# Rovibrational cooling of molecules by optical pumping

Experimental results for laser cooling of molecules

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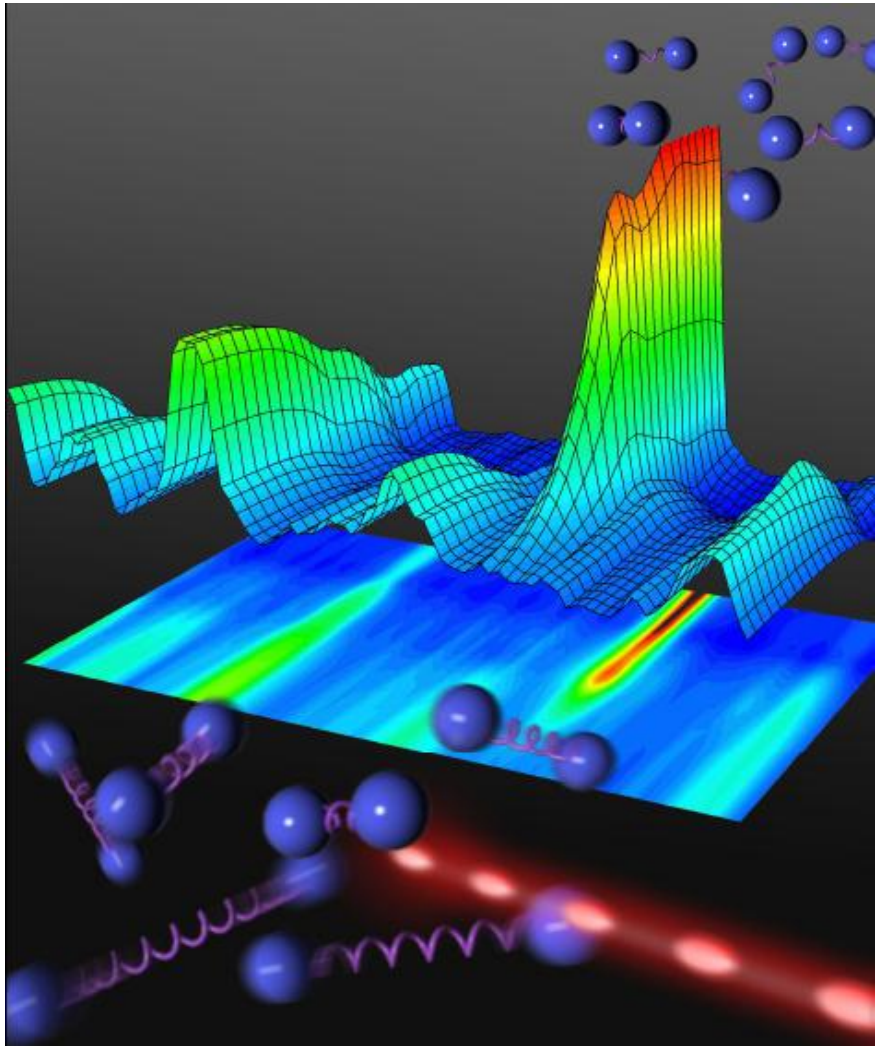
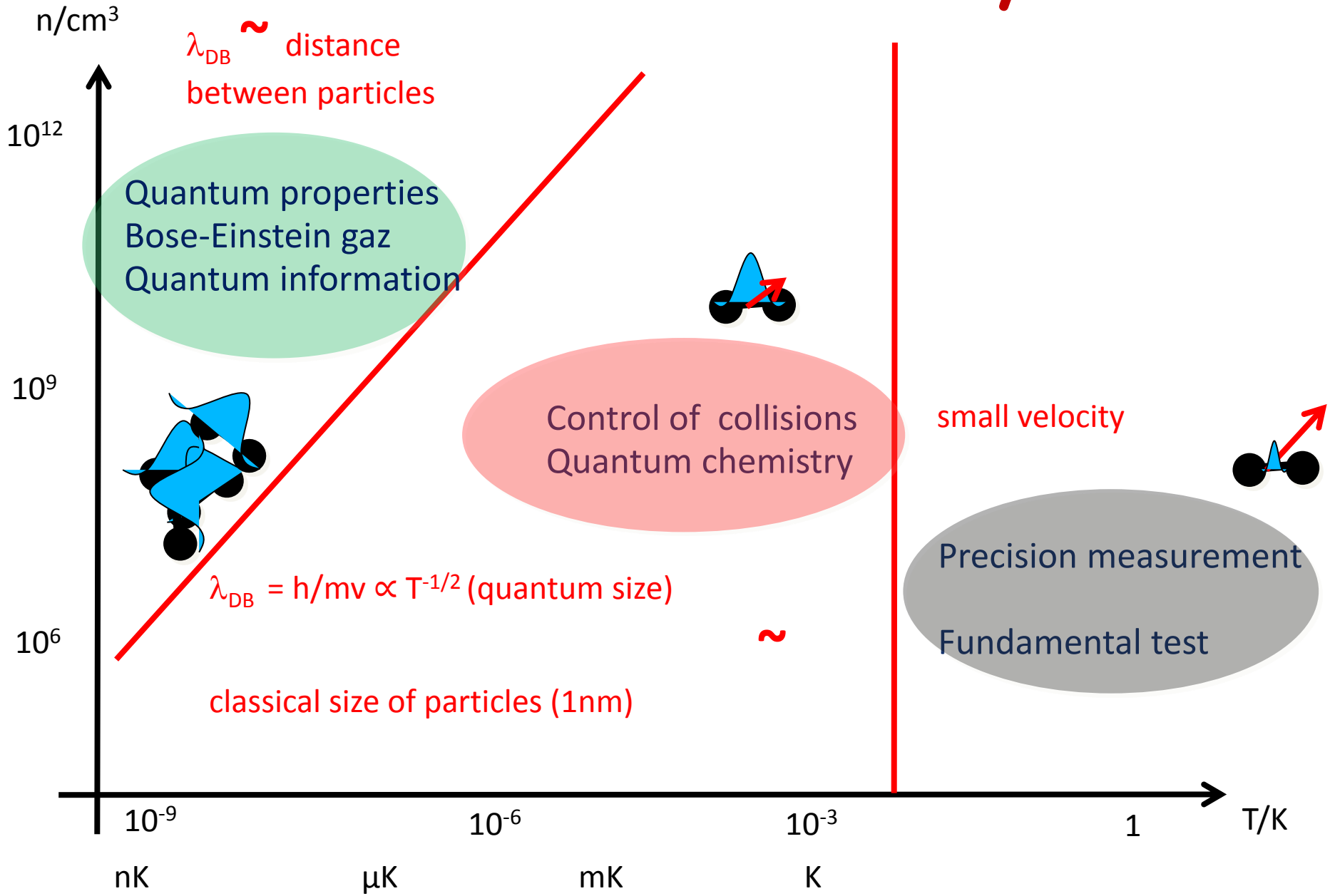


# Cold Molecules: Why?



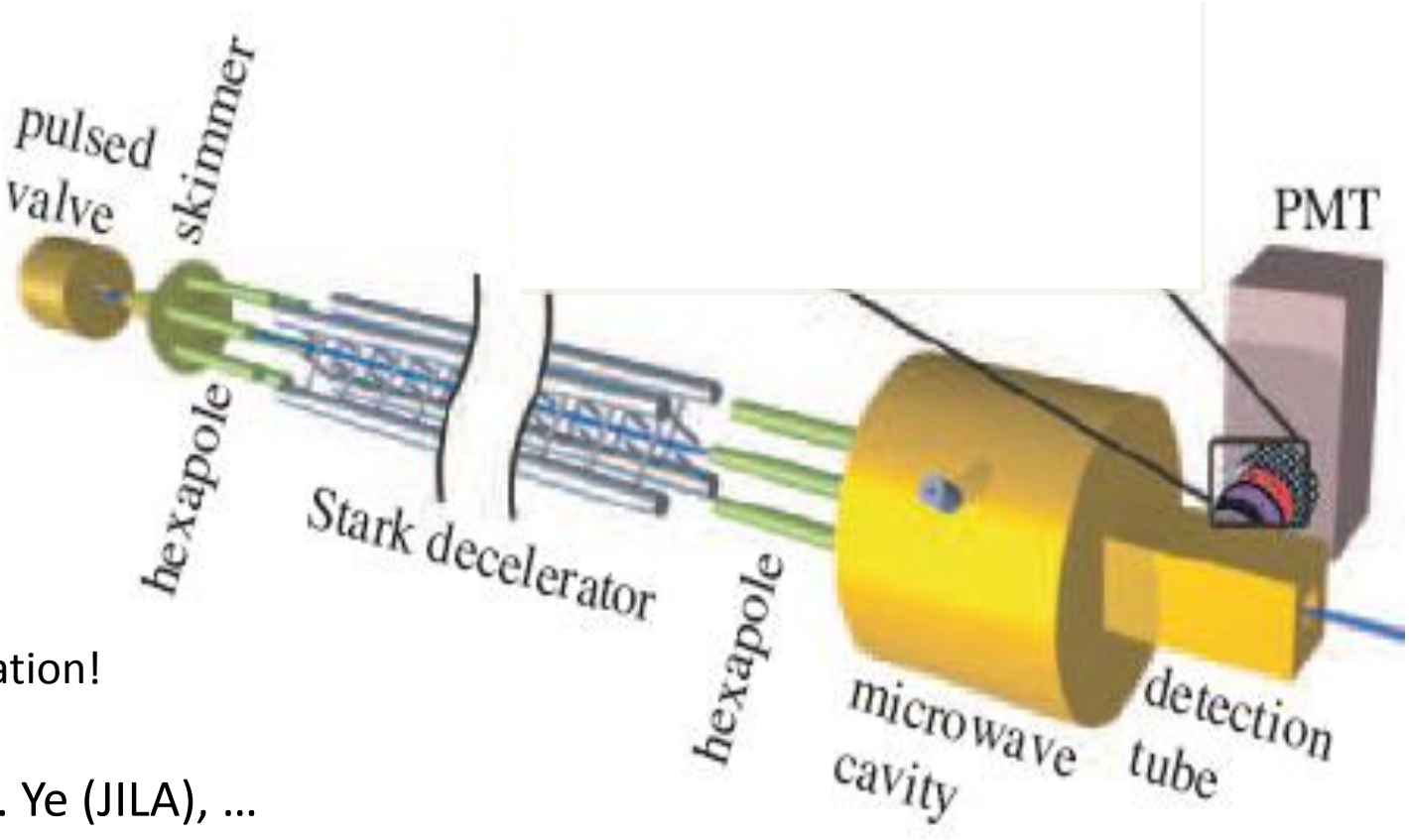
# Precise/long measurement

•Cold molecules → Slow molecules → long interaction time $\Delta t$

OH, CO, … radiative lifetime (50ms) PRL 95 013003 (2005)

•$\Delta E \sim h/\Delta t$ → precise measurement (atomic clock)

ND3 hyperfine structure (Hz) EPJD 31 337349 (2004)




Effect of black body radiation!

G. Meijer (Berlin) , J. Ye (JILA), …

# Fundamental test

Variation of constant ($10^3$ better than atoms): PRL 99, 150801 (2007)

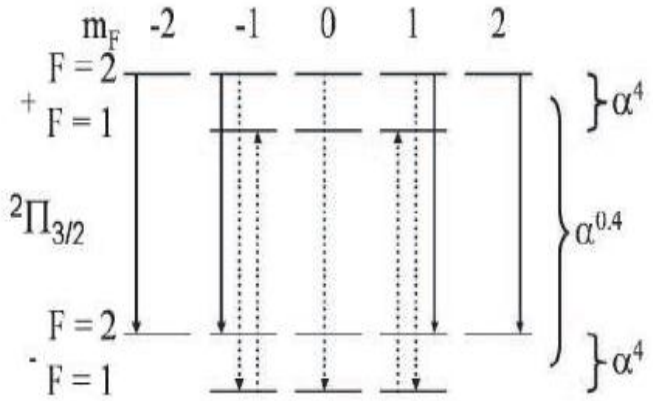
* OH electronic level PRL 96 143004 (2006).

