ADIABATIC PASSAGE TECHNIQUES IN SIMPLE QUANTUM SYSTEMS

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Outlook

Two basic approaches

- one or more level crossings
- delayed (but overlapped) pulses

Systems

- two states
- three states
- multiple states

Types of excitation

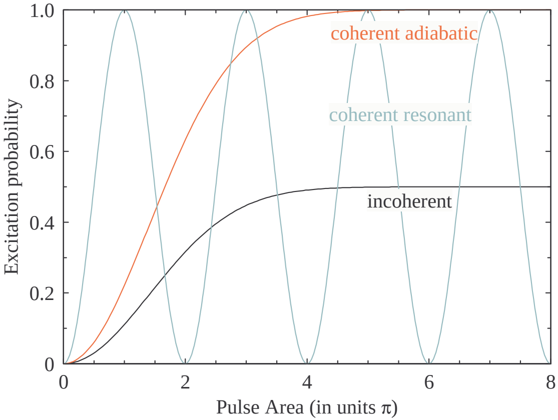


FIGURE: Incoherent excitation (black), Rabi oscillations (blue) and adiabatic passage (red).

Coherent excitation

$i\hbar \frac{d}{dt}\mathbf{c}(t) = \mathbf{H}(t)\mathbf{c}(t)$ time-dependent **Schrödinger equation**

$\mathbf{c}(t) = [c_1(t), c_2(t)]^T$ vector with the **probability amplitudes** of ψ_1 and ψ_2

$P_n(t) = |c_n(t)|^2$ $(n = 1, 2)$ **populations**

$\mathbf{H}(t) = \hbar \begin{bmatrix} 0 & \frac{1}{2}\Omega(t) \\ \frac{1}{2}\Omega(t) & \Delta(t) \end{bmatrix}$ **Hamiltonian** in Rotating-Wave Approximation

$\Omega(t)$ the **Rabi frequency**: $\Omega(t) = -\mathbf{d}_{12} \cdot \mathcal{E}(t)/\hbar$ for electric-dipole transitions, with $\mathbf{d}_{12}$ the transition dipole moment and $\mathcal{E}(t)$ the electric field envelope

$\Delta = \omega_0 - \omega$ atom-laser **detuning** between the transition frequency ω_0 and the laser frequency ω

Adiabatic states

$$\mathbf{H}(t) = \hbar \begin{bmatrix} 0 & \frac{1}{2}\Omega(t) \\ \frac{1}{2}\Omega(t) & \Delta(t) \end{bmatrix} \text{ Hamiltonian}$$

adiabatic states: the instantaneous eigenstates of the Hamiltonian

$$\varphi_+(t) = \psi_1 \sin\vartheta(t) + \psi_2 \cos\vartheta(t) \qquad \mathbf{H}(t)\varphi_+(t) = \hbar\varepsilon_+(t)\varphi_+(t)$$

$$\varphi_-(t) = \psi_1 \cos\vartheta(t) - \psi_2 \sin\vartheta(t) \qquad \mathbf{H}(t)\varphi_-(t) = \hbar\varepsilon_-(t)\varphi_-(t)$$

$$\vartheta(t) = \tfrac{1}{2}\arctan[\Omega(t)/\Delta(t)] \quad \text{mixing angle}$$

adiabatic energies: the eigenvalues of $\mathbf{H}(t)$

$$\hbar\varepsilon_\pm(t) = \tfrac{1}{2}\hbar\left[\Delta(t) \pm \sqrt{\Delta^2(t) + \Omega^2(t)}\right]$$

adiabatic evolution: no transitions between the adiabatic states

Eigenenergy splitting

Eigenenergy splitting

$$\hbar\varepsilon_+(t) - \hbar\varepsilon_-(t) = \hbar\sqrt{\Delta^2(t) + \Omega^2(t)} \geqq \hbar\Delta(t)$$

The interaction pushes away the adiabatic energies from each other!

level crossing of diabatic energies $\longrightarrow$ avoided crossing of adiabatic enegies

$$\Delta(t_c) = 0 \Longrightarrow \hbar\varepsilon_+(t_c) - \hbar\varepsilon_-(t_c) = \hbar\Omega(t_c)$$

The splitting of the avoided crossing is equal to the Rabi frequency!

Adiabatic evolution

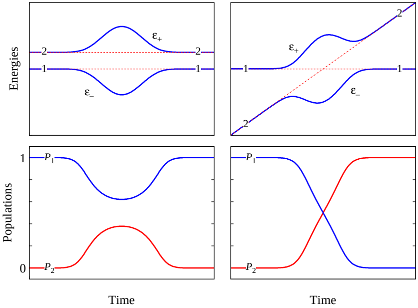


Figure: Adiabatic passage. **Left**: No crossing. **Right**: Level crossing

If the statevector $\Psi(t)$ coincides with an adiabatic state $\varphi(t)$ at some time t, then it will remain in that adiabatic state as long as the evolution is adiabatic: the statevector $\Psi(t)$ will adiabatically *follow* the adiabatic state $\varphi(t)$.

Adiabatic evolution: NO-CROSSING CASE

assumptions

- $\Delta(t) > 0$ (no crossing)
- $\Omega(t) > 0$ (the transition probability does not depend on the overall signs)
- $\Omega(t) \xrightarrow{t\to\pm\infty} 0$ (pulsed field)

$$\Downarrow$$

$$\infty \xleftarrow{-\infty\leftarrow t} \Delta(t)/\Omega(t) \xrightarrow{t\to+\infty} \infty$$

$$0 \xleftarrow{-\infty\leftarrow t} \vartheta(t) = \tfrac{1}{2}\arctan[\Omega(t)/\Delta(t)] \xrightarrow{t\to+\infty} 0$$

Asymptotically, each adiabatic state tends to the same unperturbed state

$$\psi_2 \xleftarrow{-\infty\leftarrow t} \quad \varphi_+(t) = \psi_1 \sin\vartheta(t) + \psi_2 \cos\vartheta(t) \quad \xrightarrow{t\to+\infty} \psi_2$$

$$\psi_1 \xleftarrow{-\infty\leftarrow t} \quad \varphi_-(t) = \psi_1 \cos\vartheta(t) - \psi_2 \sin\vartheta(t) \quad \xrightarrow{t\to+\infty} \psi_1$$

$\Longrightarrow$ starting from the ground state ψ_1 initially, the population makes a partial excursion into the excited state ψ_2 at intermediate times and eventually returns to ψ_1 in the end: **complete population return**

Adiabatic evolution: LEVEL CROSSING

assumptions

- $\Delta(t_c) = 0$ (crossing at time t_c) and $\dot{\Delta}(t_c) > 0$
- $\Omega(t) > 0$ (the transition probability does not depend on the overall signs)
- $\Omega(t) \stackrel{t\to\pm\infty}{\longrightarrow} 0$ (pulsed field)

$$\Downarrow$$

$$-\infty \stackrel{-\infty\leftarrow t}{\longleftarrow} \Delta(t)/\Omega(t) \stackrel{t\to+\infty}{\longrightarrow} \infty$$

$$\tfrac{1}{2}\pi \stackrel{-\infty\leftarrow t}{\longleftarrow} \vartheta(t) = \tfrac{1}{2}\arctan[\Omega(t)/\Delta(t)] \stackrel{t\to+\infty}{\longrightarrow} 0$$

Asymptotically, each adiabatic state tends to different unperturbed state

$$\psi_1 \stackrel{-\infty\leftarrow t}{\longleftarrow} \quad \varphi_+(t) = \psi_1 \sin\vartheta(t) + \psi_2\cos\vartheta(t) \quad \stackrel{t\to+\infty}{\longrightarrow} \psi_2$$

$$-\psi_2 \stackrel{-\infty\leftarrow t}{\longleftarrow} \quad \varphi_-(t) = \psi_1 \cos\vartheta(t) - \psi_2\sin\vartheta(t) \quad \stackrel{t\to+\infty}{\longrightarrow} \psi_1$$

$\Longrightarrow$ the population passes gradually from state ψ_1 to state ψ_2

complete population transfer (inversion)

Adiabatic evolution

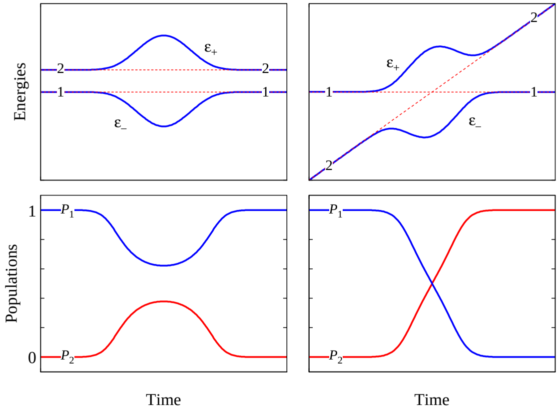


Figure: Adiabatic passage. **Left**: No crossing. **Right**: Level crossing

Adiabatic basis

Diabatic (unperturbed) basis $\mathbf{c}(t) = [c_1(t), c_2(t)]^T$

$$i\hbar \frac{d}{dt}\mathbf{c}(t) = \mathbf{H}(t)\mathbf{c}(t) \qquad \mathbf{H}(t) = \hbar \begin{bmatrix} 0 & \frac{1}{2}\Omega(t) \\ \frac{1}{2}\Omega(t) & \Delta \end{bmatrix}$$

Time-dependent rotation

$$\mathbf{c}(t) = \mathbf{R}[\vartheta(t)]\mathbf{a}(t) \qquad \mathbf{R}(\vartheta) = \begin{bmatrix} \cos\vartheta & \sin\vartheta \\ -\sin\vartheta & \cos\vartheta \end{bmatrix}$$

Adiabatic (dressed) basis $\mathbf{a}(t) = [a_-(t), a_+(t)]^T$

$$i\hbar \frac{d}{dt}\mathbf{a}(t) = \mathbf{H}_a(t)\mathbf{a}(t) \qquad \mathbf{H}_a = \hbar \begin{bmatrix} \lambda_- & -i\dot{\vartheta} \\ i\dot{\vartheta} & \lambda_+ \end{bmatrix}$$

$$\mathbf{H}_a = \mathbf{R}(-\vartheta)\mathbf{H}\mathbf{R}(\vartheta) - i\hbar\mathbf{R}(-\vartheta)\tfrac{d}{dt}\mathbf{R}(\vartheta)$$

Adiabatic solution

Adiabatic basis $\mathbf{a}(t) = [a_-(t), a_+(t)]^T$

$$i\hbar \frac{d}{dt}\mathbf{a}(t) = \mathbf{H}_a(t)\mathbf{a}(t) \qquad \mathbf{H}_a = \hbar \begin{bmatrix} \lambda_- & -i\dot{\vartheta} \\ i\dot{\vartheta} & \lambda_+ \end{bmatrix}$$

In the adiabatic limit: $|\dot{\vartheta}(t)| \ll \lambda_+ - \lambda_-$, and $\dot{\vartheta}(t)$ can be neglected. The adiabatic solution in the adiabatic basis is

$$\mathbf{U}_a(t, -\infty) = \begin{bmatrix} e^{-i\Lambda_-(t)} & 0 \\ 0 & e^{-i\Lambda_+(t)} \end{bmatrix} \qquad \Lambda_\pm = \int_{-\infty}^{t} \lambda_\pm(t')\mathrm{d}t'$$

The adiabatic amplitudes $\mathbf{a}(t) = \mathbf{U}_a(t, -\infty)\mathbf{a}(-\infty)$

The diabatic amplitudes

$$\begin{aligned} \mathbf{c}(t) &= \mathbf{R}[\vartheta(t)]\mathbf{a}(t) = \mathbf{R}[\vartheta(t)]\mathbf{U}_a(t, -\infty)\mathbf{a}(-\infty) \\ &= \mathbf{R}[\vartheta(t)]\mathbf{U}_a(t, -\infty)\mathbf{R}[-\vartheta(-\infty)]\mathbf{c}(-\infty) \end{aligned}$$

$$\mathbf{R}(\vartheta) = \begin{bmatrix} \cos\vartheta & \sin\vartheta \\ -\sin\vartheta & \cos\vartheta \end{bmatrix}$$

Adiabatic solution: Excitation probability

$$P(t) = \frac{1}{2} - \frac{\Delta(t)\Delta(t_i)}{2\lambda(t)\lambda(t_i)} - \frac{\Omega(t)\Omega(t_i)}{2\lambda(t)\lambda(t_i)} \cos\Lambda$$

$$\Lambda = \int_{-\infty}^{t} \lambda(t')\mathrm{d}t' \qquad \lambda(t) = \sqrt{\Omega^2(t) + \Delta^2(t)}$$

