



**INO - CNR**  
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56124 Pisa

## Raffreddamento laser di molecole

*Andrea Fioretti*

.....

**18 gennaio 2013**  
*Area della Ricerca di Pisa*





## SHORT STORY of AF

- Collaboration with C. Gabbanini since 1998 (IFAM, IPCF and INO)
- Last 3-years in Laboratoire Aimé Cotton, Orsay (Fr)
- Recent research activity at LAC on:
  - Monochromatic ion and electron beams from a cold atom source (collaboration with CNRS-Orsay, the private company Orsay Physics in Fuveau (FR) and the University of Pisa under a FP7-IAPP «Coldbeams»
  - Cold Rydberg atoms and cold plasma
  - Cold molecules



# OUTLINE

- Introduction. Why cold molecules?
- Introduction. Methods of production of cold molecules
- Optical pumping and vibrational cooling
- Rotational cooling
- Conclusions and perspectives

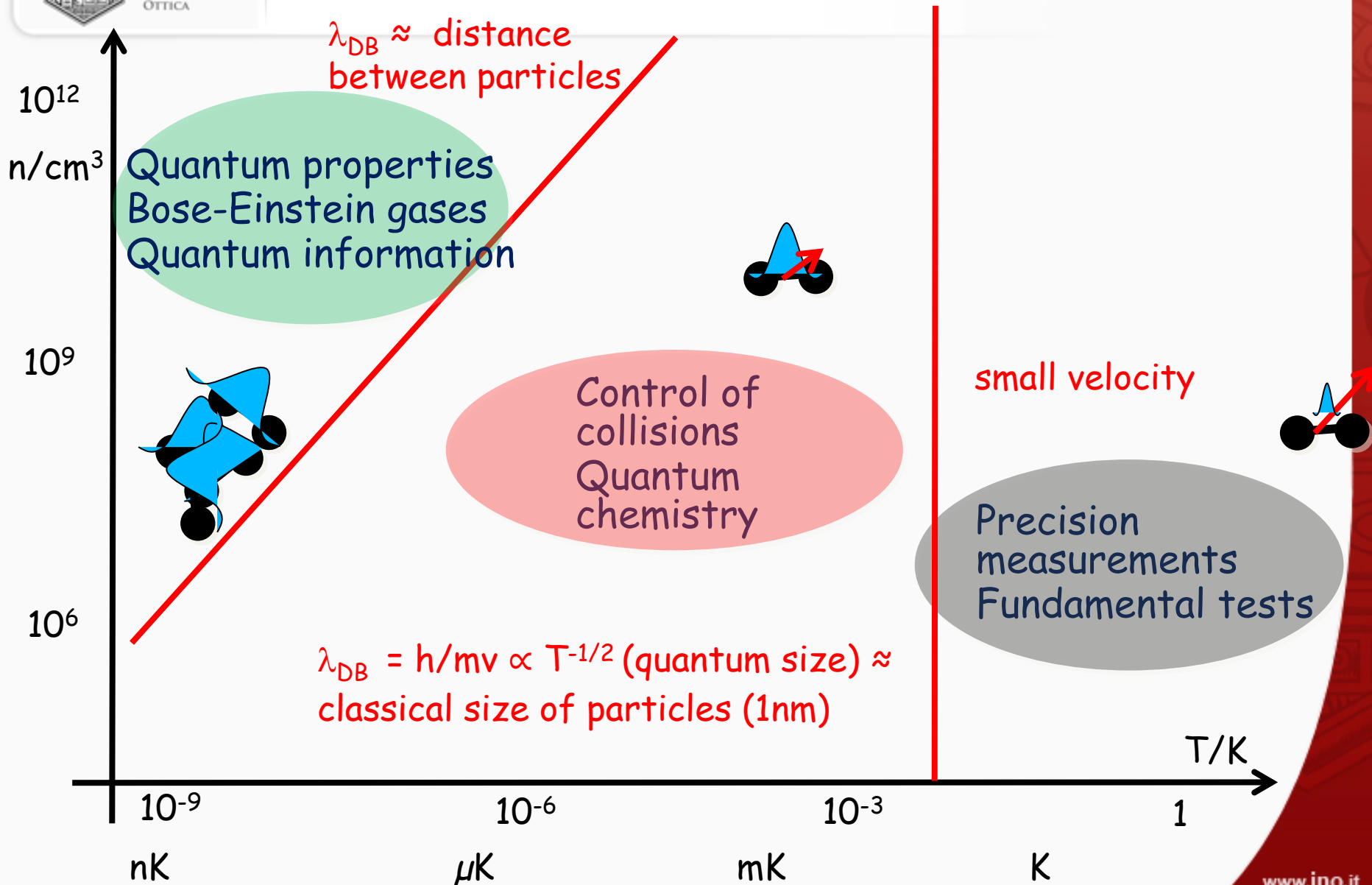


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# MOTIVATIONS FOR COLD MOLECULES





# MOTIVATIONS FOR COLD MOLECULES

- Precision measurements and fundamental tests

**Improved measurement of the shape of the electron**  
E. A. Hinds group, *Nature* **473**, 493-496 (26 May 2011)  
They use cold YbF molecules

- Quantum information with polar systems

**Quantum computation with trapped polar molecules.**

DeMille, D. , *Phys. Rev. Lett.* **88**, 067901 (2002) (PROPOSAL with RbCs)

A coherent all-electrical interface between polar molecules and mesoscopic supercond. resonators.

André, A. *et al.* *Nature Phys.* **2**, 636-642 (2006) (PROPOSAL with CaBr)

- Quantum gases, many-body physics

**A toolbox for lattice-spin models with polar molecules.**

Micheli, A., Brennen, G. K. & Zoller, P. *Nature Phys.* **2**, 341-347 (2006)  
(PROPOSAL with candidate polar molecules: CaF, CaCl and MgCl.)

- Cold collisions (instellar collisions, quantum collision regime, quantum degeneracy),... cold chemistry?

- ... **Low-energy collisions of NH<sub>3</sub> and ND<sub>3</sub> with ultracold Rb atoms.**  
Żuchowski, P. S. & Hutson, J. M. *Phys. Rev. A* **79**, 062708 (2009)



