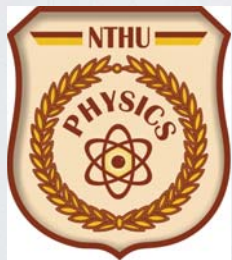# ULTRACOLD MOLECULE

劉怡維
清華大學物理系
2015
AMO Summer School

Ultracold molecule

Building **Quantum** **computer**　　**Super**chemistry
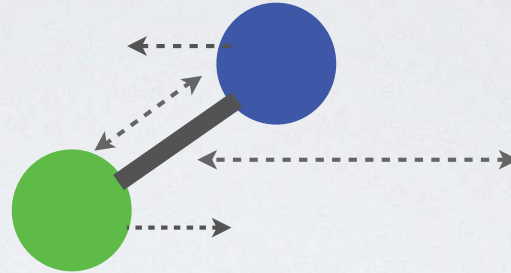
Coherent control chemical reaction

a a a a a a a

fundamental **constant** variation? Symmetry **violation**
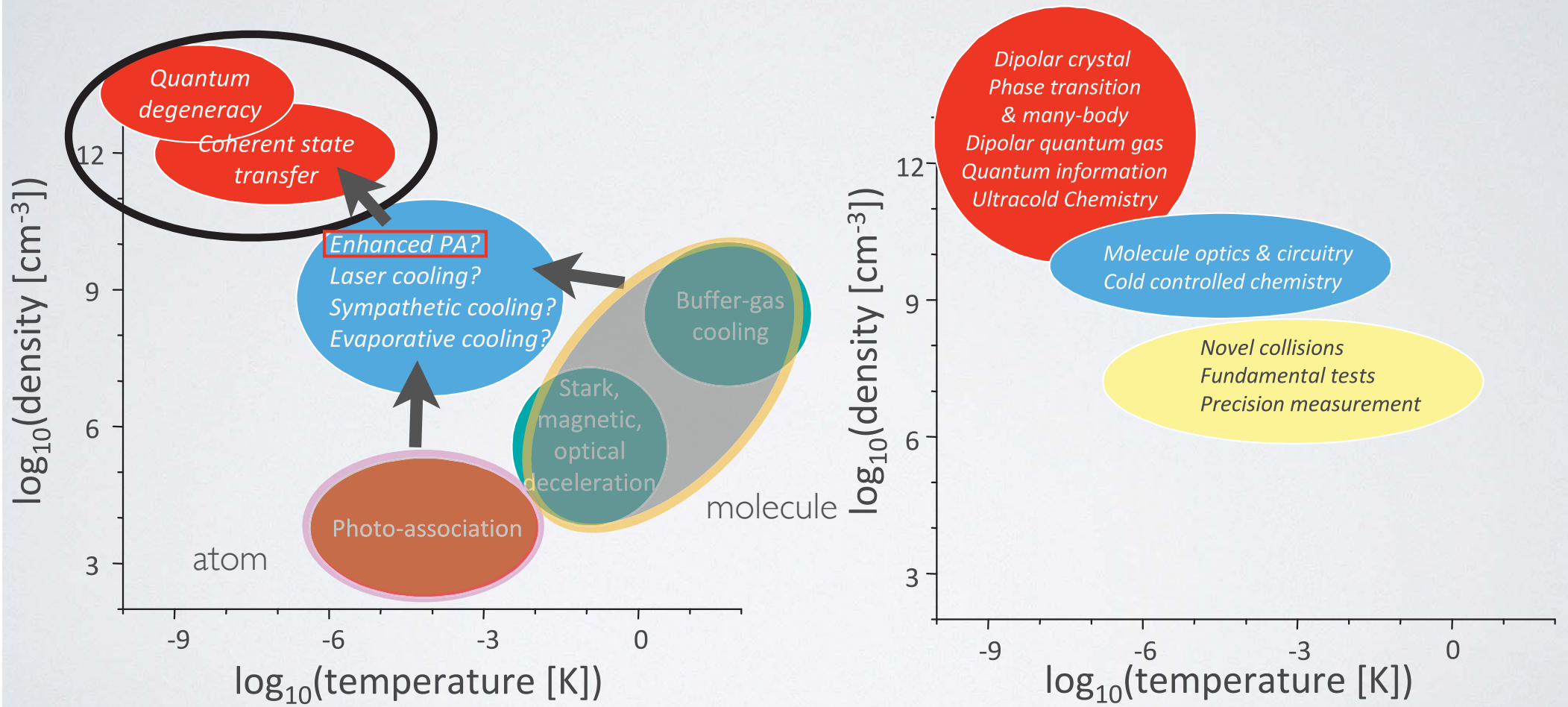

It can boot precision measurement to another regime

WHAT IS COLD MOLECULE?

- A neutral bound system with more than one atom has a very low kinetic energy (translational, vibrational, rotational)

- K_2, KRb, Rb_2, Cs_2....

New problems in molecular system

TEMPERATURE, DENSITY AND COOLING

various cooling techniques

physics using cold molecule

HEAT INCREASES INTERACTION, BUT WE ARE GOING TO COOL EVERYTHING

Heat helps chemical reaction, because:

1. higher collision rate $(r = \sigma v)$

2. higher kinetic energy to penetrate chemical energy barrier
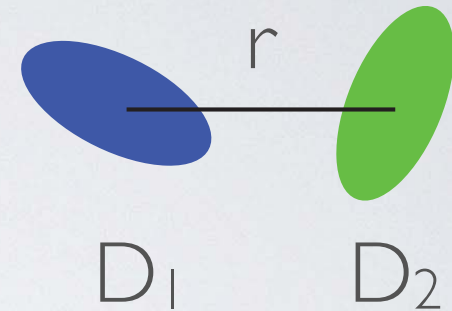
Then, why shall we go cold?

WHAT AND WHY "ULTRACOLD COLLISION"

- A+B ➔ C :chemical reaction is inelastic collision

- "Ultracold" ➔ S wave scattering ➔ increase by 10^3 ↔ It contradicts with our experience in chemical practice.

ULTRACOLD COLLISION

Dipole-Dipole interaction

$$V_d(\vec{r}) = \frac{\vec{D}_1 \cdot \vec{D}_2 - 3\left(\vec{D}_1 \cdot \hat{r}\right)\left(\vec{D}_2 \cdot \hat{r}\right)}{r^3}$$

Rank-2 Tensor: $T^{(2)}q$

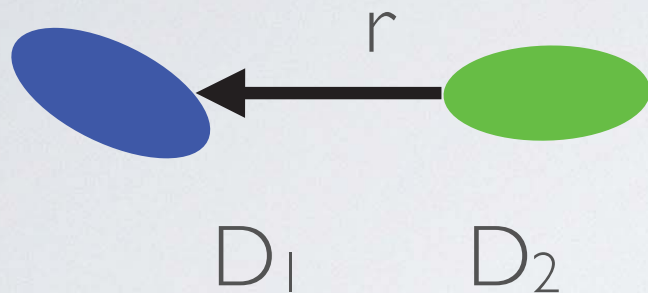Wavefunction of the complex during collision: $|l,m\rangle$

The potential is: $\langle l,m|T^{(2)}q|l,m\rangle$ $=0$

l=0, s-wave scattering

dipole-dipole interaction is vanished
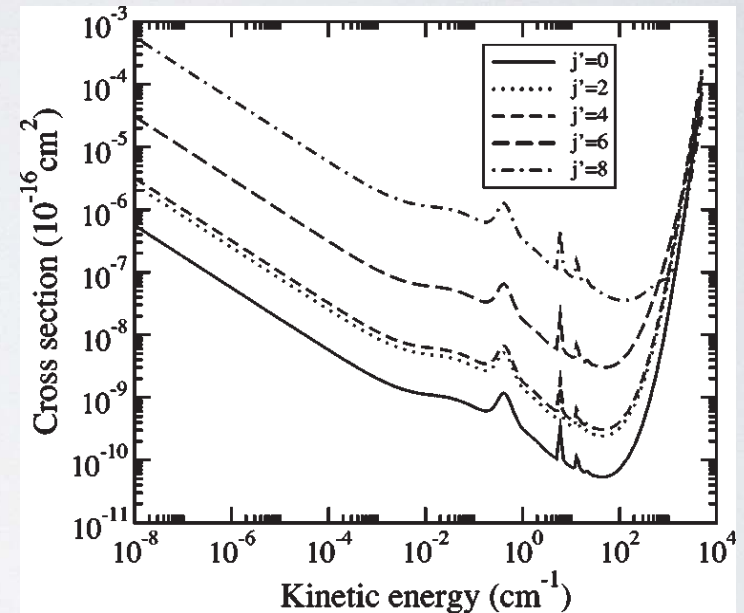
inelastic collision is enhanced
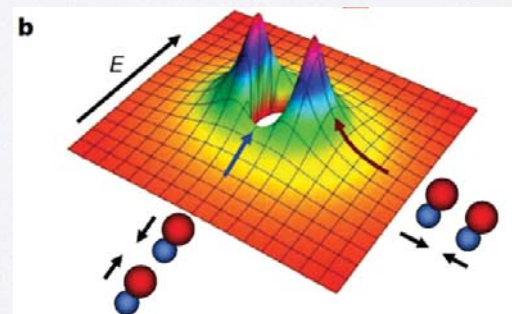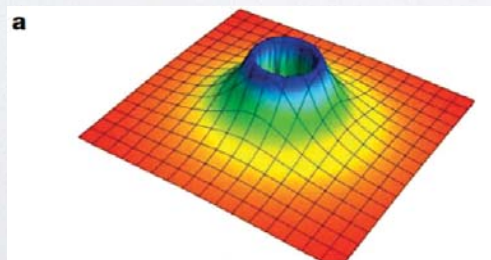
CROSS SECTION ENHANCEMENT

r

D₁   D₂

Head-to-Head collision, Averaged force is zero, No potential barrier

Furthermore, external field can be used to control the alignment, then the interaction of atoms

H₂ -Ar collision v.s temperature

Ni,K.-K. et. al. Nature 2010, 464, 1324.

PARTIAL WAVE COLLISION POTENTIAL BARRIER

$$V_{nml,nml}(R) = E_n^A + E_m^B + \frac{C_p(l,\, nm)}{R^p} + \frac{\hbar^2 l(l+1)}{2\mu R^2}$$

Neutral atom interaction : Van der Waals (R⁶)

$$V(R) = -\frac{C_6}{R^6} + \frac{\hbar^2}{2\mu}\frac{\ell(\ell+1)}{R^2}$$

centrifugal barrier (30mK-100uK)

$$E_C(\ell) = \tfrac{1}{2}[\ell(\ell+1)/3]^{3/2}E_{\mathrm{vdW}}.$$

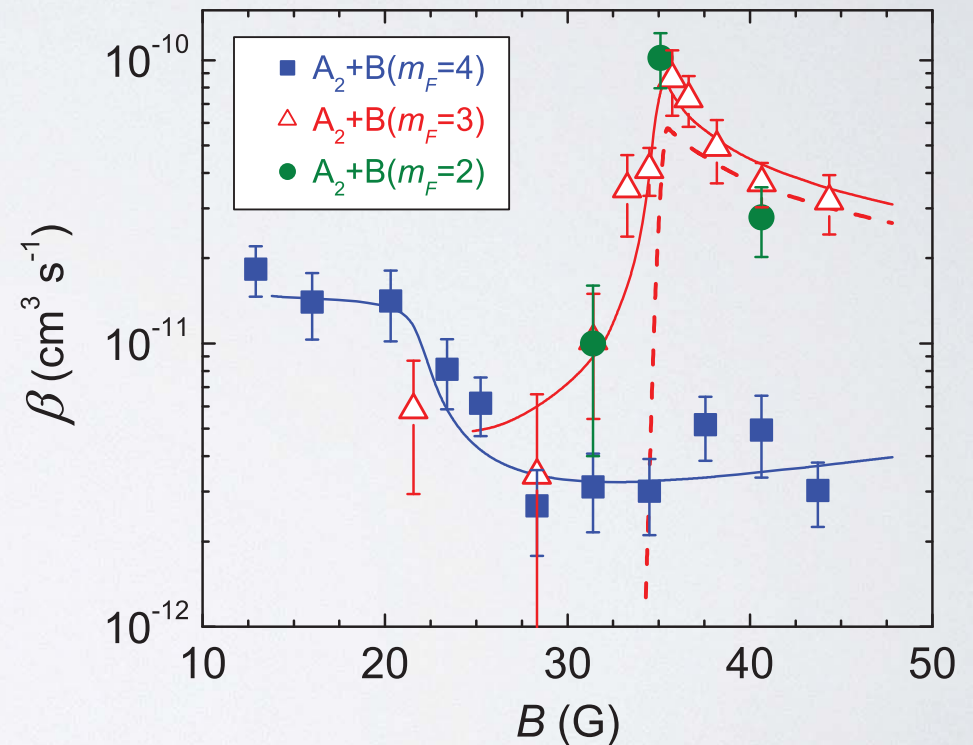| Species	Mass (amu)	C_6 (a.u.)	R_{vdW} (a_0)	E_{vdW}/k_B (mk)	E_{vdW}/h (MHz)
^{6}Li	6.0151223	1393.39[a]	31.26	29.47	614.0
^{23}Na	22.9897680	1556[b]	44.93	3.733	77.77
^{40}K	39.9639987	3897[b]	64.90	1.029	21.44
^{40}Ca	39.962591	2221[c]	56.39	1.363	28.40
^{87}Rb	86.909187	4691[b]	82.55	0.2925	6.094
^{88}Sr	87.905616	3170[c]	75.06	0.3497	7.287
^{133}Cs	132.905429	6860[d]	101.0	0.1279	2.665
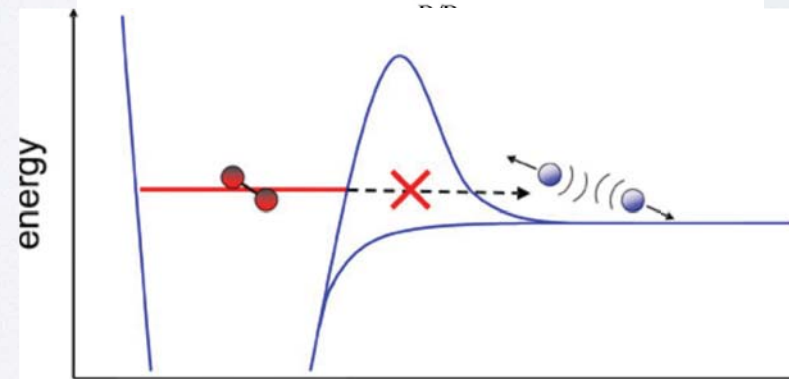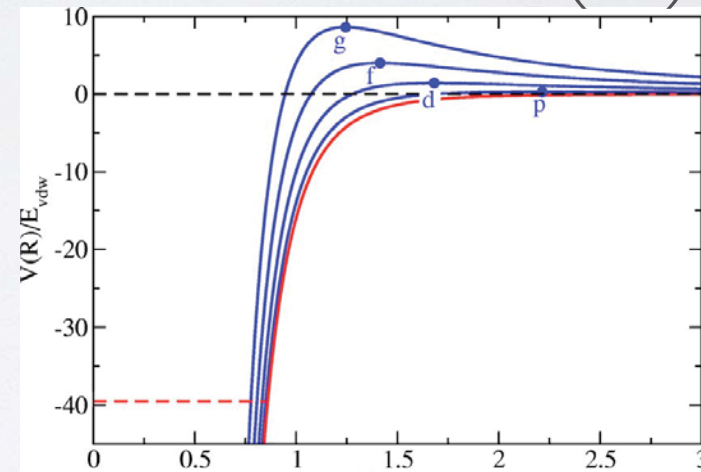
CONTROLLING CHEMICAL REACTION USING MAGNETIC FIELD
ALIGNED COLLISION

Caesium in two hyperfine levels m_F=4 and 3 as different atoms A and B

$$A_2 + B \longrightarrow A + AB$$

Inelastic collision cross section is magnetic-dependent

S. Knoop, F. Ferlaino, M. Berninger, M. Mark, H. C. Naegerl, R. Grimm, J. P. D'Incao, and B. D. Esry, Phys. Rev. Lett. **104**, 053201 (2010).

