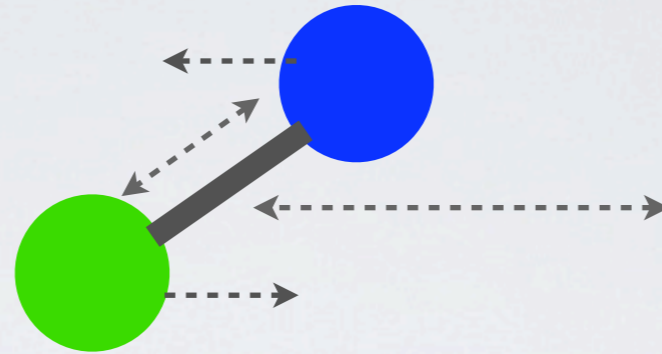


COLD MOLECULE AND PHOTOASSOCIATION

Yi-Wei Liu
Physics, NTHU
2010.10.25



WHAT IS COLD MOLECULE?



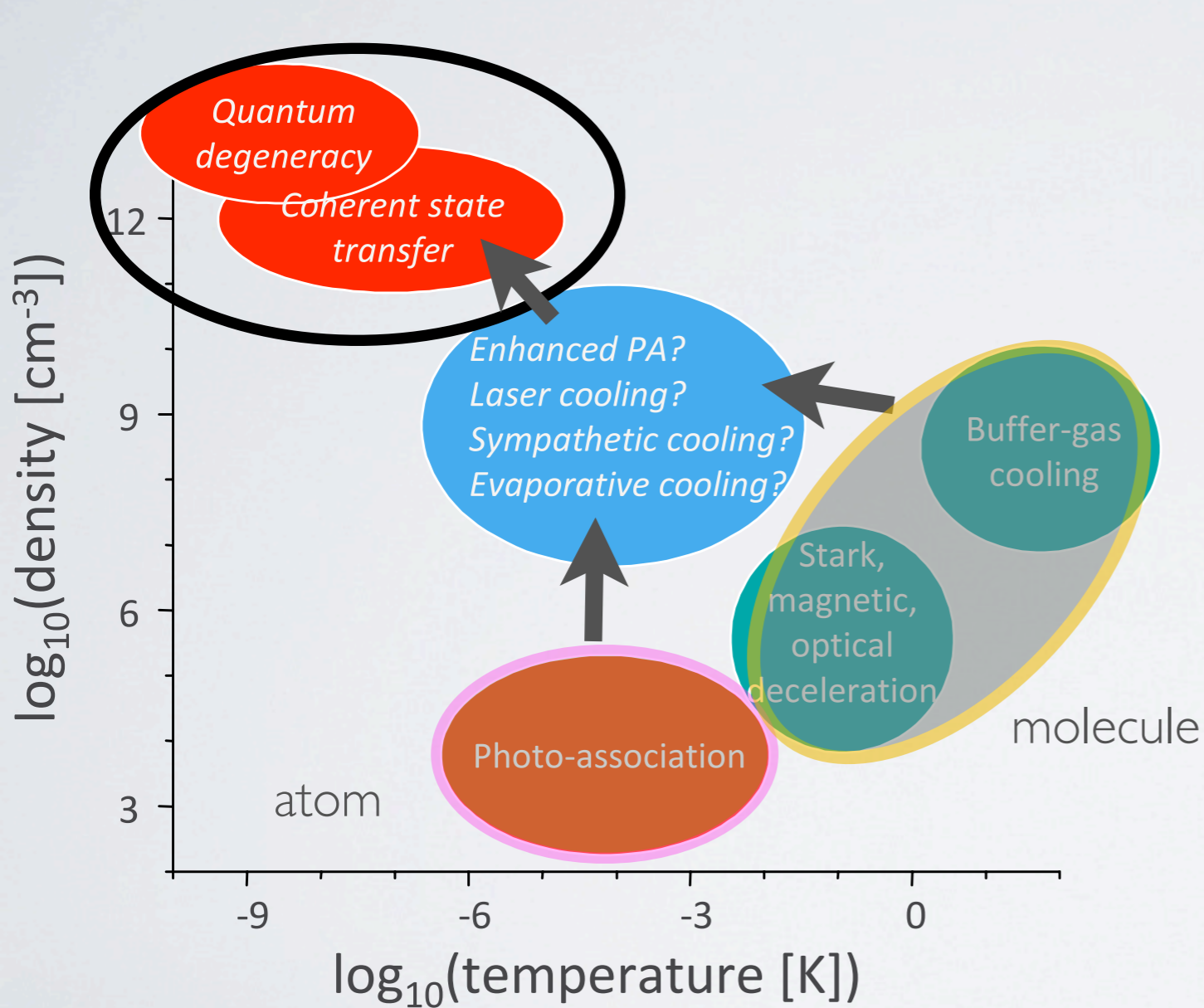
- A **neutral bound** system with more than one atom has a very low kinetic energy (**translational**, **vibrational**, **rotational**)
- K_2 , KRb, Rb₂, Cs₂....

New problems in molecular system

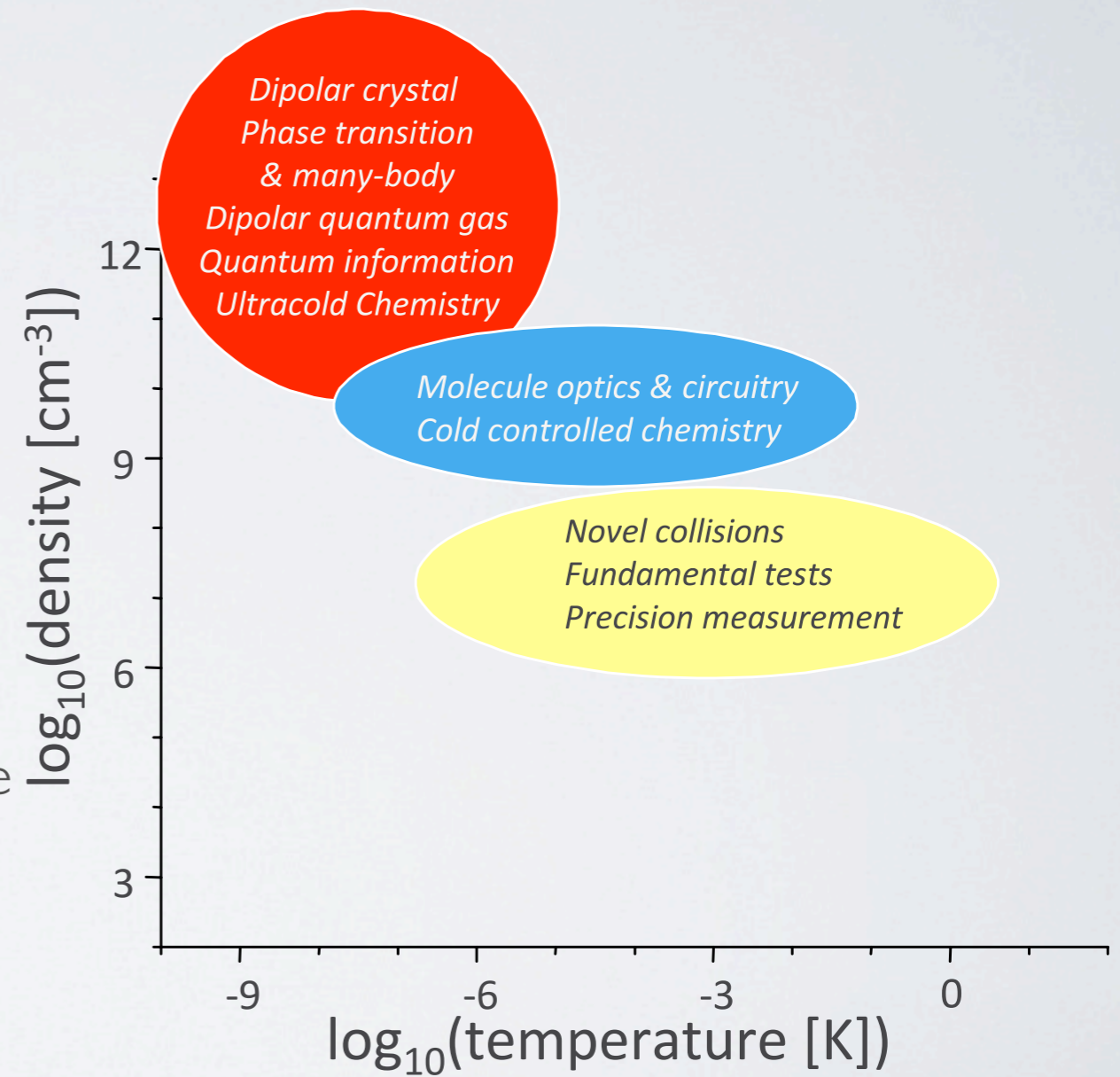
WHY COLD MOLECULE?

- **Fundamental research**: fundamental constant, EDM, PNC
- **New era of chemistry**: superchemistry
- **Quantum simulator** for solid state physics
- **Quantum Computer**.

TEMPERATURE, DENSITY AND COOLING



various cooling techniques



physics using cold molecule

TWO MAINSTREAMS IN COLD MOLECULE INDUSTRY

- Cool down a molecule
- Produce molecule from cold atoms



Association: Perform chemistry using very cold ingredients

HEAT INCREASES INTERACTION, BUT WE ARE GOING TO COOL EVERYTHING

Heat helps chemical reaction, because:

1. higher collision rate

2. higher kinetic energy to penetrate chemical energy barrier



Then, why shall we go cold?