Compare fine-structure with astrophysical data

$\Delta\alpha/\alpha < 10^{-16}$ /year

* Variation of $m_e/m_p$ or constant α in spectra.

Conincidence vibration and fine structure levels




* Electron dipole (d) moment YbF (Hinds), PbO (DeMille, Doyle),...

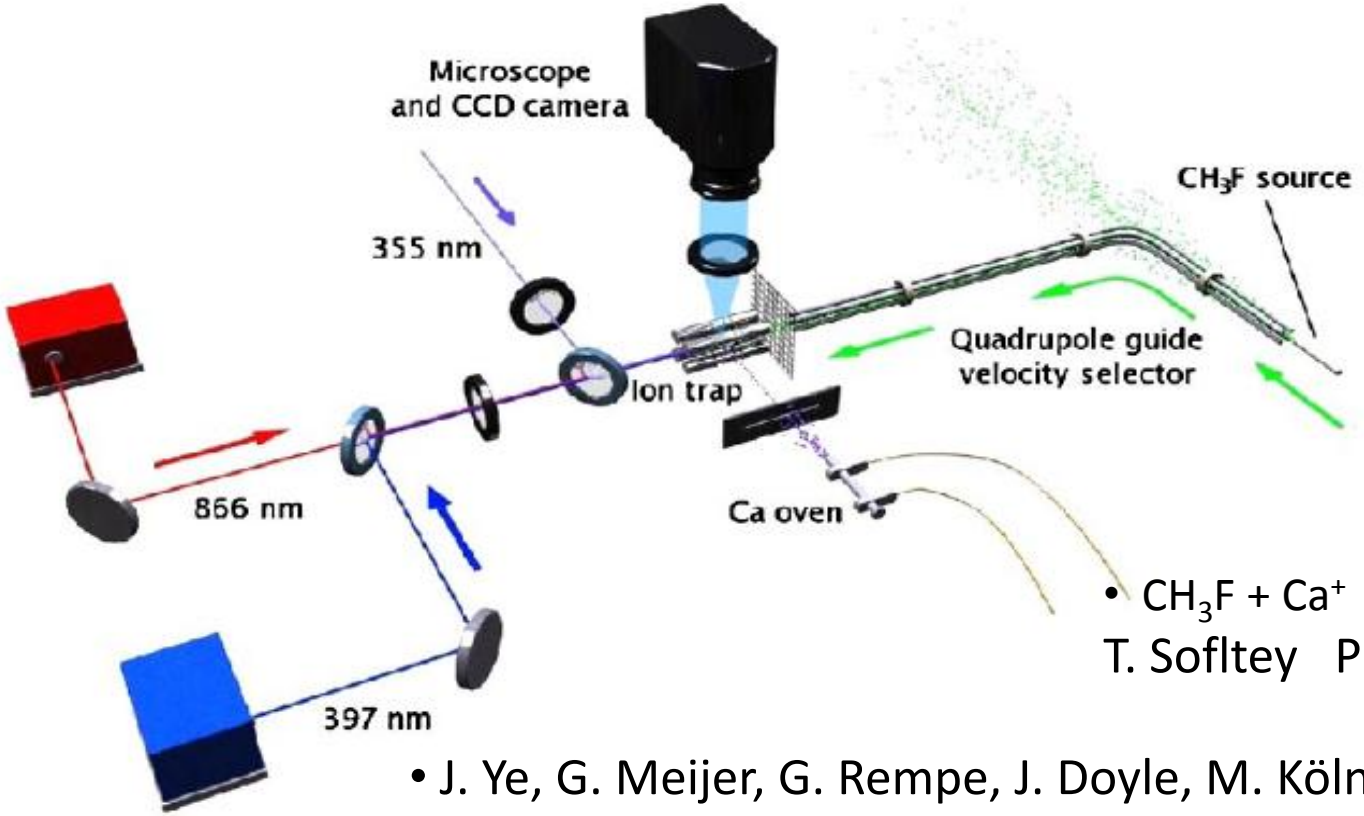
Shift –d.E due to electric E field inside the molecule GV/cm

**Improved measurement of the shape of the electron:**
Nature 473, 493 (26 May 2011)

•Chirality: BaF or HSiO D. DeMille *et al.* PRL 100, 023003 (2008)

# Control of (Reactive) collisions: quantum chemistry

- Reactions at zero temperature (Resonnance, Tunneling)
- Collisions in fields. External field to control dynamic
- Create cold atoms by photodissociation $SO_2$ → SO + O EPJD 46, 463 (2008)




- $CH_3F + Ca^+ \rightarrow CaF^+ + CH3$

T. Sofltey PRL 100, 043203 (2008)

- J. Ye, G. Meijer, G. Rempe, J. Doyle, M. Köln, H. Lewandowski …
- macromolécules (perfluorinated) M. Arndt
- Ions : M. Drewsen, S. Schiller, R.Wester, …

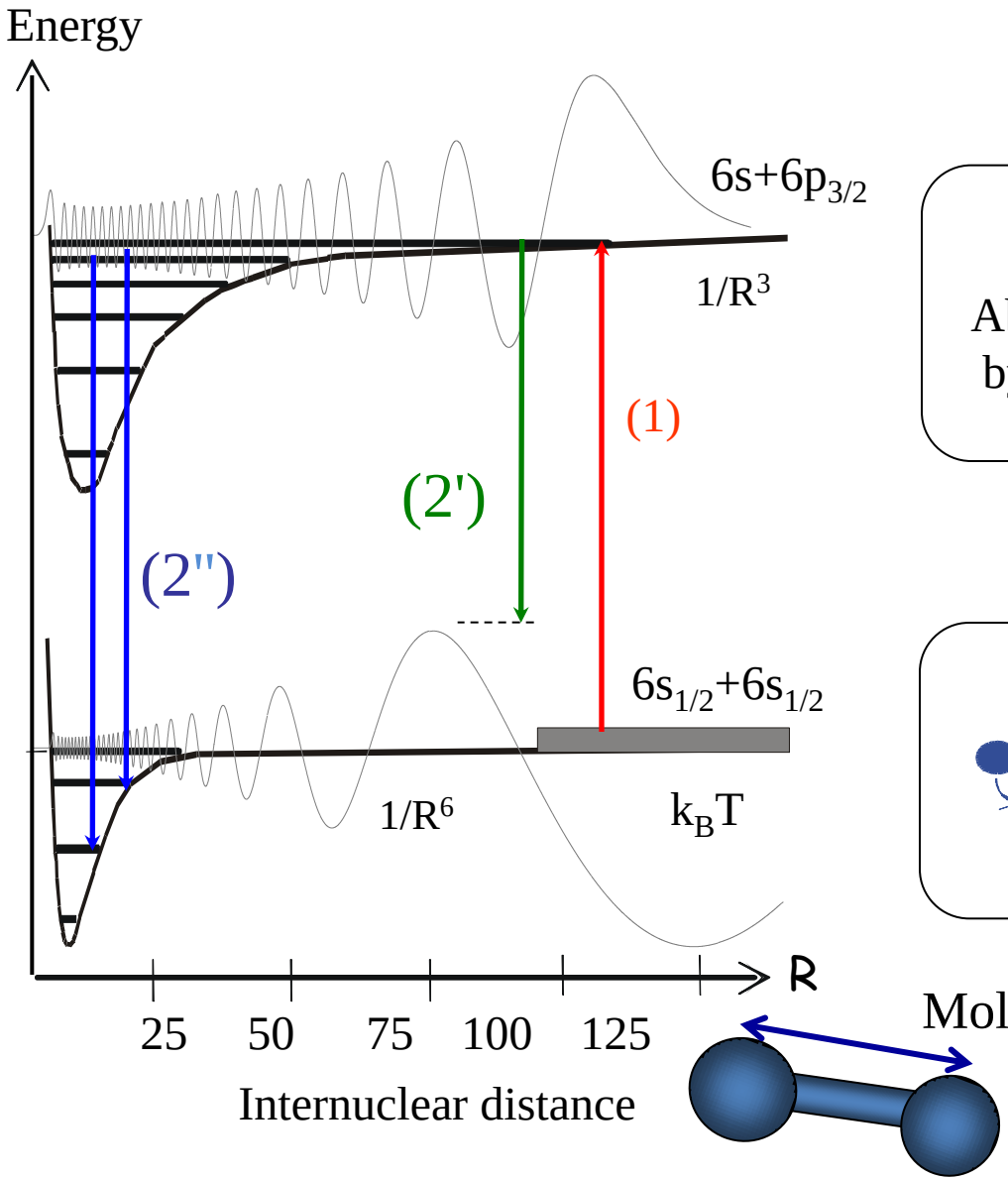
# Outline

1) Photoassociation and cold molecules

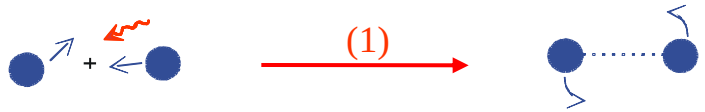
2) Rovibrational Cooling of Cesium molecules by Optical pumping

3) Conclusion

# Photoassociation

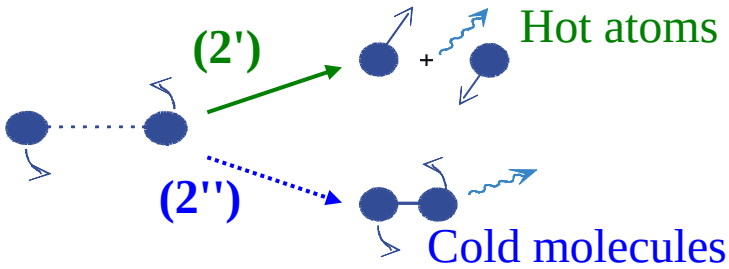



## Photoassociation




Absorption of one photon
by two cold atoms (T~100mK)