EFIMOV PHYSICS

The universality of many-body physics predicated 35 years ago now is ready to be examined by ultracold molecule collision
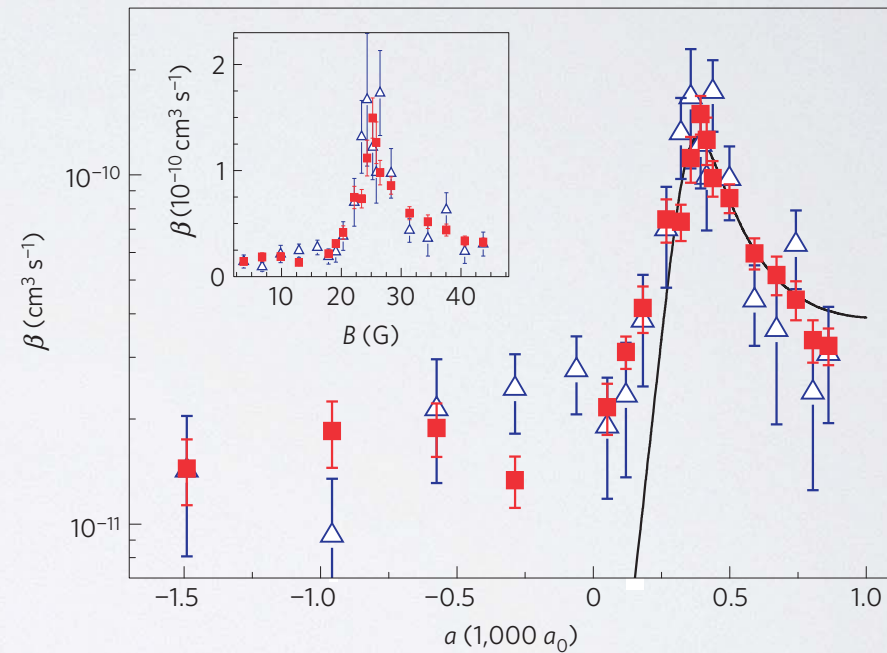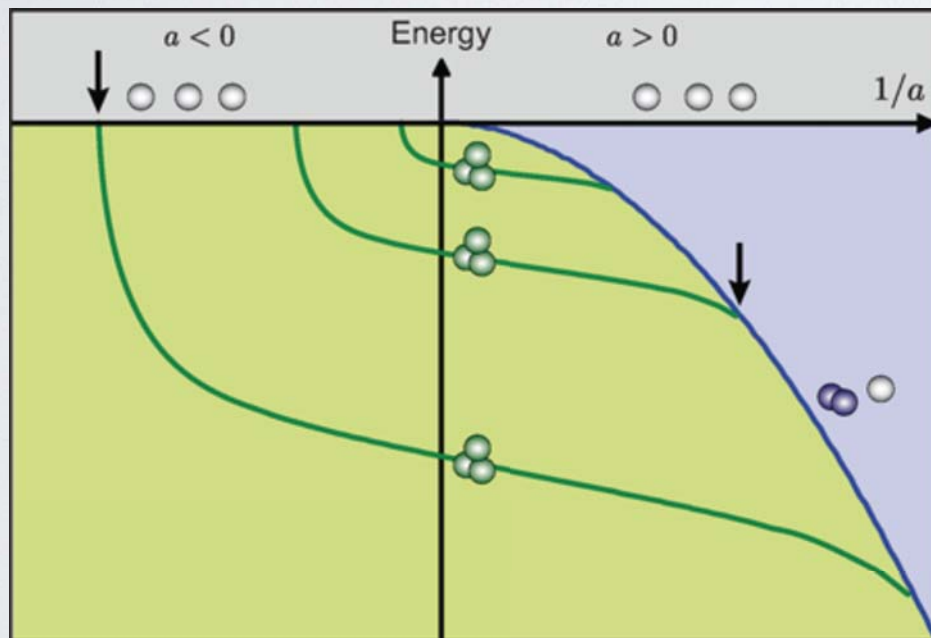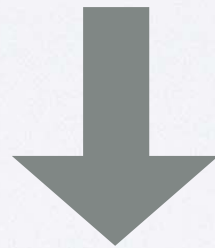

S. Knoop, F. Ferlaino, M. Mark, M. Berninger, H. Schoebel, H. C. Naegerl, and R. Grimm, Nature Phys **5**, 227 (2009).

TWO MAINSTREAMS IN COLD MOLECULE INDUSTRY

- Cool down a molecule

- Produce molecule from cold atoms

Association: Perform chemistry using very cold ingredients

How to produce molecule?
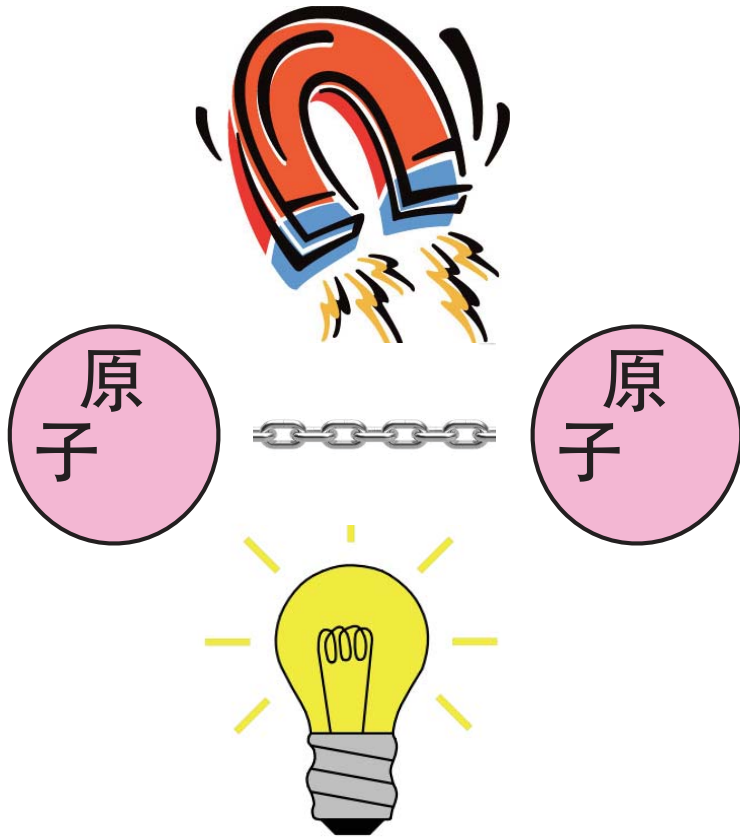
Magneto-association
(Feshbach resonance)

原子   原子

Photo-association
(PA)

Cond.	product	efficiency
nearly quantum degeneracy, nK regime	loosely bound	High
MOT μK regime	More deeply bound	Low

MAGNETO-ASSOCIATION

FESHBACH RESONANCE

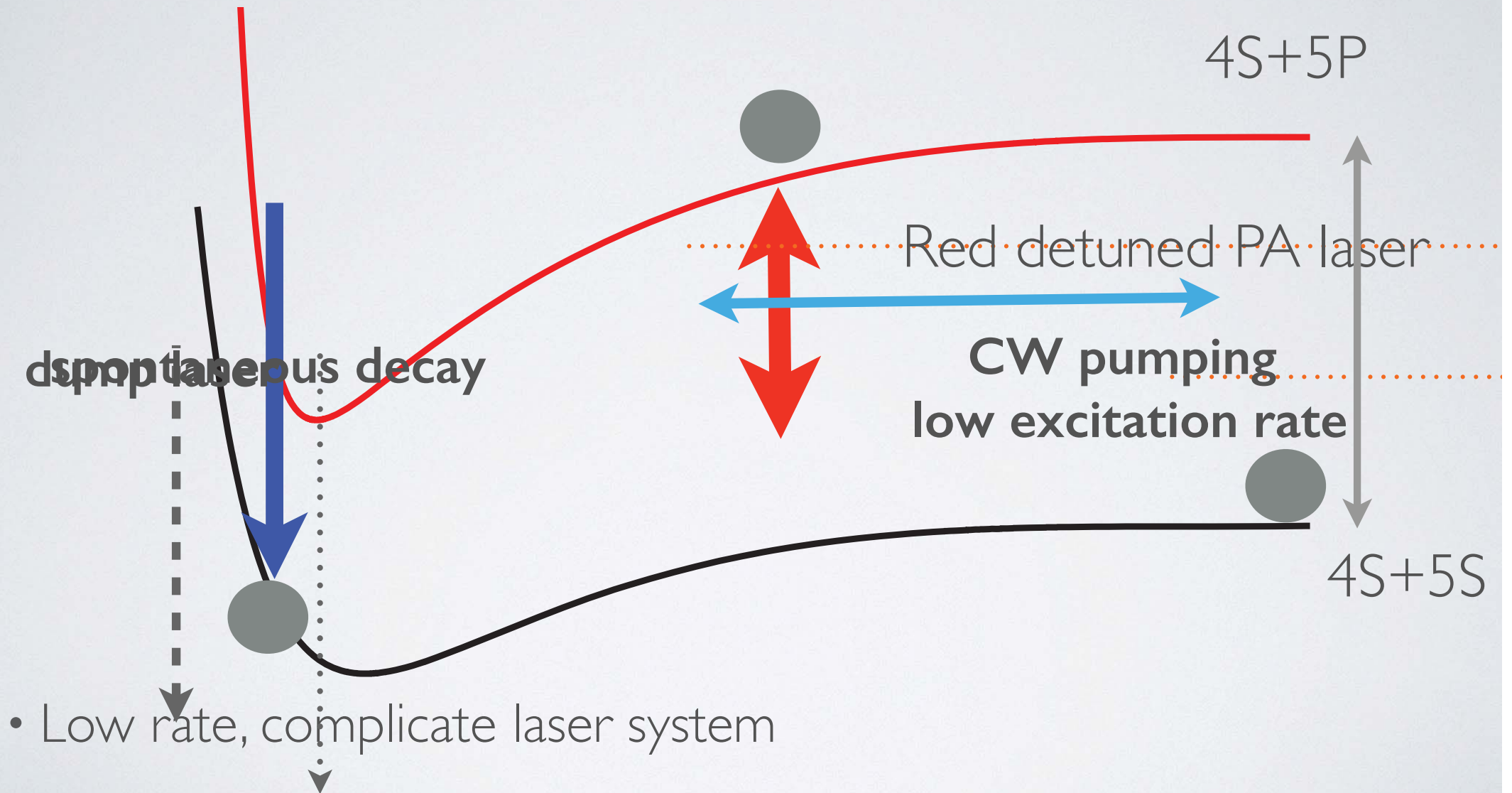Apply external field to shift energy level and tune the interaction between atoms

$K+U_{lowHF} \longrightarrow U_{highHF}$

Free $\longrightarrow$ Bound

The external fields can be: magnetic, optical, electrical

Closed channel

ΔE_{HF}

Open channel

Energy

Internuclear Distance

R

Free

$\longrightarrow$

$r<R$

Bound

PHOTOASSOCIATION

4S+5P

Red detuned PA laser

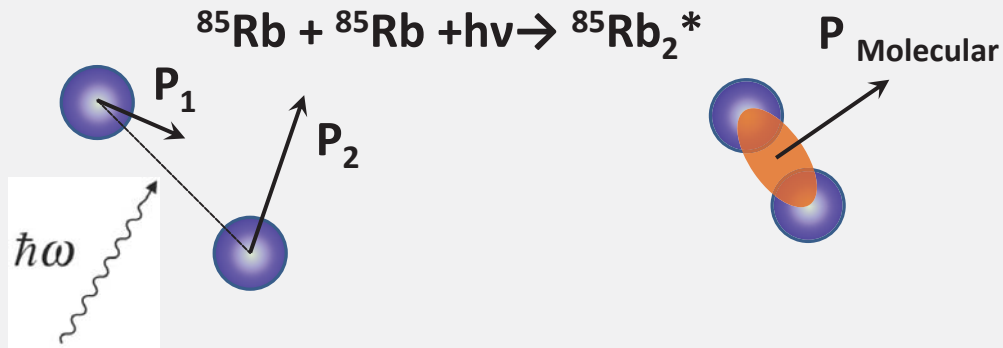CW pumping
low excitation rate

pump laser

spontaneous decay

4S+5S
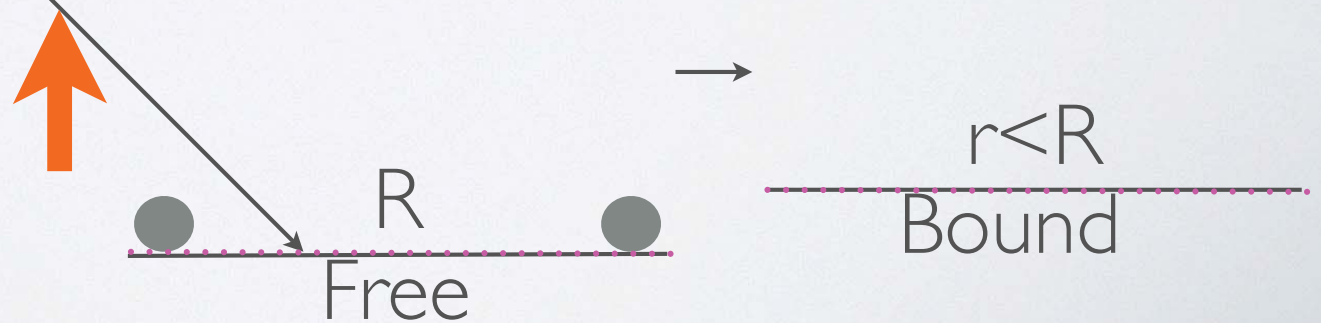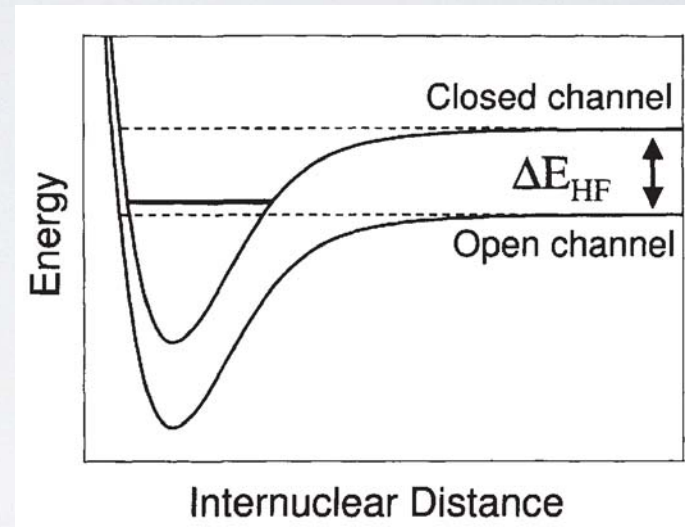
• Low rate, complicate laser system