WHAT AND WHY “ULTRACOLD”

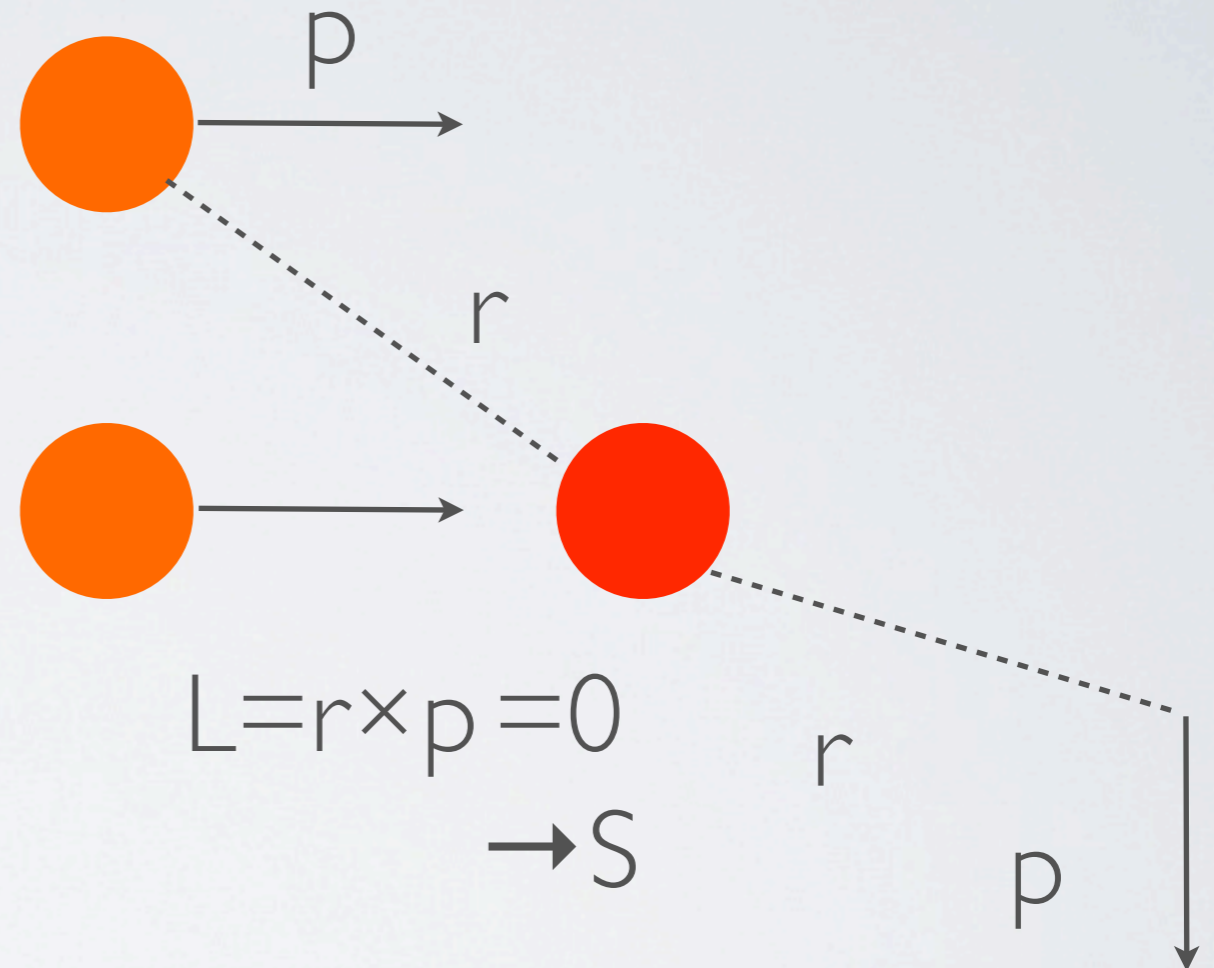
- The ingredients (atoms) must be cold to have cold products (molecules).
- “Ultracold” \rightarrow S wave scattering \rightarrow enhance inelastic collision and chemical reaction. (increase by 10^3) \leftrightarrow It contradicts with our experience in chemical practice.

SCATTERING

Scattering of various partial wave

The collision complex can be decomposed according to the **angular momentum**. s, p, d ...

During the process of the collision, the angular momentum must be **conserved**.

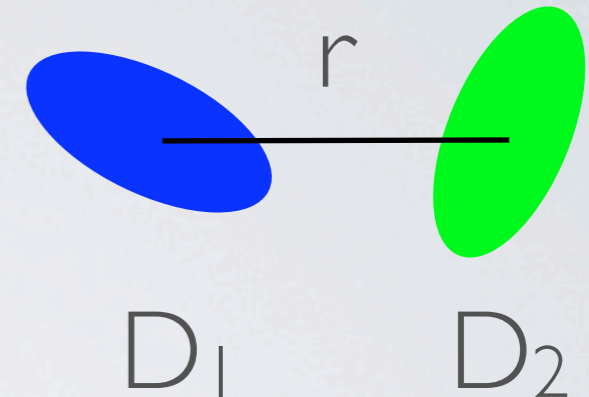


Head-to-Head collision

ULTRACOLD COLLISION

Dipole-Dipole interaction

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



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

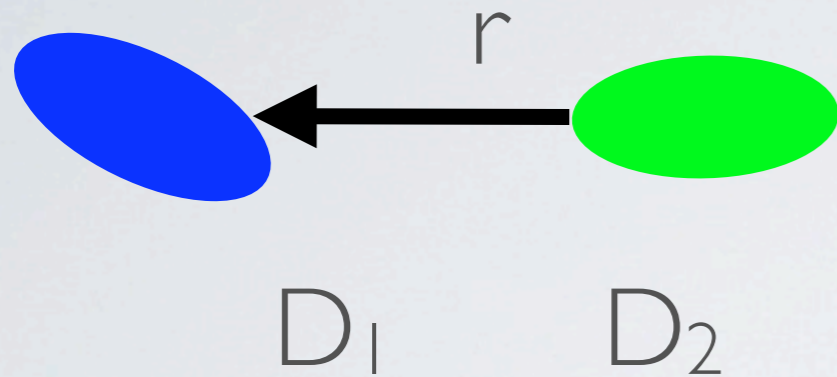
Wavefunction of the complex during collision: $|\mathbf{l}, \mathbf{m}\rangle$

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

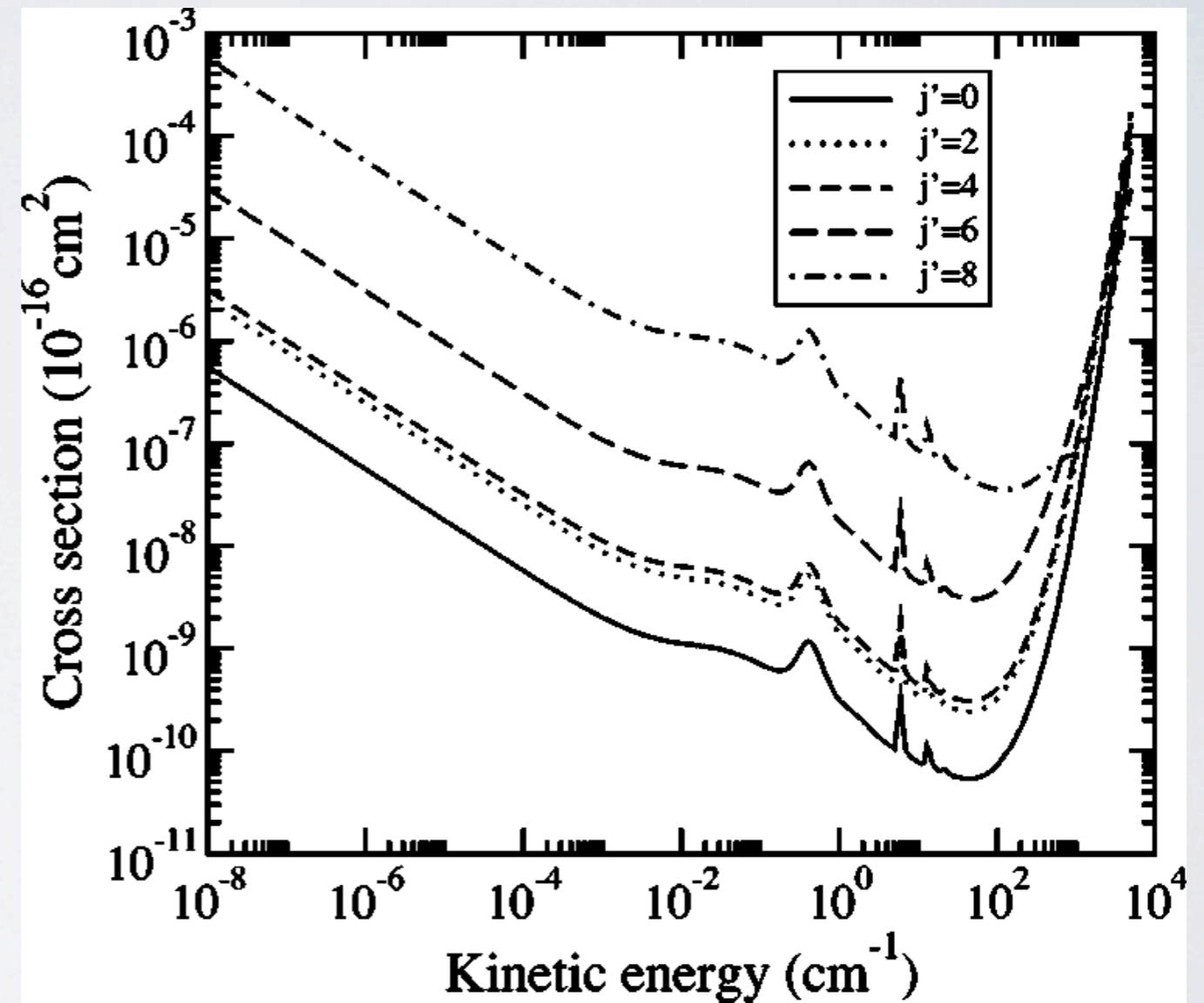
$l=0$, s-wave scattering

dipole-dipole interaction is vanished
inelastic collision is enhanced

CROSS SECTION ENHANCEMENT

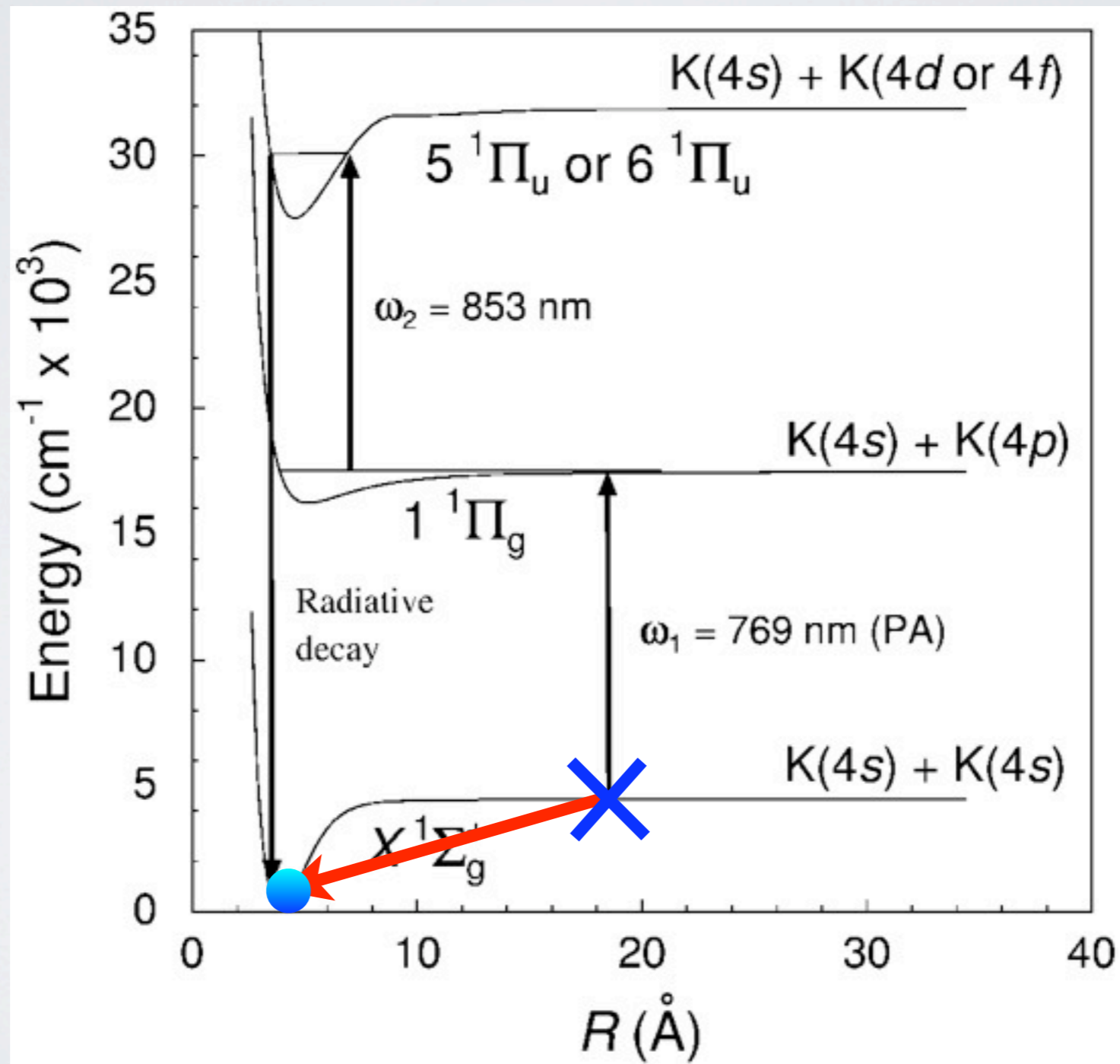


Head-to-Head collision
Averaged force is zero
No potential barrier
Furthermore, external
field can be used to
control the alignment, then
the interaction of atoms



