第一届冷原子物理和量子信息青年学者学术讨论会

A few issues on molecular condensates

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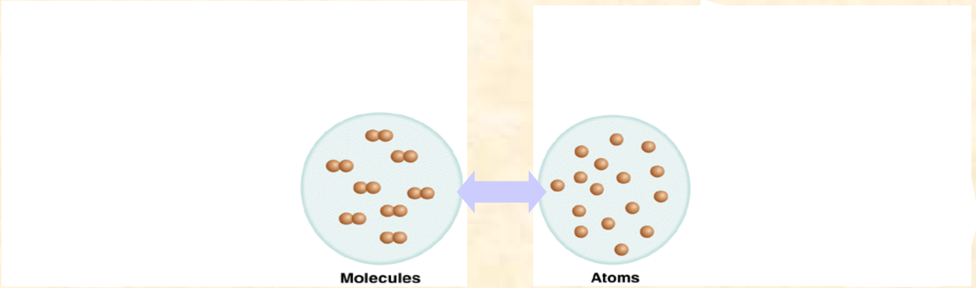
Outline

- Background
- Atomic dark state and creating ground molecular BEC
- Superchemistry and coherent atom-trimer conversion
- Quantum dynamics of a molecular matter-wave amplifier

BACKGROUND

From cold atoms to cold molecules



- **Great successes in cold atom research**: BEC, quantum degenerate fermions, atom interferometer, precision spectroscopy, vortices, optical lattice, …
  - Nobel Prize: 1997, 2001

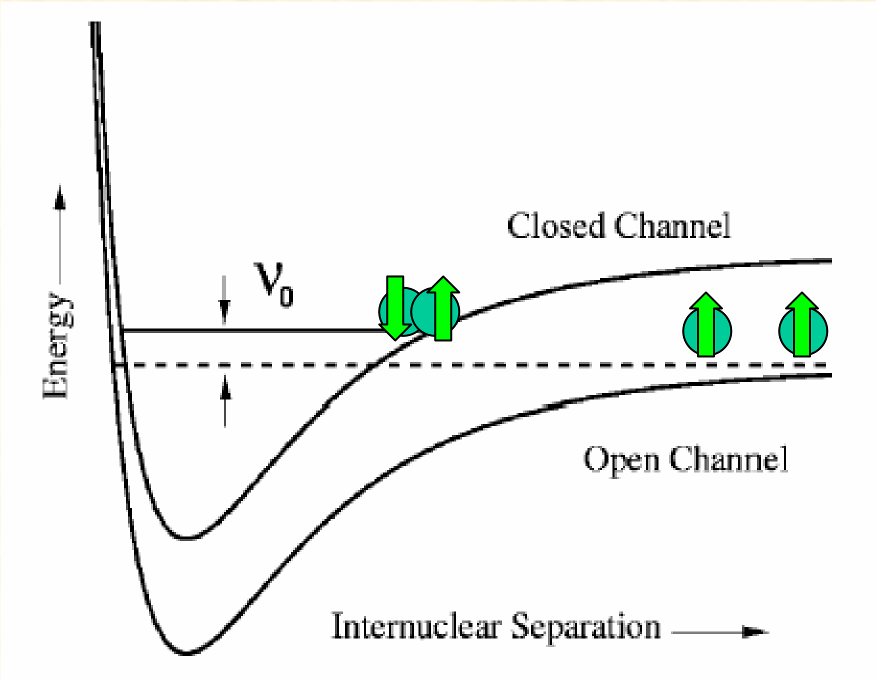
- **Play the same games with molecules**: controlled chemical reaction, collisional studies, precision molecular spectroscopy, polar molecules (large dipole moment), measurement of fundamental constants, macroscopic atom-molecule quantum coherence, …

How to get cold molecules?

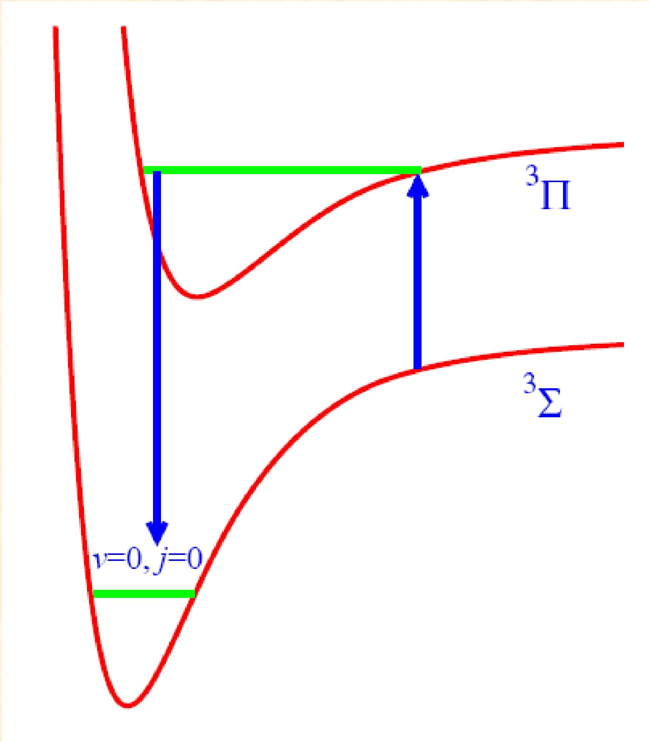
- Molecules are harder to cool than atoms are. (Lack of cycling transitions: direct laser cooling)

- Methods for creating cold molecules:
  - Direct Cooling
    - Buffer gas cooling (Doyle)
    - Stark deceleration (Meijer, Hinds)
    - Velocity-selective cooling (Rempe)
  - Indirect Cooling (cold atom and cold molecule)
    - Photo-association (Heinzen, Hulet, Walraven, Bigelow, Gould, Stwalley, Pillet, Knize, Rolston, Phillips, Lett, DeMille,…)
    - Feshbach resonance (Jin, Wieman, Grimm, Hulet, Thomas, Ketterle, Saloman, Rempe…)

Feshbach Resonance & Photoassociation: Atom-Molecule Coupling



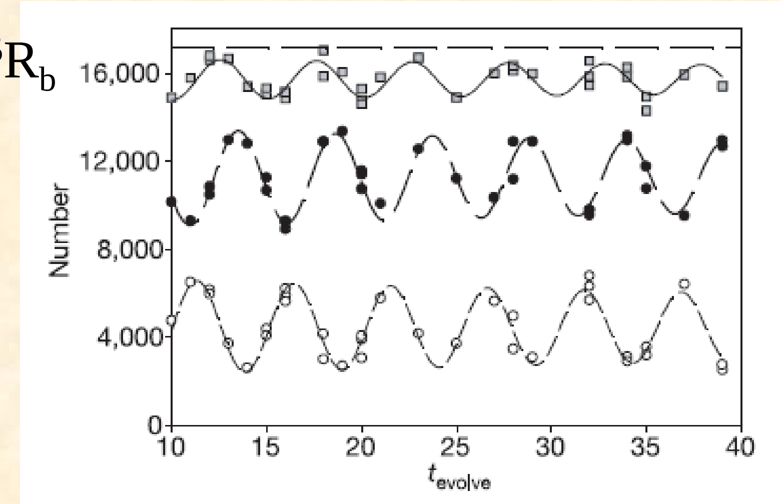
Feshbach Resonance



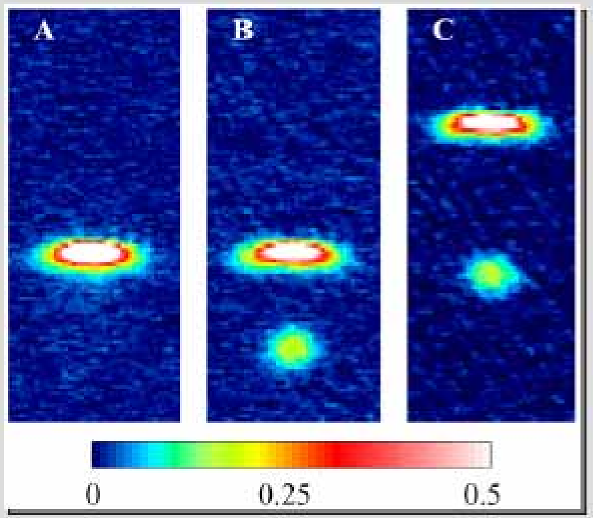
Photoassociation

making molecules from BECs

$^{85}R_b$

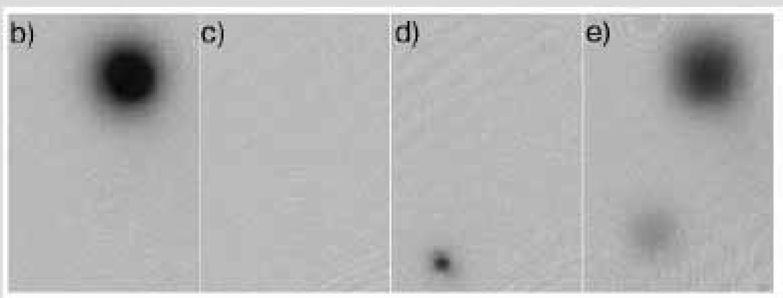


Atom–molecule coherence in a BEC
JILA, Nature 417, 529 (2002)



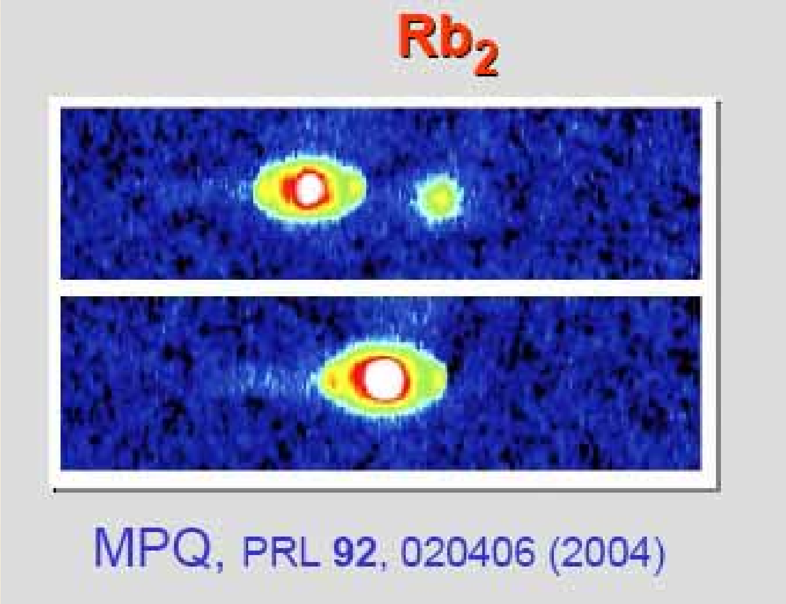
Cs_2

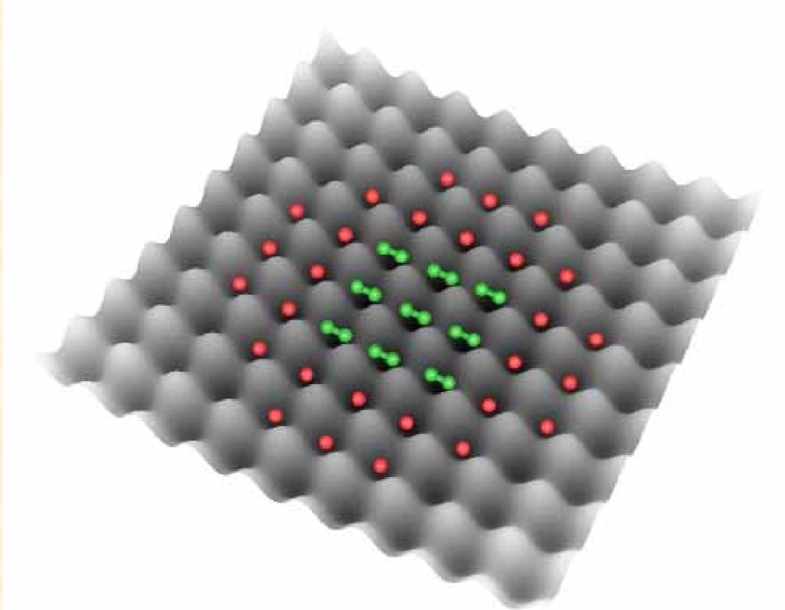
Innsbruck, Science **301**, 1510 (2003)



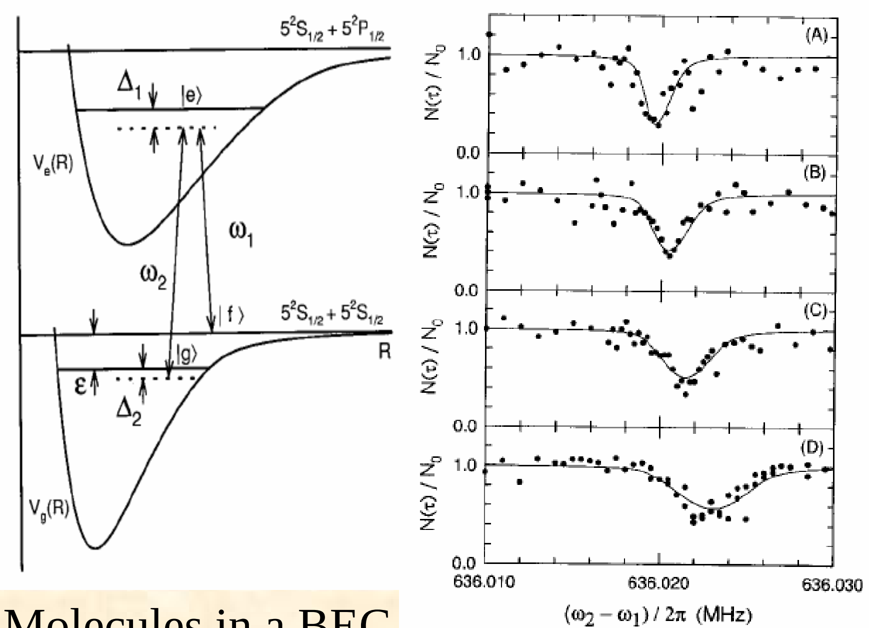
Na_2

MIT, PRL **91**, 210402 (2003)

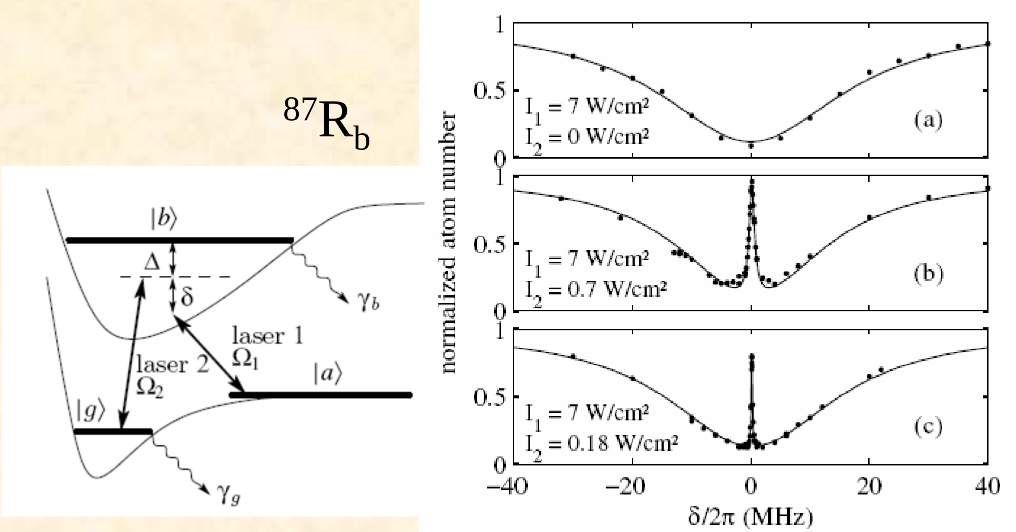




One molecule at each site of an optical lattice.
MPQ, Nature Phys. 2, 692 (2006)

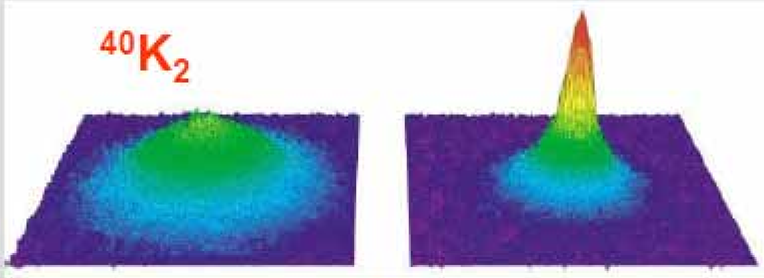


Molecules in a BEC
UT Austin, Science, 287, 1086 (2000)



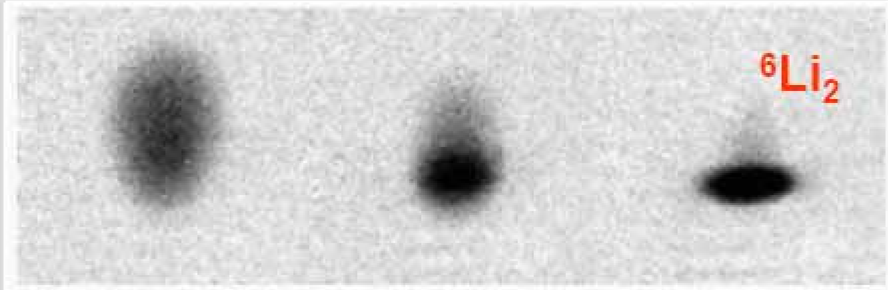
Atom-Molecule Dark States in a BEC
Innsbruck PRL, 95, 063202 (2005)

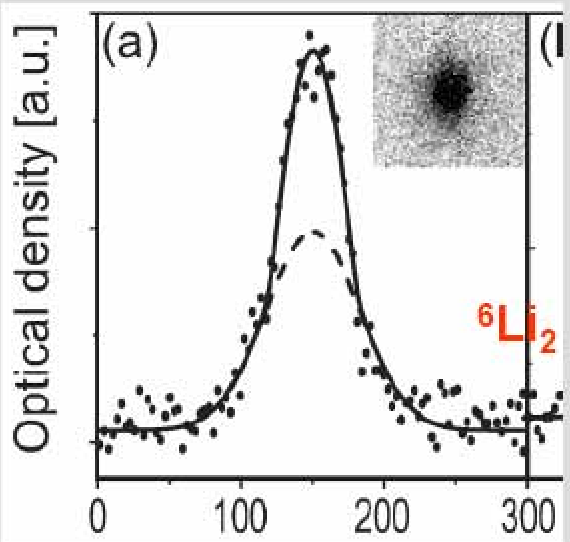
(fermion) molecular BEC gallery



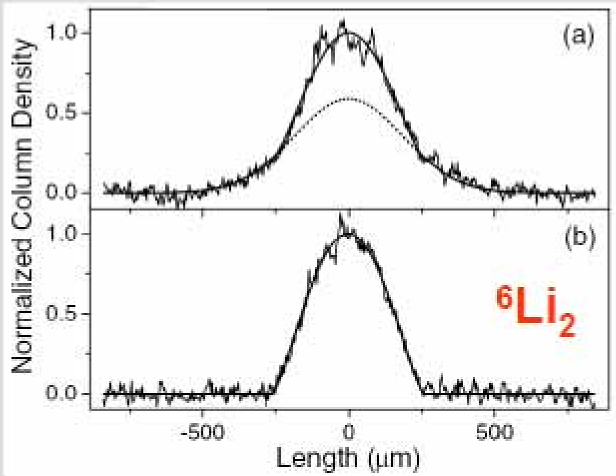
JILA, Jin et al.