KINETIC ENERGY IS A PART OF EXCITATIION

Formation of Ultracold Rb2 and Photoassociation Process

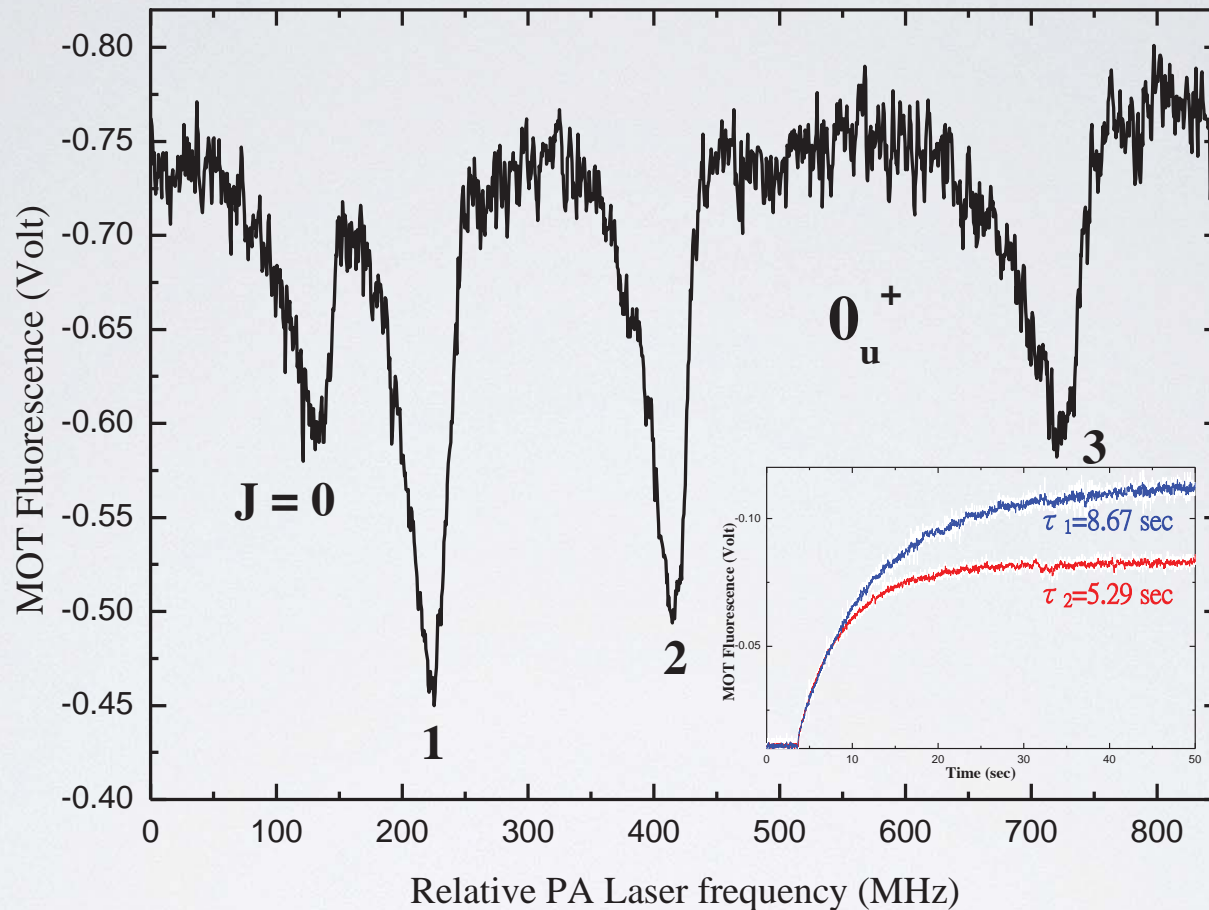$$^{85}Rb + {}^{85}Rb + h\nu \rightarrow {}^{85}Rb_2^*$$

P_1

P_2

$\hbar\omega$

$P_{Molecular}$

$$m\mathbf{v_1} + m\mathbf{v_2} = 2m\mathbf{v}$$

$$\frac{1}{2}m\mathbf{v_1}^2 + \frac{1}{2}m\mathbf{v_2}^2 + h\nu = \frac{1}{2}(2m)\,\mathbf{v}^2 + E_e$$
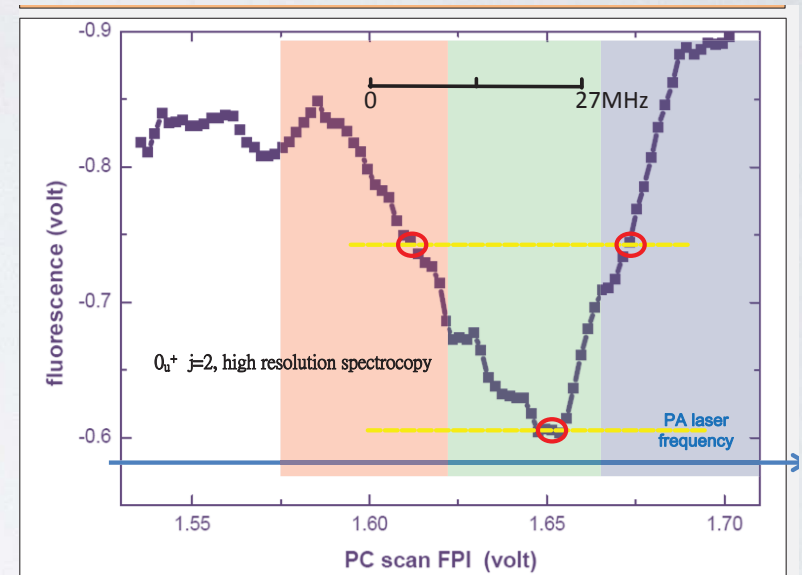
$$\frac{1}{2}m(\mathbf{v_1}-\mathbf{v_2})^2 + h\nu = E_e$$

Excited state

$|e\rangle$ Δ_e Δ

E_e $\hbar\omega$

E_A

ε_r

Ground state

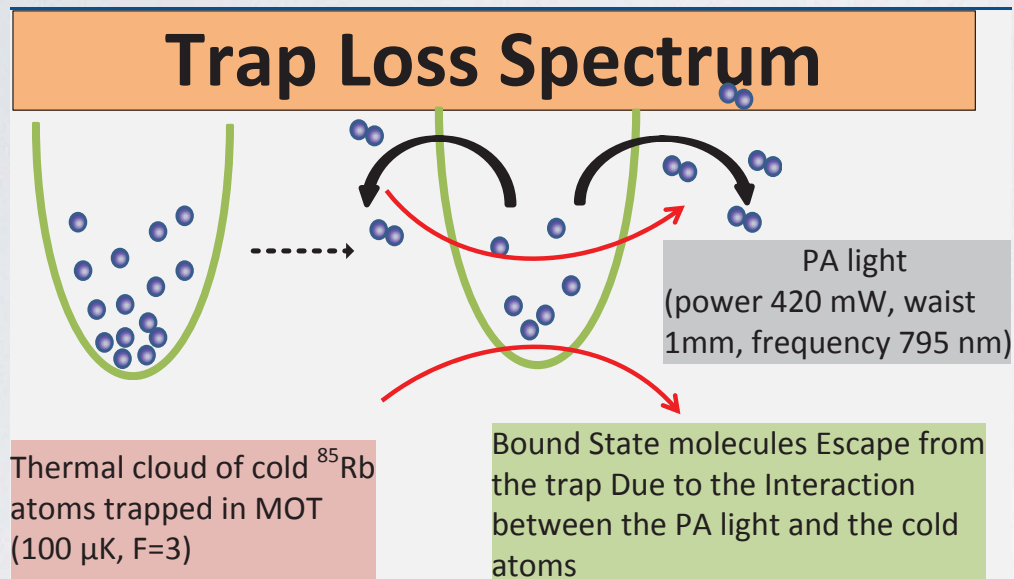$\Gamma_{nat}/(2\pi) = 5$ MHz

$k_BT/h = 10$ MHz
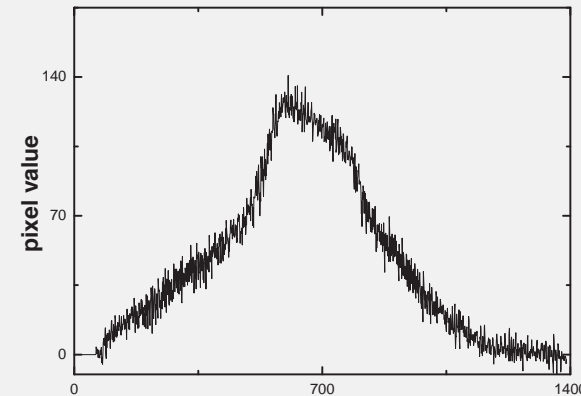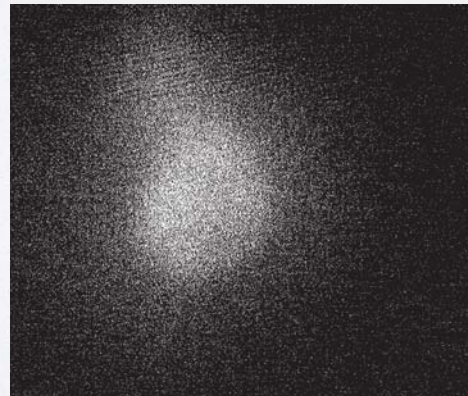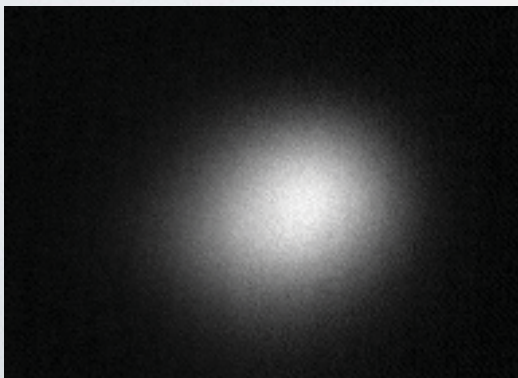
Signal (arb. units)
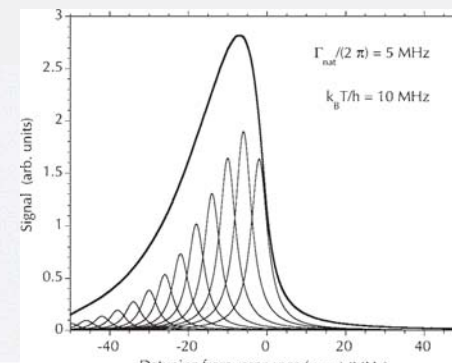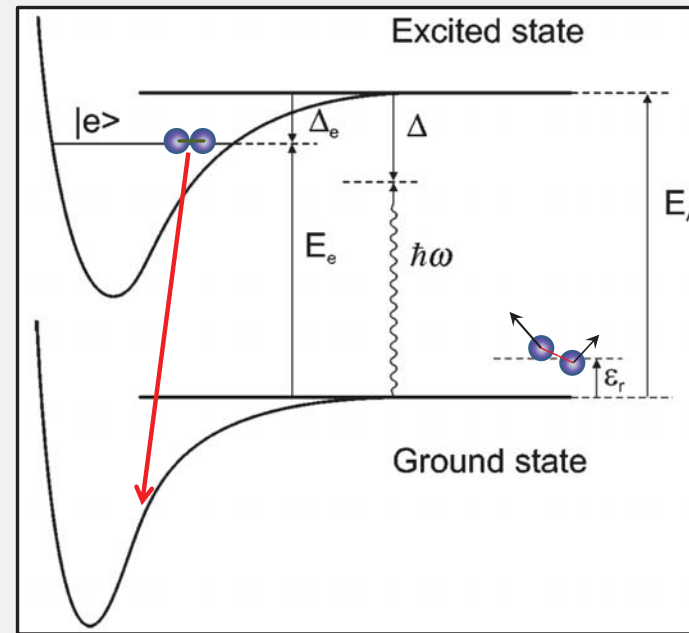
MOT PA TRAP LOSS

The photoassociation spectrum of ^{85}Rb (0^+_u, $v' = 49$)
The maximum loss rate is 40%. PA rate of 1.6×10^6 molecules /sec.
J=2 transition was used to produce ^{85}Rb$_2$ in $X^1\Sigma^+_g$ at v'' = 112 − 116.