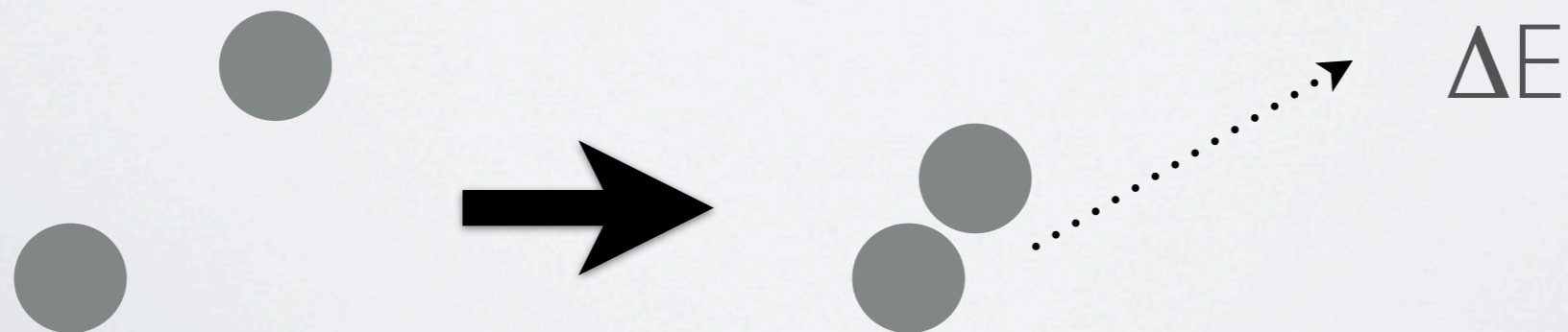
H_2 -Ar collision v.s temperature

OUR ULTIMATE GOAL : TO THE GROUND STATE



FROM COLD ATOMS TO COLD MOLECULES

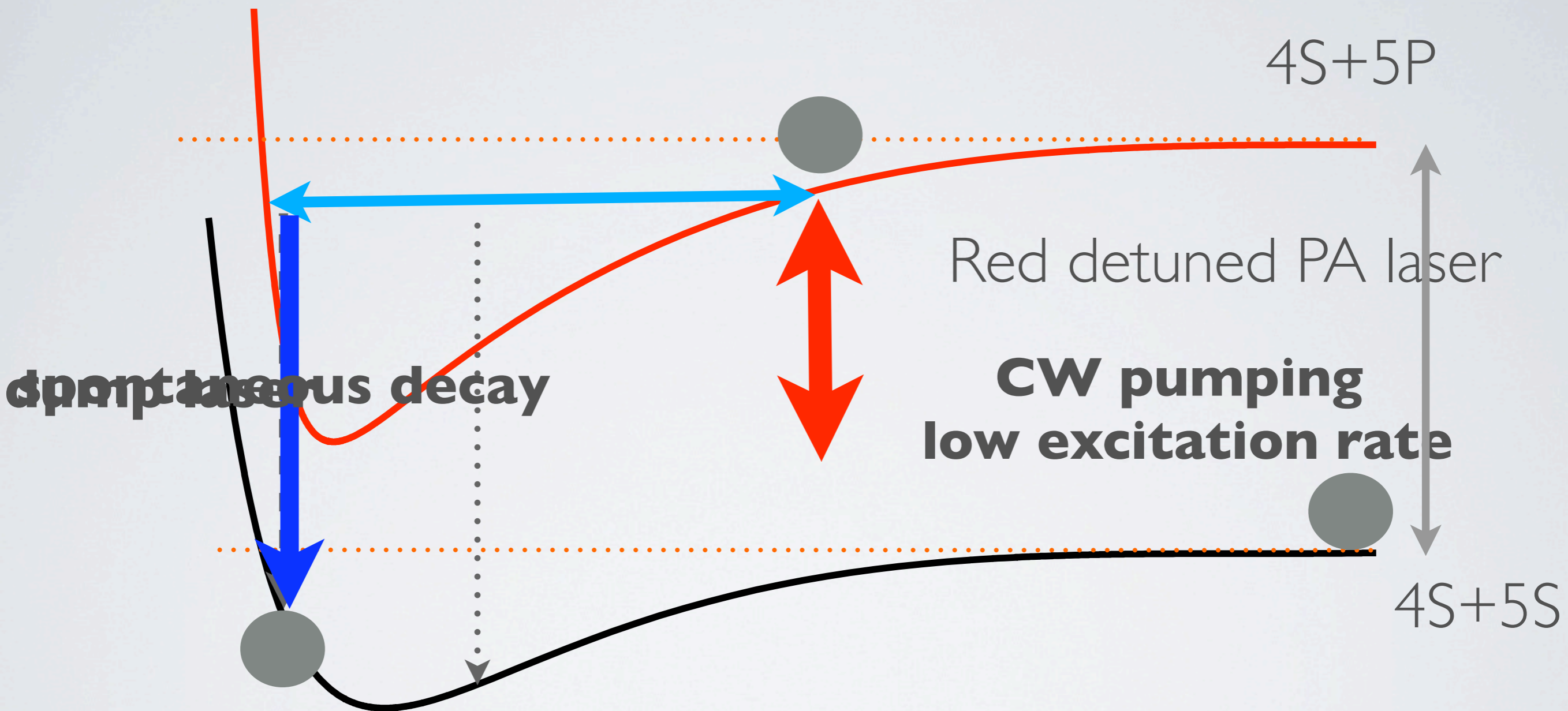
- Energy conservation: How to remove internal energy?
- Vibration of neutral particles can not emit photons \rightarrow no radiative decay!
- The third party is needed: photon or particle (three-body collision)



AVAILABLE APPROACHES

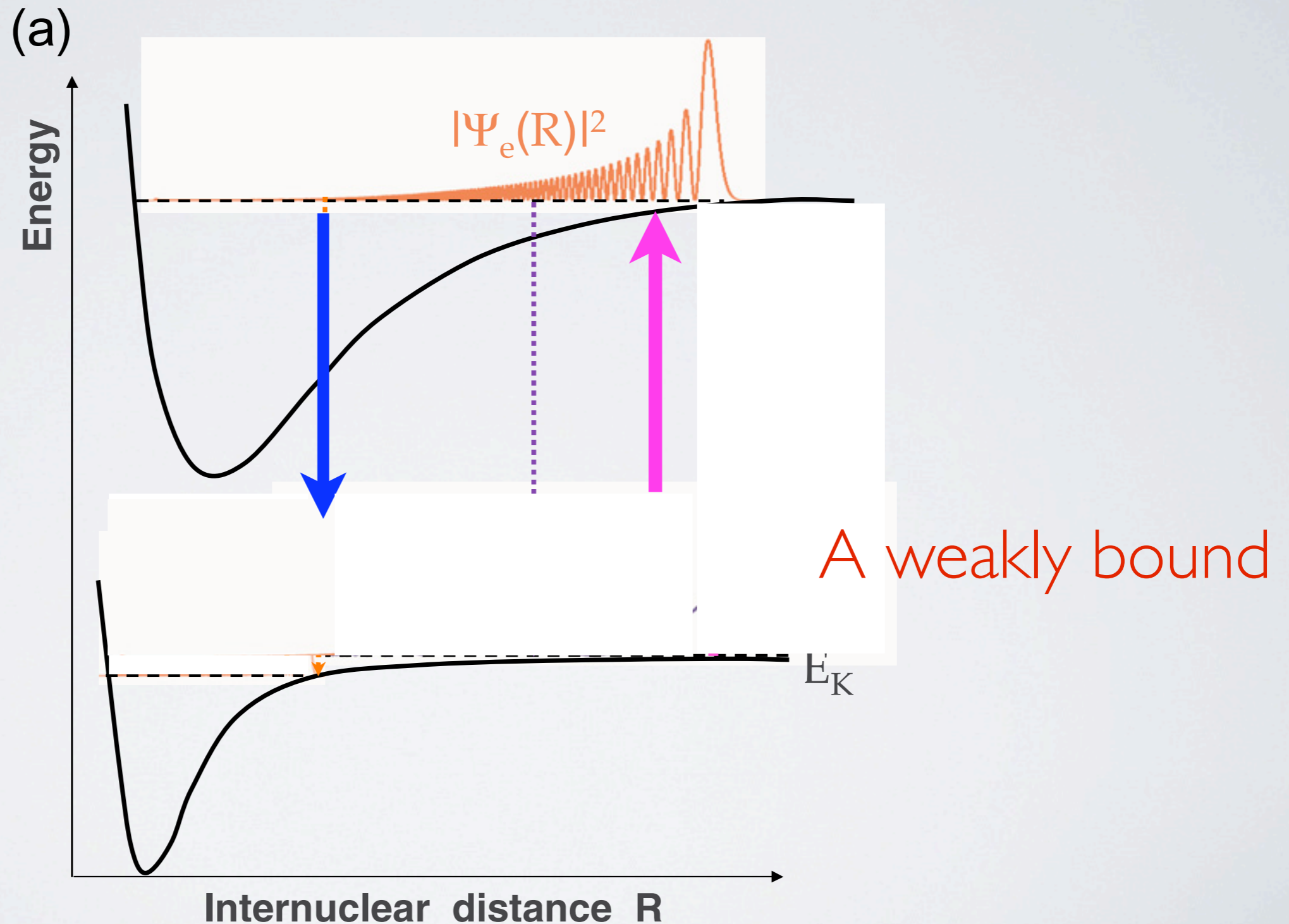
- Feshbach resonance : tuning interaction by external field, transfer kinetic energy to internal hyperfine(**Adiabatic**)
- Photoassociation : using photons to take out energy.(**Adiabatic or Not**)