If $\Omega(t_i) = 0$ (pulsed field) then $P(t) = \frac{1}{2} - \frac{\Delta(t)\Delta(t_i)}{2\lambda(t)|\Delta(t_i)|}$

No-crossing case $(\Delta(t) > 0$ for definiteness$)$

$$P_2(t) = \frac{1}{2} - \frac{\Delta(t)}{2\sqrt{\Omega^2(t) + \Delta^2(t)}} \xrightarrow{t\to+\infty} 0$$

Level crossing $(\dot{\Delta}(t) > 0$ for definiteness $\Longrightarrow \Delta(t_i) < 0)$

$$P_2(t) = \frac{1}{2} + \frac{\Delta(t)}{2\sqrt{\Omega^2(t) + \Delta^2(t)}} \xrightarrow{t\to+\infty} 1$$

Adiabatic condition

$$i\hbar\frac{d}{dt}\mathbf{a}(t) = \mathbf{H}_a(t)\mathbf{a}(t) \qquad \mathbf{H}_a = \hbar\begin{bmatrix} \lambda_- & -i\dot\vartheta \\ i\dot\vartheta & \lambda_+ \end{bmatrix}$$

adiabatic evolution: no transitions between the adiabatic states

$\Longrightarrow$ adiabatic condition $|\dot\vartheta(t)| \ll \lambda_+(t) - \lambda_-(t)$ (coupling $\ll$ splitting)

$$\Longrightarrow |\Delta\dot\Omega(t) - \dot\Delta(t)\Omega(t)| \ll [\Omega^2(t) + \Delta^2]^{3/2}$$

adiabaticity function $\qquad f(t) = \dfrac{\Delta\dot\Omega(t)}{[\Omega^2(t) + \Delta^2]^{3/2}}$

adiabatic evolution $\Longleftrightarrow |f(t)|_{\max} < \epsilon \ll 1$

The adiabatic condition for each specific pair of $[\Omega(t), \Delta(t)]$ is derived by analyzing the function $f(t)$.
Generally adiabatic evolution requires smooth time dependences, long interaction time and large Rabi frequency and/or large detuning.

Adiabatic condition: Examples

Rosen-Zener model

$$\Omega(t) = \Omega_0 \operatorname{sech}(t/T) \qquad \Delta(t) = \text{const}$$

$$|\Delta| \gg \Delta_0 = \frac{1}{T}$$

the evolution is adiabatic for $|\Delta| \gtrsim \Delta_0$ and nonadiabatic for $|\Delta| \lesssim \Delta_0$
the adiabatic condition does *not* depend on the peak Rabi frequency Ω_0!

Gaussian model

$$\Omega(t) = \Omega_0 \exp(-t^2/T^2) \qquad \Delta(t) = \text{const}$$

$$\Delta \gg \Delta_0 \approx \frac{1}{T}\sqrt{\tfrac{4}{27}\ln(\Omega_0 T\sqrt{2})}$$

the evolution is adiabatic for $|\Delta| \gtrsim \Delta_0$ and nonadiabatic for $|\Delta| \lesssim \Delta_0$
the adiabatic condition depends only logarithmically on Ω_0

Adiabatic condition: Examples

Allen-Eberly model

$$\Omega(t) = \Omega_0 \text{sech}\,(t/T) \qquad \Delta(t) = B\tanh(t/T) \quad \text{chirp rate } B/T$$

$$\Omega_0^2 \gg B/T \qquad (\Omega_0 < B)$$
$$BT \gg 1 \qquad (\Omega_0 > B)$$

Gaussian model

$$\Omega(t) = \Omega_0 \exp(-t^2/T^2) \qquad \Delta(t) = Bt/T \quad \text{chirp rate } B/T$$

$$\Omega_0^2 \gg B/T \qquad (\Omega_0\sqrt{2} \leqq B)$$
$$BT \gg 1 \qquad (\Omega_0\sqrt{2} > B)$$

Landau-Zener model

$$\Omega(t) = \text{const} \qquad \Delta(t) = Bt/T$$
$$\Omega_0^2 \gg B/T$$

Exactly soluble two-state models

Rabi:	$\Omega(t) = \Omega_0 (t \leqq T/2), \quad \Delta(t) = \Delta_0$
Landau-Zener:	$\Omega(t) = \Omega_0, \quad \Delta(t) = Bt$
Rosen-Zener:	$\Omega(t) = \Omega_0 \operatorname{sech} t/T, \quad \Delta(t) = \Delta_0$
Allen-Eberly:	$\Omega(t) = \Omega_0 \operatorname{sech} t/T, \quad \Delta(t) = B \tanh t/T$
Bambini-Berman:	$\Omega(t) = \Omega_0 \operatorname{sech} t/T, \quad \Delta(t) = B(1 + \tanh t/T)$
Demkov-Kunike:	$\Omega(t) = \Omega_0 \operatorname{sech} t/T, \quad \Delta(t) = \Delta_0 + B \tanh t/T$
Hioe-Carroll:	$\Omega(t) = \Omega_0 \operatorname{sech} t/T, \quad \Delta(t) = S \operatorname{sech} t/T + B \tanh t/T$
Heun:	$\Omega(t) = \Omega_0 \operatorname{sech} t/T, \quad \Delta(t) = \Delta_0 + S \operatorname{sech} t/T + B \tanh t/T$
Demkov:	$\Omega(t) = \Omega_0 \exp(-\|t\|/T), \quad \Delta(t) = \Delta_0$
Nikitin:	$\Omega(t) = \Omega_0 \exp(-\|t\|/T), \quad \Delta(t) = \Delta_0 + B \exp(-\|t\|/T)$
Carroll-Hioe:	$\Omega(t) = \Omega_0 \exp(-\|t\|/T), \quad \Delta(t) = \Delta_0 + B \exp(-2\|t\|/T)$

allow to study the full ranges of parameters, beyond the adiabatic regime

Adiabatic passage: No crossing

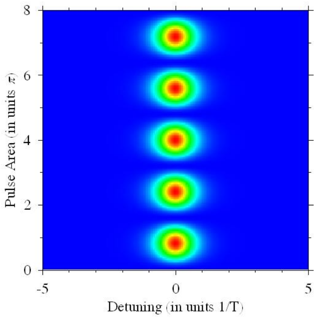


Figure: **Left — Rosen-Zener model:** $\Omega(t) = \Omega_0 \text{sech}\,(t/T),\ \Delta(t) = \text{const.}$
Right — Gaussian model: $\Omega(t) = \Omega_0 \exp(-t^2/T^2),\ \Delta(t) = \text{const.}$

Adiabatic passage: Level crossing

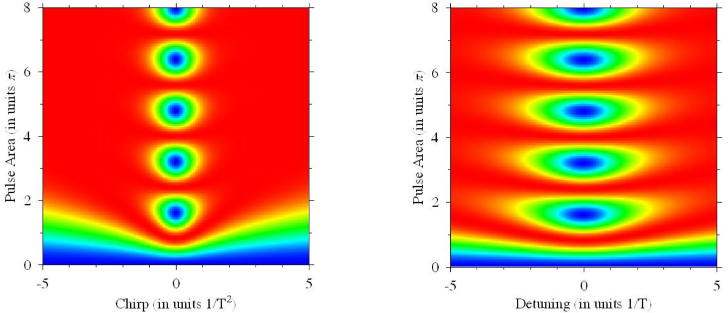


Figure: **Left — Allen-Eberly model:** $\Omega(t) = \Omega_0 \text{sech}\,(t/T),\ \Delta(t) = B\tanh(t/T)$.
Right — chirped Gaussian model: $\Omega(t) = \Omega_0 \exp(-t^2/T^2),\ \Delta(t) = Bt/T$.

Adiabatic evolution: Summary

- no transitions in the adiabatic basis (between the adiabatic states)
- for no-crossing energies in the original basis: complete population return
- for crossing energies in the original basis: complete population transfer
- population transfer robust agains variations of the interaction parameters (Rabi frequency, chirp rate, interaction duration): significant advantage over Rabi cycling
- adiabatic passage does not depend on the sign of the chirp: $\dot{\Delta}(t) > 0$ or $\dot{\Delta}(t) < 0$
- adiabatic passage does not depend on the sign of the Rabi frequency Ω
- adiabatic evolution requires smooth time dependences and large Rabi frequency and/or large detuning (the adiabatic condition may vary!)

Power broadening in steady excitation

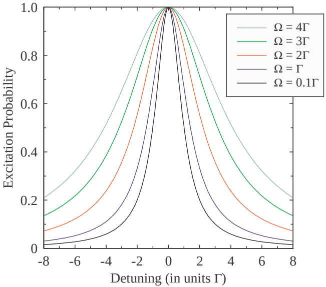


FIGURE: Lineshapes in steady excitation: typical power broadening.

Steady-state excitation of a two-state atom driven by cw laser field

$$P \propto \frac{1}{\Delta^2 + \Gamma^2/4 + \Omega^2/4}$$

POWER *non*-BROADENING IN PULSED EXCITATION

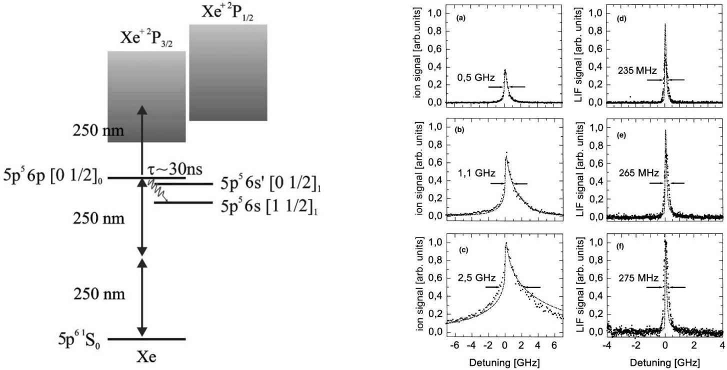


FIGURE: Ionization signal (left) and fluorescence (right) in xenon for pump intensities 0.75 GW/cm^2 (a,d), 2.5 GW/cm^2 (b,e) and 7.5 GW/cm^2 (c,f).

T. Halfmann, T. Rickes, N.V.V. and K. Bergmann, Opt. Commun. 220, 353 (2003)

Power *non*-broadening in pulsed excitation

Line width measured during excitation

$$I = \int_{-\infty}^{+\infty} \Gamma(t) P_2(t) \mathrm{d}t$$

The signal increases with the Rabi frequency [because $P_2(t)$ increases with Ω]
$\Longrightarrow$ typical power broadening.

Line width measured after excitation

Excitation probability in the adiabatic limit

$$P_2(t) = \frac{1}{2} - \frac{\Delta(t)}{2\sqrt{\Omega^2(t) + \Delta^2(t)}} \xrightarrow{t\to+\infty} 0$$

No excitation outside the nonadiabatic region $|\Delta| \lesssim 1/T$
$\Longrightarrow$no dependence on the Rabi frequency Ω_0!
$\Longrightarrow$ no power broadening!