## Desexcitation

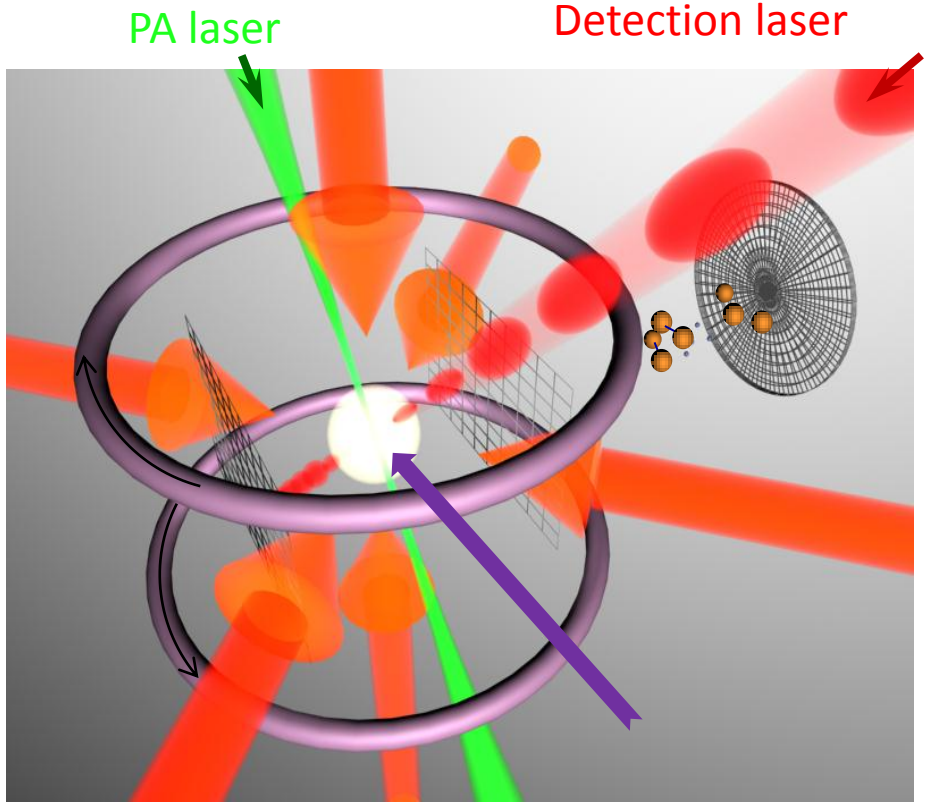



Molecules in several vibrational levels

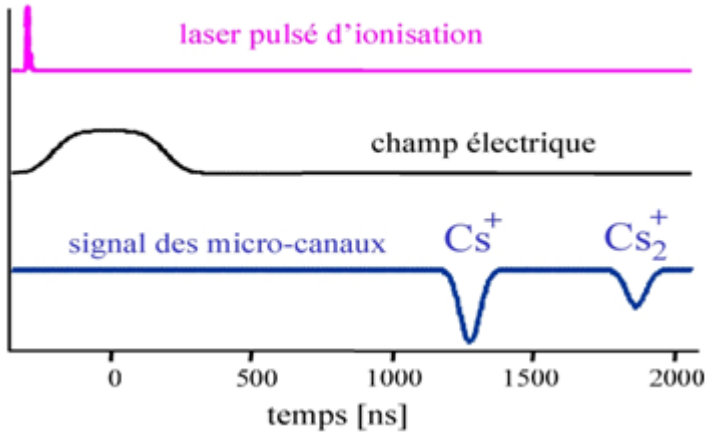
# Experimental Setup

Cs - MOT

~ 5 $\cdot 10^7$ atoms ~$10^{11}$at/cm$^3$ ~100µK

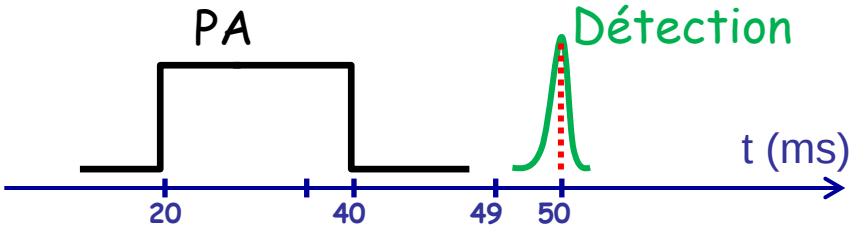


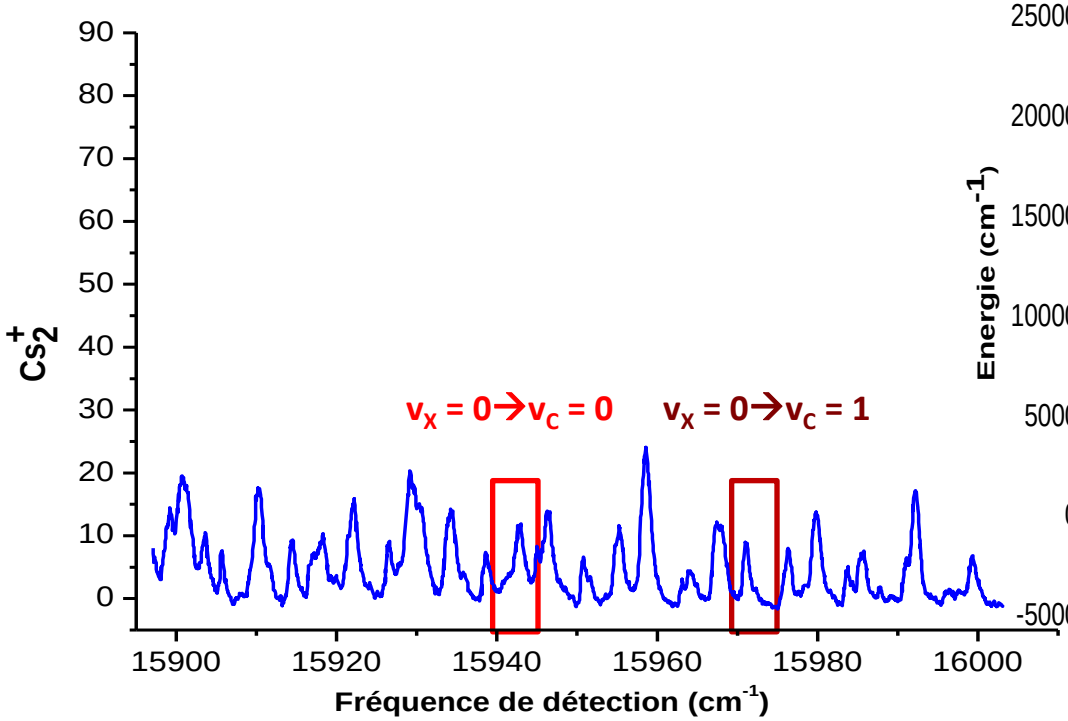

- PA: Ti:Sa continu, 852 nm, ~1W
- Detection (ionization): pulsed laser
  10 Hz-7ns, 5-10mJ/pulse

