UNITED NATIONS EDUCATIONAL, SCIENTIFIC AND CULTURAL ORGANIZATION
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INTERNATIONAL CENTRE FOR THEORETICAL PHYSICS
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Winter College on Quantum Optics: Novel Radiation Sources

3-21 March 1997

Manupulation of atomic and molecular level population by coherent radiation fields

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Manipulation of atomic and molecular level population by coherent radiation fields

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estonishing properties, such as:

- population transfer between quantum states:
  by design
  (nearly) 100% efficiency
- elimination of spontaneous emission
- new type of spectroscopy
- new type of collision dynamics experiment
- new type of atom-optic elements
- changing optical properties of matter:
  making opaque media transparent
  changing non-linear optical properties
- designing photon fields in high-Q cavities
- many others

Lecture 1:

mainly: two-level systems

Lecture 2:

mainly: three-level systems
(coherent population transfer by **St**imulated **R**aman Scattering with **A**diabatic **P**assage -- STIRAP)

Lecture 3:

mainly: multi-level systems and applications to atom-optics

STIRAP-Papers from the Bergmann-Group (2/97)

1. U. Gaubatz, P. Rudecki, M. Becker, S. Schiemann, M. Külz and K. Bergmann
„Population Switching Between Vibrational Levels in Molecular Beams"
Chem Phys. Lett. 149, 463-468 (1988)

2. J.K. Kuklinski, U. Gaubatz, F.T. Hioe, and K. Bergmann
„Adiabatic Population Transfer in a Three Level System Driven by Delayed Laser Pulses"
Phys. Rev. A 40, 6741-6744 (1989)

3. U. Gaubatz, P. Rudecki, S. Schiemann, and K. Bergmann
„Population Transfer Between Molecular Vibrational Levels by Stimulated Raman Scattering with Partially Overlapping Laser: A New Concept and Experimental Results"
J. Chem. Phys. 92, 5363-5376 (1990)

4. G.Z. He, A. Kuhn, S. Schiemann, and K. Bergmann
„Population Transfer by Stimulated Raman Scattering with Delayed Pulses and by the SEP Method: A Comparative Study"
J. Opt. Soc. Am. B7, 1960-1969 (1990)

5. H.-G. Rubahn, E. Konz, S. Schiemann, and K. Bergmann
„Alignment of Electronic Angular Momentum by Stimulated Raman Scattering with Delayed Pulses"
Z. Phys. D 22, 401-406 (1991)

6. B.W. Shore, K. Bergmann, J. Oreg, and S. Rosenwaks
„Multilevel Adiabatic Population Transfer"
Phys. Rev. A 44, 7442-7447 (1991)

7. G. Coulston and K. Bergmann
„Population Transfer by Stimulated Raman Scattering with Delayed Pulses: Analytical Results for Multilevel Systems"
J. Chem. Phys. 96, 3467-3475 (1992)

8. A. Kuhn. S. Schiemann, G.Z. He, G. Coulston, W.S. Warren, and K. Bergmann
„Population Transfer by Stimulated Raman Scattering with Delayed Pulses using Spectrally Broad Light"
J. Chem. Phys. 96, 4215-4223 (1992)

9. J. Oreg, K. Bergmann, B.W. Shore, and S. Rosenwaks
„Population Transfer with Delayed Pulses in Four-State Systems"
Phys. Rev. A 45, 4888-4896 (1992)

10. B.W. Shore, K. Bergmann, A. Kuhn, S. Schiemann, J. Oreg, and J.H. Eberly
„Laser-induced Population Transfer in Multistate Systems: A comparative Study"
Phys. Rev. A 45, 5297-5300 (1992)

11. B.W. Shore, K. Bergmann, and J. Oreg
„Coherent Population Transfer: Stimulated Raman Adiabatic Passage and the Landau-Zener Picture"
Z. Phys. D 23, 33-39 (1992)

12. S. Schiemann, A. Kuhn, St. Steuerwald, and K. Bergmann
„Coherent Population Transfer in NO with Pulsed Lasers"
Phys. Rev. Lett. 71, 3637-3640 (1993)

13. B.W. Shore, J. Martin, M.P. Fewell, and K. Bergmann
„Coherent Population Transfer in Multilevel Systems with Magnetic Sublevels I: Numerical Studies"
Phys. Rev. A 52, 566-582 (1995)

14. J. Martin, B.W. Shore, and K. Bergmann
„Coherent Population Transfer in Multilevel Systems with Magnetic Sublevels II: Algebraic Analysis"
Phys. Rev. A 52, 583-593 (1995)

15. K. Bergmann and B.W. Shore
„Coherent Population Transfer" in:
„Molecular Dynamics and Spectroscopy by Stimulated Emission Pumping", eds. H.L. Dai and R.W. Field in: Advances in Phys. Chemistry, (World Scientific), page 315-373 (1995)

16. T. Halfmann and K. Bergmann
„Coherent Population Transfer and Dark Resonances in SO_2 Molecules"
J. Chem. Phys. 104, 7068-7073 (1996)

17. J. Martin, B.W. Shore, and K. Bergmann
„Coherent Population Transfer in Multilevel Systems with Magnetic Sublevels III: Experimental Results"
Phys. Rev. A 54, 1556-1569 (1996)

18. P. Fewell, B.W. Shore, and K. Bergmann
„Coherent Population Transfer among Three States: Full Algebraic Solutions and the Relevance of Diabatic Processes"
Austr. J. Phys., (in press)

19. L. Yatsenko. R. Unanyan, K. Bergmann, T. Halfmann, and B.W. Shore
„Population Transfer through the Continuum using Laser-controlled Stark Shifts"
Opt. Commun. 135, 406-412 (1997)

20. R. Unanyan, L. Yatsenko, K. Bergmann, and B.W. Shore
„Population in Versions using Counterintuitively Ordered Laser and Quasistatic Magnetic Field Pulses
Opt. Commun., (submitted 08/96)

21. R. Unanyan, L. Yatsenko, K. Bergmann, and B.W. Shore
„Laser-induced Atomic Reorientation with Control of Diabatic Losses"
Opt. Commun., in press

22. A. Kuhn and K. Bergmann
„*Crossed Population Transfer in NO with Pulsed Lasers: and the Consequences of Doppler Broadening*"
in preparation

23. H. Theuer and K. Bergmann
„*Efficient Atomic Mirror based on Coherent Population Transfer with Adiabatic Passage*"
in preparation

Applications of STIRAP to collision dynamics

1. P. Dittmann, F.P. Pesl, J. Martin, G. Coulston, G.Z. He, and K. Bergmann
„*The Effect of Vibrational Excitation ($3 \leq v'' \leq 19$) on the Chemiluminescent Channels of the Reaction $Na_2(v'')+Cl \rightarrow NaCl + Na^*(3p)$*"
J. Chem. Phys. 97, 9472-9475 (1992)

2. M. Külz, A. Kortyna, M. Keil, B. Schellhaaß, J. Hauck, K. Bergmann, D. Weyh, and W. Meyer
„*Dissociative Attachment of Low Energy Electrons to State Selected Diatomic Molecules*"
Phys. Rev. A 53, 3324-3334 (1996)

(1) Two-level systems

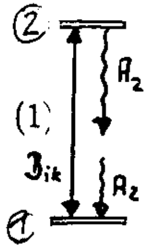
(a) Rate equations - Schrödinger equation
Evolution of population
Rabi oscillations
Damping of Rabi oscillations

(b) Dressed states, dressed state eigenvalues
dynamic Stark splitting

(c) Adiabatic passage in a two-level system
frequency chirping

(d) Coherent population return

Rate equation solution for a two-level system



$$\frac{d}{dt}P_1(t) = -B_{12}I(t)P_1(t) + B_{21}I(t)P_2(t) + A_{21}P_2(t)\,. \qquad (1)$$

$$\frac{d}{dt}P_2(t) = +B_{12}I(t)P_1(t) - B_{21}I(t)P_2(t) - A_2P_2(t)\,. \qquad (2)$$

$$\frac{d}{dt}[P_1(t) + P_2(t)] = -(A_2 - A_{21})P_2(t)\,. \qquad (3)$$

$$(t) = \frac{BI}{2\sqrt{(BI)^2 + BIA_{21} + \left(\frac{1}{2}A_2\right)^2}}[\exp(R_+t) - \exp(R_-t)]\,, \qquad (4)$$

(for $B_{12} = B_{21}$)

$$R_\pm = -\left(BI + \frac{1}{2}A_2\right) \pm \sqrt{(BI)^2 + BIA_{21} + \left(\frac{1}{2}A_2\right)^2}\,. \qquad (5)$$

$$P_2(t) = \frac{BI}{(2BI + A_2)}[1 - \exp(-(2BI + A_2)t)]\,. \qquad (6)$$

(for $A_{12} = A_2$)

$P_2(t) < 0.5$

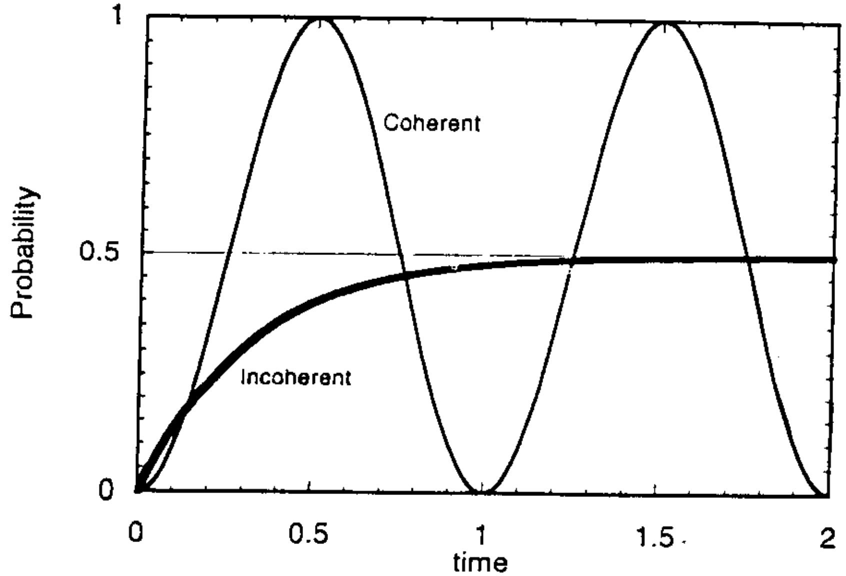


Figure 1. Time dependence of the probability of finding a two-state atom in the excited state, under conditions governed by rate equations (the monotonically rising incoherent curve) and under conditions governed by the time-dependent Schrödinger equation (the sinusoidal coherent curve).

Two-Level System: Coherent Excitation

Schrödinger Equation:

$$\hbar \frac{\partial}{\partial t} |\Psi(t)\rangle = -i\, H(t)\, |\Psi(t)\rangle$$

$$H(t) = H_o + V(t) + ((\, H_{el.mag}))$$

$$V(t) = -\vec{\mathbf{d}} \cdot \vec{\mathbf{E}}(t) \qquad \vec{\mathbf{E}}(t) = \hat{\mathbf{e}}\mathcal{E}(t)\cos(\omega t + \varphi)$$

$$\hbar\Omega(t) = -\langle 2|\mathbf{d}\cdot\mathbf{e}|1\rangle \mathcal{E}(t)\exp(-i\varphi)$$ Rabi frequency Ω

$$A(t) = \int_0^t \Omega(t')\, dt'$$ $A\,(t = \infty) \mathrel{\widehat{=}}$ Pulse Area

State vector $|\Psi\rangle$ is linear combination of the two basis states $|1\rangle$ and $|2\rangle$

$$|\Psi(t)\rangle = |1\rangle C_1(t)\exp[-i\zeta_1(t)] + |2\rangle C_2(t)\exp[-i\zeta_2($$

written in vector form $|\Psi(t)\rangle = \begin{pmatrix} C_1(t) \\ C_2(t) \end{pmatrix}$

matrix elements of H

$$H_{11} = E_1 \qquad H_{21} = \hbar\Omega(t)e^{i\varphi}\cos(\omega t + \varphi)$$

$$H_{22} = E_2 \qquad H_{12} = H_{21}^* \,.$$

choice: $\zeta_1 - \zeta_2 = \omega t$ $\quad h\,\zeta_1 = E_1 t$ $\quad E_1 = 0$

$$\hbar\Delta(t) = E_2 - (E_1 + \hbar\omega)$$

$$|\Psi(t)\rangle = |1\rangle C_1(t) + |2\rangle C_2(t)\exp[-i\omega t]$$

$$\frac{d}{dt}\begin{bmatrix} C_1(t) \\ C_2(t) \end{bmatrix} = -\frac{i}{2}\begin{bmatrix} 0 & \Omega(t)^* f(t)^* \\ \Omega(t) f(t) & 2\Delta(t) \end{bmatrix}\begin{bmatrix} C_1(t) \\ C_2(t) \end{bmatrix}$$

$$f(t) = 1 + \exp[2i\omega t + 2i\varphi]$$

$f(t) \approx 1$ $\hat{=}$ rotating wave approximation (RWA)
(neglect of rapidly oscillating terms

$$\frac{d}{dt}\begin{bmatrix} C_1(t) \\ C_2(t) \end{bmatrix} = -\frac{i}{2}\begin{bmatrix} 0 & \Omega(t)^* \\ \Omega(t) & 2\Delta(t) \end{bmatrix}\begin{bmatrix} C_1(t) \\ C_2(t) \end{bmatrix}$$

$$\frac{d}{dt}\mathbf{C}(t) = -iW(t)\mathbf{C}(t)$$

for resonant pulses, $\Delta = 0$

with $dA = \Omega(t)\, dt$

$$\frac{d}{dA}\begin{bmatrix} C_1(t) \\ C_2(t) \end{bmatrix} = -\frac{i}{2}\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}\begin{bmatrix} C_1(t) \\ C_2(t) \end{bmatrix}$$

the solution is

$$P_2(t) = \frac{1}{2}[1 - \cos(A)]\,.$$

with $C_2^2(t) = P_2(t)$, independent of shape A(t).

For $A = \pi, 3\pi, \ldots. (2n+1)\pi$:

all the population is in state 2.

If $\Omega(t) = \Omega_0$, then pulse area $A(t) = \Omega_0 t$ and

$P_2(t) = 0.5\ (1 - \cos \Omega_0 t)$

observed: regular „Rabi ocillations“

Diagonalization of Hamiltonian yields

„dressed“ eigenstates and eigenvalues

$$|\Phi_-\rangle = \cos\Theta\,|1\rangle - \exp(-i\varphi)\sin\Theta\,|2\rangle$$

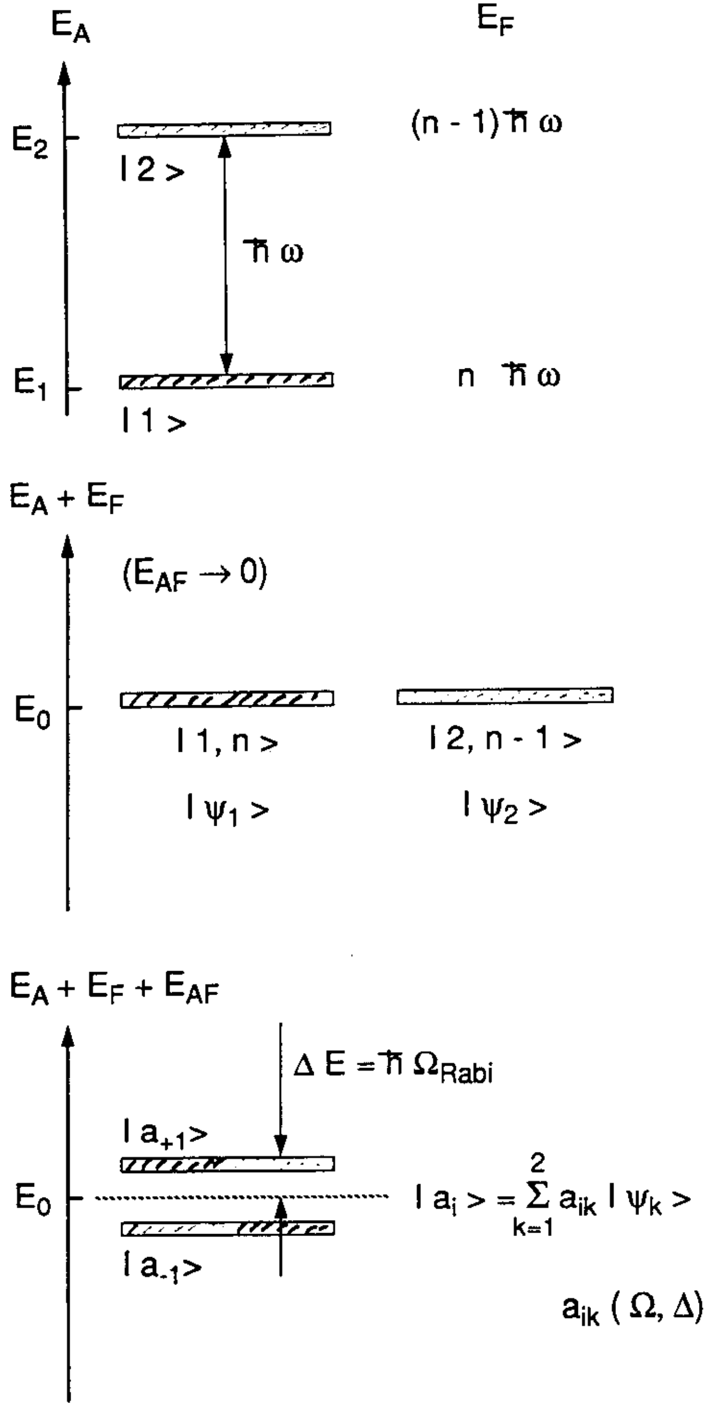
$$|\Phi_+\rangle = \exp(i\varphi)\sin\Theta\,|1\rangle + \cos\Theta\,|2\rangle$$

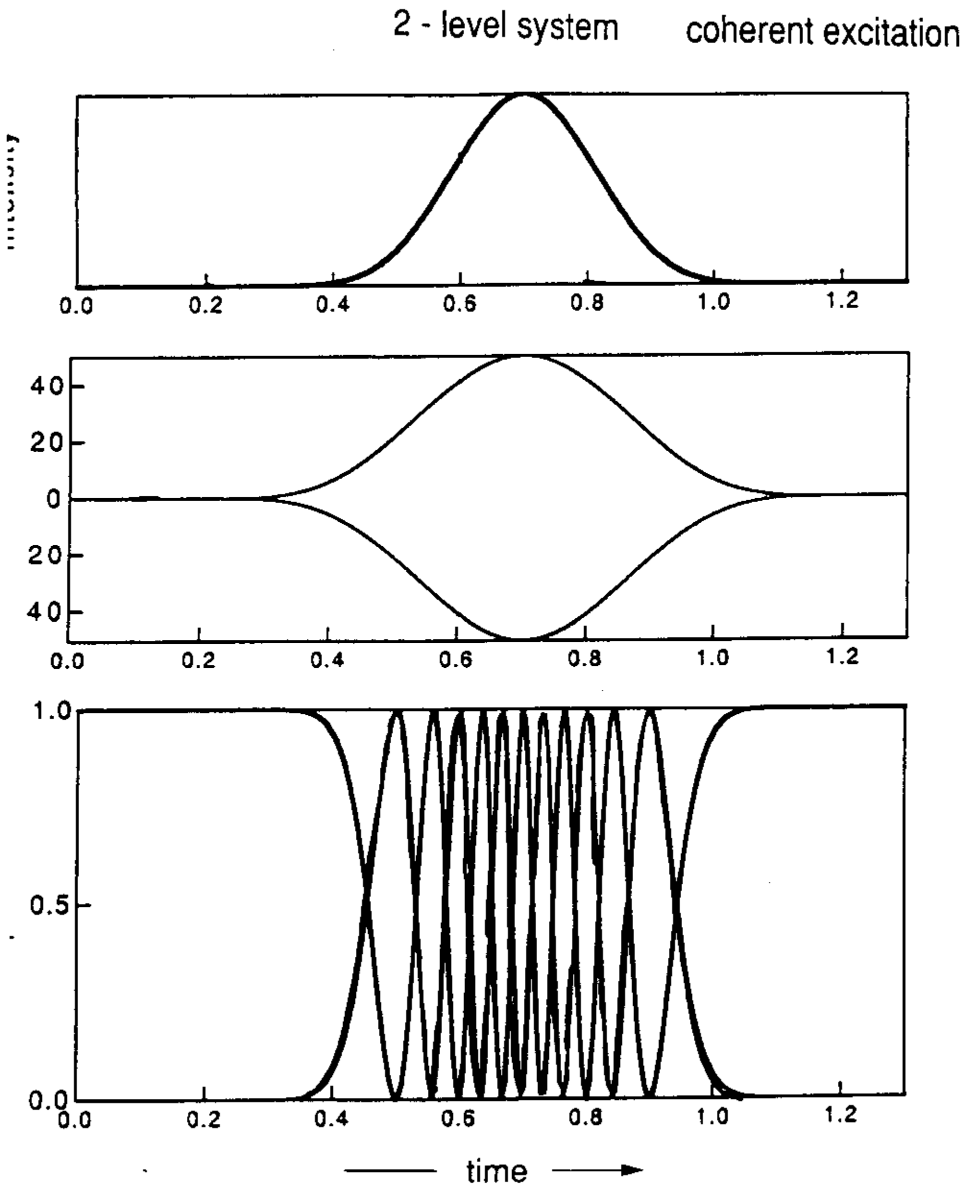
$$\langle\Phi_-|\Phi_+\rangle = 0\,, \quad \langle\Phi_-|\Phi_-\rangle = \langle\Phi_+|\Phi_+\rangle = 1$$

$$\omega_\pm = \frac{1}{2}\left[\Delta \pm \sqrt{|\Omega|^2 + \Delta^2}\right]$$

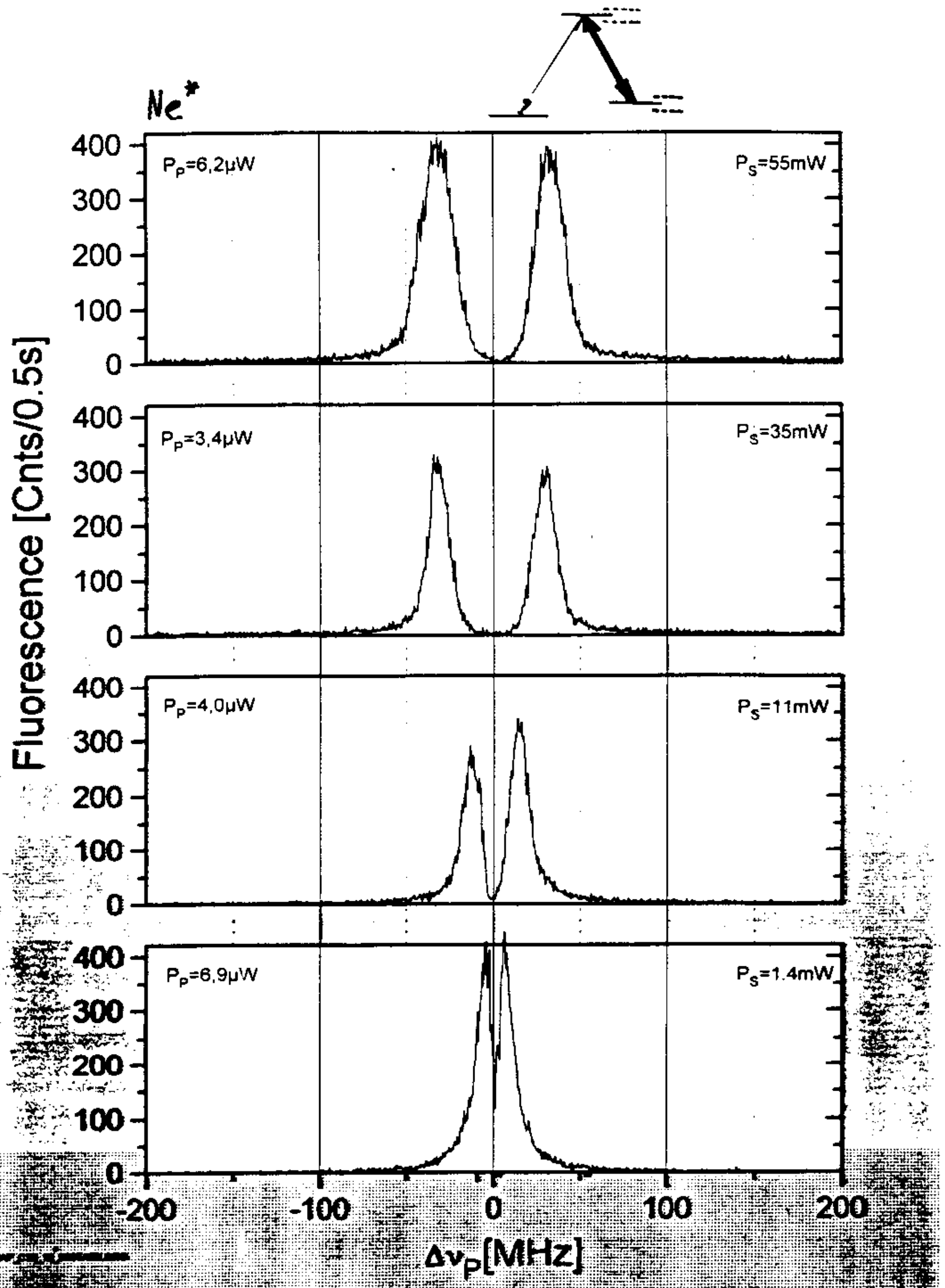
$$\cos(2\Theta) = \frac{\Delta}{\sqrt{\Delta^2 + |\Omega|^2}}\,, \quad \sin(2\Theta) = \frac{|\Omega|}{\sqrt{\Delta^2 + |\Omega|^2}}$$

character of dressed eigenstates changes with detuning Δ and Rabi frequency Ω

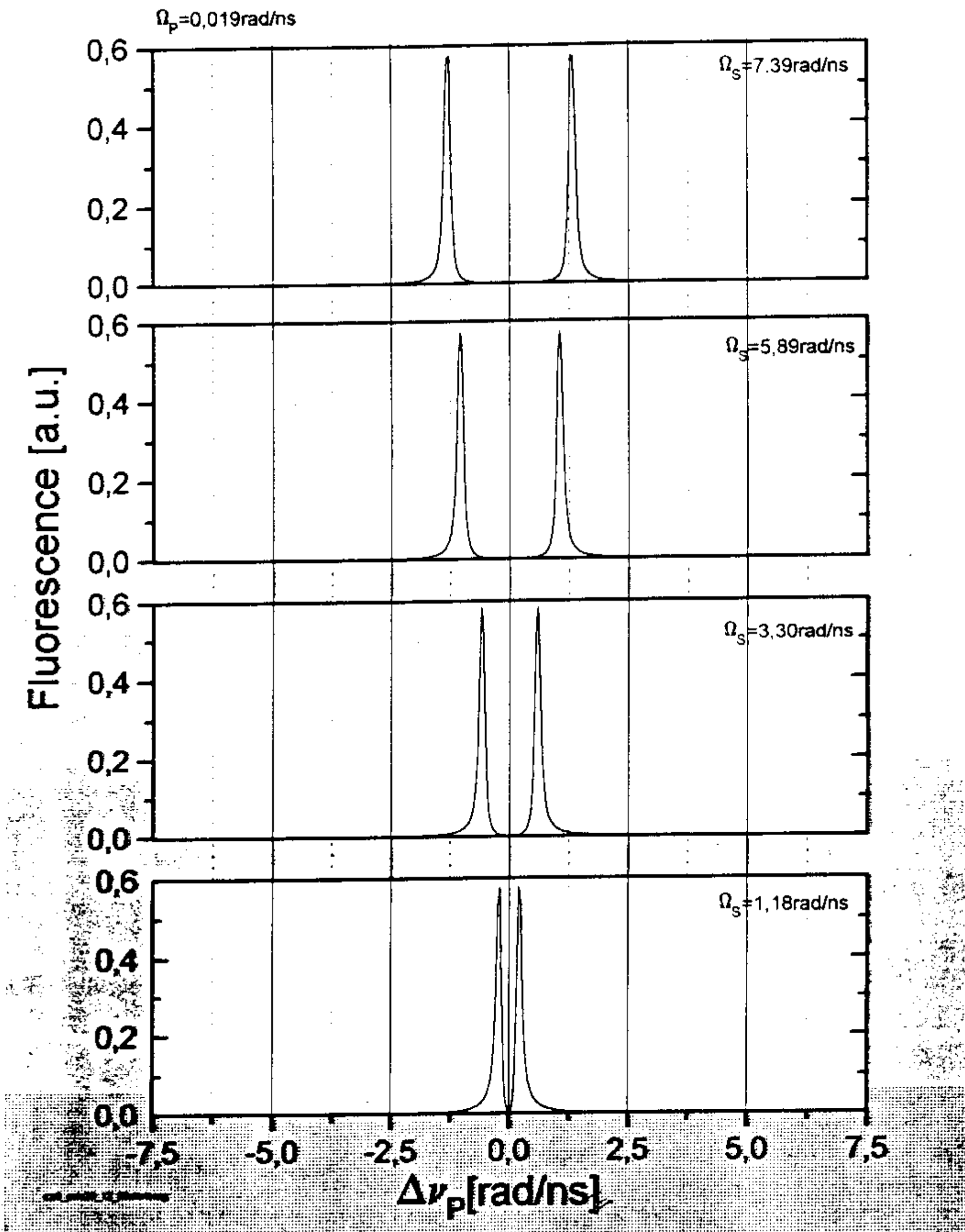




Experimental demonstration
of dynamic Stark splitting

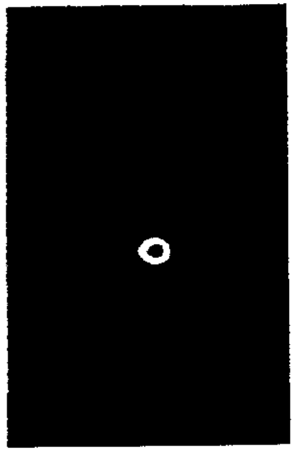


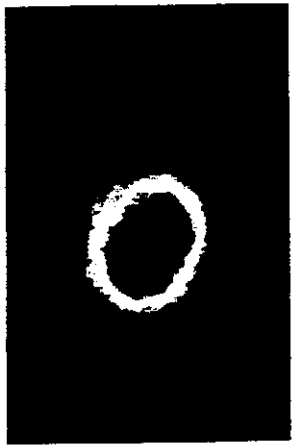
Ne*
P_P=6,2µW
P_S=55mW
P_P=3,4µW
P_S=35mW
P_P=4,0µW
P_S=11mW
P_P=6,9µW
P_S=1.4mW
Fluorescence [Cnts/0.5s]
Δν_P[MHz]



Ω_P=0,019rad/ns
Ω_S=7.39rad/ns
Ω_S=5,89rad/ns
Ω_S=3,30rad/ns
Ω_S=1,18rad/ns
Fluorescence [a.u.]
Δν_P[rad/ns]

Pumplaser





Stokeslaser

Die beiden Strahlprofile wurden bei max. Leistung aufgenommen, also

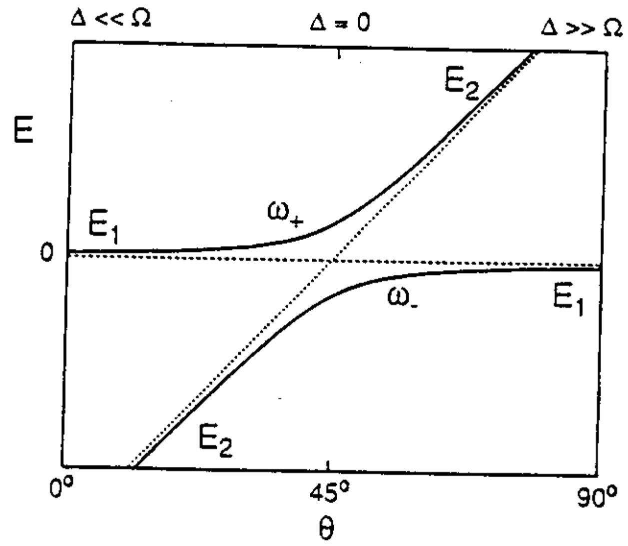
$P_{Pump} = 26\,mW$

$P_{Stokes} = 44\,mW$

HJ

Consequences of
detuning and chirping

Adabatic passage



$$\omega_\pm = \frac{1}{2}\left(\Delta \pm \sqrt{\Omega^2 + \Delta^2}\right)$$

$\Delta = 0$: $\omega_+ = \frac{1}{2}\Omega \qquad \omega_- = -\frac{1}{2}\Omega$

$|\Delta| \gg \Omega$: $\omega_\pm = \frac{1}{2}\left(\Delta \pm |\Delta|\right)$

$\Delta < 0 \qquad \omega_+ = 0 \qquad \omega_- = -\Delta$

$\Delta > 0 \qquad \omega_+ = \Delta \qquad \omega_- = 0$

Character of dressed eigenstates and adiabatic population transfer

$$\cos(2\theta) = \frac{\Delta}{\sqrt{\Delta^2 + \Omega^2}} \qquad \sin(2\theta) = \frac{|\Omega|}{\sqrt{\Delta^2 + \Omega^2}}$$

$$|\Phi_-\rangle = \cos\theta\,|1\rangle - e^{-i\varphi}\sin\theta\,|2\rangle$$

$$|\Phi_+\rangle = e^{i\varphi}\sin\theta\,|1\rangle + \cos\theta\,|2\rangle$$

$\Delta = 0 \qquad \cos 2\theta = 0, \quad \sin 2\theta = 1 \Rightarrow \theta = 45°$

$$|\Phi_-\rangle = \frac{1}{\sqrt{2}}\left(|1\rangle - e^{-i\varphi}|2\rangle\right)$$

$$|\Phi_+\rangle = \frac{1}{\sqrt{2}}\left(e^{i\varphi}|1\rangle + |2\rangle\right)$$

$|\Delta| \gg \Omega$

$\Delta < 0 \qquad \cos 2\theta = -1, \quad \sin 2\theta = 0 \Rightarrow \theta = 90°$

$$|\Phi_-\rangle = -e^{-i\varphi}|2\rangle; \quad |\Phi_+\rangle = |1\rangle e^{i\varphi}$$

$\Delta > 0 \qquad \cos 2\theta = 1, \quad \sin 2\theta = 0 \Rightarrow \theta = 0°$

$$|\Phi_-\rangle = |1\rangle; \quad |\Phi_+\rangle = |2\rangle$$

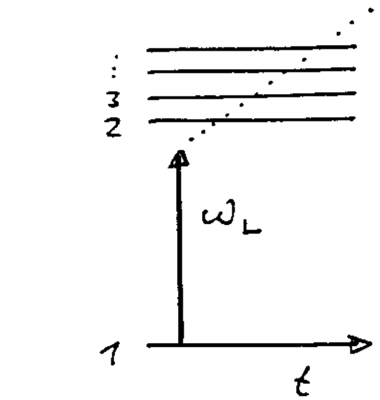
Initially: population in $|1\rangle$

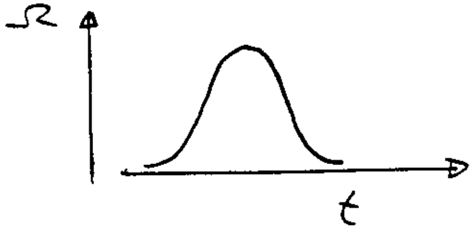
$|\Delta| \gg \Omega \quad \Delta > 0 \quad |\langle 1|\Phi_-\rangle|^2 = 1$

$\Downarrow \qquad\qquad \Downarrow$

$|\Delta| \gg \Omega \quad \Delta < 0 \quad |\langle 2|\Phi_-\rangle|^2 = 1$

Adiabatic passage to a single level in a multilevel system by chirping (pulsed lasers)





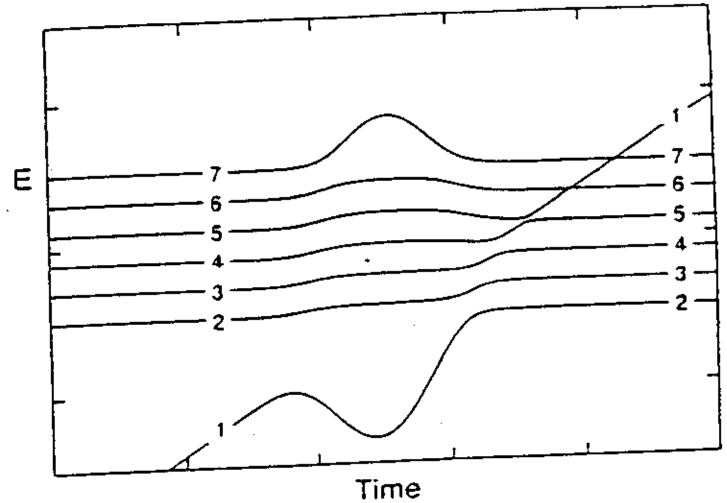


Fig. 4. Variation of the dressed state eigenvalues with time for a chirped laser pulse when level 2 (see Fig. 2) is replaced by a group of 6 evenly spaced levels. Labels at left and right identify the unperturbed states associated with these eigenvalues; adiabatic following will transfer all population from state 1 to state 2. The variation of the eigenvalues due to the pulse envelope produces the bulge. The frequency chirping is evidenced by the line crossing the group of levels. Early there occurs a pronounced avoided crossing between states 1 and 2. The interaction at the subsequent crossings is smaller because the Rabi frequency is smaller.