N.V.V., B.W.Shore, L.Yatsenko, K.Böhmer, T.Halfmann, T.Rickes, K.Bergmann, Opt. Commun. 199, 117 (2001)

Level crossing: implementations

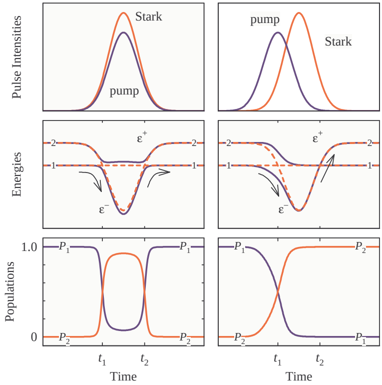
Traditional techniques

- pulse shaping for femtosecond and picosecond pulses (chirp $\omega_L(t)$)
- AOM and EOM for microsecond pulses (chirp $\omega_L(t)$)
- electric fields (chirp $\omega_A(t)$)
- magnetic fields (chirp $\omega_A(t)$)

Other methods (suitable for nanosecond pulses)

- Stark-chirped rapid adiabatic passage (SCRAP) (laser-induced dynamic Stark shift in ω_A)
- retroreflection-induced bichromatic adiabatic passage (RIBAP) (crossing of Floquet energies)
- superadiabatic passage (SAP) (crossing in adiabatic basis)

SCRAP: Stark-chirped rapid adiabatic passage



Evolution of the laser pulses, the level energies and the populations for coincident and delayed pump and Stark pulses

- a strong far-off-resonant laser pulse Stark shifts the level energies
- a nearly-resonant pump laser pulse is applied with a suitable detuning
- the Stark pulse and the pump detuning create two level crossings
- the driving pulse is applied at one of the induced level crossings
- the laser parameters are chosen such that ensure diabatic evolution at one crossing and adiabatic at the other
- this adiabatic-diabatic scenario produces complete population transfer

SCRAP: EXPERIMENT

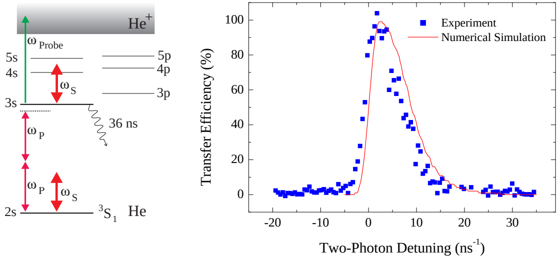
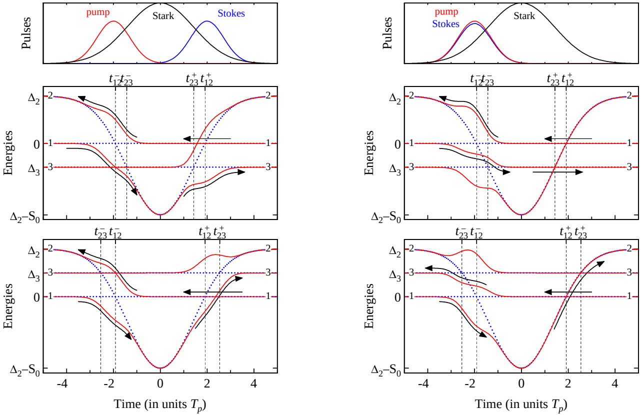


FIGURE: Experimental demonstration of SCRAP in helium

T.Rickes,L.P.Yatsenko,S.Steuerwald,T.Halfmann,B.W.Shore,N.V.V.,K.Bergmann, J.Chem.Phys. 113, 534 (2000)

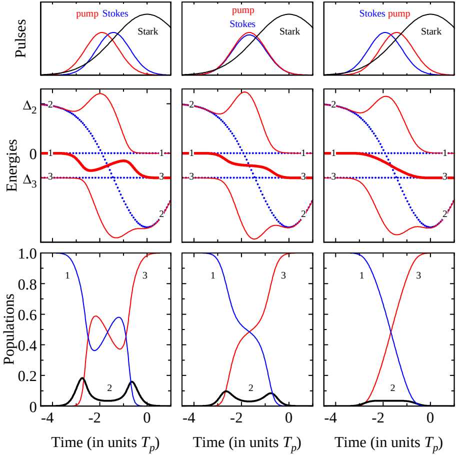
L.P.Yatsenko, A.Vardi, T.Halfmann, B.W.Shore, K.Bergmann, Phys. Rev. A 60, R4237 (1999)

Three-state SCRAP: Theory



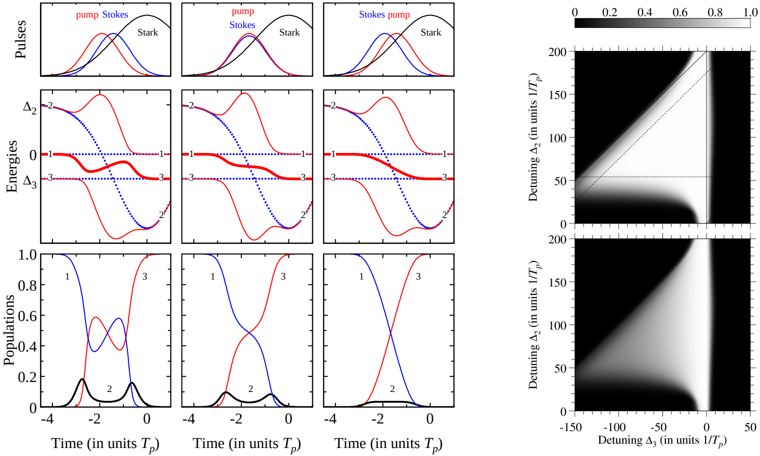
A.A.Rangelov, N.V.V., L.P.Yatsenko, B.W.Shore, T.Halfmann, K.Bergmann, Phys. Rev. A 72, 053403 (2005)

Three-state SCRAP: Optimization



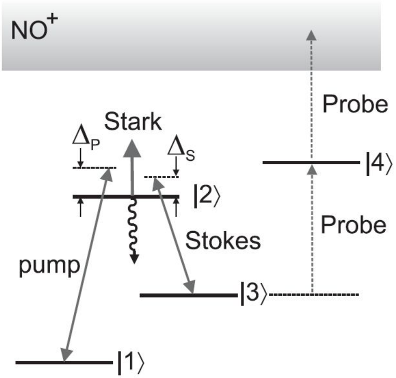
A.A.Rangelov, N.V.V., L.P.Yatsenko, B.W.Shore, T.Halfmann, K.Bergmann, Phys. Rev. A 72, 053403 (2005)

Three-state SCRAP: Robustness



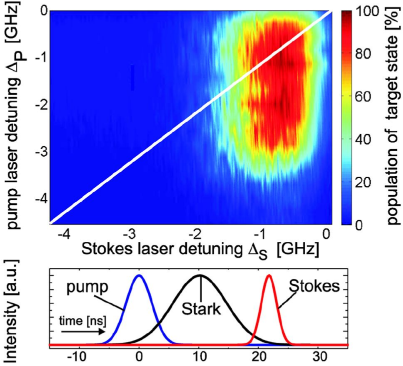
A.A.Rangelov, N.V.V., L.P.Yatsenko, B.W.Shore, T.Halfmann, K.Bergmann, Phys. Rev. A 72, 053403 (2005)

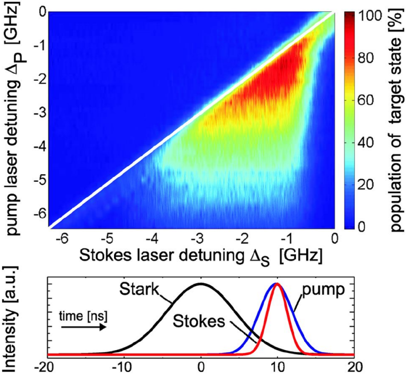
Three-state SCRAP: Experiment



M. Oberst, H. Muench, T. Halfmann, PRL 99, 173001 (2007)

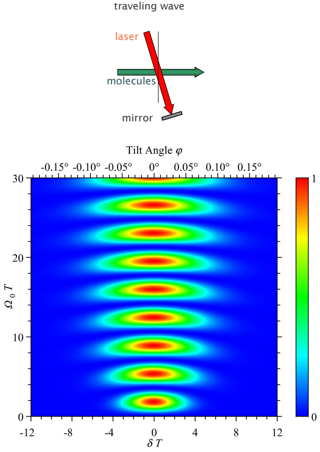
Three-state SCRAP: Experiment

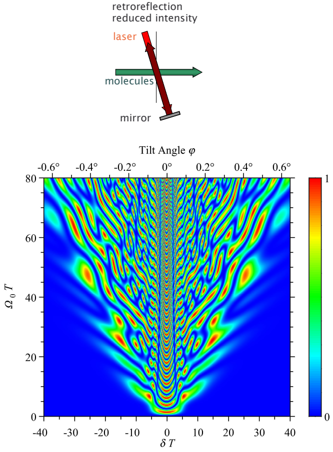




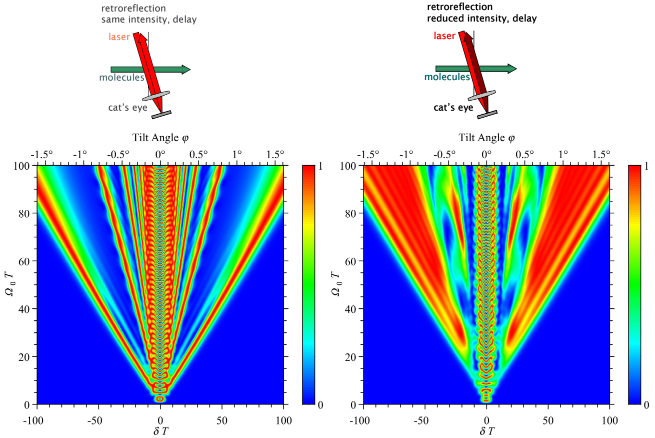
M. Oberst, H. Muench, T. Halfmann, PRL 99, 173001 (2007)

RIBAP: Retroreflection-induced bichromatic adiabatic passage





RIBAP: Retroreflection-induced bichromatic adiabatic passage



S. Guérin, L.P. Yatsenko, H.R. Jauslin, Phys. Rev. A 63, 031403 (2001)

L.P. Yatsenko, B.W. Shore, N.V.V., K. Bergmann, Phys. Rev. A 68, 043405 (2003)

RIBAP: Floquet picture

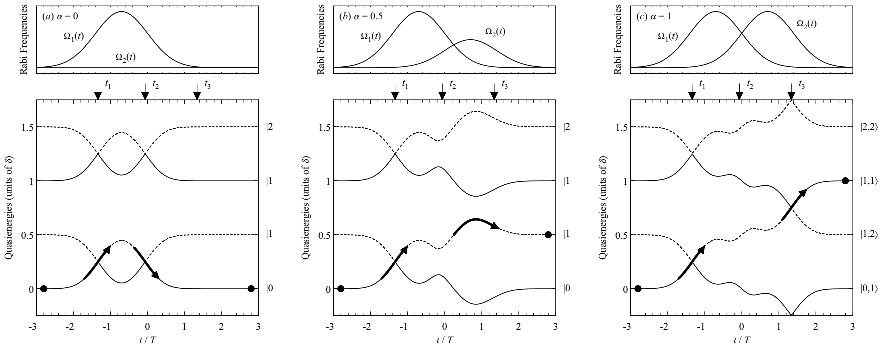


Figure: Two pairs of Floquet quasienergies for (a) $\alpha = 0$, (b) $\alpha = 0.5$, (c) $\alpha = 1$. Full curves: quasienergies for ψ_1; dashed curves: quasienergies for ψ_2.

S. Guérin, L.P. Yatsenko, H.R. Jauslin, Phys. Rev. A 63, 031403 (2001)
L.P. Yatsenko, B.W. Shore, N.V.V., K. Bergmann, Phys. Rev. A 68, 043405 (2003)

RIBAP: EXPERIMENT IN HELIUM

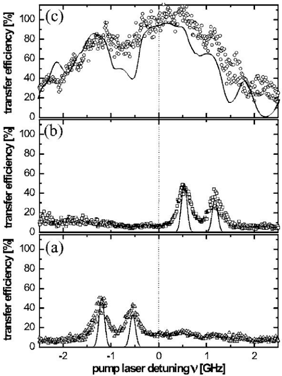
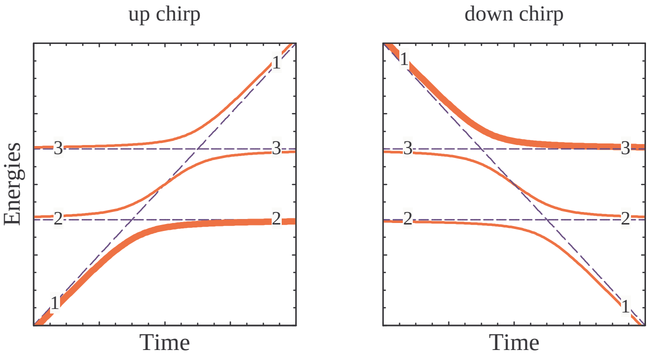


FIG. 4. Ion signal versus pump laser tuning for the atomic system, interacting with the strong pump laser pulse alone (a), the weak pump laser pulse alone (b), and simultaneously with both pump lasers pulses (c). Solid lines show numerical simulations.