VELOCITY SELECTION USING PHOTOASSOCIATION

Trap Loss Spectrum

PA light
(power 420 mW, waist 1mm, frequency 795 nm)

Thermal cloud of cold ^{85}Rb atoms trapped in MOT (100 μK, F=3)

Bound State molecules Escape from the trap Due to the Interaction between the PA light and the cold atoms

0_u^+ j=2, high resolution spectrocopy

PA laser frequency

Non-Gaussian momentum distribution

(a)

$|\Psi_e(R)|^2$

A weakly bound

E_K

Energy

Internuclear distance R
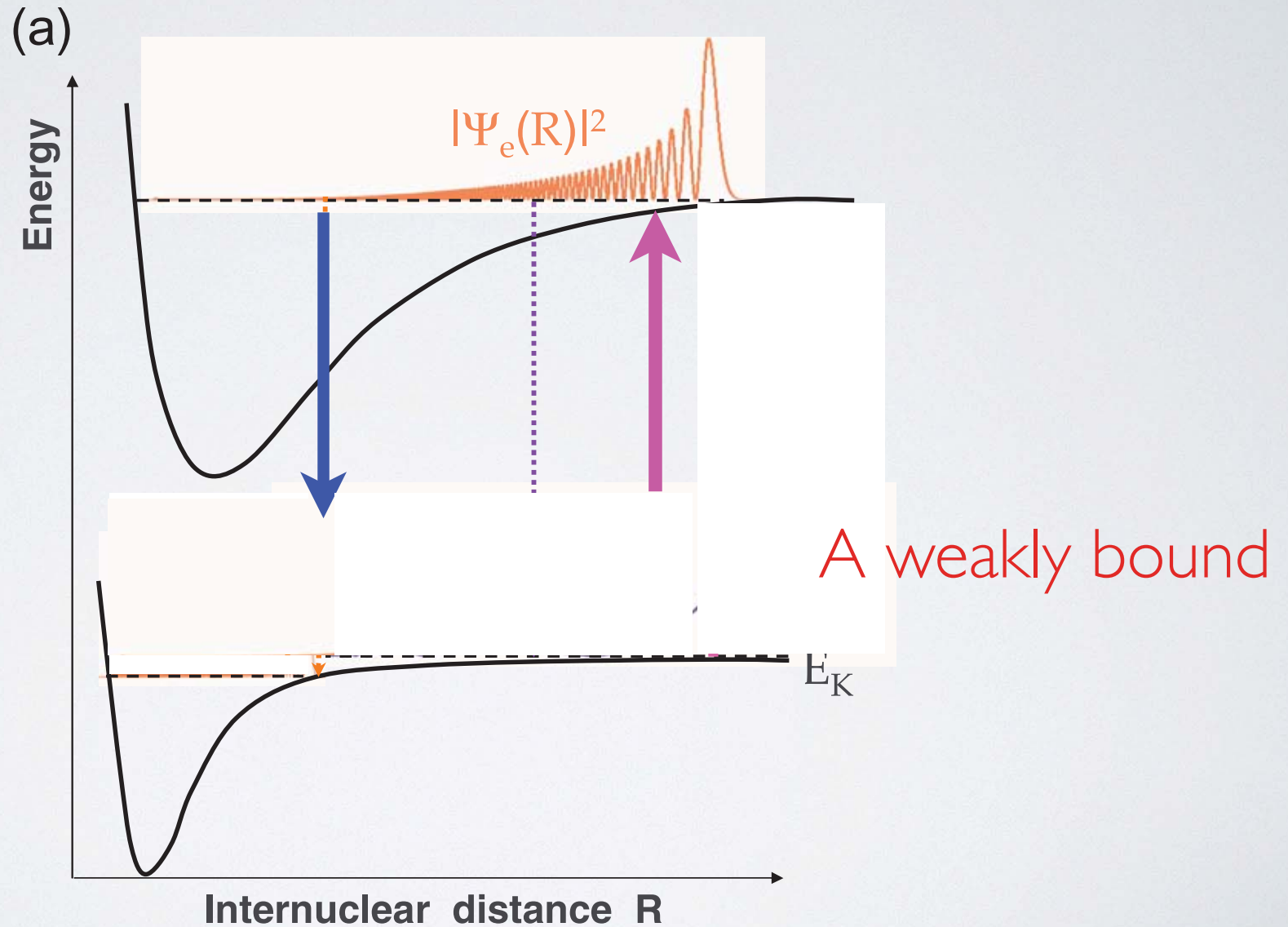
STIMULATED RAMAN ADIABATIC PASSAGE: **STIRAP**

$|A> = (a_1 |1> + a_2|3>)e^{i\alpha t}$:coupled by laser 1

$|B> = (b_1 |2> + b_2|3>)e^{i\beta t}$:coupled by laser 2

If laser 1,2 are **coherent**,

then $|1>$ and $|2>$ are coupled into a coherent dark state

$|coherent\ dark\ state> = (r_1 |1> + r_2|2>)e^{i\gamma t}$

Population can be transfered between $|1>$ and $|2>$ with no access to $|3>$, therefore no spontaneous decay
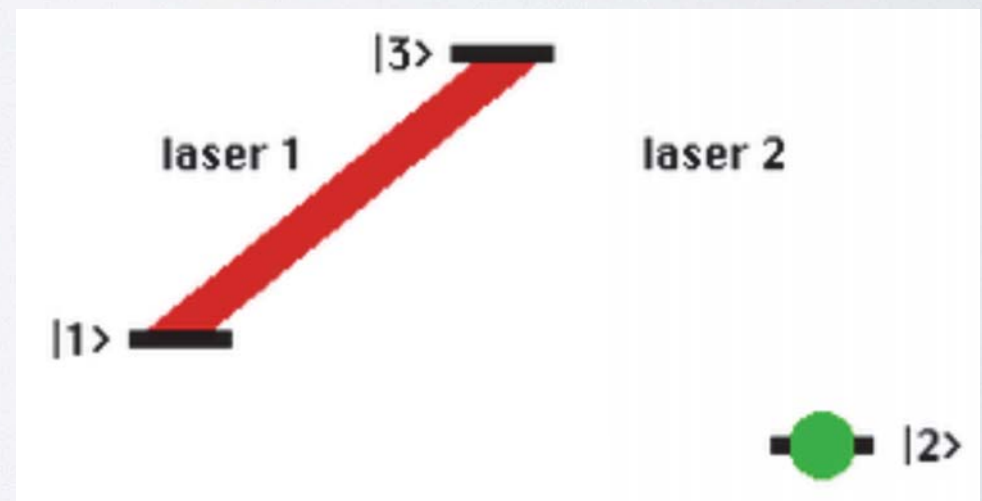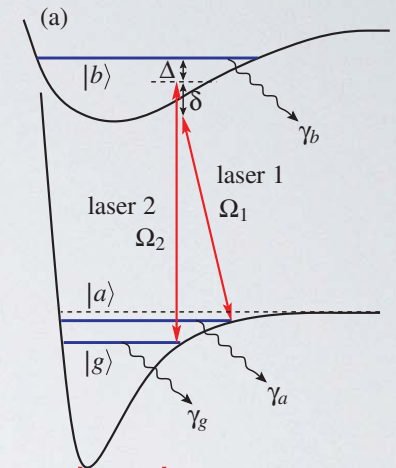
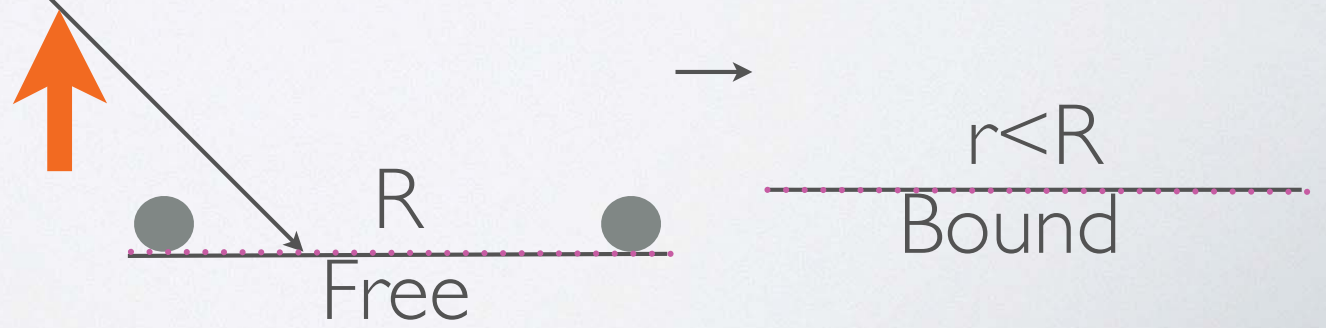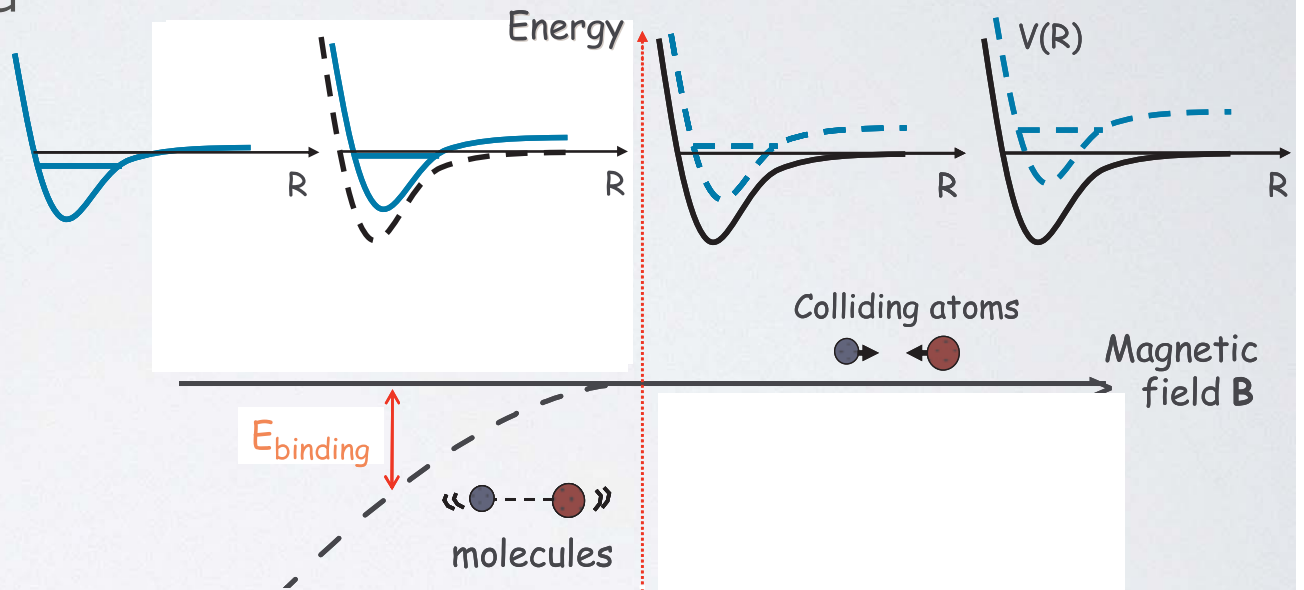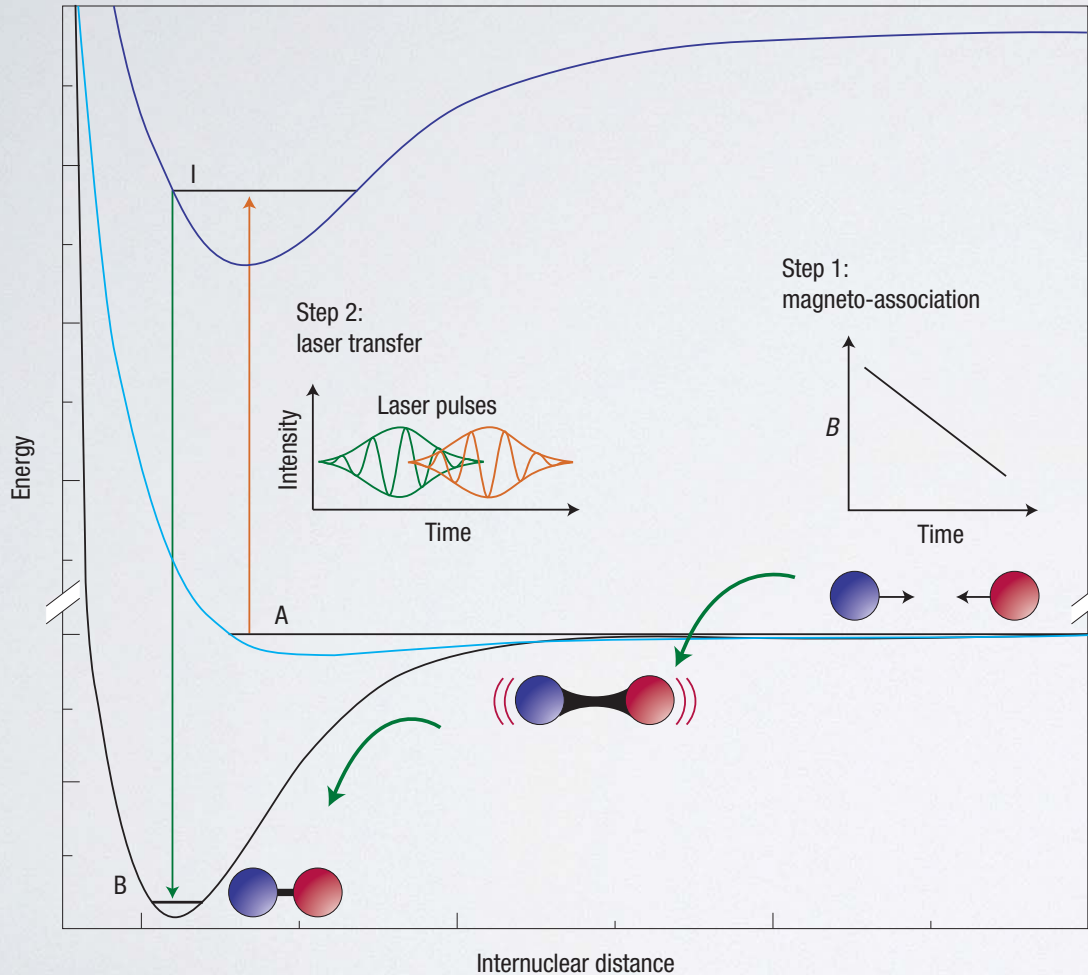# FESHBACH RESONANCE

Apply external field to shift energy level and tune the interaction between atoms

$$K+U_{lowHF} \rightarrow U_{highHF}$$

$$Free \rightarrow Bound$$

The external fields can be:
magnetic,
optical,
electrical

Energy

V(R)

R   R   R   R

Colliding atoms

Magnetic field **B**

$E_{binding}$

molecules

R

Free

r<R

Bound

THE BEST SOLUTION SO FAR

- Feshbach resonance+STRAP (stimulated Raman adiabatic passage)

- Form a very large (R, and high ν) molecule, then remove vibrational energy by stimulation emission.

- CsRb and KRb have been successfully produced (Ni and et al, JILA, Science, 2008. Sage and et al, Yale, PRL, 2005)

Very classical, and low rate.......
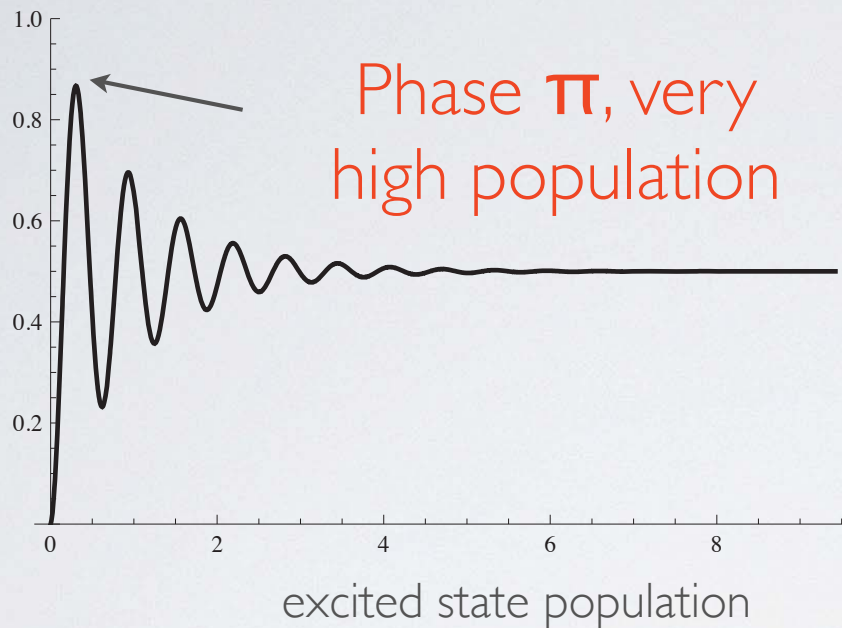
A PROPOSAL TO INCREASE PA RATE

- Can we drive population in all levels using one laser?