PHOTOASSOCIATION



- Low rate, complicate laser system

A WEAKLY BOUND SYSTEM CAN INCREASE PA RATE



STIMULATED RAMAN ADIABATIC PASSAGE: **STIRAP**

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

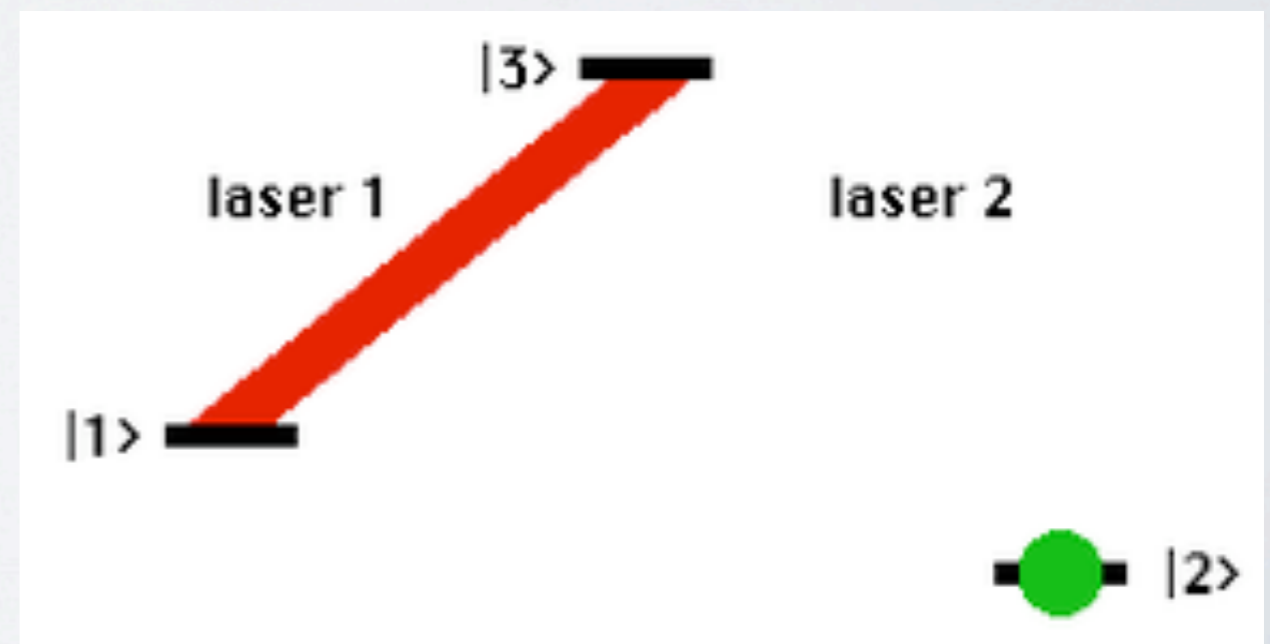
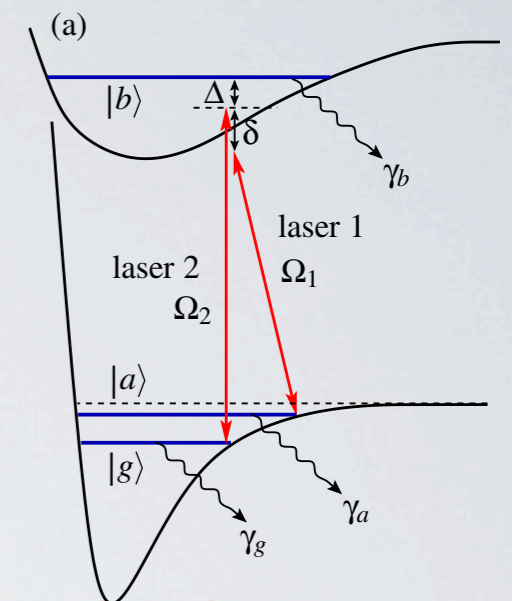
$|B\rangle = (b_1 |2\rangle + b_2 |3\rangle) e^{i\beta t}$: coupled by laser 2

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

then $|1\rangle$ and $|2\rangle$ are coupled into a **coherent dark state**

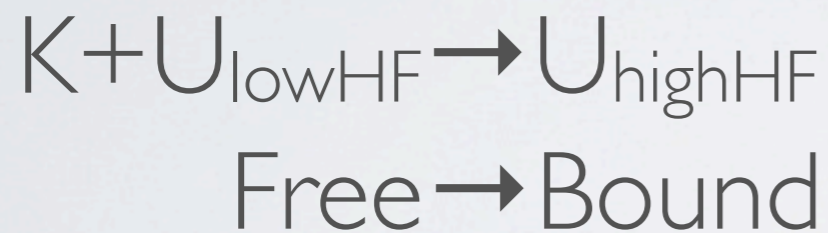
$$|\text{coherent dark state}\rangle = (r_1 |1\rangle + r_2 |2\rangle) e^{i\gamma t}$$

Population can be transferred between $|1\rangle$ and $|2\rangle$ with no access to $|3\rangle$, therefore no spontaneous decay



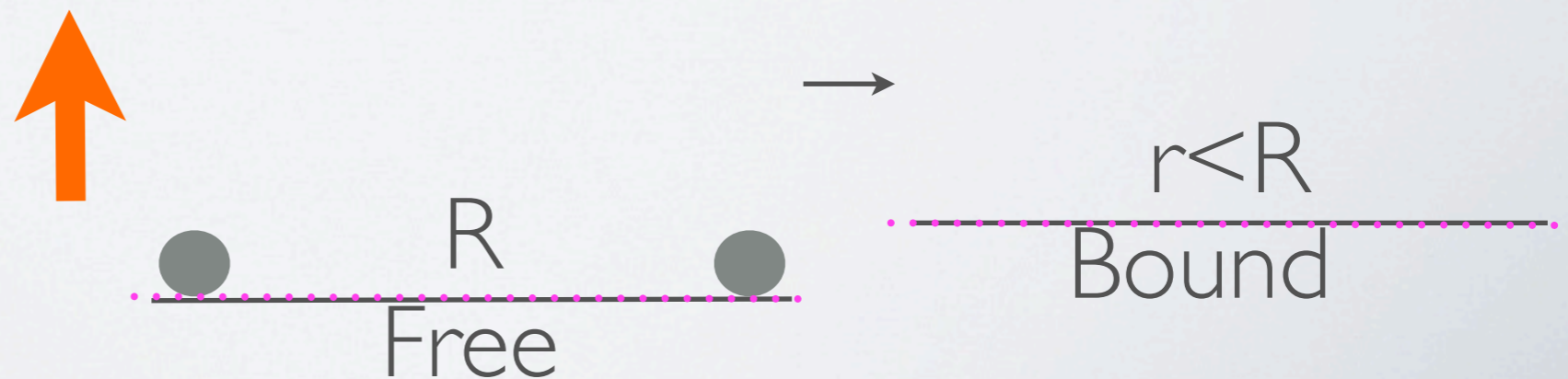
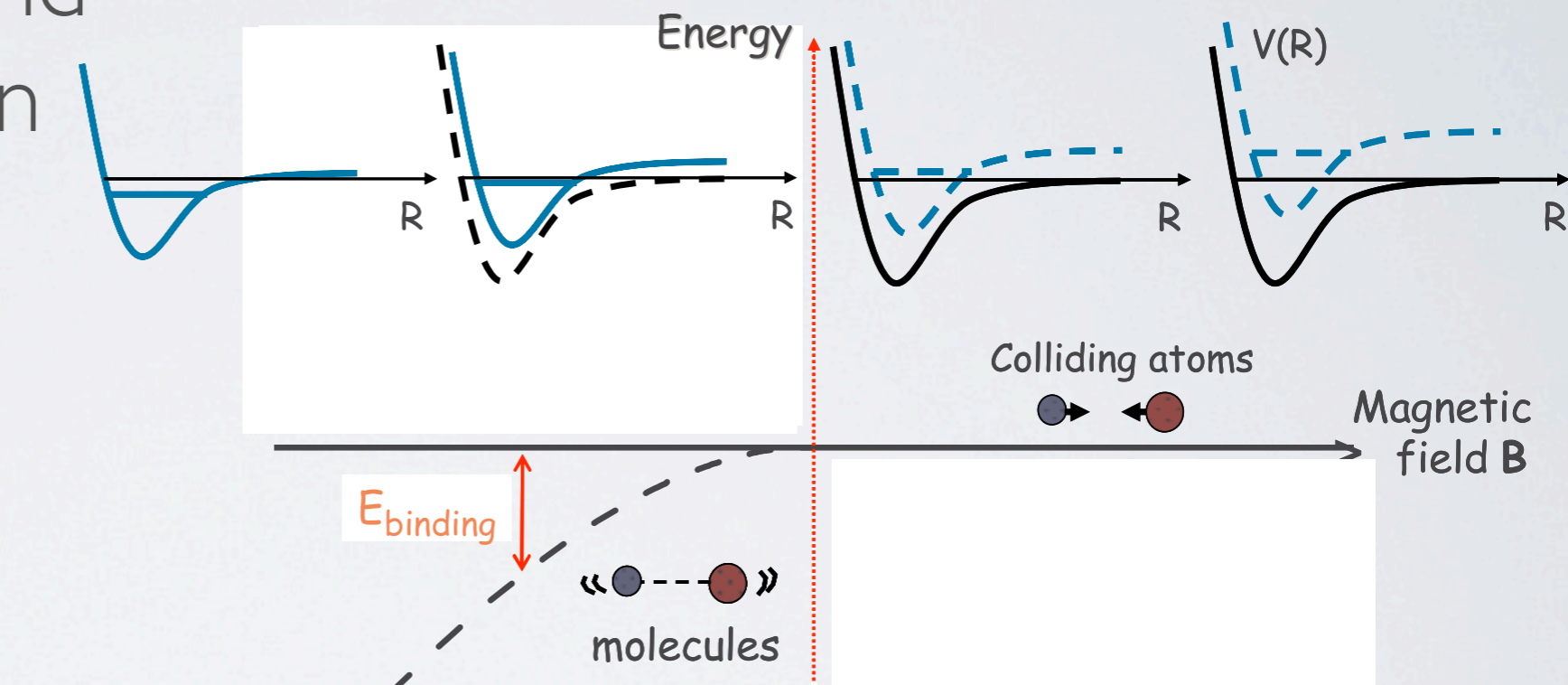
FESHBACH RESONANCE