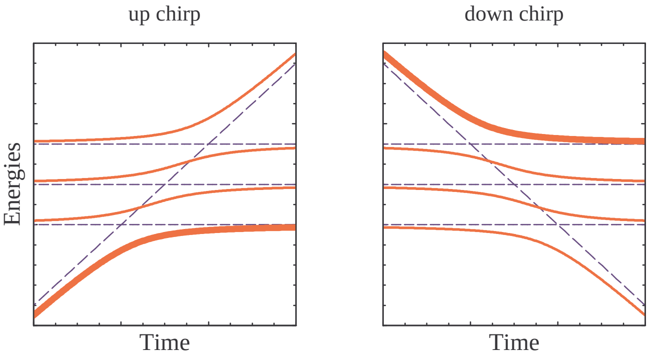
A.P. Conde, L.P. Yatsenko, J. Klein, M. Oberst, T. Halfmann, Phys. Rev. A 72, 053808 (2005)

Multiple Crossings

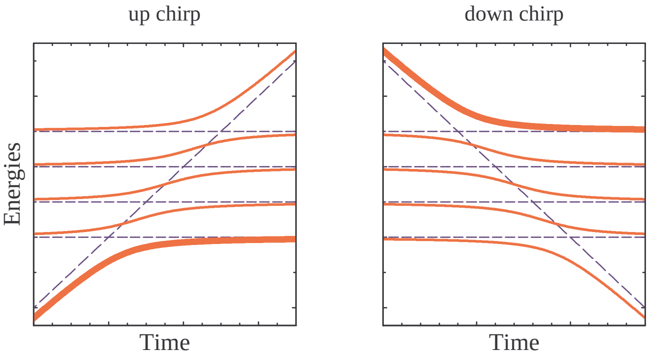
Selectivity of adiabatic passage



Selectivity of adiabatic passage



Selectivity of adiabatic passage



Selectivity of adiabatic passage

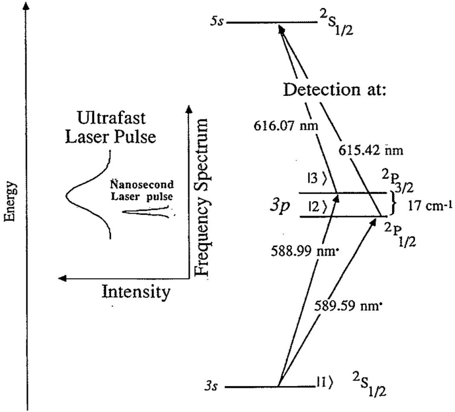


Figure: Sodium experiment of Melinger et al (1992).

J.S. Melinger, S.R. Gandhi, A. Hariharan, J.X. Tull, and W.S. Warren, Phys. Rev. Lett. 68, 2000 (1992).

Selectivity of adiabatic passage

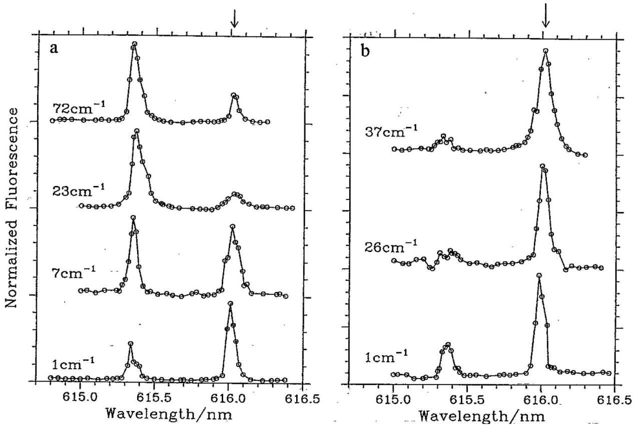
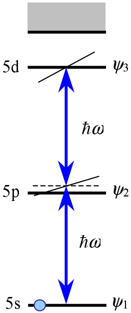


Figure: Sodium experiment of Melinger et al (1992).

J.S. Melinger, S.R. Gandhi, A. Hariharan, J.X. Tull, and W.S. Warren, Phys. Rev. Lett. 68, 2000 (1992).

Ladder climbing



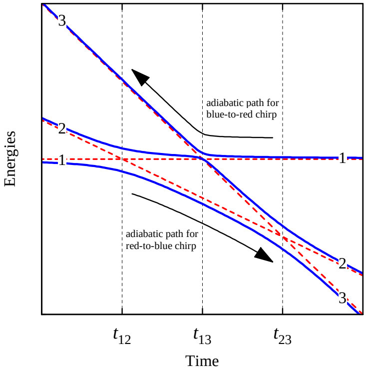


Figure: Three-state ladder climbing in rubidium.

Ladder climbing

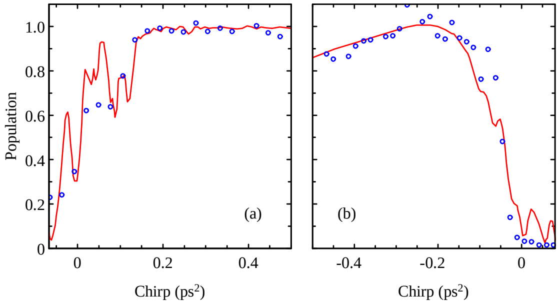


Figure: Three-state ladder climbing in rubidium: experiment.

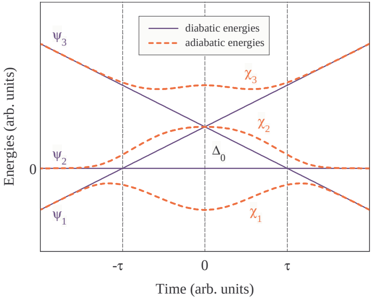
B. Broers, H. B. van Linden van den Heuvell, and L. D. Noordam, Phys. Rev. Lett. 69, 2062 (1992)

B. Broers B, L. D. Noordam, and H. B. van Linden van den Heuvell, Phys. Rev. A 46, 2749 (1992)

Level Crossing: Analytic Tools

- two crossing levels: Landau-Zener and Demkov-Kunike models
- three crossing levels: Carroll-Hioe model
- multiple crossings: Demkov-Osherov, Demkov-Ostrovsky, bowtie, degenerate Landau-Zener

Three crossing levels



$$\mathbf{H}(t) = \begin{bmatrix} \Delta_0 + At & \frac{1}{2}\Omega_{12}(t) & 0 \\ \frac{1}{2}\Omega_{12}(t) & 0 & \frac{1}{2}\Omega_{23}(t) \\ 0 & \frac{1}{2}\Omega_{23}(t) & \Delta_0 - At \end{bmatrix}$$

the propagator

$$\mathbf{U}(\infty,-\infty) = \mathbf{R}(\infty)\mathbf{U}^A(\infty,-\infty)\mathbf{R}^T(-\infty)$$

$$\mathbf{U}^A(\infty,-\infty) = \mathbf{M}(\infty,\tau)\mathbf{U}_{LZ}(\tau)\mathbf{M}(\tau,0)\mathbf{U}_{LZ}(0)\mathbf{M}(0,-\tau)\mathbf{U}_{LZ}(-\tau)\mathbf{M}(-\tau,-\infty)$$

Three crossing levels: LZ approach

The transition probability matrix

$$\mathbf{P} = \begin{bmatrix} pp_0 & qp_0 & q_0 \\ qp + pq_0q + 2qp\sqrt{q_0}\cos\gamma & p^2 + q^2q_0 - 2qp\sqrt{q_0}\cos\gamma & qp_0 \\ q^2 + q_0p^2 - 2qp\sqrt{q_0}\cos\gamma & qp + pq_0q + 2qp\sqrt{q_0}\cos\gamma & pp_0 \end{bmatrix}$$

$$p_\kappa = e^{-\pi a_\kappa^2}, \quad q_\kappa = 1 - p_\kappa \quad (\kappa = +, -, 0)$$

$$a_- = \Omega_{12}(-\tau)/(2A)^{1/2}, \quad a_0 = \Omega_{\text{eff}}(0)/2A^{1/2}, \quad a_+ = \Omega_{23}(\tau)/(2A)^{1/2}$$

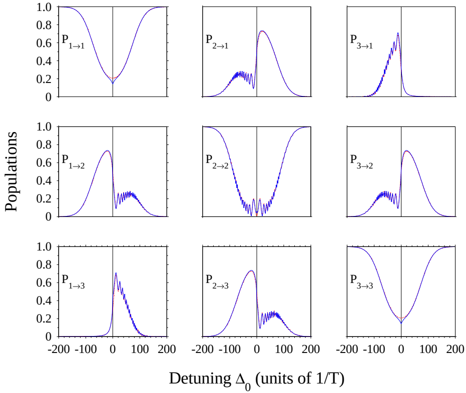
$$\Omega_{\text{eff}}(0) = \lambda_2(0) - \lambda_3(0) = -\tfrac{1}{2}\Delta_0 + \tfrac{1}{2}\sqrt{\Delta_0^2 + 2\Omega_0^2}$$

$$\phi_\kappa = \arg\Gamma(1 - ia_\kappa^2) + \frac{\pi}{4} + a_\kappa^2(\ln a_\kappa^2 - 1)$$

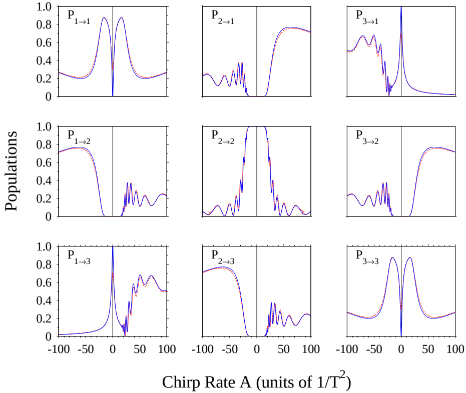
$$\gamma = 2\phi - \phi_0 + 2\varphi_1$$

S.S. Ivanov and N.V.V., Phys. Rev. A 77, 023406 (2008)

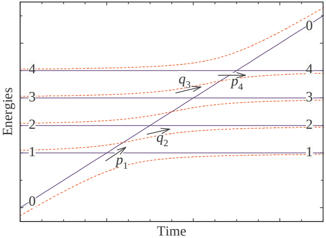
Three crossing states: asymmetry vs detuning



Three crossing states: asymmetry vs chirp

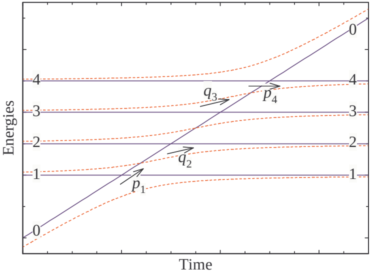


Demkov-Osherov model



$$\mathbf{H} = \begin{bmatrix} \beta t & \frac{1}{2}\Omega_1 & \frac{1}{2}\Omega_2 & \frac{1}{2}\Omega_3 & \cdots & \frac{1}{2}\Omega_N \\ \frac{1}{2}\Omega_1 & \Delta_1 & 0 & 0 & \cdots & 0 \\ \frac{1}{2}\Omega_2 & 0 & \Delta_2 & 0 & \cdots & 0 \\ \frac{1}{2}\Omega_3 & 0 & 0 & \Delta_3 & \cdots & 0 \\ \vdots & \vdots & \vdots & \vdots & \ddots & \vdots \\ \frac{1}{2}\Omega_N & 0 & 0 & 0 & \cdots & \Delta_N \end{bmatrix}$$

Demkov-Osherov model



Amazingly: the exact result coincides with the naive classical multiplication of probabilities!

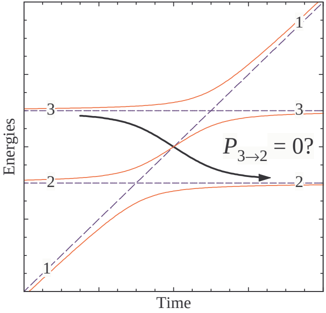
$p_n = \exp\left(-\pi\Omega_n/2\beta\right)$
no-transition probability

$q_n = 1 - p_n = 1 - \exp\left(-\pi\Omega_n/2\beta\right)$
transition probability

$\Psi(-\infty) = \|0\rangle$	$\Psi(-\infty) = \|1\rangle$	$\Psi(-\infty) = \|2\rangle$	$\cdots$
$P_{0\to1} = q_1$	$P_{1\to1} = p_1$	$P_{2\to1} = 0!$	$\cdots$
$P_{0\to2} = p_1 q_2$	$P_{1\to2} = q_1 q_2$	$P_{2\to2} = p_2$	$\cdots$
$P_{0\to3} = p_1 p_2 q_3$	$P_{1\to3} = q_1 p_2 q_3$	$P_{2\to3} = q_2 q_3$	$\cdots$
$P_{0\to4} = p_1 p_2 p_3 q_4$	$P_{1\to4} = q_1 p_2 p_3 q_4$	$P_{2\to4} = q_2 p_3 q_4$	$\cdots$
$P_{0\to0} = p_1 p_2 p_3 p_4$	$P_{1\to0} = q_1 p_2 p_3 p_4$	$P_{2\to0} = q_2 p_3 p_4$	$\cdots$

Demkov, Osherov. Sov. Phys. JETP 26, 916 (1968), Demkov, Ostrovsky. J. Phys. B 28, 403 (1995)

Counterintuitive transitions



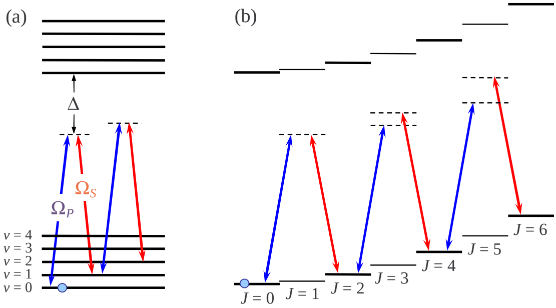
The zero probability for counterintuitive transitions applies only to this model!