- *A broad band laser!*

- Can we pump up population only, without stimulating it down?

A pulse laser!

$\Rightarrow$ *The femtosecond laser*

MAXIMUM INVERSION USING Π

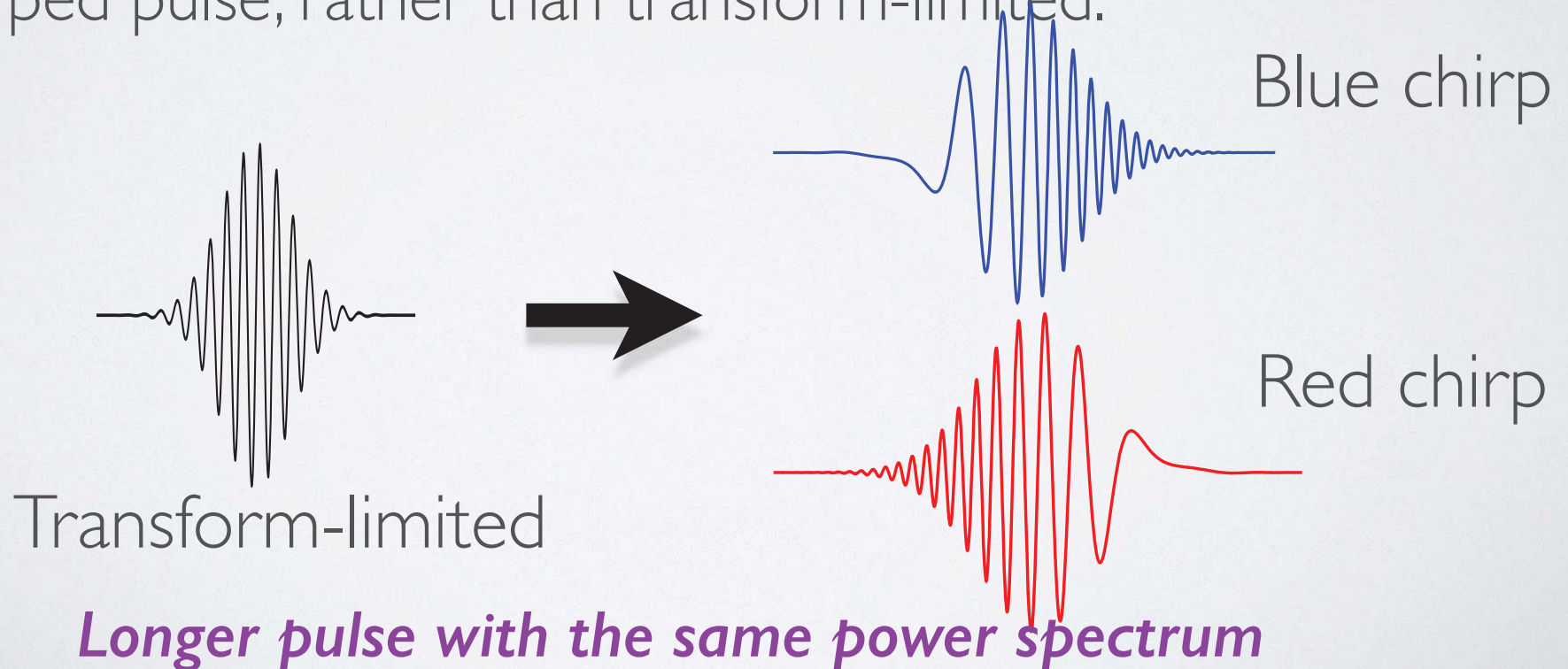Phase **π**, very high population
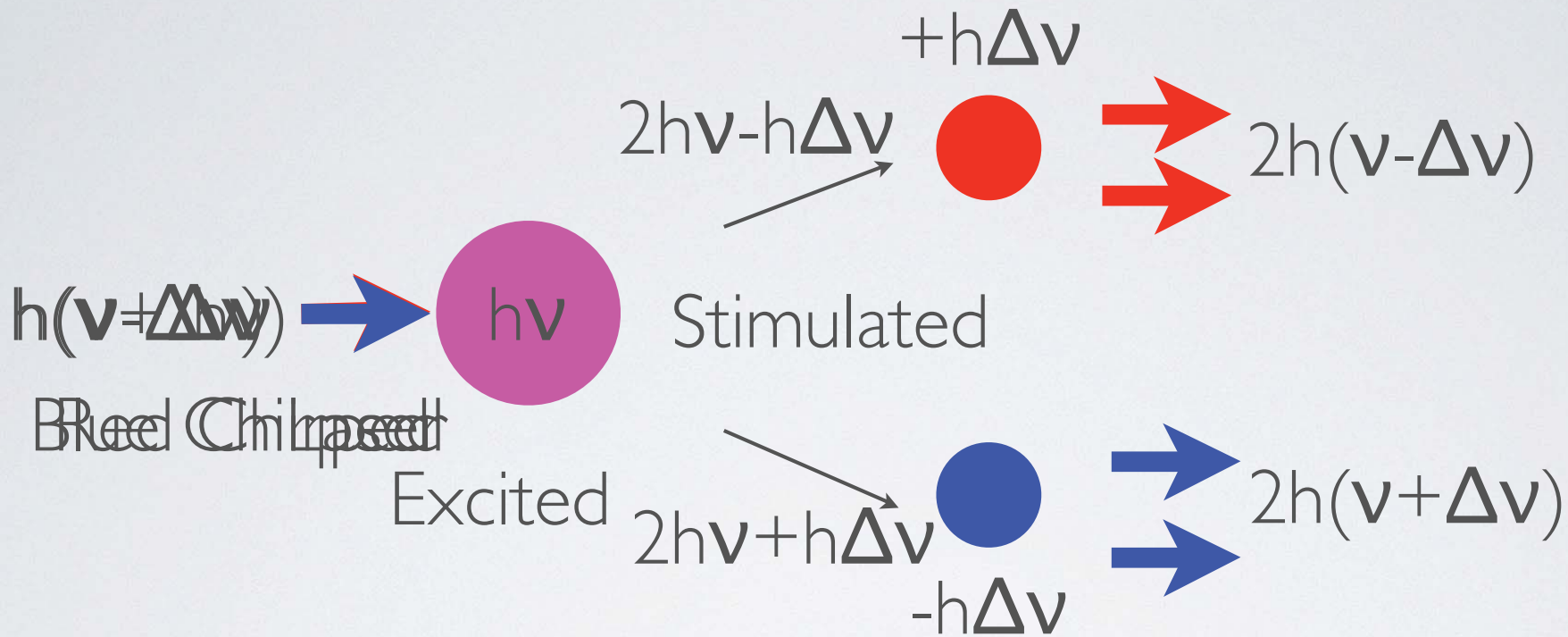
Rabi Osc

excited state population

- Short pulse to perform **π** population transfer. $\tau E_0 \mu / \hbar = \pi$

  • Problems: very large $E_0 \sim 10^{12}$-10^{14} W/cm^2. Many subtle effects should be taken into account, such as multi-photon transition

CHIRPED FEMTOSECOND LASER

- $\tau E_0 \mu / \hbar = \pi$, a longer τ can lower required E_0

- To maintain the same $\Delta\nu$ with a long τ $(>1/\Delta\nu)$, we need a chirped pulse, rather than transform-limited.

Blue chirp

Red chirp

Transform-limited

Longer pulse with the same power spectrum

BLUE OR RED?

$+h\Delta\nu$

$2h\nu-h\Delta\nu$ ● → $2h(\nu-\Delta\nu)$

$h(\nu+\Delta\nu)$ → hν Stimulated

Blue Chirped

Excited $2h\nu+h\Delta\nu$ ● → $2h(\nu+\Delta\nu)$

$-h\Delta\nu$

- **The Blue Chirped pulse can remove energy** *(proposed by J. Cao et al, PRL 1998)*

- just like Raman cooling (transfer to a lower kinetic energy level)

CHIRP EXPERIMENT

**Coherent Control of Ultracold Molecule Dynamics in a Magneto-Optical Trap
by Use of Chirped Femtosecond Laser Pulses**

Benjamin L. Brown,[1,2,*] Alexander J. Dicks,[1] and Ian A. Walmsley[1]

[1]*Clarendon Laboratory, Department of Physics, University of Oxford, Oxford, OX1 3PU, United Kingdom*
[2]*The Institute of Optics, University of Rochester, Rochester, New York 14627, USA*
(Received 13 September 2005; published 5 May 2006)
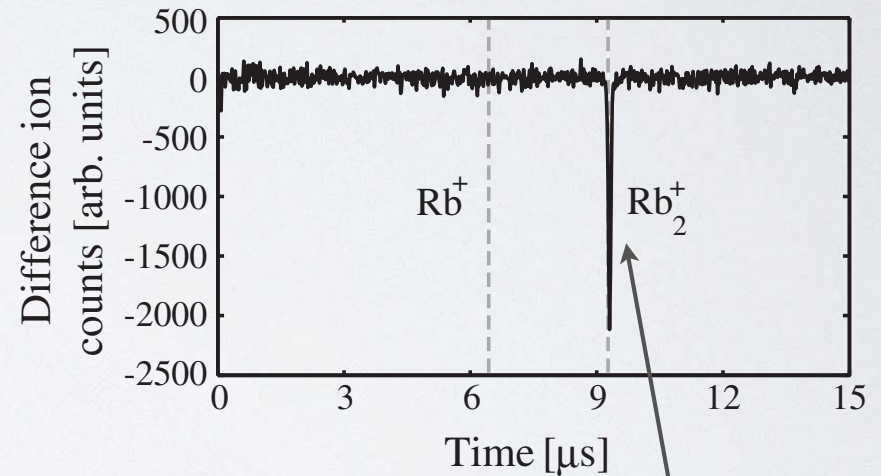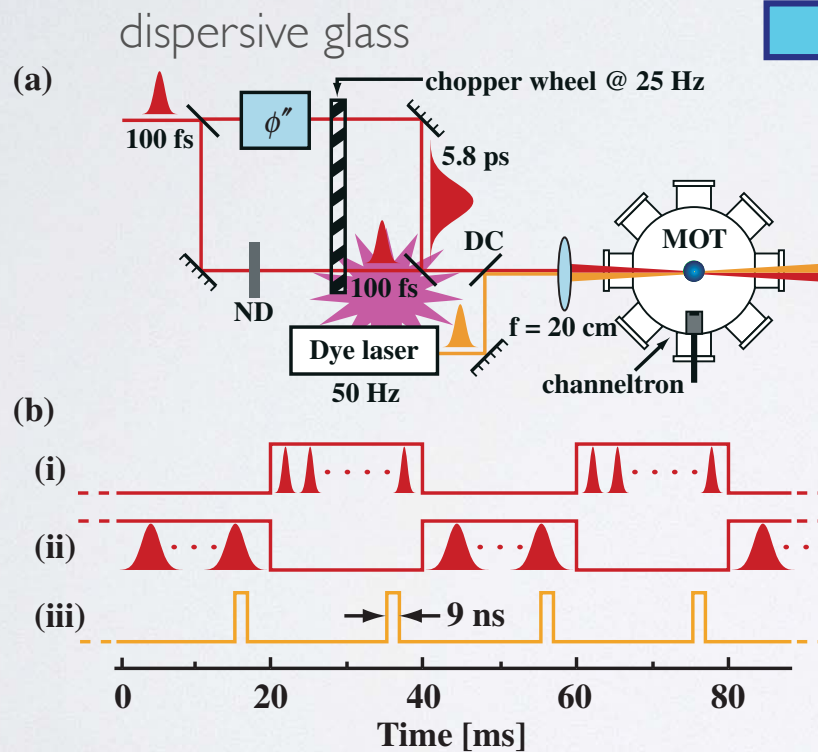
The rate decreased !! Very sad!!

(fs laser, not comb, no stabilization)

Many exciting experiments are on going, including the

comb laser excitation and pulse shape feedback control

COHERENT EXCITATION

## Coherent Transients in the Femtosecond Photoassociation of Ultracold Molecules

W. Salzmann, T. Mullins, J. Eng, M. Albert, R. Wester, and M. Weidemüller[*]

Physikalisches Institut, Universität Freiburg, Hermann Herder Strasse 3, D-79104 Freiburg im Breisgau, Germany