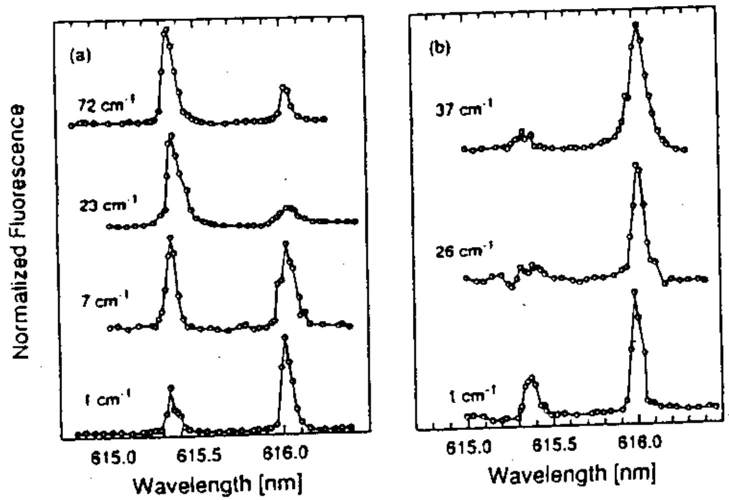
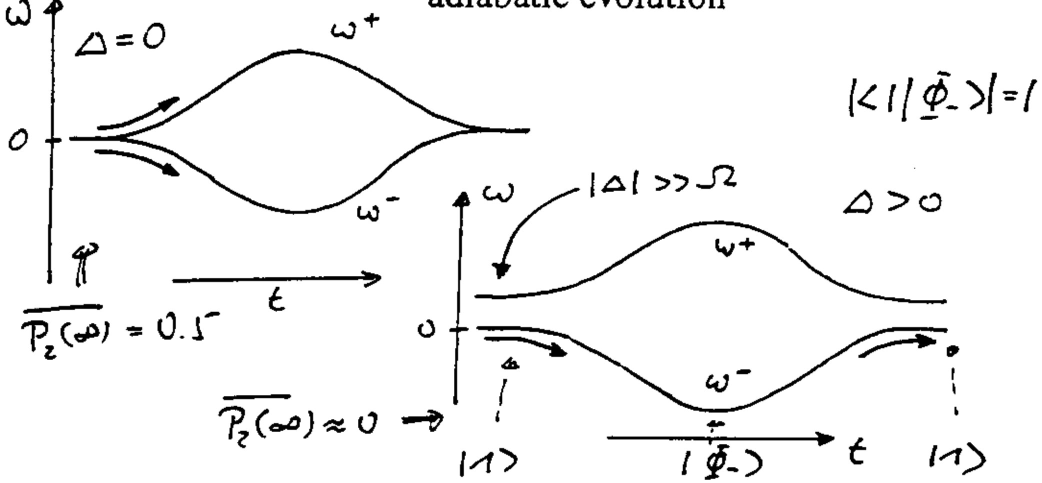


Fig. 6. Normalized relative intensities of the fluorescence from the 5s level of Na populated by the probe laser from the 3p ($^2P_{1/2}$) at 615.42 nm or 3p ($^2P_{3/2}$) at 616.07 nm. In frame (a) the chirp goes from red to blue and produces predominantly population of the lower fine structure level. In frame (b) the chirp goes from blue to red and leads to predominant population of the upper fine structure level. The numbers in ... Rabi frequencies. Adapted from Melinger et al.[42] and

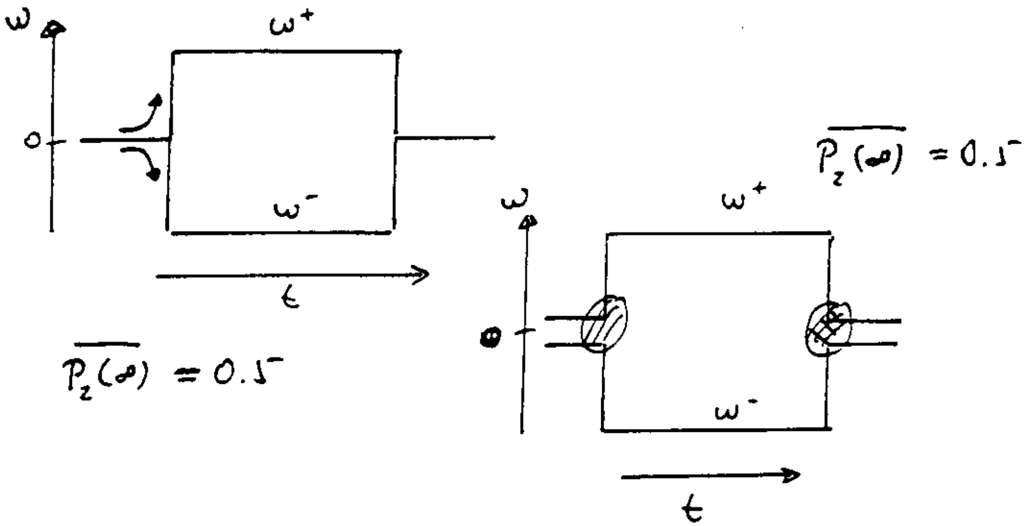
Coherent Population Return
elimination of saturation broadening

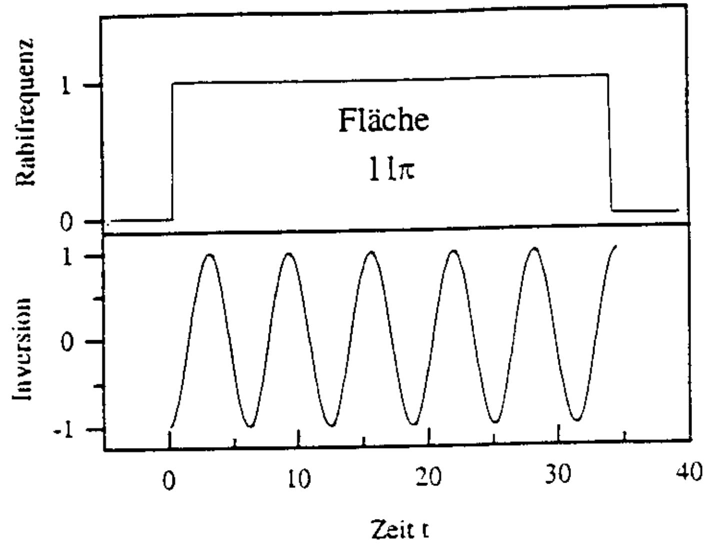
pulsed excitation, $\Delta\tau_{pulse} << \tau_{spont}$.

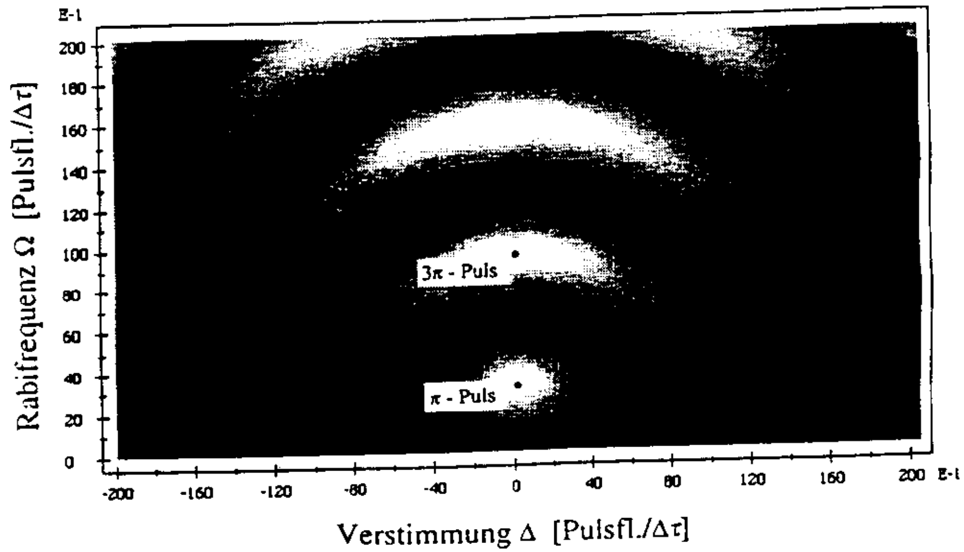
Gaussian pulses: smooth change of $\Omega(t)$
adiabatic evolution



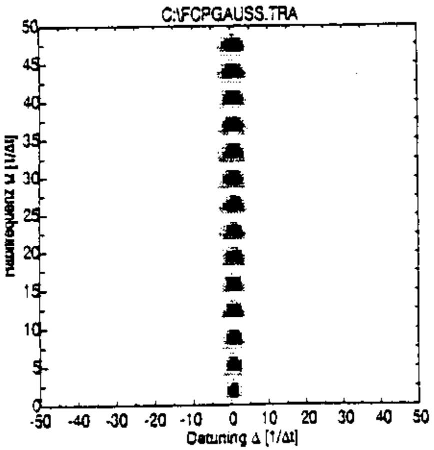
Rectangular pulses: sudden changes of $\Omega(t)$
strong diabatic coupling



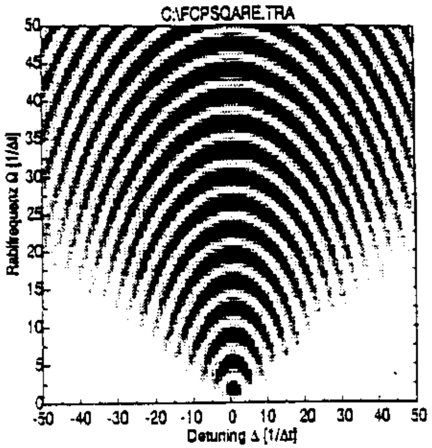




Damping of Rabi oscillations due to pulse to pulse intensity fluctuations



Gaussian pulse



Rectangular pulse

$\Delta = 0$

Rabi oscillations for both pulse shapes

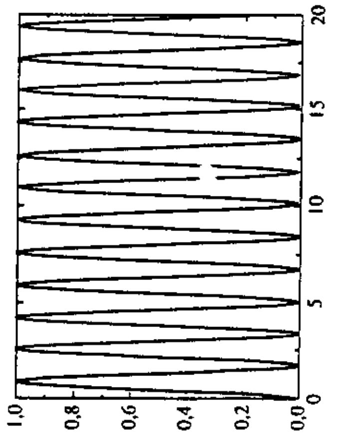
$\Delta \neq 0$

Rectangular pulse: sudden change of Ω
Strong diabatic coupling
both dressed states populated
population in 2 depends on phase of Rabi oscillation

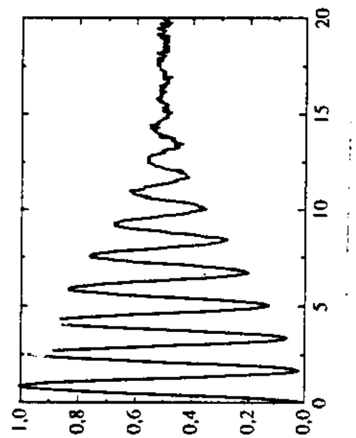
Gaussian pulse: at early times: $|\Delta| >> \Omega$
only one dressed state populated
smooth change of Ω
adiabatic evolution
remaining in given dresse state
„late“: same situation as „early“
no population remains in $|2>$

Population transfer in a two-level system and the consequences of intensity fluctuations

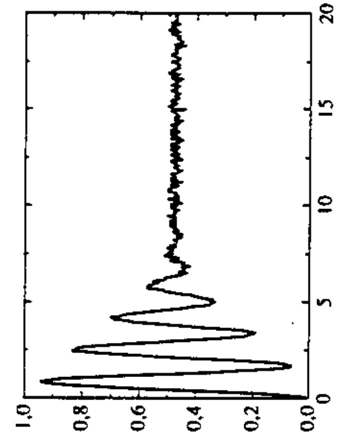
$\Delta = 0$



Pump Rabifreq. Ω_P (GHz)
Fluktuations of Ω_P: +/- 0 %



Pump Rabifreq. Ω_P (GHz)
Fluktuations of Ω_P: +/- 5 %



Pump Rabifreq. Ω_P (GHz)
Fluktuations of Ω_P: +/- 10 %

(2) Three-level systems

(a) Rate equations - Schrödinger equation
    Dressed states

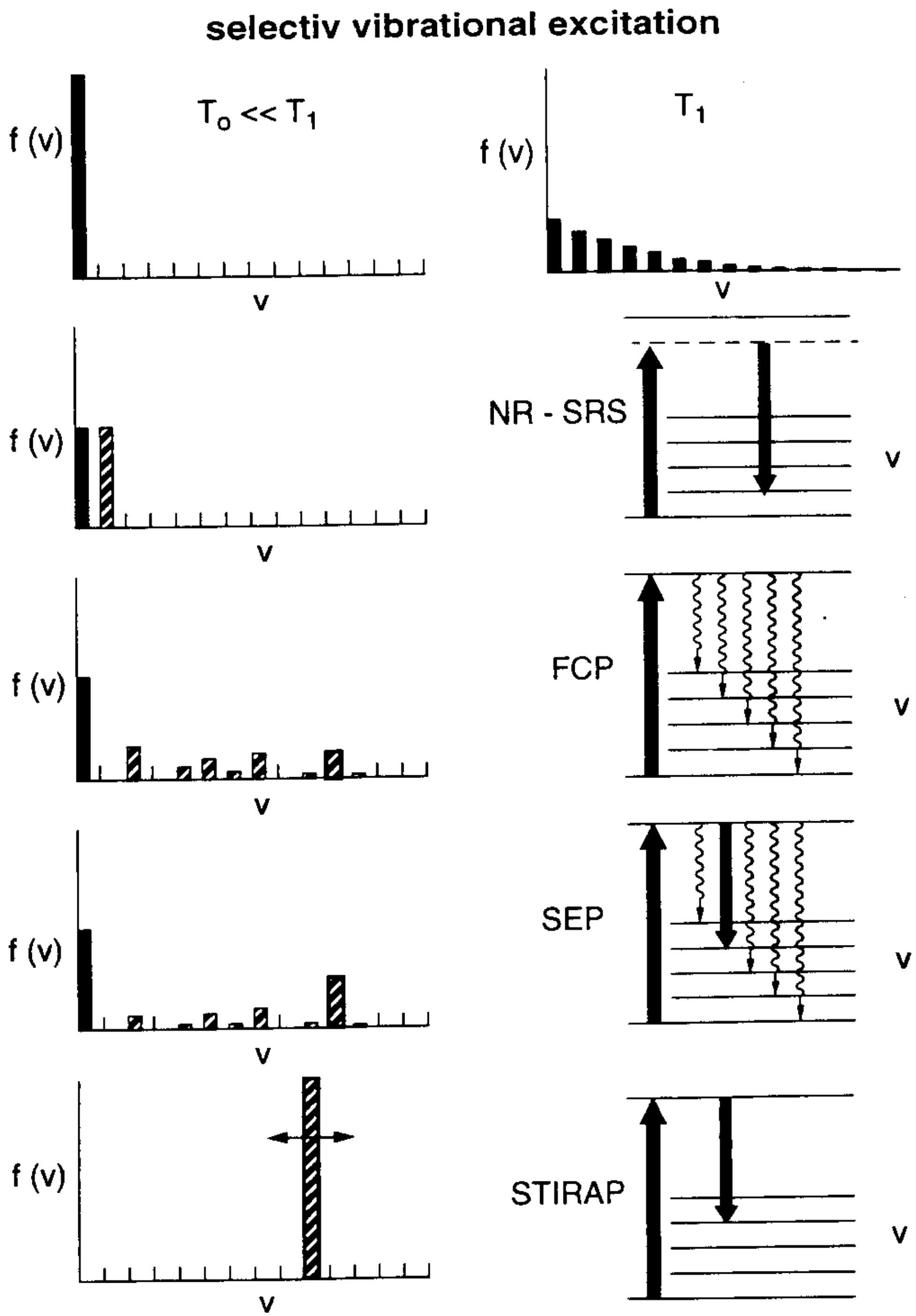
(b) The STIRAP approach
    Dark resonances
    The robustness of the transfer
    Example 1: Ne*
    Example 2: NO
    Example 3: SO_2

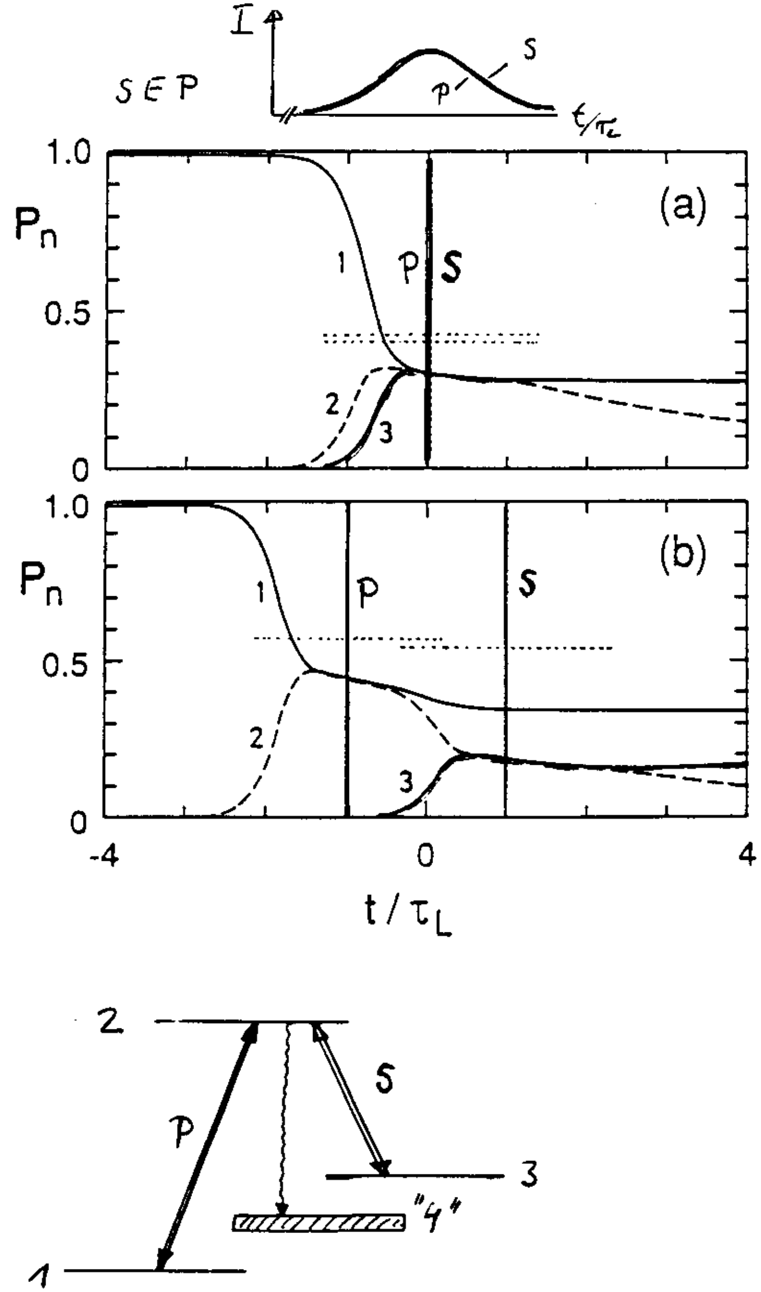
(c) Conditions for adiabatic following

(d) The consequences of two-photon detuning

A motivation

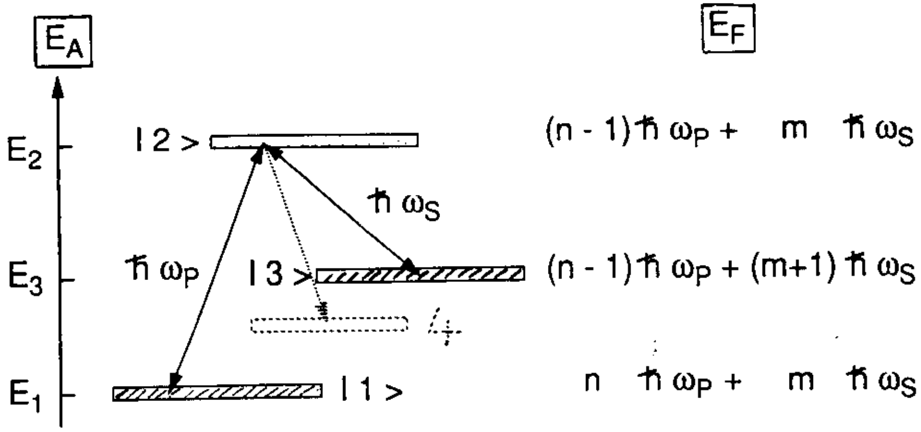
Rate equation solutions

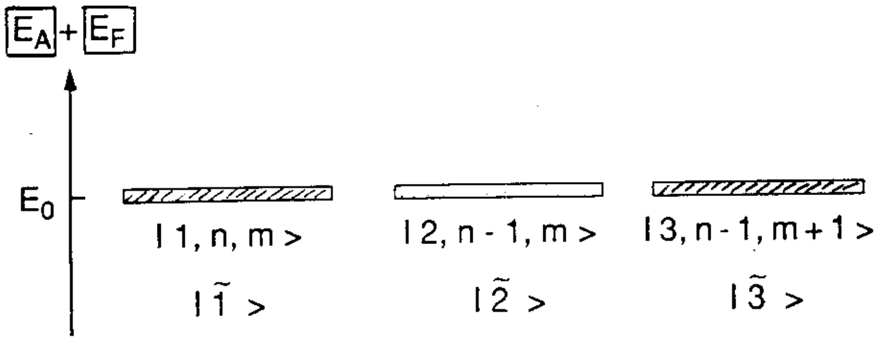




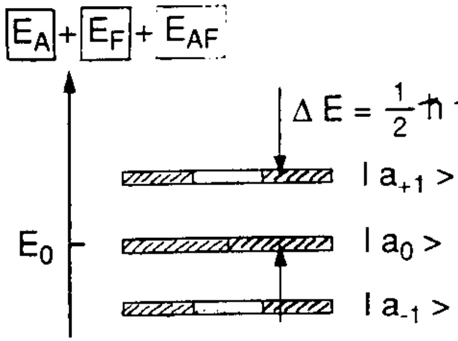
Three-level dressed states

"dressed states"   3 - Niveau - System

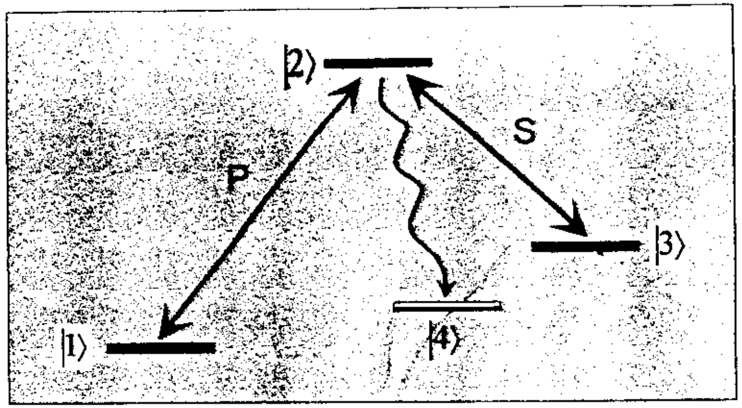




$$\Delta E = \frac{1}{2}\hbar\sqrt{\Omega_P^2 + \Omega_S^2} = \Delta E(t)$$



$$|a_i> = \sum_{k=1}^{3} a_{ik}(\Omega_P, \Omega_S, \Delta)\,|\tilde{k}>$$



RWA-Hamiltonian:

$$H = \frac{\hbar}{2}\begin{bmatrix} 0 & \Omega_P(t) & 0 \\ \Omega_P(t) & 2\Delta_P & \Omega_S(t) \\ 0 & \Omega_S(t) & 2(\Delta_P - \Delta_S) \end{bmatrix}$$

with Rabi-frequency $\Omega_i = \frac{\mu E_i}{\hbar}$ and laser-detuning Δ_i

$\Longrightarrow$ **dressed states** $|a^+\rangle, |a^-\rangle, |a^0\rangle$ on two photon resonance:

$$|a^+\rangle = \sin\Theta\sin\Psi|1\rangle - \cos\Psi|2\rangle + \cos\Theta\sin\Psi|3\rangle$$

$$|a^-\rangle = \sin\Theta\cos\Psi|1\rangle - \sin\Psi|2\rangle + \cos\Theta\cos\Psi|3\rangle$$

TRAPPED STATE $\longrightarrow$ $|a^0\rangle = \cos\Theta|1\rangle - \sin\Theta|3\rangle$ $\longleftarrow$

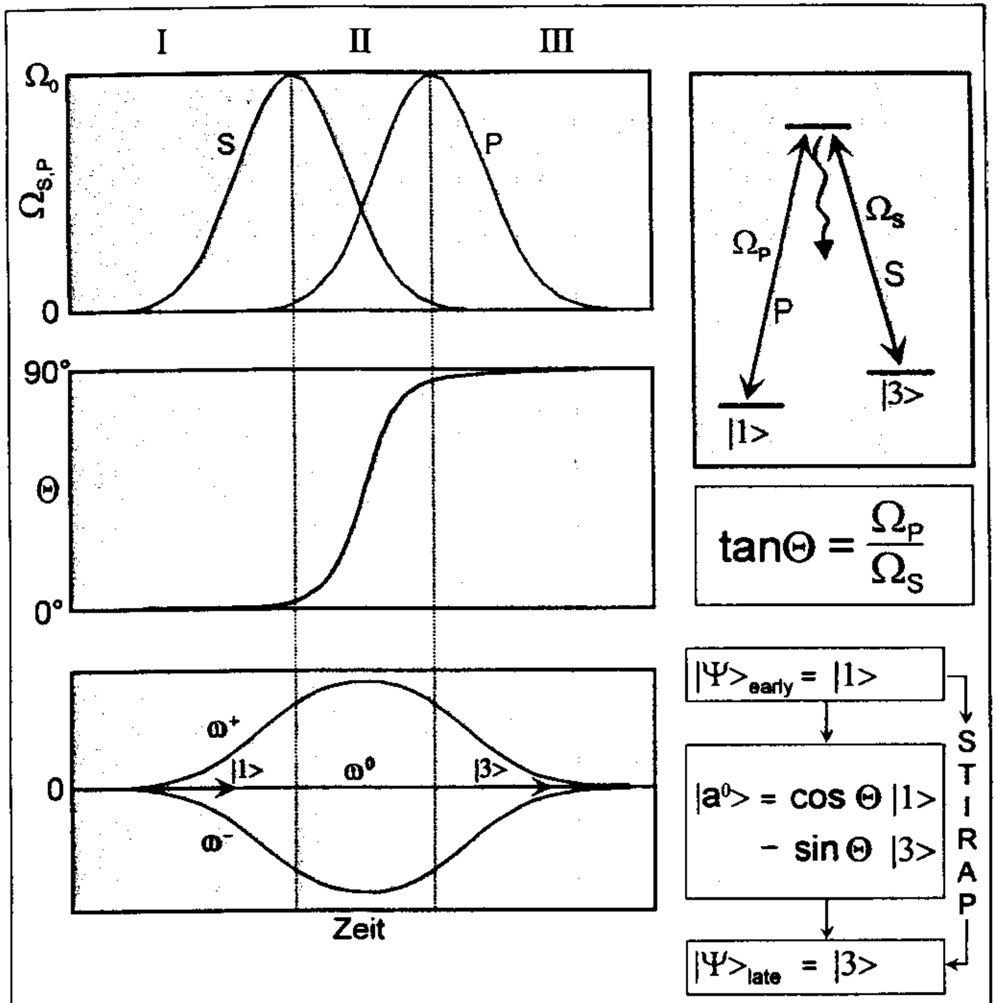
mixing angles: $\Psi = \Psi(\Omega_P, \Omega_S, \Delta; t)$ $\quad \tan\Theta(t) = \frac{\Omega_P(t)}{\Omega_S(t)}$

control of Θ: |1> $\xrightarrow[\text{transfer}]{\text{population}}$ |3>

J. Chem. Phys. 92, 5363 (1990)   Phys. Rev. A 52, 566 (1995)

Time Evolution of Ω, Θ and $\omega^{0,\pm}$

three-level system



eigenvalues:

$$\omega^{\pm} = \Delta_P \pm \sqrt{\Delta_P^2 + \Omega_P^2 + \Omega_S^2}$$

$$\longrightarrow \quad \omega^0 = 0 \quad \longleftarrow$$

STIRAP = <u>STI</u>mulated <u>R</u>aman <u>A</u>diabatic <u>P</u>assage

$$\hbar \frac{\partial}{\partial t} |\Psi(t)\rangle = H(t)\,|\Psi(t)\rangle$$

$$H(t)\,|\Phi(t)\rangle = \omega(t)\,|\Phi(t)\rangle$$

($|\Phi\rangle$ same as $|a\rangle$)

consequences of $\omega = 0$ for all t:

