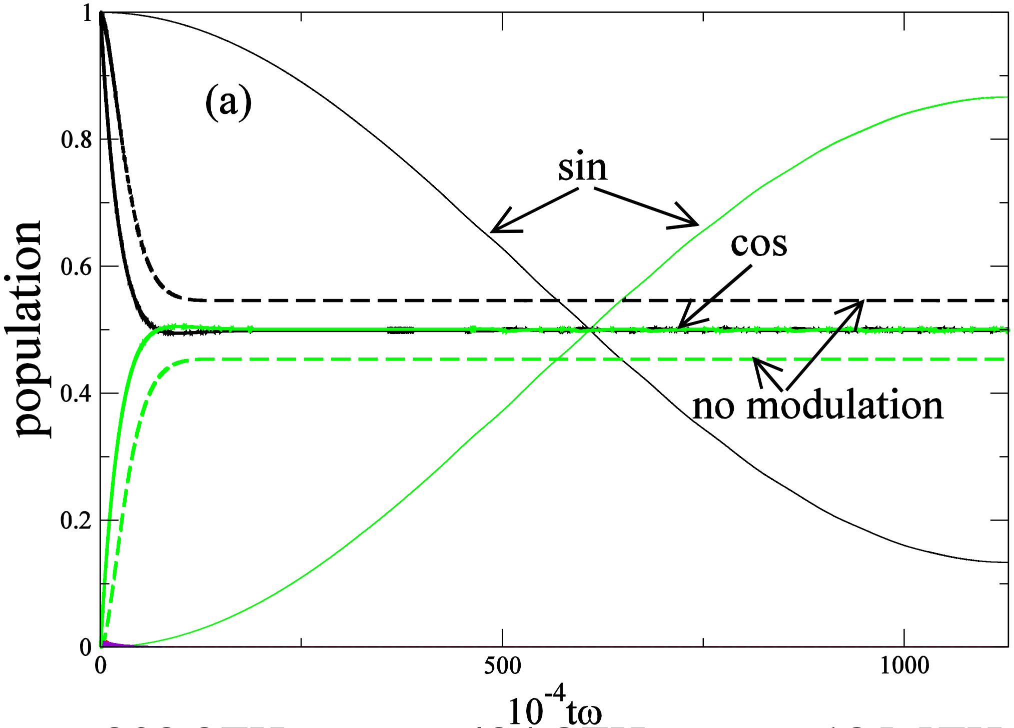
Population dynamics induced by sine/cosine and standard OFCs