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

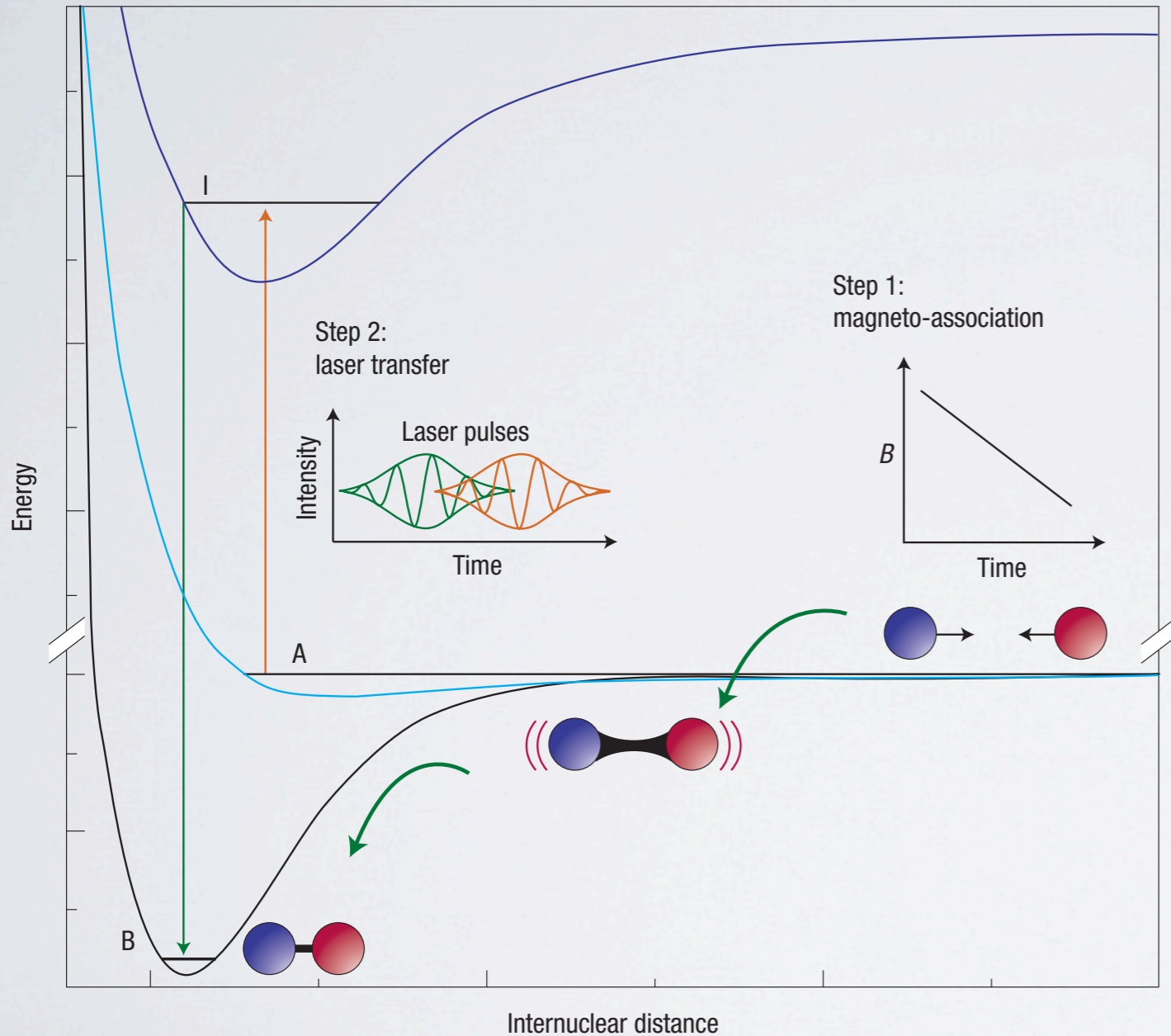


The external fields can be:

magnetic,
optical,
electrical



THE BEST SOLUTION SO FAR



- **Feshbach** resonance + **STRAP** (stimulated Raman adiabatic passage)
- Form a very large (R , and high \mathbf{v}) 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.....

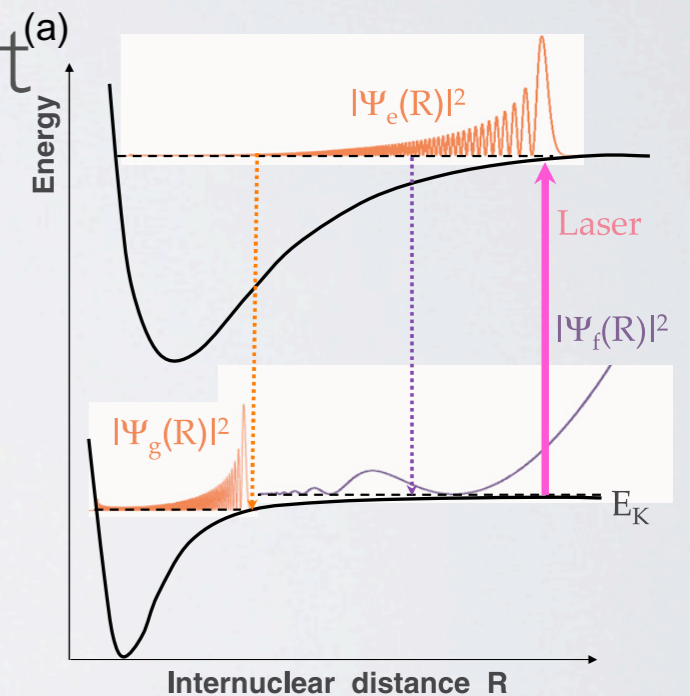
A PROPOSAL TO INCREASE PA RATE

- 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!

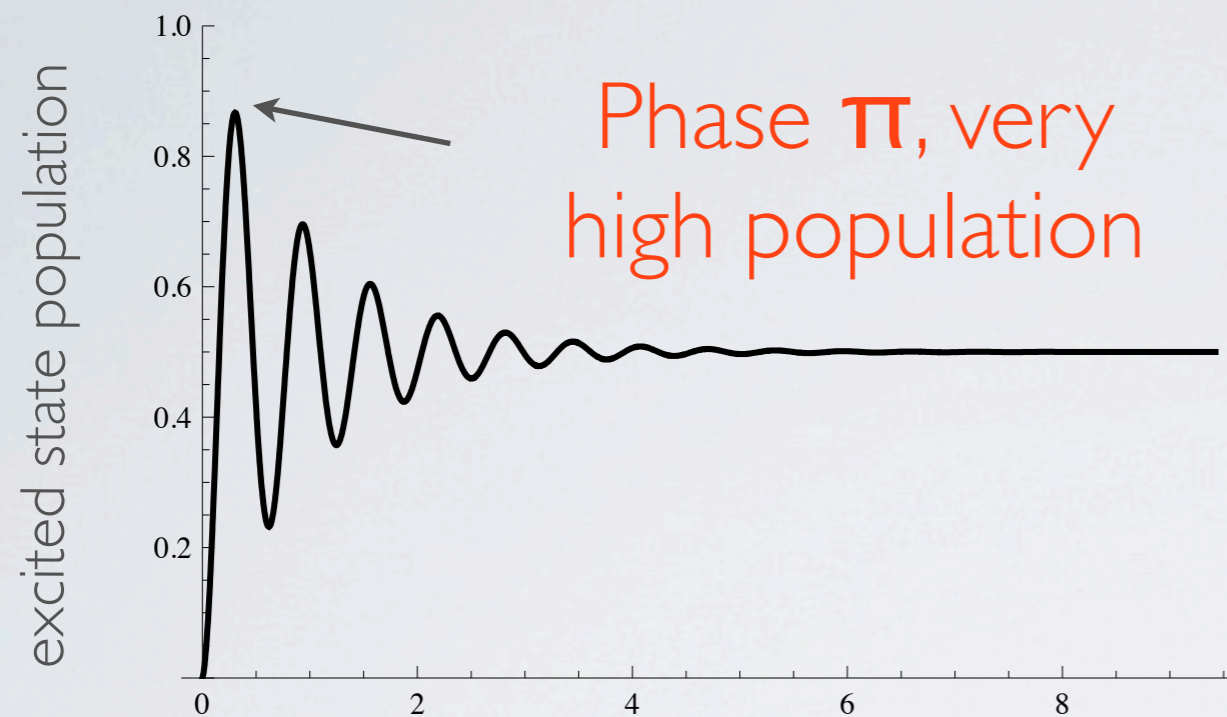


⇒ The femtosecond laser

BANDWIDTH OF PUMP LASER

- The typical linewidth of molecular absorption without overlap with neighboring band →
 $\Delta\nu = 10^2 - 10^3 \text{ cm}^{-1}$
- By uncertainty principle (Fourier transform-limited),
 $\tau = \text{femtosecond}$

MAXIMUM INVERSION USING π

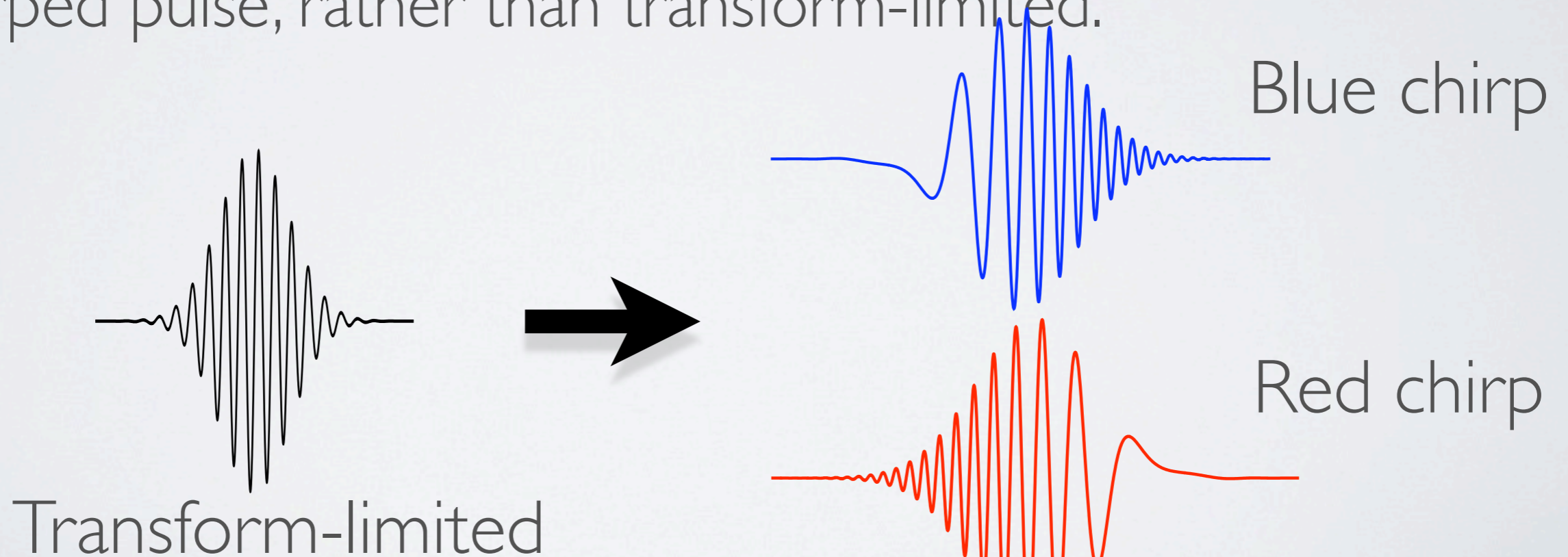


Rabi Oscillation

- Short pulse to perform π population transfer.
- $\tau E_0 \mu / \hbar = \pi$
 - Problems: very large $E_0 \sim 10^{12}-10^{14}$ W/cm². Many subtle effects should be taken into account, such as multi-photon transition