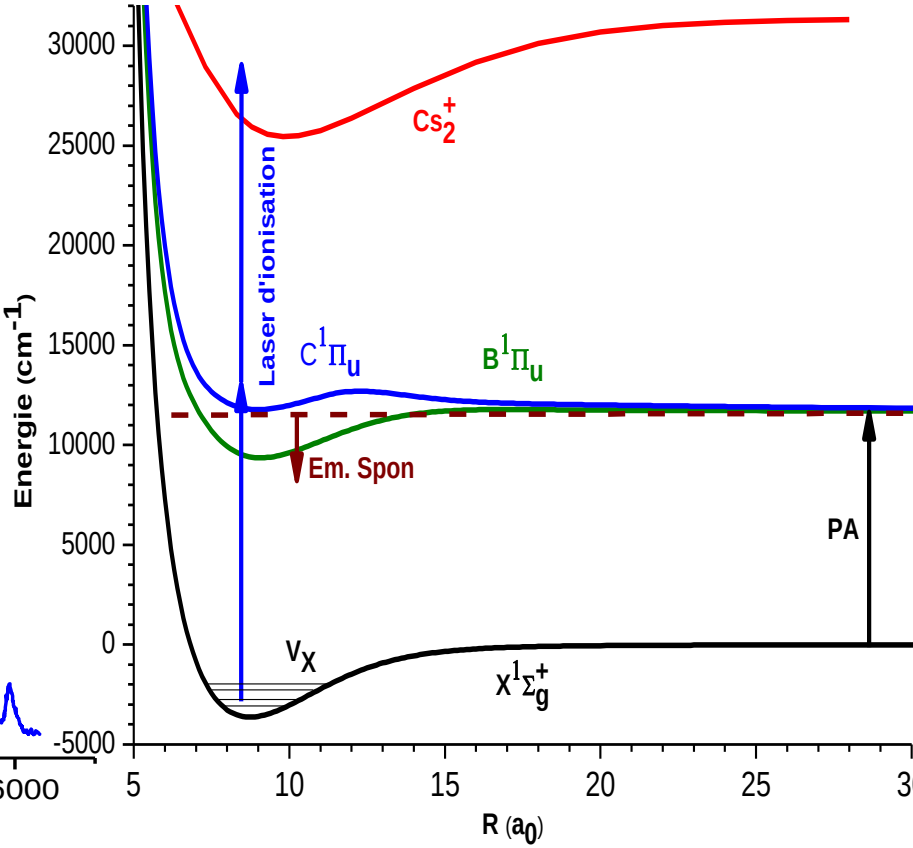



- Optical pumping: femtoseconde laser,
  12.5 ns, 770 nm, 1W

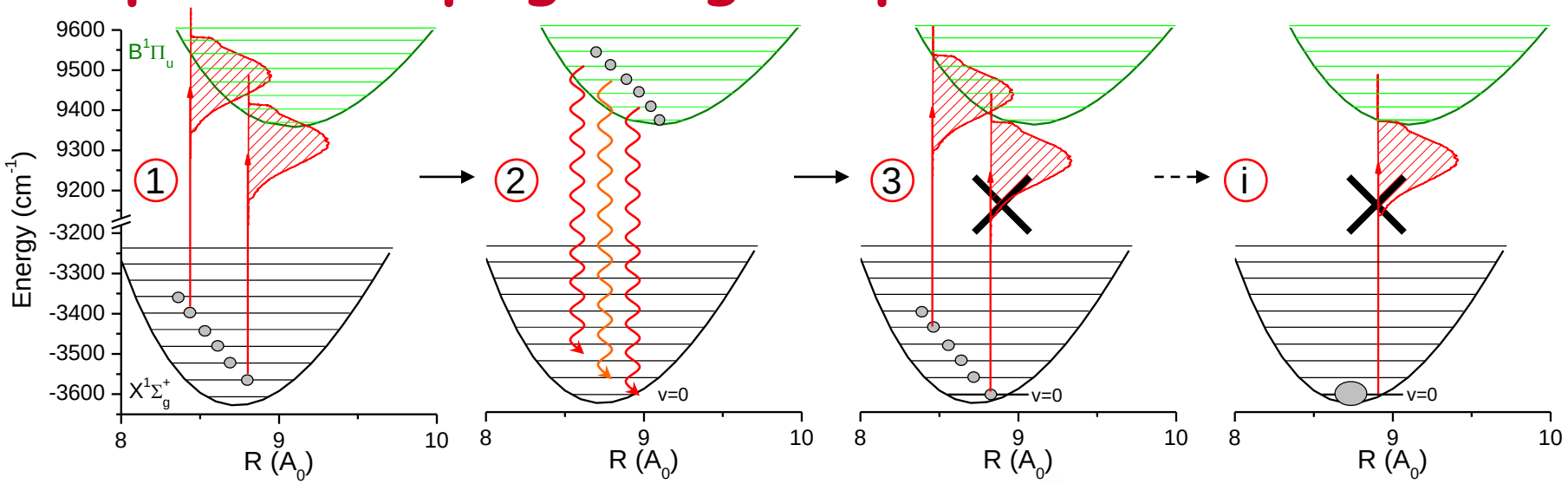
# Population after Photoassociation

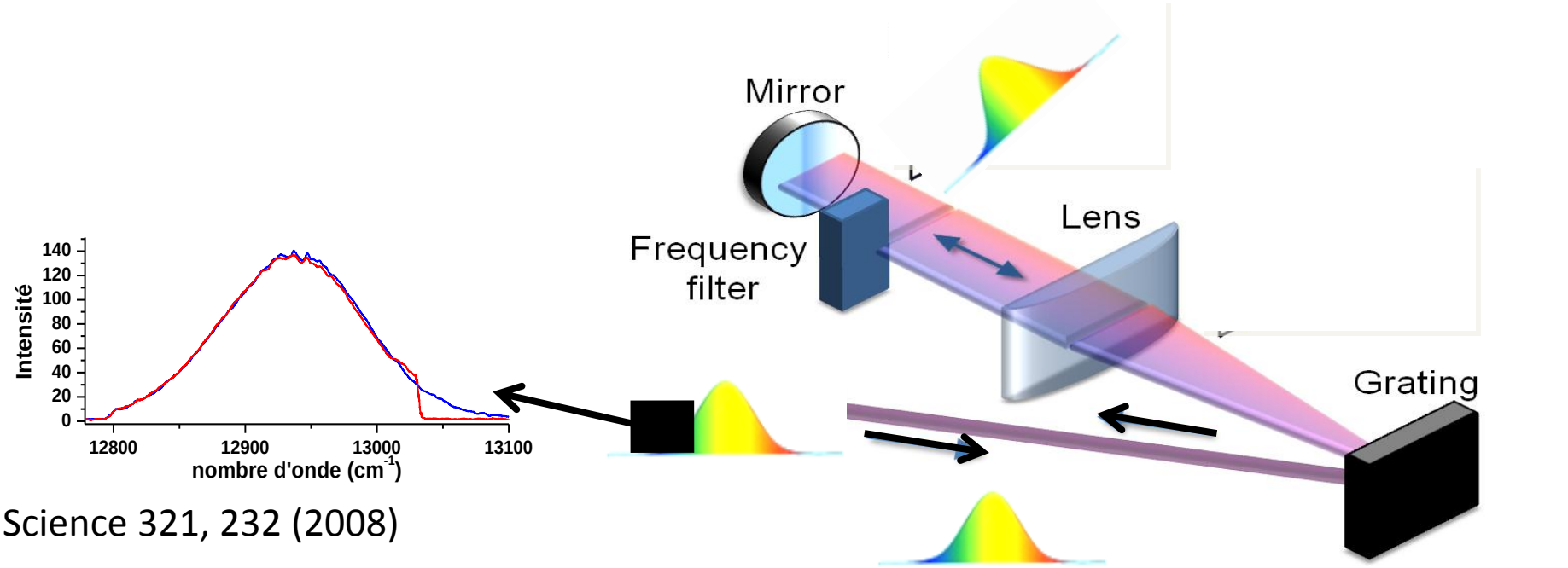


# Need: Vibrational cooling
# Optical Pumping using shaped broadband laser



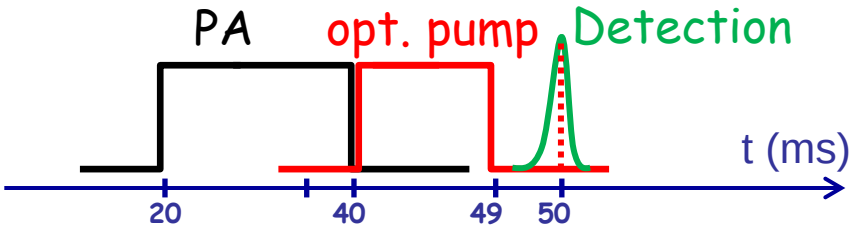



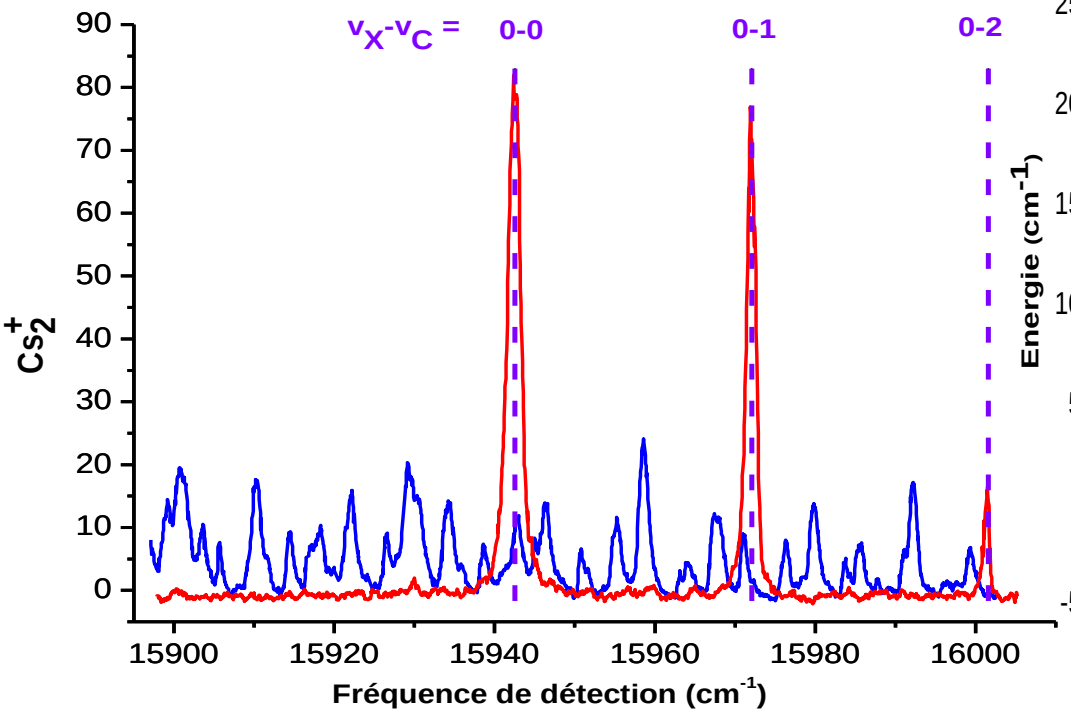

Science 321, 232 (2008)