Nonzero probability if

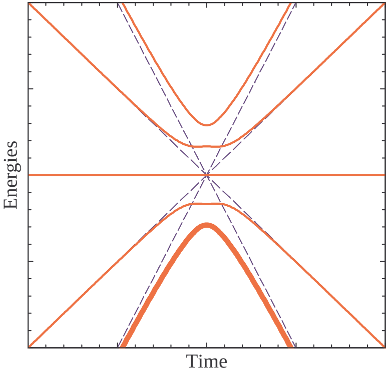
- finite duration
- pulsed interaction
- nonlinear energies

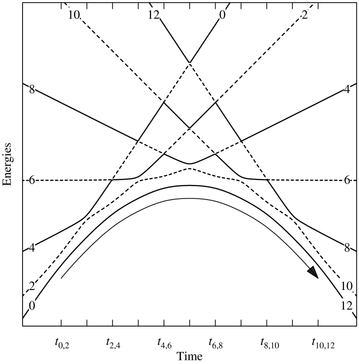
A.A.Rangelov, J.Piilo, N.V.V., Phys. Rev. A 72, 053404(9) (2005)

Vibrational and rotational ladder climbing



Bow-tie and "triangular" models





Bow-tie and "triangle" models: Applications

Bow-tie model

- molecules: vibrational ladder climbing
- entangled Dicke states of trapped ions

  I.E. Linington and N.V.V., Phys. Rev. A 77, 010302(R) (2008)

"Triangle" model

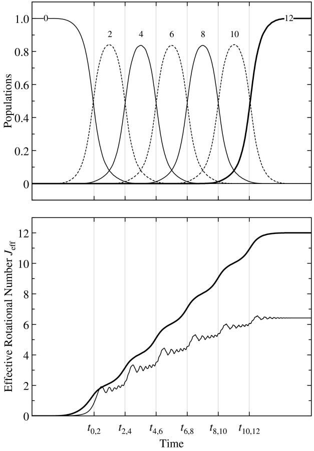
- molecules: rotational ladder climbing (optical centrifuge)

  Karczmarek, Wright, Corkum, Ivanov, Phys. Rev. Lett. 82, 3420 (1999); Villeneuve, Aseyev, Dietrich, Spanner, Ivanov, Corkum, Phys. Rev. Lett. 85, 542 (2000); N.V.V. and B. Girard, Phys. Rev. A 69, 033409(13) (2004)
- molecules: vibrational ladder climbing (anharmonic)

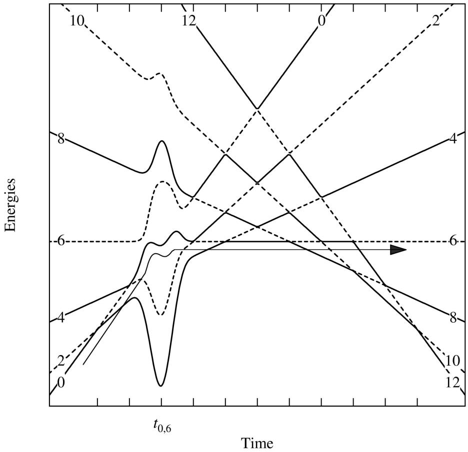
  Chelkowski, Bandrauk, Corkum, Melinger, Warren, Phys. Rev. Lett. 65, 2355 (1990); J. Chem. Phys. 99, 4279 (1993); 95, 2210 (1991); 101, 6439 (1994); Phys. Rev. A 52, R3417 (1995); J. Raman Spectrosc. 28, 459 (1997); J. Chem. Phys. 110, 4229 (1999)
- entangled Dicke states of coupled spins

  R.G. Unanyan, N.V.V., K. Bergmann, Phys. Rev. Lett. 87, 137902 (2001); Phys. Rev. A 66, 042101 (2002)

Optical centrifuge for molecules



Superrotors



Stimulated Raman Adiabatic Passage STIRAP

STIRAP

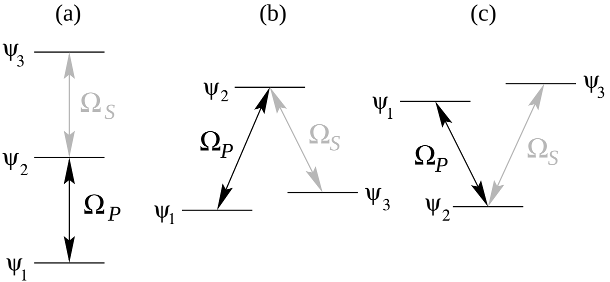
Stimulated Raman Adiabatic Passage

- Basic three-state STIRAP
- Multistate STIRAP
- Fractional STIRAP
- STIRAP via continuum

N.V.V., M. Fleischhauer, B.W. Shore, K. Bergmann, Adv. At. Mol. Opt. Phys. 46, 55-190 (2001)
Coherent manipulation of atoms and molecules by sequential pulses

N.V.V., T. Halfmann, B.W. Shore, K. Bergmann, Annu. Rev. Phys. Chem. 52, 763-809 (2001)
Laser-induced population transfer by adiabatic passage techniques

Three-state chains $\psi_1 \leftrightarrow \psi_2 \leftrightarrow \psi_3$



$$\mathbf{H}(t) = \hbar \begin{bmatrix} 0 & \frac{1}{2}\Omega_p(t) & 0 \\ \frac{1}{2}\Omega_p(t) & \Delta_2 & \frac{1}{2}\Omega_s(t) \\ 0 & \frac{1}{2}\Omega_s(t) & \Delta_3 \end{bmatrix}$$

pump Rabi freq $\Omega_p(t) = -\mathbf{d}_{12} \cdot \mathcal{E}_p(t)/\hbar$
Stokes Rabi freq $\Omega_s(t) = -\mathbf{d}_{23} \cdot \mathcal{E}_s(t)/\hbar$
transition dipole moments $\mathbf{d}_{mn}$
electric-field envelopes $\mathcal{E}_m(t)$

ladder: $\hbar\Delta_2 = E_2 - E_1 - \hbar\omega_1 \quad \hbar\Delta_3 = E_3 - E_1 - \hbar\omega_1 - \hbar\omega_2$
lambda: $\hbar\Delta_2 = E_2 - E_1 - \hbar\omega_1 \quad \hbar\Delta_3 = E_3 - E_1 - \hbar\omega_1 + \hbar\omega_2$

Three-state system: STIRAP

population transfer $\psi_1 \longrightarrow \psi_3$

- sequential π pulses (pump-Stokes): $\psi_1 \xrightarrow{\pi} \psi_2 \xrightarrow{\pi} \psi_3$
- simultaneous pump and Stokes (generalized π pulse): $\psi_1 \longrightarrow \psi_2 \longrightarrow \psi_3$
- SEP: stimulated-emission pumping (pump-Stokes): $\psi_1 \xrightarrow{50\%} \psi_2 \xrightarrow{50\%} \psi_3$
- STIRAP: stimulated Raman adiabatic passage (Stokes-pump): $\psi_1 \xrightarrow{100\%} \psi_3$

STIRAP requires

- two-photon resonance between ψ_1 and ψ_3
- counterintuitive pulse order Stokes-pump (sufficient overlap)
- adiabatic evolution (large pulse areas)

STIRAP delivers

- complete population transfer from ψ_1 to ψ_3
- no transient population in ψ_2

STIRAP: Theory

The Schrödinger equation: $i\frac{d}{dt}\mathbf{c}(t) = \mathbf{H}(t)\mathbf{c}(t) \qquad \mathbf{c}(t) = [c_1(t), c_2(t), c_3(t)]^T$

$$\begin{array}{lll} \text{initially:} & \Psi(-\infty) \equiv \psi_1 & \mathbf{c}(-\infty) = [1,0,0]^T \\ \text{objective:} & \Psi(+\infty) \equiv \psi_3 & \mathbf{c}(+\infty) = [0,0,1]^T \end{array}$$

RWA Hamiltonian for coherent excitation

$$\mathbf{H}(t) = \hbar \begin{bmatrix} 0 & \frac{1}{2}\Omega_p(t) & 0 \\ \frac{1}{2}\Omega_p(t)^* & \Delta_p & \frac{1}{2}\Omega_s(t) \\ 0 & \frac{1}{2}\Omega_s(t)^* & \Delta_p - \Delta_s \end{bmatrix}$$

detunings

$$\hbar\Delta_p = E_2 - E_1 - \hbar\omega_p$$
$$\hbar\Delta_s = E_2 - E_3 - \hbar\omega_s$$

Essential condition for STIRAP: two-photon resonance between ψ_1 and ψ_3

$$\Delta_p = \Delta_s \equiv \Delta$$

$$\mathbf{H}(t) = \hbar \begin{bmatrix} 0 & \frac{1}{2}\Omega_p(t) & 0 \\ \frac{1}{2}\Omega_p(t) & \Delta & \frac{1}{2}\Omega_s(t) \\ 0 & \frac{1}{2}\Omega_s(t) & 0 \end{bmatrix}$$

Adiabatic basis

adiabatic states: instantaneous eigenstates of $\mathbf{H}(t)$

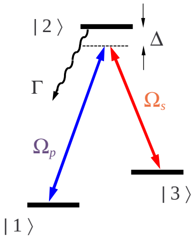
$$\begin{aligned}
\chi_+(t) &= \psi_1 \sin\vartheta(t) \sin\varphi(t) + \psi_2 \cos\varphi(t) + \psi_3 \cos\vartheta(t) \sin\varphi(t) \\
\chi_0(t) &= \psi_1 \cos\vartheta(t) - \psi_3 \sin\vartheta(t) \quad \text{no component from } \psi_2\text{!!!} \\
\chi_-(t) &= \psi_1 \sin\vartheta(t) \cos\varphi(t) - \psi_2 \sin\varphi(t) + \psi_3 \cos\vartheta(t) \cos\varphi(t)
\end{aligned}$$

adiabatic energies: the eigenvalues of $\mathbf{H}(t)$ $\quad \hbar\varepsilon_-,\ \hbar\varepsilon_0,\ \hbar\varepsilon_0$

$$\begin{aligned}
\varepsilon_+(t) &= \tfrac{1}{2}\Delta + \tfrac{1}{2}\sqrt{\Delta^2 + \Omega^2(t)} = \tfrac{1}{2}\Omega(t) \cot\varphi(t) \\
\varepsilon_0(t) &= 0 \\
\varepsilon_-(t) &= \tfrac{1}{2}\Delta - \tfrac{1}{2}\sqrt{\Delta^2 + \Omega^2(t)} = -\tfrac{1}{2}\Omega(t) \tan\varphi(t)
\end{aligned}$$

$$\tan\vartheta(t) = \frac{\Omega_p(t)}{\Omega_s(t)} \qquad \tan 2\varphi(t) = \frac{\Omega(t)}{\Delta} \qquad \Omega(t) = \sqrt{\Omega_p^2(t) + \Omega_s^2(t)}$$

STIRAP: MECHANISM



dark state $\chi_0(t) = \psi_1 \cos\vartheta(t) - \psi_3 \sin\vartheta(t)$

$$= \frac{\Omega_s(t)}{\Omega(t)}\psi_1 - \frac{\Omega_p(t)}{\Omega(t)}\psi_3$$

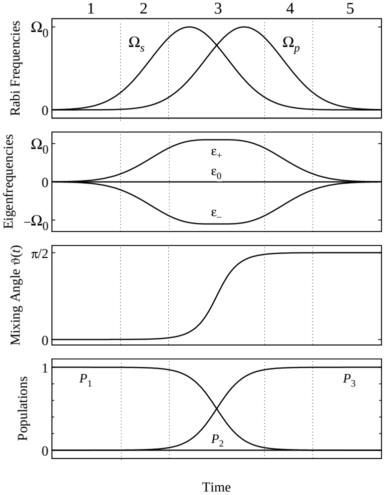
$$\tan\vartheta(t) = \frac{\Omega_p(t)}{\Omega_s(t)} \qquad \Omega(t) = \sqrt{\Omega_p^2(t) + \Omega_s^2(t)}$$