MIT, Ketterle et al.



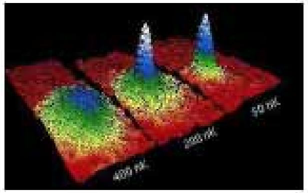


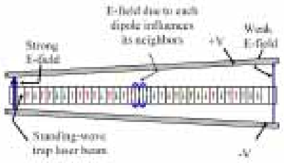
ENS Paris, Salomon et al.



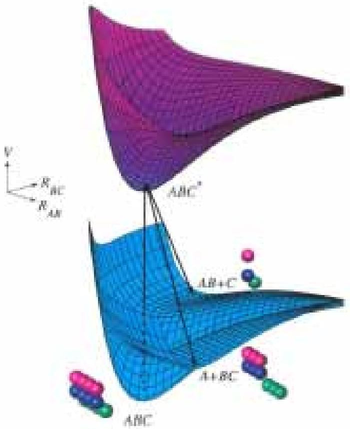
Rice,
Hulet et al.

coupled atom-molecule system





$\frac{d\alpha}{dt} \neq$ constant?



New phases of matter
Molecular BEC
BEC of polar species
(Ketterle, Wieman, Cornell, Jin, Pfau, Doyle ...)

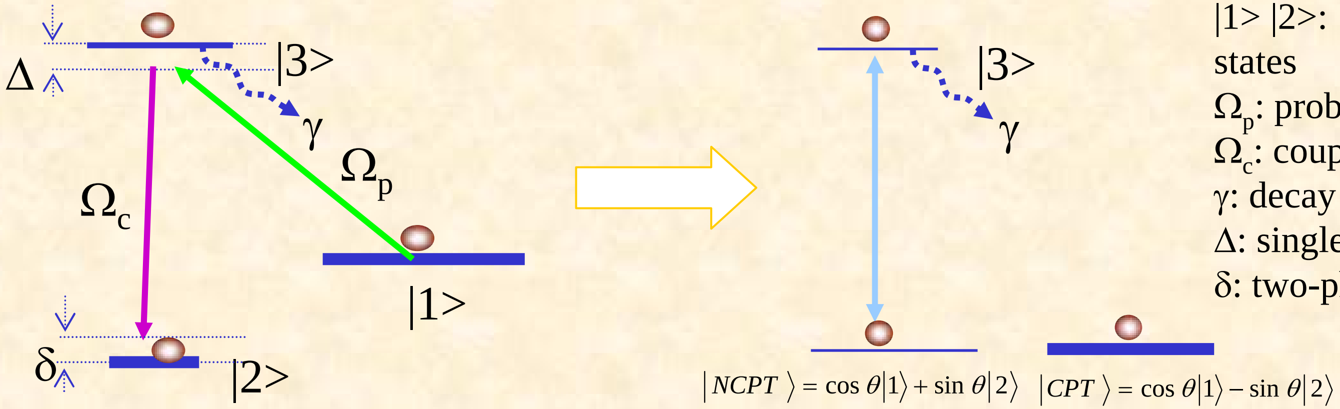
Quantum computation with
cold trapped molecules
(DeMille, Lukin, Doyle ...)

Test of fundamental symmetries
Search for time variation of
fundamental constants
(DeMille, Ye, Prentiss, Flambaum ...)

Chemistry in the quantum regime
Bose-enhanced chemistry
Controlled molecular dynamics
(Balakrishnan, Bohn, Hutson, Dalgarno, Kosloff, Doyle ...)

Atomic dark state and creating ground molecular BEC

Concept of Dark State or Coherent Population Trapping (CPT) State



|3>: unstable excited state
|1> |2>: two lower (stable) states
Ω_p: probe Rabi frequency
Ω_c: coupling Rabi frequency
γ: decay rate of the excited state
Δ: single-photon detuning
δ: two-photon detuning

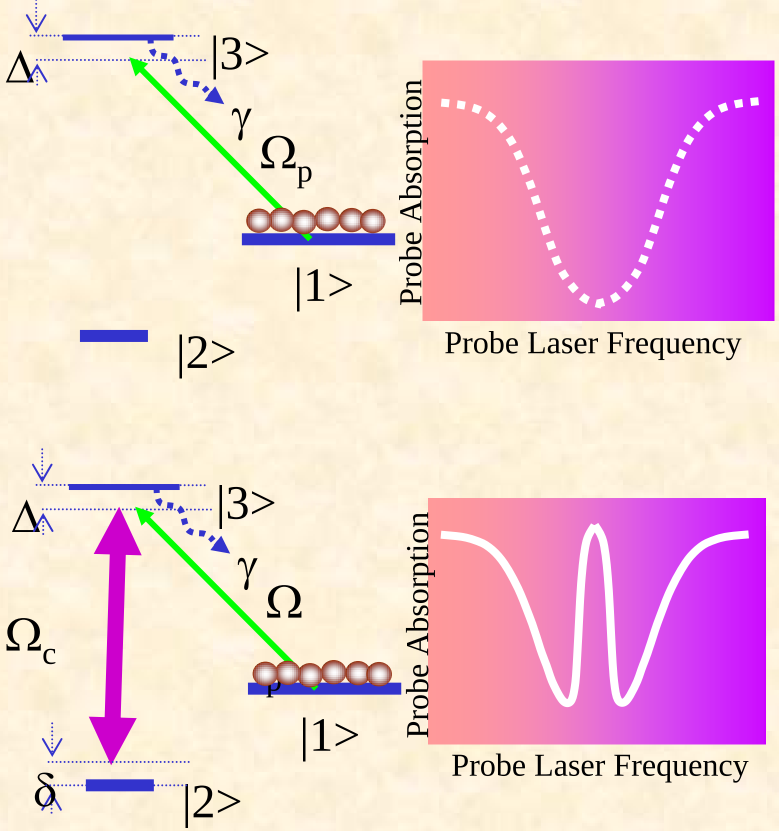
Two-Photon Resonance Condition: $\delta = 0$

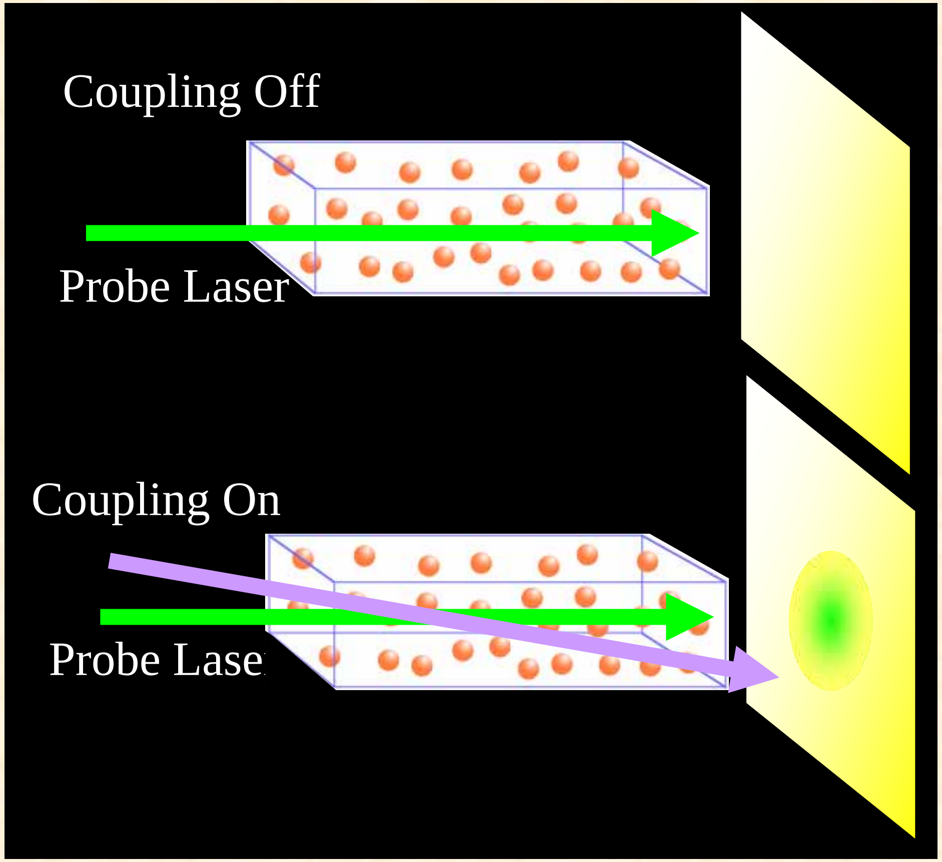
CPT or dark state $|CPT\rangle = \cos\theta|1\rangle - \sin\theta|2\rangle$

$$\tan\theta = \Omega_p / \Omega_c$$

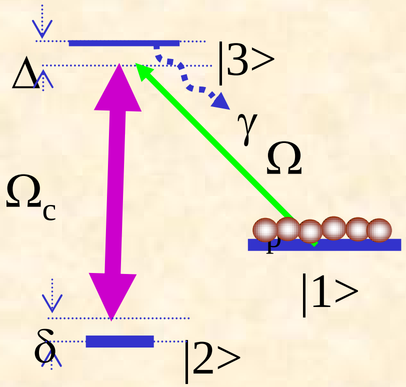
CPT Properties: (a) phase coherent
(b) immune to the spontaneous emission

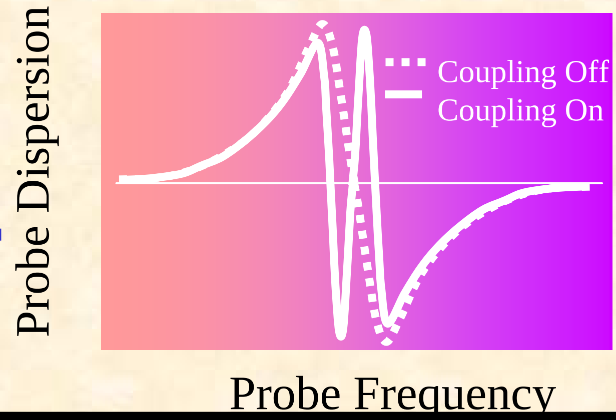
Electromagnetically Induced Transparency



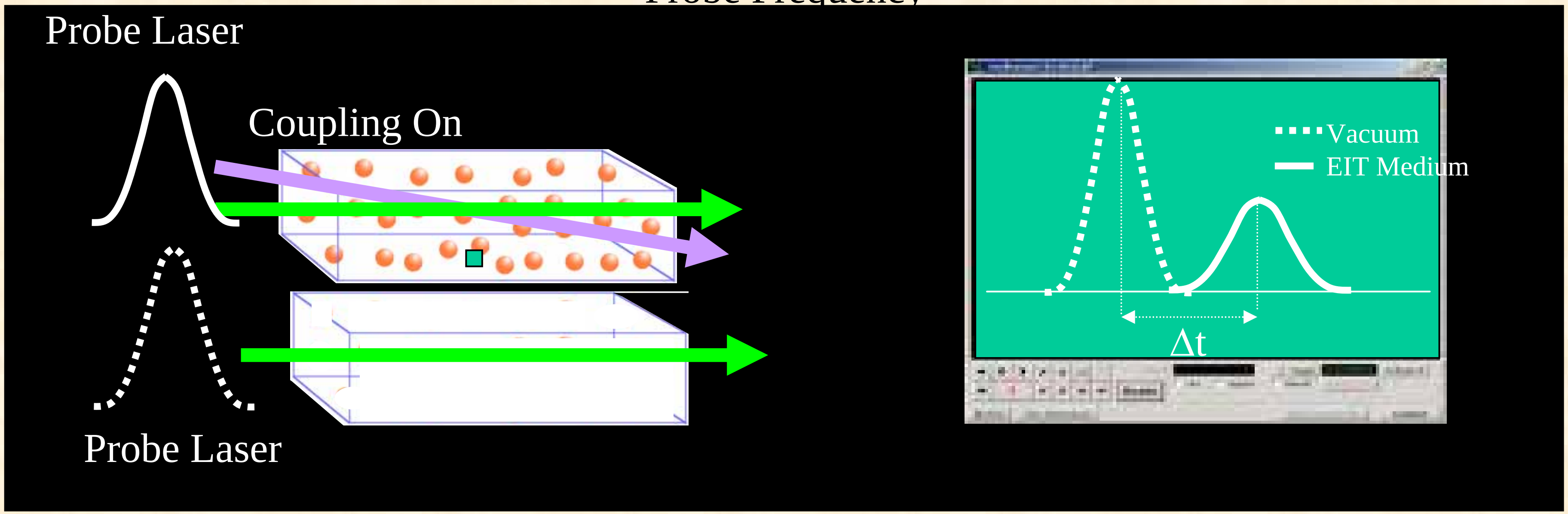


Slow Light





$$v_g = \frac{c}{n + \omega \frac{dn}{d\omega}}$$

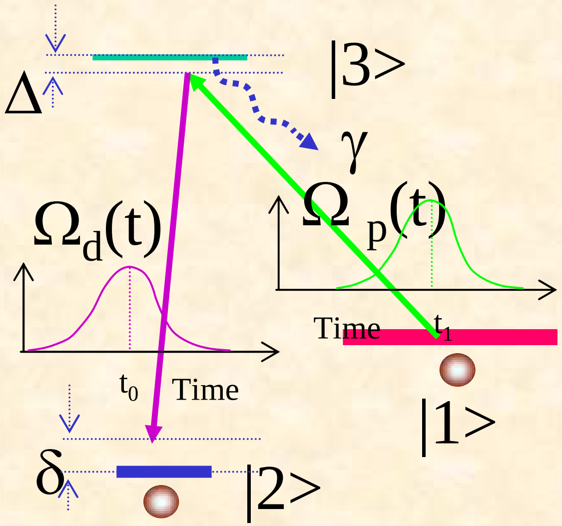


L. V. Hau, et al., Nature 397, 594 (1999)

STIRAP (STImulated Raman Adiabatic Passage)

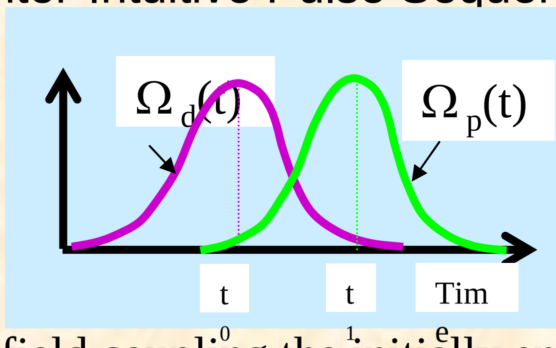
$$\left|CPT\right\rangle = \cos\theta\left|1\right\rangle - \sin\theta\left|2\right\rangle$$

$$n_1 = \frac{1}{1+|\Omega_p/\Omega_d|^2}, n_2 = \frac{|\Omega_p/\Omega_d|^2}{1+|\Omega_p/\Omega_d|^2}$$

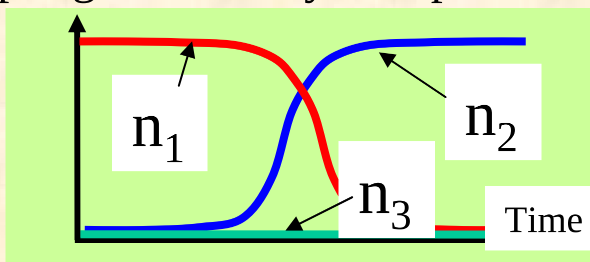


As $\Omega_p(t)/\Omega_d(t)$ ->0 to ∞, n_1 changes from 1 to 0, n_2 changes from 0 to 1, and n_2 remains empty

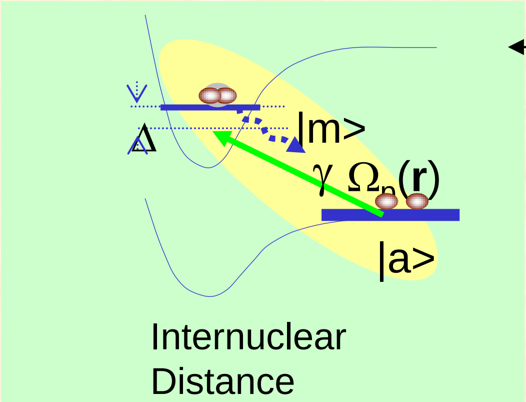
Counter-Intuitive Pulse Sequence



The field coupling the initially empty states is applied before the field coupling the initially occupied states.



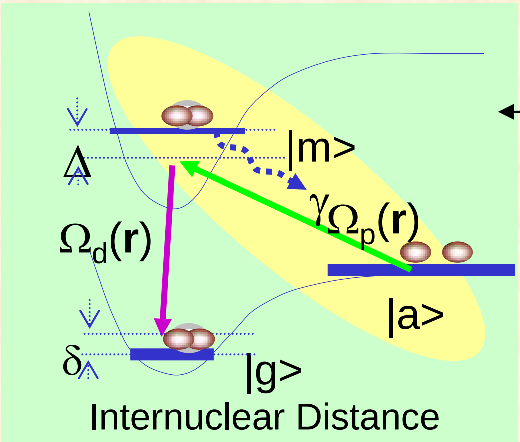
Photoassociation and Two-Color Raman photoassociation models



Photoassociation

A pair of free atoms in state |a> is brought into a bound molecule of state |m> by absorption of a photon through a dipole transition. Δ: single-photon detuning, controlled by the pump laser of Rabi frequency Ω_p

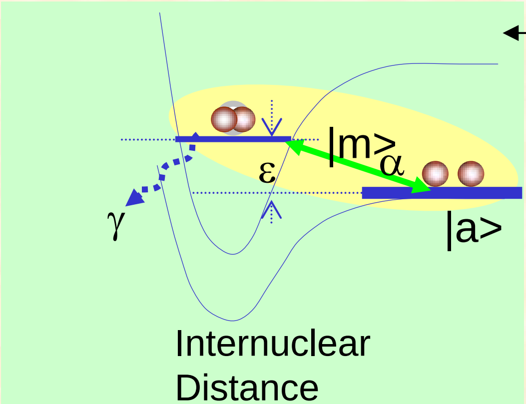
! |m> electronically excited: unstable and short-lived due to large Inelastic loss rate.



Two-Color Raman Photoassociation Model

A dump laser field of Rabi frequency Ω_d is introduced to drive molecules in |m> to a stable (ground) molecular state |g>. δ is the two-photon detuning.

Magnetoassociation and Feshbach-Assisted Raman Model

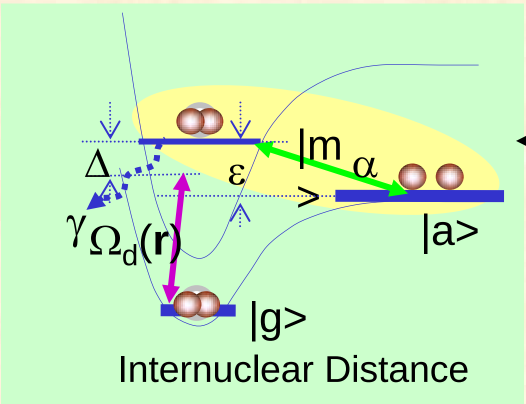


Feshbach Resonance

Atom pairs in an open channel |a> are converted into molecules of state |m> in a closed channel through the hyperfine spin interaction of strength α. ε is the Feshbach detuning, controlled by an external magnetic field.