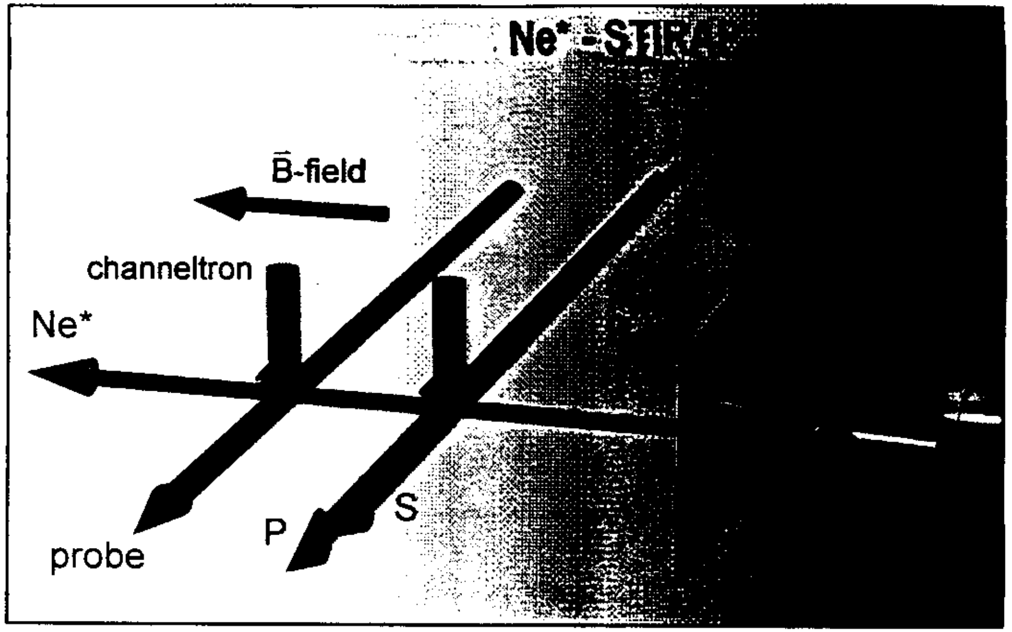
$$H(t)\,|\Phi^0(t)\rangle = \hbar \frac{\partial}{\partial t} |\Phi^0(t)\rangle = 0$$

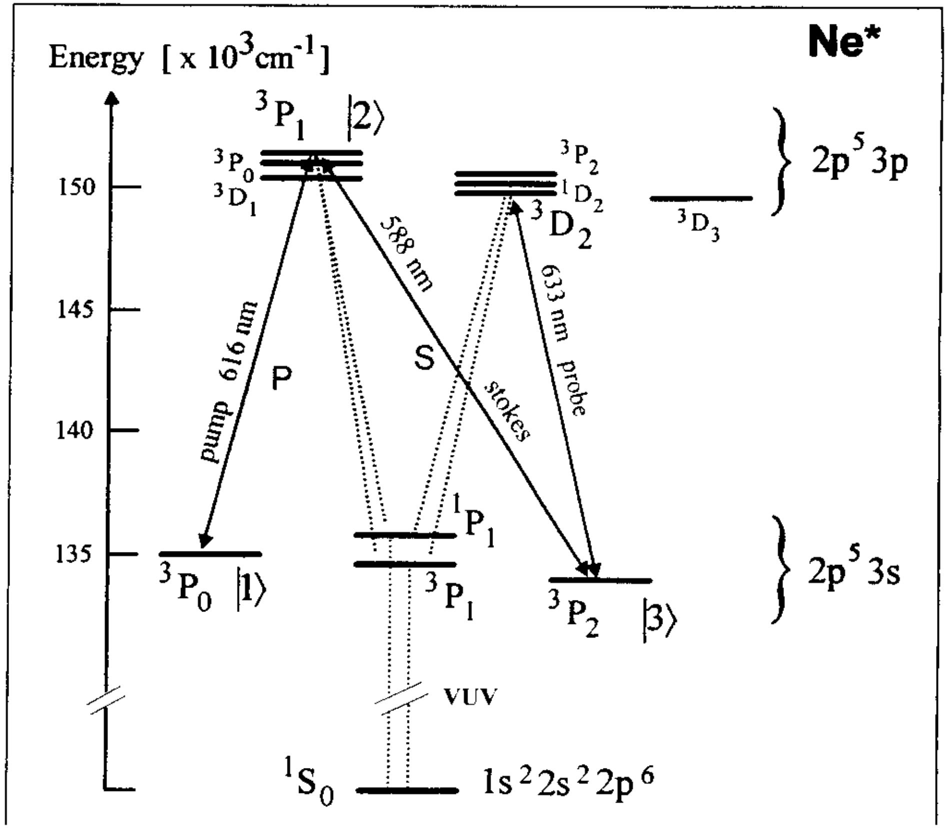
no change of stationary state $|\Phi^0(t)\rangle$

condition for existence of zero-energy states:

$$\mathrm{Det}\,|H(t)| = 0$$

Verification for population transfer between metastable states of Ne*



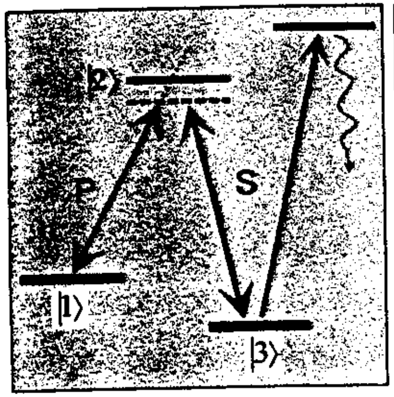


Ne* in an atomic beam

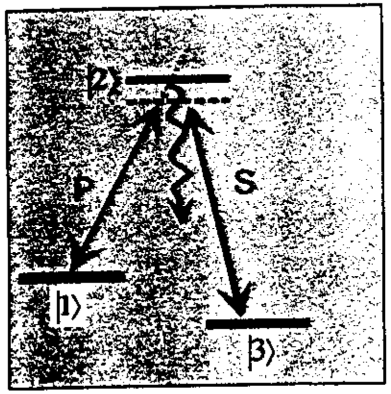
[illegible]

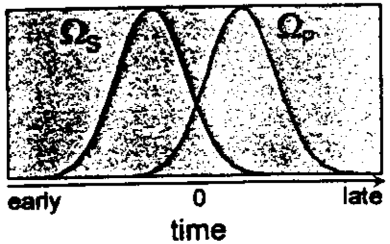
POPULATION TRANSFER and related **DARK RESONANCE**

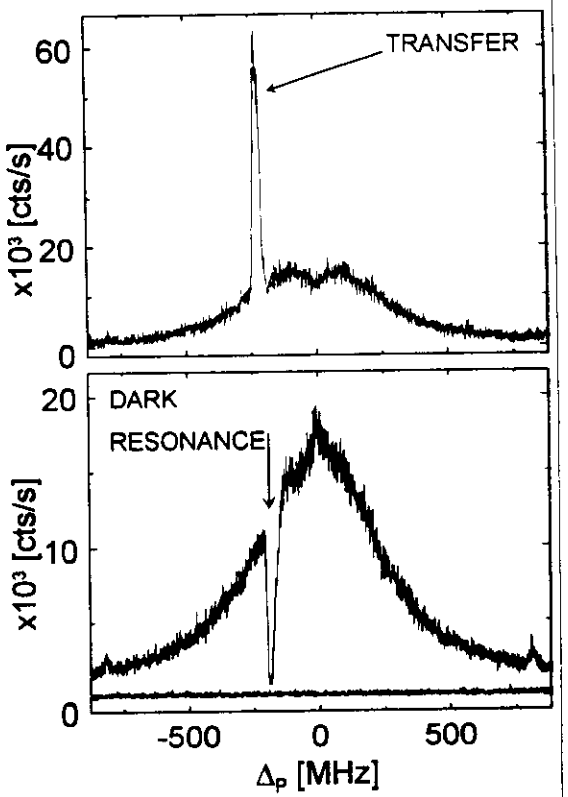
— the three level situation—



$\vec{E}_P \parallel \vec{E}_S \parallel \vec{B}$







· Situation in the experiment:

! interaction time ≈ $20 \cdot \tau_{radiative}$ **!**

Phys. Rev. A 54, 1556 (1996)

First observation
(and interpretation)
of dark resonances

ulti-mode
aser -
xcitation
f Na in
apour

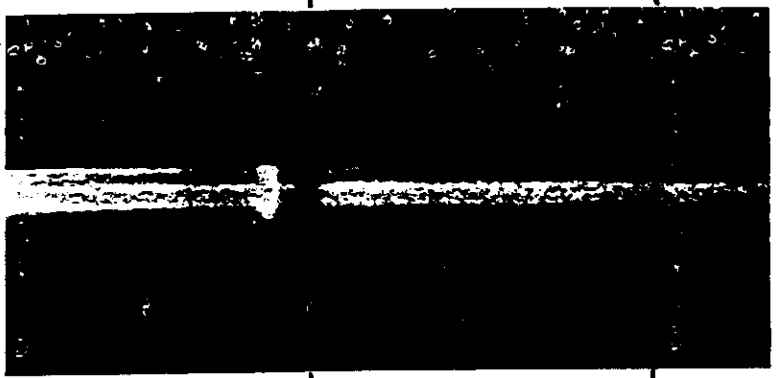
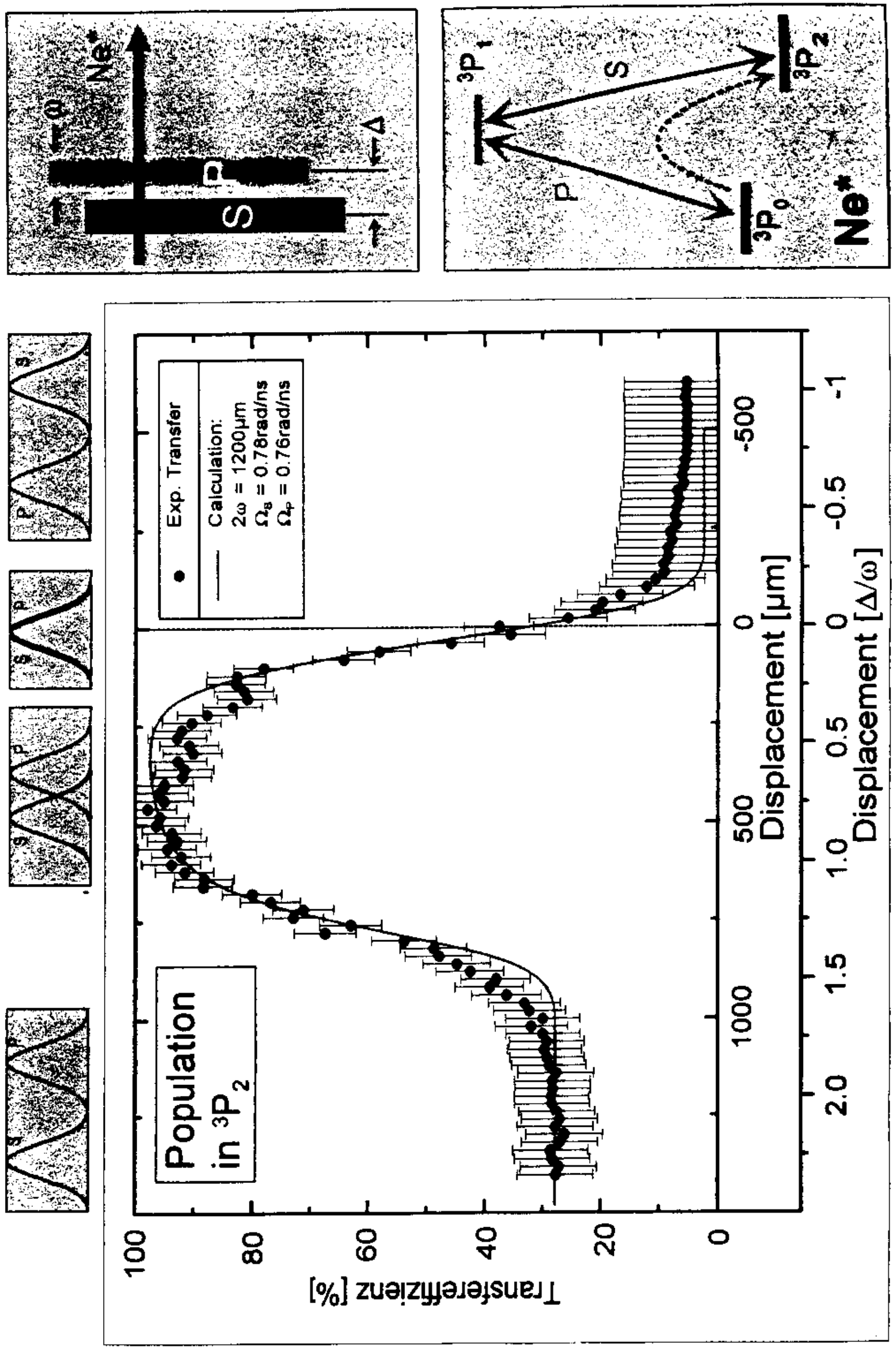


Fig. 12. – Black lines as observed by tilting the magnetic field to make an angle of ~10° with the direction of the pumping radiation. The bright spot, due to the (2, −2; 2, −1) r.f. transition, is used for calibrating purposes.

→ B field gradient



Nonabsorbing Atomic Coherences by Coherent Two-Photon Transitions in a Three-Level Optical Pumping.

E. Arimondo

Istituto di Fisica dell'Università - Pisa

G. Orriols

Laboratorio di Fisica Atomica e Molecolare del C.N.R. - Pisa
Departament d'Optica, Universitat de Barcelona - Barcelona

Lett.Nuov.Cim. 17, 333 (1976)

(ricevuto il 30 Agosto 1976)

population
in the
excited
state

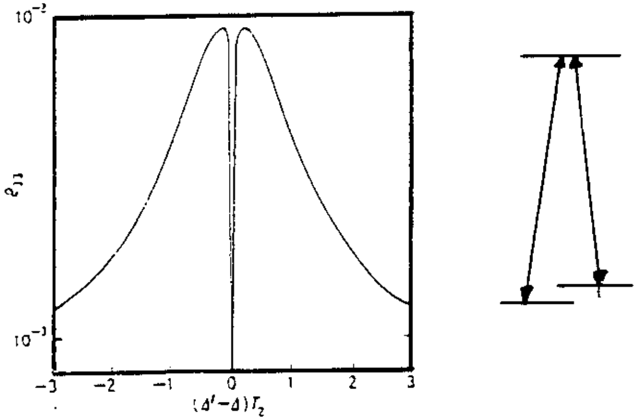
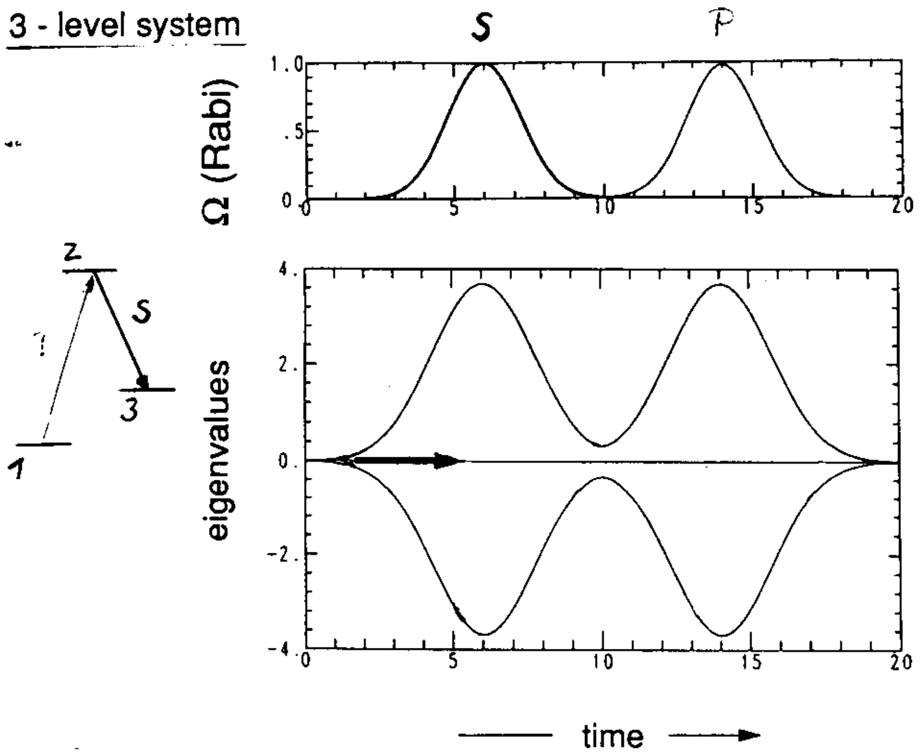
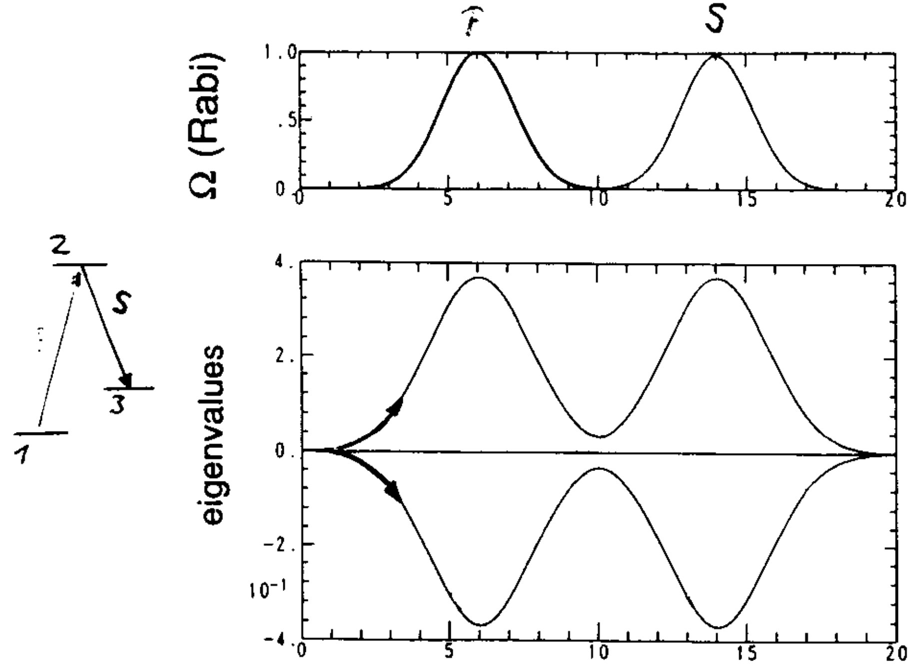


Fig. 1. – Excited-state occupation as a function of the resonance parameter $(\Delta' - \Delta)T_2$, for $\Delta = 0$, $\alpha T_2 = |\beta| T_2 = 0.1$, $\Phi = 0$, $\tau_1 = \tau_2 = 10^4 T_2$, $T_1 = T_2/2$. The minimum value of ϱ_{33} in $9\cdot10^{-4}$.

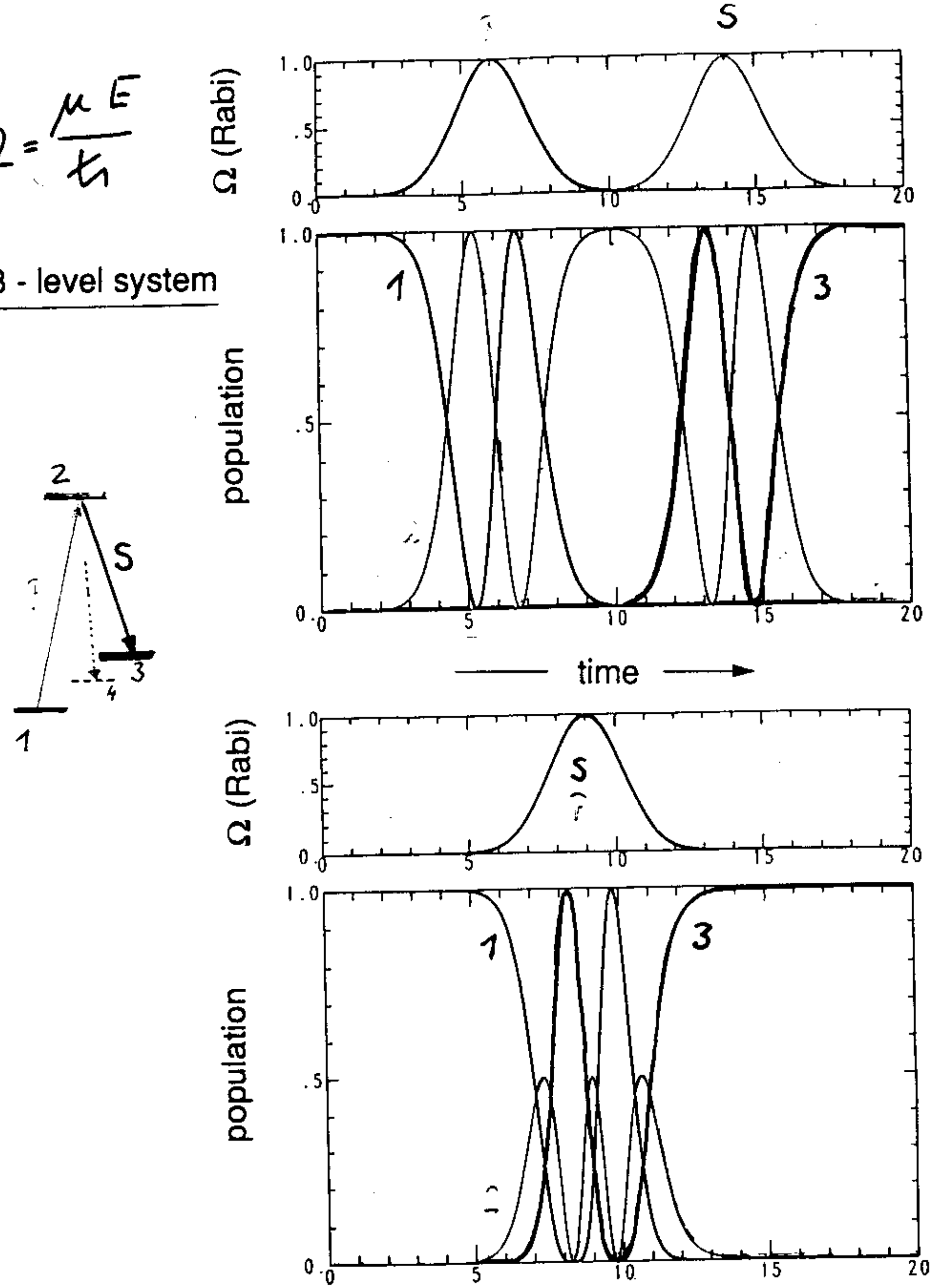
3 - level system





$$\Omega = \frac{\mu E}{\hbar}$$

3 - level system

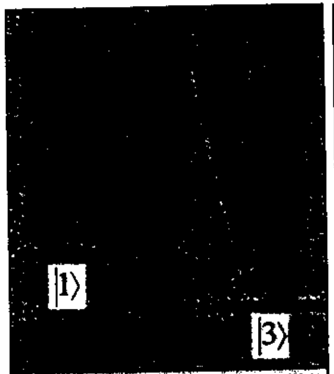


$$A(t) = \int_0^t \Omega(t')\,dt'$$

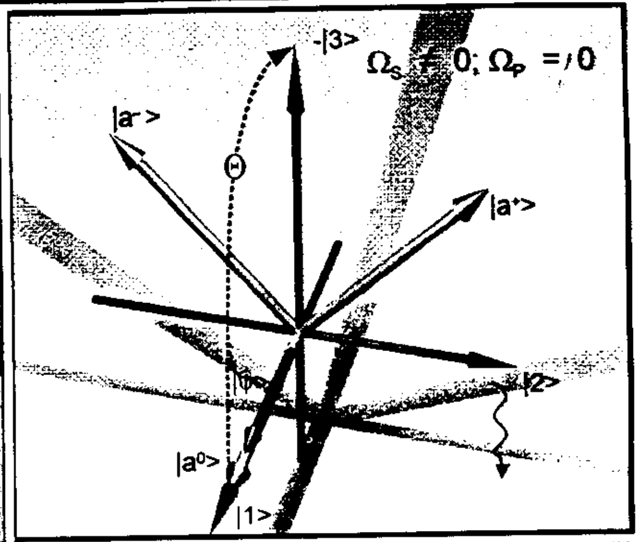
$t \to \infty$

Conditions for
Adiabatic Following

STIRAP: the main features



- on two-photon resonance
- on one-photon resonance



$$\tan\Theta(t) = \frac{\Omega_P(t)}{\Omega_S(t)}$$

|a⁰>, |a⁺>, |a⁻> : dressed states

|1>, |2>, |3> : bare states

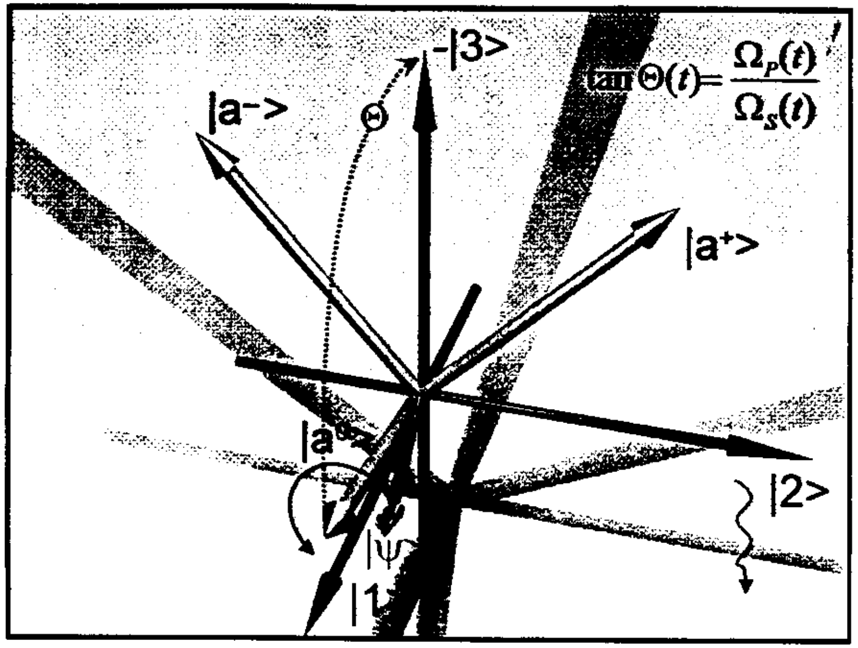
adiabatic evolution - if

$$|\dot{\Theta}| \ll |\omega^0 - \omega^{\pm}|$$

then

- efficiency → **100%**
- selectivity → **100%**
- experimentally robust against small fluctuations of
  - → intensity
  - → frequency
  - → overlap

Adiabatic following



general:

$$|\langle a^{\pm}|\dot{a}^0\rangle| \ll |\omega^0 - \omega^{\pm}|$$

in terms of the mixing angle Θ (in |1>–|3> plane)

$$\longrightarrow \quad |\dot{\Theta}| \ll |\omega^0 - \omega^{\pm}|$$

"global":

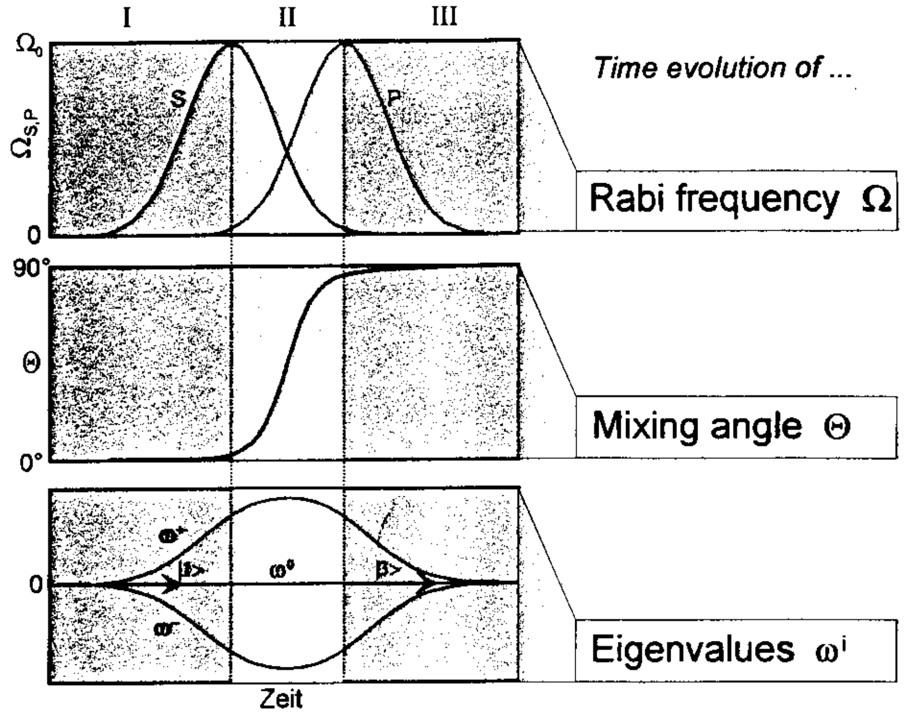
for $\Omega_{P,max} \sim \Omega_{S,max} = \Omega$ and ~ gaussian pulses

$$\longrightarrow \quad \Omega\,\Delta\tau > 10$$

rewritten in terms of pulse energy and inclusion of phase-fluctuations:

$$\Omega^2\Delta\tau > (100/\Delta\tau)\,(1 + (\Delta\omega_L/\Delta\omega_{FTL})^2)$$

J. Chem. Phys. 97, 4251, 1992



	$\lvert\langle a^{\pm}\lvert\dot{a}^0\rangle\rvert \ll \lvert\omega^0 - \omega^{\pm}\rvert$	$\lvert\dot{\Theta}\rvert \ll \lvert\omega^0 - \omega^{\pm}\rvert$
I	≈0	small ⟶ 0
II	large	very large
III	≈0	small ⟶ 0

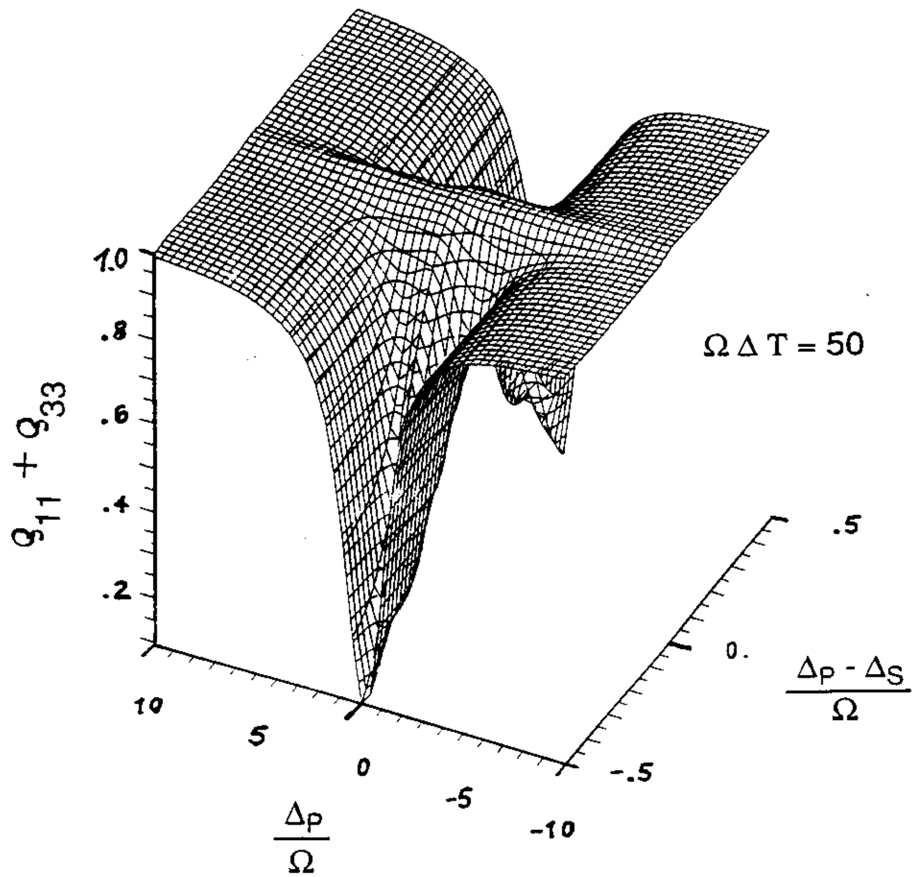
Adiabatic following

$\Omega\,\Delta\tau > 10$ $\Omega^2\Delta\tau > (100/\Delta\tau)$	transforme limited pulse
$\Omega^2\Delta\tau > (100/\Delta\tau)\,(1 + (\Delta\omega_L/\Delta\omega_{FTL})^2)$	with phase fluctuations