A. Merli, S. M. Weber, F. Sauer, M. Plewicki, F. Weise, L. Wöste, and A. Lindinger[†]
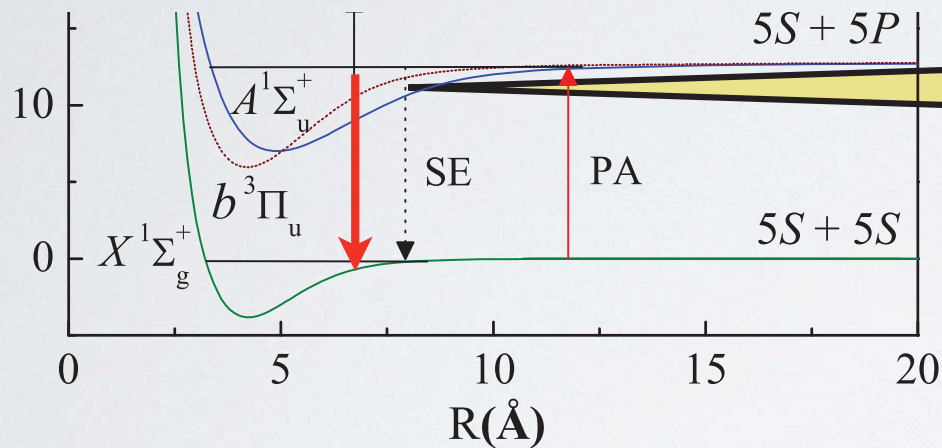
Institut für Experimentalphysik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin, Germany
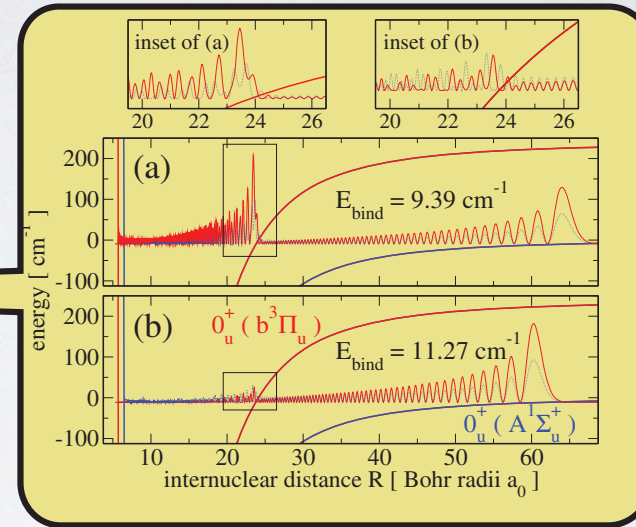(Received 11 December 2007; published 13 June 2008)
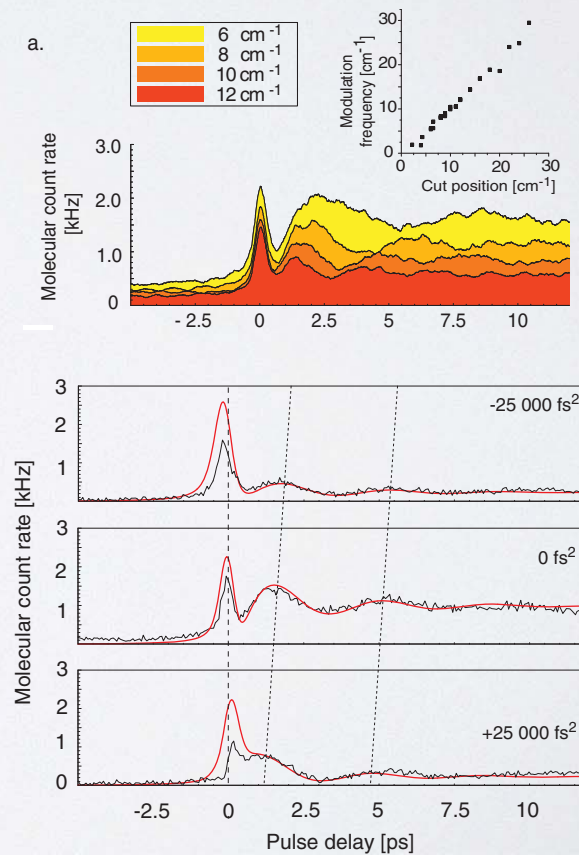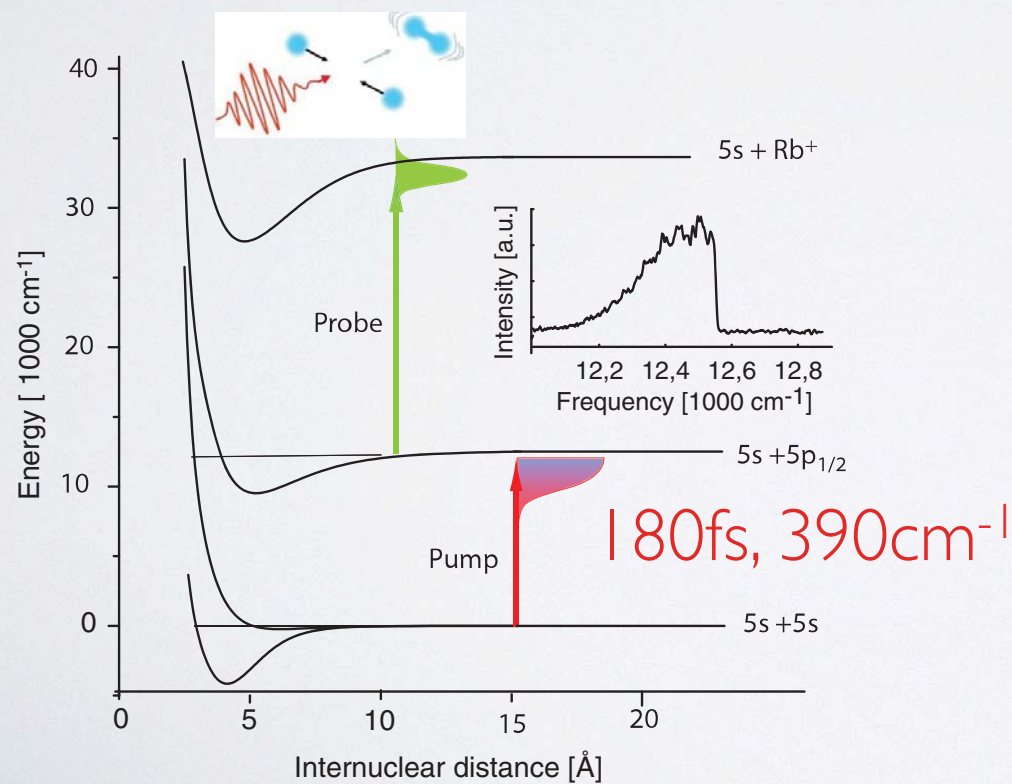
RESONANCE COUPLING ENHANCEMENT

H. Pechkis, D. Wang, Y. Huang, E. Eyler, P. Gould, W. Stwalley, and C. Koch, Phys. Rev. A **76**, 022504 (2007).
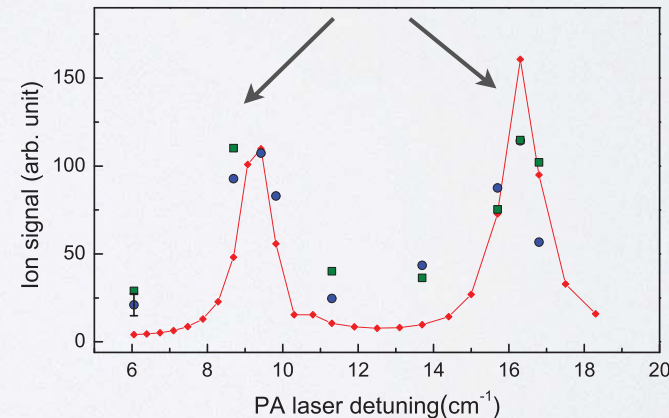

Spin-obit coupling

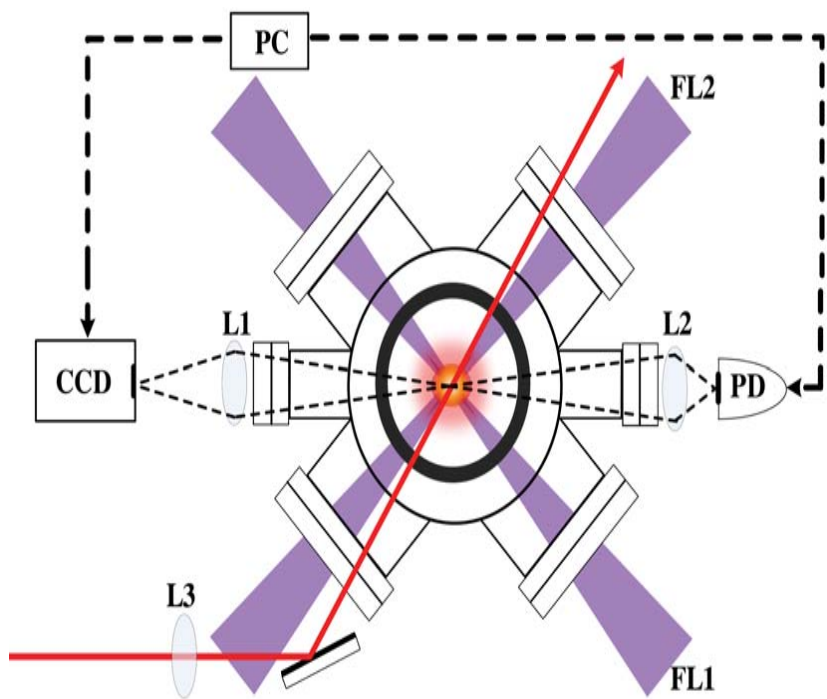$$\hat{H}_e = \begin{pmatrix} \hat{T} + V_{A^1\Sigma_u^+}(R) & \\ & \hat{T} + V_{b^3\Pi_u}(R) \end{pmatrix}$$
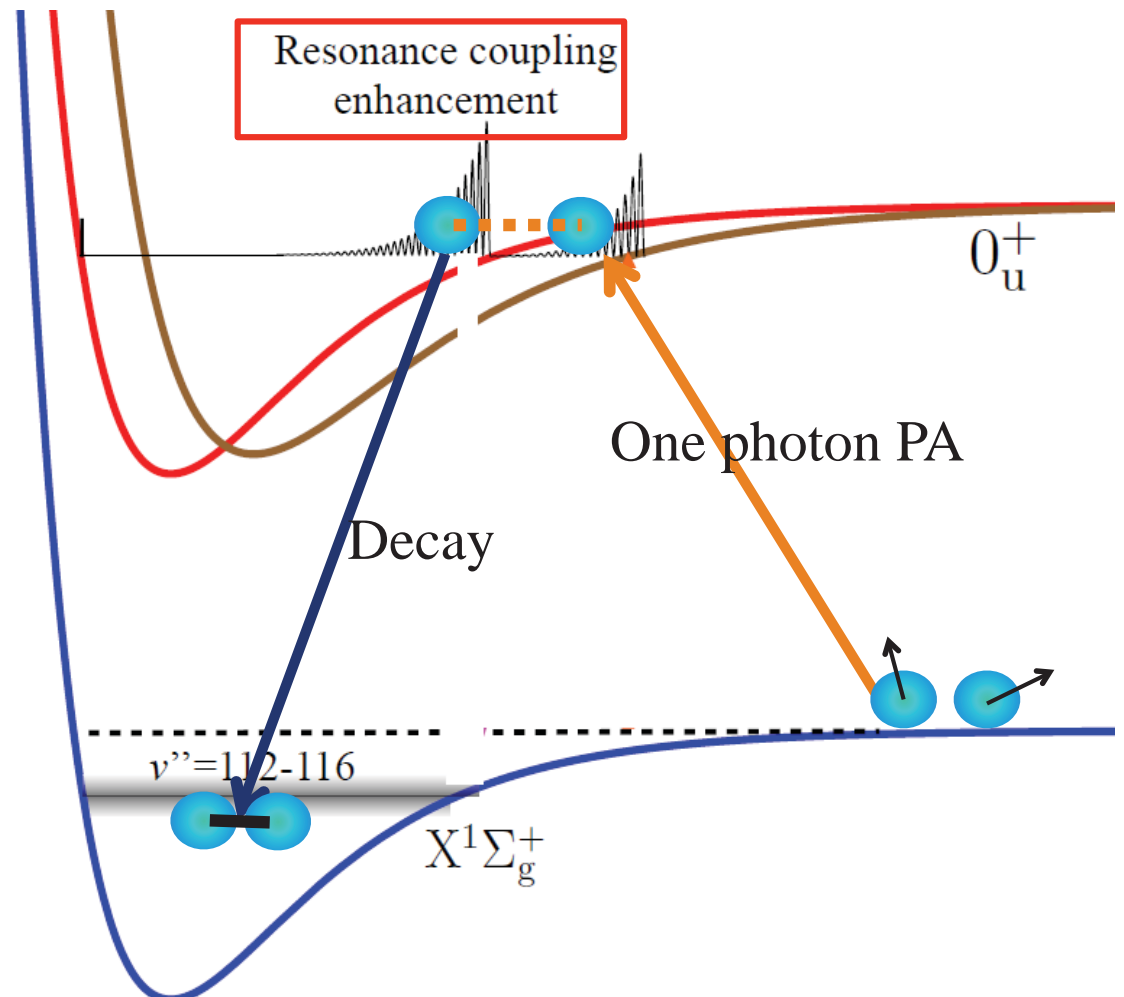
enhancement for vibrational states

If molecules are accumulated with such a high production rate, we can have a lot of them

Resonance coupling enhancement in PA

$n_m = 5.2 \times 10^{11} \text{cm}^{-3} \sim n_a$

**efficiently one-photoassociated
to the deeply bound singlet ground state**

The second challenge...
HOW TO DETECT MOLECULE

There are difficulties

- Very small amount, low density,

- Much more complex internal energy structure (vibrational, rotational ....)

Current solutions

- Ionization mass spectrometer (REMPI) →high energy resolution, but invasive

- Absorption image->molecule must be very well understood, and delicate

Molecule must be very well understood before detection applied.

More lasers, more delicate detectors.....

A more versital detection is required

MOLECULE DETECTION

1. Number density, spatial and momentum distribution can all be measured

2. Difficult to find a proper cycling transition (with a high **η**)
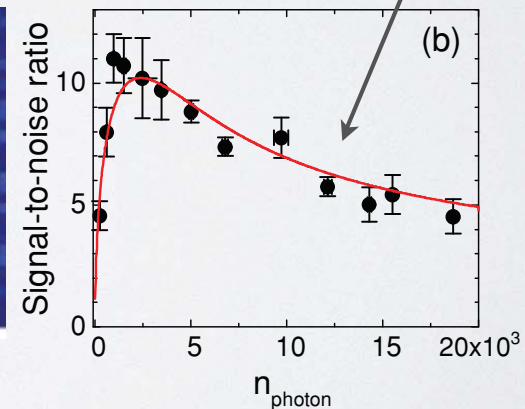

η=0.14
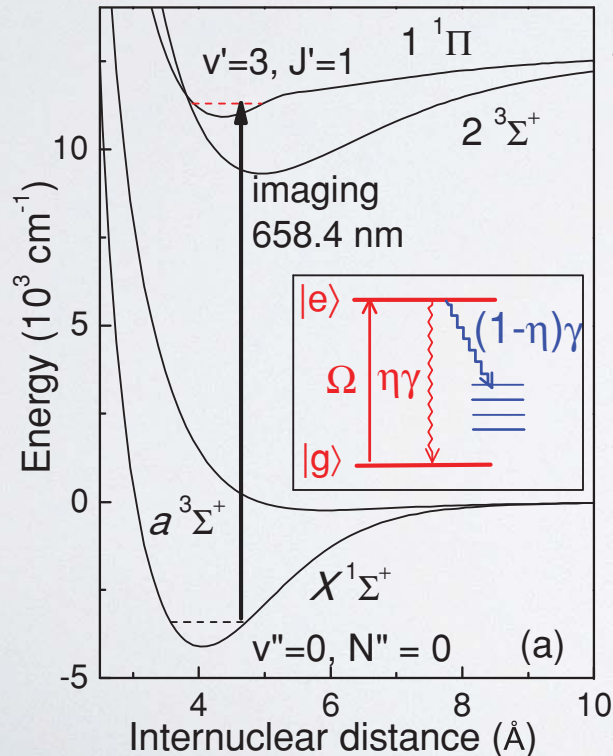
atomic system **η~1**

$$N_{\text{signal}} = \frac{N_{\text{molecule}}}{1 - \eta}\left[1 - \exp^{-4\pi \frac{d^2(1-\eta)}{\hbar\epsilon_0 \lambda A \gamma} n_{\text{photon}}}\right]$$

low photon number
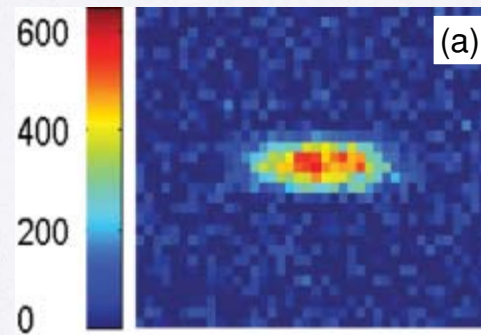
DETECT MOLECULE BY ATOM
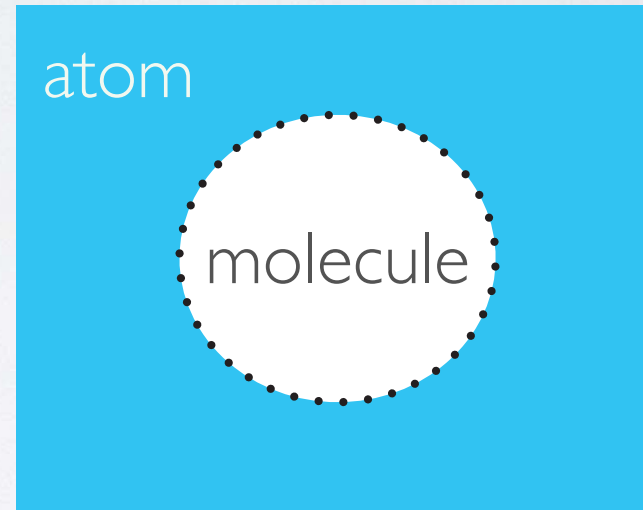
The "invisible" molecule will come to the world through atom-molecule collision
Experimental setup smilier to atom imaging, no additional laser and detector