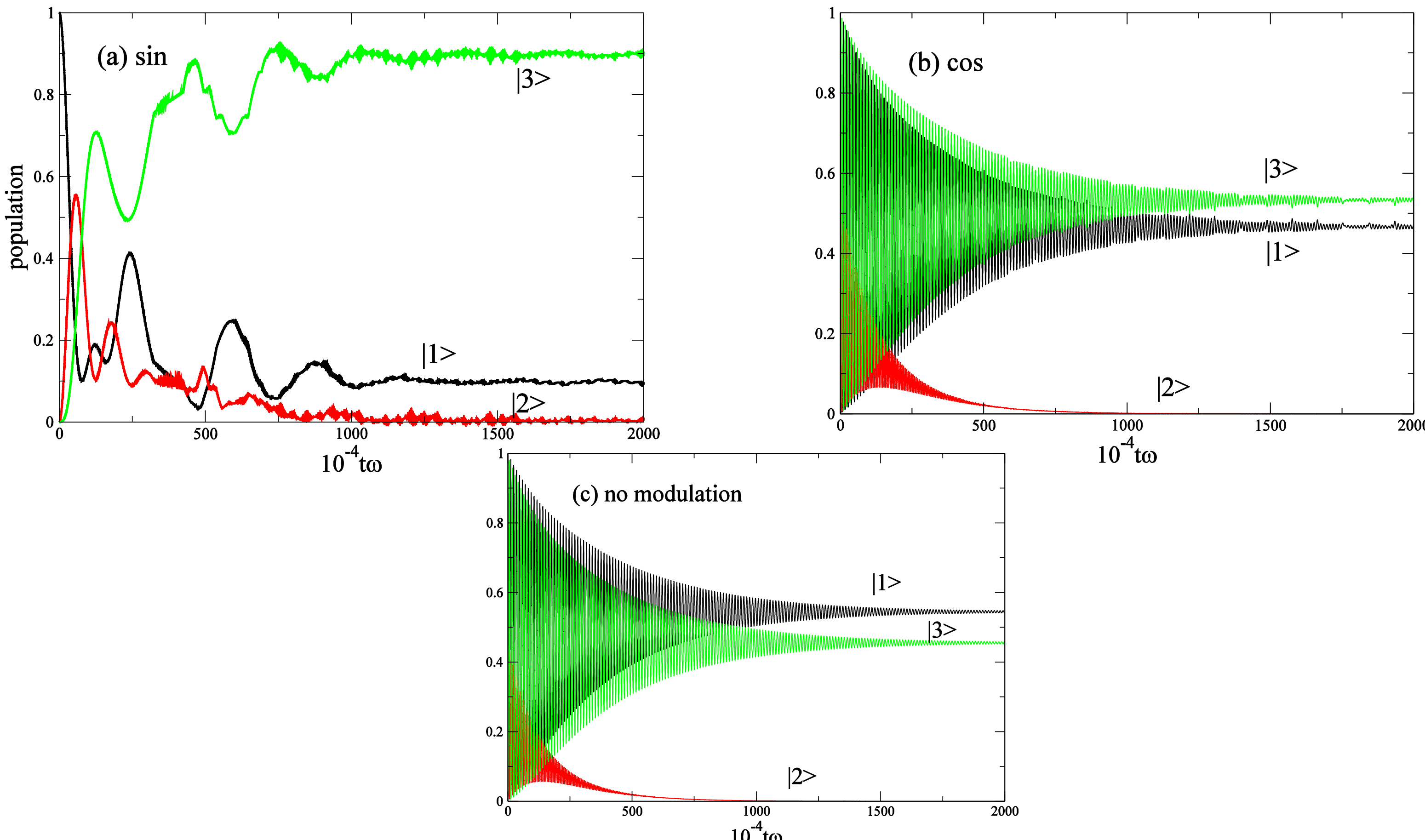


$$J.Ye:\omega_{21}=309.3THz \quad \omega_{32}=434.8THz \quad \omega_{31}=125.5THz$$

$$\omega_L=\omega_{32},\ \Omega=\omega_{21},\ \Omega_R=12.5THz \quad \gamma_{21}=\gamma_{23}=10^{-3},\ \Gamma_{32}=\Gamma_{12}=10^{-3}$$

S.A. Malinovskaya, S.L. Horton, "Rovibrational cooling using optical frequency combs in the presence of decoherence," J. Opt. Soc. Am. B 30, 482 (2013)

Population dynamics in the presence of experimental decoherence



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

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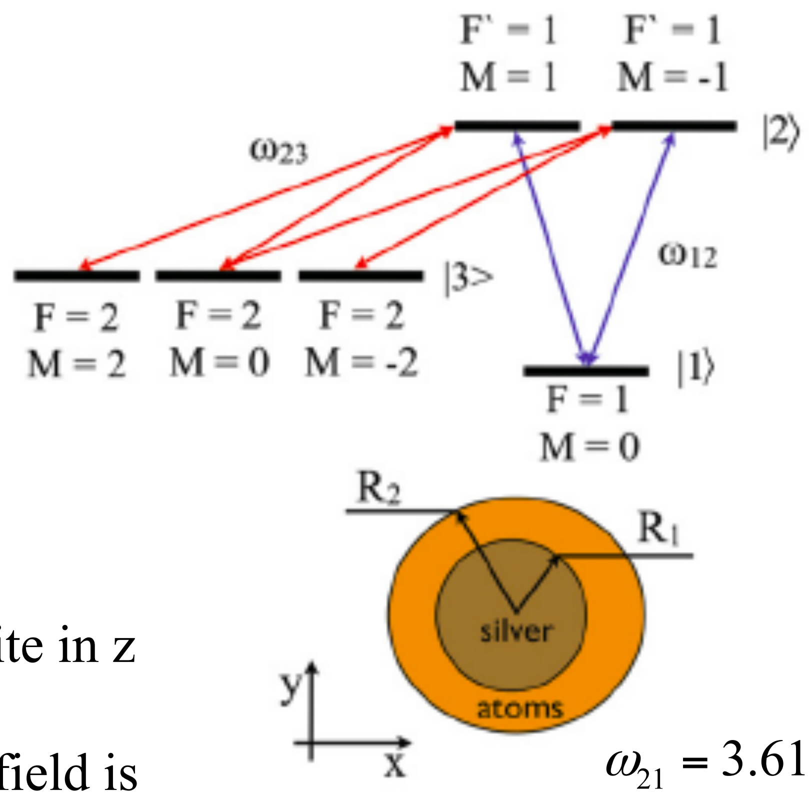
Silver nanowire covered by a thin layer of three-level atoms