J. Chem. Phys. 97, 4251, 1992

Various other aspects

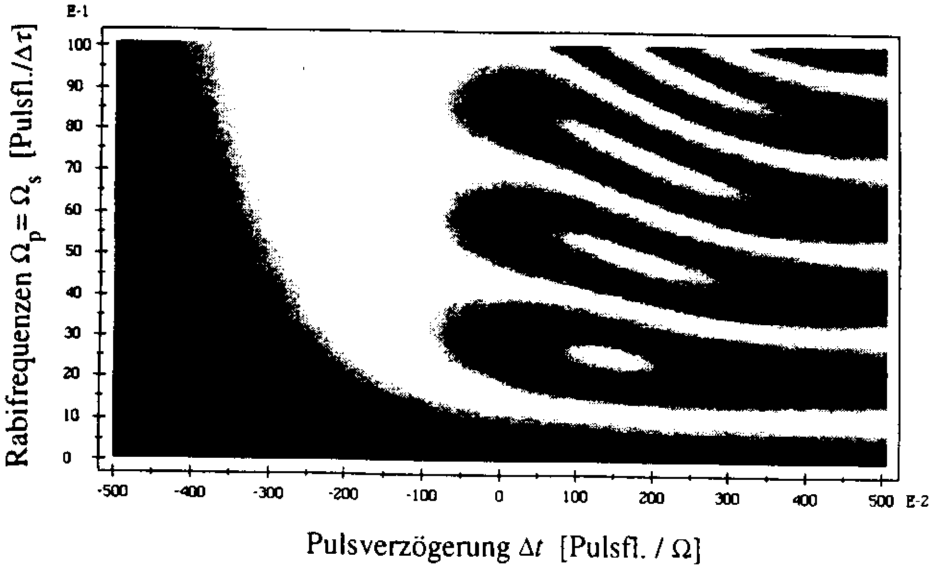
Population remaining in the system



Loss from three-level system (only) by spontaneous emission from level 2

„STIRAP-ridge" shows trapped state character of superposition state

Population Transfer by STIRAP



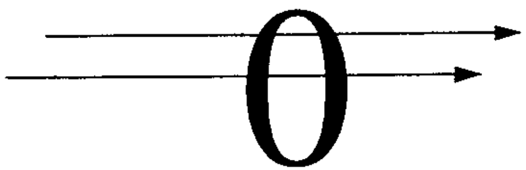
Pulse delay

Robustness of STIRAP

$$\Omega_m \Delta\tau >> 1$$

$$\Omega_m(t) = \frac{\mu(m)\, E(t)}{\hbar}$$

$$A_m = \frac{1}{\hbar} \mu(m) \int E(t)\, dt$$

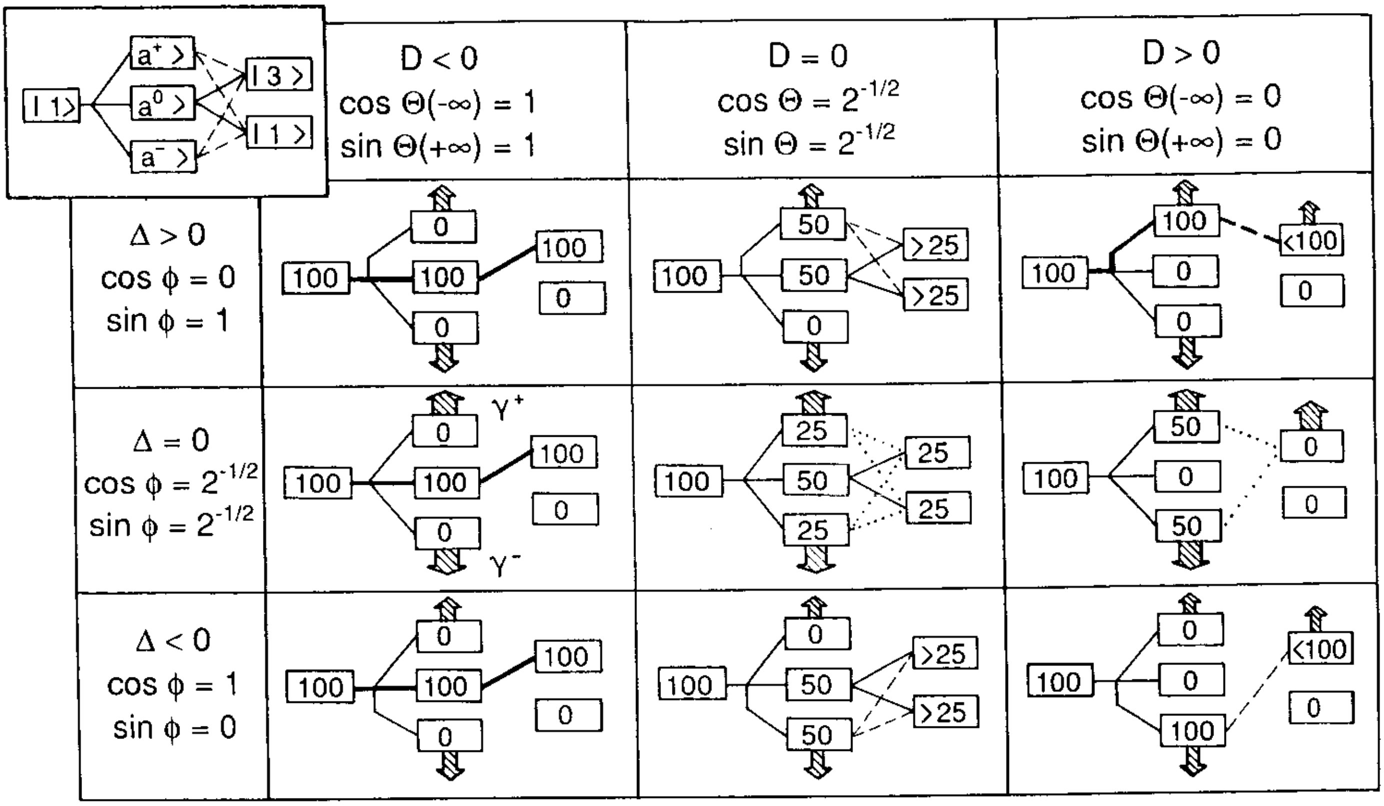


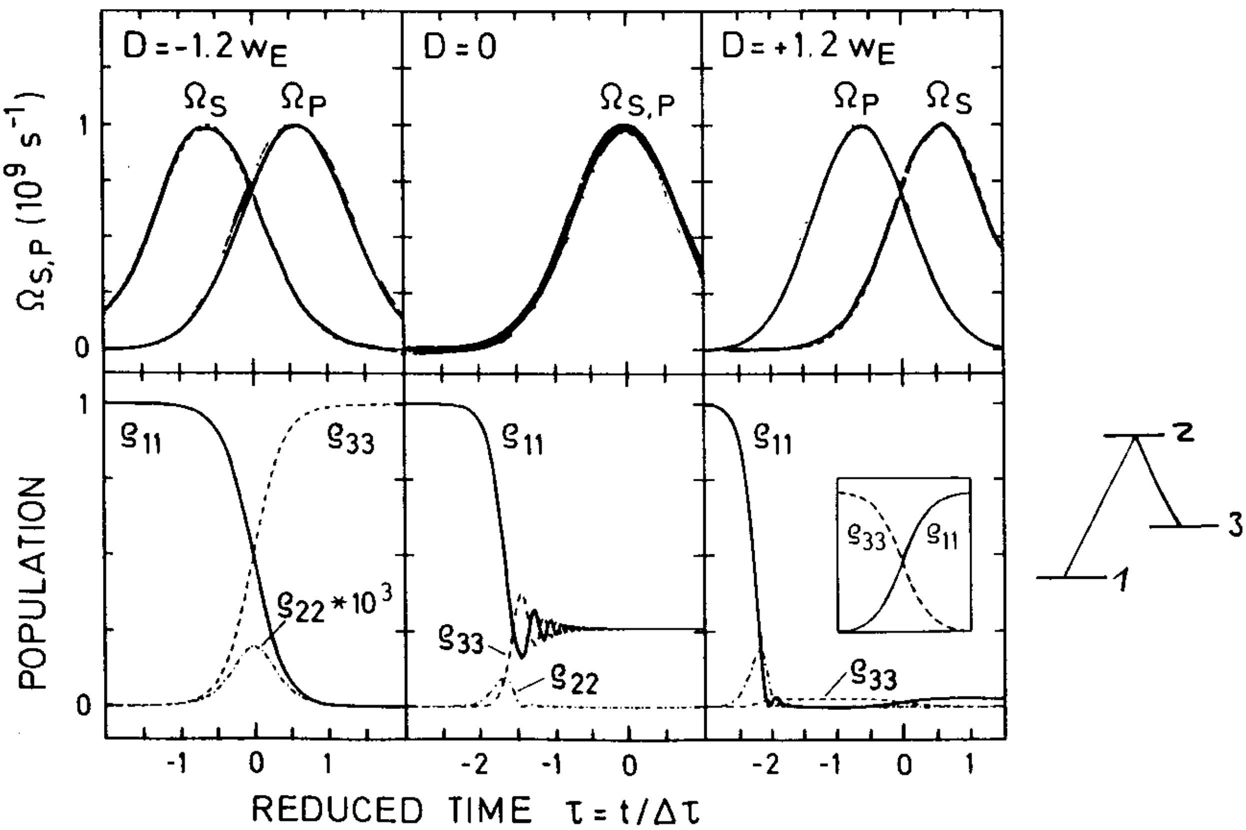
Transfer efficiency is insensitive to

m_j-state and

location of path through laser beam,

provide (1) is satisfied.


|1>
|a+>
|a0>
|a->
|3>
|1>
D < 0
cos Θ(-∞) = 1
sin Θ(+∞) = 1
D = 0
cos Θ = 2^-1/2
sin Θ = 2^-1/2
D > 0
cos Θ(-∞) = 0
sin Θ(+∞) = 0
Δ > 0
cos φ = 0
sin φ = 1
Δ = 0
cos φ = 2^-1/2
sin φ = 2^-1/2
Δ < 0
cos φ = 1
sin φ = 0
γ+
γ-


D = -1.2 wE
D = 0
D = +1.2 wE
ΩS
ΩP
ΩS,P
ΩS,P (10^9 s^-1)
POPULATION
ϱ11
ϱ33
ϱ22 * 10^3
ϱ22
REDUCED TIME τ = t/Δτ
1
2
3

classification of excitation schemes

J. Opt. Soc. Am. B/Vol. 7, No. 9/September 1990

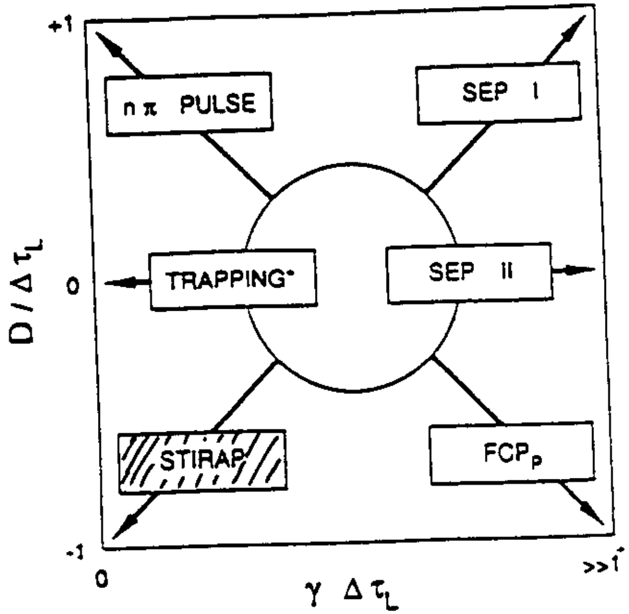


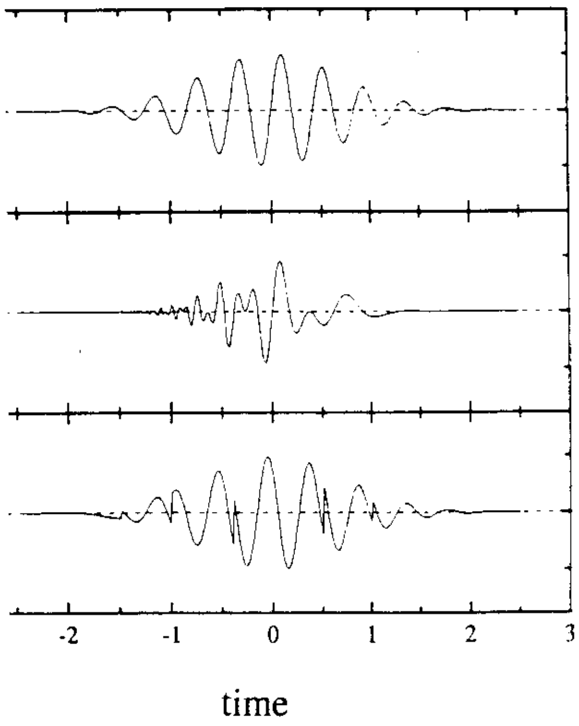
Fig. 10. Classification of the various regimes of operation according to the delay $D/\Delta\tau_L$ between pump laser and Stokes laser and the relaxation rate, given in reduced units $\gamma\Delta\tau_L$. The asterisk indicates a short electronic lifetime, $\tau_{sp}/\Delta\tau_L < 1$.

Implementation of coherent population (STIRAP) transfer with pulsed lasers

- UV needed for many molecules of interest

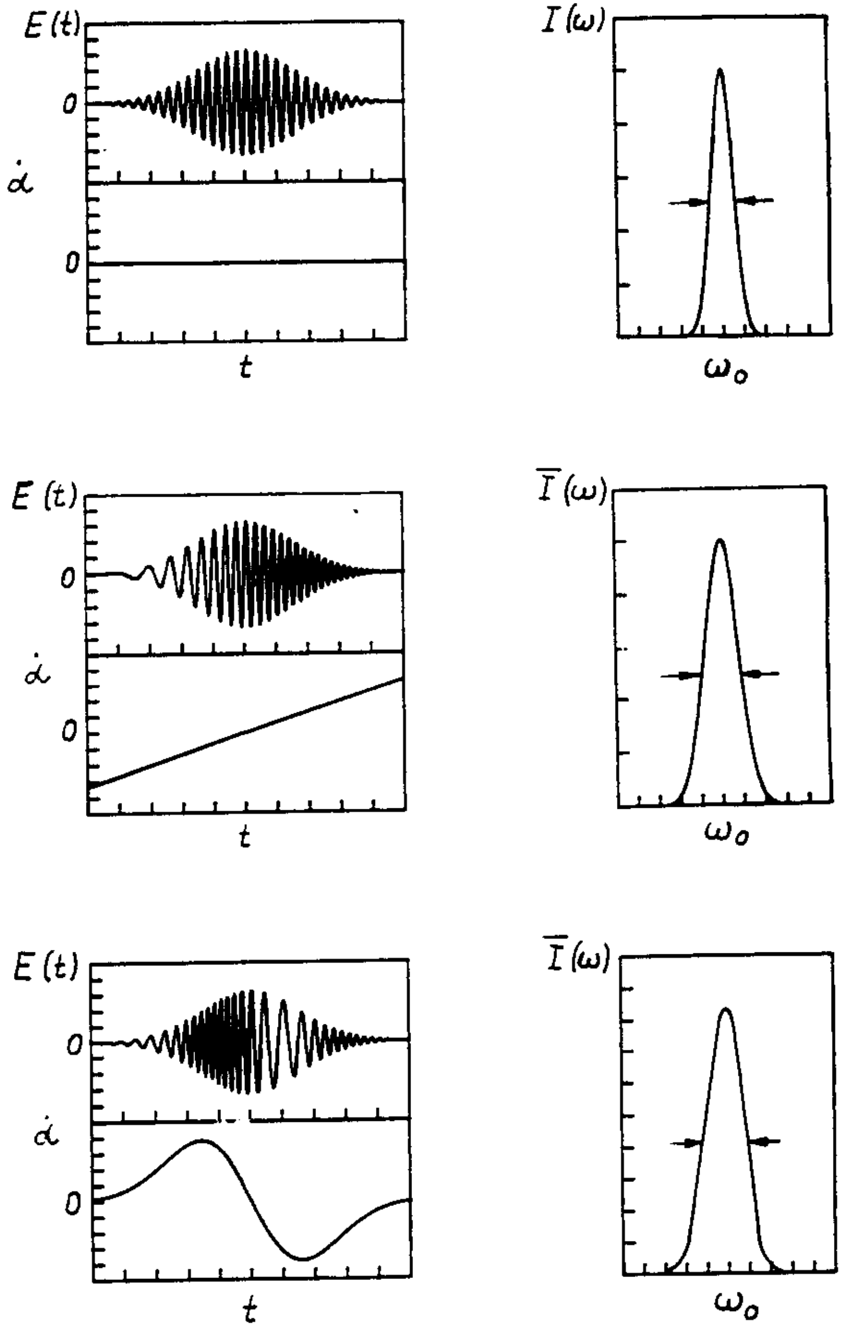
- required power only available from pulsed lasers

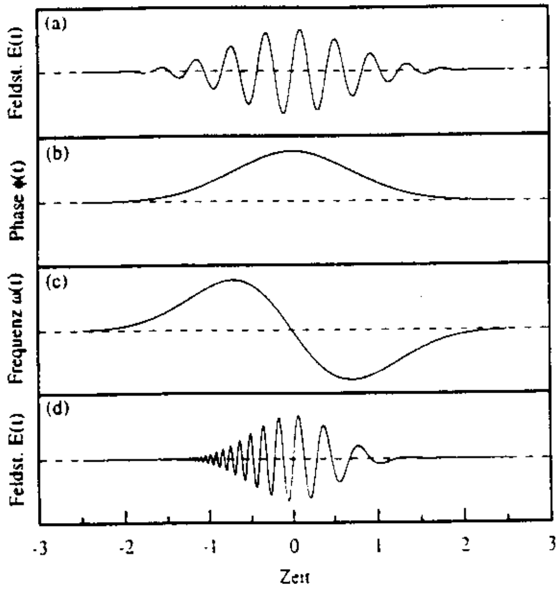
- pulsed lasers have inferior coherence properties



- coherent pulse
- pulse with phase drift
- pulse with phase jumps

$$E(t) = E_0(t)\, e^{i(\omega t + \alpha(t))} \qquad \dot{\alpha}(t) \mathrel{\hat{=}} \Delta\omega$$





self-phase modulation

Abb. 3.1: Schematische Darstellung der Selbstphasenmodulation und daraus resultierenden Pulskompression eines kohärenten Gaußpulses beim Durchgang durch ein Medium mit intensitätsabhängigem Brechungsindex: (a) Pulsverlauf vor Durchgang durchs Medium, (b) Phasenverlauf des Pulses im Medium, (c) zugehöriger Frequenzverlauf des Pulses im Medium, (d) Pulsverlauf nach Durchgang durchs Medium.

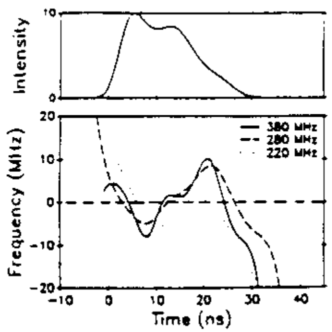


Abb. 3.2: Aus Heterodyn-Messungen rekonstruierter Intensitäts- und Frequenzverlauf eines selbstphasenmodulierten Pulses, der durch Pulsverstärkung von cw-Strahlung in Farbstoff-Zellen erhalten wurde. Pumplaser der Zellen ist ein Excimer-Laser. (Abbildung aus [48], Mischfrequenzen 220 – 380MHz)

conditions for adiabatic following

- general:

$$|\langle a^{+/-} | \dot{a}^{o} \rangle|^2 << | \omega^{o} - \omega^{+\backslash -} |^2 \qquad (= |\Delta\omega|^2)$$

- „local“ : PRA 40, 6741 (89)

$$\dot{\Theta} \approx \frac{\dot{\Omega}_P(t)\,\Omega_S(t) - \Omega_P(t)\,\dot{\Omega}_S(t)}{\Omega^2_P(t) + \Omega^2_S(t)} << |\Delta\omega|$$

- „global“: JCP 92, 5363 (90)

  for $\Omega_{P,max} \sim \Omega_{S,max} = \Omega$ and ~ gaussian pulses

  $\Omega\,\Delta\tau > 10$

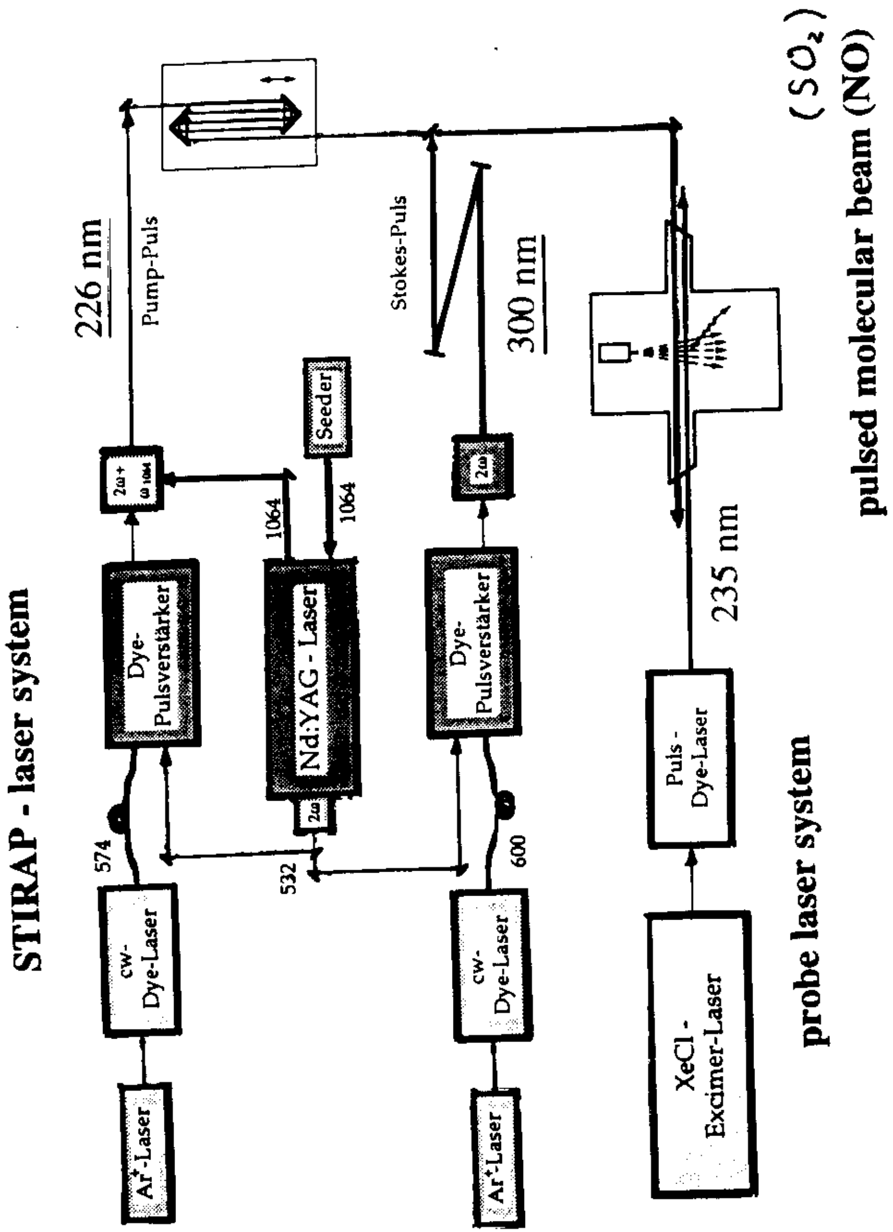
- rewritten in terms of pulse energy:

  $\Omega^2\,\Delta\tau > (100 / \Delta\tau)$

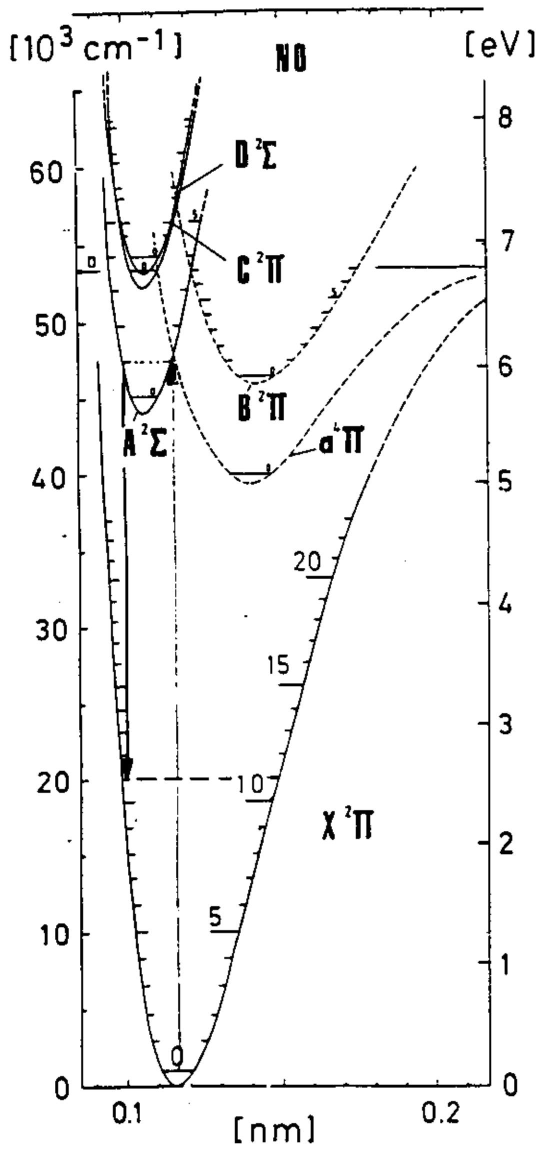
- and inclusion of phase-fluctuations: JCP 97, 4251 (92)

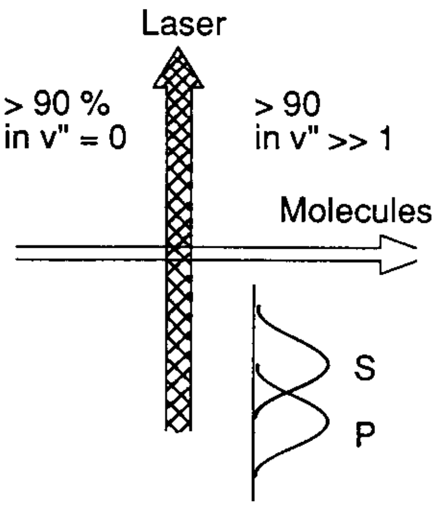
  $\Omega^2\,\Delta\tau > (100 / \Delta\tau)\,(1 + N^2)\,\Gamma$

  with $N = (\Delta\omega_L / \Delta\omega_{FTL})$



Examples: NO and SO_2

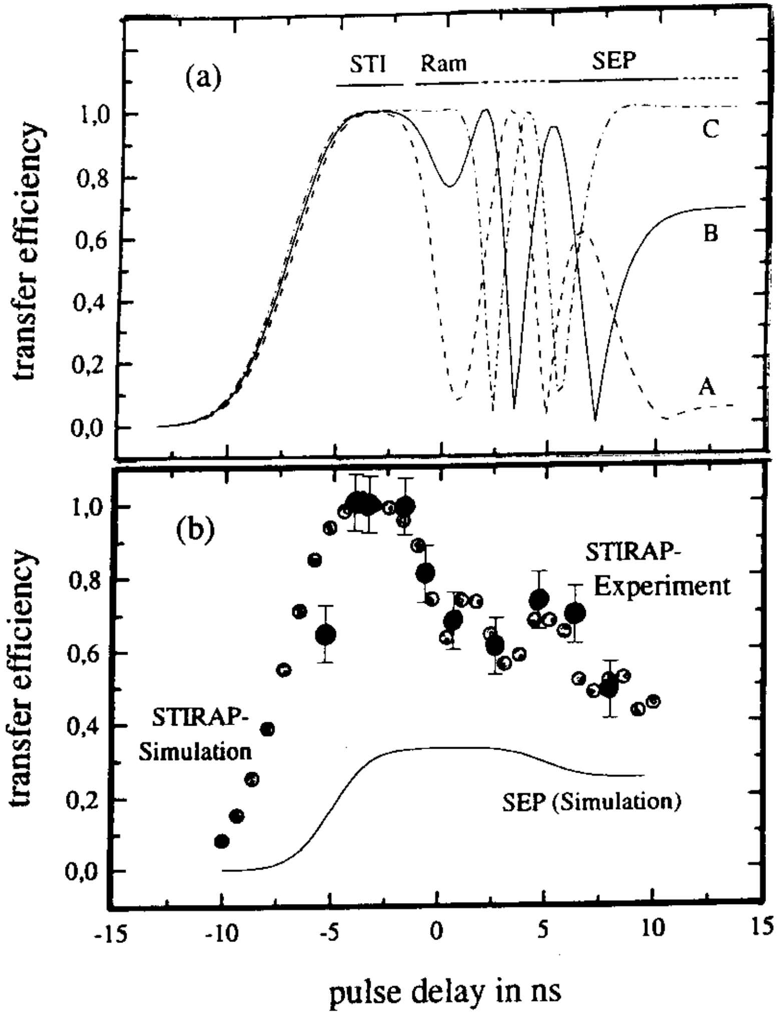


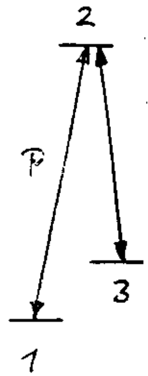


- Variability
- Effiziency
- Selectivity

"STIRAP":

e.g.:



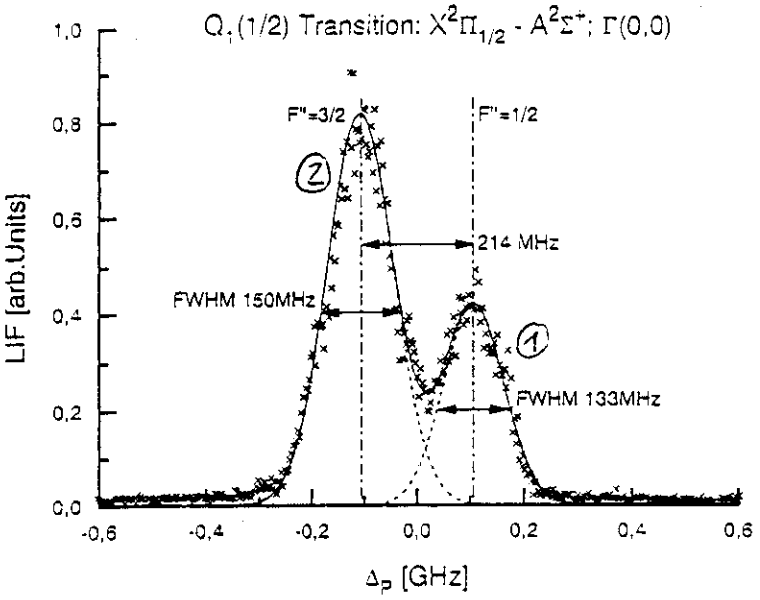


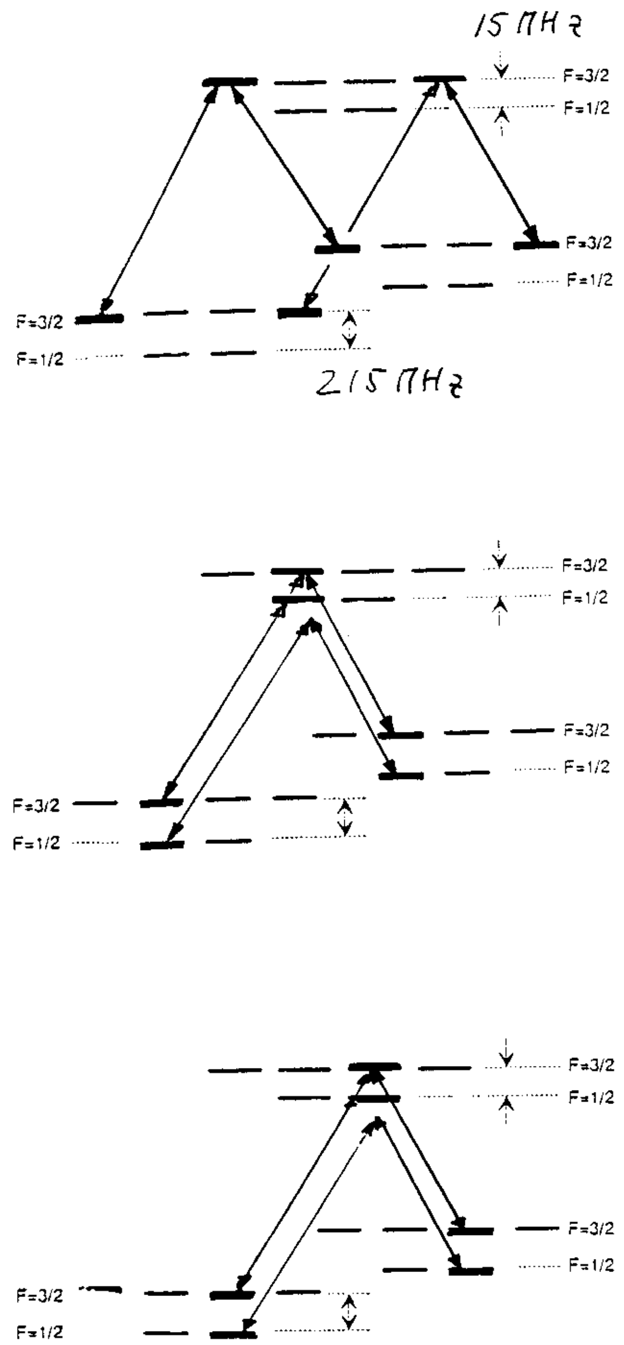
S···P S P P···S

Stimulated Raman Scattering with "adiabatic passage"