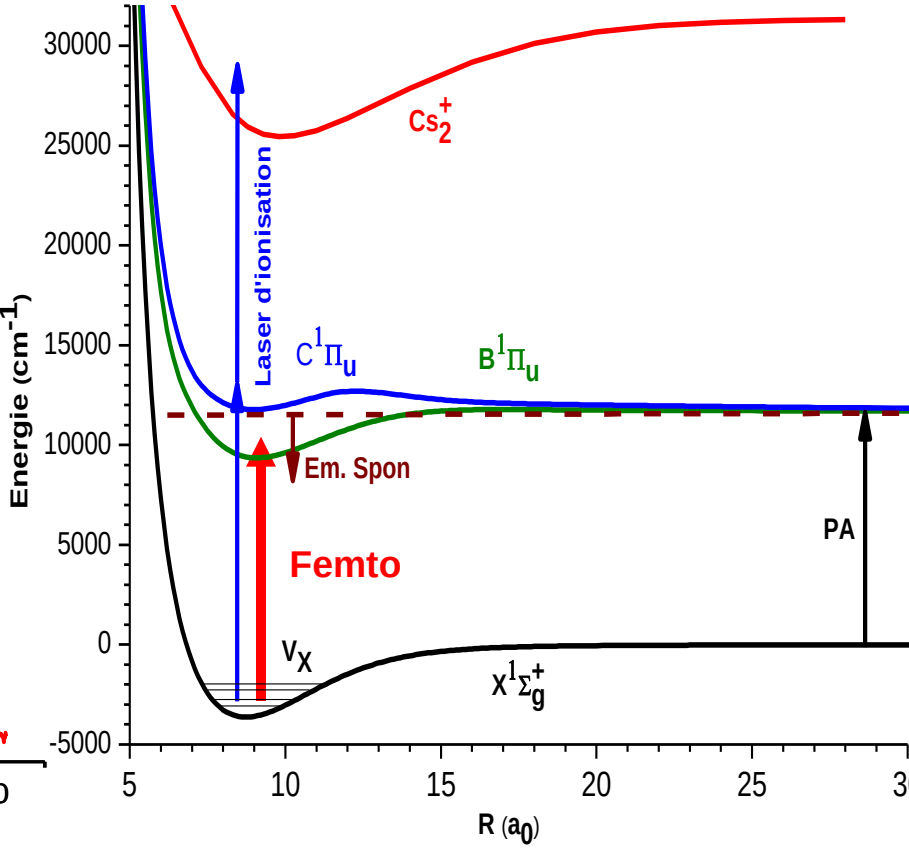
# Population after pumping

- Optical pumping

→Efficiency ~ 65 %

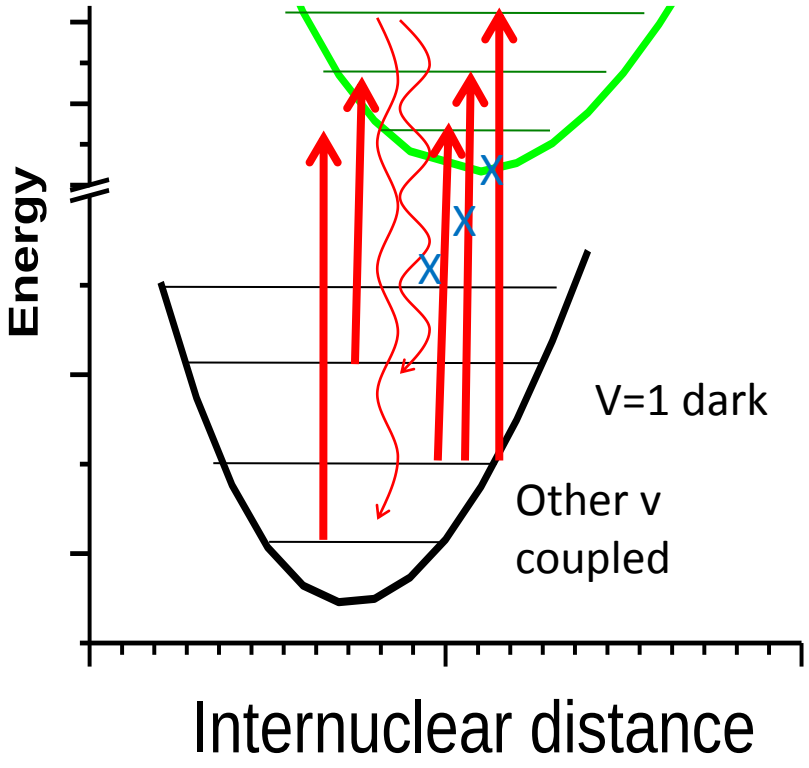


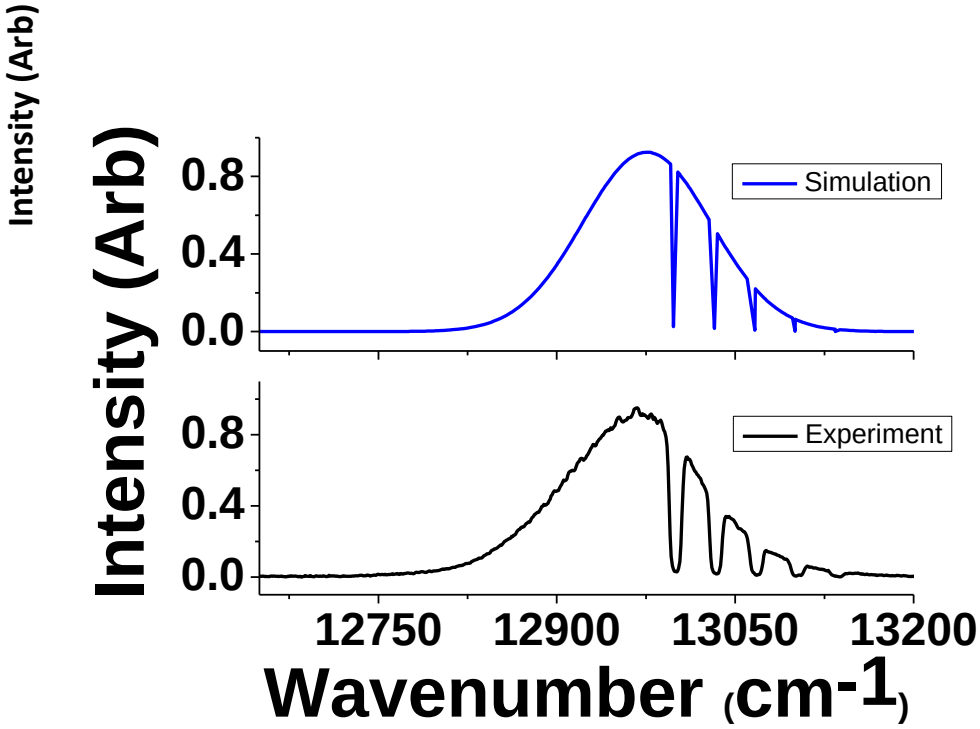



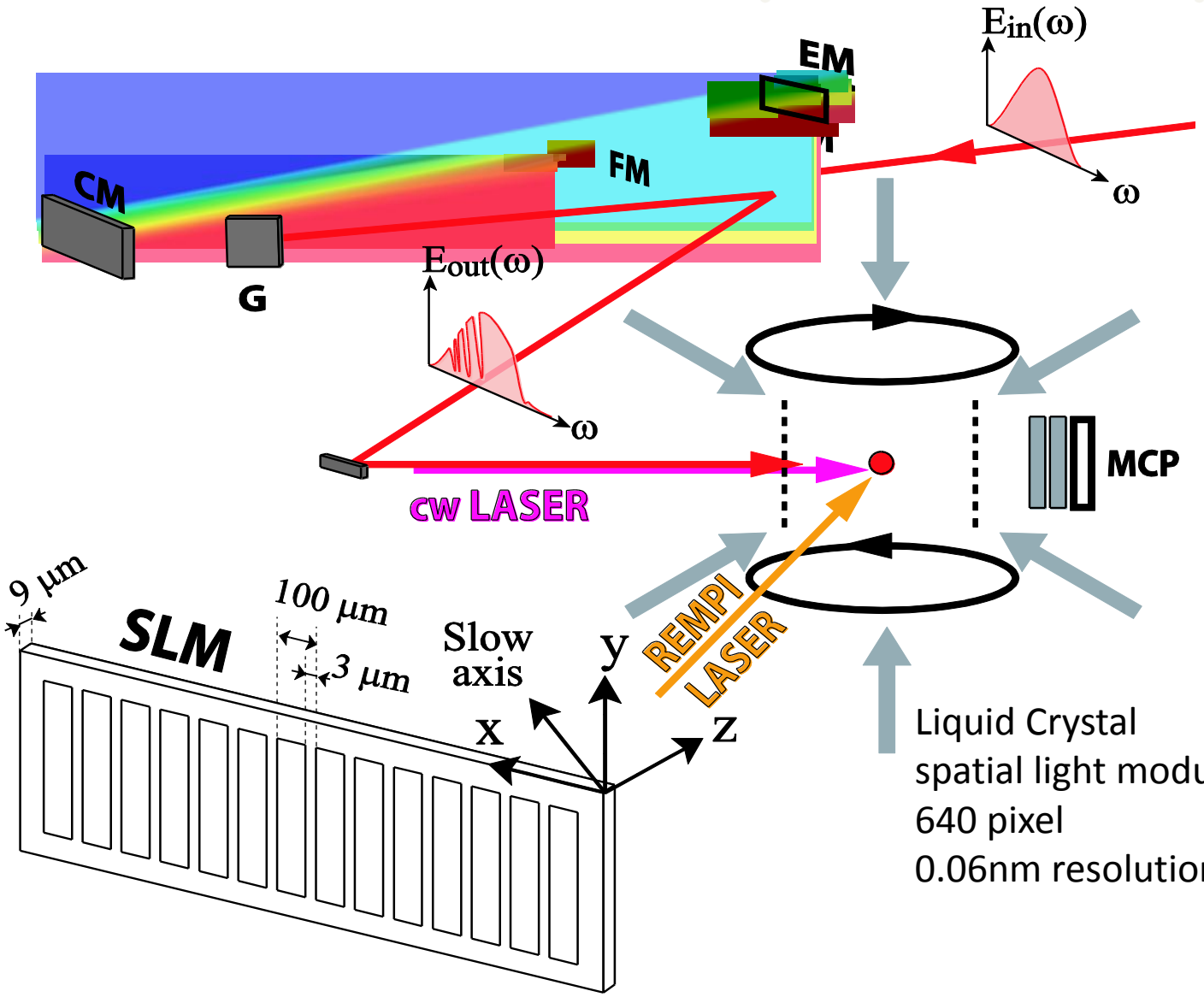
# Pumping to the dark v=1

NJP **11** 055037 (2009)



# Better amplitude shaping

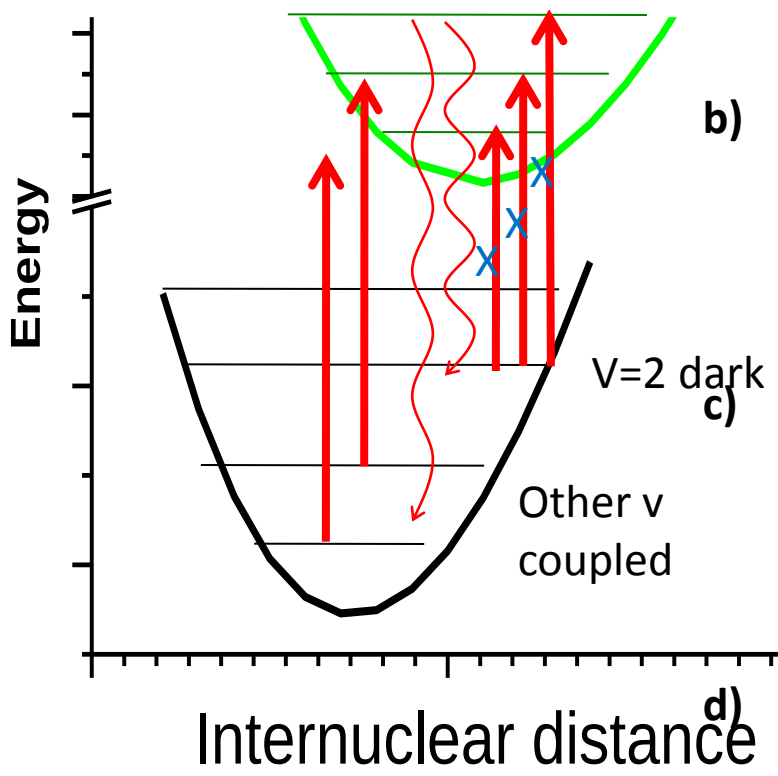


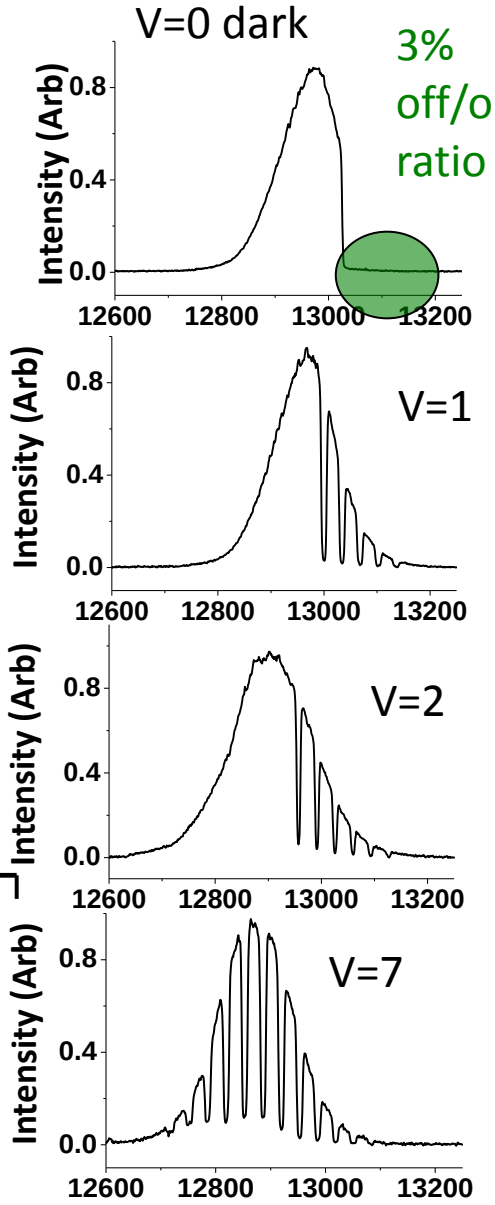

Liquid Crystal
spatial light modulator (SLM)
640 pixel
0.06nm resolution ~ 1 $cm^{-1}$

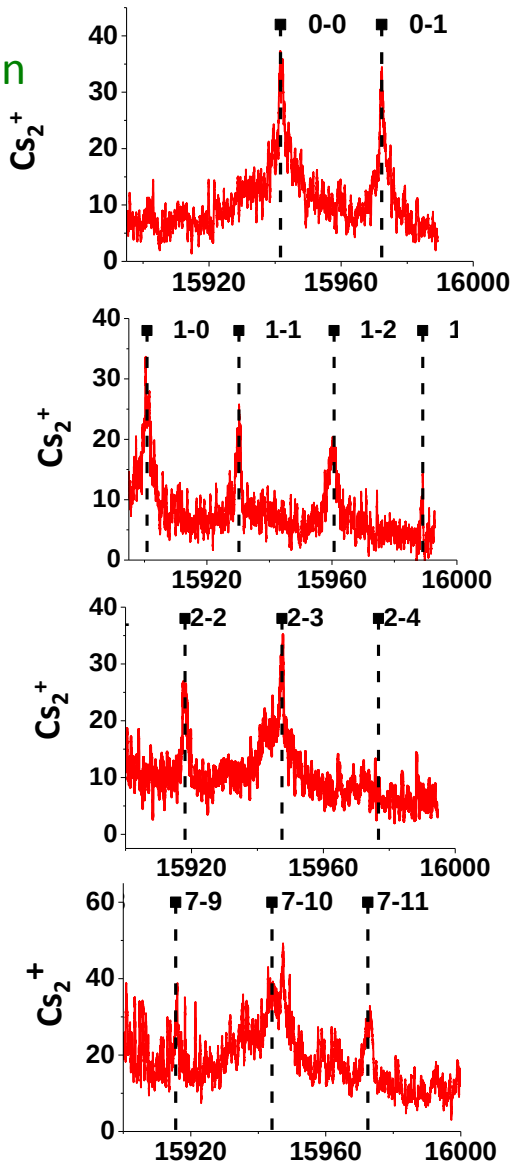
Collaboration with Béatrice Chatel, Laboratoire Collision Agrégat Réactivité, Toulouse

# Pumping to a chosen dark state !

NJP **11** 055037 (2009)