☑ High efficiency. No need for strong laser fields.

✘ |m> **large** vibrational quantum number: unstable and short-lived due to large Inelastic loss rate.

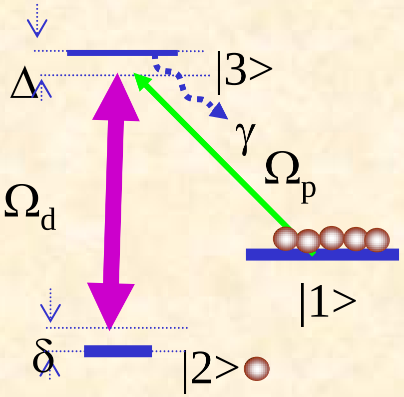


Feshbach-Assisted Raman Model

A dump laser field of Rabi frequency Ω_d is introduced to drive molecules in |m> to a stable (ground) molecular state |g>. Δ is the single-photon detuning.

?

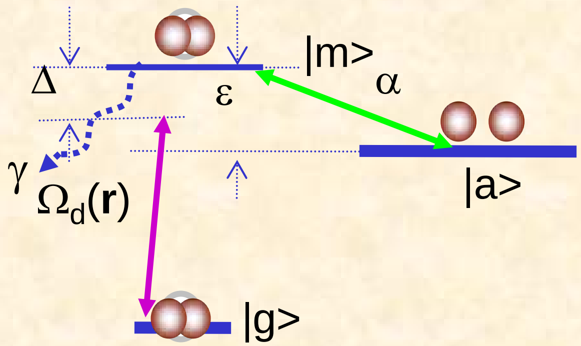
Three-Level Atomic Λ System



Linear System

It supports a CPT superposition, which has found a widespread of applications in the past few decades

Λ-type Coupled Atomic-Molecular Condensate System



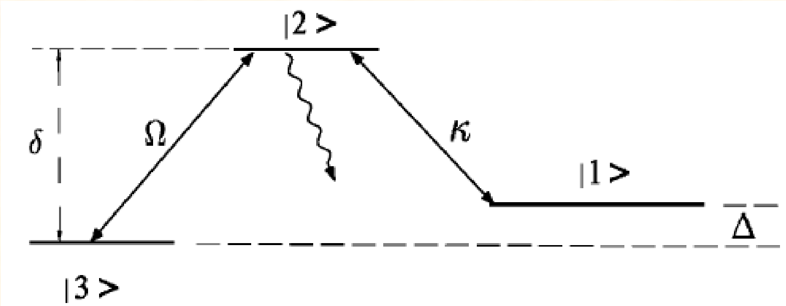
Nonlinear System

? Does this system support a CPT state, which is a coherent superposition between an atomic and a ground molecular condensate state.

VOLUME 84, NUMBER 17 PHYSICAL REVIEW LETTERS 24 APRIL 2000

Bose-Stimulated Raman Adiabatic Passage in Photoassociation

Matt Mackie, Ryan Kowalski, and Juha Javanainen
Department of Physics, University of Connecticut, Storrs, Connecticut 06269-3046



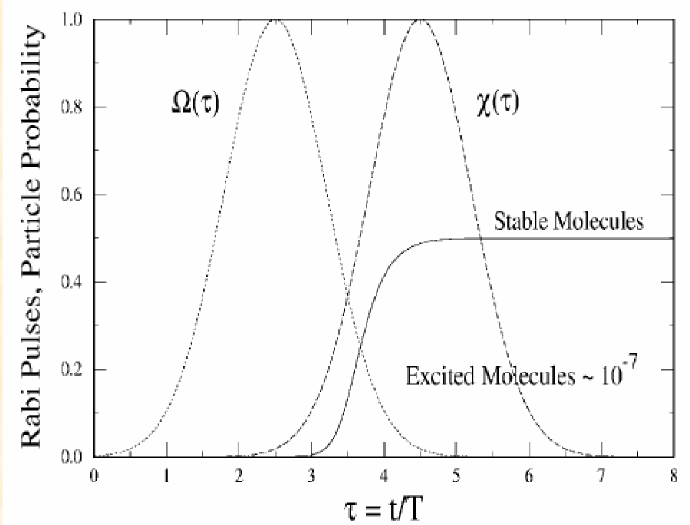
CPT solutions

$$\mu_0 = 0\,,\qquad a_0 = \sqrt{\bar\Omega\,(\sqrt{2+\bar\Omega^2}-\bar\Omega)}\,,\qquad b_0 = 0\,,\qquad g_0 = -\frac{1}{2}\,(\sqrt{2+\bar\Omega^2}-\bar\Omega)\,;$$

$$\frac{H}{\hbar} = \frac{H_0}{\hbar} - \tfrac{1}{2}\kappa(aab^\dagger + a^\dagger a^\dagger b) - \tfrac{1}{2}\Omega(bg^\dagger + b^\dagger g)$$

$$i\dot a = \tfrac{1}{2}\Delta a - \chi a^\dagger b\,,$$
$$i\dot b = \delta b - \tfrac{1}{2}(\chi aa + \Omega g)\,,$$
$$i\dot g = -\tfrac{1}{2}\Omega b\,.$$

$\chi = \sqrt{N}\,\kappa$ Bose enhancement!



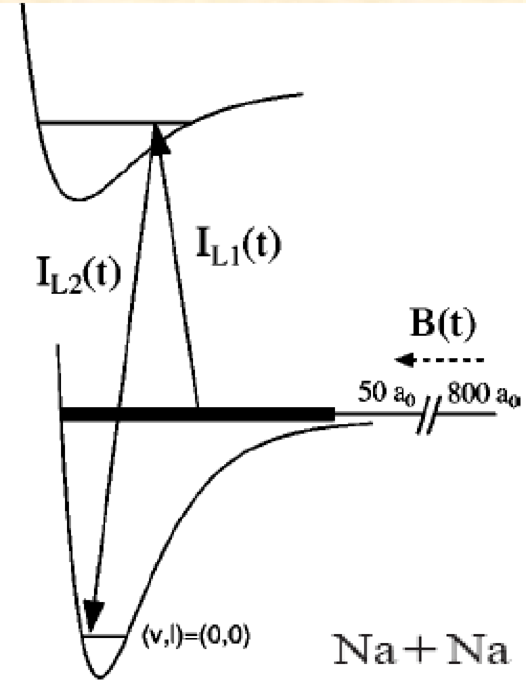
Neglect the collisions and decays, if there is no intermediate-state population (no photodissociation) and time is much shorter than the time scales for collisions

PHYSICAL REVIEW A, VOLUME 63, 031601(R)

Formation of a Bose condensate of stable molecules via a Feshbach resonance

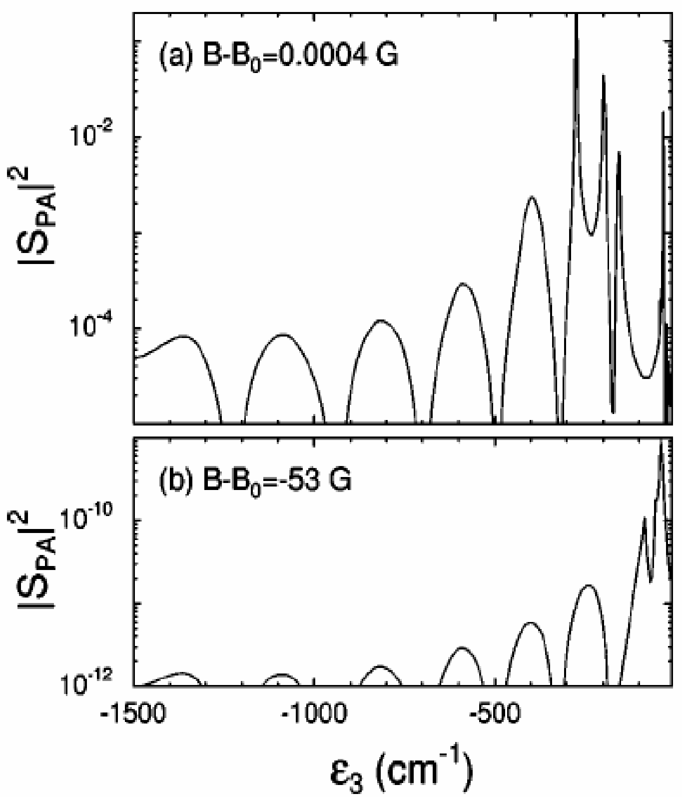
S. J. J. M. F. Kokkelmans, H. M. J. Vissers, and B. J. Verhaar
Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands

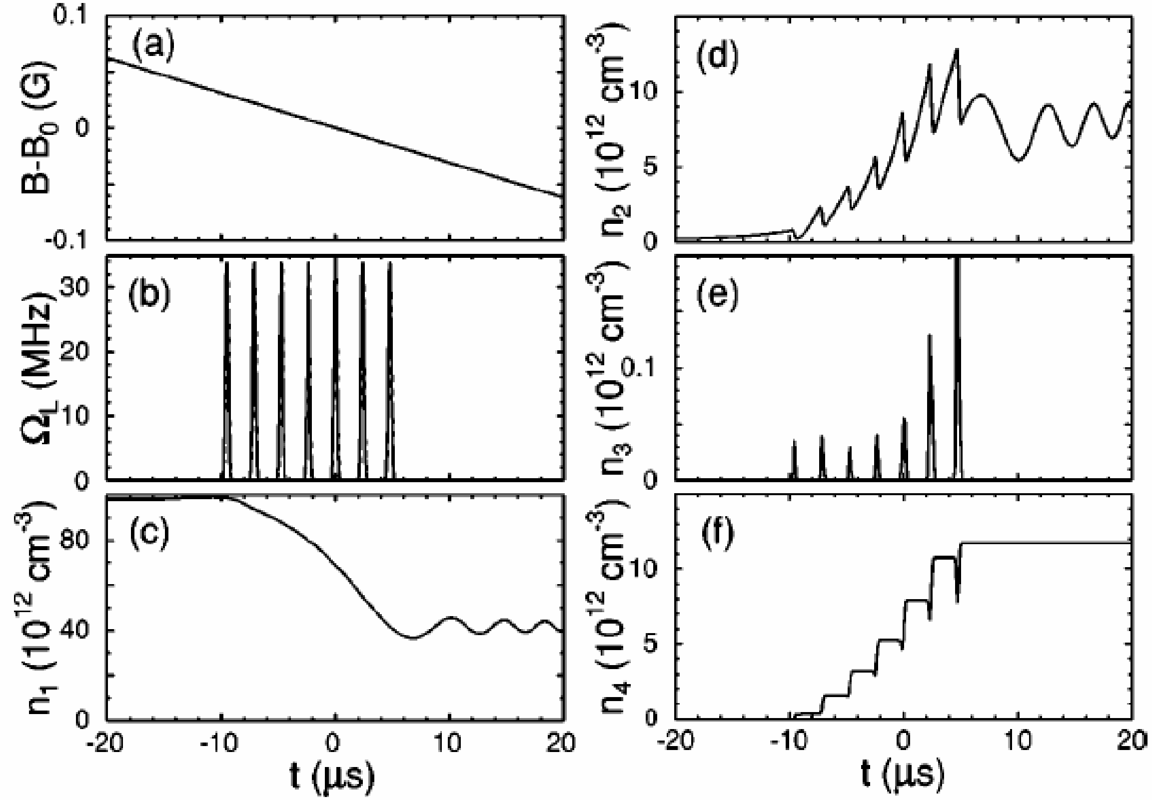
Feshbach + Raman technique



$$
\begin{aligned}
i\hbar\,\dot{\phi}_1 &= U_0|\phi_1|^2\phi_1 + 2\alpha\phi_1^*\phi_2\,,\\
i\hbar\,\dot{\phi}_2 &= \left(\varepsilon_2 - \frac{i}{2}\gamma_0\right)\phi_2 + \alpha\phi_1^2 + \frac{1}{2}\hbar\Omega_{L1}\phi_3\,,\\
i\hbar\,\dot{\phi}_3 &= \left(\varepsilon_3 - \frac{i}{2}\gamma_{sp} - \hbar\omega_{L1}\right)\phi_3 + \frac{1}{2}\hbar\Omega_{L1}\phi_2 + \frac{1}{2}\hbar\Omega_{L2}\phi_4\,,\\
i\hbar\,\dot{\phi}_4 &= (\varepsilon_4 - \hbar\omega_{L1} + \hbar\omega_{L2})\phi_4 + \frac{1}{2}\hbar\Omega_{L2}\phi_3\,,
\end{aligned}
\tag{1}
$$

Including atomic collision and decays


(a) B-B0=0.0004 G
(b) B-B0=-53 G
|SPA|2
10-2
10-4
10-10
10-12
-1500
-1000
-500
ε3 (cm-1)


(a)
(b)
(c)
(d)
(e)
(f)
B-B0 (G)
ΩL (MHz)
n1 (1012 cm-3)
n2 (1012 cm-3)
n3 (1012 cm-3)
n4 (1012 cm-3)
0.1
0
-0.1
30
20
10
80
40
0.1
5
-20
-10
0
10
20
t (μs)
t (μs)

PHYSICAL REVIEW A, VOLUME 65, 063619

Stimulated Raman adiabatic passage from an atomic to a molecular Bose-Einstein condensate

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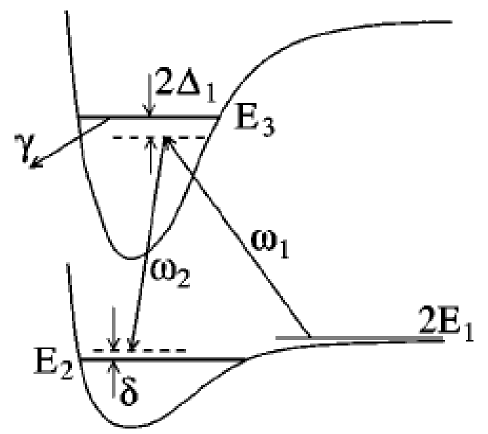
Department of Physics, The University of Queensland, Brisbane, Queensland 4072, Australia

D. J. Heinzen and R. H. Wynar
Department of Physics, University of Texas, Austin, Texas 78712



$$\hat{H}^{(0)} = \int d^3\mathbf{x} \sum_{i=1}^{3} \left[\frac{\hbar^2}{2m_i} |\nabla \hat{\Psi}_i(\mathbf{x})|^2 + V_i(\mathbf{x}) \hat{\Psi}_i^\dagger(\mathbf{x}) \hat{\Psi}_i(\mathbf{x}) \right]$$

$$\hat{H}_{int}^{(s)} = \frac{\hbar}{2} \int d^3\mathbf{x} \sum_{ij} U_{ij} \hat{\Psi}_i^\dagger(\mathbf{x}) \hat{\Psi}_j^\dagger(\mathbf{x}) \hat{\Psi}_j(\mathbf{x}) \hat{\Psi}_i(\mathbf{x}),$$

$$\hat{H}_{int}^{(1-3)} = \int d^3\mathbf{x} \left[\frac{-\hbar\Omega_1}{2\sqrt{2}} e^{-i\omega_1 t} \hat{\Psi}_1^2(\mathbf{x}) \hat{\Psi}_3^\dagger(\mathbf{x}) + \mathrm{H.c.} \right],$$

$$\hat{H}_{int}^{(2-3)} = \int d^3\mathbf{x} \left[\frac{-\hbar\Omega_2}{2} e^{-i\omega_2 t} \hat{\Psi}_2(\mathbf{x}) \hat{\Psi}_3^\dagger(\mathbf{x}) + \mathrm{H.c.} \right].$$

$$\Omega_i = \int d^3\mathbf{R} \Omega_i^{(el)}(\mathbf{R})\, u_3^*(R) u_i(R) \approx \overline{\Omega_i^{(el)}} I_{i,3} \quad (i=1,2)$$

Franck-Condon overlap integrals $I_{i,3} = \int d^3\mathbf{R} u_3^*(R) u_i(R)$

$$2\Delta_1(\mathbf{x}) = [E_3 - 2V_1(\mathbf{x})]/\hbar - \omega_1,$$

$$\Delta_2(\mathbf{x}) = [E_3 - V_2(\mathbf{x})]/\hbar - \omega_2,$$

$$\Delta_3(\mathbf{x}) = [E_3 - V_3(\mathbf{x})]/\hbar.$$

$$\Delta_j^{GP}(\mathbf{x},t) = \Delta_j(\mathbf{x}) + \frac{\hbar}{2m_j}\nabla^2 - \sum_{k=1}^{3} U_{jk}|\psi_k|^2 + i\frac{\gamma_j}{2}.$$

$$\delta \equiv \Delta_2(0) - 2\Delta_1(0) = -(E_2 - 2E_1)/\hbar + (\omega_1 - \omega_2).$$

$$\frac{\partial\psi_1(\mathbf{x},t)}{\partial t} = i\Delta_1^{GP}\psi_1 + \frac{i\Omega_1^*}{\sqrt{2}}\psi_3\psi_1^*,$$

$$\frac{\partial\psi_2(\mathbf{x},t)}{\partial t} = i\Delta_2^{GP}\psi_2 + \frac{i\Omega_2^*}{2}\psi_3,$$

$$\frac{\partial\psi_3(\mathbf{x},t)}{\partial t} = i\Delta_3^{GP}\psi_3 + \frac{i\Omega_1}{2\sqrt{2}}\psi_1^2 + \frac{i\Omega_2}{2}\psi_2,$$

Neglecting kinetic energy terms, i.e., in the Thomas-Fermi limit of large, relatively dense condensates, which is a regime of many experiments.

$$\Delta_j^{GP}(\mathbf{x},t) \longrightarrow \Delta_j^{TF}(\mathbf{x},t) = \Delta_j(\mathbf{x}) - \sum_{k=1}^{3} U_{jk}|\psi_k|^2 + i\frac{\gamma_j}{2}.$$

$$\psi_m = \psi_1/\sqrt{2}$$

$$\tilde{\Omega}_1 = \psi_1\Omega_1$$

Bose enhancement!

$$\frac{\partial\psi_m(\mathbf{x},t)}{\partial t} = i\Delta_1^{TF}\psi_m + \frac{i\tilde{\Omega}_1^*}{2}\psi_3$$

$$\frac{\partial\psi_2(\mathbf{x},t)}{\partial t} = i\Delta_2^{TF}\psi_2 + \frac{i\Omega_2^*}{2}\psi_3$$

$$\frac{\partial\psi_3(\mathbf{x},t)}{\partial t} = i\Delta_3^{TF}\psi_3 + \frac{i\tilde{\Omega}_1}{2}\psi_m + \frac{i\Omega_2}{2}\psi_2$$