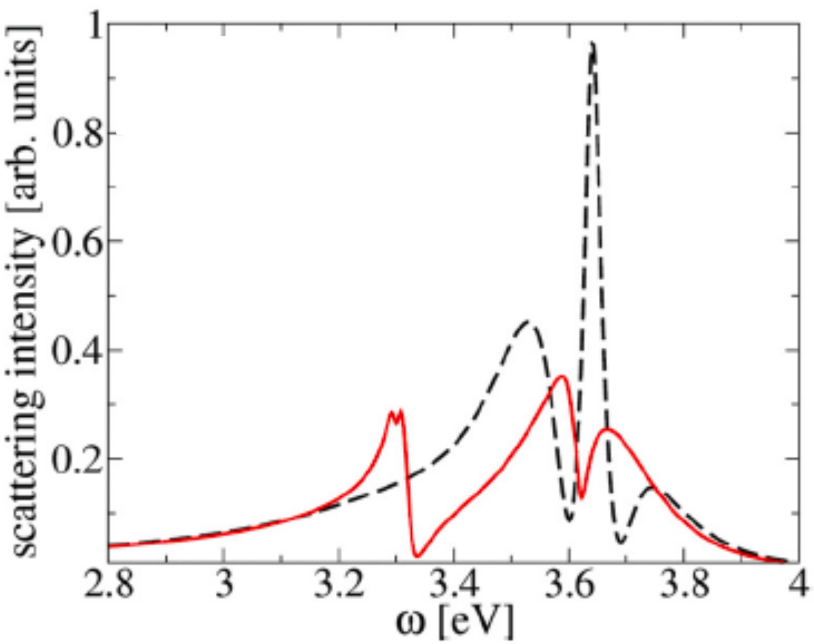


The wire is infinite in z direction;
Incident electric field is polarized along x-axis, and propagates along y-axis

$$\omega_{21} = 3.61eV, \omega_{32} = 3.3eV$$
$$R_1 = 20nm, R_2 = 35nm$$

Scattering intensity of the core-shell nanowire after STIRAP

M. Sukharev, S.A. Malinovskaya, PRA 86, 043406 (2012)



$$n_a = 5\times10^{27} m^{-3}, black - before\ STIRAP$$
$$red - after\ STIRAP$$

- The spectrum after STIRAP (red) differs from that before (black)
- The Rabi splitting from 216 meV reduced to 78meV
- The collective atom-plasmon mode is not seen
- A new resonance near atomic frequency ω_{23} is observed due to the presence of inverted atoms
- Tiny Rabi splitting in 3meV near the later resonance
- So, the scattering spectrum manifests double Rabi splittings associated with two atomic transitions

Thank you!

- Gengyuan Liu, PhD student
- Vishesha Patel, PhD 2010, Financial Software Trainer at NumeriX
- Thomas Collins, PhD 2012, Assistant Professor, Hofstra University
- Elena Kuznetzova, ITAMP, Harvard-Smithsonian Center for Astrophysics Cambridge, MA 02138, USA, Russian Quantum Center, Skolkovo, Russia
- Vladimir Malinovsky, Army Research Lab, Stevens Institute of Technology
- Maxim Sukharev, Arizona State University
- Phil Gould, University of Connecticut

National Science Foundation
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