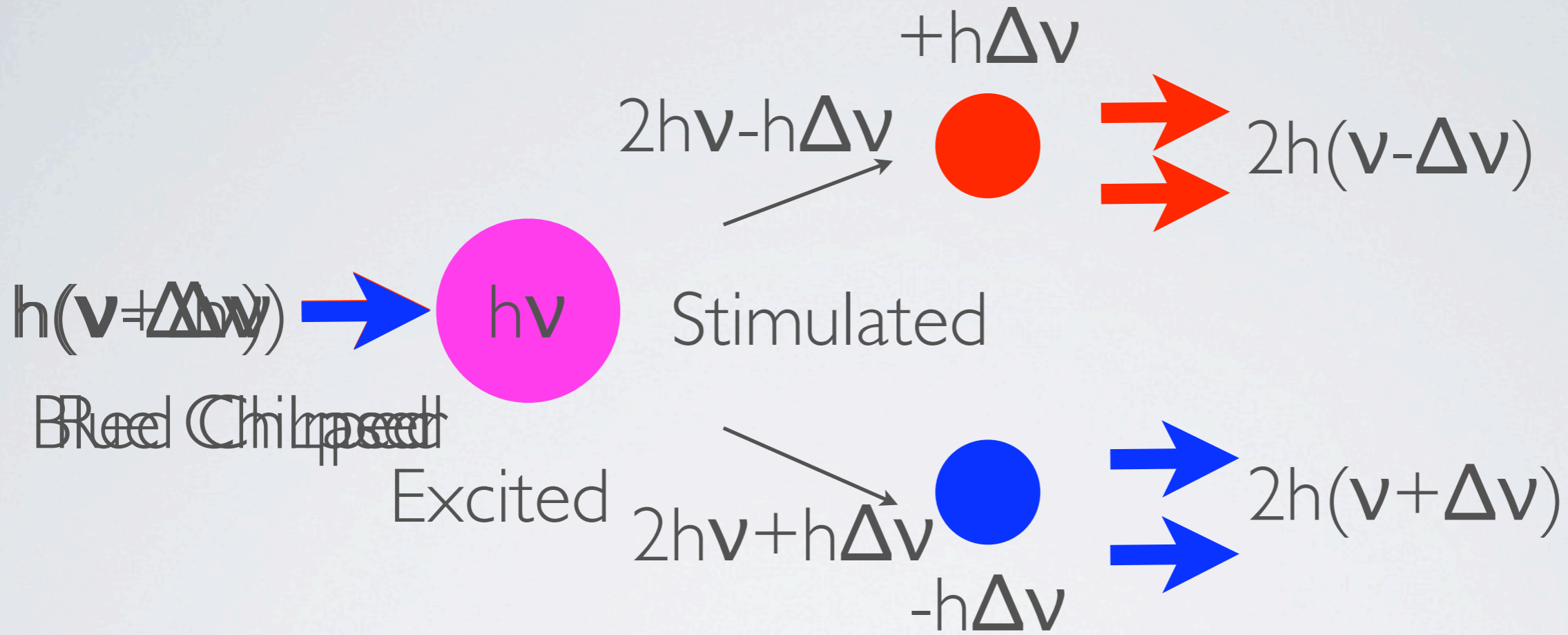
CHIRPED FEMTOSECOND LASER

- $\tau E_0 \mu / \hbar = \pi$, a longer τ can lower required E_0
- To maintain the same $\Delta\nu$ with a long $\tau (> 1/\Delta\nu)$, we need a chirped pulse, rather than transform-limited.



Longer pulse with the same power spectrum

BLUE OR RED?



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

- just like Raman cooling

CHIRP EXPERIMENT

PRL 96, 173002 (2006)

PHYSICAL REVIEW LETTERS

week ending
5 MAY 2006

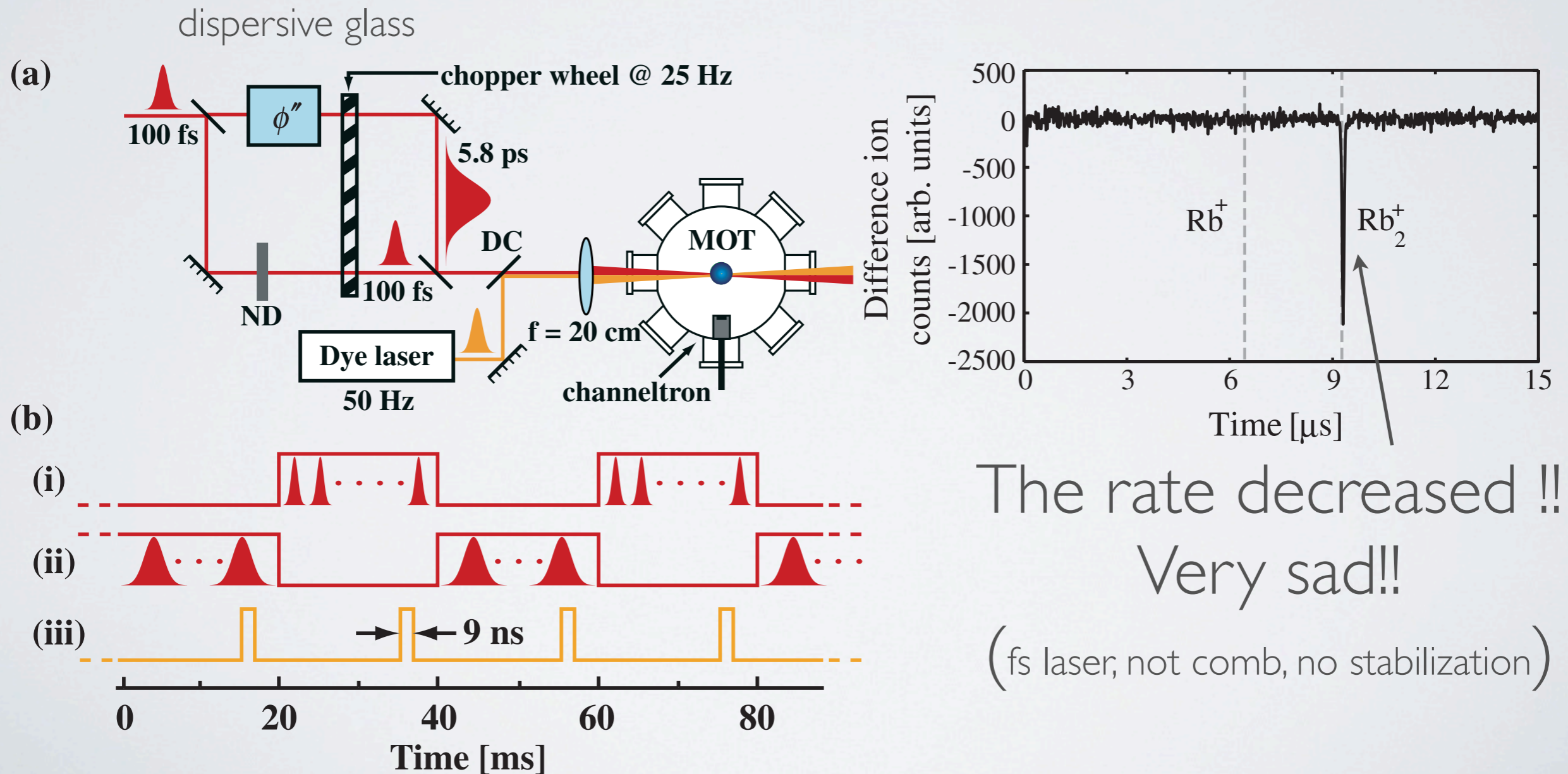
Coherent Control of Ultracold Molecule Dynamics in a Magneto-Optical Trap by Use of Chirped Femtosecond Laser Pulses

Benjamin L. Brown,^{1,2,*} Alexander J. Dicks,¹ and Ian A. Walmsley¹

¹Clarendon Laboratory, Department of Physics, University of Oxford, Oxford, OX1 3PU, United Kingdom

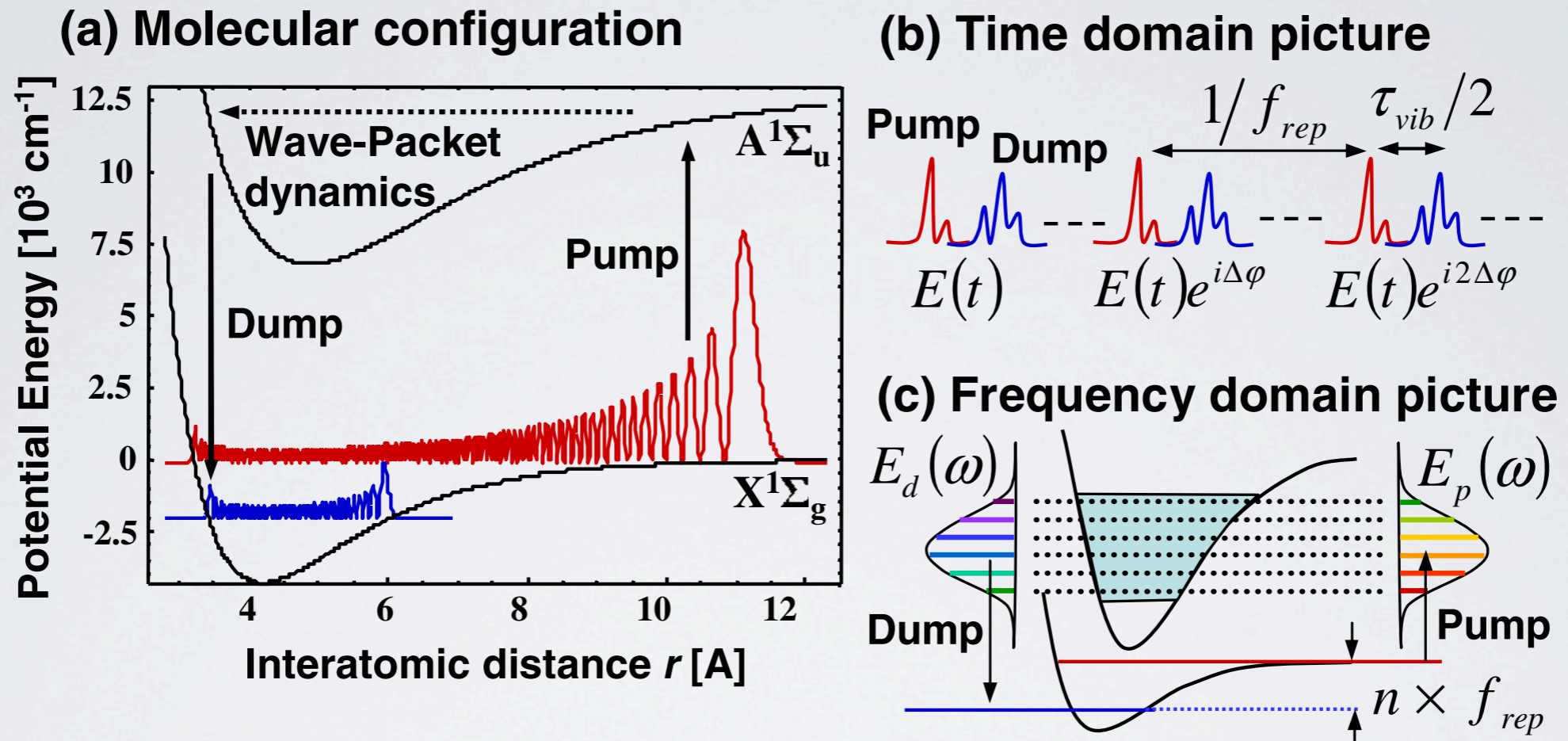
²The Institute of Optics, University of Rochester, Rochester, New York 14627, USA