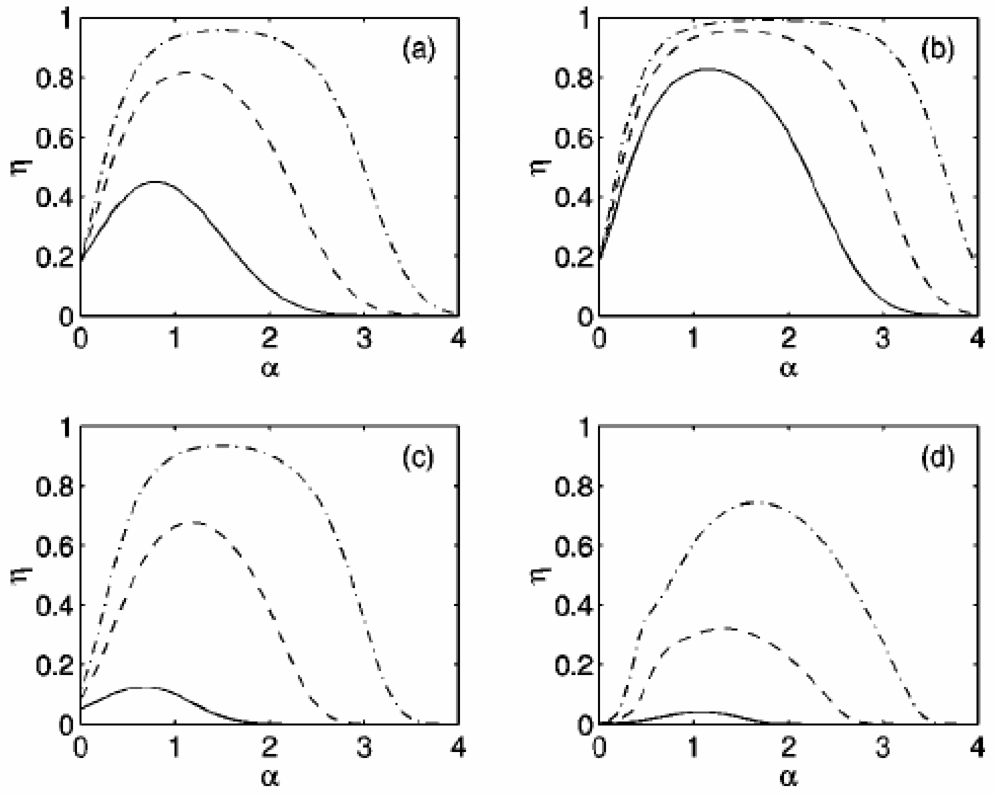
STIRAP solution

$$\psi_1(\mathbf{x},t) = \psi_1(\mathbf{x},0)\cos(\theta),$$

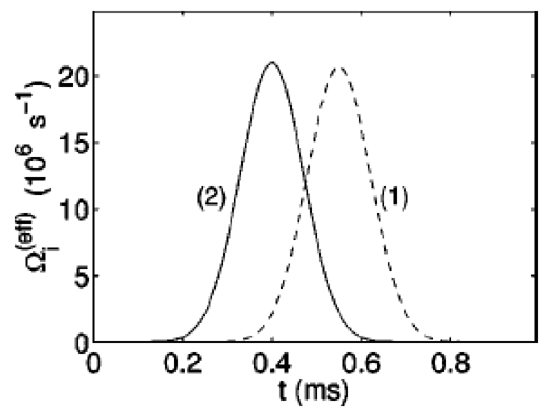
$$\psi_2(\mathbf{x},t) = -\psi_1(\mathbf{x},0)\sin(\theta)/\sqrt{2},$$

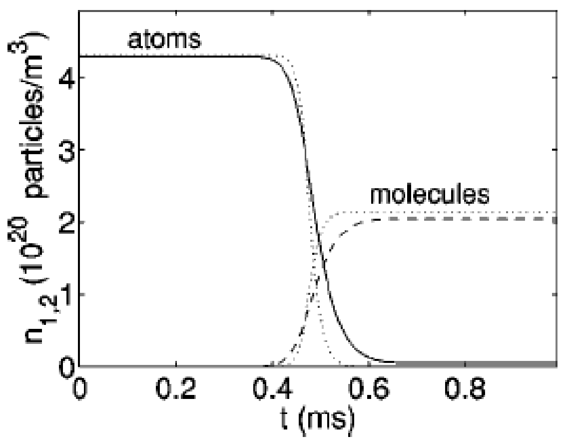
$$\psi_3(\mathbf{x},t) = 0.$$

$$\tan(\theta) = \frac{\tilde{\Omega}_1}{\Omega_2} = \frac{\psi_1\Omega_1}{\Omega_2} = \left[\sqrt{\frac{\psi_1^2(\mathbf{x},0)\Omega_1^2}{\Omega_2^2} + \frac{1}{4}} - \frac{1}{2}\right]^{1/2}$$

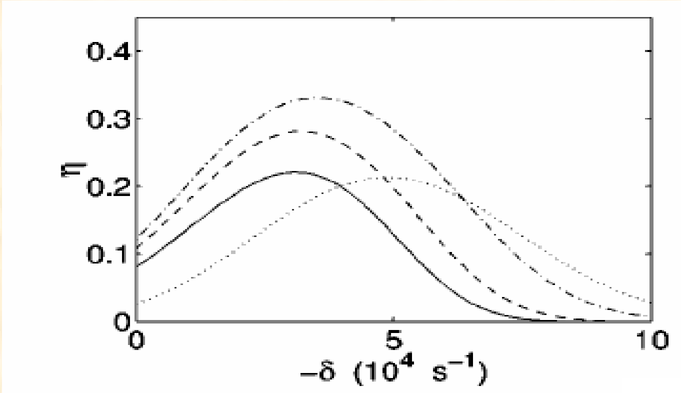


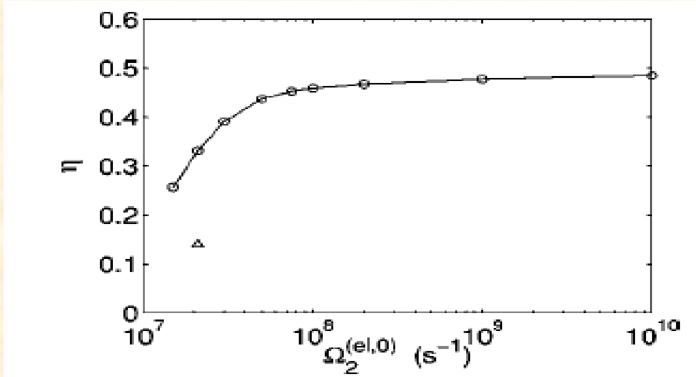
Increasing pulse separation, increasing Rabi frequency, (b,d) for larger pulse width, (c,d) including collisions.





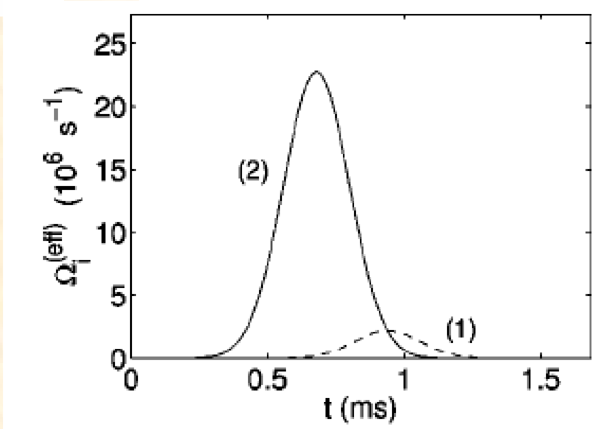
Franck-Condon integrals, $|I_{1,3}| \simeq 10^{-14}$ m$^{3/2}$ and $|I_{2,3}| \simeq 0.1$ [1], the magnitudes of $\Omega_1^{(eff,0)} = \Omega_2^{(eff,0)} = 2.1\times10^7$ s^{-1} translate to peak values of the bare Rabi frequencies equal to $\Omega_1^{(el,0)} = 10^{11}$ s^{-1} [for $n_1(0) = 4.3\times10^{20}$ m^{-3}] and $\Omega_2^{(el,0)} = 2.1\times10^8$ s^{-1}. The peak Rabi frequency of $\Omega_1^{(el,0)} = 10^{11}$ s^{-1} for the free-bound transition would be realized with a 1 W laser power and a waist size of about 10 μm, off-resonance operation, tuning the two-photon detuning to compensate the collision

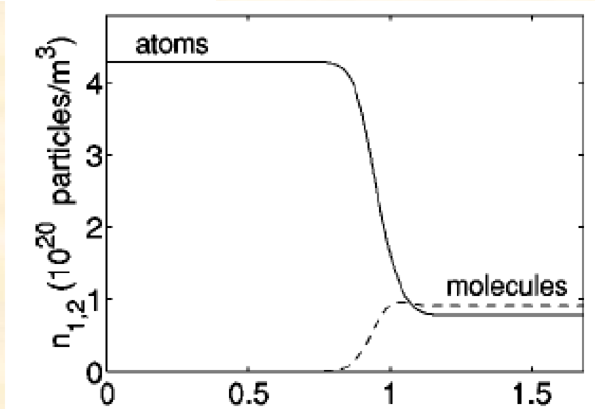




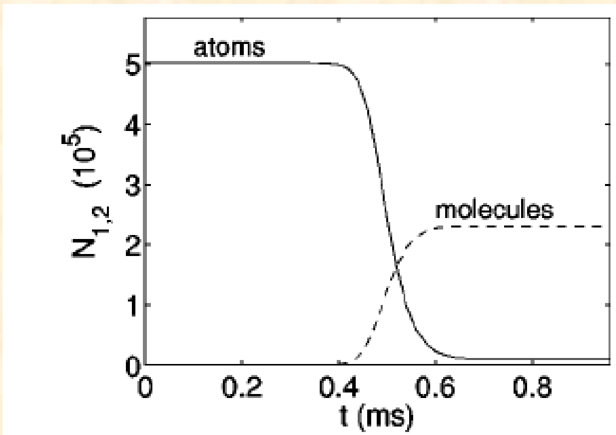
Uniform multilevel model

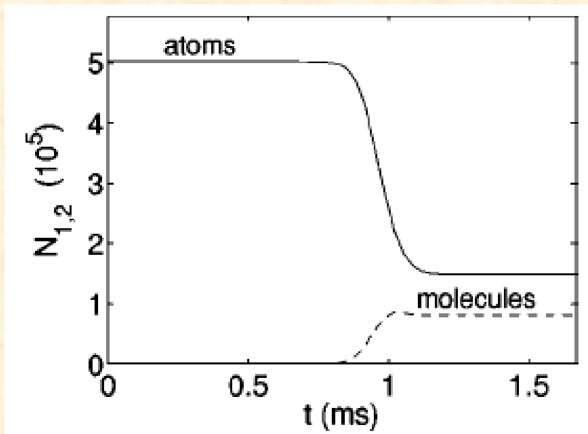
Increasing the detuning to one transition may bring the laser frequency to a resonance with respect to the nearby level





Nonuniform condensates



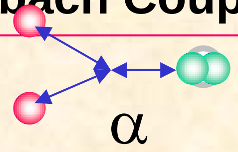


PRL **93,** 250403 (2004) PHYSICAL REVIEW LETTERS week ending 17 DECEMBER 2004

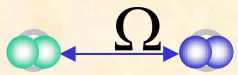
Creating a Stable Molecular Condensate Using a Generalized Raman Adiabatic Passage Scheme

Hong Y. Ling,[1] Han Pu,[2] and Brian Seaman[3]

Feshbach Couping



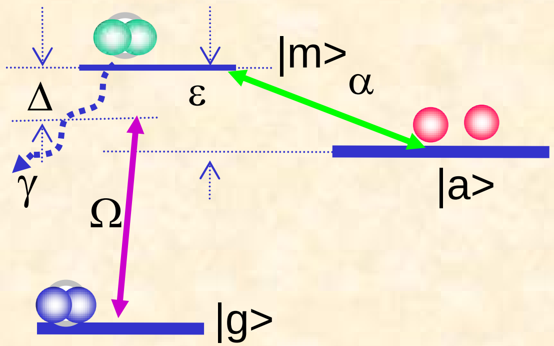
Laser Coupling



$$\frac{d\psi_a}{dt} = -i\omega_a\psi_a - i\alpha\psi_m\psi_a^*$$

$$\frac{d\psi_m}{dt} = -i\varepsilon\psi_m - i\omega_m\psi_a - i\frac{\alpha}{2}\psi_a^2 + i\frac{\Omega}{2}\psi_g - \gamma\psi_m$$

$$\frac{d\psi_g}{dt} = -i\left(\Delta+\varepsilon\right)\psi_g - i\omega_g\psi_g + i\frac{\Omega}{2}\psi_m$$

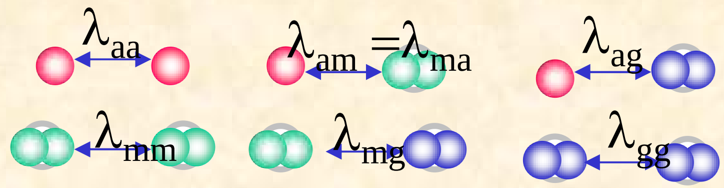


Feshbach resonance assisted stimulated adiabatic passage

where ω_i: mean-field phase shift defined as

$$\omega_i = \lambda_{ia}\left|\psi_a\right|^2 + \lambda_{im}\left|\psi_m\right|^2 + \lambda_{ig}\left|\psi_g\right|^2, i = a, m, g$$

with λ_{ij} representing inter- and intra-species collisional strengths associated to various s-wave scattering lengths.



CPT Conditions

The dark or CPT state is a stationary solution of the mean field equation in the form of

$$\psi_m(t) = 0,$$
$$\psi_a(t) = \psi_a^0 e^{i\mu_a t}, \psi_g(t) = \psi_g^0 e^{i\mu_g t}$$

where $\mu_a, \mu_g, \psi_a^0, \psi_g^0$ are determined from the coupled stationary Gross-Pitaevskii's equations

Two-Photon Resonance (Energy Conservation) Condition

$$\Delta + \varepsilon = 2\mu_a - \mu_g$$

Atom-Molecule Dark (or CPT) State

Dark State Population Distribution:

$$\psi_m^0 = 0$$

$$n_a = \left|\psi_a^0\right|^2 = \frac{2}{1+\sqrt{1+8\left(\alpha/\Omega\right)^2}}$$

$$n_g = \left|\psi_g^0\right|^2 = \frac{\sqrt{1+8\left(\alpha/\Omega\right)^2}-1}{1+\sqrt{1+8\left(\alpha/\Omega\right)^2}}$$

Dark State Condition:

$$\Delta + \varepsilon = \left(2\lambda_{ag} - \lambda_g\right)\left|\psi_g^0\right|^2 + \left(2\lambda_a - \lambda_{ag}\right)\left|\psi_a^0\right|^2$$

- Generalized two-photon resonance condition.

- As populations change in state a and g, detunings needed to be adjusted accordingly to compensate for the collisional shifts.

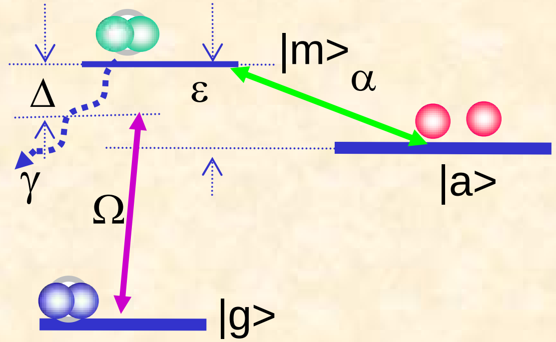
- Collisional phase shifts: Can two-photon resonance be maintained?

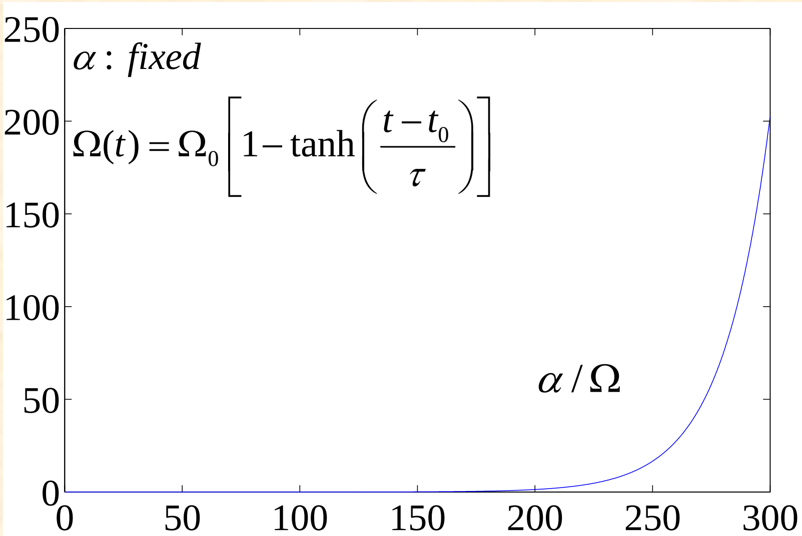
☑ **YES!** (via frequency chirp)

Atom-Molecule Chirped STIRAP

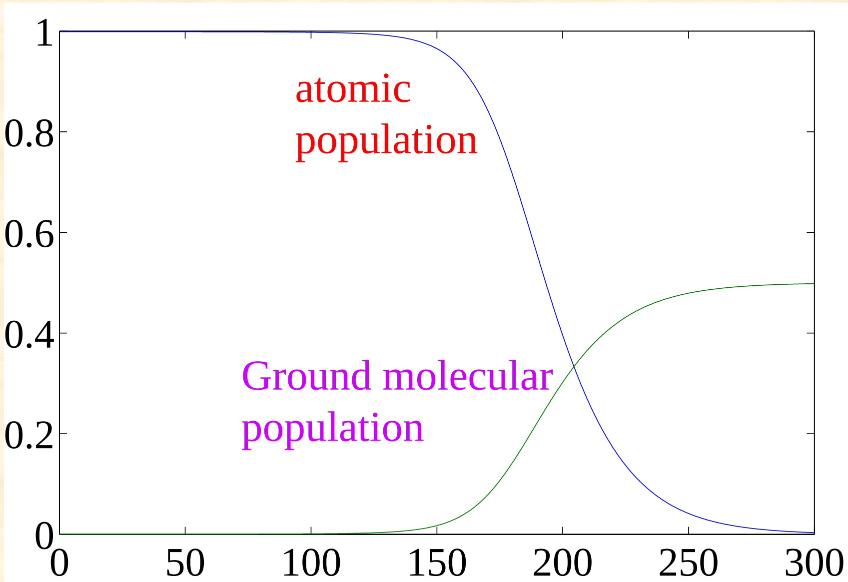
ε : *fixed*

$$\Delta = -\varepsilon + \left(2\lambda_{ag} - \lambda_g\right)\left|\psi_g^0\right|^2 + \left(2\lambda_a - \lambda_{ag}\right)\left|\psi_a^0\right|^2$$



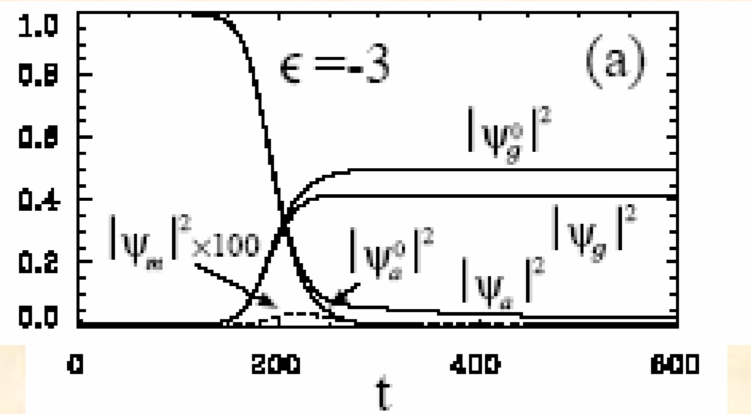


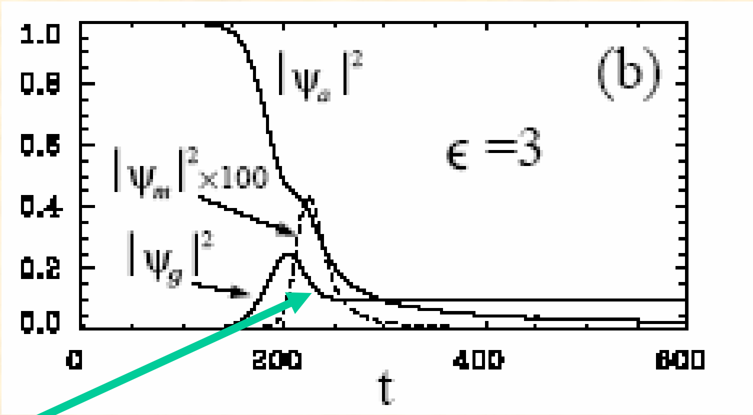
time



time

An Example of Population Dynamics





The departure from the CPT solution can be caused by

(a) Dynamical Instability
(b) Insufficient Adiabaticity

Parameters:

$n=5x10^{20}m^{-3}$, $\alpha=9.436x10^4\ s^{-1}$, $\Omega_{max}=40\alpha$, $t_0=120/\alpha$, $\tau = 40/\alpha$, $\lambda_{aa}=5.9x10^4 s^{-1}=0.652\alpha$, $\lambda_{mm}= \lambda_{gg}= 0.1875$, $\lambda_{am} = \lambda_{ag} = \lambda_{mg} = 0.1875$, t is in the unit of 0.01 ms.