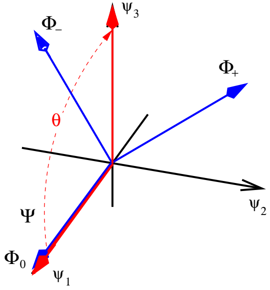
$$0 \xleftarrow{-\infty\leftarrow t} \frac{\Omega_p(t)}{\Omega_s(t)} \xrightarrow{t\to+\infty} \infty \quad \Longrightarrow \quad 0 \xleftarrow{-\infty\leftarrow t} \vartheta(t) \xrightarrow{t\to+\infty} \tfrac{1}{2}\pi$$

counterintuitive $(S - P)$ $\Longrightarrow$ $\psi_1 \xleftarrow{-\infty\leftarrow t} \chi_0(t) \xrightarrow{t\to+\infty} -\psi_3$

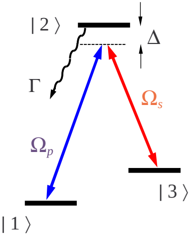
- two-photon resonance $\Longrightarrow$ $\mathbf{H}(t)$ possesses a dark state
- counterintuitive pulse sequence $\Longrightarrow$ $\Psi(-\infty) \equiv \chi_0(-\infty)$
- adiabatic evolution $\Longrightarrow$ the system stays in $\chi_0(t)$: $\Psi(t) = \chi_0(t) \xrightarrow{t\to+\infty} -\psi_3$

STIRAP: MECHANISM





STIRAP: DARK STATE



$$\mathbf{H}(t) = \hbar \begin{bmatrix} 0 & \frac{1}{2}\Omega_p(t) & 0 \\ \frac{1}{2}\Omega_p(t) & \Delta - i\Gamma/2 & \frac{1}{2}\Omega_s(t) \\ 0 & \frac{1}{2}\Omega_s(t) & 0 \end{bmatrix}$$

two-photon resonance between ψ_1 and ψ_3

dark (trapping, trapped) state

$$\chi_0(t) = \psi_1 \cos\vartheta(t) - \psi_3 \sin\vartheta(t)$$

no component from $\psi_2 \Longrightarrow$ the (lossy) state ψ_2 remains unpopulated in the adiabatic regime if the system stays in $\chi_0(t)$: $\Psi(t) = \chi_0(t)$!

$\Longrightarrow$ the properties of ψ_2 (detuning Δ, loss Γ) do not affect STIRAP!

Bergmann group: STIRAP with 100% efficiency even for $T \sim 100/\Gamma$!

STIRAP SIGNATURE

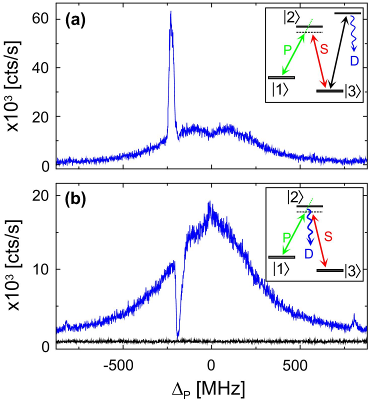


FIGURE: STIRAP signature in Ne*: Increase of signal from 3 and decrease of signal from 2.

Intuitive vs counterintuitive pulse sequence

RESONANCE $(\Delta = 0)$

- counterintuitive sequence: complete population transfer to ψ_3
- intuitive pulse sequence: generalized Rabi oscillations

$$\Delta = 0 \Longrightarrow \varphi \equiv \pi/4 \qquad \text{for intuitive ordering: } \vartheta(-\infty) = \pi/2,\ \vartheta(+\infty) = 0$$

$$\begin{aligned} \tfrac{1}{\sqrt{2}}(\psi_1 + \psi_2) \xleftarrow{-\infty} \quad \chi_+(t) \quad &\xrightarrow{+\infty} \tfrac{1}{\sqrt{2}}(\psi_2 + \psi_3), \\ -\psi_3 \xleftarrow{-\infty} \quad \chi_0(t) \quad &\xrightarrow{+\infty} \psi_1, \\ \tfrac{1}{\sqrt{2}}(\psi_1 - \psi_2) \xleftarrow{-\infty} \quad \chi_-(t) \quad &\xrightarrow{+\infty} \tfrac{1}{\sqrt{2}}(-\psi_2 + \psi_3). \end{aligned}$$

$\Longrightarrow$ initially both states $\chi_+(t)$ and $\chi_-(t)$ are populated
interference between two different paths from ψ_1 to ψ_3 $\Longrightarrow$ oscillations

$$P_1 = 0, \quad P_2 = \sin^2 \tfrac{1}{2}A, \quad P_3 = \cos^2 \tfrac{1}{2}A \qquad A = \int_{-\infty}^{+\infty} \Omega(t)\mathrm{d}t$$

N.V.V. & S. Stenholm, Phys. Rev. A 55, 648 (1997)

Intuitive vs counterintuitive pulse sequence

OFF RESONANCE ($\Delta \neq 0$)

- counterintuitive sequence: complete population transfer to ψ_3
- intuitive pulse sequence: complete population transfer to ψ_3

for the intuitive ordering: $\vartheta(-\infty) = \pi/2$, $\vartheta(+\infty) = 0$, $\varphi(-\infty) = \varphi(+\infty) = 0$

$$\psi_1 \xleftarrow{-\infty} \chi_-(t) \xrightarrow{+\infty} \psi_3$$

however the intermediate state receives a significant transient population:

$$P_2 = \sin^2 \varphi(t)$$

If the lifetime of ψ_2 is short compared to the excitation duration, then population transfer can be achieved only with the counterintuitive sequence. If the lifetime of ψ_2 is sufficiently long then complete population transfer can be achieved with either pulse orderings.

N.V.V. & S. Stenholm, Phys. Rev. A 55, 648 (1997)

Intuitive vs counterintuitive pulse sequence

FAR OFF RESONANCE ($|\Delta| \gg \Omega_{p,s}(t)$)

- counterintuitive sequence: complete population transfer to ψ_3
- intuitive pulse sequence: complete population transfer to ψ_3

$|\Delta| \gg \Omega_{p,s}(t) \Longrightarrow$ the intermediate state ψ_2 can be eliminated adiabatically
$\Longrightarrow$ effective two-state model

$$\Omega_{\text{eff}}(t) = -\frac{\Omega_p(t)\Omega_s(t)}{2\Delta} \qquad \Delta_{\text{eff}}(t) = \frac{\Omega_p^2(t) - \Omega_s^2(t)}{2\Delta}$$

delayed pulses: the detuning $\Delta_{\text{eff}}(t)$ crosses resonance when $\Omega_p(t_0) = \Omega_s(t_0)$
$\Longrightarrow$ complete population transfer for both pulse orderings in the adiabatic limit
(the ordering reversal leads to the unimportant change of sign in $\Delta_{\text{eff}}(t)$)

B.W. Shore, K. Bergmann, A. Kuhn, S. Schiemann, J. Oreg, J.H. Eberly, Phys. Rev. A 45, 5297 (1992)

N.V.V. & S. Stenholm, Phys. Rev. A 55, 648 (1997)

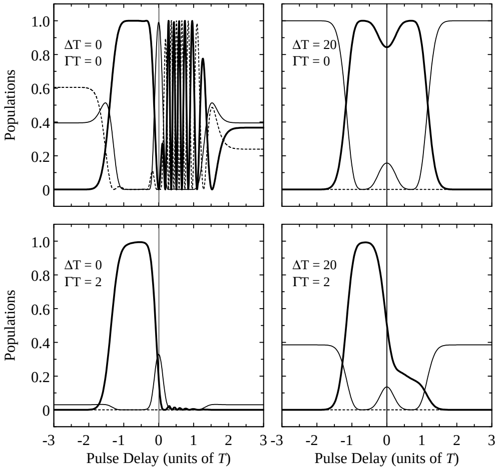


FIGURE: Populations of the initial state (thin solid line), the intermediate state (dashed line), and the final state (thick solid line) against the pulse delay for Gaussian pulse shapes: $\Omega_p(t) = \Omega_0 e^{-(t-\tau)^2/T^2}$, $\Omega_s(t) = \Omega_0 e^{-(t+\tau)^2/T^2}$, with $\Omega_0 T = 40$.

STIRAP: experiment in Ne*

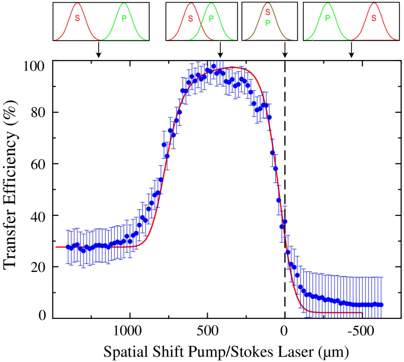


FIGURE: STIRAP experiment in Ne*: efficiency vs delay.

K. Bergmann, H. Theuer, B.W. Shore, Rev. Mod. Phys. 70, 1003 (1998)

STIRAP: experiment in NO

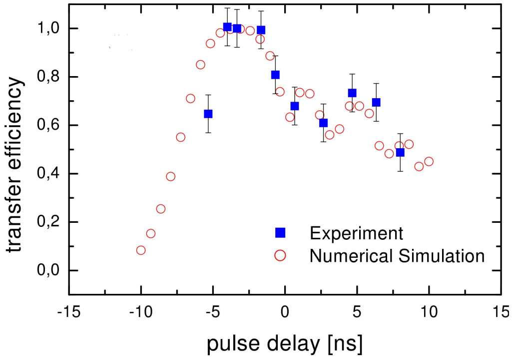


FIGURE: STIRAP experiment with pulsed lasers in NO: efficiency vs delay.

S. Schiemann, A. Kuhn, S. Steuerwald, K. Bergmann, Phys. Rev. Lett. 71, 3637 (1993)

STIRAP: EXPERIMENT IN SO_2

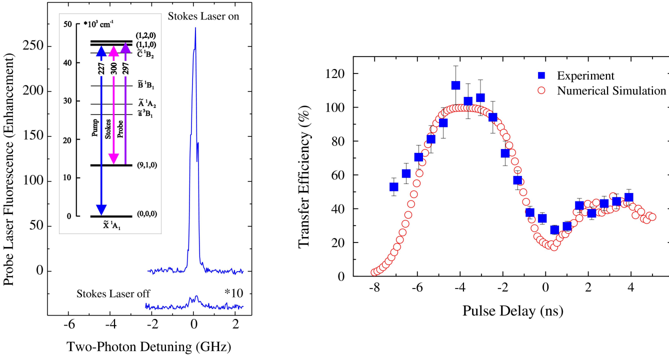


FIGURE: STIRAP experiment with pulsed lasers in SO_2: efficiency vs delay.

T. Halfmann and K. Bergmann, J. Chem. Phys. 104, 7068 (1996)

STIRAP: ADIABATIC BASIS & CONDITIONS

amplitudes of the adiabatic states $\mathbf{b}(t) = [b_+(t), b_0(t), b_-(t)]^T$

orthogonal transformation $\mathbf{c}(t) = \mathbf{R}(t)\mathbf{b}(t)$

rotation matrix $\mathbf{R}(t) = \begin{bmatrix} \sin\vartheta\sin\varphi & \cos\vartheta & \sin\vartheta\cos\varphi \\ \cos\varphi & 0 & -\sin\varphi \\ \cos\vartheta\sin\varphi & -\sin\vartheta & \cos\vartheta\cos\varphi \end{bmatrix}$

Schrödinger equation in the adiabatic basis $i\hbar\dfrac{d}{dt}\mathbf{b}(t) = \mathbf{H}_b(t)\mathbf{b}(t)$

Hamiltonian $\mathbf{H}_b = \mathbf{R}^{-1}\mathbf{H}\mathbf{R} - \mathbf{R}^{-1}\dot{\mathbf{R}} = \hbar\begin{bmatrix} \frac{1}{2}\Omega\cot\varphi & i\dot{\vartheta}\sin\varphi & i\dot{\varphi} \\ -i\dot{\vartheta}\sin\varphi & 0 & -i\dot{\vartheta}\cos\varphi \\ -i\dot{\varphi} & i\dot{\vartheta}\cos\varphi & -\frac{1}{2}\Omega\tan\varphi \end{bmatrix}$

adiabatic conditions $|\frac{1}{2}\Omega\cot\varphi| \gg |\dot{\vartheta}\sin\varphi|$

$|\frac{1}{2}\Omega\tan\varphi| \gg |\dot{\vartheta}\cos\varphi|$ *stronger!*

Adiabatic conditions: local and global

adiabatic evolution $\Longleftarrow$ negligible coupling between each pair of adiabatic states compared to their energy difference

$$\text{with respect to the dark state } \chi_0(t)\text{:} \quad |\langle\dot{\chi}_0|\chi_\pm\rangle| \ll |\varepsilon_0 - \varepsilon_\pm|$$

$$\left|\dot{\vartheta}\frac{\sin^2\varphi}{\cos\varphi}\right| \ll \tfrac{1}{2}\Omega, \qquad \left|\dot{\vartheta}\frac{\cos^2\varphi}{\sin\varphi}\right| \ll \tfrac{1}{2}\Omega \quad (\textit{stronger})$$