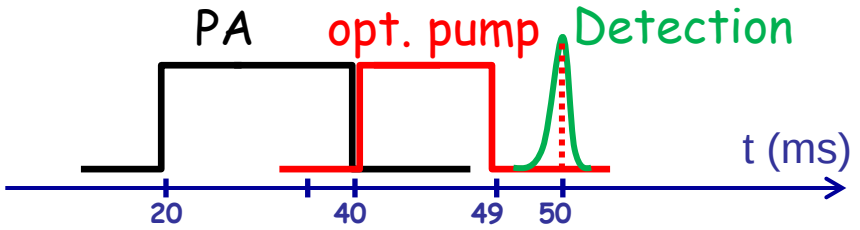




Efficiency ~ 60% with SLM

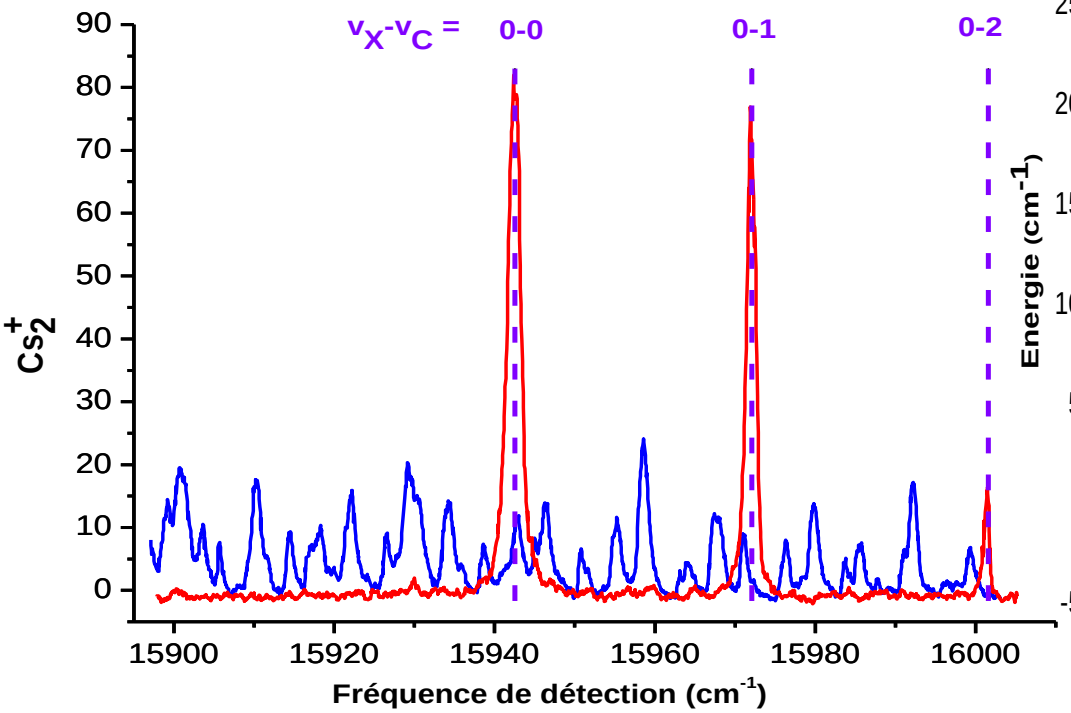
# Population after pumping

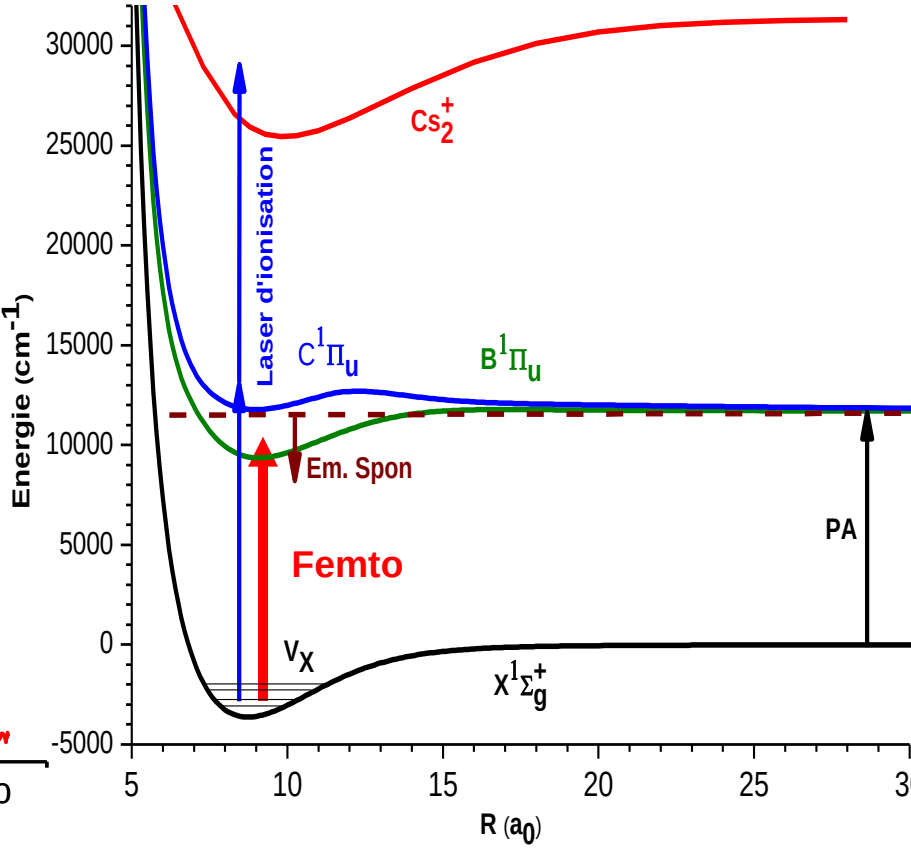
- Optical pumping

→Efficiency ~ 65 %

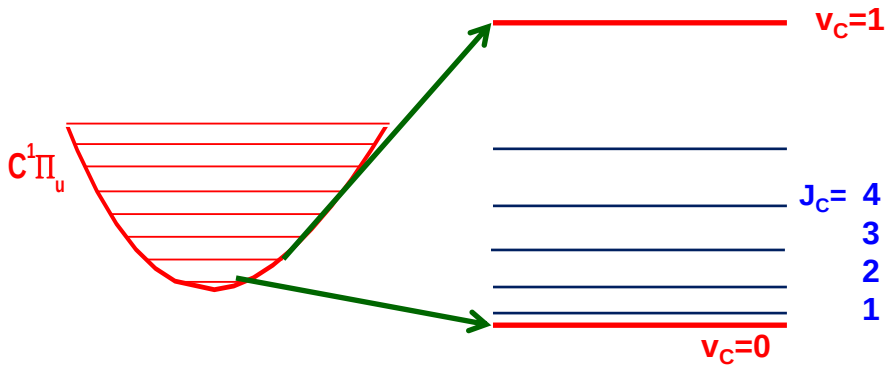



**In one vibrational level many rotational levels are populated**



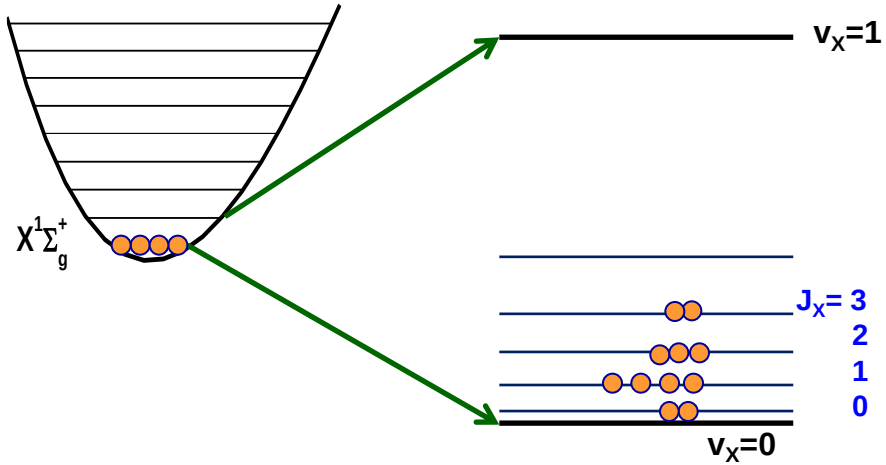
# Detection of the rotation




➢ Detection:

- For one vibrational level many rotational levels are populated
- Rotational separation (~600MHz)

→Unresolved with the REMPI detection (3 GHz)



# Detection of the rotation

➢ Detection:

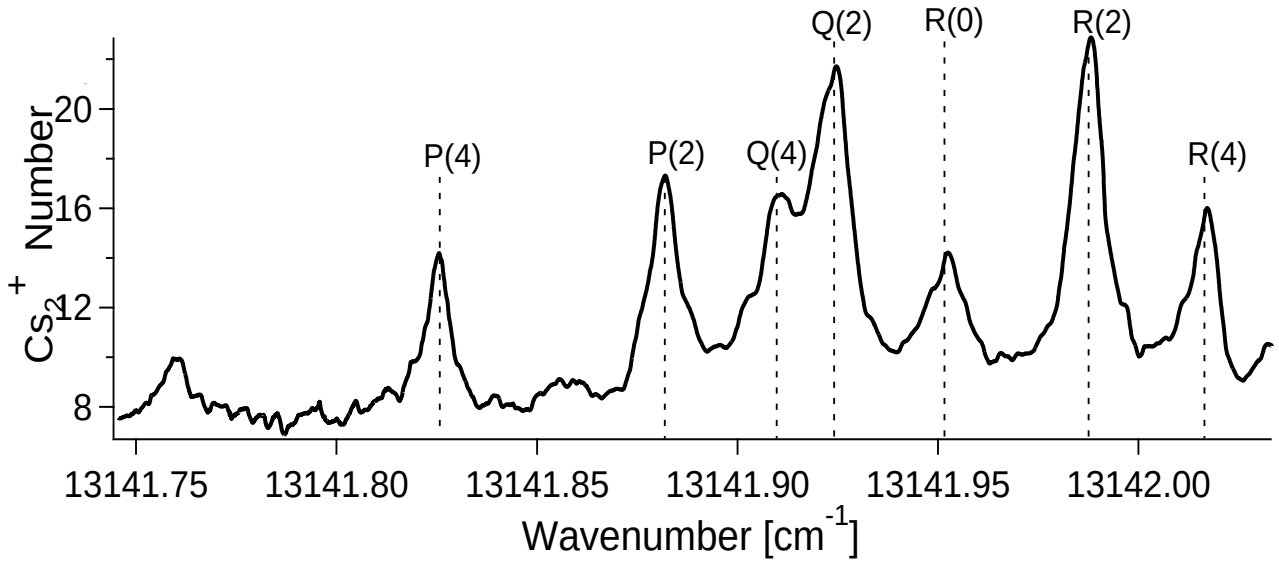
- For one vibrational level many rotational levels are populated
- Rotational separation (~600MHz)

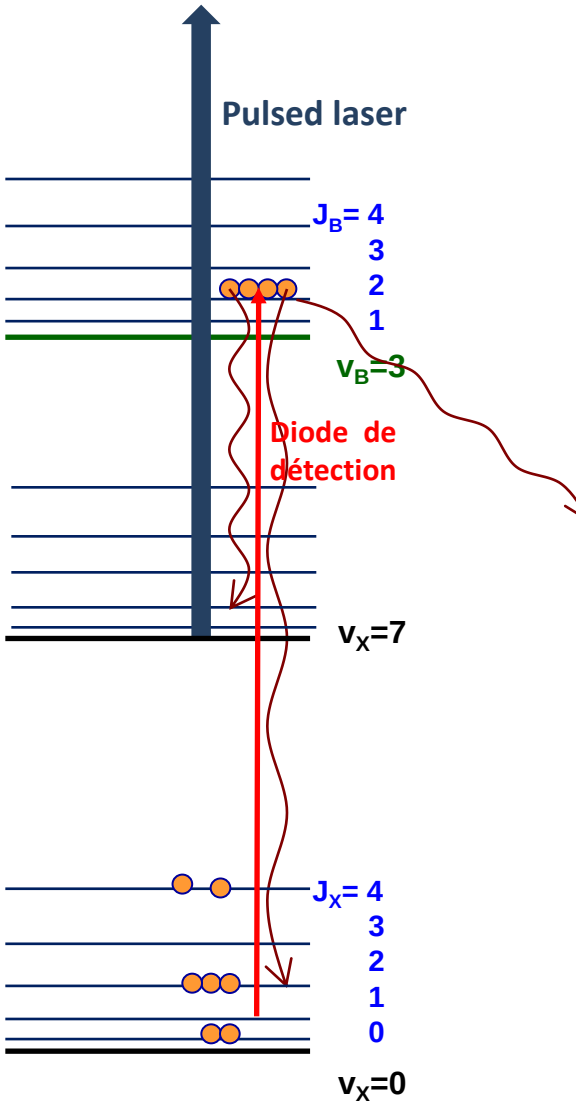
→Unresolved with the REMPI detection (3 GHz)

→Detection with a narrowband laser

→Desexcitation to many vibrational levels

→REMPI detection in $v_X = 7$



Selection rules : $\Delta J = 0, \pm 1$ $+ \leftrightarrow -$

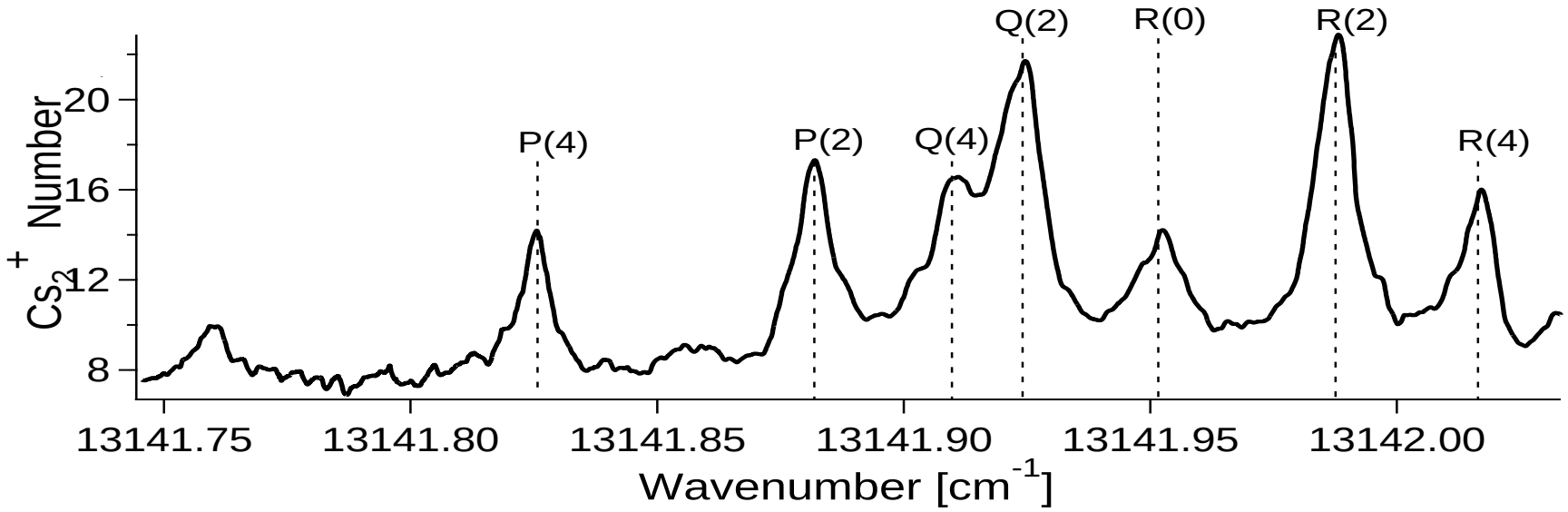
- P : J decrease by 1 in absorption
- Q : J constant
- R : J increase by 1 in absorption