H. Y. Ling, H. Pu, and B. Seaman, Phys. Rev. Lett. 93, 250403 (2004)

How to control the instability ?

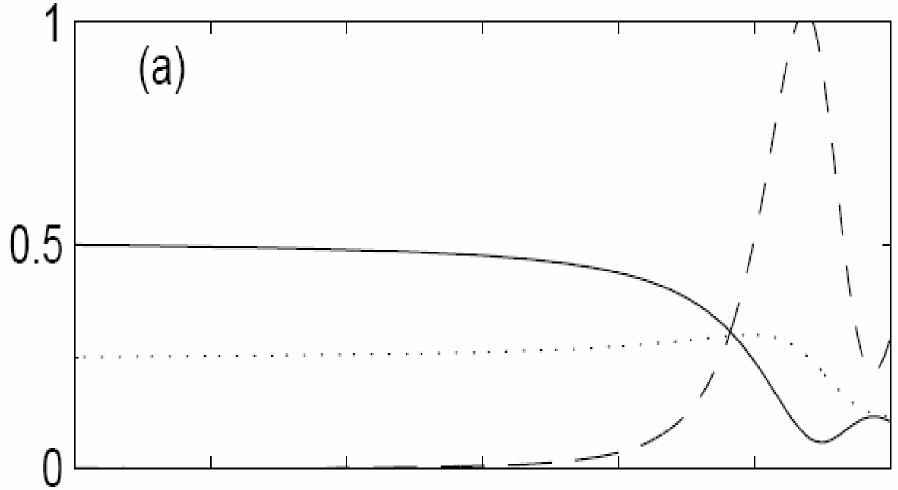
Cheng, Han, Yan, PRA **73**, 035601

The condition for the coherent population trapping state

$$\Delta = -\epsilon + (2\lambda_{ag} - \lambda_g)|\psi_g^0|^2 + (2\lambda_a - \lambda_{ag})|\psi_a^0|^2$$

$$|\psi_m^0|^2 = 0 \qquad |\psi_a^0|^2 = 1 - 2|\psi_g^0|^2 = \frac{2}{\sqrt{1+8\,(\alpha/\Omega)^2}+1}$$

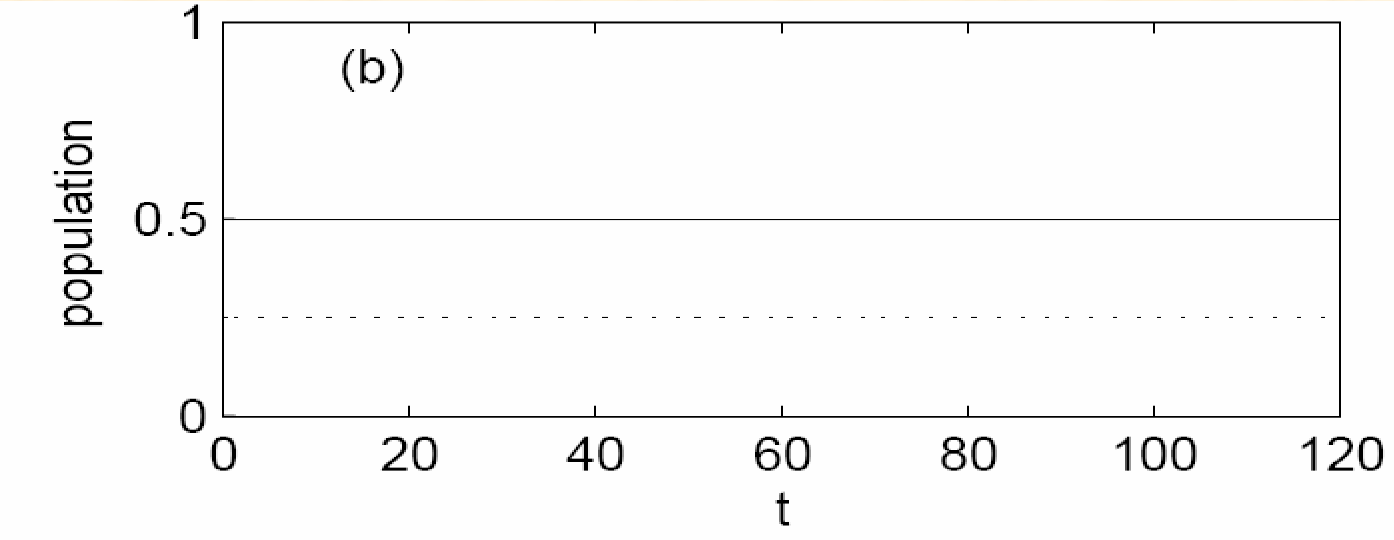
Δ is determined directly from $\Omega(t)$ corresponding to a kind of ***open loop control strategy***, suffering instability generally



We propose a kind of ***feedback control strategy*** to suppression the dynamical instability

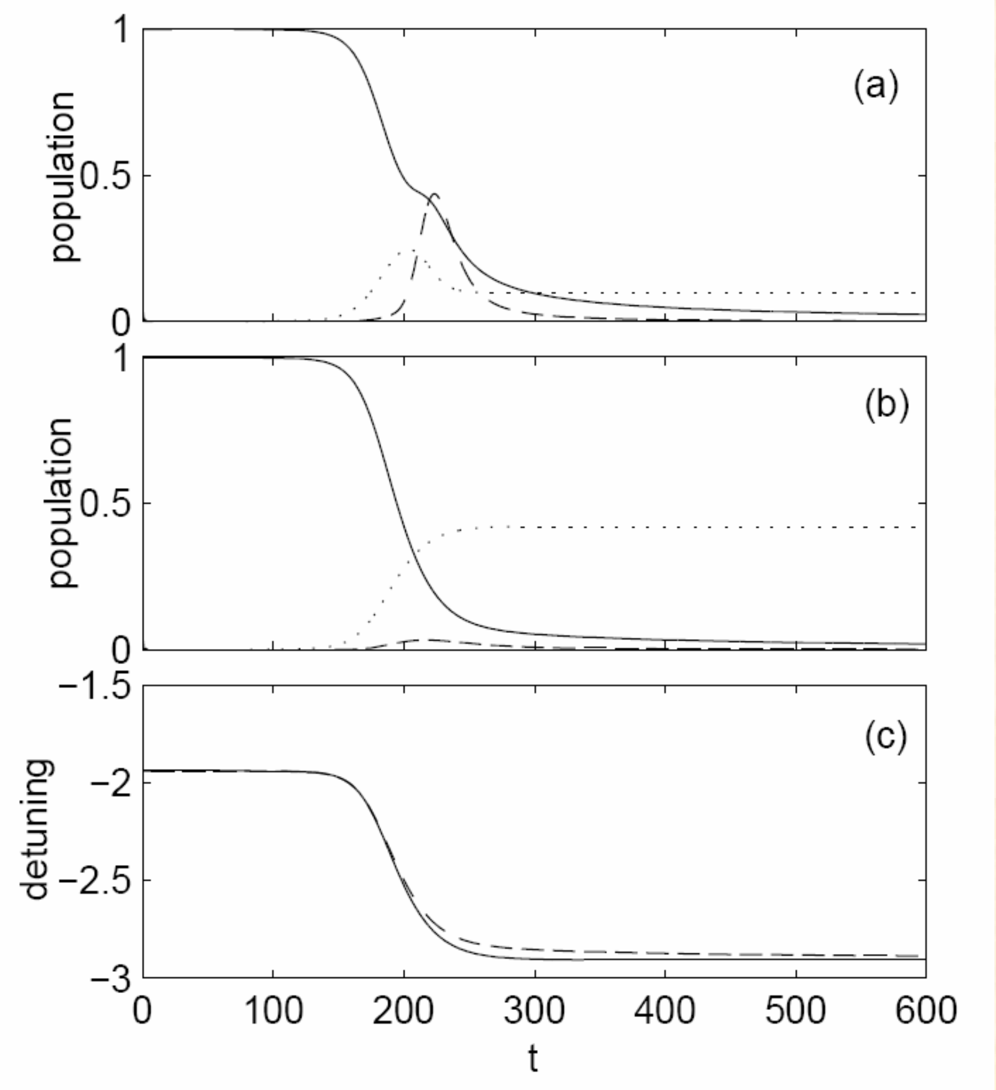
$$\Delta(t) = -\epsilon + \frac{1}{2}(2\lambda_{ag} - \lambda_g)(1 - |\psi_a(t)|^2) + (2\lambda_a - \lambda_{ag})|\psi_a(t)|^2,$$

Δ is determined from the instantaneous atomic density



This kind of adaptive laser detuning feedback control completely suppresses the dynamical instability in the CPT state !

An example

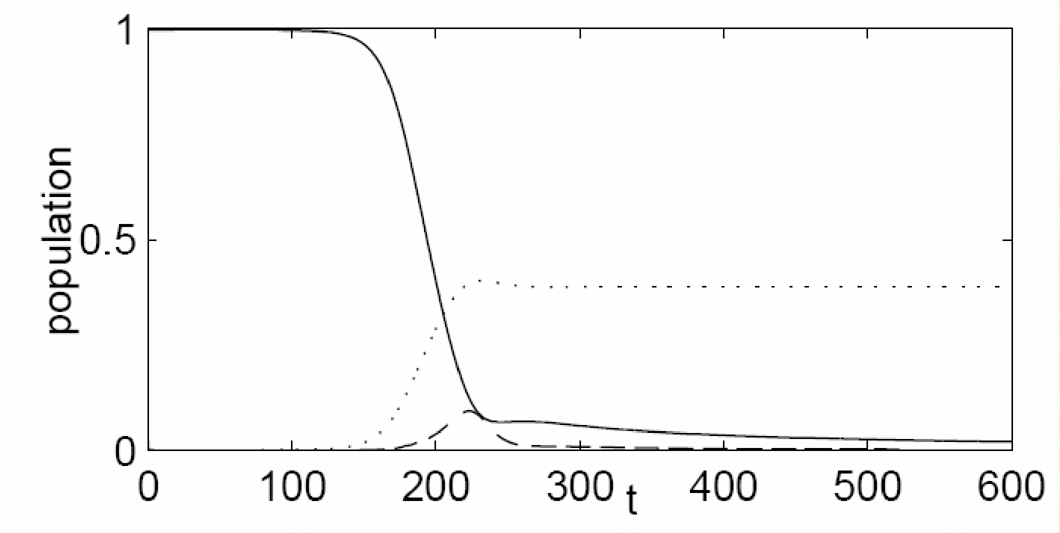


from the direct control scheme

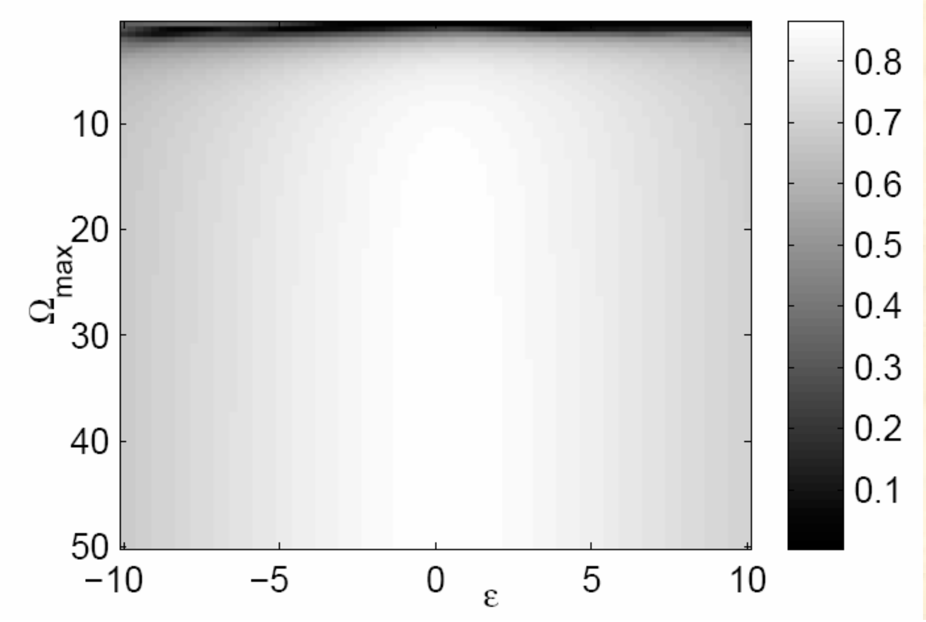
from the feedback control scheme

time-dependent laser detuning

robustness of the feedback control method



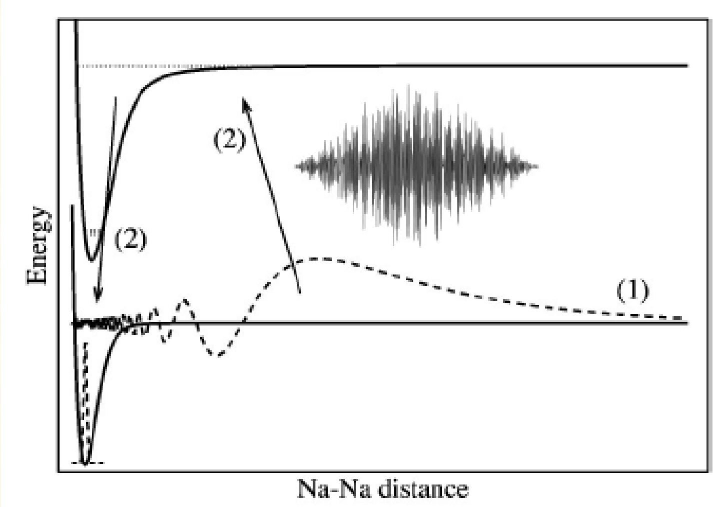
2% white-noise fluctuation $\delta\Delta(t)$



Cheng, Han, Yan, PRA **73**, 035601

using optimal control theory to create stable molecules

Koch, Palao, Kosloff, and Masnou-Seeuws, PRA 70,013402



Optimal control theory (OCT) offers the prospect of driving an atomic or molecular system to an arbitrary, desired state due to the interaction with an external field.

Optimal control has been intensely studied both theoretically and experimentally in many areas of physical chemistry.

exist a route from the last bound levels to *v*=0.

In this scheme, ground state molecules from a molecular beam with $v=0, J=0$ are excited by a CW laser ($\lambda=610$ nm) to the $A^1\Sigma_u^+$ excited state ($v'=15$). Those molecules which decay to the $v=29$ level of the ground state are excited by a second CW laser ($\lambda=540$ nm) to excited-state levels with $v'=100-140$. A third CW laser ($\lambda=595$ nm) probes the transition between these excited-state levels and the last bound levels ($v=61-65$) of the ground state.

two-step scheme for the production of ultracold molecules

(1) loosely bound molecules are created by Feshbach resonance or enhanced three-body recombination or photoassociation.

(2) a shaped laser pulse is applied to transfer the highly excited molecules to v=0

Model: radial Schrödinger equation of two channels

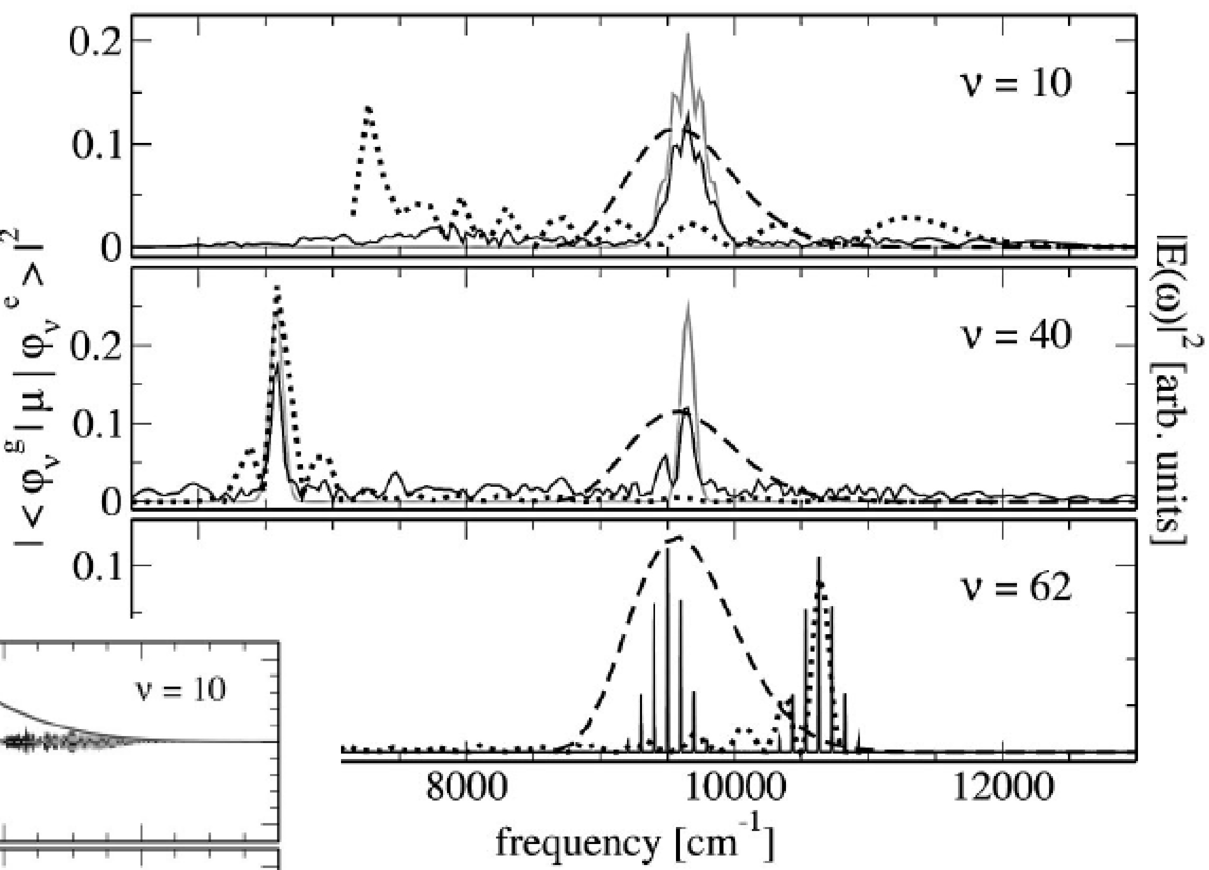
$$i\hbar\frac{\partial}{\partial t}\varphi(R,t)=\hat{\mathbf{H}}\varphi(R,t), \qquad \varphi=\begin{pmatrix}\varphi_g\\ \varphi_e\end{pmatrix}$$

$$\hat{\mathbf{H}}=\begin{pmatrix}\hat{\mathbf{T}}+\hat{\mathbf{V}}_g & 0\\ 0 & \hat{\mathbf{T}}+\hat{\mathbf{V}}_e\end{pmatrix}+\begin{pmatrix}0 & \hat{\mu}\varepsilon(t)\\ \hat{\mu}\varepsilon(t)^* & 0\end{pmatrix}$$