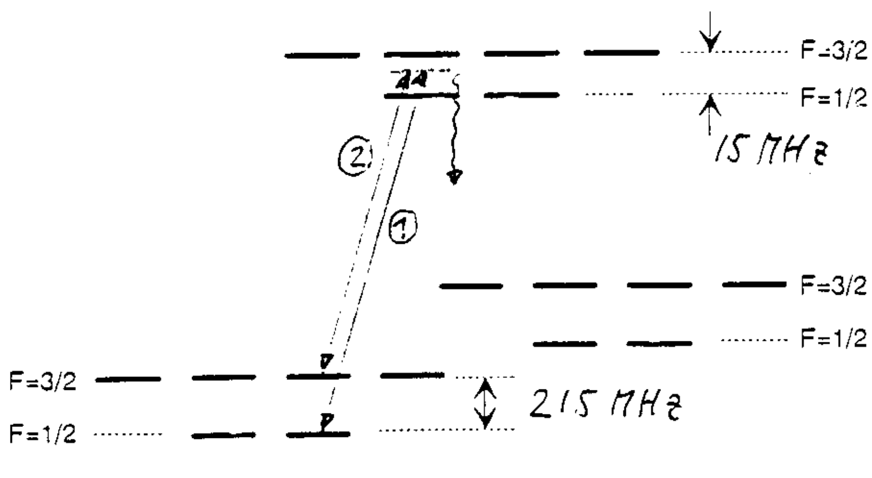
NO: m_j - degeneracy and HFS splitting „decoupling“ by choice of polarization

$\Delta m = 0$

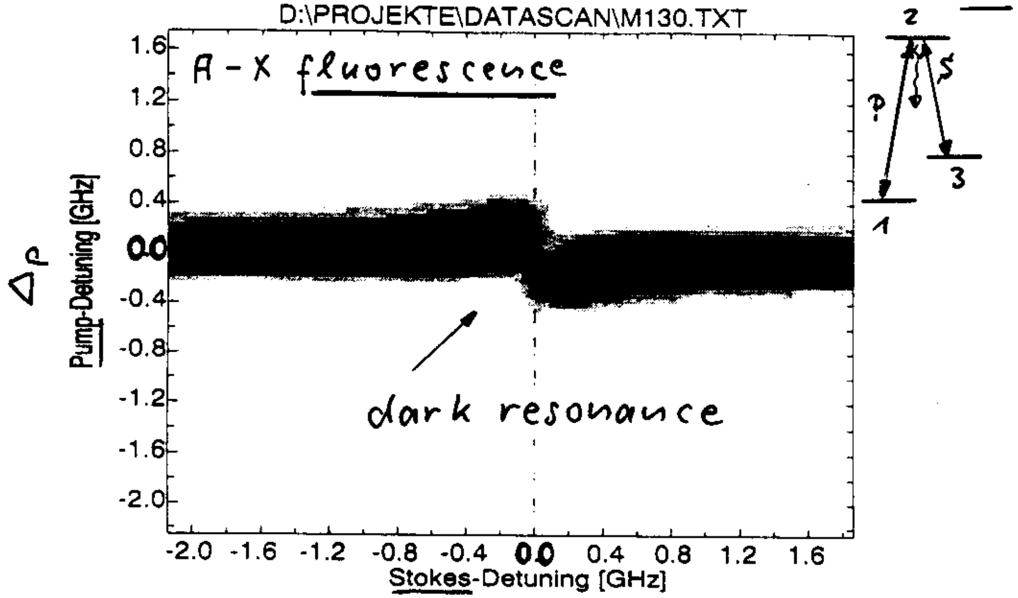


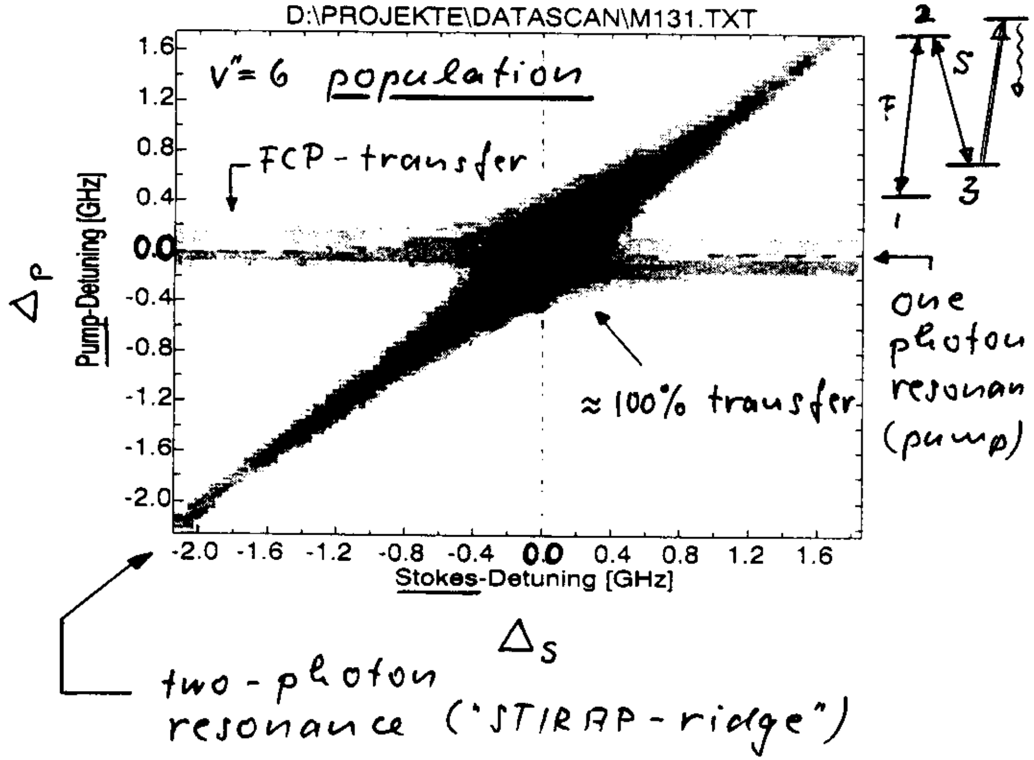


NO: resolution of HFS splitting



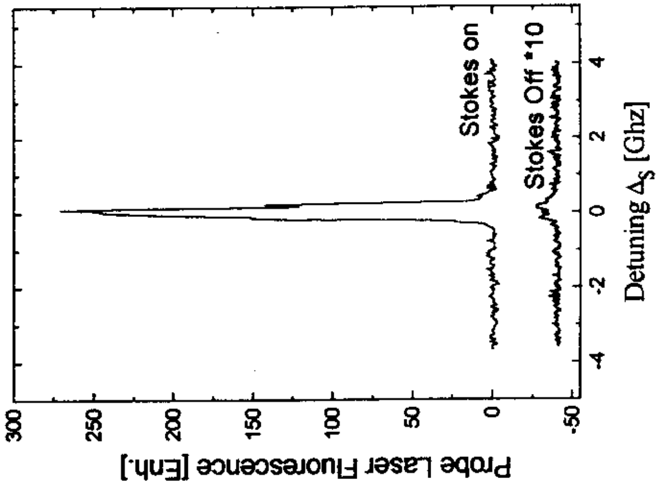
NO - experimental data

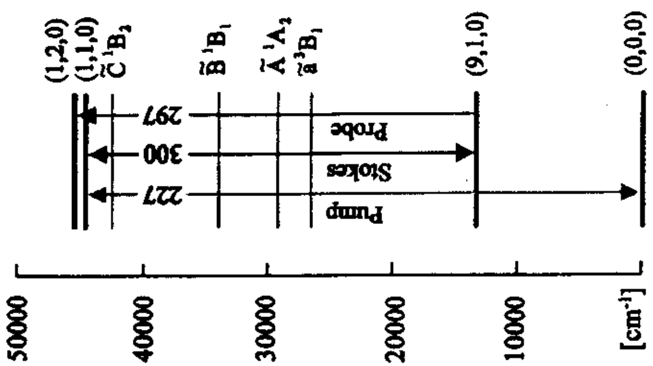




SO_2

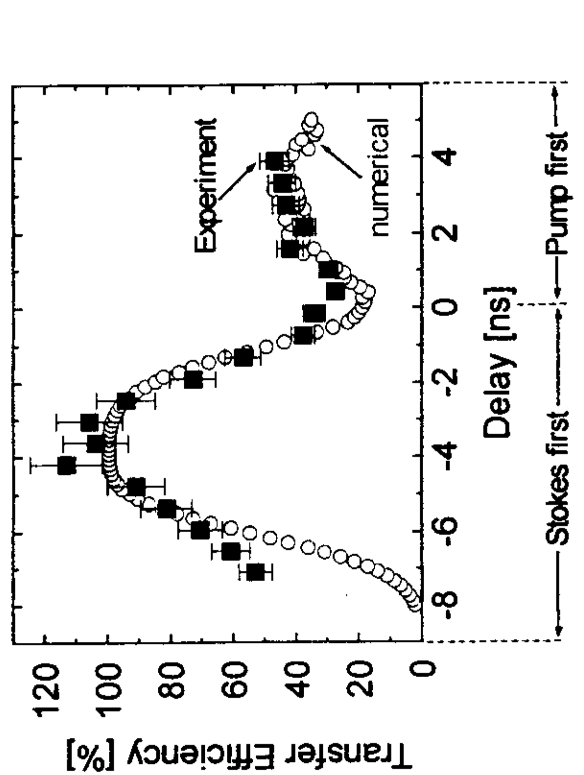
pulse energy 0.5 mJ
pulse width 3 ns
laser dia. 1 mm²



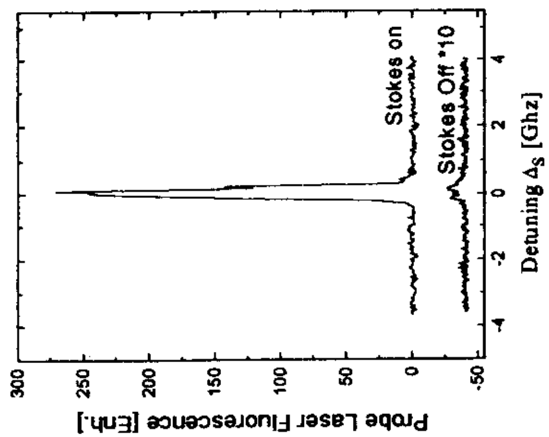


J. Chem. Phys. 104, 7068 (1996)

SO_2



J. Chem. Phys. 104, 7068 (1996)

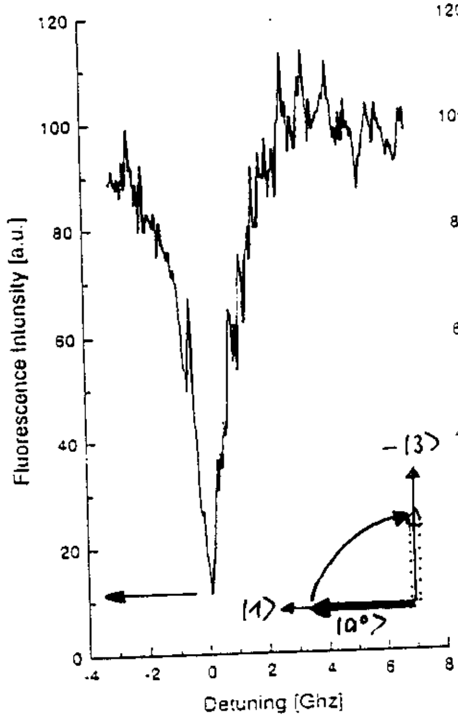


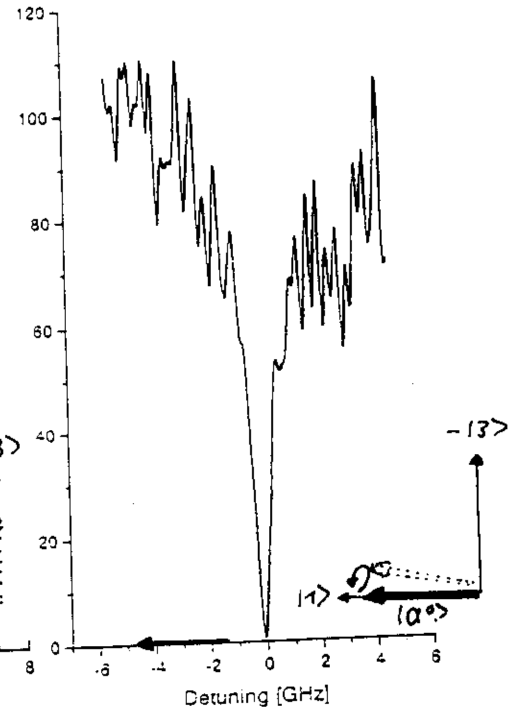
SO_2: X(0,0,0) -- 225 nm -- C(1,2,0) -- 300 nm -- X(8,4,0)

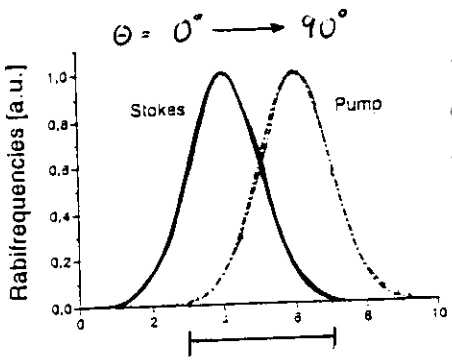
dark resonance in configuration:

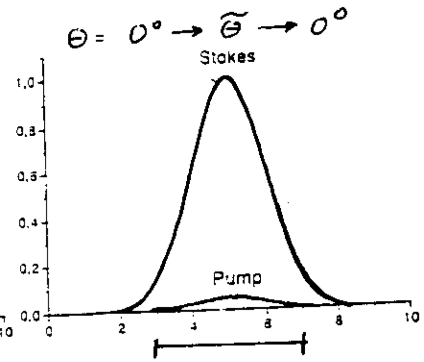
→ STIRAP (for population transfer)

→ Raman (for spectroscopy)





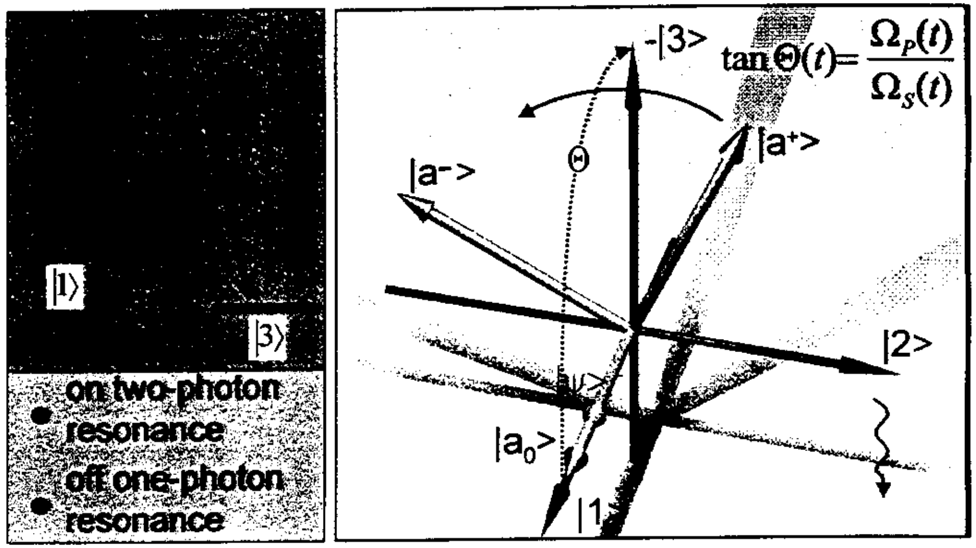


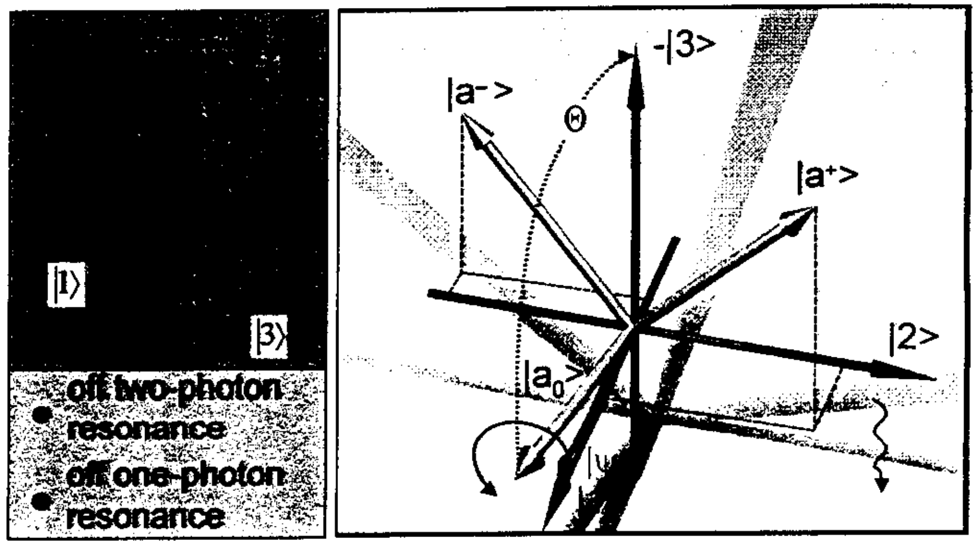


$|\langle a^{\pm}|\dot{a}^{0}\rangle| \ll |\Delta\omega|$

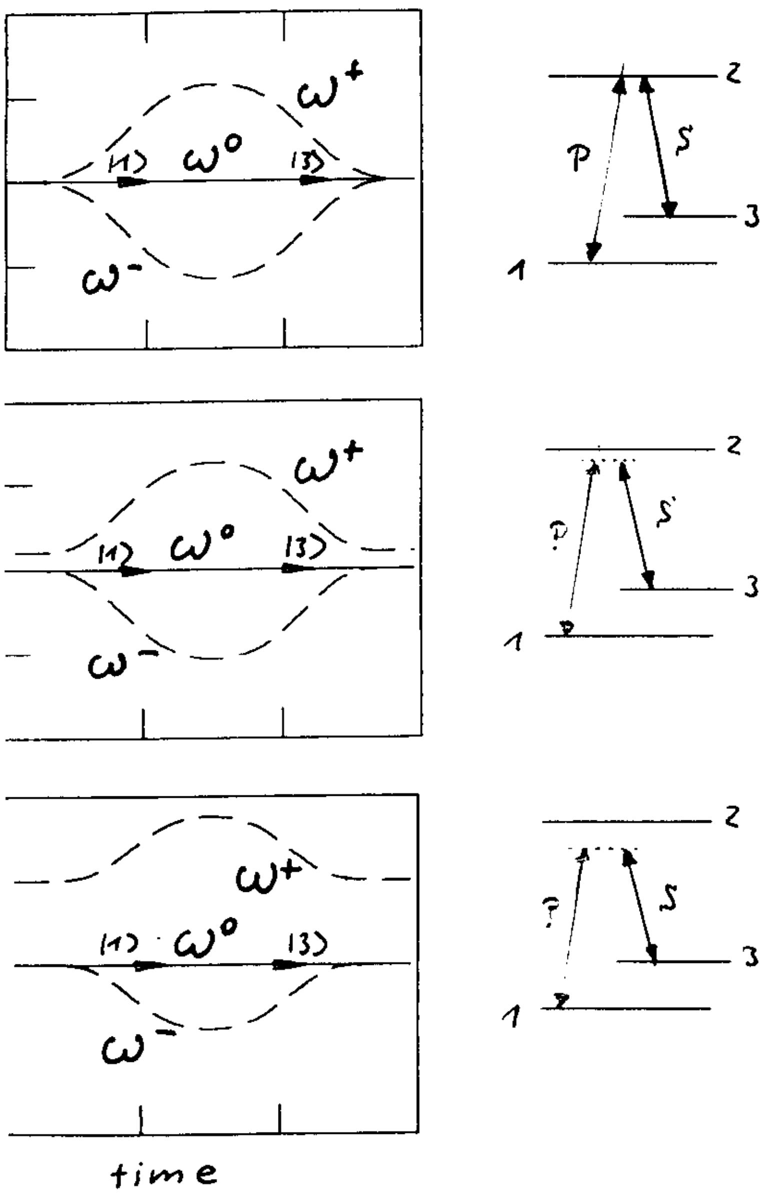
Consequences of detuning from
the two-photon resonance

The three-level dressed states (graphical)

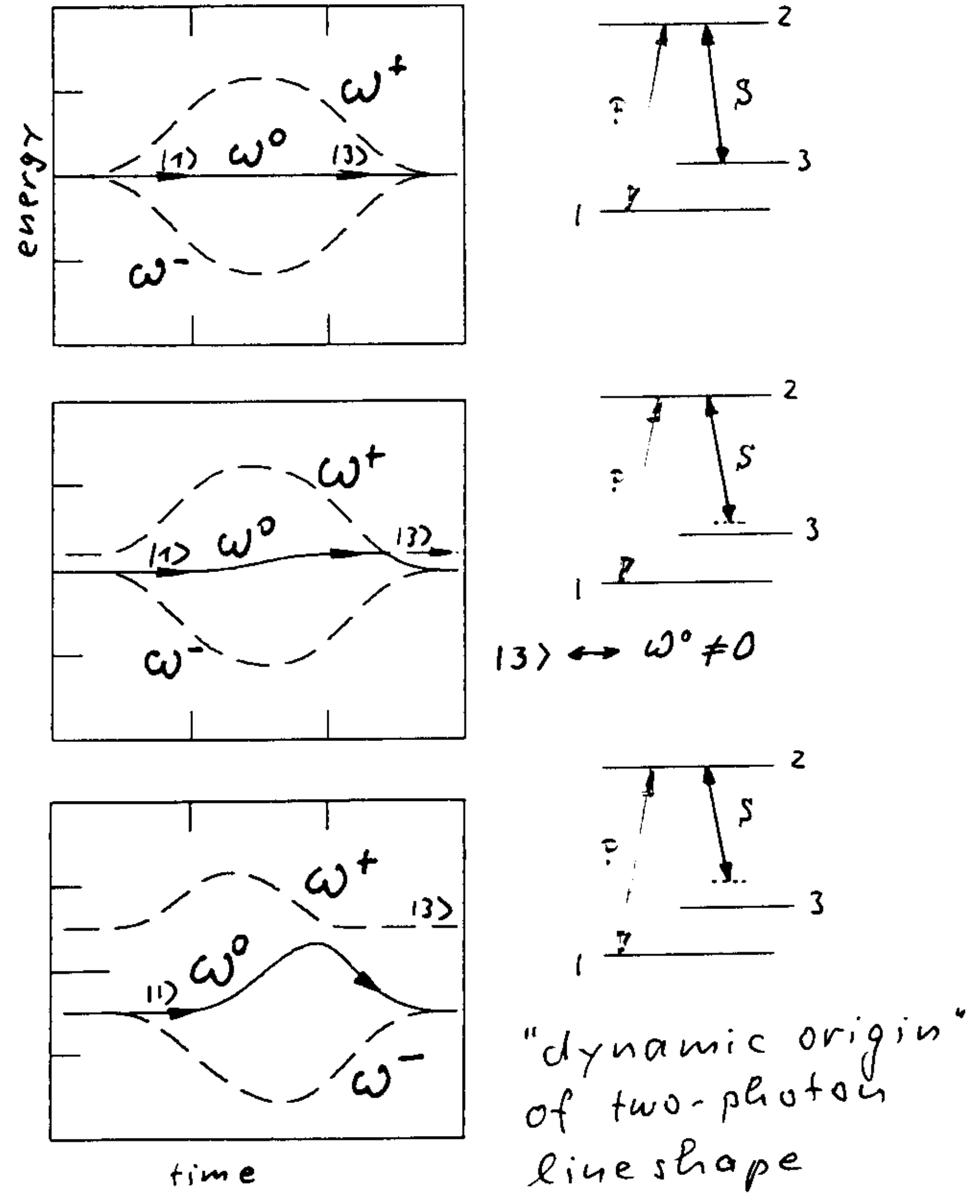




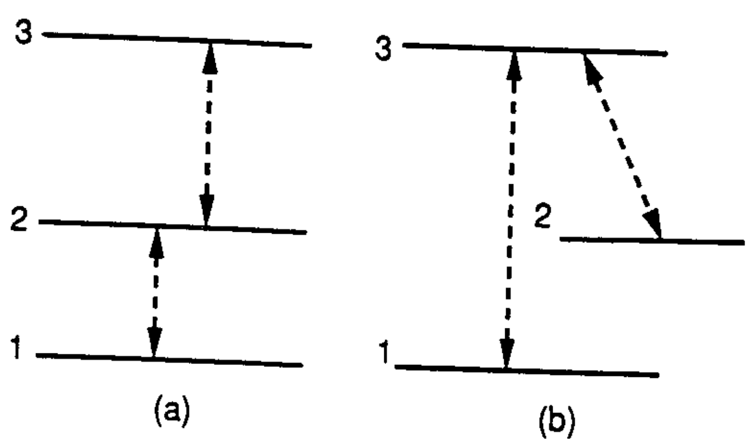
Detuning off one - photon resonance
adiabatic evolution



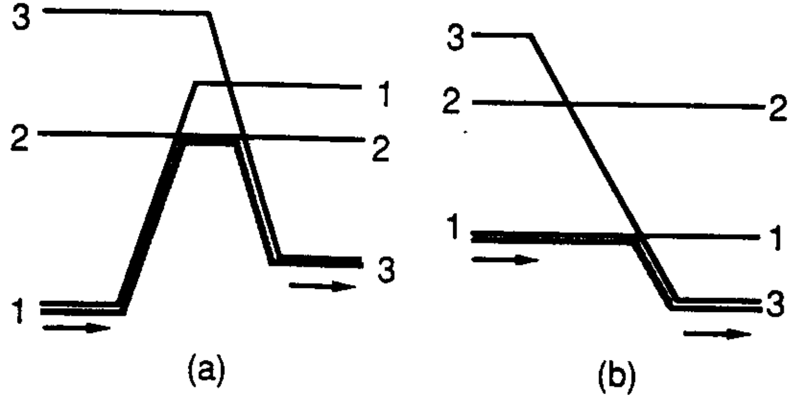
Detuning off two - photon resonance
adiabatic vs diabatic evolution



Population Transfer via Frequency chirping



(a) Ladder configuration and (b) Λ configuration.



(a) Frequency variation of RWA detunings needed to produce indirect population transfer 1-2-3. (b) Frequency variation of RWA detunings needed to produce direct population transfer 1-3. Population follows shaded path if excitation is adiabatic.

(3) Multi-level systems

(a) Ne* -Zeeman levels as a model system
- Multi-level dressed state eigenvalues
  - Blocking of adiabatic path

(b) Application to atom cptics
- Deflection by a dissipative process
- Deflection by coherent momentum transfer
  - Conditions for trapped state formation
  - Energy balance
- Atomic mirror

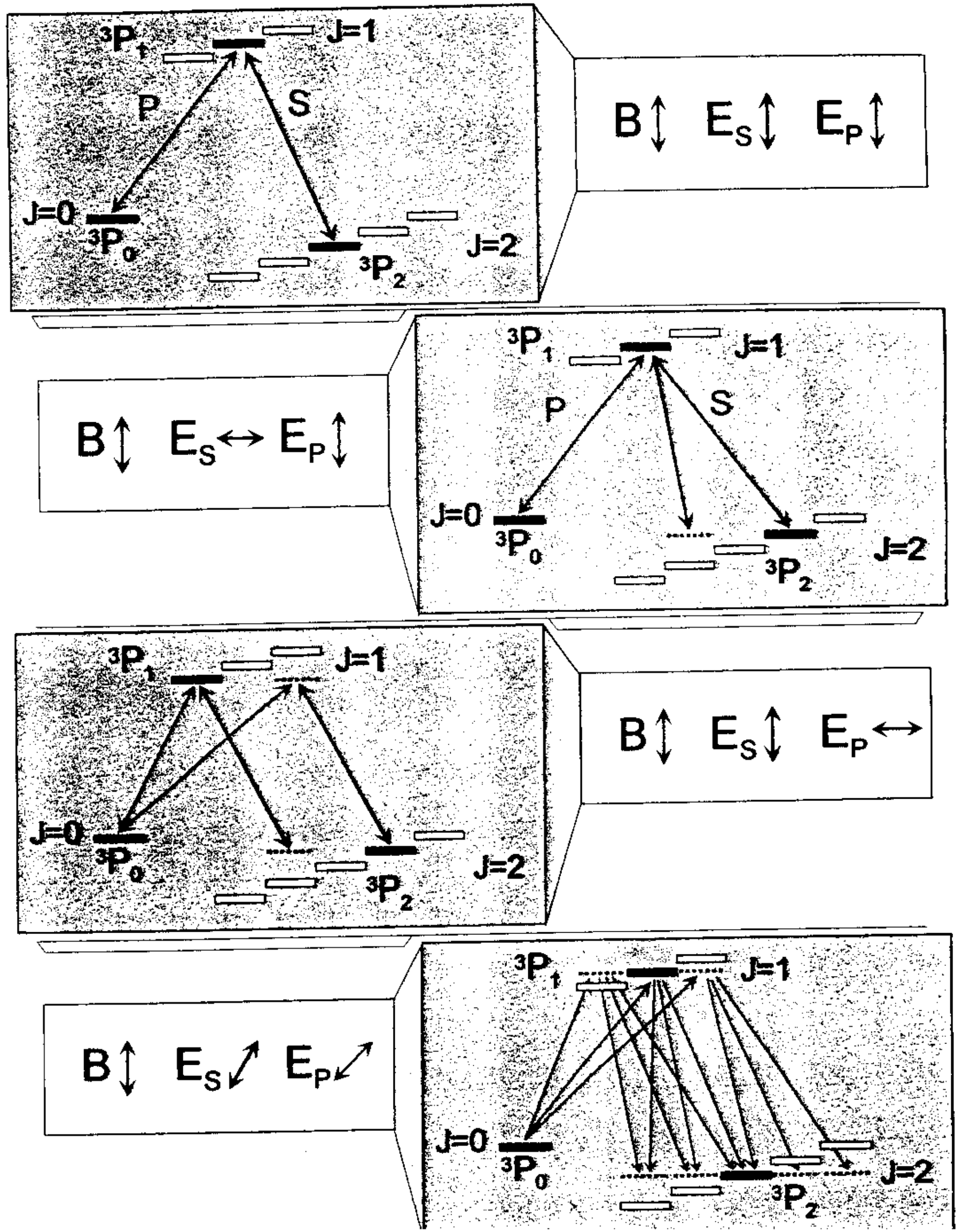
(c) Measurement of small B-fields
- by optical pumping depletion
- by Larmor velocity filter

(d) The concept of a dark state
- atomic interferometer

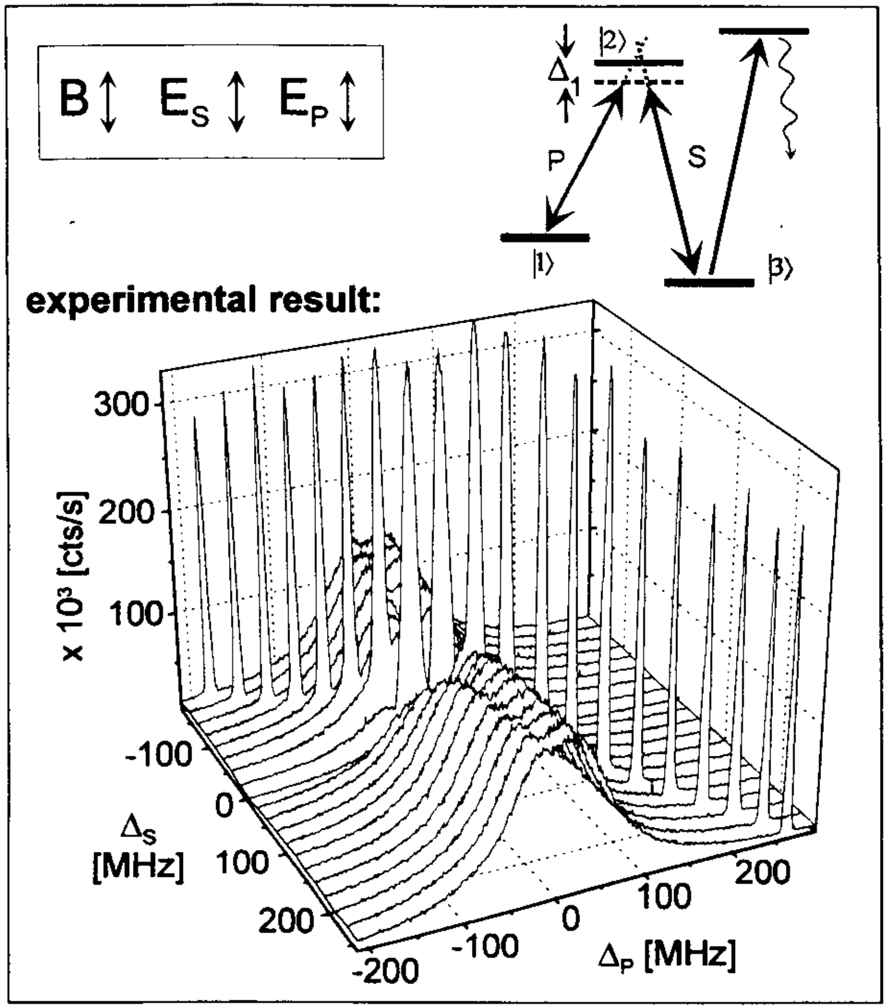
Metastable Ne* atoms
in a magnetic field
as a model system
for the study of
coherent population transfer
in multi-level systems