On one-photon resonance ($\Delta = 0$): $\varphi = \pi/4$ and hence (T is the pulse width)

$$\text{local}: \quad \Omega(t) \gg |\dot{\vartheta}(t)| \propto T^{-1} \qquad\qquad \Omega(t) = \sqrt{\Omega_p^2(t) + \Omega_s^2(t)}$$

Integration over t:

$$\text{global}: \quad A \propto \Omega_0 T \gg 1$$

$\Longrightarrow$ adiabaticity demands a large pulse area A

Adiabatic conditions: pulsed lasers

cw lasers have almost perfect coherence properties
pulsed lasers: the adiabatic conditions need to be modified

perfectly coherent pulsed lasers: the adiabatic condition is written as

energy $\propto \Omega_0^2 T > 100/T \Longrightarrow$ lower limit on pulse energy

$\Longrightarrow$ the needed laser energy grows rapidly when the pulse duration decreases

phase fluctuations $\Longrightarrow$ the actual bandwidth $\Delta\omega$ exceeds $\omega_{\mathrm{TL}} = 1/T$

modified adiabaticity condition

$$\Omega_0 T \gg \sqrt{1 + (\Delta\omega/\Delta\omega_{\mathrm{TL}})^2}$$

phase fluctuations $\Longrightarrow$ time-dependent changes in the laser frequencies
$\Longrightarrow$ two-photon detuning $\Longrightarrow$ reduced efficiency

A. Kuhn, G. Coulston, G.Z. He, S. Schiemann, K. Bergmann, W.S. Warren, J. Chem. Phys. 96, 4215 (1992)

Transition time in STIRAP

Define: T_{STIRAP} is the time during which $P_3(t)$ rises from ϵ to $1-\epsilon$ ($\epsilon \ll 1$)

For Gaussian pulses $\Omega_p(t) = \Omega_{p0} e^{-(t-\tau/2)^2/T^2}$ and $\Omega_s(t) = \Omega_{s0} e^{-(t+\tau/2)^2/T^2}$:

$$P_3(t) = \sin^2 \vartheta(t) = \frac{1}{1 + (\Omega_{s0}/\Omega_{p0})^2 \, e^{-4\tau t/T^2}}$$

$$\Longrightarrow T_{STIRAP} = t_{1-\epsilon} - t_{\epsilon} = \frac{T^2}{2\tau} \ln\left(\frac{1-\epsilon}{\epsilon}\right)$$

$$T_{STIRAP} = t_{0.9} - t_{0.1} = \frac{T^2}{\tau} \ln 3, \quad T_{STIRAP} = t_{0.99} - t_{0.01} \approx \frac{T^2}{\tau} \ln 10$$

naive expectation: $T_{STIRAP} \propto T_{\text{interaction}} \propto \tau + 2T$
difference between interaction time and transition time!

$T_{STIRAP} \propto 1/\tau \Longrightarrow$ the population transfer proceeds faster for larger delay!

P.A. Ivanov, N.V.V., K. Bergmann, Phys. Rev. A 70, 063409 (2004)

Sensitivity/Robustness of STIRAP to parameter fluctuations

N.V.V., M. Fleischhauer, B.W. Shore, K. Bergmann, Adv. At. Mol. Opt. Phys. 46, 55-190 (2001)

Sensitivity to single-photon detuning

The single-photon detuning Δ does not affect the dark state $\chi_0(t) = \psi_1 \cos\vartheta(t) - \psi_3 \sin\vartheta(t)$ because $\vartheta(t) = \arctan[\Omega_p(t)/\Omega_s(t)]$

However, Δ affects the adiabatic conditions:

$$\left|\dot{\vartheta}\frac{\sin^2\varphi}{\cos\varphi}\right| \ll \tfrac{1}{2}\Omega; \qquad \left|\dot{\vartheta}\frac{\cos^2\varphi}{\sin\varphi}\right| \ll \tfrac{1}{2}\Omega \quad (\text{stronger because } |\varphi(t)| \leqq \pi/4)$$

$$\tan 2\varphi(t) = \Omega(t)/\Delta \qquad \Omega(t) = \sqrt{\Omega_p^2(t) + \Omega_s^2(t)}$$

φ is a decreasing function of $\Delta \Longrightarrow$ the LHS increases with Δ
$\Longrightarrow$ adiabaticity deteriorates $\Longrightarrow$ transfer efficiency decreases

scaling properties of the FWHM $\Delta_{1/2}$ of the single-photon line profile $P_3(\Delta)$

$$\Delta_{1/2} = D(\tau)\Omega_0^2 \propto \text{peak laser intensity}$$

Sensitivity to detunings: Theory

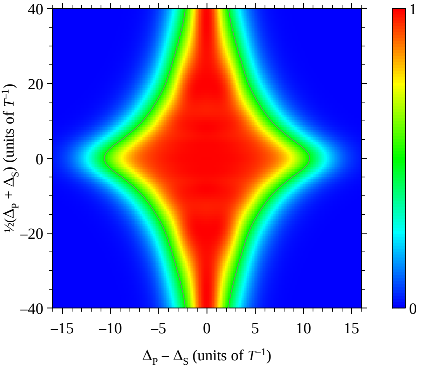


Figure: Transfer efficiency vs the single-photon and two-photon detunings for Gaussian pulses, $\Omega_p = \Omega_0 e^{-(t-\tau/2)^2/T^2}$, $\Omega_s = \Omega_0 e^{-(t+\tau/2)^2/T^2}$, with $\Omega_0 T = 20$, $\tau = 1T$.

Sensitivity to detunings: Experiment

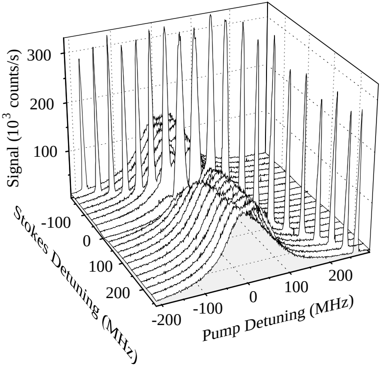


Figure: Experimentally measured transfer efficiency of STIRAP in neon.

J. Martin, B.W. Shore, K. Bergmann, Phys. Rev. A 54, 1556 (1996)

Sensitivity to two-photon detuning

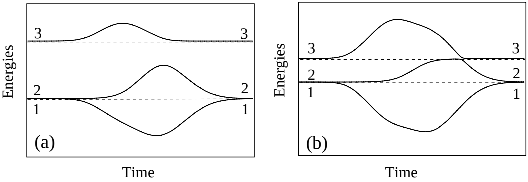
STIRAP is much more sensitive to the two-photon detuning $\delta = \Delta_p - \Delta_s$ because it prevents the exclusive population of the dark state.

Two approaches:

- analysis of transitions between the adiabatic states
- analysis of adiabatic conditions

Sensitivity to two-photon detuning

- the dark state $\chi_0(t)$ is no longer an eigenstate of $\mathbf{H}(t)$
- for nonzero δ each of the three eigenstates of $\mathbf{H}(t)$ connects to *the same* bare state at both $t = -\infty$ and $t = +\infty$
- no adiabatic connection from state ψ_1 to state ψ_3
- adiabatic evolution leads to complete population return to ψ_1
- the only mechanism, by which population transfer to state ψ_3 can occur, is by nonadiabatic transitions between the adiabatic states
- such transitions can take place for small values of δ when there are narrow avoided crossings between the adiabatic eigenvalues



Sensitivity to two-photon detuning

Alternative approach: uses the adiabatic condition in the basis of the eigenstates of $\mathbf{H}(t)$ for $\delta = 0$ In this basis the effect of nonzero two-photon detuning δ shows up in additional terms proportional to δ

$$\mathbf{H}_b(\delta) = \mathbf{H}_b(\delta = 0) + \tfrac{1}{2}\hbar\delta\cos^2\vartheta \begin{bmatrix} 1 & -\sqrt{2}\tan\vartheta & 1 \\ -\sqrt{2}\tan\vartheta & 2\tan^2\vartheta & -\sqrt{2}\tan\vartheta \\ 1 & -\sqrt{2}\tan\vartheta & 1 \end{bmatrix},$$

The effect of δ shows up as additional nonadiabatic couplings (which do not vanish in the adiabatic limit!) between the $\delta = 0$ adiabatic states.

Modified adiabatic condition: $\delta \sin\vartheta\cos\vartheta \ll \frac{1}{\sqrt{2}}\Omega$

The two-photon line profile $P_3(\delta)$ must scale as

$$\delta_{1/2} = d(\tau)\Omega_0 \propto \sqrt{\text{laser intensity}}$$

Sensitivity of STIRAP to decoherence

- irreversible population losses outside the system

$$\Gamma_{1/2} \approx G(\tau/T)(\Omega_0 T)^2, \qquad G(\tau/T) \approx \frac{3(\tau/T)\ln 2}{8(\tau/T)^2 + \pi/2} e^{-2(\tau/T)^2}$$

Fleischhauer & Manka, Phys. Rev. A 54, 794 (1996); N.V.V. & Stenholm, Phys. Rev. A 56, 1463 (1997)

- dephasing (Liouville equation)

$$P_3 = \frac{1}{3} + \frac{2}{3}\exp(-3\gamma_{13}T^2/4\tau) \qquad T_{STIRAP} \propto T^2/4\tau!$$

P.A. Ivanov, N.V.V., K. Bergmann, Phys. Rev. A 70, 063409 (2004)

- spontaneous emission within the system (Liouville equation)
  main loss mechanism: overdamping!

P.A. Ivanov, N.V.V., K. Bergmann, Phys. Rev. A 72, 053412 (2005)

Fractional STIRAP

Idea: As in STIRAP, the Stokes pulse arrives before the pump pulse, but unlike STIRAP, here the two pulses vanish simultaneously, while maintaining a constant finite ratio of amplitudes:

$$\lim_{t\to-\infty} \frac{\Omega_P(t)}{\Omega_S(t)} = 0 \qquad \lim_{t\to+\infty} \frac{\Omega_P(t)}{\Omega_S(t)} = \tan\alpha$$

$$\chi_0(t) = \frac{\Omega_S(t)}{\Omega(t)} e^{-i\phi_S}\psi_1 - \frac{\Omega_P(t)}{\Omega(t)} e^{i\phi_P}\psi_3 \qquad \left(\Omega(t) = \sqrt{\Omega_P^2(t) + \Omega_S^2(t)}\right)$$

$$\Longrightarrow \quad e^{-i\phi_S}\psi_1 \overset{-\infty\leftarrow t}{\longleftarrow} \chi_0(t) \overset{t+\infty}{\longrightarrow} e^{-i\phi_S}\left[\psi_1\cos\alpha - \psi_3 e^{i\phi}\sin\alpha\right] \qquad (\phi = \phi_P + \phi_S)$$

A coherent superposition of states ψ_1 and ψ_3 is created:

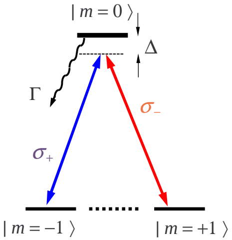
$$\Psi(+\infty) = \psi_1\cos\alpha - \psi_3 e^{i\phi}\sin\alpha$$

$$\alpha = \pi/4 \Longrightarrow \Psi(+\infty) = \tfrac{1}{\sqrt{2}}(\psi_1 - \psi_3)$$

Hadamard gate in quantum information; beam splitter in atom optics

P. Marte, P. Zoller, and J.L. Hall, Phys. Rev. A 44, R4118 (1991); M. Weitz, B.C. Young, S. Chu, Phys. Rev. Lett. 73, 2563 (1994); N.V.V., K.-A. Suominen, and B.W. Shore, J. Phys. B 32, 4535 (1999)

Fractional STIRAP

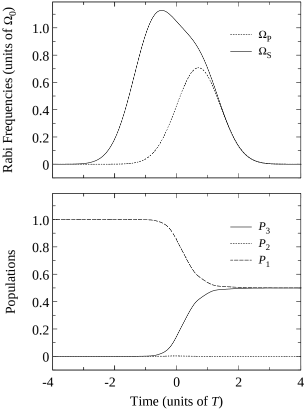


If σ_+ and σ_- propagate in opposite directions $\Longrightarrow$ momentum transfer $2\hbar k$