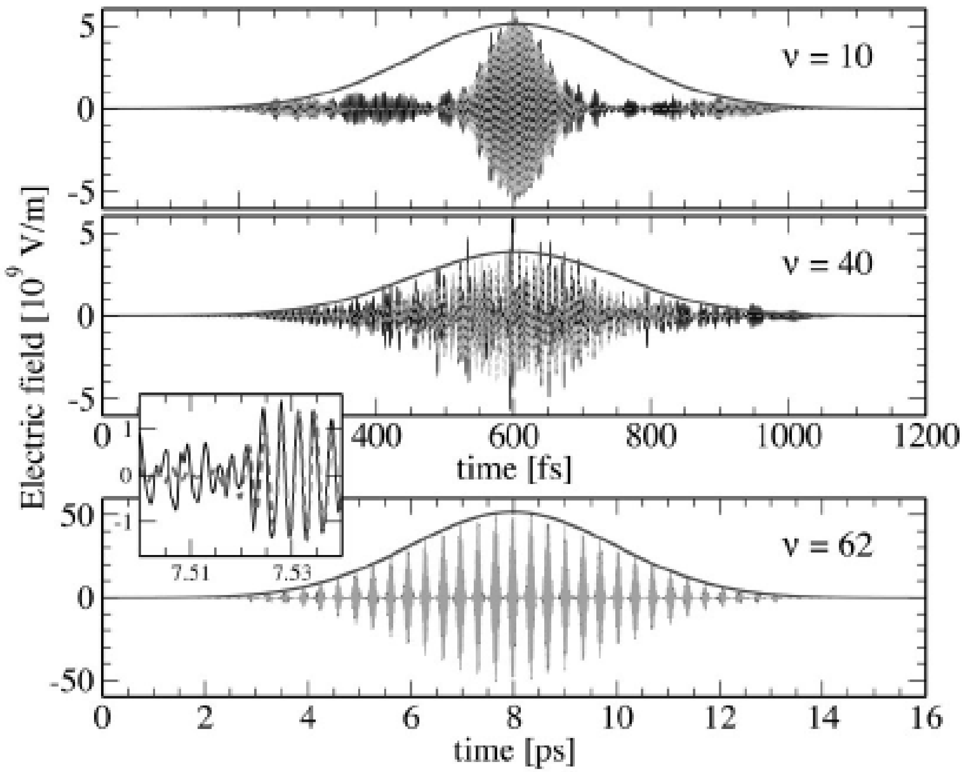
Finding a field by maximizing $F=\left|\langle\varphi_i|\hat{\mathbf{U}}^+(T,0;\varepsilon)|\varphi_f\rangle\right|^2$

Or the optimal field is found by minimization of $J=-F+\int_0^T g(\varepsilon,\varphi)dt$

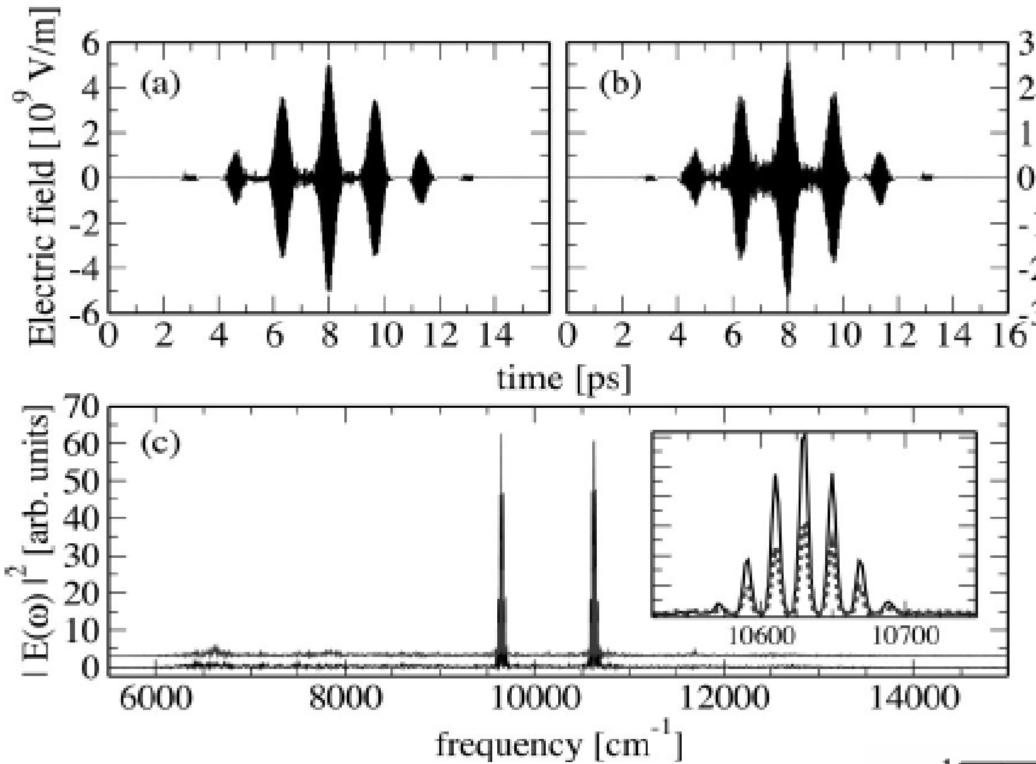
$$g(\varepsilon,\varphi)=g(\varepsilon)=\frac{\alpha}{S(t)}[\varepsilon(t)-\tilde{\varepsilon}(t)]^2$$



ν = 10
ν = 40
ν = 62
0.2
0.1
0
|< φ_ν^g | μ | φ_ν^e >|^2
|E(ω)|^2 [arb. units]
8000
10000
12000
frequency [cm^-1]

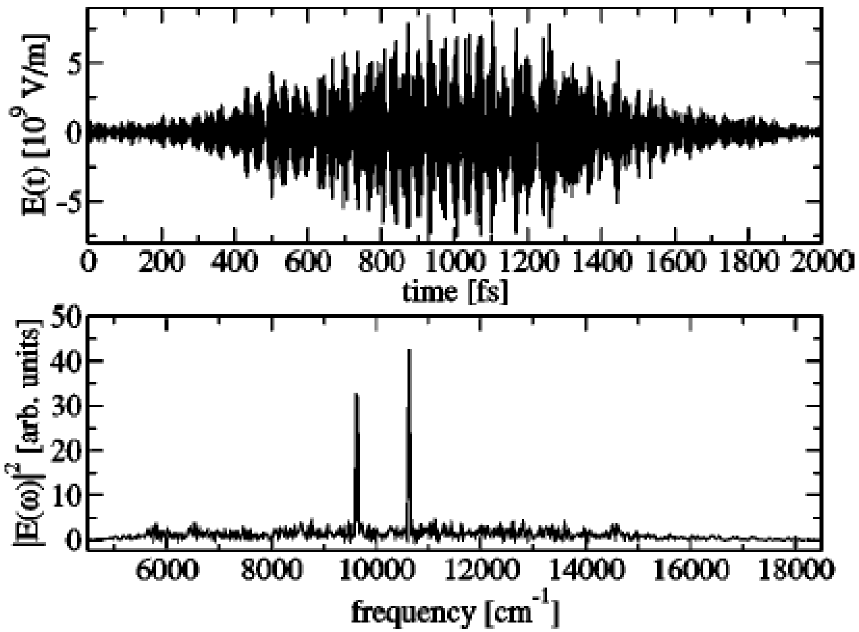


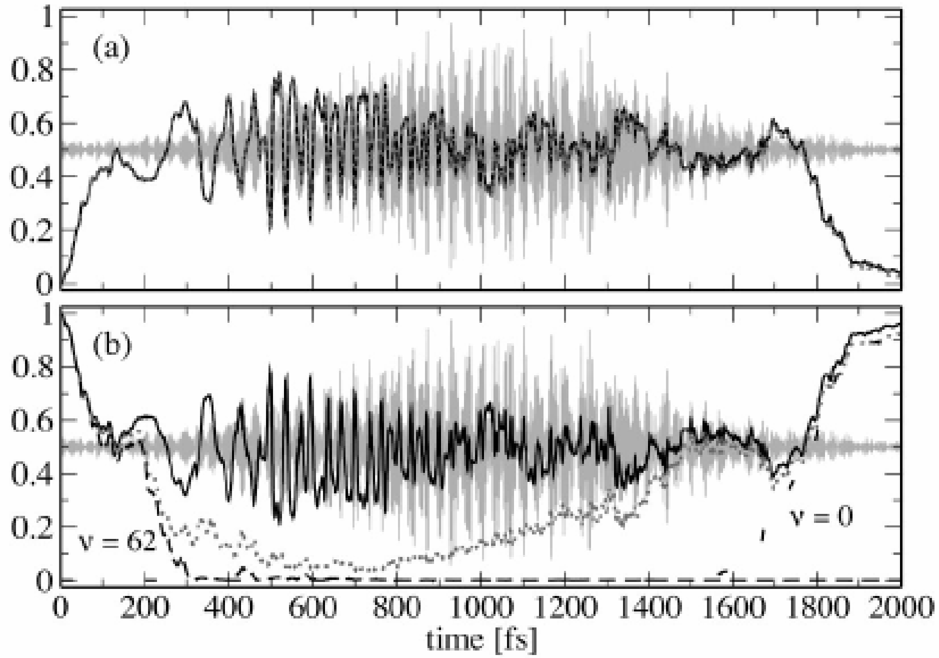
ν = 10
ν = 40
ν = 62
Electric field [10^9 V/m]
5
0
-5
50
-50
0
400
600
800
1000
1200
time [fs]
2
4
6
8
10
12
14
16
time [ps]
1
-1
7.51
7.53



$E_0 = 0.01$ a.u. $\mathcal{E}_{\text{pulse}} = 16$ mJ

$E_0 = 0.005$ a.u. $\mathcal{E}_{\text{pulse}} = 4$ mJ





Superchemistry and coherent atom-trimer conversion

Superchemistry:

Bose-enhanced nonlinear coherent chemistry at zero

- **《SC: Dynamics of Coupled Atomic and Molecular Bose Condensates 》**
- ***D. J. Heinzen*, et al. PRL 84, 5029 (2000)**
- Classical Arrhenius chemical kinetics: not depends on product numbers & go to ***zero*** <u>at low temperatures</u> (Arrhenius law or Boltzmann kenetics).
- **Low-temperature quantum effects: like *super-conductivity*!**

- Largely Bose enhanced A-M conversion at zero temperature, induced by a weak photoassociation (PA) light or FR…

coherent oscillations of A-M species;

long-time molecular damping and atomic revivals; … …

- not observed so far …

The attitude of chemists on superchemistry?

- **“External fields (at subkelvin temperatures) may therefore be used to … stimulate forbidden electric transitions, … or tune Feshbach resonances that enhance chemical reactivity.”**

- **“Possibilities of chemical research with cold and ultracold molecules are boundless and enticing. Particularly appealing are the prospects to explore Bose-enhanced chemistry. Selectivity of chemical reactions and branching ratios of photodissociation may be greatly enhanced in a MBEC due to collective dynamics of condensed molecules.”**

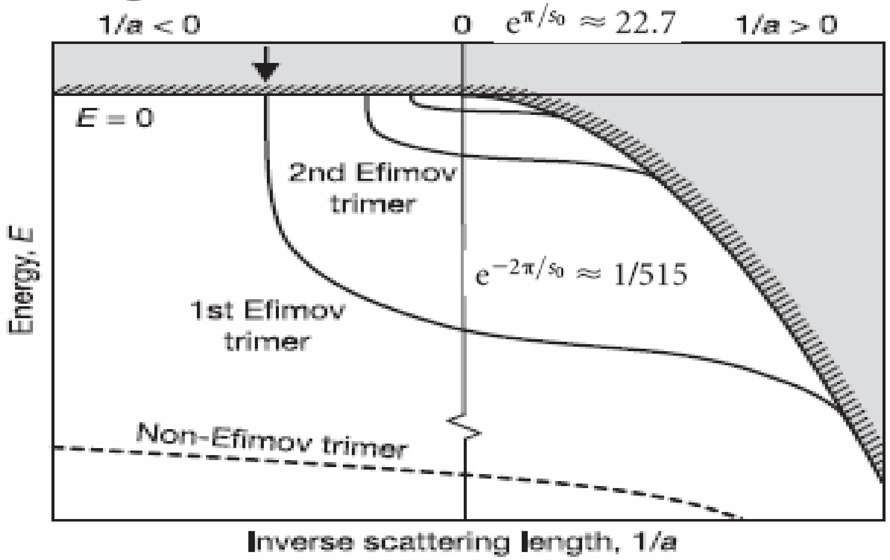
- **“Experiments with ultracold molecules will test the applicability limits of conventional molecular dynamics theories as it does not account for quantum effects in molecular interactions.”**

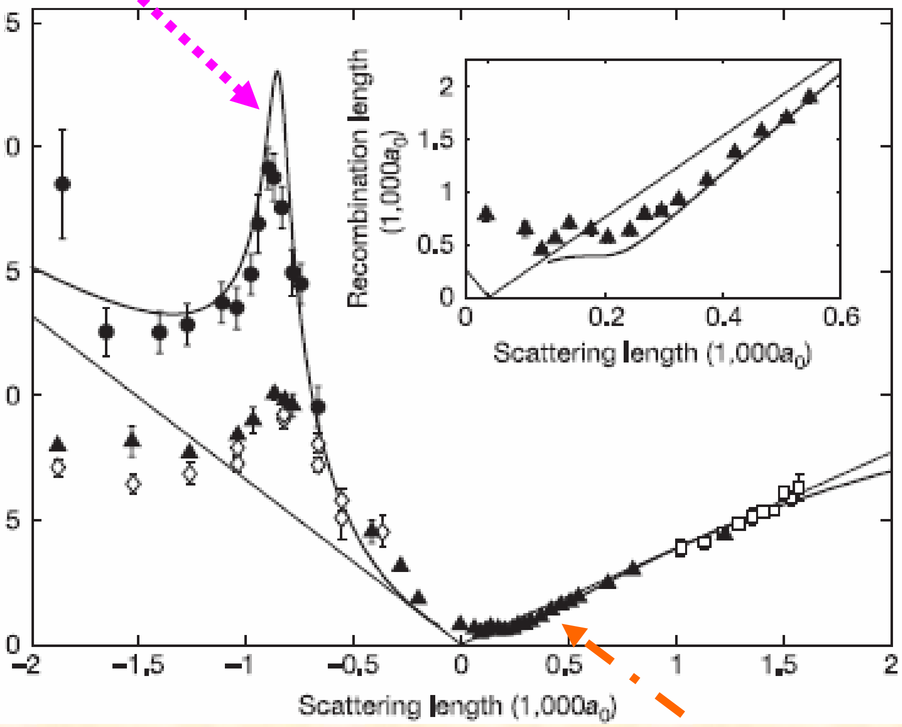
- *R. V. Krems*, **International Reviews in Physical Chemistry 24 (2005) 99–118.**

Evidence for Efimov quantum states in an ultracold gas of caesium atoms

Efimov resonance!

T. Kraemer[1], M. Mark[1], P. Waldburger[1], J. G. Danzl[1], C. Chin[1,2], B. Engeser[1], A. D. Lange[1], K. Pilch[1], A. Jaakkola[1], H.-C. Nägerl[1] & R. Grimm[1,3]





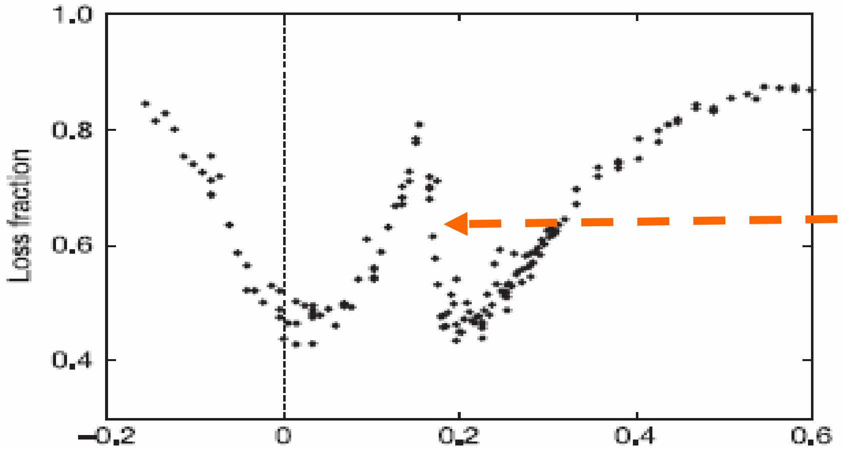


Figure 3 | Atom loss for small scattering lengths.

Atom-Dimer Feshbach Resonance!