Control of the linkage pattern by polarization

Ne*: ($^3P_0 \Longleftrightarrow {}^3P_1 \Longleftrightarrow {}^3P_2$)



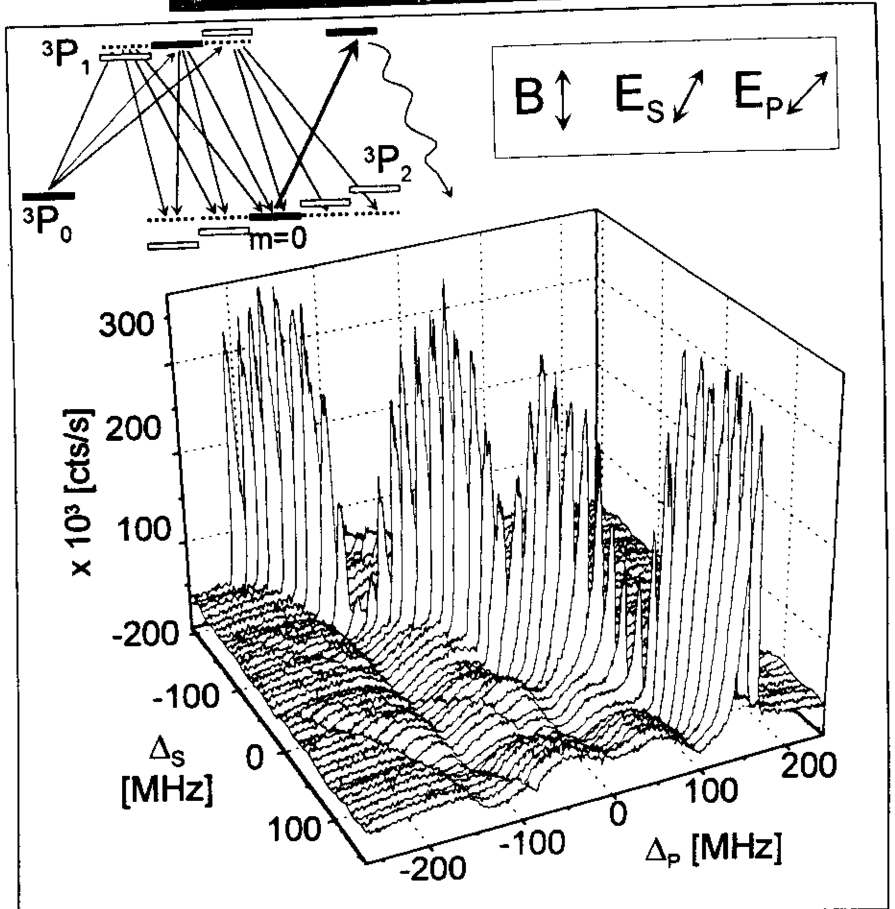
STIRAP: 3 - states coupled



MAIN CHARACTERISTICS:

- three dressed state eigenvalues
- one zero-energy eigenvalue throughout
  ⟶ trapped state exsists
- adiabatic transfer successfull also for $\Delta_1=0$

STIRAP: 9 - states coupled



MAIN CHARACTERISTICS:

- nine dressed state eigenvalues
- energy levels may cross
- avoided crossings may block adiabatic transfer path
- trapped state may not exsist throughout the interaction
  (⟶ some radiative losses)

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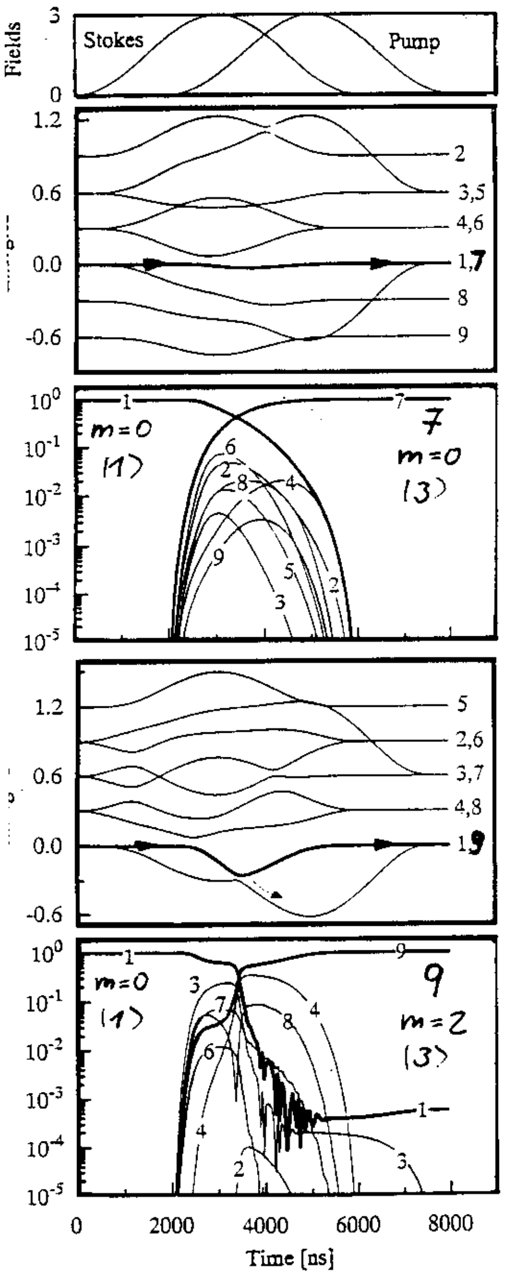
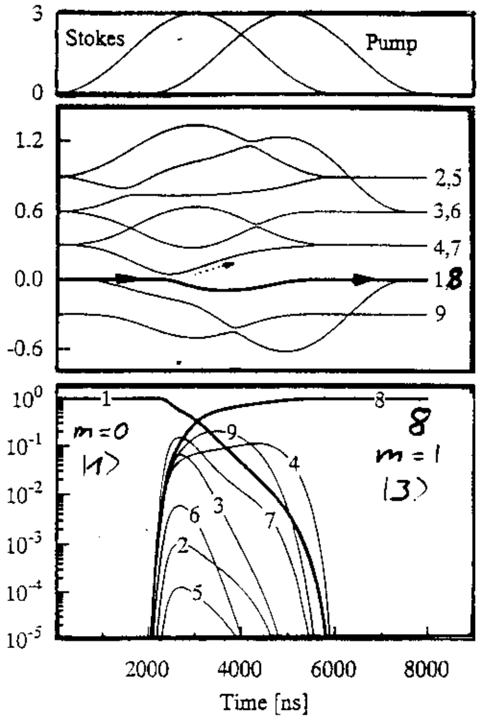
$j=0 \leftrightarrow j=1 \leftrightarrow j=2$

the general case: dressed state spaghetti

B ⟷ P ⤢ S ⤡ 9-level-system





$|1\rangle \hat{=} {}^3P_0 \quad |3\rangle \hat{=} {}^3P_2$

- relevant eigenvalue not zero throughout

- crossings
- avoided crossings

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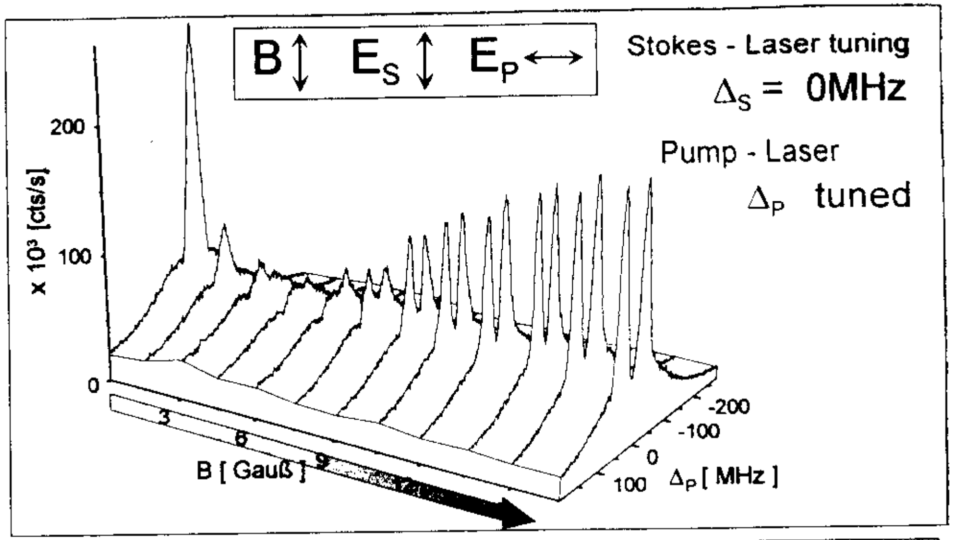
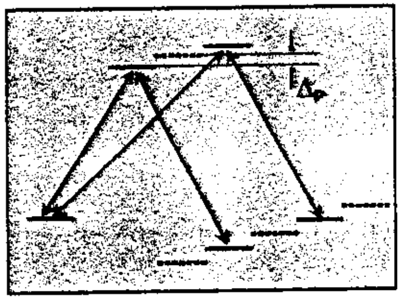
566 (1995)
583 (1995)

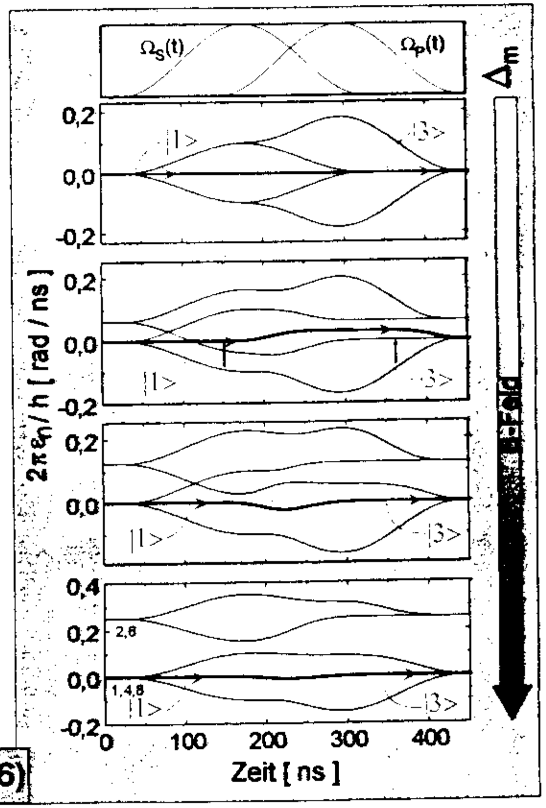




Demonstration of the Connectivity Problem

Variation of Transfer in Ne* with B-field

five - level situation



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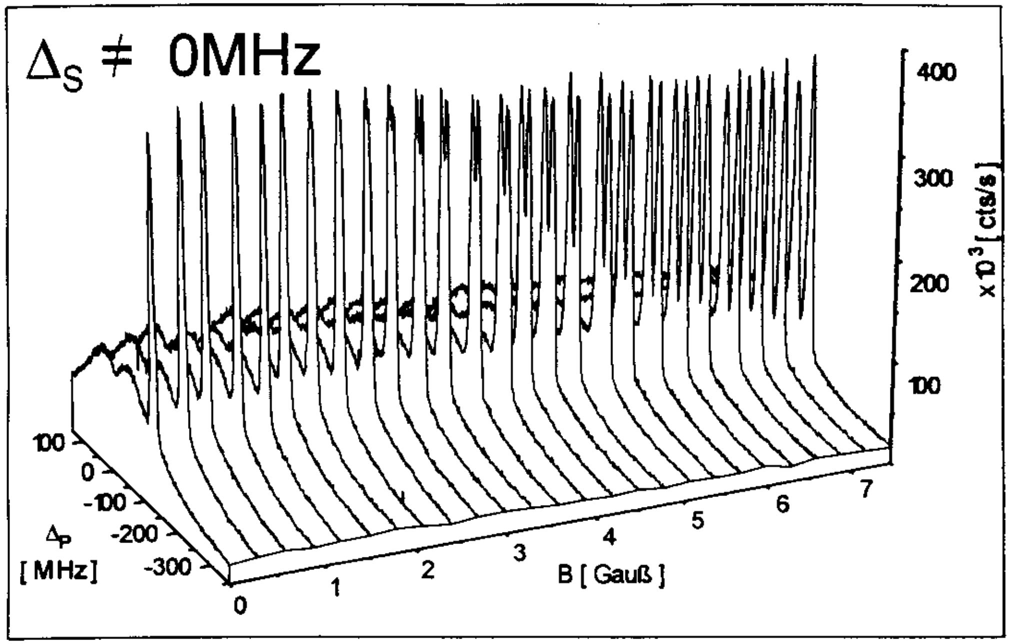
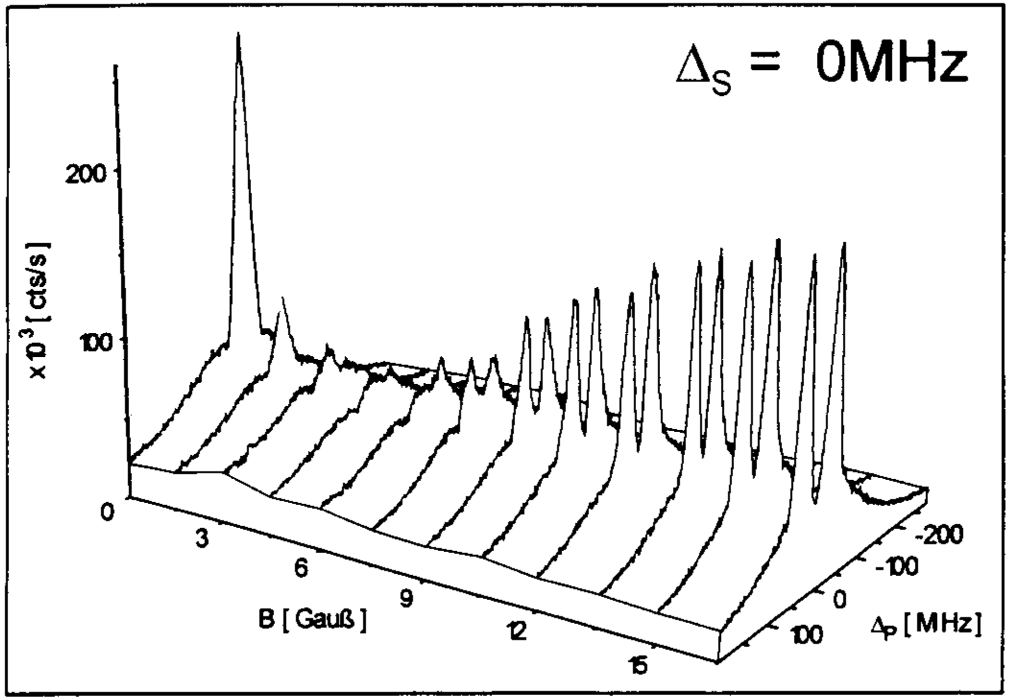
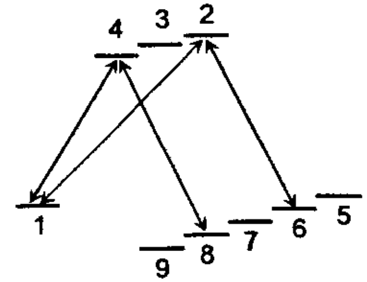
STIRAP in a 5-level-system

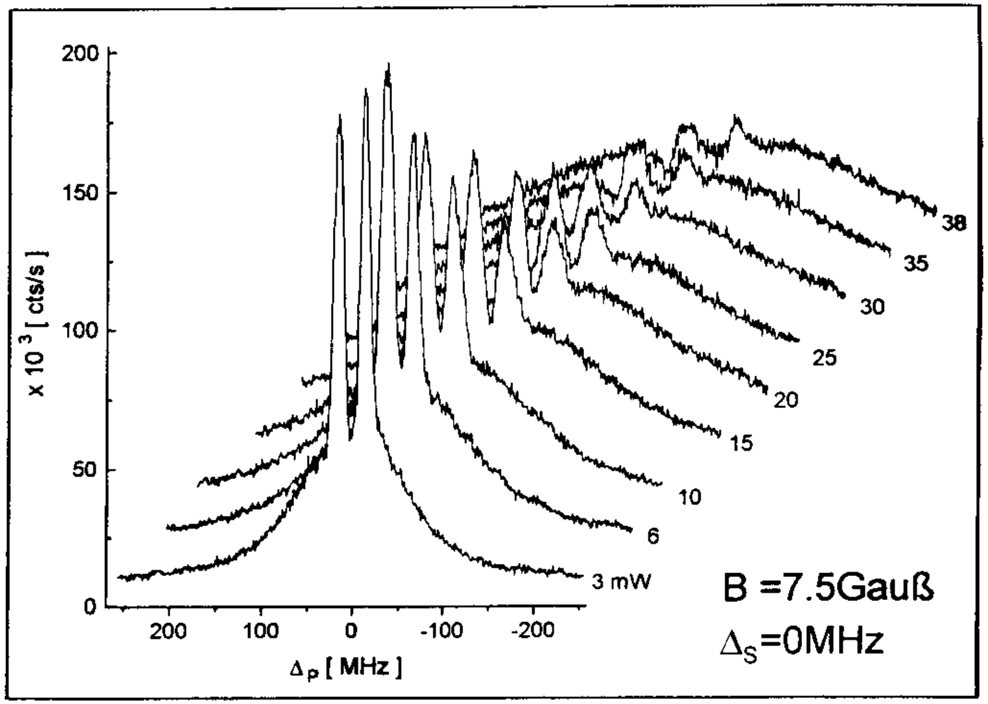




Dependence on the Rabi-frequency

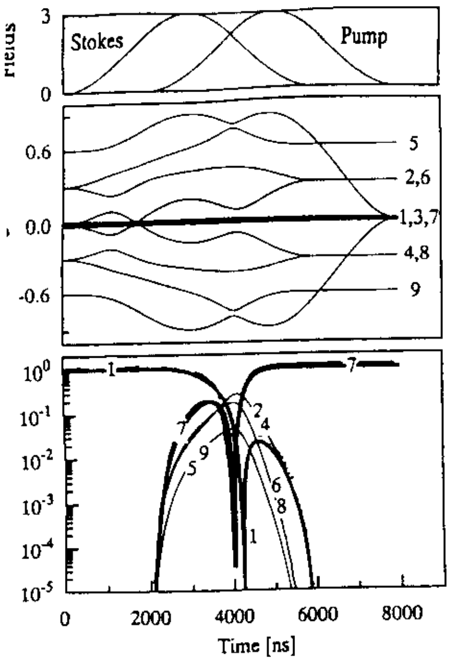
Example: 5-level-system
(two intermediate
states)

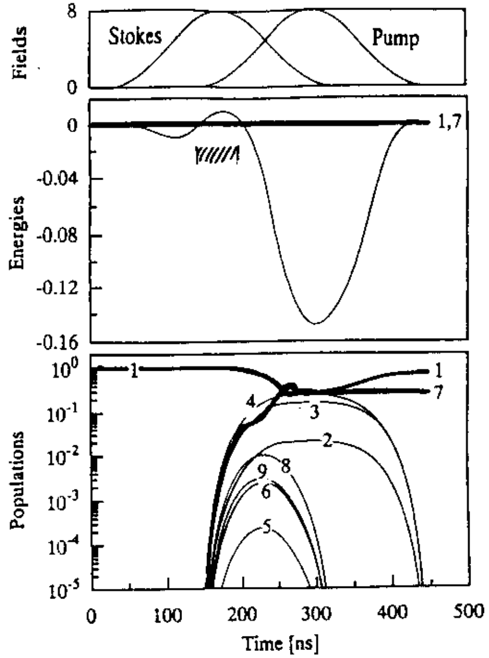


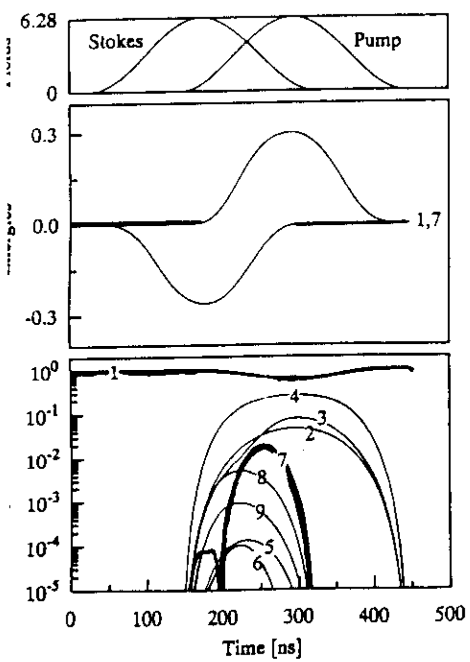


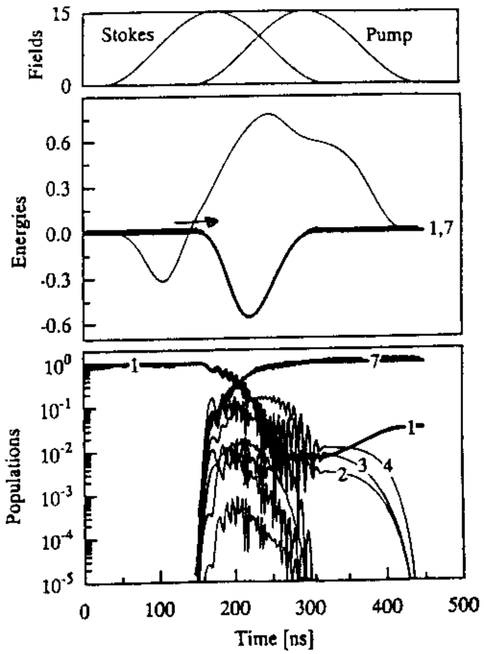
- In contrast to the 3-level-system:

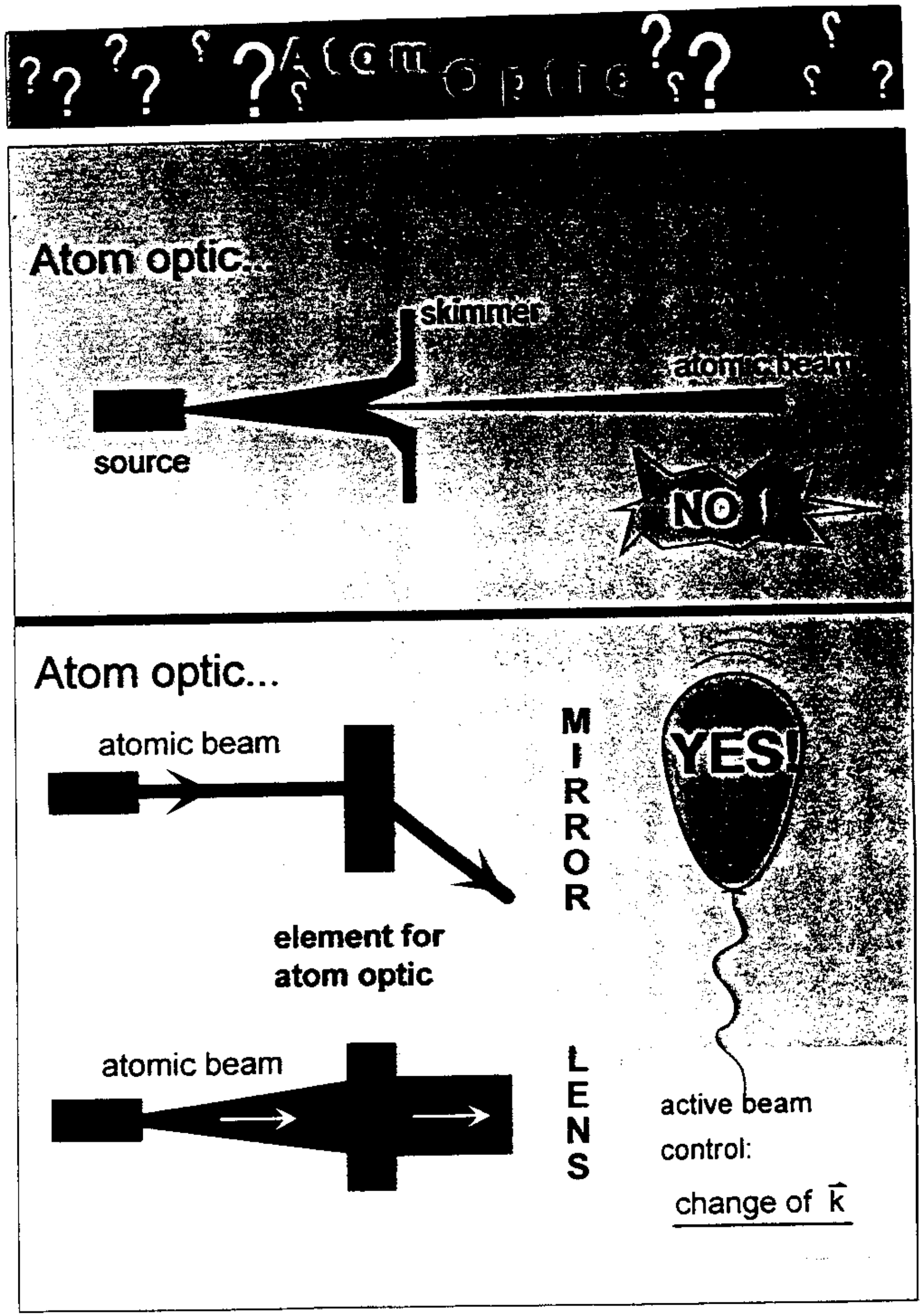
  Breakdown of transfer-efficiency with increasing Rabi-frequency


Stokes
Pump
5
2,6
1,3,7
4,8
9
Time [ns]


Fields
Stokes
Pump
Energies
1,7
Populations
Time [ns]

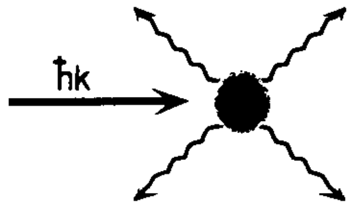

Stokes
Pump
1,7
Time [ns]

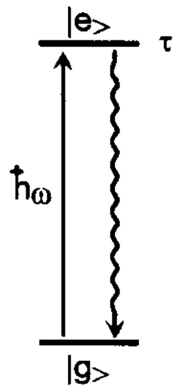

Fields
Stokes
Pump
Energies
1,7
Populations
Time [ns]


Atom optic...
skimmer
atomic beam
source
NO !
Atom optic...
atomic beam
MIRROR
element for
atom optic
atomic beam
LENS
YES!
active beam
control:
change of $\vec{k}$

The simplest element for atom optic: The mirror

Force by cycled absorption - spontaneous emission:





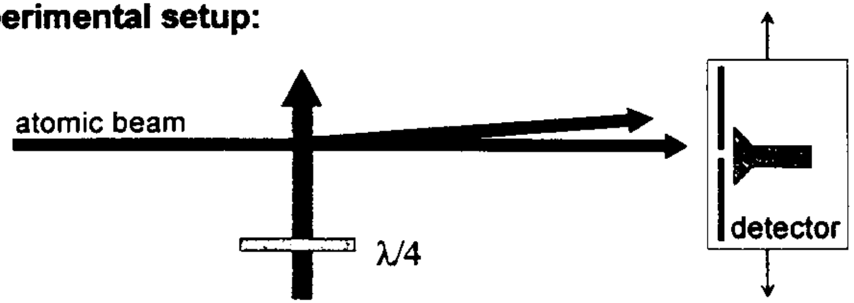
Force in stationary state:

$$F = \frac{\hbar k}{2\tau} \cdot \frac{2S_0}{1+2S_0+\Delta^2}$$

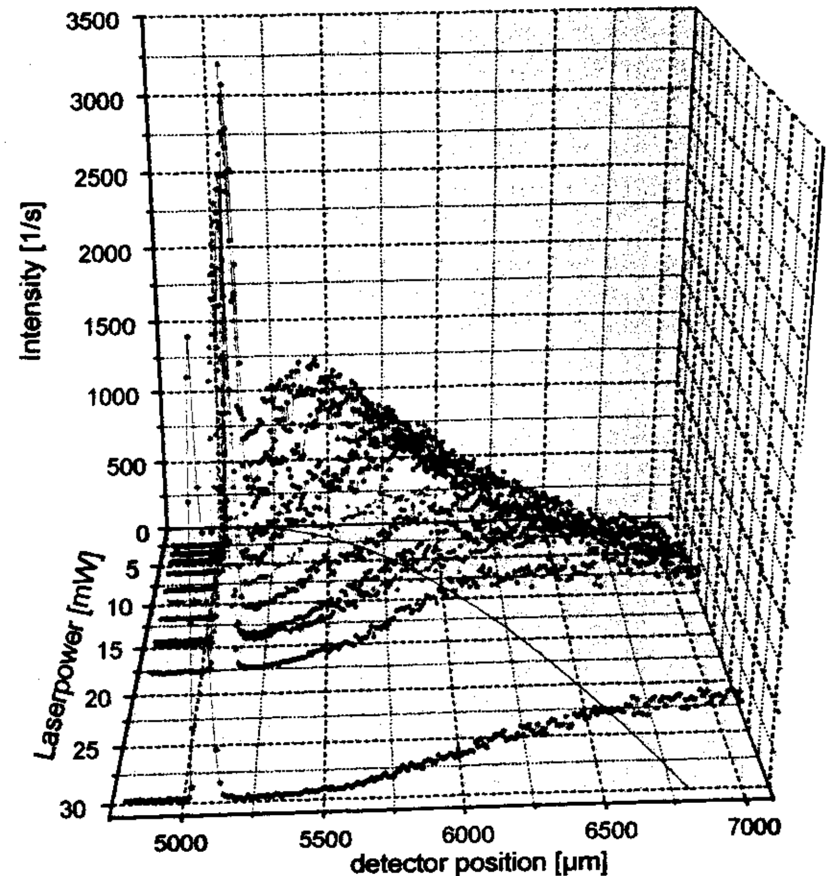
Δ : detuning
S_0 : saturation parameter

- Force saturates with increasing $S_0 = \frac{I}{I_0}$
- Acceleration for Ne*: approx. **80000g** !

Experimental setup:



experimental . . .



Disadvantages of this method:

spontaneous emission

↓ heating of transversal motion

↓ loss of coherence

So ⟶ STIRAP

Coherent Population Transfer in Multilevel - Systems

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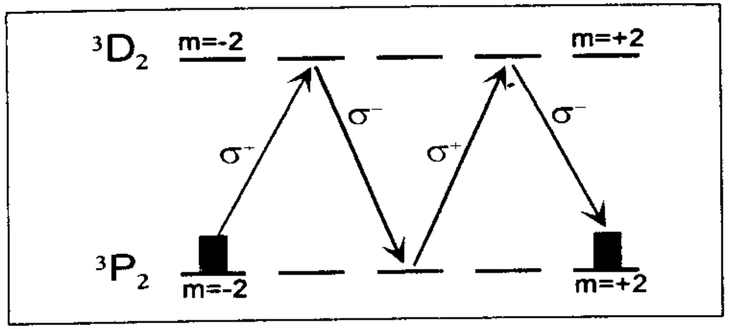
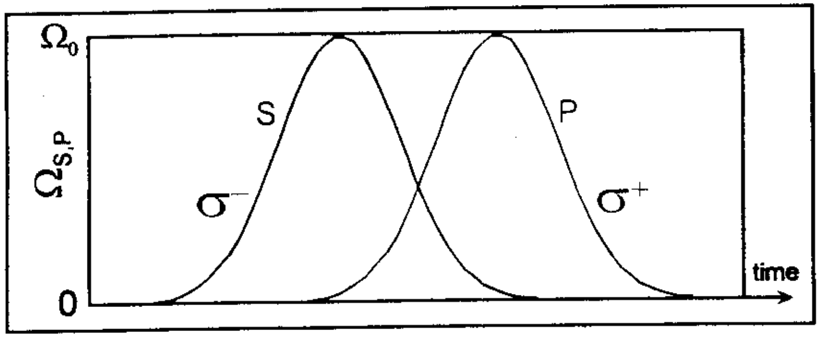
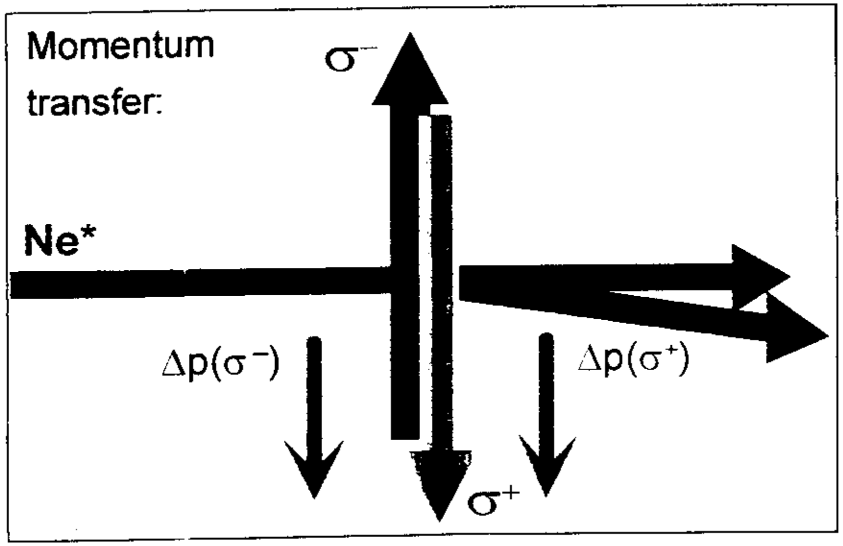
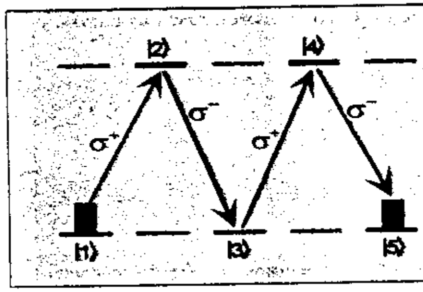
Application to Atom - Optics







some aspects of multi - level - STIRAP



existence of

trapped states ?