- full STIRAP: atomic mirror
- half-STIRAP: atomic beam splitter

M. Weitz, B.C. Young, S. Chu, Phys. Rev. Lett. 73, 2563 (1994)

Fractional STIRAP



$$\Omega_P(t) = \Omega_0 \sin\alpha\ \mathrm{e}^{-(t-\tau)^2/T^2}$$

$$\Omega_S(t) = \Omega_0 \mathrm{e}^{-(t+\tau)^2/T^2} + \Omega_0 \cos\alpha\ \mathrm{e}^{-(t-\tau)^2/T^2}$$

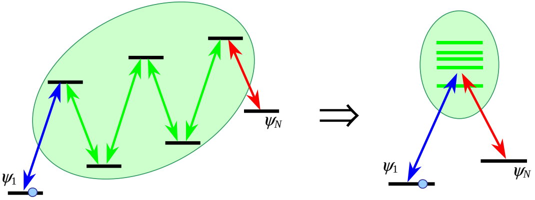
can be achieved by using only two pulses:
one with σ^- polarization
and Rabi frequency $\Omega_0 \mathrm{e}^{-(t+\tau)^2/T^2}$,
and another with Rabi freq $\Omega_0 \mathrm{e}^{-(t-\tau)^2/T^2}$
and elliptic polarization,
with angle of rotation of the ellipse $\phi/2$
and axial ratio $|1 - \tan\alpha|/(1 + \tan\alpha)$

beam splitter: $\sigma_+ \rightarrow \pi$

This technique can be extended to multistate systems (higher J)

N.V.V., K.-A. Suominen, and B.W. Shore, J. Phys. B 32, 4535 (1999)

STIRAP in multistate chains



$$\psi_1 \leftrightarrow \psi_2 \leftrightarrow \psi_3 \leftrightarrow \ldots \leftrightarrow \psi_N$$

$$\mathbf{H} = \frac{\hbar}{2} \begin{bmatrix} 0 & \Omega_{1,2} & 0 & \cdots & 0 & 0 \\ \Omega_{1,2} & 2\Delta_2 & \Omega_{2,3} & \cdots & 0 & 0 \\ 0 & \Omega_{2,3} & 2\Delta_3 & \cdots & 0 & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & 0 & \cdots & 2\Delta_{N-1} & \Omega_{N-1,N} \\ 0 & 0 & 0 & \cdots & \Omega_{N-1,N} & 0 \end{bmatrix}$$

$(N-1)$-photon resonance

such chains behave differently when they involve odd or even number of states

STIRAP in multistate chains: Resonance

Odd number of states $(N = 2n+1)$: STIRAP possible!

$$\mathbf{H} = \frac{\hbar}{2}\begin{bmatrix} 0 & \Omega_{1,2} & 0 & 0 & 0 \\ \Omega_{1,2} & 2\Delta & \Omega_{2,3} & 0 & 0 \\ 0 & \Omega_{2,3} & 0 & \Omega_{3,4} & 0 \\ 0 & 0 & \Omega_{3,4} & 2\Delta & \Omega_{4,5} \\ 0 & 0 & 0 & \Omega_{4,5} & 0 \end{bmatrix}$$

Hamiltonian for $N = 5$

a zero eigenvalue $\Longrightarrow$ multistate dark state

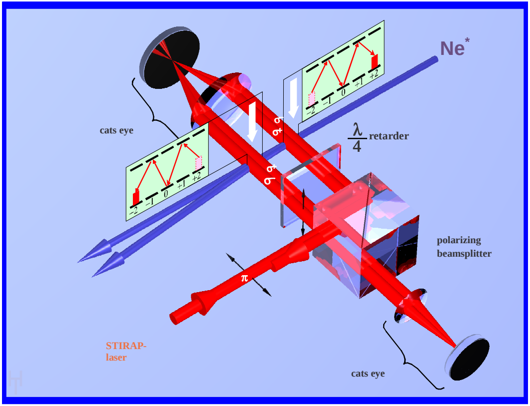
$$\chi_0(t) = \frac{1}{\mathcal{N}(t)}[\Omega_{2,3}(t)\Omega_{4,5}(t)\psi_1 - \Omega_{1,2}(t)\Omega_{4,5}(t)\psi_3 + \Omega_{1,2}(t)\Omega_{3,4}(t)\psi_5]$$

adiabatic connection between ψ_1 and ψ_N: $\psi_1 \overset{-\infty\leftarrow t}{\longleftarrow} \chi_0(t) \overset{t\to+\infty}{\longrightarrow} \psi_N$

$\chi_0(t)$ survives also when (only) the even states in the chain are detuned

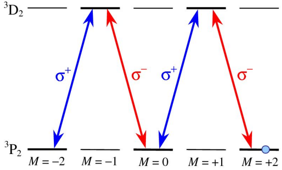
P. Marte, P. Zoller, and J.L. Hall, Phys. Rev. A 44, R4118 (1991)
B.W. Shore, K. Bergmann, J. Oreg, S. Rosenwaks, Phys. Rev. A 44, 7442 (1991)

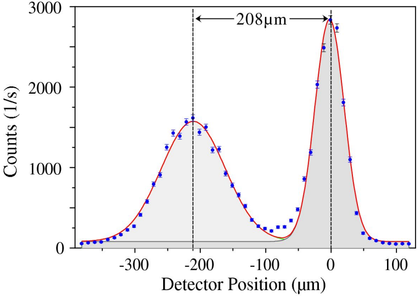
STIRAP in multistate chains: Atomic Mirror



K. Bergmann, H. Theuer, B.W. Shore, Rev. Mod. Phys. 70, 1003 (1998)

STIRAP in multistate chains: Atomic Mirror





$8\hbar k$ momentum transfer in double-STIRAP: $|m=-2\rangle \leftrightarrows |m=+2\rangle$
transient population of $|m=0\rangle$ not a problem

K. Bergmann, H. Theuer, B.W. Shore, Rev. Mod. Phys. 70, 1003 (1998)

STIRAP in multistate chains: Resonance

Even number of states: STIRAP impossible!
all intermediate-state detunings vanish: $\Delta_2 = \Delta_3 = \cdots = \Delta_{N-1} = 0$
$\Longrightarrow$ the Hamiltonian does not have a zero eigenvalue $\Longrightarrow$ no dark state
More importantly: $\mathbf{H}(t)$ does not possess even a more general adiabatic-transfer (AT) state that provides an adiabatic connection $\psi_1 \to \psi_N$:

$$\psi_1 \overset{-\infty\leftarrow t}{\longleftarrow} \chi_T(t) \overset{t+\infty}{\longrightarrow} \psi_N$$

$\chi_T(t)$ (AT state) may have nonzero contributions from all states
$\chi_0(t)$ (the dark state) is an AT state with contributions only from ground states

Example: resonantly driven $(\Omega_p, \Omega_i, \Omega_s)$ four-state chain $\to$ oscillations

$$P_1(\infty) \approx \cos^2\vartheta\cos^2\varphi$$
$$P_2(\infty) \approx 0$$
$$P_3(\infty) \approx \sin^2\vartheta\cos^2\varphi$$
$$P_4(\infty) \approx \sin^2\varphi$$

$$\tan\vartheta = \lim_{t\to+\infty}[\Omega_p(t)/\Omega_i(t)]$$
$$\varphi = \int_{-\infty}^{\infty}\sqrt{\tfrac{1}{2}\left(\Omega^2 - \sqrt{\Omega^4 - 4\Omega_p^2\Omega_s^2}\right)}\,dt$$
$$\Omega^2 = \Omega_p^2 + \Omega_i^2 + \Omega_s^2$$

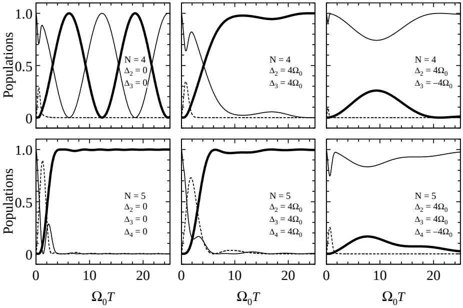
N.V.V., Phys. Rev. A 58, 2295 (1998)

STIRAP in multistate chains: Off resonance

generally nonzero intermediate-state detunings $\Delta_2, \Delta_3, \ldots, \Delta_{N-1}$

chains with even and odd N behave similarly: AT state may or may not exist!

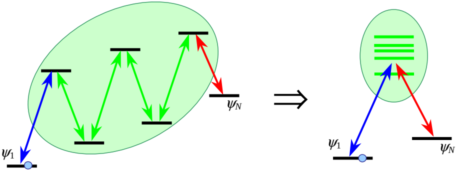
$$\psi_1 \xleftarrow{-\infty\leftarrow t} \chi_T(t) \xrightarrow{t+\infty} \psi_N$$



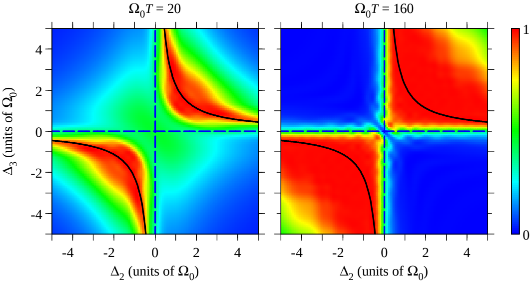
If all $\Delta_2, \Delta_3, \ldots, \Delta_{N-1} \neq 0$
the condition for AT state is
$\Delta_2 \Delta_{N-1} > 0$

N.V.V., Phys. Rev. A 58, 2295 (1998)

STIRAP in multistate chains: Optimization

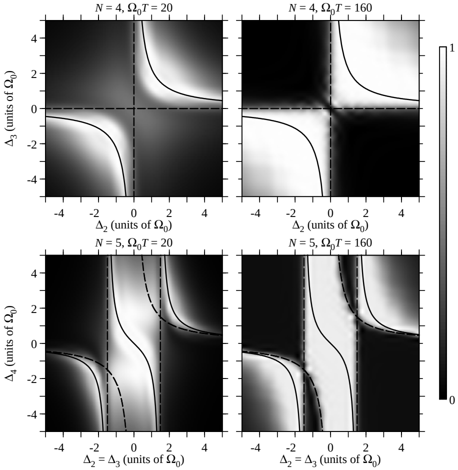


tune to a dressed resonance!



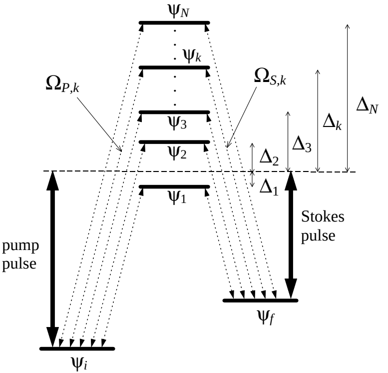
N.V.V., B.W. Shore, K. Bergmann, Eur. Phys. J. D 4, 15 (1998)

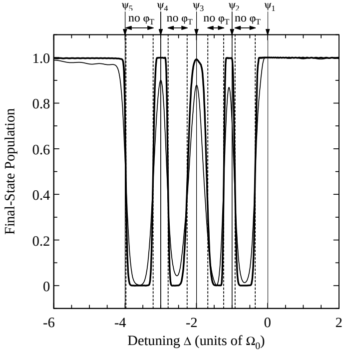
STIRAP in multistate chains: Optimization



N.V.V., B.W. Shore, K. Bergmann, Eur. Phys. J. D 4, 15 (1998)

STIRAP in parallel multi-Λ system





States	ψ_1	ψ_2	ψ_3	ψ_4	ψ_5
α_k	1.0000	0.2505	0.2786	0.7596	0.1505
β_k	1.0000	0.7878	1.3739	0.4621	1.5730

N.V.V. and S. Stenholm, Phys. Rev. A 60, 3820 (1999)

Variations of STIRAP

- STIRAP in a tripod system (3 ground + 1 excited): two dark states $\Longrightarrow$ creation of superpositions (Unanyan, Theuer, Bergmann)
- STIRAP between degenerate levels (Shore, Bergmann, Shapiro, Kis)
- cavity-STIRAP (pump-vacuum or vacuum-Stokes): single photon pistol (Kuhn, Rempe, Guérin)
- STIRAP via continuum (1 - continuum - 3) (Yatsenko, Halfmann, Knight)
- STIRAP into continuum (Rangelov)
- STIRAP in multiparticle systems: creation of entangled states (Unanyan: spins, Linington: ions)
- STIRAP in classical systems (Yatsenko: pendula, Rangelov: ED, Coriolis)
- ....

MERCI BEAUCOUP!
THANK YOU!