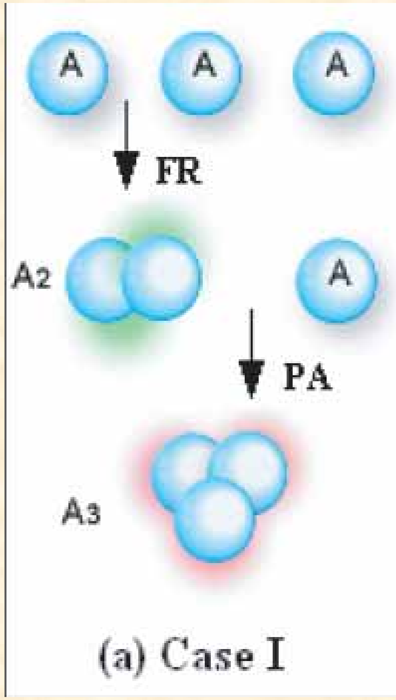
Two regimes:

Strong attractive atom-atom-atom;

Weak repulsive atom-dimer->trimer

Coherent atom-trimer conversion in repulsive BECs

Jing, Cheng, Meystre, quant-ph/0703247



(a) Case I

$$\hat{\mathcal{H}}_I = -\hbar \int dr \Big\{ \sum_{i,j} \chi'_{ij} \hat{\psi}_i^\dagger(r) \hat{\psi}_j^\dagger(r) \hat{\psi}_j(r) \hat{\psi}_i(r) + \delta \hat{\psi}_d^\dagger(r) \hat{\psi}_d(r) + \lambda'_1 \left[\hat{\psi}_d^\dagger(r) \hat{\psi}_a(r) \hat{\psi}_a(r) + h.c. \right] + (\Delta + \delta) \hat{\psi}_g^\dagger(r) \hat{\psi}_g(r) - \lambda'_2 \left[\hat{\psi}_d^\dagger \hat{\psi}_a^\dagger \hat{\psi}_g(r) + h.c. \right] \Big\}$$

$$\frac{d\psi_a}{dt} = 2i\tilde{\chi}_a \psi_a + 2i\lambda_1 \psi_d \psi_a^* - i\lambda_2 \psi_d^* \psi_g,$$

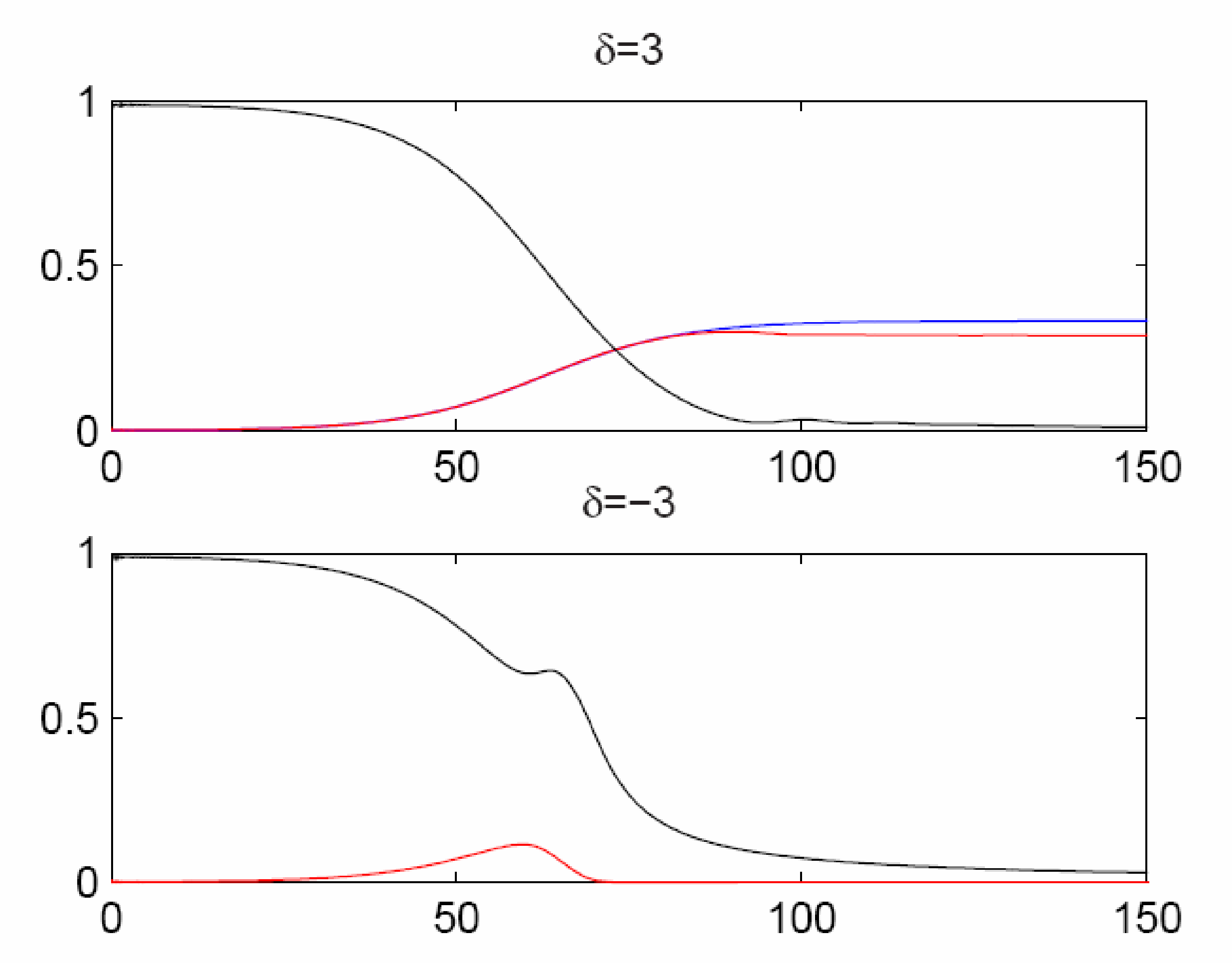
$$\frac{d\psi_d}{dt} = -\gamma \psi_d + i\delta \psi_d + 2i\tilde{\chi}_d \psi_d + i\lambda_1 \psi_a^2 - i\lambda_2 \psi_a^* \psi_g,$$

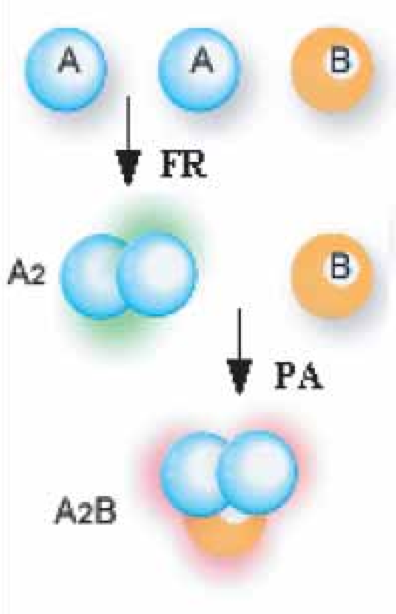
$$\frac{d\psi_g}{dt} = 2i\tilde{\chi}_g \psi_g + i(\Delta + \delta)\psi_g - i\lambda_2 \psi_d \psi_a,$$

coherent population trapping (CPT) state

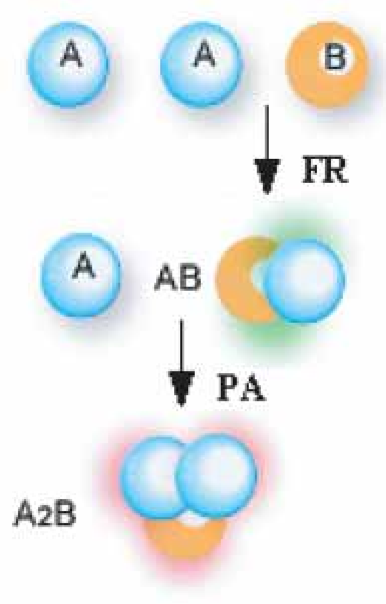
$$\bar{N}_a^0 = [1 + 3(\lambda_1/\lambda_2)^2]^{-1} = 1 - 3\bar{N}_g^0 \quad \bar{N}_d^0 = 0$$

$$\Delta_I = -\delta + (6\chi_{ag} - \chi_g)N_g^0 + (6\chi_a - 2\chi_{ag})N_a^0$$





(b) Case II



(c) Case III

$$\hat{\mathcal{H}} = -\hbar \int dr \Big\{ \sum_{i,j} \chi_{ij} \hat{\psi}_i^\dagger(r) \hat{\psi}_j^\dagger(r) \hat{\psi}_j(r) \hat{\psi}_i(r) + \delta \hat{\psi}_d^\dagger(r) \hat{\psi}_d(r) + \lambda_1' \left[\hat{\psi}_d^\dagger(r) \hat{\psi}_a(r) \hat{\psi}_a(r) + h.c. \right] + (\Delta + \delta) \hat{\psi}_g^\dagger(r) \hat{\psi}_g(r) - \Omega_1' \left[\hat{\psi}_g^\dagger(r) \hat{\psi}_d \hat{\psi}_b + h.c. \right] \Big\}$$

$$\begin{aligned}
\frac{d\psi_a}{dt} &= 2in \sum_j \chi_{aj} |\psi_j|^2 \psi_a + 2i\lambda_1 \psi_d \psi_a^*, \\
\frac{d\psi_b}{dt} &= 2in \sum_j \chi_{bj} |\psi_j|^2 \psi_b - i\Omega_1 \psi_d^* \psi_g, \\
\frac{d\psi_d}{dt} &= -(\gamma - i\delta)\psi_d + 2in \sum_j \chi_{dj} |\psi_j|^2 \chi_d \psi_d + i\lambda_1 \psi_a^2 \\
&\quad - i\Omega_1 \psi_b^* \psi_g, \qquad (1) \\
\frac{d\psi_g}{dt} &= 2in \sum_j \chi_{gj} |\psi_j|^2 \psi_g + i(\Delta + \delta)\psi_g - i\Omega_1 \psi_d \psi_b,
\end{aligned}$$

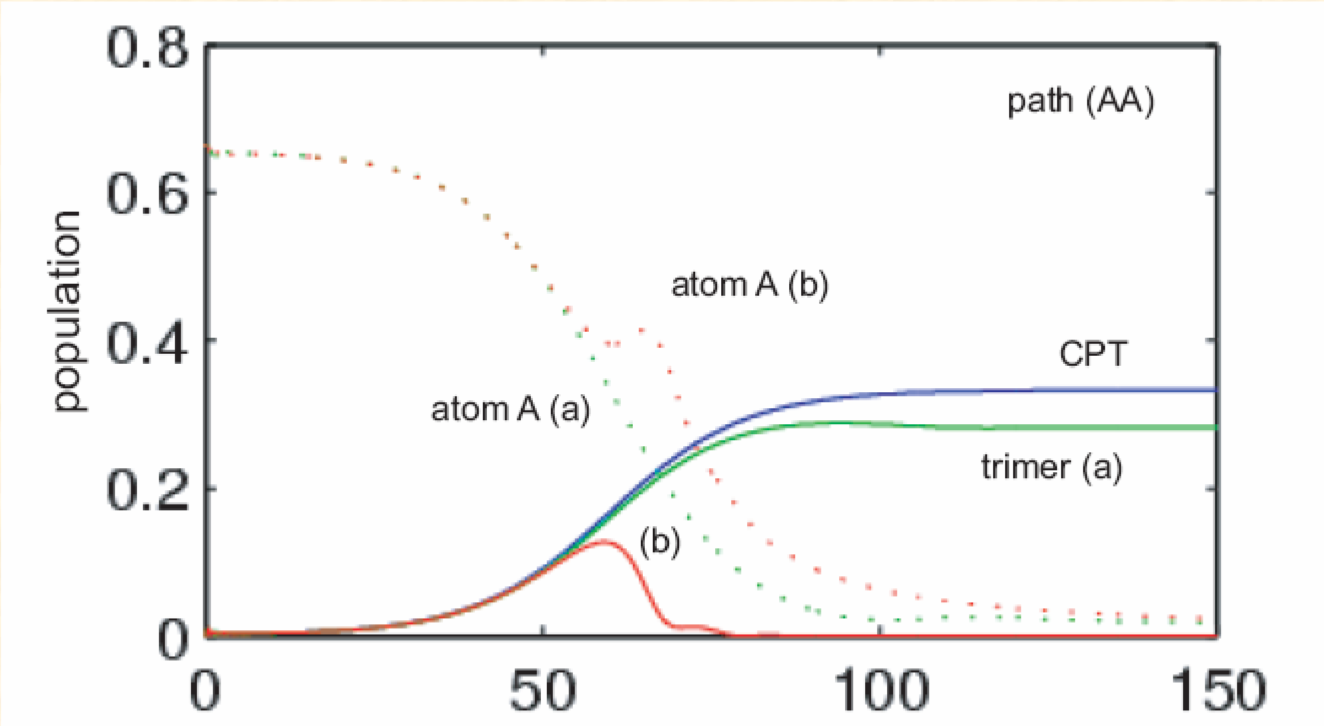
Case II, path AA

$$\Delta = -\delta + 2(2\chi_{ag} + \chi_{bg} - \chi_{gg})nN_{g,s} + (4\chi_{aa} - 2\chi_{ag} + 4\chi_{ab} + \chi_{bb} - \chi_{bg})nN_{a,s}$$

$$N_{g,s} = \frac{1}{3}\left(\frac{k(\lambda_i/\Omega_i)^2}{1 + k(\lambda_i/\Omega_i)^2}\right)$$

CPT state, for matched atom numbers $N_a=2N_b$

$i = 1$ and $k = 4$

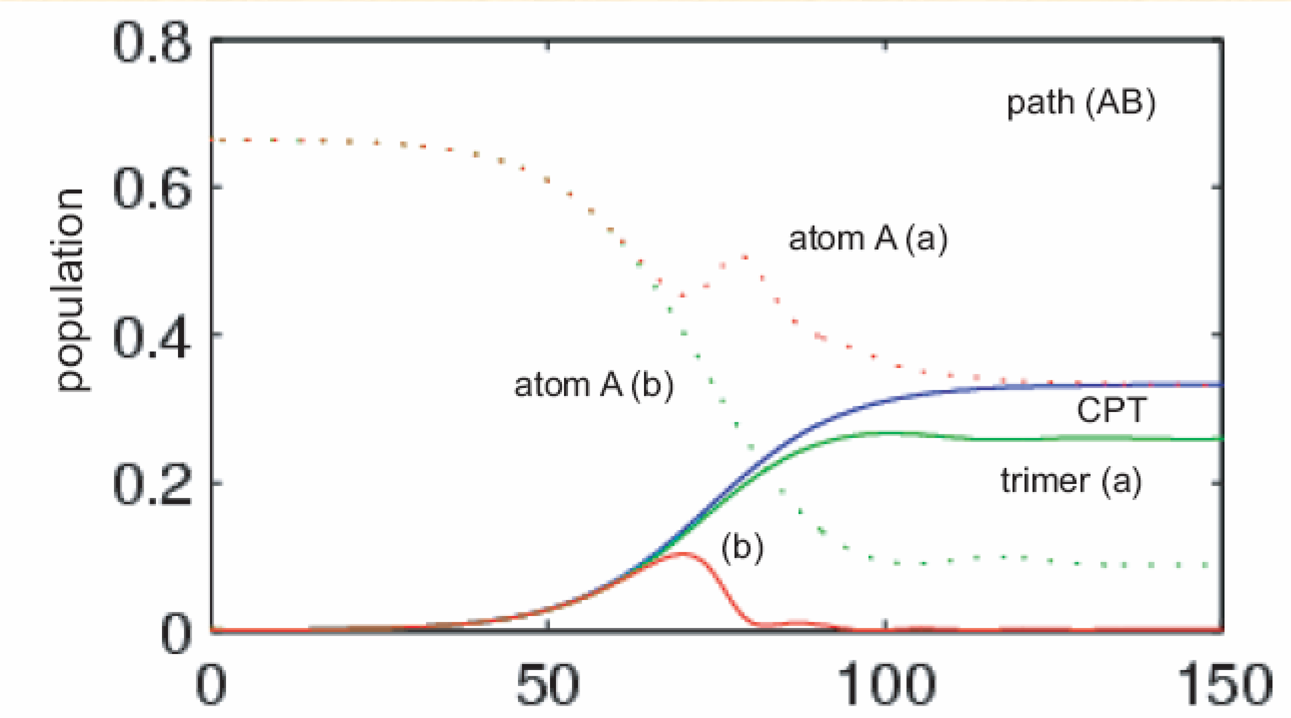


Case III, path AB

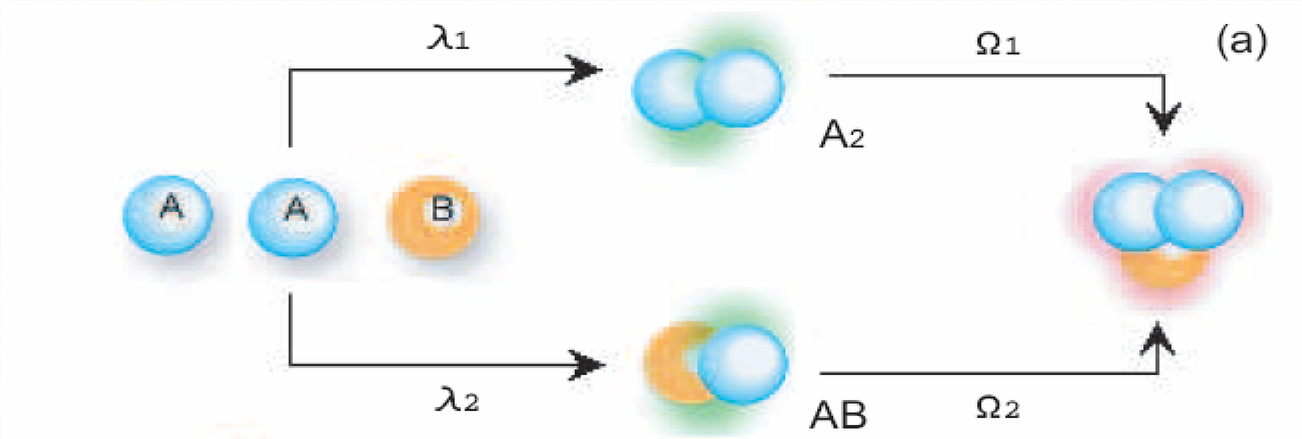
$$\Delta = -\delta + 2(2\chi_{ag} + \chi_{bg} - \chi_{gg})nN_{g,s} + (4\chi_{aa} - 2\chi_{ag} + 4\chi_{ab} + \chi_{bb} - \chi_{bg})nN_{a,s}$$

$$N_{g,s} = \frac{1}{3}\left(\frac{k(\lambda_i/\Omega_i)^2}{1+k(\lambda_i/\Omega_i)^2}\right)$$

CPT state, for matched atom numbers $N_a=2N_b$ $i = 2$ and $k = 1$



the coexistence of the two channels provides considerable additional flexibility in approaching the ideal CPT value for trimer formation