(Received 13 September 2005; published 5 May 2006)



COMB LASER

CONTROL PHASE OF WAVE PACKET USING COHERENCE BETWEEN PULSES



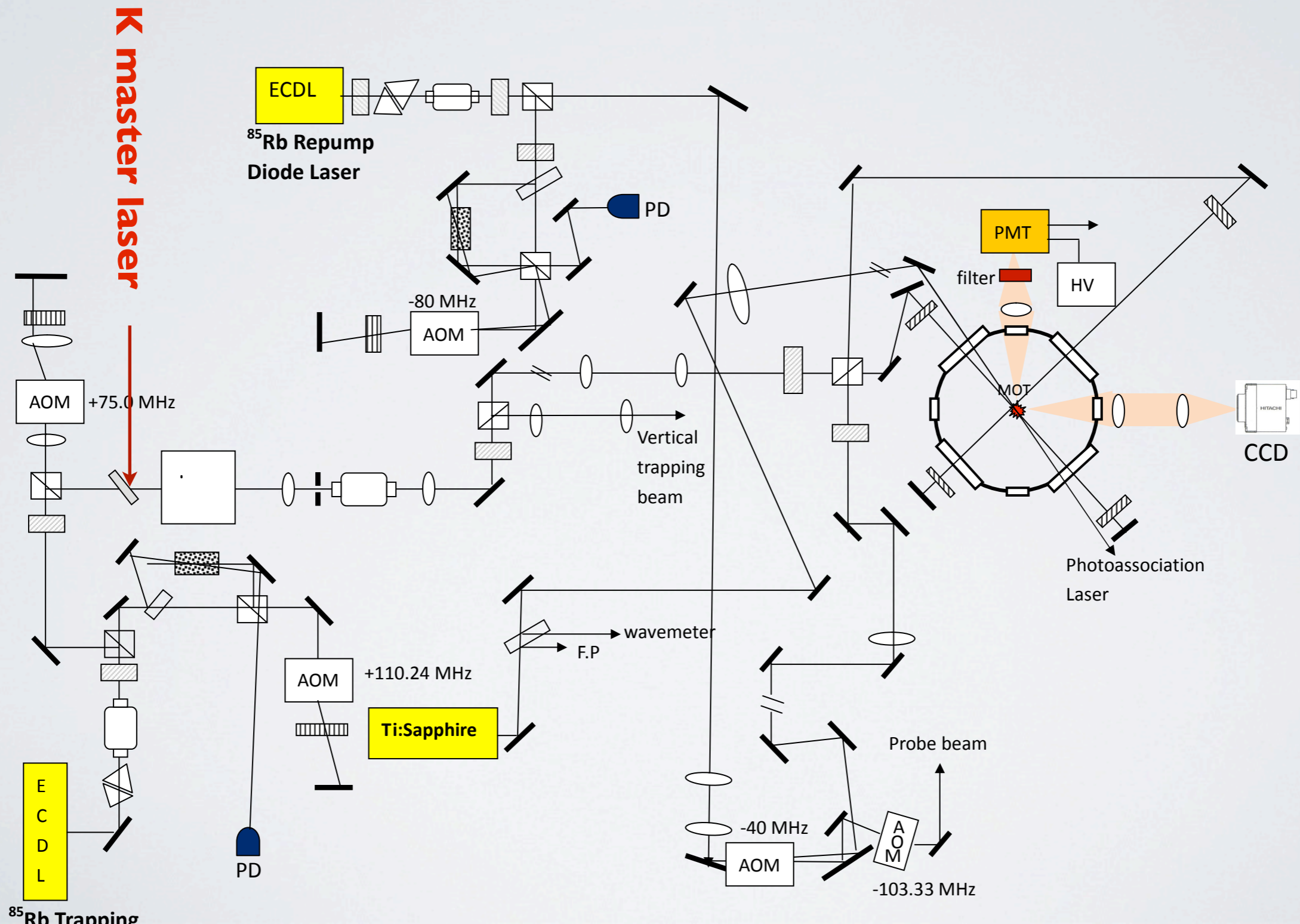
- The phase of the final products from each pulse is fixed, due to the coherent nature of comb laser.
- $|\Phi\rangle_f = e^{i\theta}|\Phi\rangle + e^{i2\theta}|\Phi\rangle + e^{i3\theta}|\Phi\rangle + \dots$ interference of grating

Proposed by Pe'er and et al, JILA, PRL, 2007

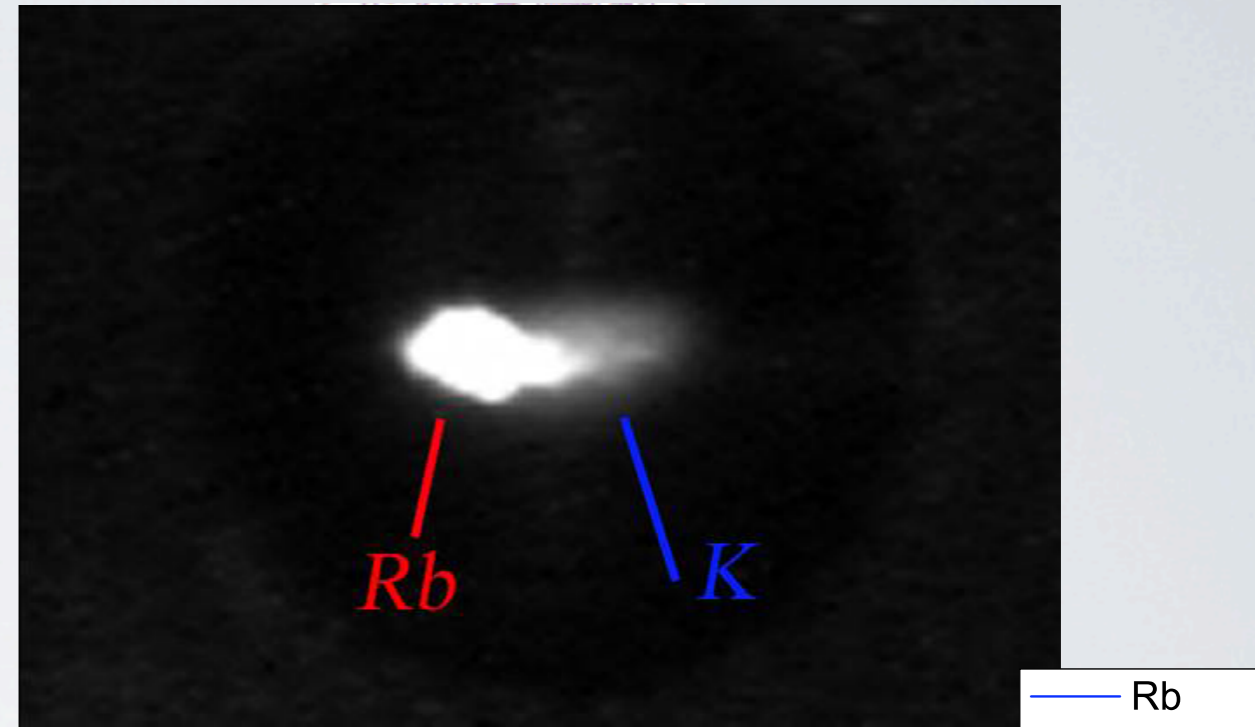
OUR APPROACH

- KRb mixture
- Polarization Gradient cooling
- Dipole trap
- Feshbach resonance combination
- Photo-association (pulsed coherent Raman)