**Too cool the rotation we use excite the P branch**

2) Rotational cooling a modifies the vibration
→ vib. cooling needed

In practice:

$Cs_2$ rotational structure
too small to be shaped with grating




Only Even rotational distribution are populated
The rotational states have a (+/-) parity given by the sign of $(-1)^{J'+1}$ in $0_g^-$ and $(-1)^J$ in $^1X\sum_g$

# Rotational cooling

Selection rules : $\Delta J = 0, \pm 1$ + ←→ -

- P : J decrease by 1 in absorption
- Q : J constant
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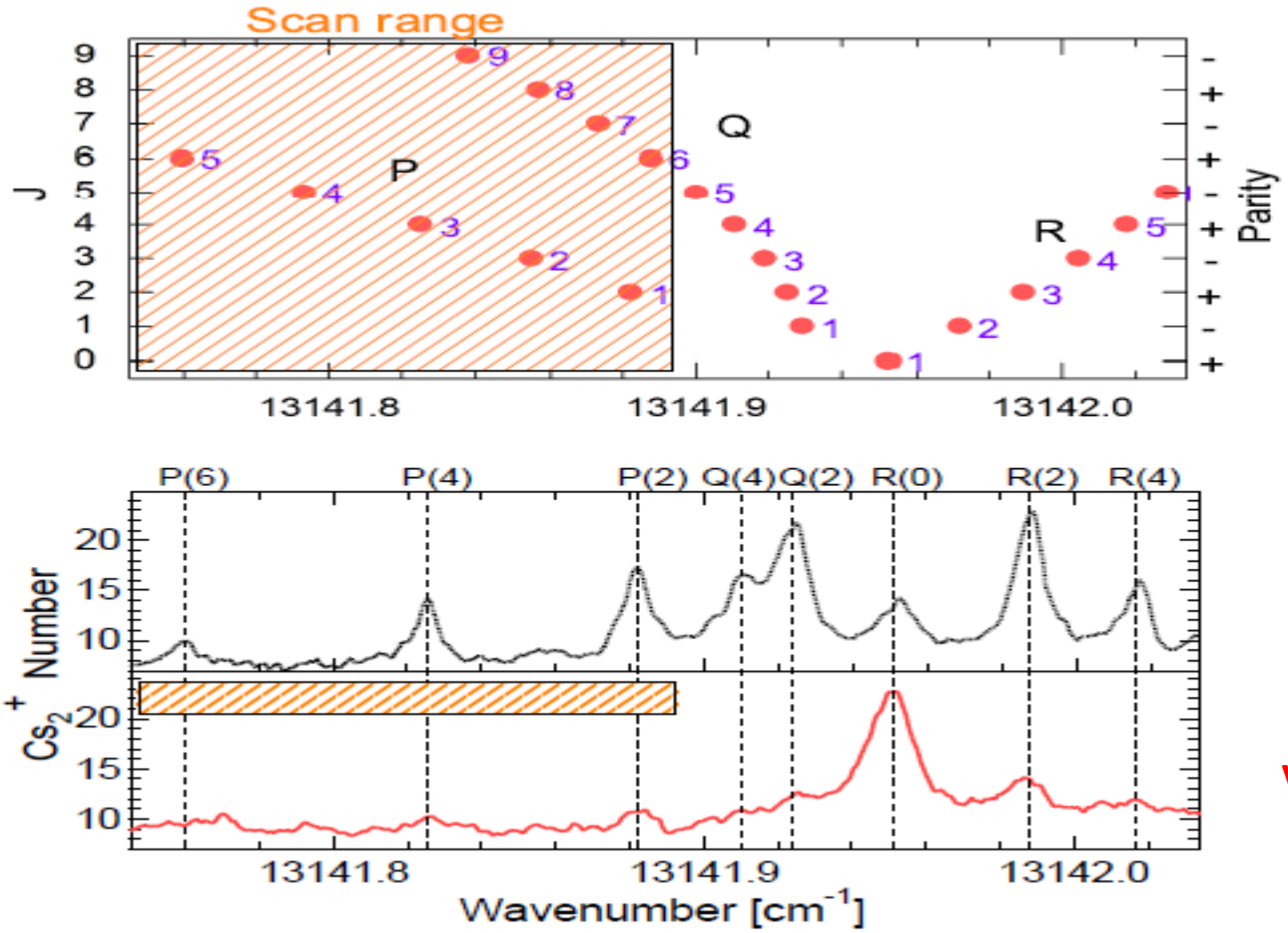
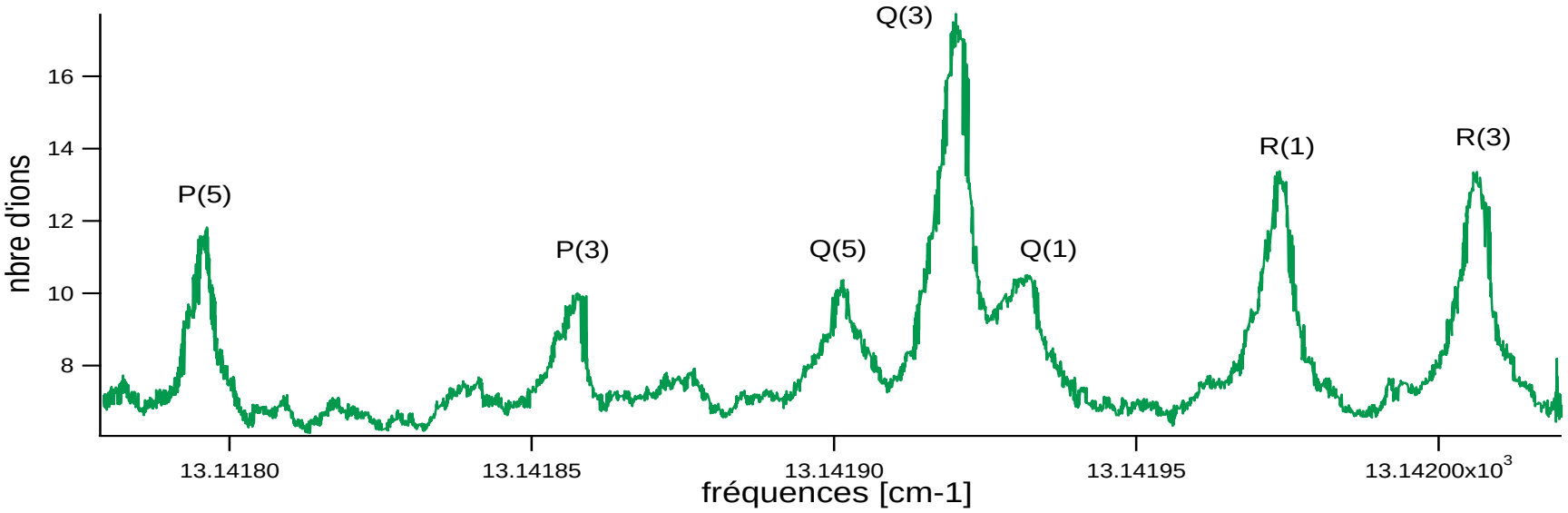