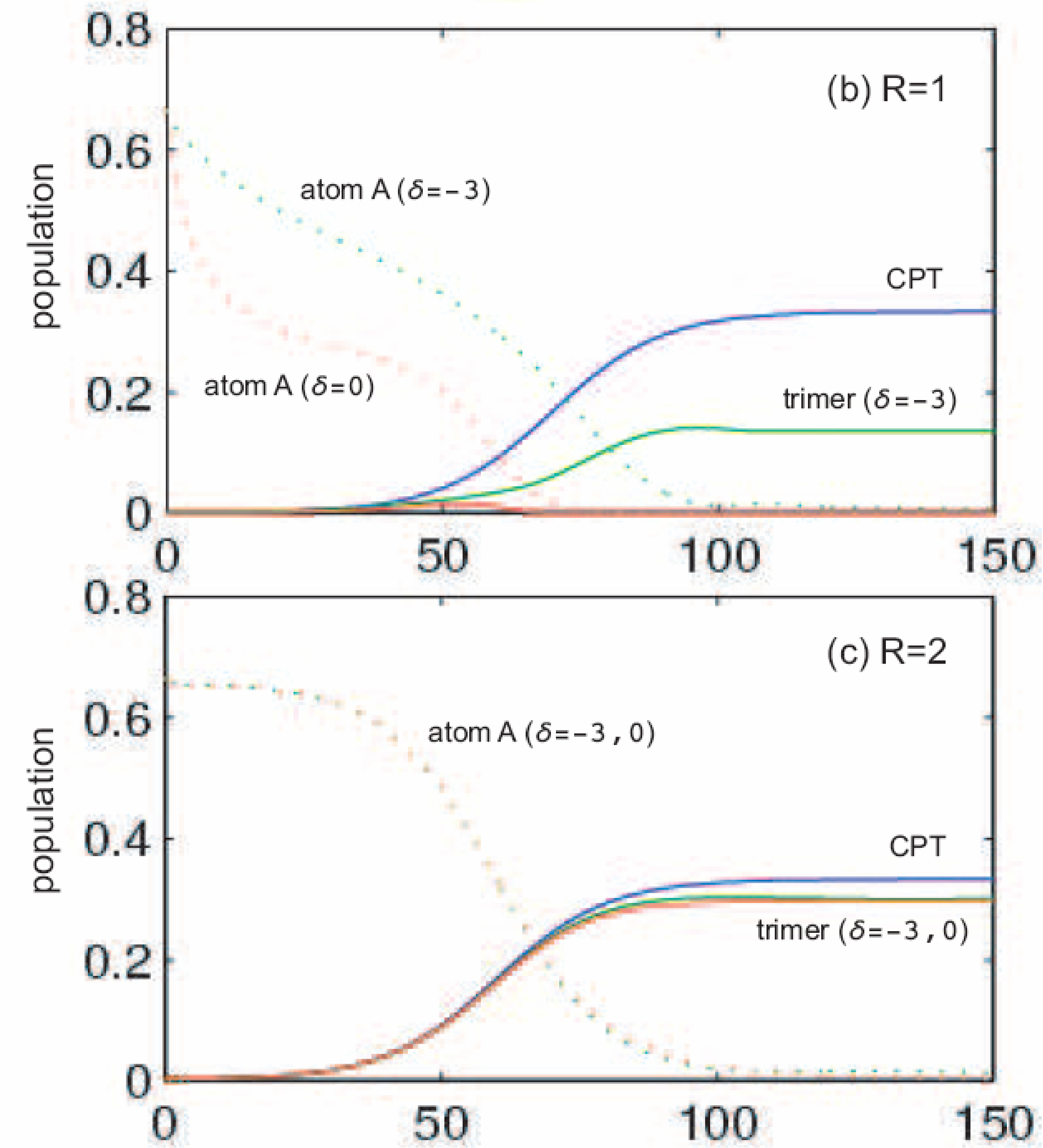


$$\eta_l = \lambda_l/\Omega_l, l = 1, 2, \qquad R = \eta_2/\eta_1$$

$$N_{g,s} = \frac{(\lambda_1/\Omega_1)(\lambda_2/\Omega_2)^2}{\lambda_1/\Omega_1 + \lambda_2/\Omega_2 + 3(\lambda_1/\Omega_1)(\lambda_2/\Omega_2)^2}$$

$$\begin{aligned}\Delta =& -\delta + 2(2\chi_{ag} + \chi_{bg} - \chi_{gg})nN_{g,s} \\ &+ (4\chi_{aa} - 2\chi_{ag} + 4\chi_{ab} + \chi_{bb} - \chi_{bg})nN_{a,s}\end{aligned}$$

$N_a = 2N_b$



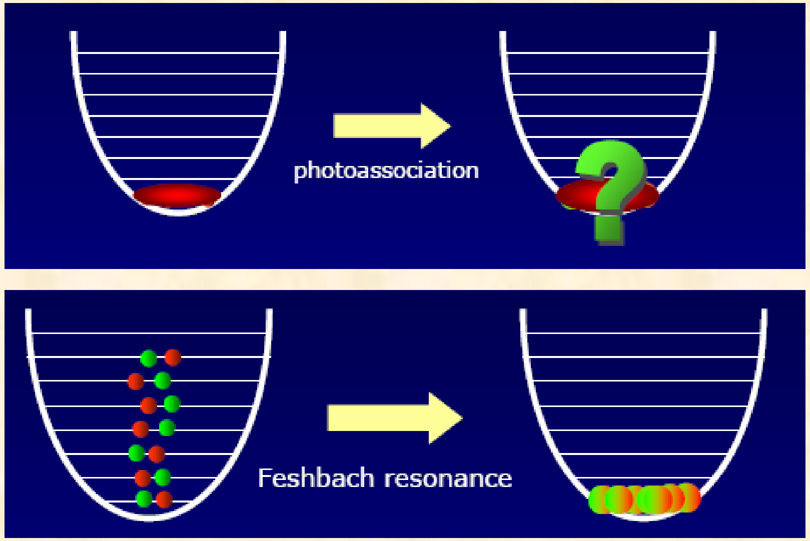
0.8
0.6
0.4
0.2
0
population
(b) R=1
atom A (δ=−3)
CPT
atom A (δ=0)
trimer (δ=−3)
0
50
100
150
0.8
0.6
0.4
0.2
0
population
(c) R=2
atom A (δ=−3 , 0)
CPT
trimer (δ=−3 , 0)
0
50
100
150

Quantum dynamics of a molecular matter-wave amplifier

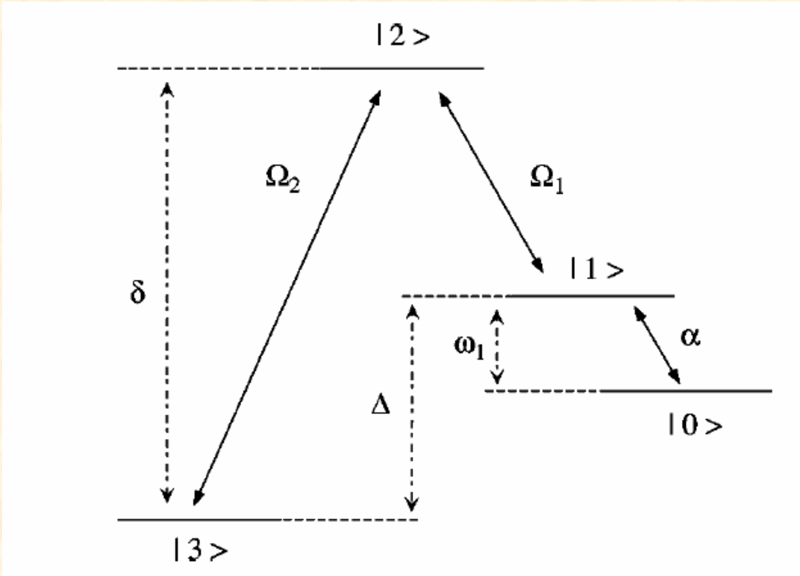
分子物质波放大器中的量子涨落

两个动机

超冷分子的量子统计



高效产生稳定分子凝聚体

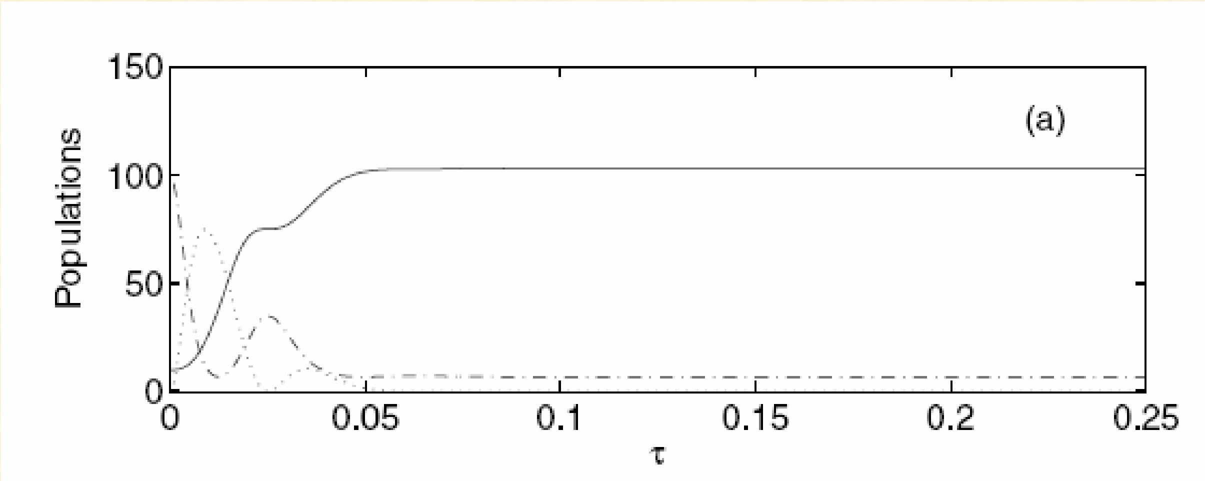


quantum statistics in atom-molecule BECs，fluctuation, correlation, entanglement, number statistics
Meystre, Drummond, Pu Han, …

two-photon Raman PA, STIRAP, Feshbach stimulated Raman photoproduction
Mackie, Drummond, Ling HY,…

分子物质波放大器 (molecular matter-wave amplifier)
模型 ——Search & Meystre, PRL 93, 140405 (2004)

特点: 缔合—经典Pump—量子**Dump**，
大失谐，坏腔，量子耗散=>单向放大基态分子数目



平均场求解主方程

$$\dot{\rho} = -i[H_{01} + \delta \hat{b}_3^\dagger \hat{b}_3, \rho] + \kappa|\beta|^2(\hat{b}_1 \hat{b}_3^\dagger \rho \hat{b}_3 \hat{b}_1^\dagger - \hat{b}_3 \hat{b}_1^\dagger \hat{b}_1 \hat{b}_3^\dagger \rho + \text{H.c.})$$

分子物质波放大器的量子统计：粒子数涨落，二阶关联函数——三个物质波场之间的自相关和互相关

方法：正P表示，随机微分方程——短时有效，长时发散

量子迹，MC波函数——任意时间，粒子数目限制

Lindblad形式的主方程
$$\dot{\rho} = i[\rho, H_s] + L_{relax}(\rho)$$

$$L_{relax}(\rho) = -\frac{1}{2}(\hat{C}^\dagger \hat{C}\rho + \rho\hat{C}^\dagger \hat{C} + 2\hat{C}^\dagger \rho\hat{C})$$
$$\hat{C} = \sqrt{2\kappa|\beta|^2}\hat{b}_1\hat{b}_3^\dagger$$

波函数表示
$$|\phi(t)\rangle = \sum_{n=0}^{M}\sum_{m=0}^{M-n} c_{nm}(t)|2n\rangle_0|m\rangle_1|M-n-m\rangle_3$$

$$|\phi(t+\delta t)\rangle = \begin{cases} \frac{(1-iH\delta t)|\phi(t)\rangle}{\sqrt{1-\delta p}}, & \text{with probability } 1-\delta p \\ \frac{\hat{C}|\phi(t)\rangle}{\sqrt{\delta p/\delta t}}, & \text{with probability } \delta p \end{cases}$$

$$\delta p = \delta t\langle\phi(t)|\hat{C}^\dagger\hat{C}|\phi(t)\rangle$$
$$H = H_s - i\hat{C}^\dagger\hat{C}/2$$

布居数 $n_0(t) = \mathrm{Tr}[\hat{b}_0^\dagger \hat{b}_0 \rho] = \frac{1}{N}\sum_{k=0}^{N}\left\{\sum_{n=0}^{M}\sum_{m=0}^{M-n} 2n|c_{nm}^{\{k\}}|^2\right\}$ $n_1(t)$ $n_3(t)$

单模二阶关联，自相关 $G_j^{(2)}(t) = \mathrm{Tr}[\hat{b}_j^\dagger \hat{b}_j^\dagger \hat{b}_j \hat{b}_j \rho]$

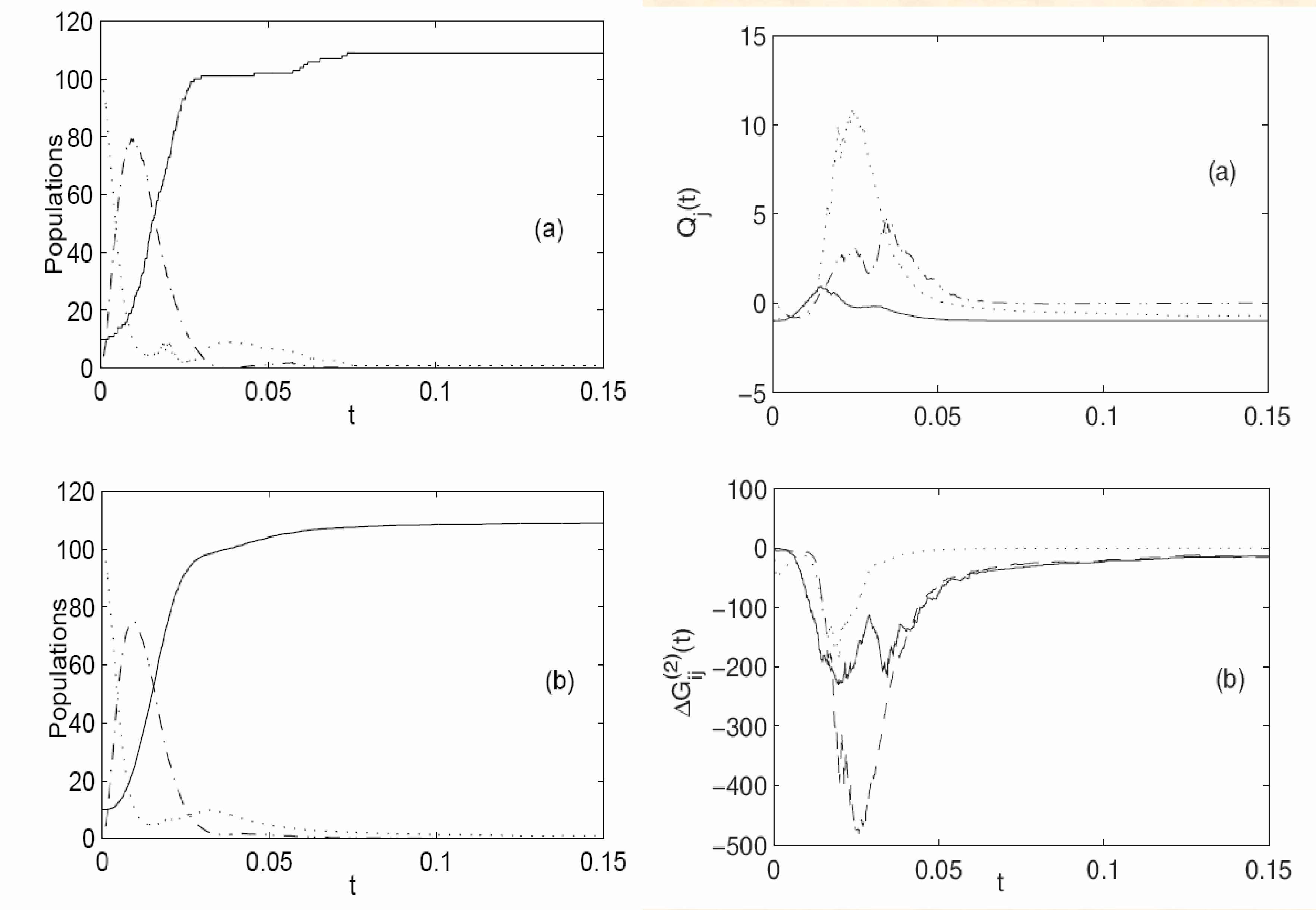
双模二阶关联，互相关 $G_{ij}^{(2)}(t) = \mathrm{Tr}[\hat{b}_i^\dagger \hat{b}_j^\dagger \hat{b}_j \hat{b}_i \rho]$

$G_j^{(2)} < n_j^2$ →亚泊松统计 $Q_j(t) = \frac{G_j^{(2)}(t) - n_j^2(t)}{n_j(t)}$

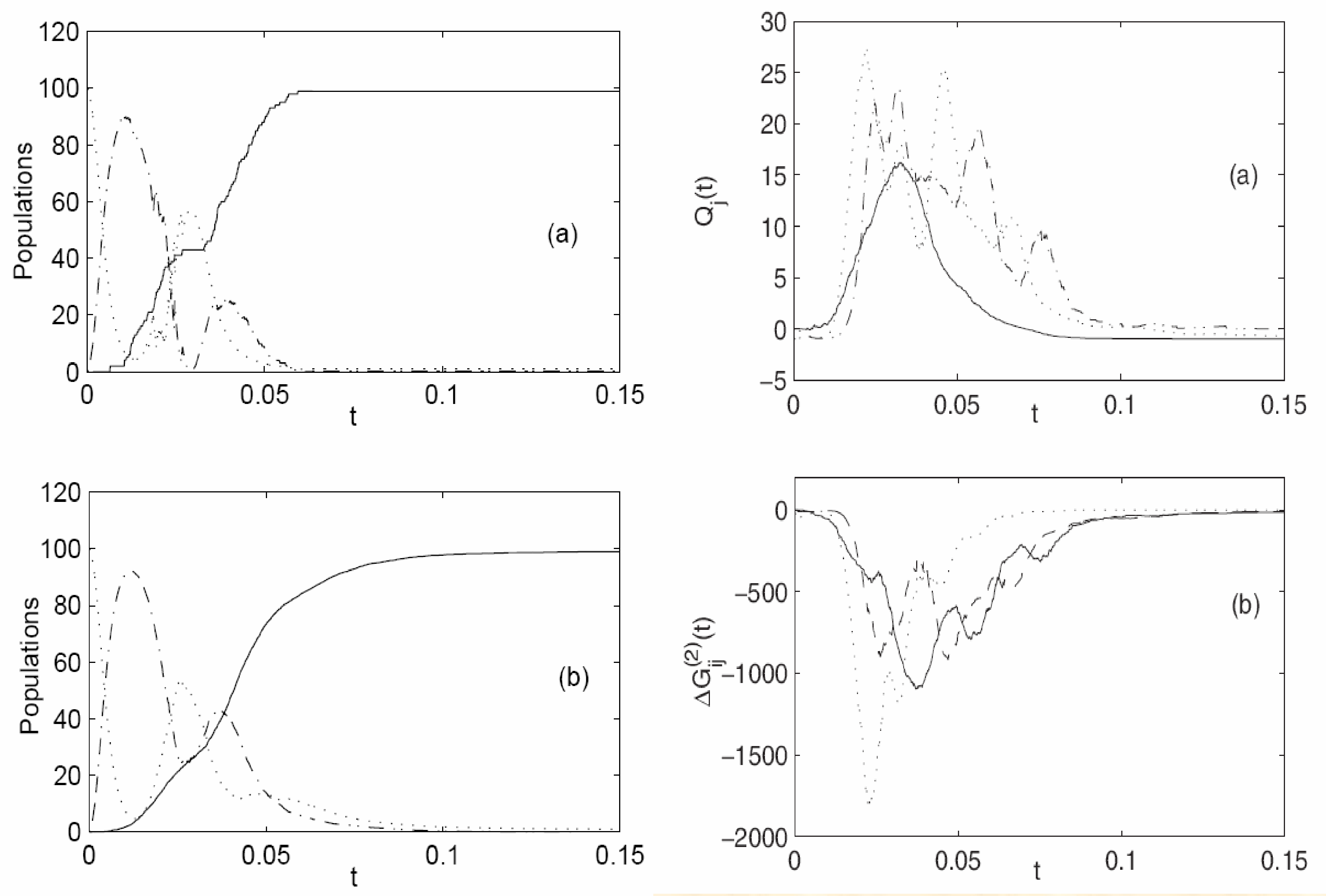
$G_{ij}^{(2)} > \sqrt{G_i^{(2)} G_j^{(2)}}$ →双模非经典关联，bunching

分子物质波的最终演化定态是亚泊松统计
任意两个物质波场之间的关联函数显现反聚束 性质

$|\phi(0)\rangle = |200\rangle_0 |0\rangle_1 |10\rangle_3$ $\quad |\phi_f\rangle = |0\rangle_0 |0\rangle_1 |110\rangle_3$



$$|\phi(0)\rangle = |200\rangle_0 |0\rangle_1 |0\rangle_3$$



Cheng, Yan, PRA **75**, 033614


Thanks !