Imaging molecule through atom

Probe beam

NOPA

PA

CCD

Rb$_2$ molecules   Cold Rb atoms

TIMING SEQUENCE

Rb and Rb$_2$ Collecting	PGC	Optical Pumping	Rb and Rb$_2$ Collision in ODT

Trap laser	On		Off
Magnetic field	On		Off
Repump laser	On		Off
PA laser	On		Off
ODT	On		On
CCD			

Time
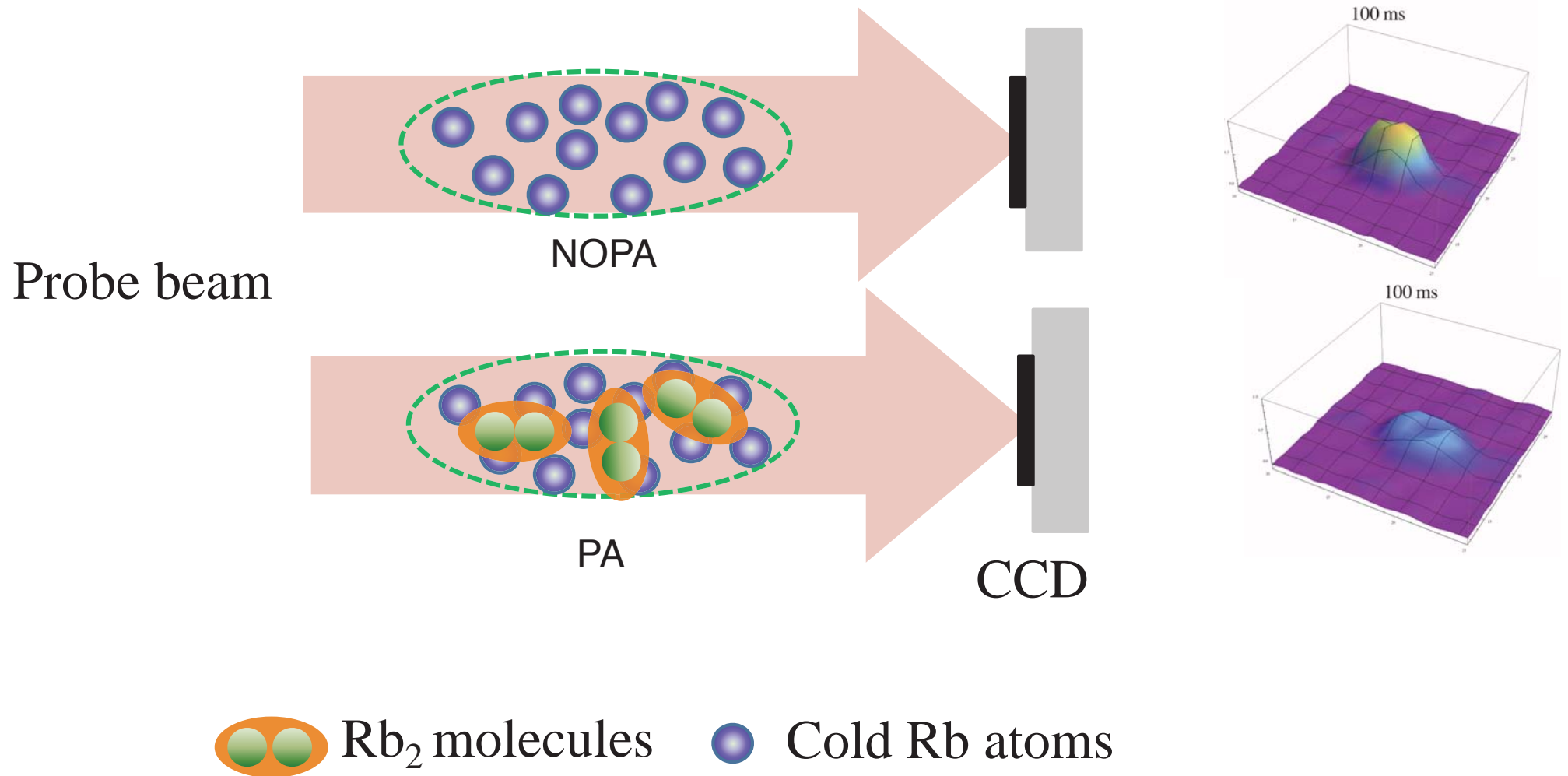
Holding Time
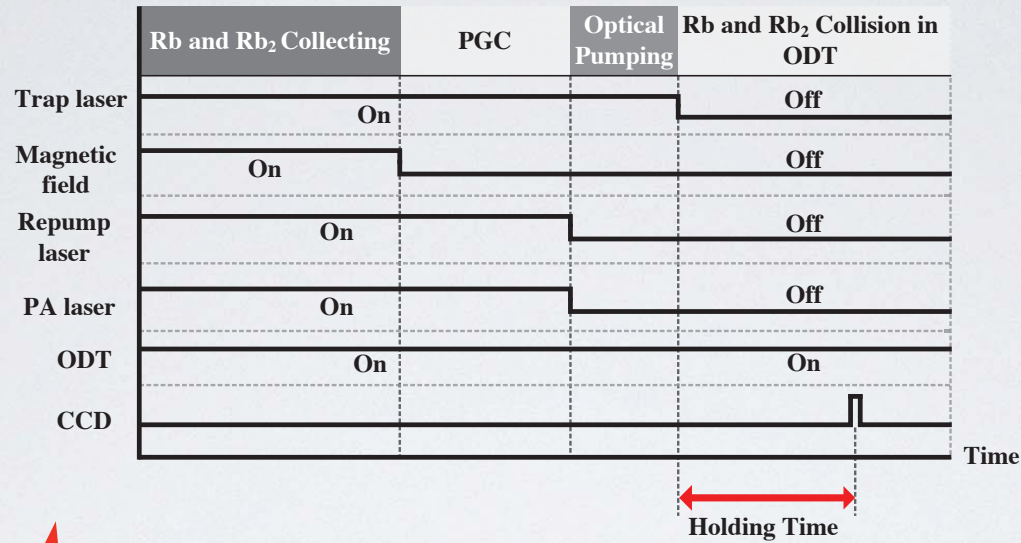
PA Laser
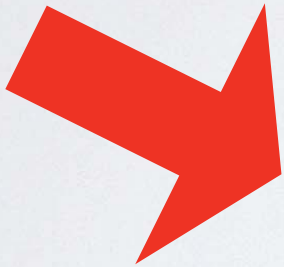
Dipole trap
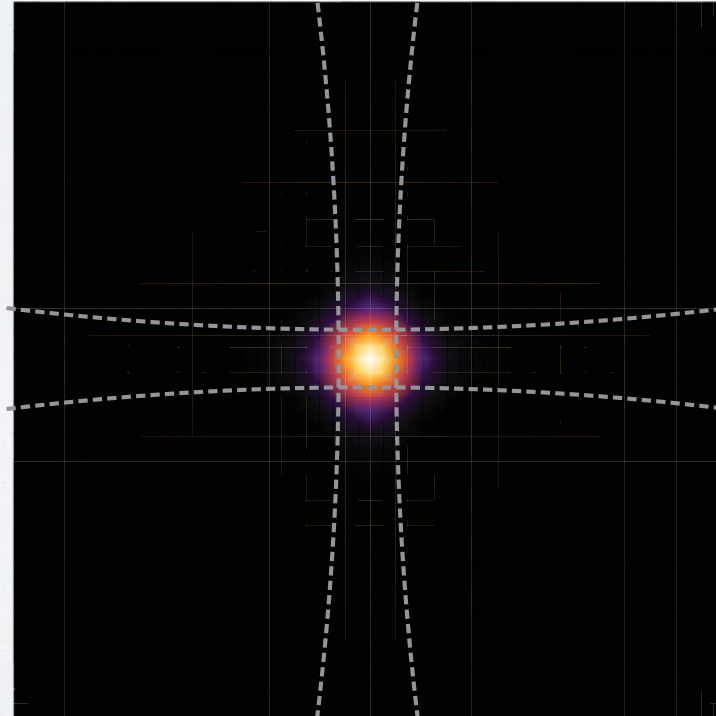
The evolution of the absorption image of rubidium atoms