Schrödinger-equation gives:

$$\frac{d}{dt}\vec{C}(t) = -\frac{i}{\hbar}\hat{H}(t)\vec{C}(t)$$

look for solutions

$$\frac{d}{dt}\vec{C}(t) = 0$$

$$\boxed{\det \hat{H}(t) = 0} \qquad -\infty < t < +\infty$$

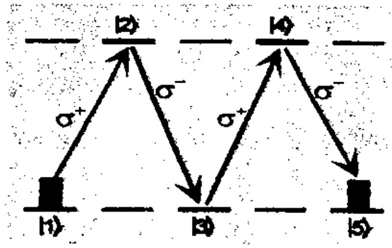
RWA - Hamilton matrix:

$$\hat{H}(t) = \frac{\hbar}{2}\cdot\begin{bmatrix} 0 & \Omega_{12} & 0 & 0 & 0 \\ \Omega_{21} & 2\Delta_{12} & \Omega_{23} & 0 & 0 \\ 0 & \Omega_{32} & 2\Delta_{13} & \Omega_{34} & 0 \\ 0 & 0 & \Omega_{43} & 2\Delta_{14} & \Omega_{45} \\ 0 & 0 & 0 & \Omega_{54} & 2\Delta_{15} \end{bmatrix}$$

evaluate...

$$\boxed{\det \hat{H}(t) = 2\Delta_{15}\det \hat{H}_4(t) - \Omega_{45}^2 \det \hat{H}_3(t)}$$

more . . . multi-level-STIRAP



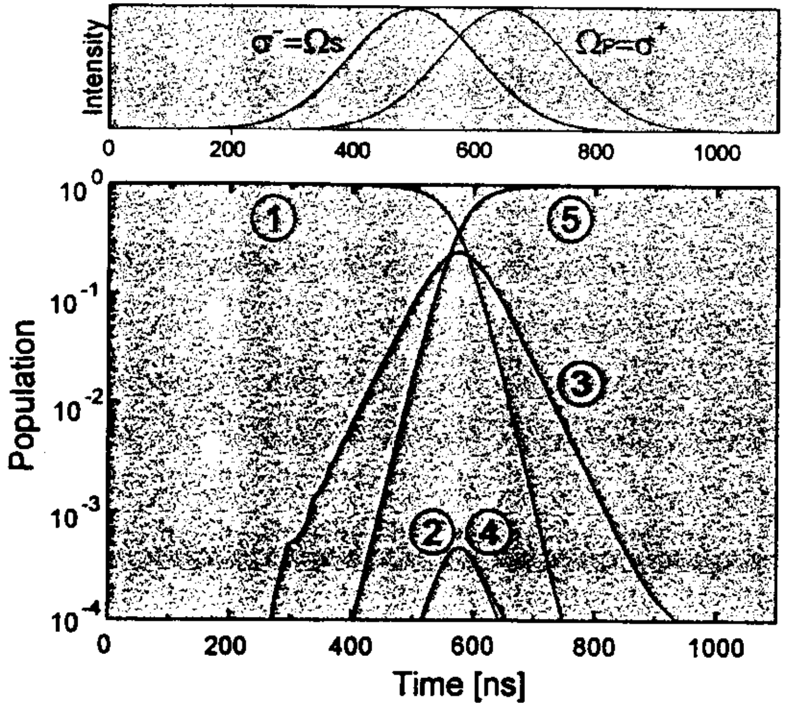
trapped state for

$0 = \Delta_{13} = \Delta_{15}$

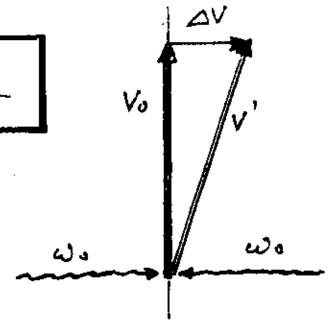
$|2\rangle$ $|4\rangle$ are not populated

Simulation:

$\tau = 19{,}6\,\text{ns}$



Energy balance



A) $E' = \frac{1}{2} m v'^2 \qquad v' = \sqrt{v_0^2 + \Delta v^2}$

$E' = \frac{1}{2} m v_0^2 + \frac{1}{2} m \Delta v^2$

- $\Delta E_R = E' - E_0 = \frac{1}{2} m \Delta v^2 = \frac{\Delta p^2}{2m} = \frac{(2\hbar k)^2}{2m}$ recoil energy

$\Delta E_R (600\,\text{nm}, Ne^*) = h\,\Delta\nu_{recoil} \qquad \Delta\nu_{recoil} \approx 75\,\text{kHz}$

$\Delta\nu_n \approx 15\,\text{MHz} \approx 200\,\Delta\nu_{recoil}$

B)

$\omega' = \omega_0 + k_0 v_0 \qquad \omega'' = \omega_0 - k_0 v_0 \qquad \underline{\Delta E_{ph} = 2 v_0 \hbar k_0}$

$\Delta p = \hbar (k' + k'') = \hbar \left(\frac{\omega_0 + v_0 k_0}{c} + \frac{\omega_0 - v_0 k_0}{c} \right)$

$= 2\hbar \frac{\omega_0}{c} = 2\hbar k_0$

$E' = \frac{1}{2} m v'^2 = \frac{1}{2} m (v_0 + \Delta v)^2 = \frac{1}{2} m v_0^2 + m v_0 \Delta v + \frac{1}{2} m \Delta v^2$

$\Delta E_R = m v_0 \Delta v + \frac{1}{2} m \Delta v^2 = v_0 \Delta p + \frac{\Delta p^2}{2m}$

- $\Delta E_R = \underline{2 v_0 \hbar k_0} + \frac{\Delta p^2}{2m}$ ← recoil energy

↳ Energy difference of Doppler-shifted photons

. . . implementation ⇐?⇒ problems

- very high collimation of the atomic beam

  passiv: 50µm - slits (or less)
  activ: transversal laser cooling (lin⊥lin)

  ⟶ collimation: better than 1 : 30000

- high resolution, low noise detector

  movable channeltron (resolution <10µm)
  Vacuum: UHV $p \leq 1 \cdot 10^{-8}$ mbar (with beam)

- creating and handling of cicularly polarized light

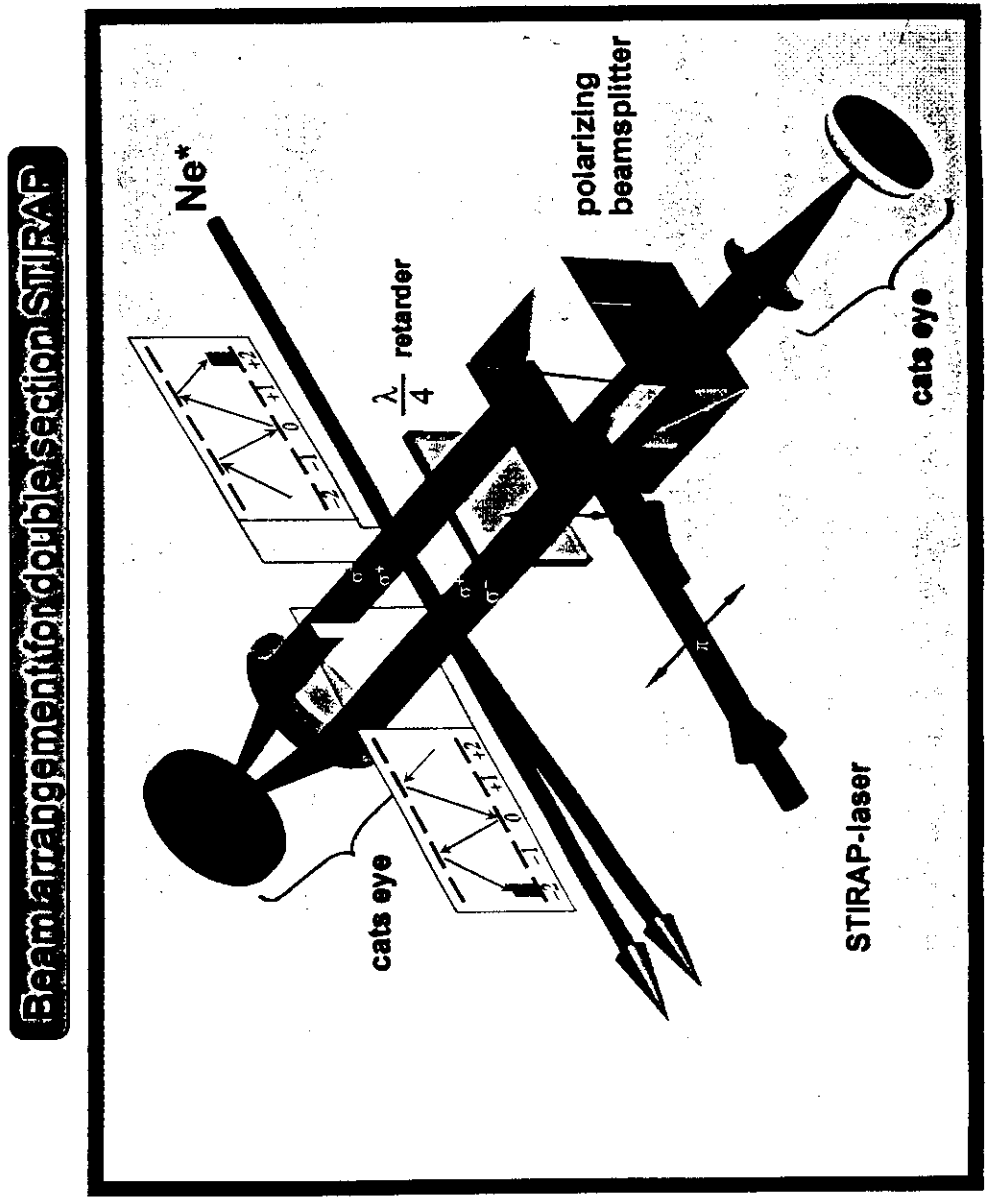
  suitable optics . . .

- region with low magnetic field $B << B_{earth}$

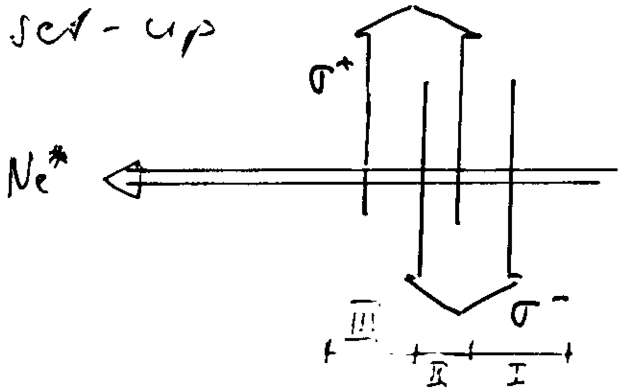
  three pairs of (Helmholtz-) coils } long term
  sensors, electronics } stability

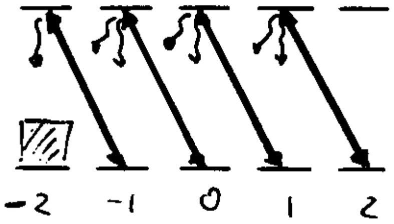
  reduction: $B \longrightarrow 0$?

  ⟶ Physics !

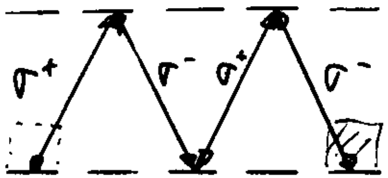


inherent preparation of m=-2 state in STIRAP set-up

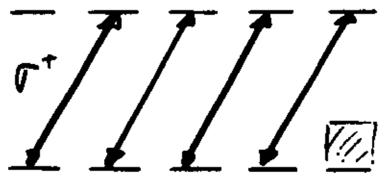




σ− zone I

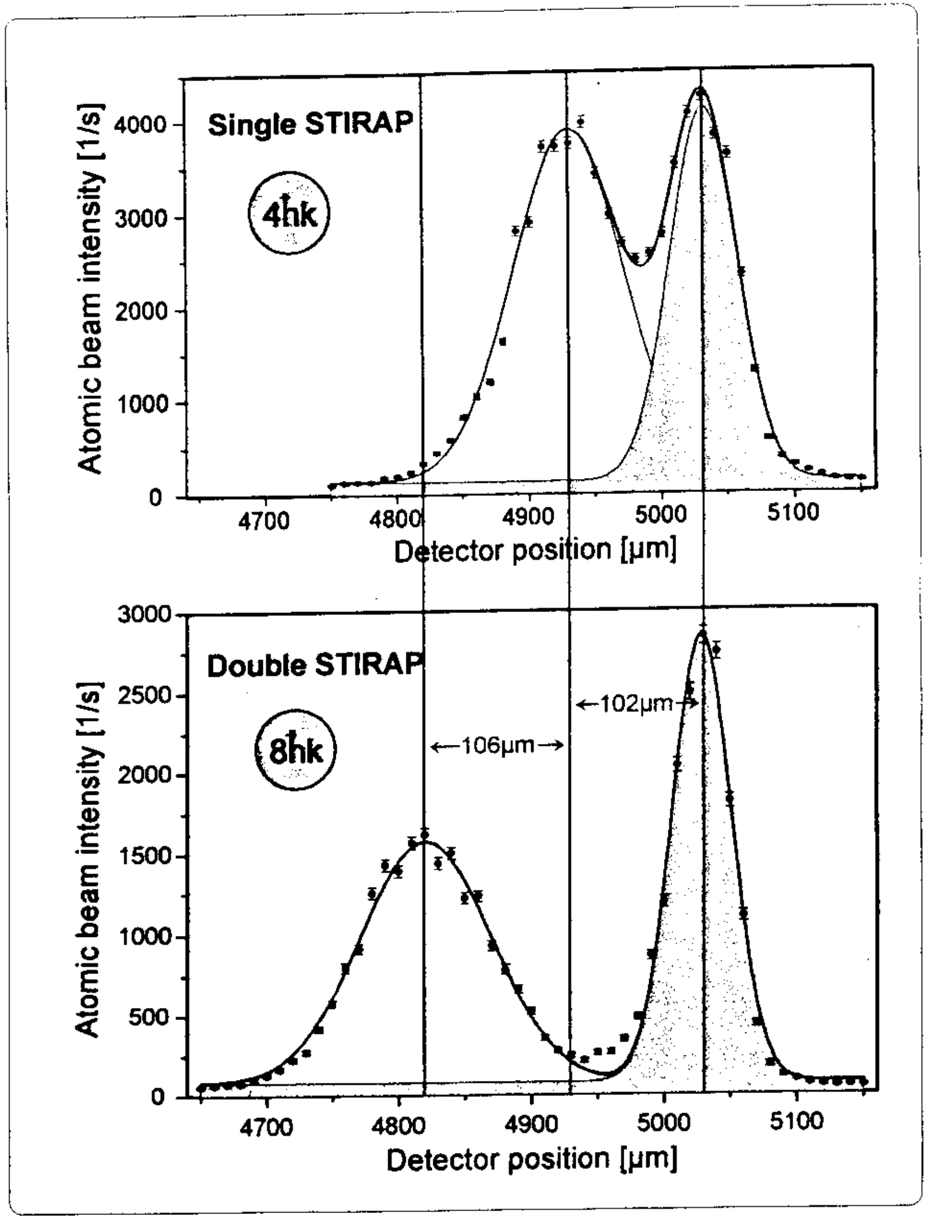


σ+/σ− zone II



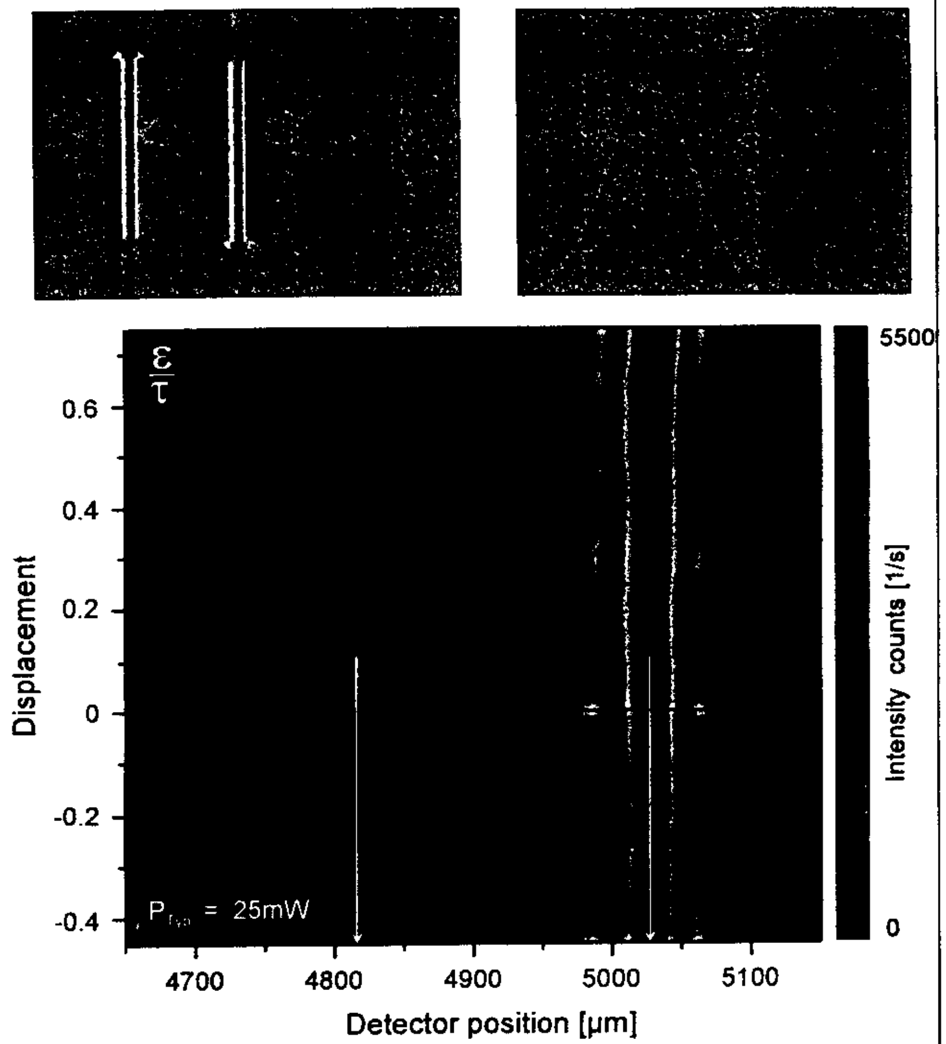
σ+ zone III

Multisection STIRAP Atomic Beam Deflection



STIRAP atomic beam mirror

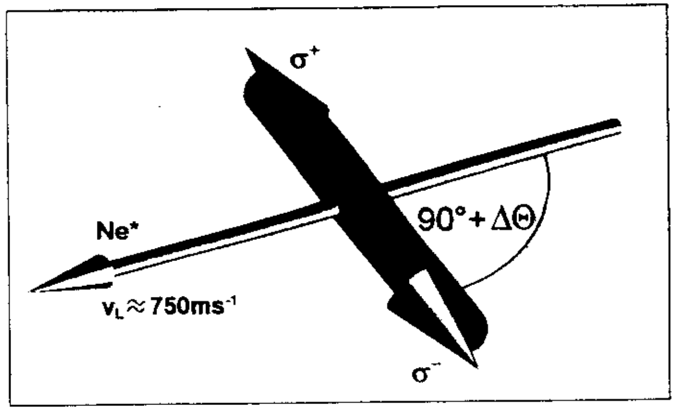
Sensitivity on displacement variation



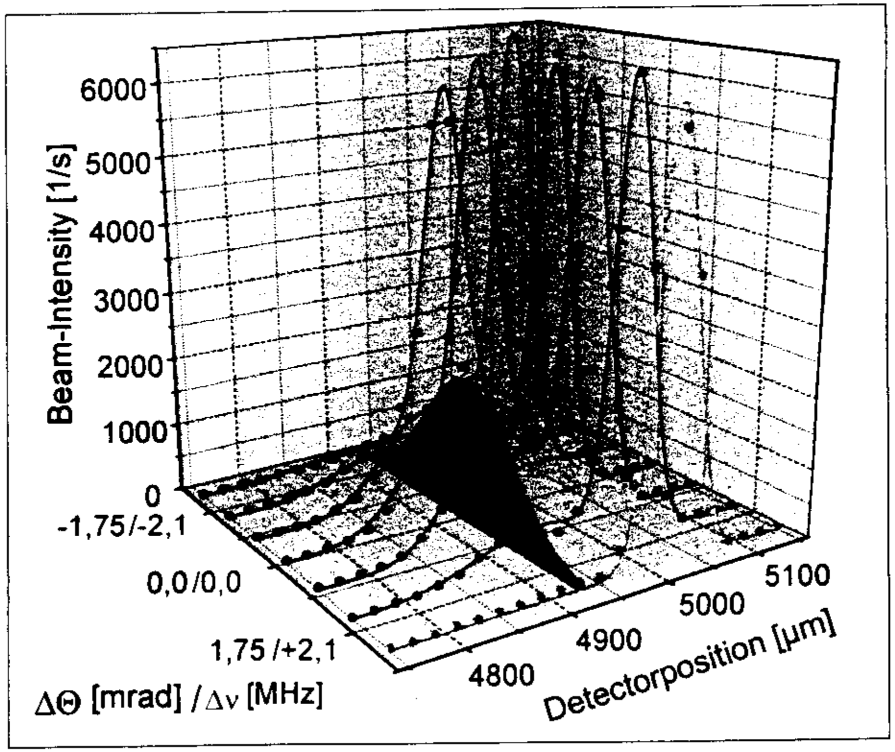
Deflection:

- State selective
- State conservative !

STIRAP atomic beam mirror and misalignment



- Single LASER-STIRAP deflection only possible, if
  Misalignment induced Dopplershift < Two photon linewidth
- Natural linewidth of $^3D_2$ - level: 8,1MHz



summary: atomic beam mirror

- large deflection angle

  → multi - zone - STIRAP

- reflectivity > 80%

  to be improved . . .

- coherence maintained

  → STIRAP

- one laser only → stability

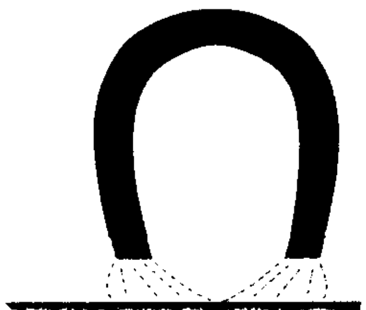
  single - laser - STIRAP

- low power, typ. 20 - 30mW

- accurate alignment required

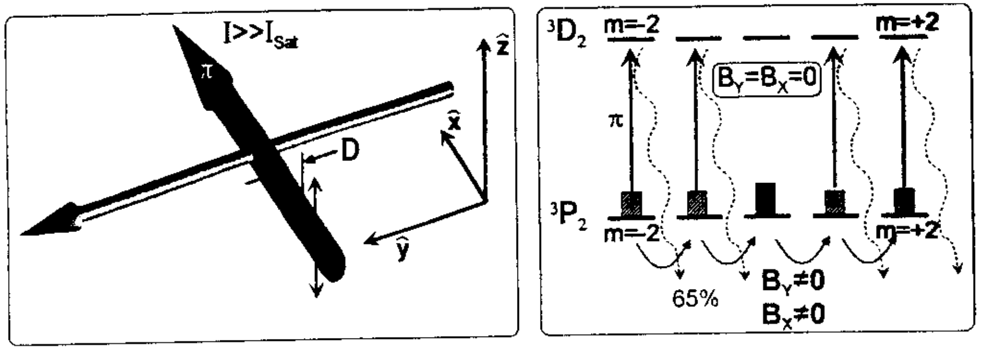
  better than 2mrad

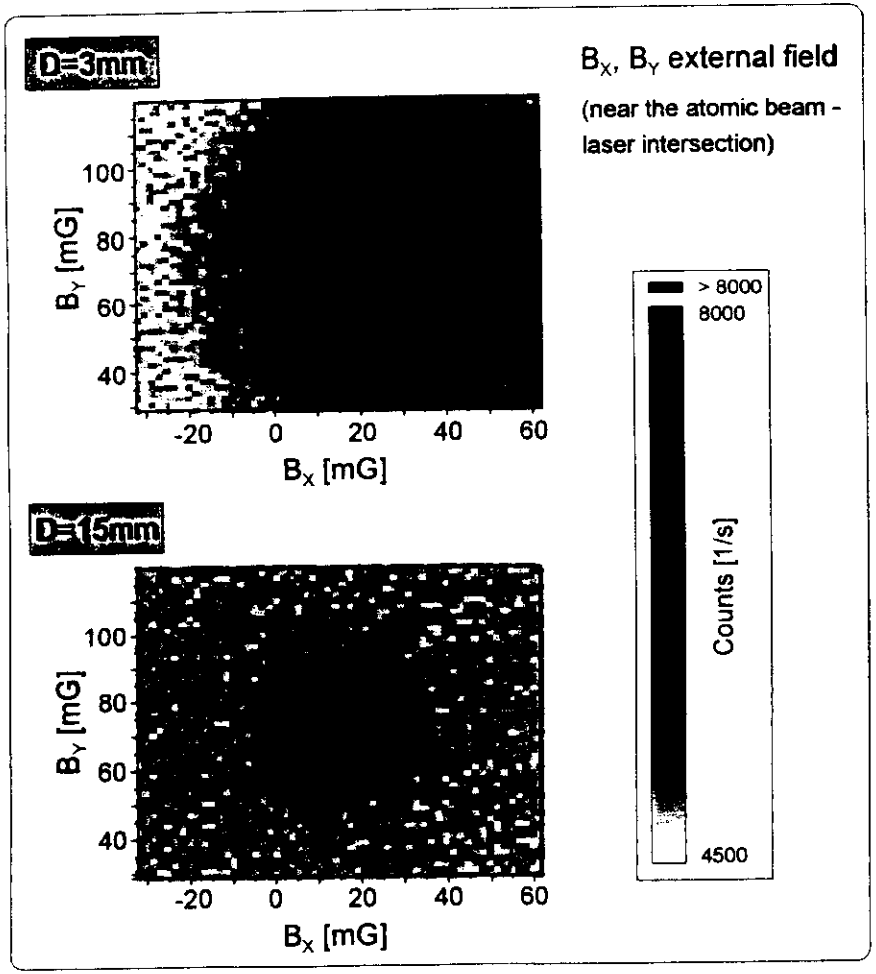
- effect of magnetic field → more physics !



Measurement of small magnetic fields

Probing Magnetic Fields through Larmor Precession

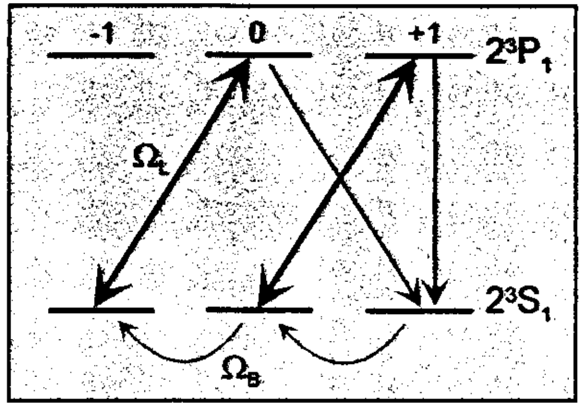


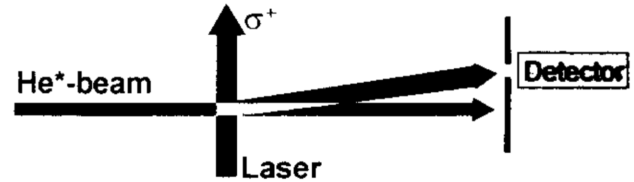


Mechanical Hanle-effect

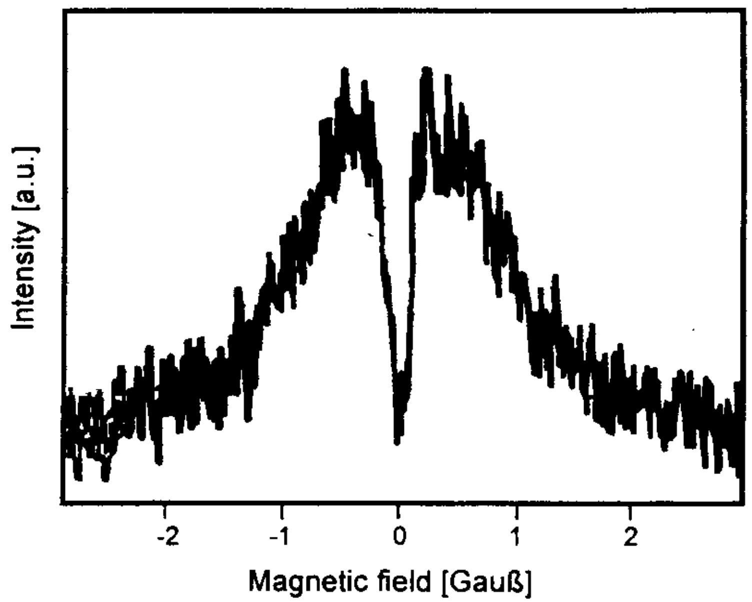
R. Kaiser et al., Z. Phys. D 18, 17 (1991)

Radiative coupling in metastable Helium:

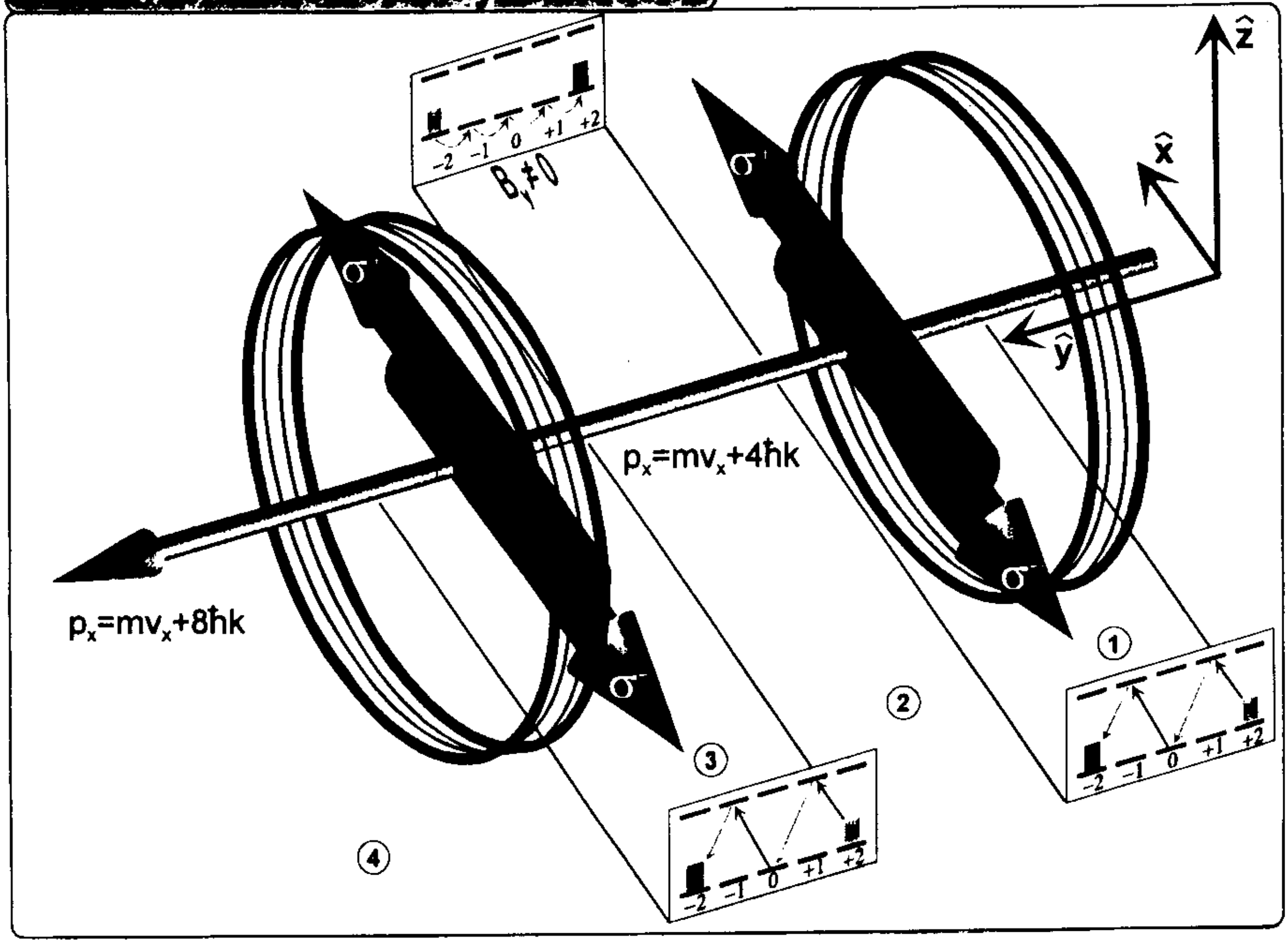




Experimental result:

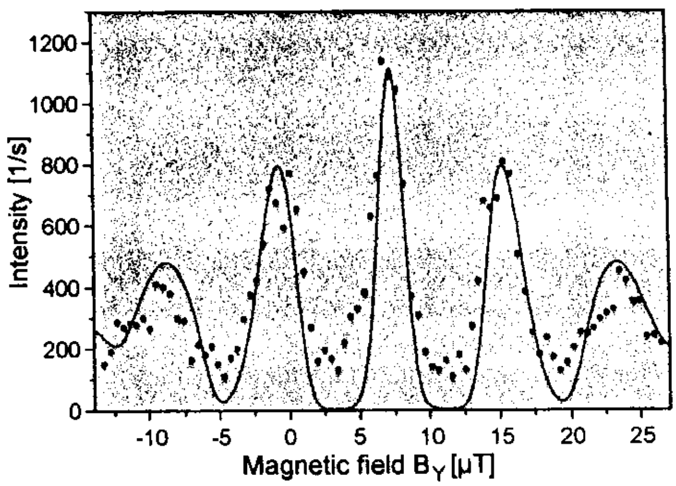


Larmor Velocity Filter



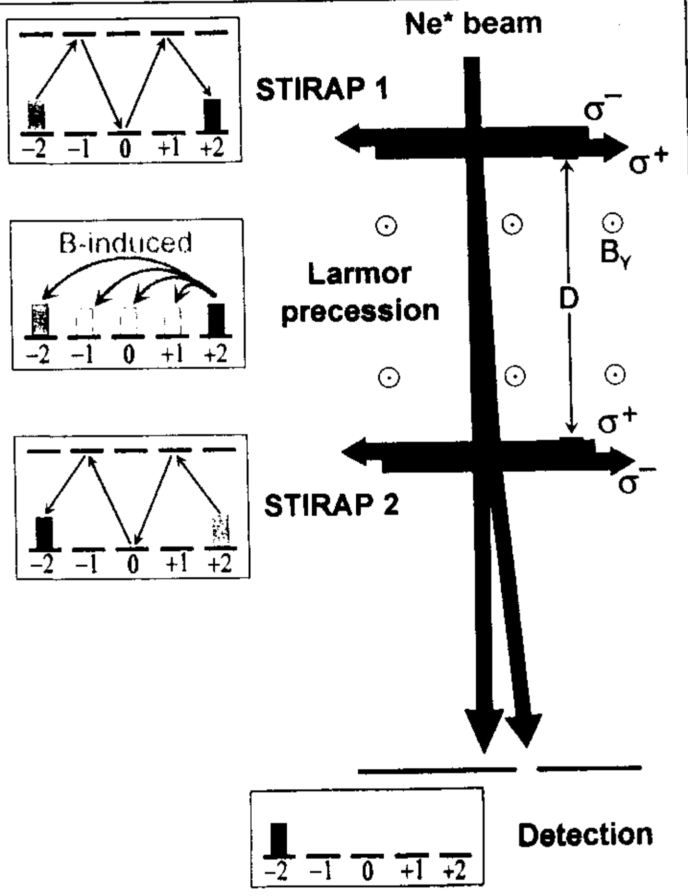
Larmor velocity filter – using deflection

Experimental result • compared with Simulation ——

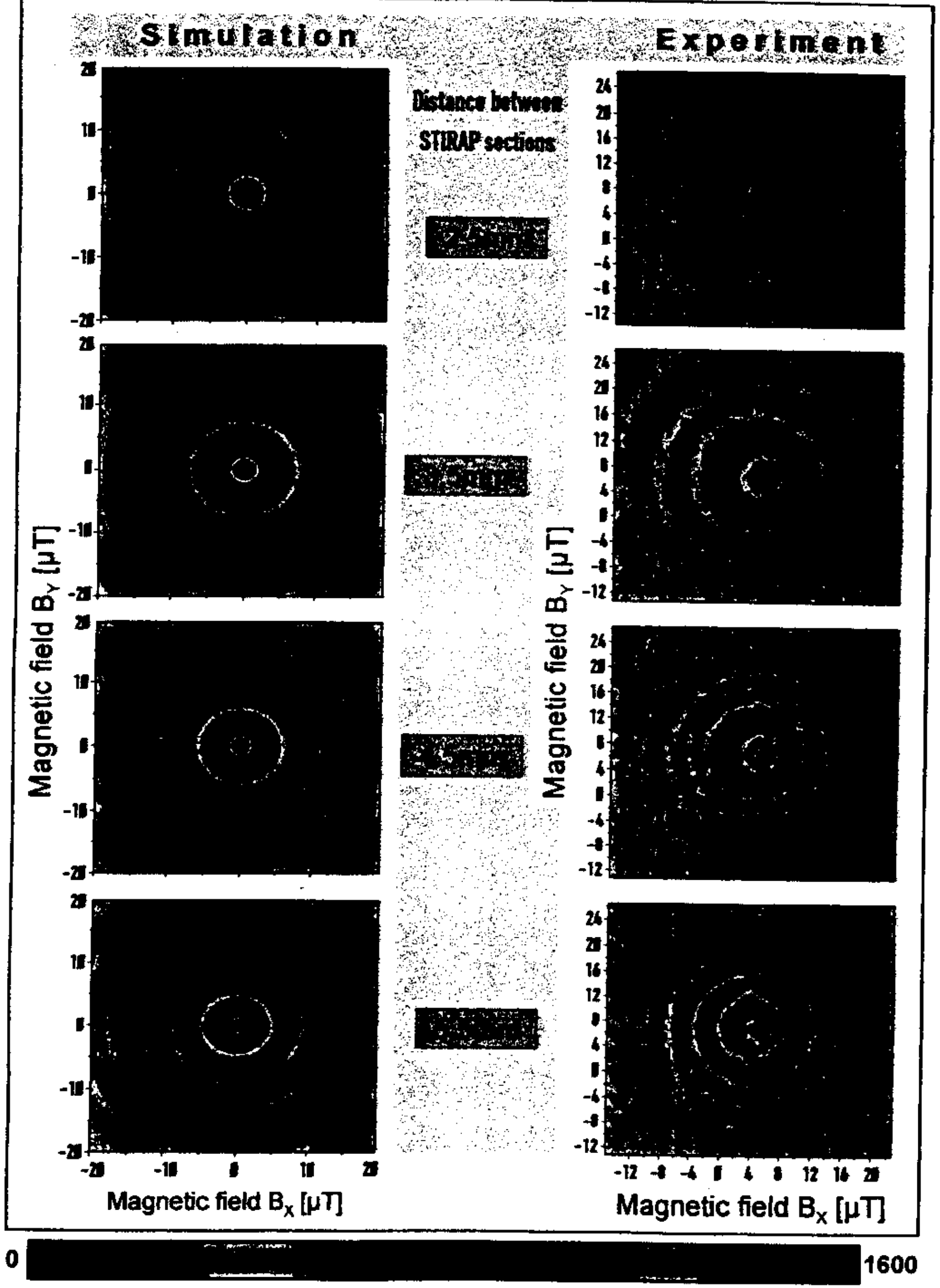


Population in m = -2:

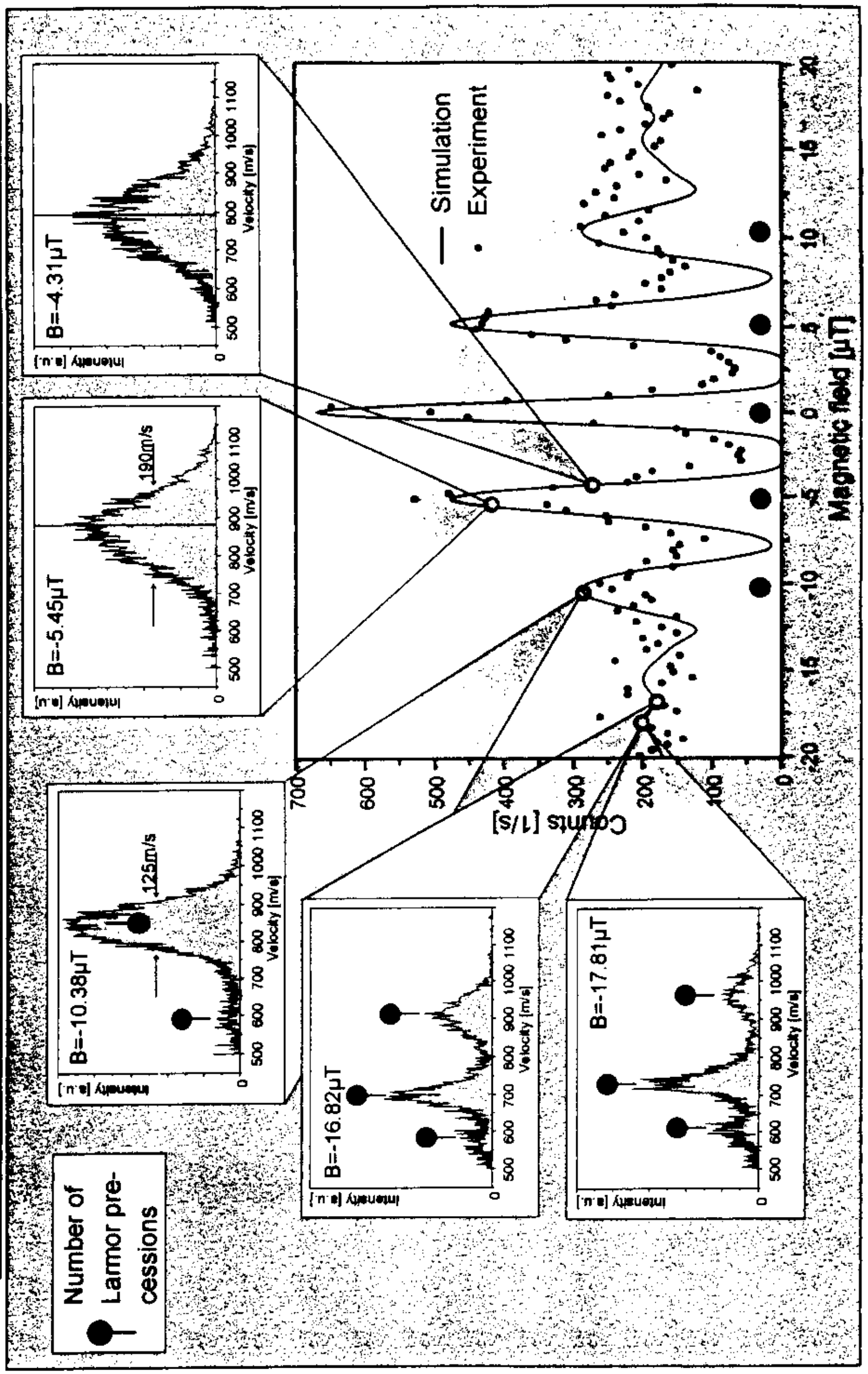
$$|c^{-2}|^2 \sim \left(\cos\frac{\omega_{Larmor}}{2}\cdot\frac{D}{v}\right)^8$$

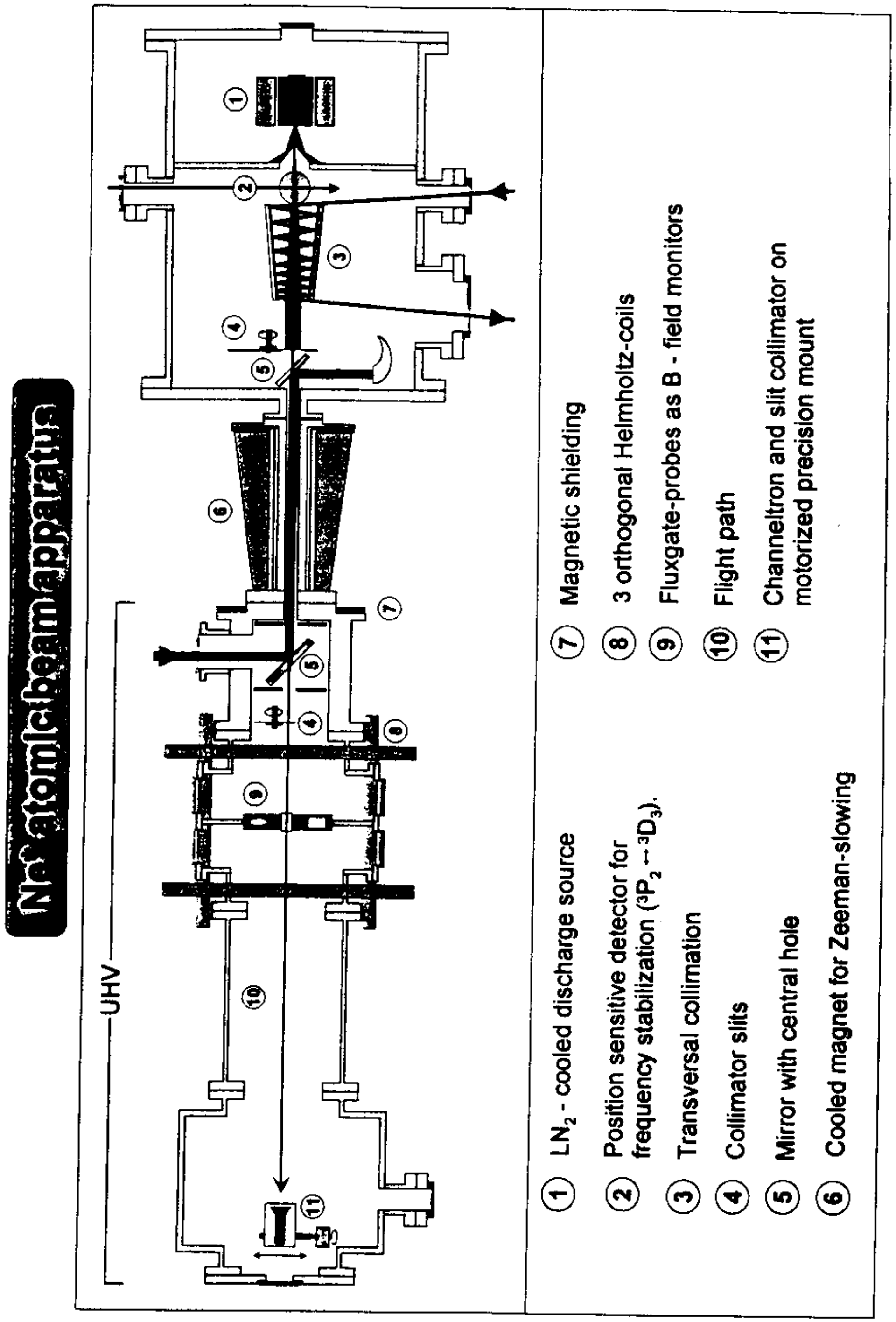


"Inverted" Larmor Velocity Filter



Inverted Larmor Velocity Filter: Time of Flight - first results





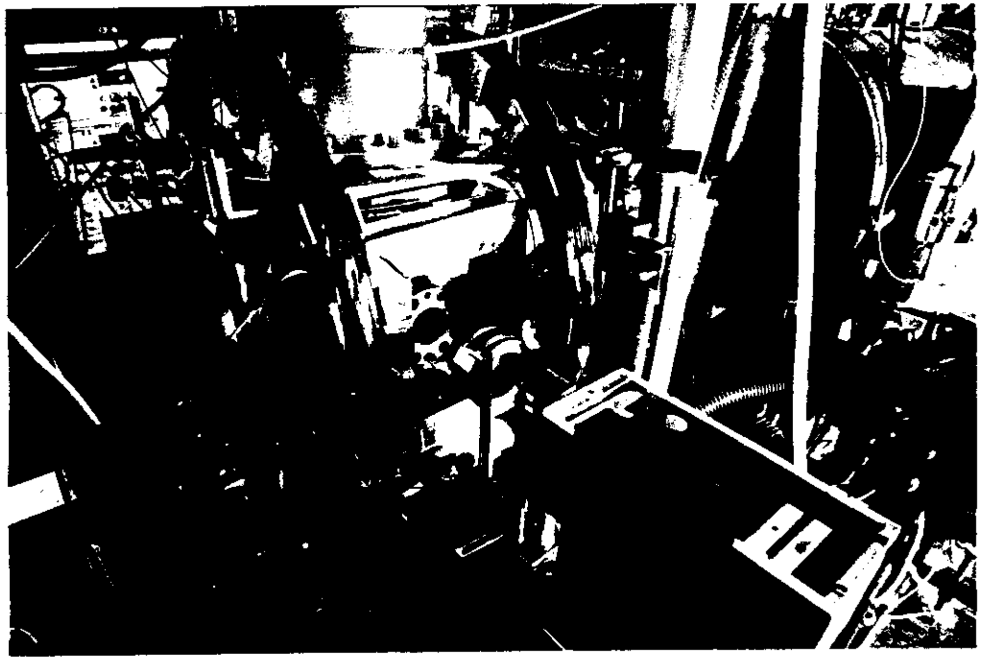
Towards a dark state (STIRAP-)
atomic interferometer

all optical components
insensitive to intensity variations
(dark state character)

insensitive to (small) frequency variations
(adiabatic following)

high contrast ratio

next steps...



- Integration of Zeeman-slower
  → higher accuracy of the Larmor-Velocity-Filter

- 1D ⤻ 2D polarization gradient cooling
  → Ne* - beam brightening

- σ- / π - atomic beam mirror and beam splitter

STIRAP - Interferometer ✓

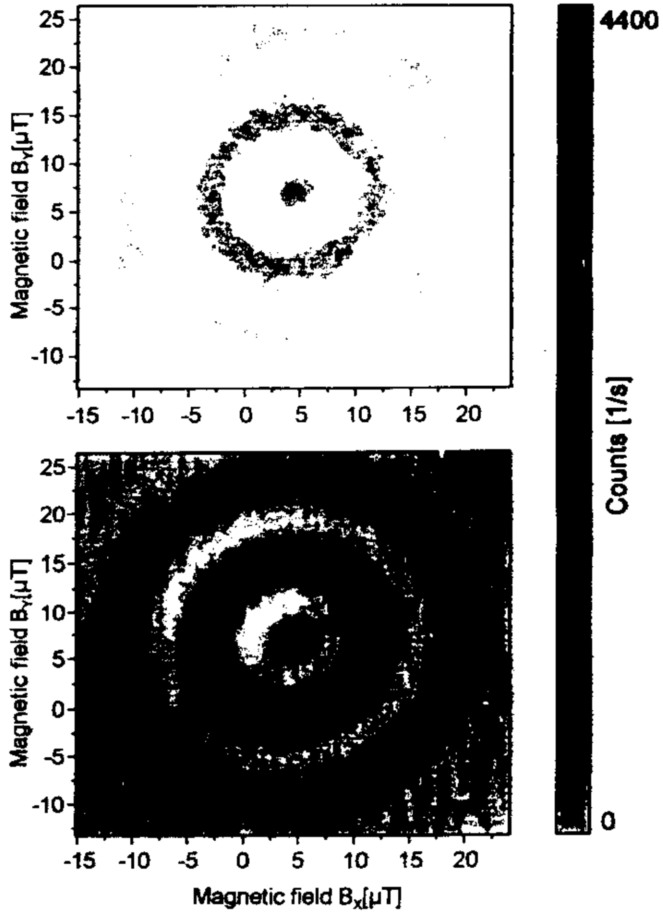
Next steps...

- slowing down the atoms
  → increasing the sensitivity of the Larmor velocity filter
- transversal laser cooling techniques
  → brightening the atomic beam

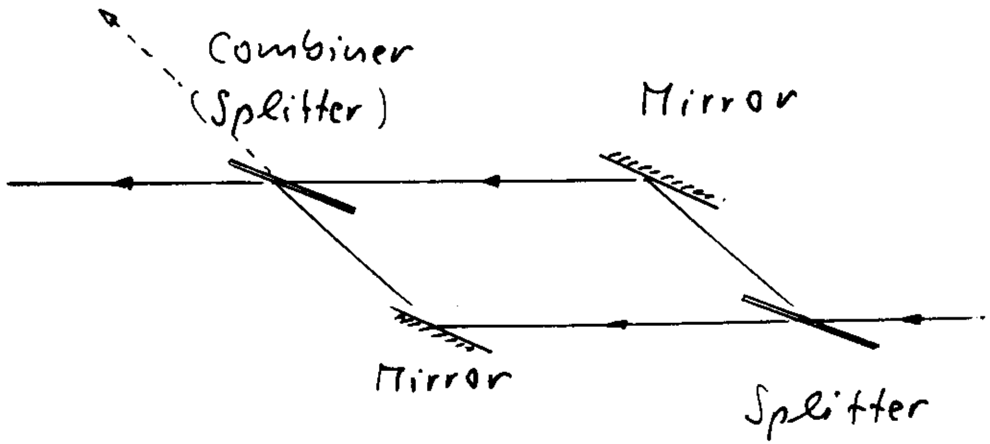
First impression of transversal cooling:

without cooling

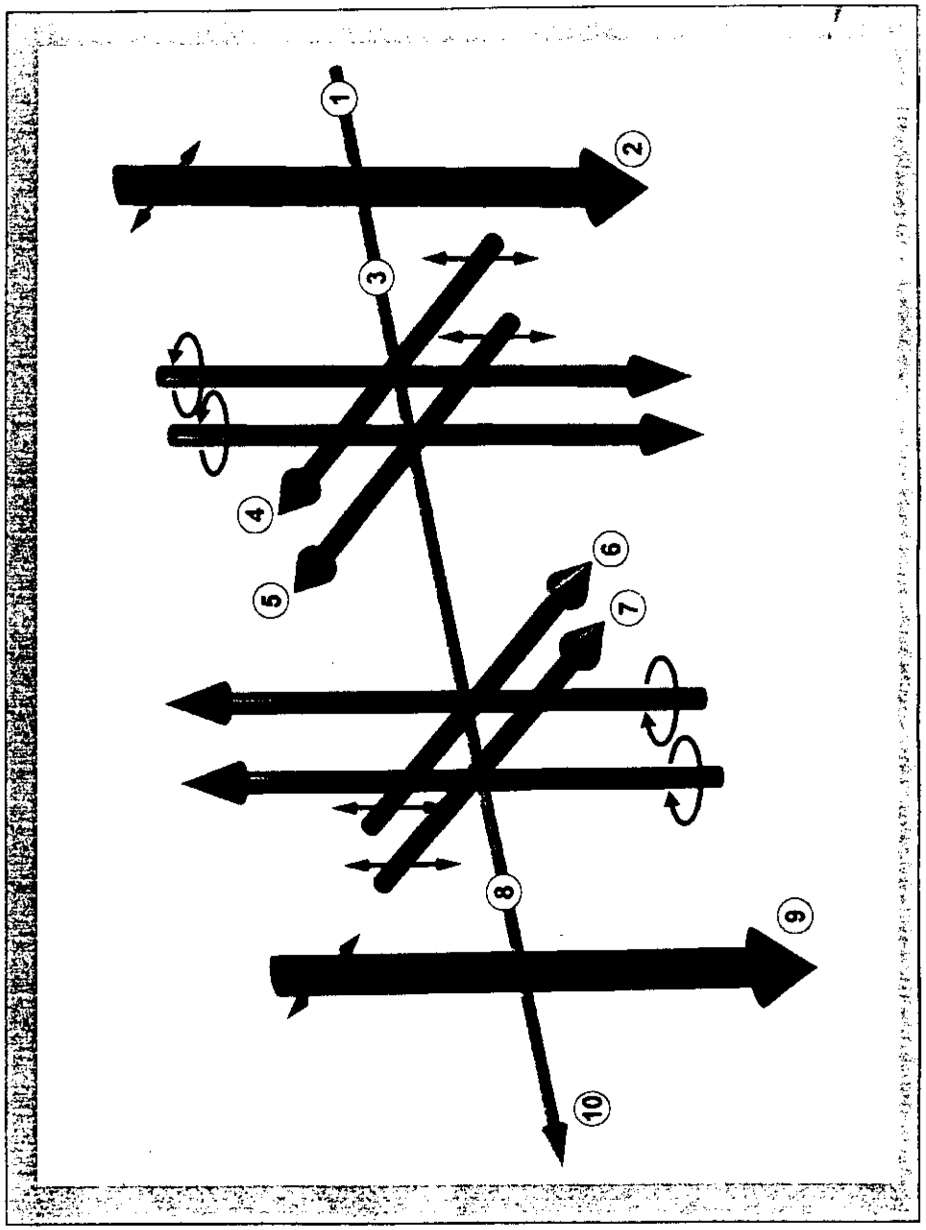
with cooling technique



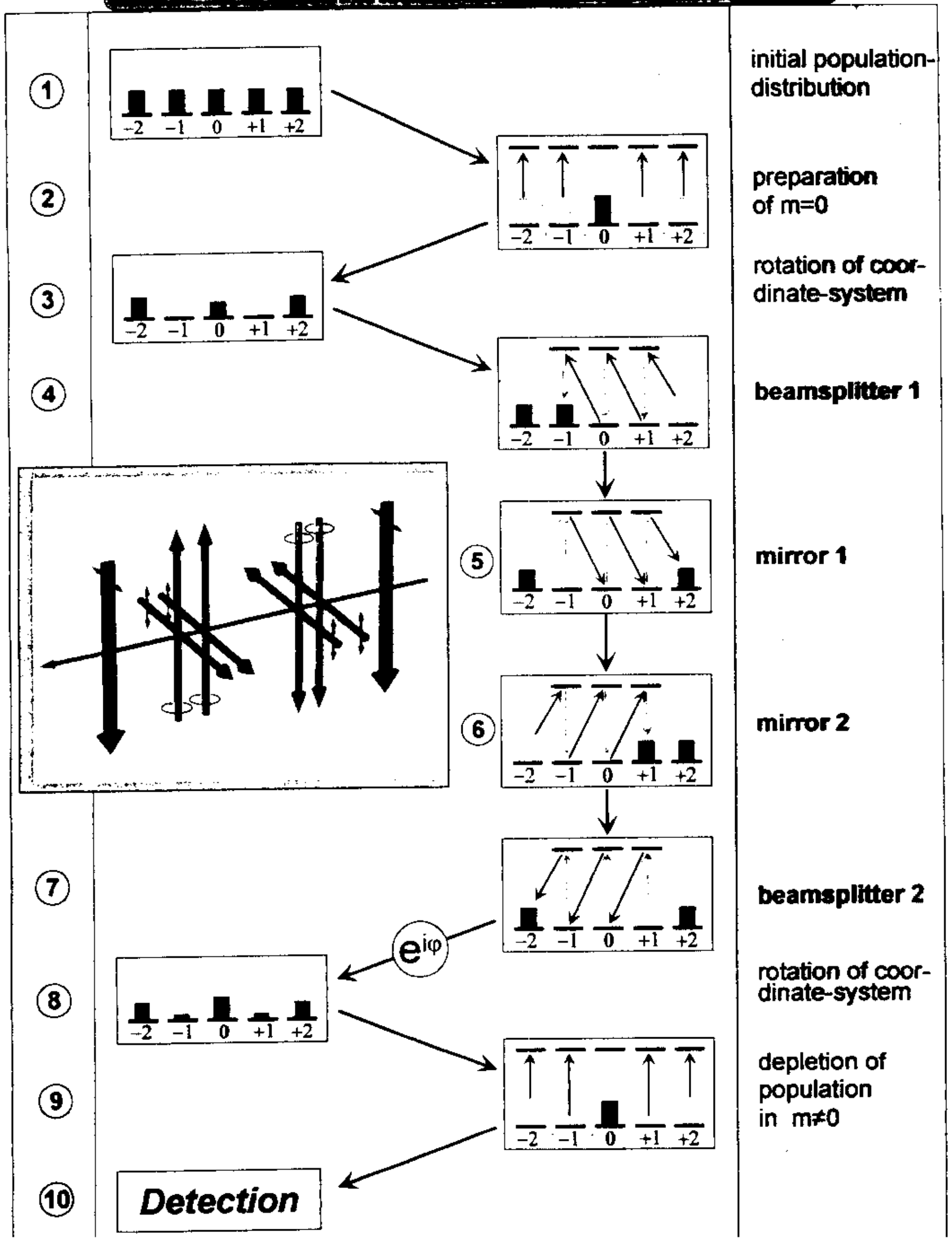
→ **ATOM INTERFEROMETER** ←

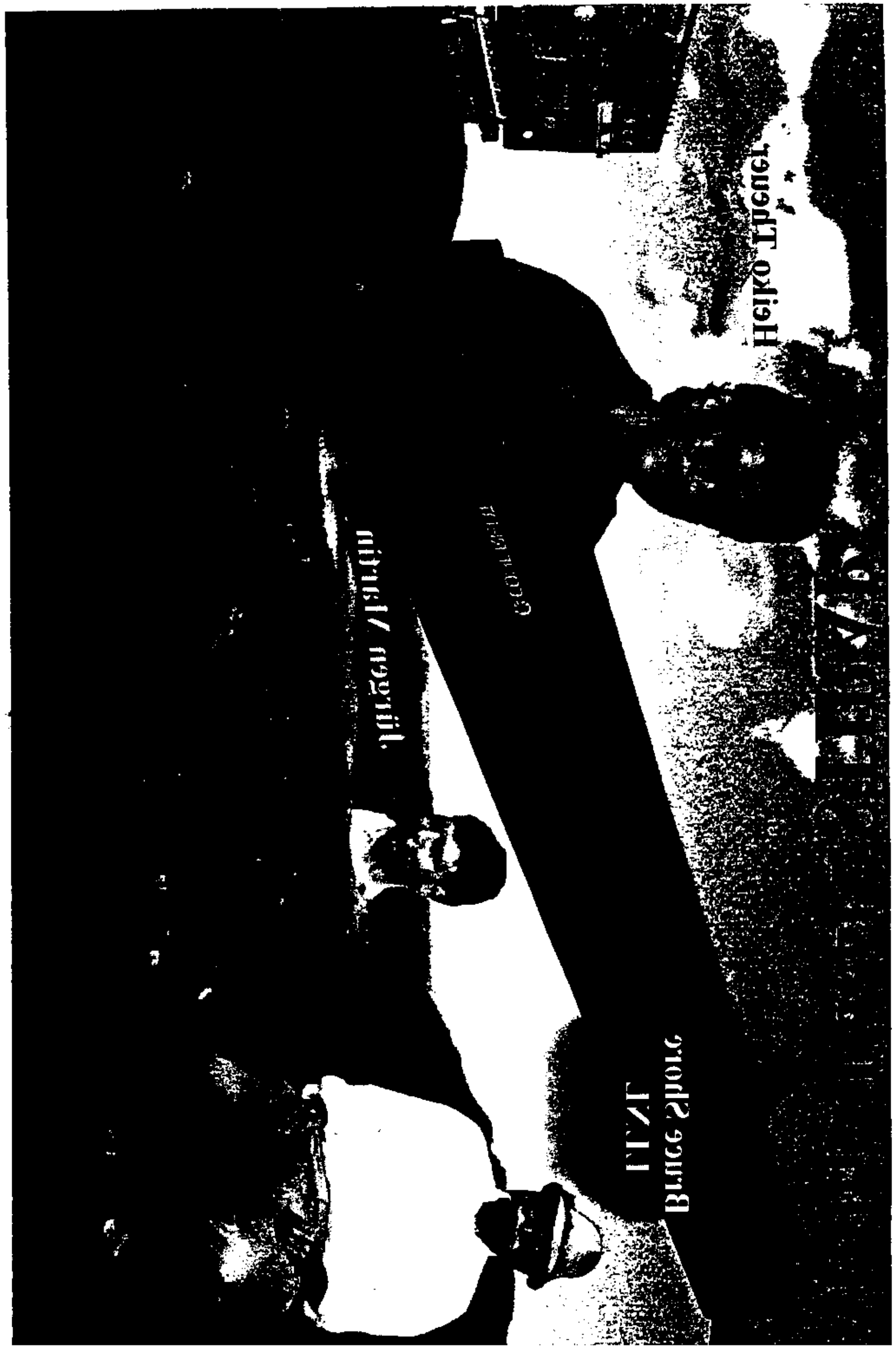


STIRAP - Atominterferometer



STIRAP - Atominterferometer
Transfer of population between m-states





Summary

- transfer of population "by design"
- process is (nearly) free of dissipation
- process is experimentally very robust
  - caution: case of multilevel systems (N > 3)
  - caution: implementation with pulsed lasers

- new attractive applications:
  - ⟶ collision dynamics
  - ⟶ spectroscopy
  - ⟶ magnetometry
  - ⟶ atom - optics
  - ⟶ laser cooling
  - ⟶ cavity - QED
  - ⟶ others . . . .