LASER SYSTEM FOR DOUBLE SPECIES COOLING SYSTEM



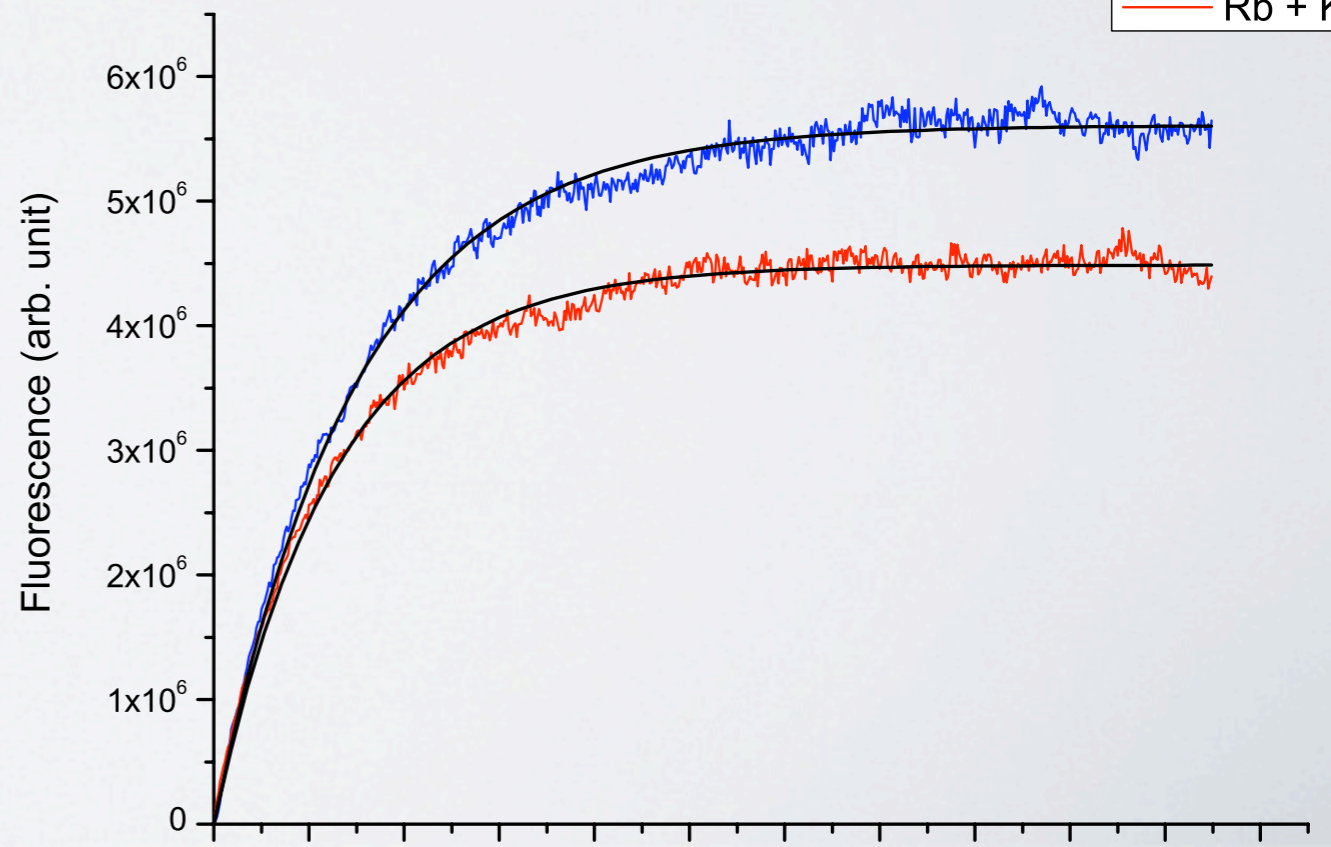
K AND Rb MIXTURE



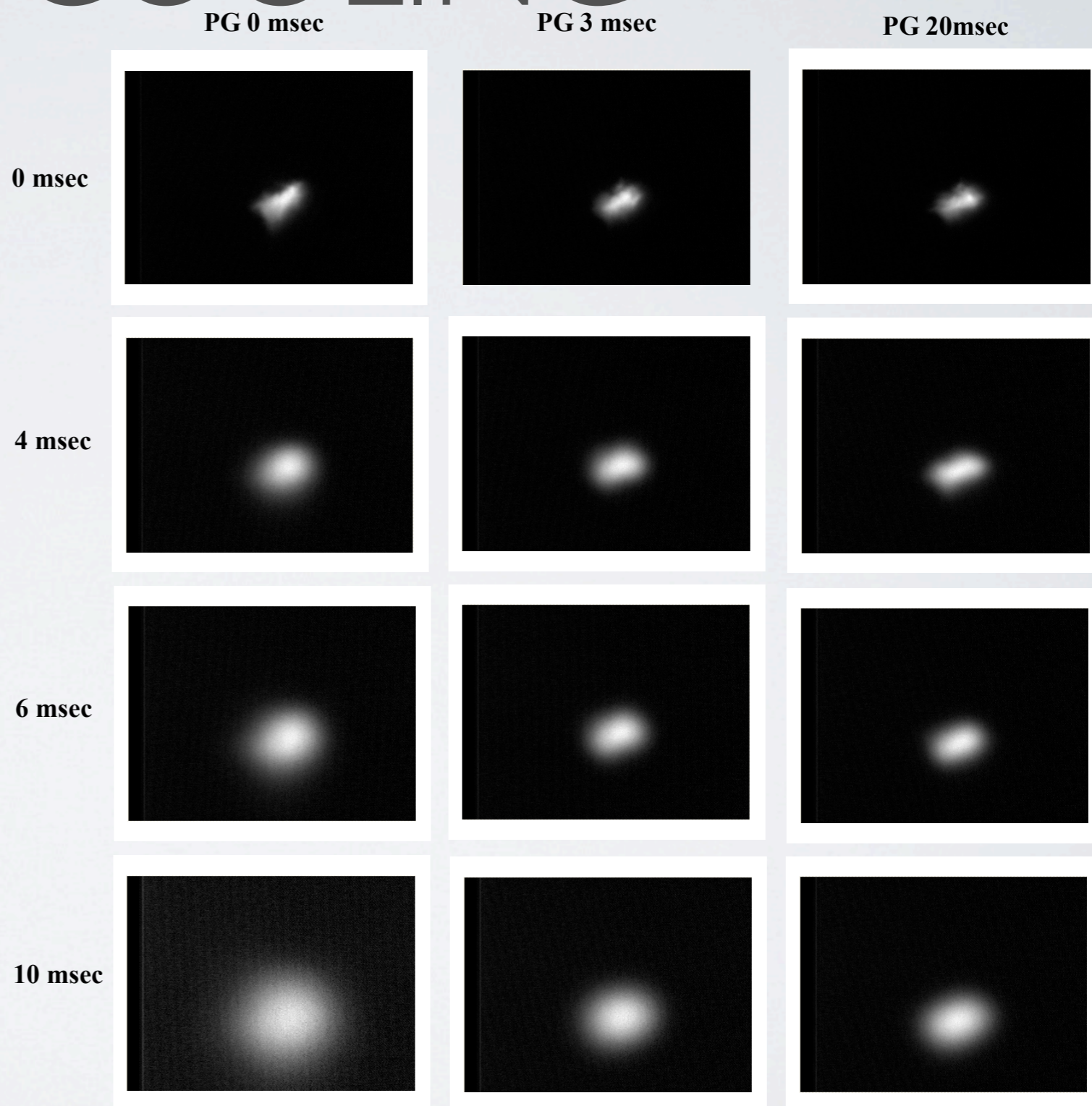
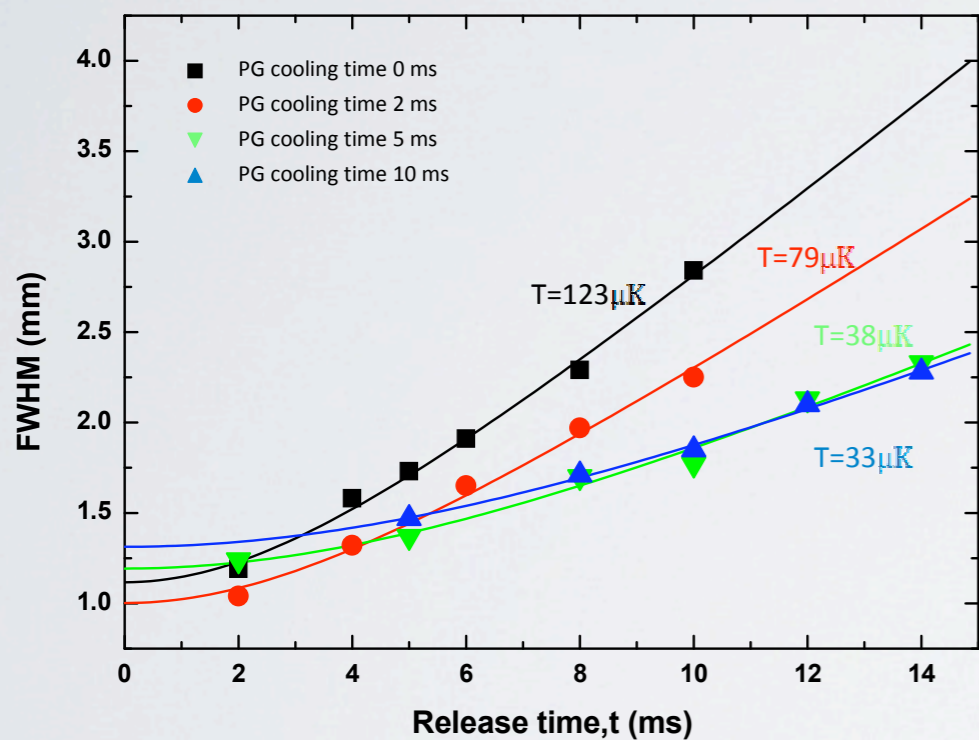
$$\frac{dN_K}{dt} = L - \gamma N_K - \beta n_K N_K - \beta' n_{Rb} N_K$$

$$N_K = N_0 \{1 - \exp[-(\gamma + \beta n_K + \beta' n_{Rb})t]\}$$

$$N_0 = L / (\gamma + \beta n_{Rb} + \beta' n_K)$$



A LITTLE BIT COLDER USING PG COOLING



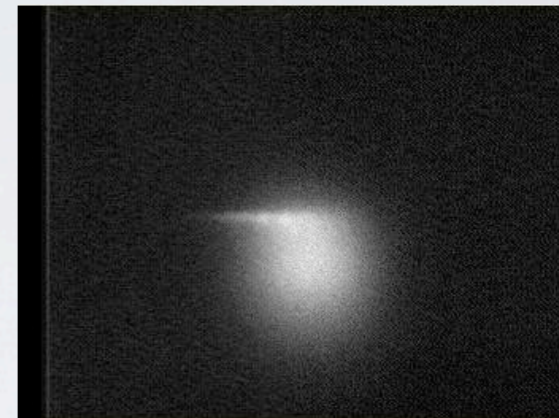
DIPOLE TRAP FOR ULTRACOLD ATOM



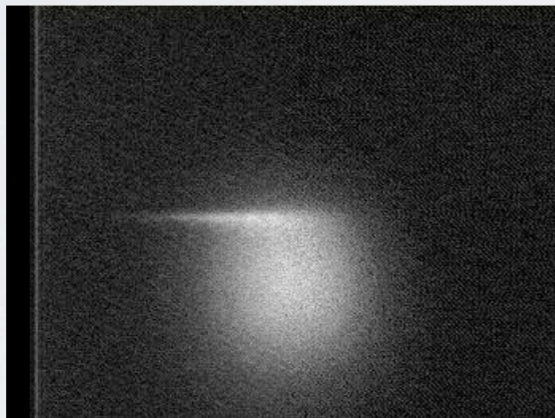
release time 0 ms



release time 5 ms



release time 10 ms



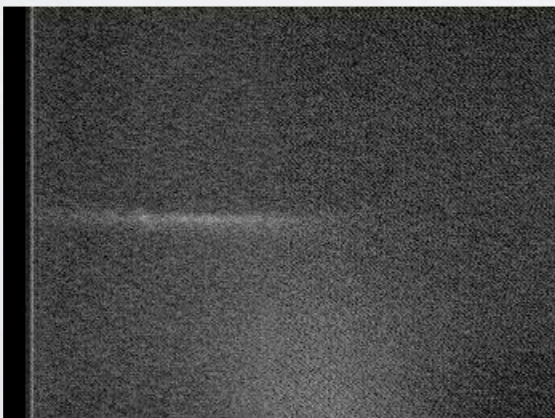
release time 15 ms



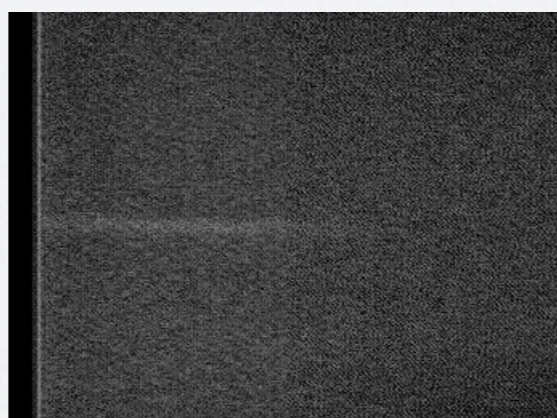
release time 20 ms



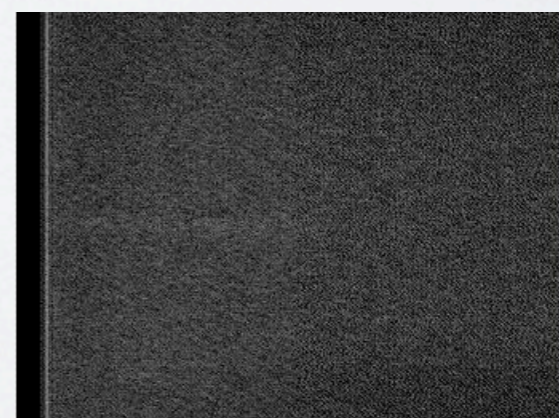
release time 25 ms



release time 30 ms



release time 40 ms



release time 50 ms

CONCLUSIONS

- JILA experiment (KRb) is very **successful**, but the laser system of two cw lasers, one comb laser, and one ionization laser is very **complicate**.
- Direct comb driving PA is **attractive and promising**. Although, the oxford experiment discouraged. How to optimize pulse shape and spectrum is still **unclear**. There is a long way to go.