40 ms
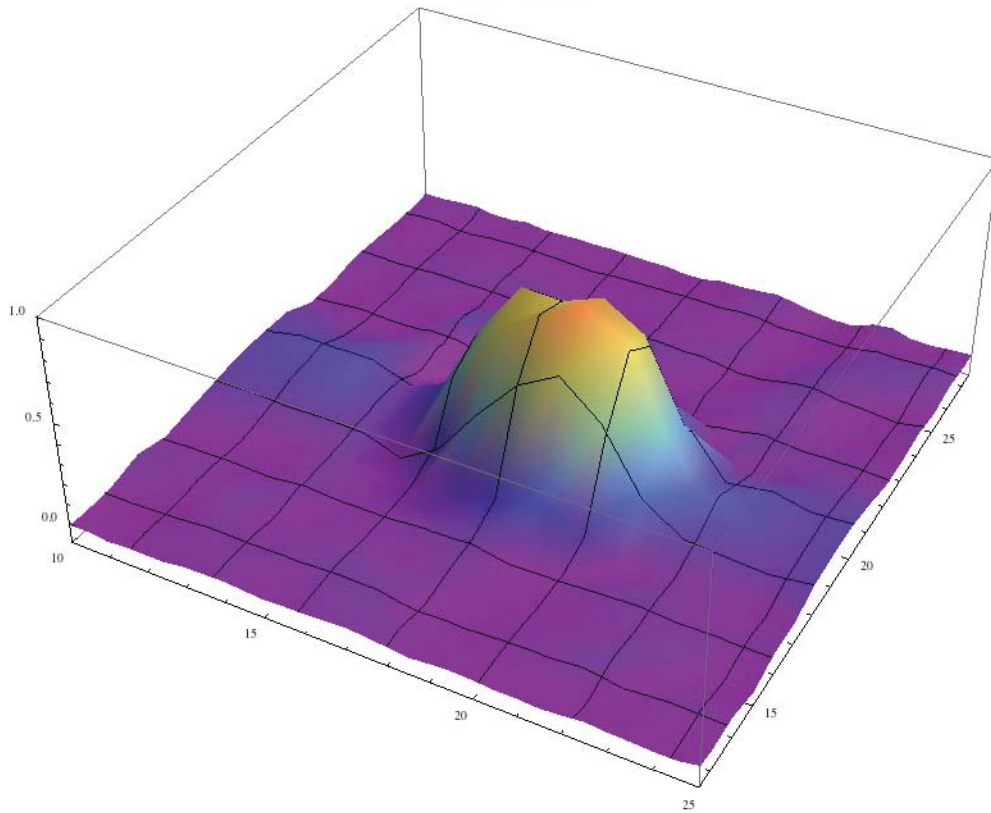
NO PA

40 ms
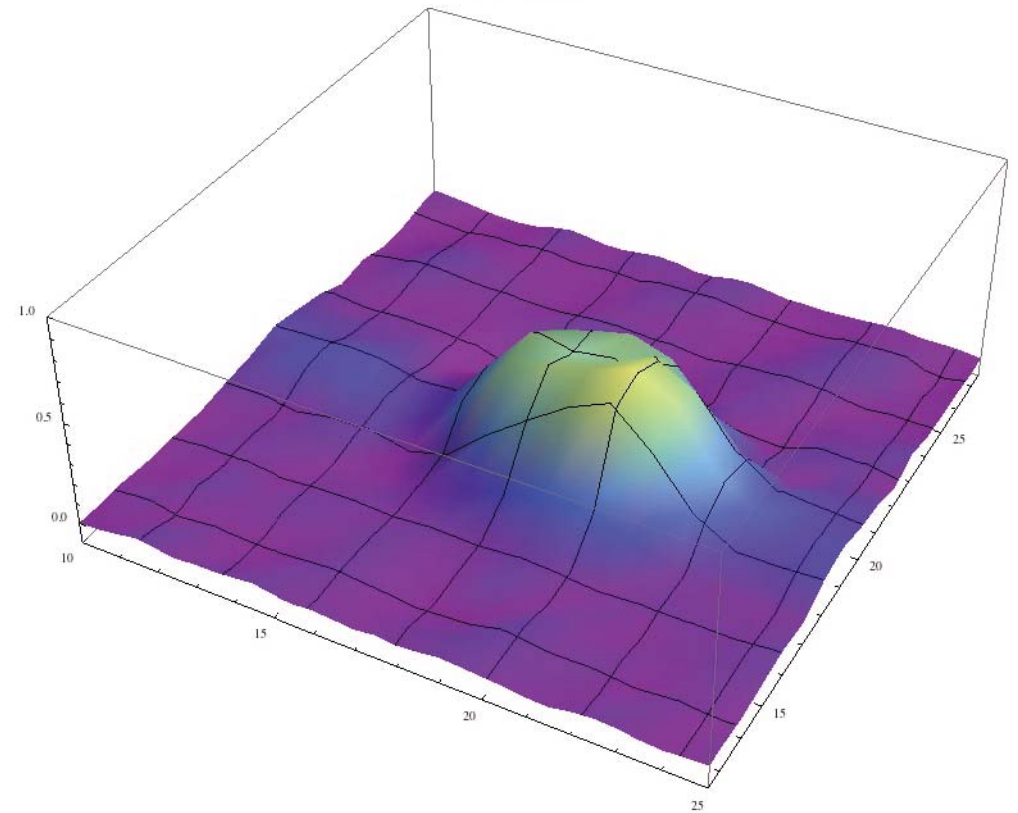
PA

The evolution of the atom number

$$\frac{\mathrm{d}N_a}{\mathrm{dt}} = -[\Gamma + \frac{\beta}{V(t)}N_m]N_a$$

$$\frac{\mathrm{d}N_m}{\mathrm{dt}} = -[\Gamma_m + \frac{\beta}{V(t)}N_a]N_m$$

$\Gamma_{eff} = 7.3 \text{ s}^{-1} \text{ (Rb-Rb)}$

$\Gamma_{eff} = 13 \text{ s}^{-1} \text{(Rb-Rb}_2\text{)}$

Atom number

Holding time (ms)
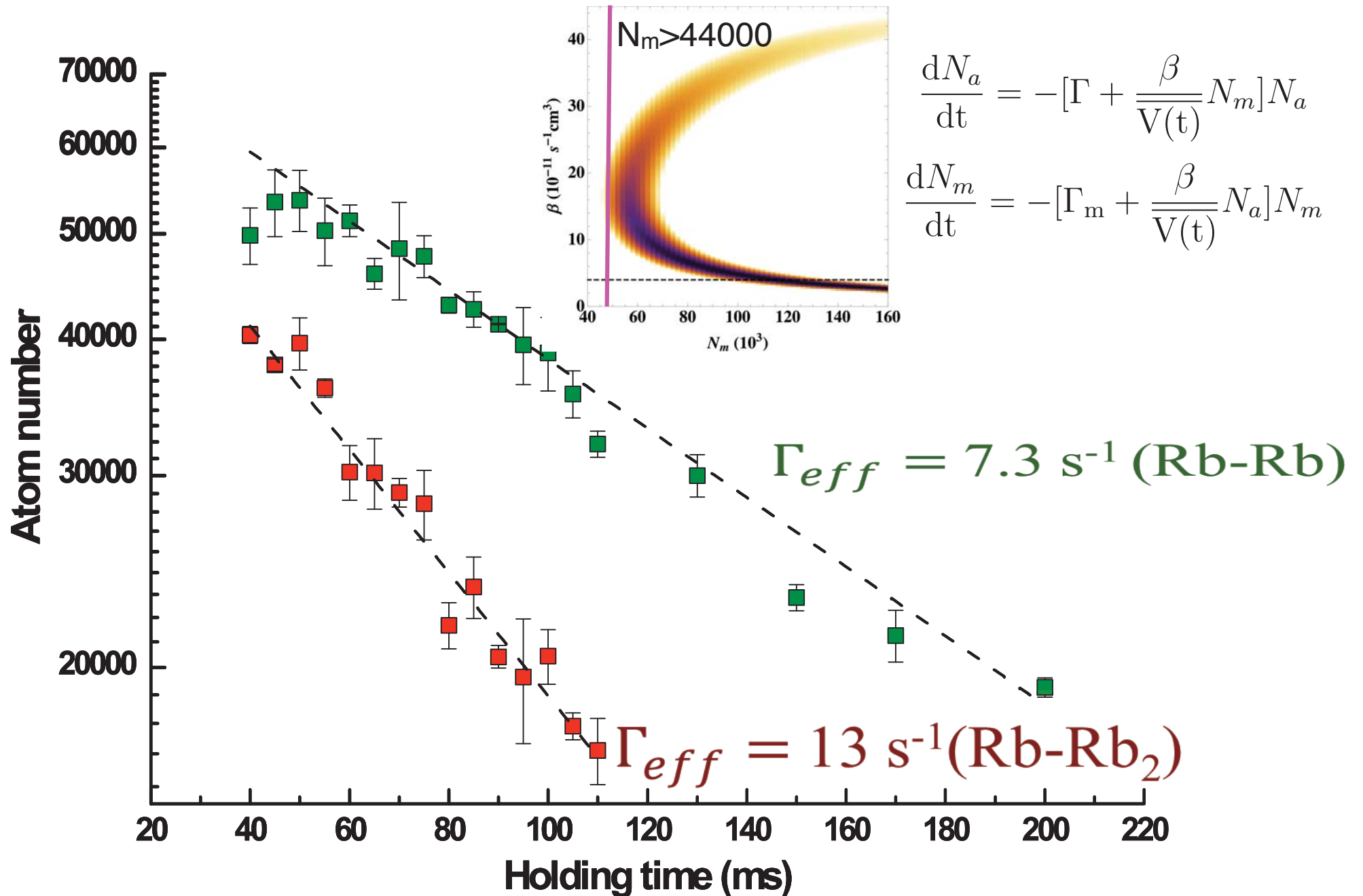
The evolution of the atomic temperature (size)

Atoms are heated to higher temperature due to the existence of molecule

PA

NO PA
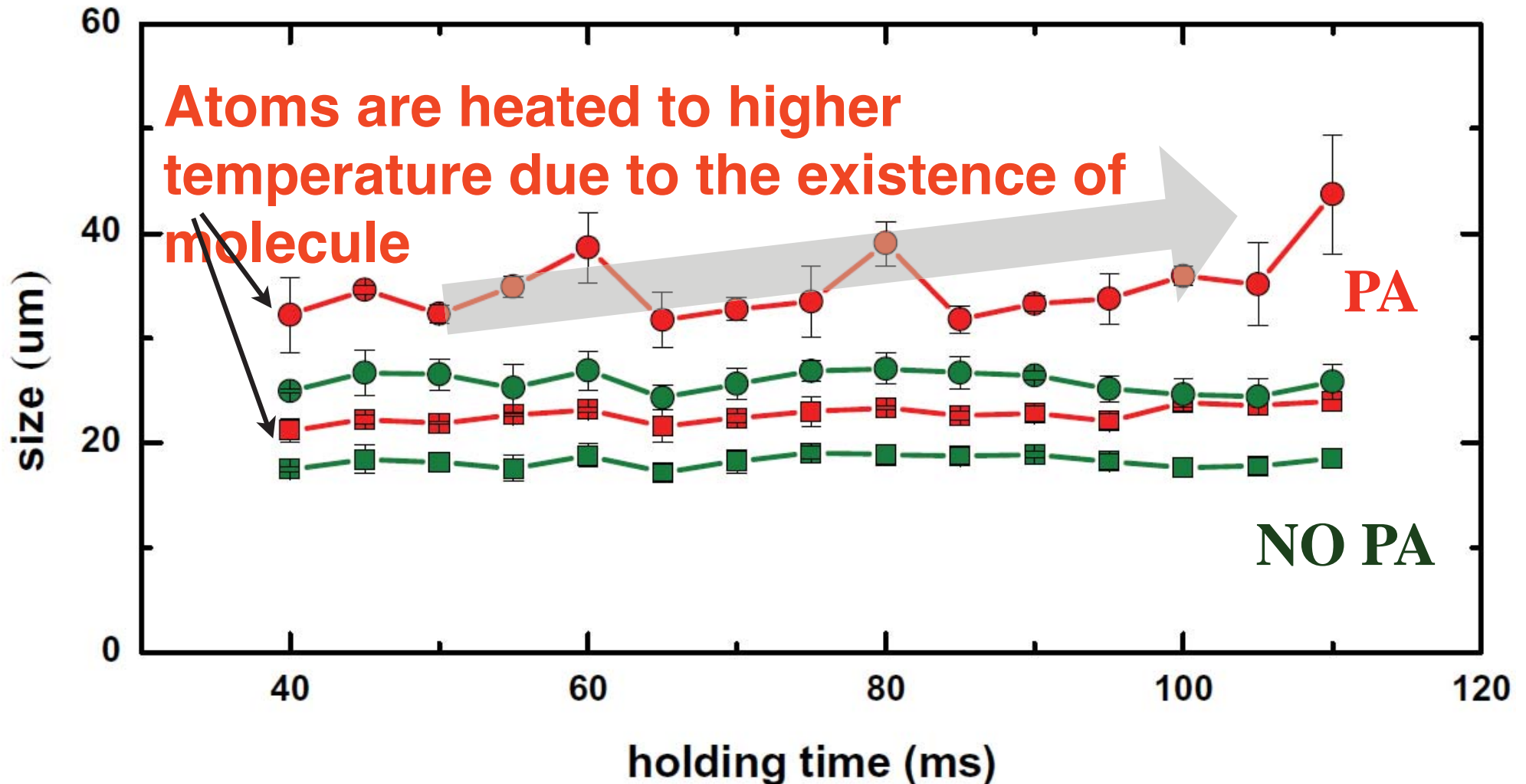
Conclusions

- A high number density of $^{85}Rb_2$ dimers $> 5.2\times10^{11}$ cm^{-3} has been produced and trapped in a crossed optical dipole trap. An order of magnitude higher then previous report.

- In the future, it could combine with the non-destructive atomic image technique for in-situ molecular observation and the magnetic control chemical reaction

FOR EVEN COLDER LOWER TEMPERATURE

- Reduce background collision, longer lifetime, further cooling process.

- Move the atoms to a higher vacuum using magnetic transfer
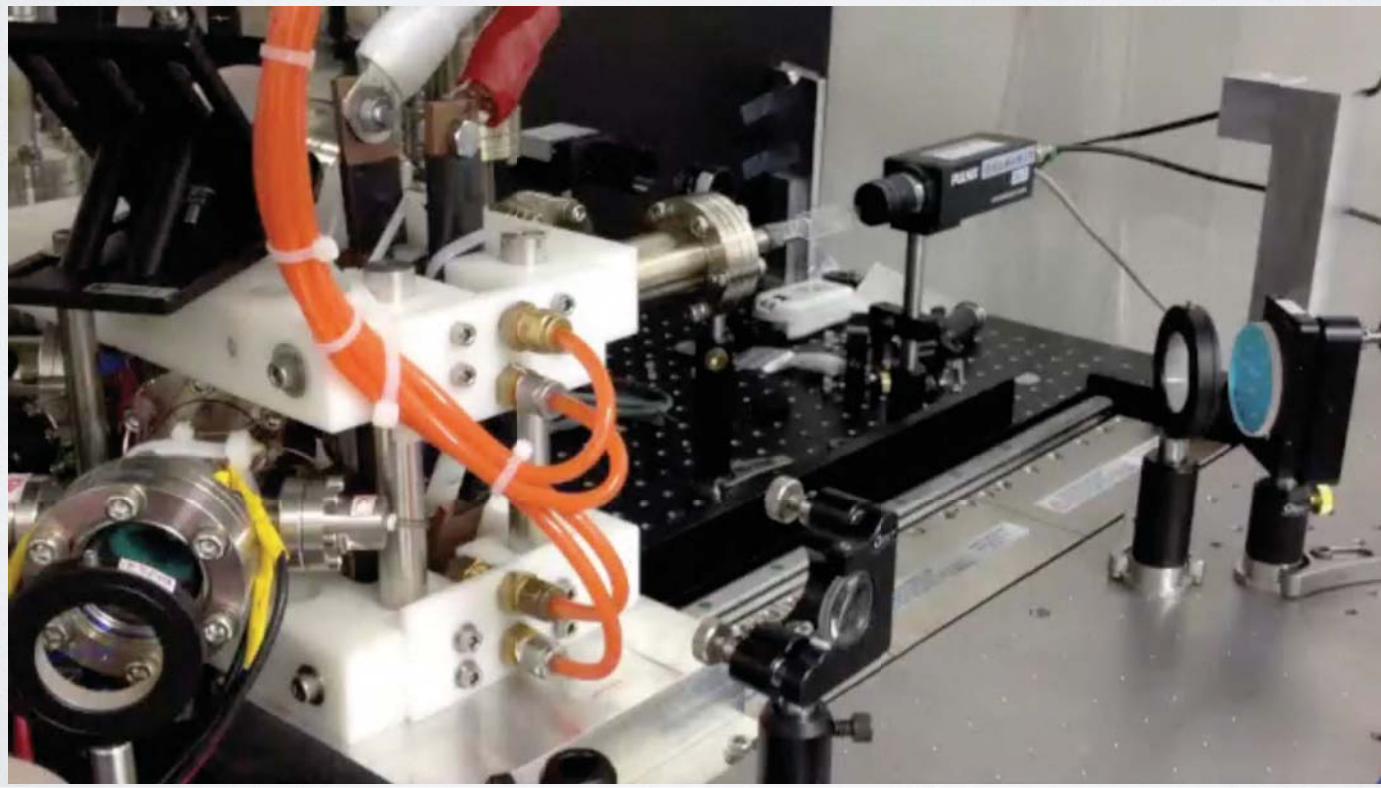
Table 1. List of Ultracold Polar Molecules of Current Experimental Interest[a]

polar molecule	city/country of the group	T	comments
$^{40}K^{87}Rb$	Boulder, CO, U.S.A.	~250 nK	GS, QR[1,114,119]
$^{40}K^{87}Rb$	Boulder, CO, U.S.A.	≃300 nK	not GS but coherent population transfer, QR[118]
$^{41}K^{87}Rb$	Tokyo, Japan	~130 μK	GS, not QR[120]
$Sr^{19}F$	New Haven, CT, U.S.A.	~300 μK	GS, not QR[91,121]
$^{85}Rb^{133}Cs$	New Haven, CT, U.S.A.	~100 μK	GS but not ground rotational state, not QR[107]
$^{7}Li^{133}Cs$	Heidelberg, Germany	~260 μK	GS, but not coherent population transfer, not QR[101]
$^{85}Rb^{133}Cs$	New Haven, CT, U.S.A.		not GS[122]
$^{39}K^{85}Rb$	São Paulo, Brazil	~150 μK	not GS[123]
$^{23}Na^{133}Cs$	Rochester, NY, U.S.A.	~260 ± 130 μK	not GS[124−126]
$^{39}K^{85}Rb$	Storrs, CT, U.S.A.		not GS[127]
$^{174/176}Yb^{87}Rb$	Düsseldorf, Germany		not GS[128]
$^{85}Rb^{133}Cs$	Pisa, Italy		not GS[129]
$^{6}Li^{40}K$	Paris, France		not GS[130]
$^{85}Rb^{133}Cs$	Taiyuan, China.		not GS[131]
$^{87}Rb−^{133}Cs$	Innsbruck, Austria		AAM[115−117]
$^{87}Rb−^{133}Cs$	Durham, U.K.		AAM[132,133]
$^{23}Na−^{40}K$	Cambridge, MA, U.S.A.		AAM[134]
$^{6}Li−^{85}Rb$	Vancouver, Canada		AAM[135]
$^{6}Li−^{173/174}Yb$	Kyoto, Japan		AAM[136]
$^{6}Li−^{174}Yb$	Seattle, WA, USA		AAM[137,138]
$^{87}Rb−^{176}Yb$	Düsseldorf, Germany		AAM[139]
$^{6}Li−^{23}Na$	Cambridge, MA, U.S.A.		AAM[140]