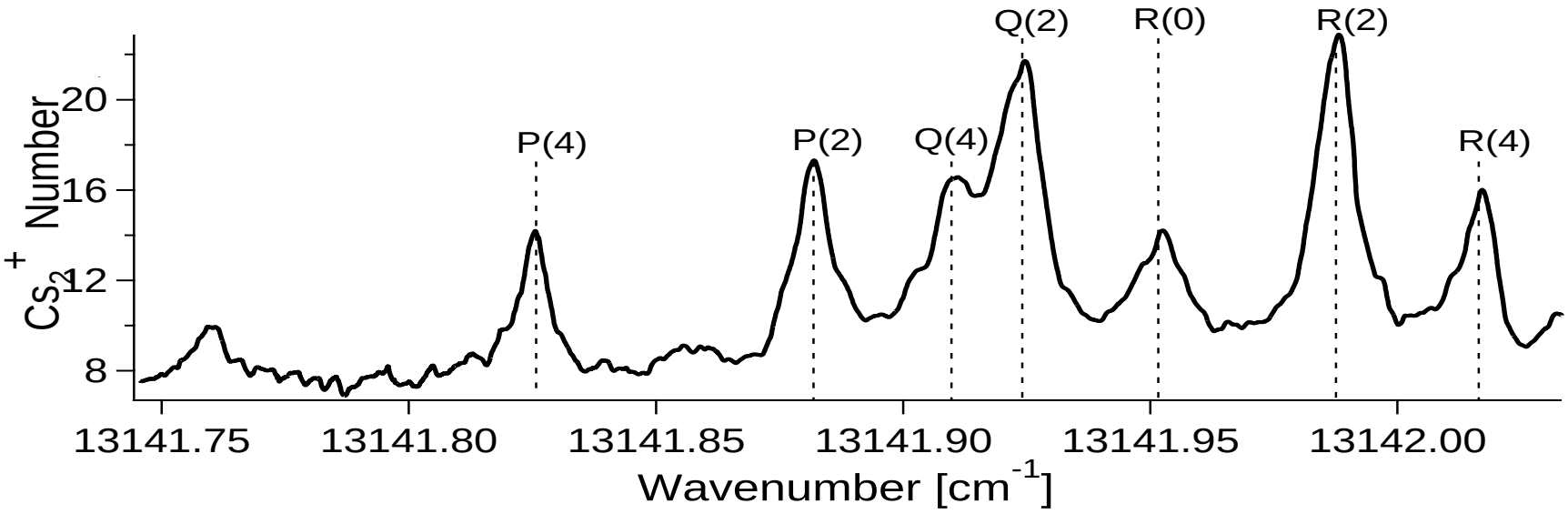

**V= 0 , J = 0**

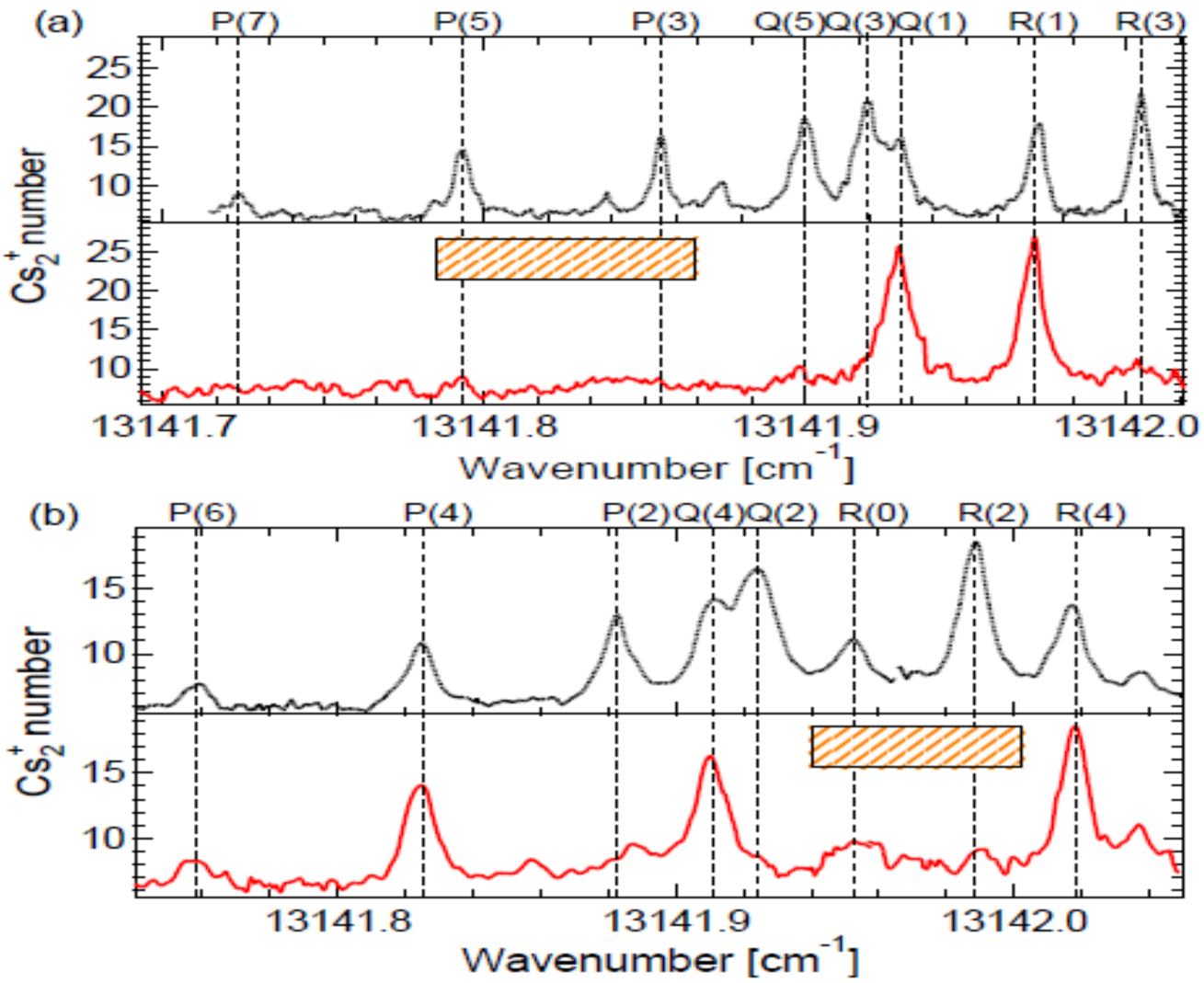
The green spectrum : only odd rotational levels are populated

The black one : only even rotational levels are populated

The rotational states have a (+/-) parity given by the sign of $(-1)^{J'+1}$ in $0_g^-$ and $(-1)^J$ in $^1X\sum_g$




(a)
P(7)
P(5)
P(3)
Q(5)
Q(3)
Q(1)
R(1)
R(3)
$Cs_2^+$ number
25
20
15
10
13141.7
13141.8
13141.9
13142.0
Wavenumber [$cm^{-1}$]
(b)
P(6)
P(4)
P(2)
Q(4)
Q(2)
R(0)
R(2)
R(4)
$Cs_2^+$ number
15
10
13141.8
13141.9
13142.0
Wavenumber [$cm^{-1}$]

V= 0 , J = 1

V= 0 , J = 4

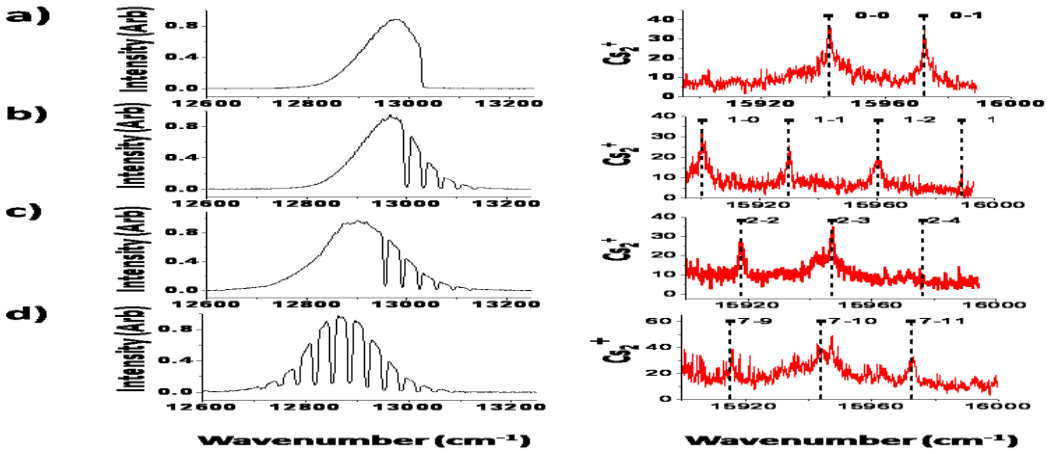
# Conclusion

- Accumulation of molecules in a choosen vibrational level using a shaped broadband femtoseconde laser

New Journal of Physics, 11(5)(2009)

Journal of Modern Optics, 56:2089-2099,(2009).

Molecular Physics, 108 :795{810, (2010)



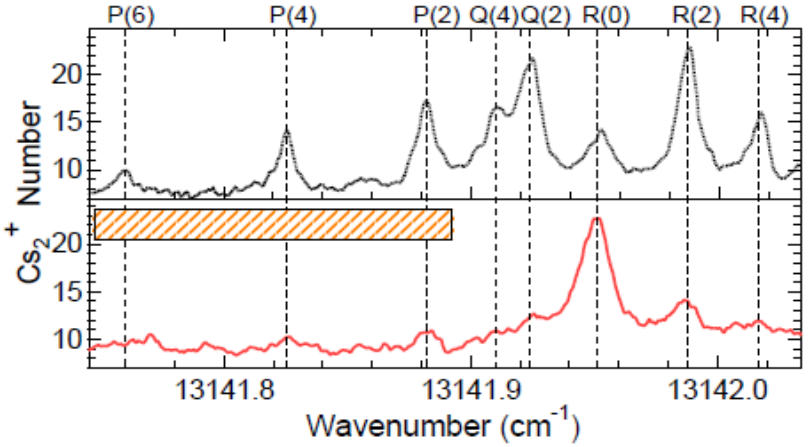

Our vibrational cooling method is genaral method and can be used in any molecules, demonstrated recently in Bigelow group for NaCs molecules

Optics Express, Vol. 20, No. 14, (2012)

# Conclusion

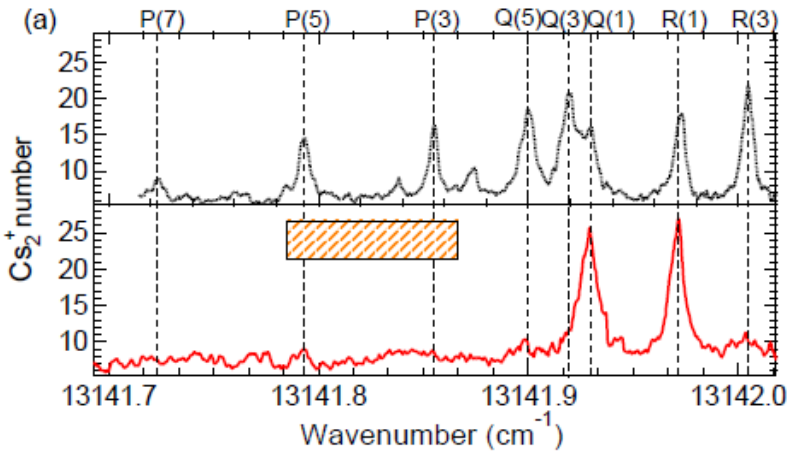
- Rovibrational cooling of $Cs_2$ molecules are also demonstrate

**Accepted in PRL**

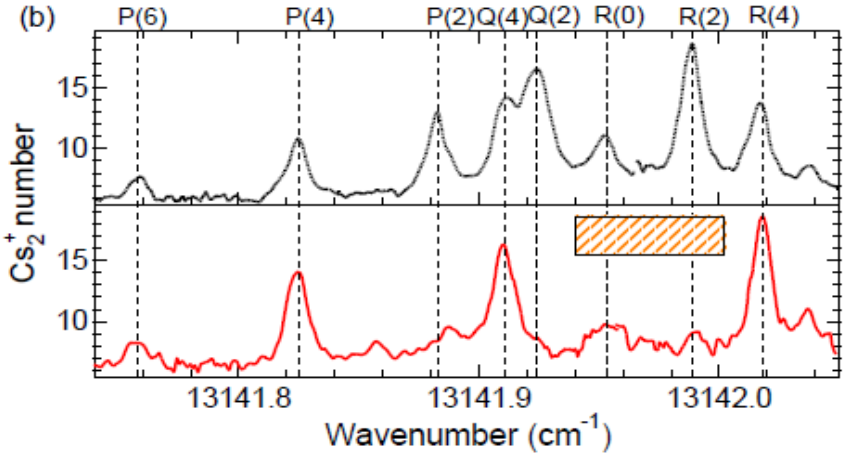



**V= 0 , J = 0**

Transfert of molecules in a choosen rotational level




**V= 0 , J = 1**



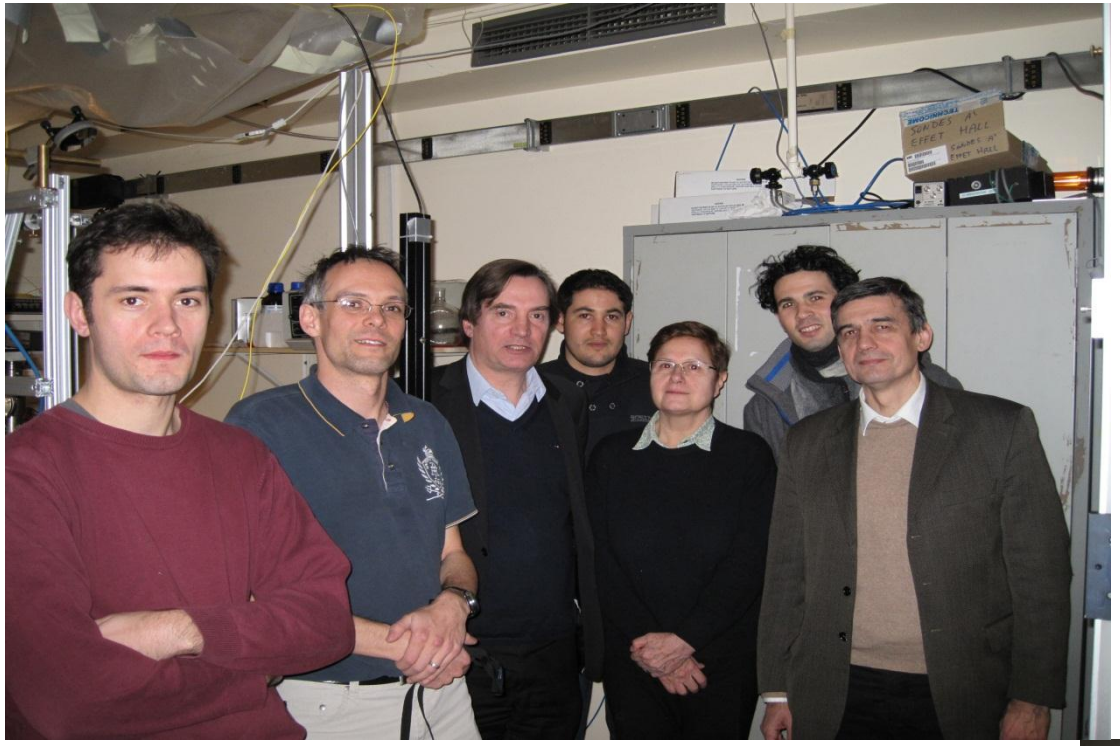

**V= 0 , J = 4**

# Perspectives

* With rovibrational cooling collision between molecules and atoms can be studied.

* The method of rovibrational cooling could be extended to other molecules and molecular beams.

* And also opens up general perspectives in laser cooling the external degrees of freedom of molecules.



## Visitors

- Marin Pichler
- Maria Allegrini
- Goran Pichler
- Emiliya Dimova
- Lirong Wang

## Theory LAC

- Nadia Bouloufa
- Olivier Dulieu

## Experiment LAC

- Isam Manai
- Ridha Horchani
- Mehdi Hamamda
- Daniel Comparat
- Hans Lignier
- Pierre Pillet

**Thank you for your attention !**

## Collaboration

- Béatrice Chatel
- Sébastien Weber

(LCAM, Toulouse, France)