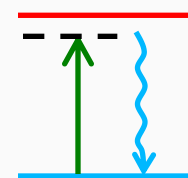
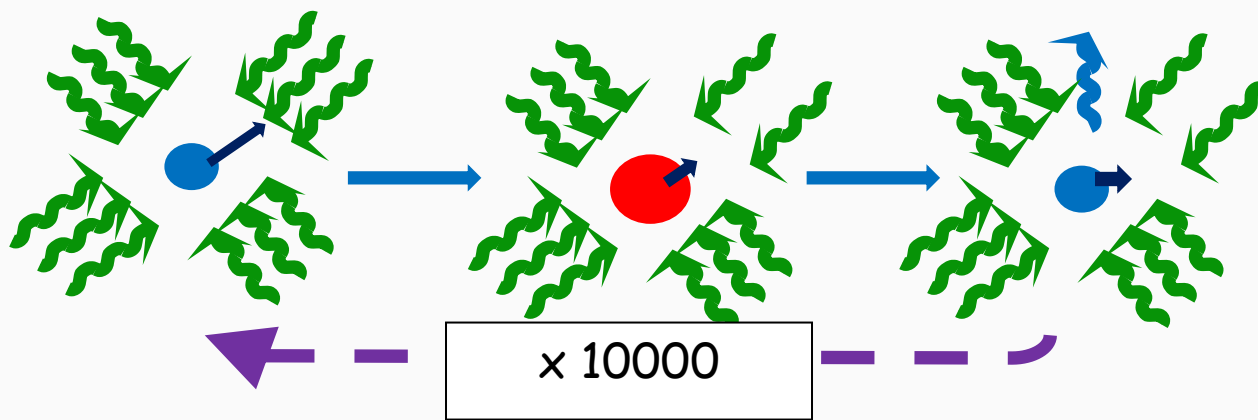
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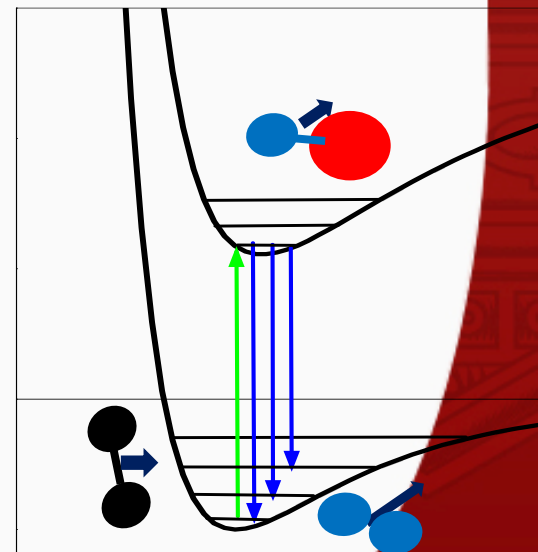
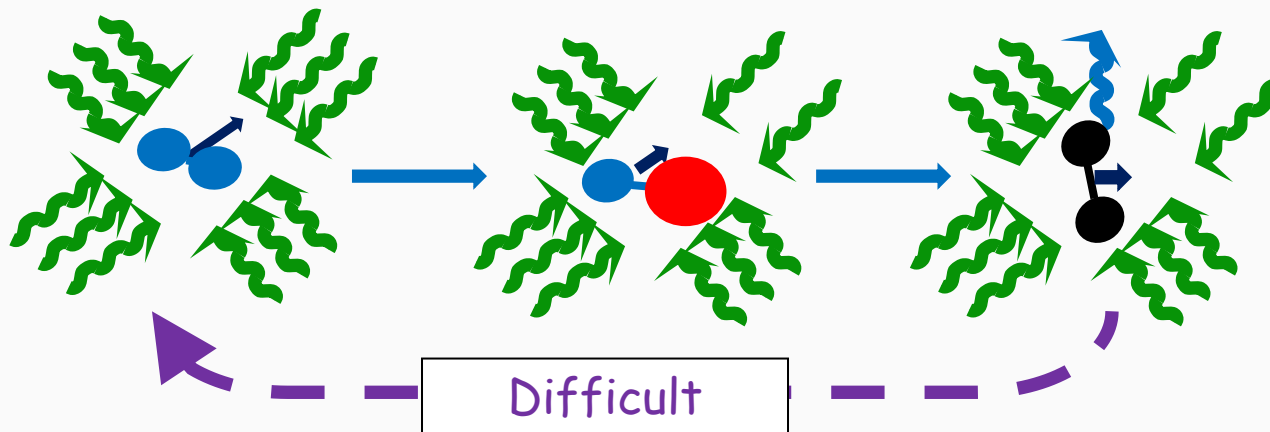


# LASER COOLING OF MOLECULES: POSSIBLE?

Laser cooling of atoms: many absorption-spontaneous emission cycles



Laser cooling of molecules?

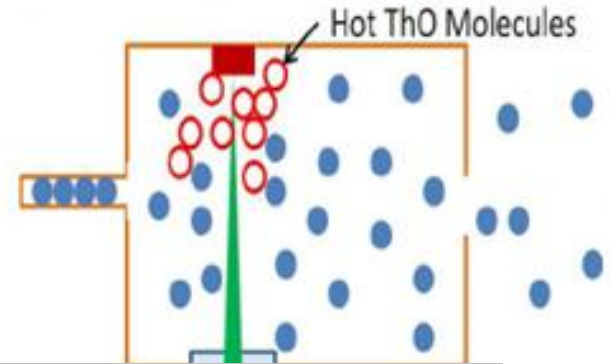






# COLD MOLECULE FORMATION 1

## from molecules



- Cryogenic method: Buffer gas cooling + magnetic trapping  
CaH, PbO, O<sub>2</sub>, ND<sub>3</sub>, ... (Doyle, Harvard 1998)

Recent achievements in direct laser cooling of molecules:

- **Transverse laser cooling and longitudinal slowing of SrF**

E. Shuman, J. Barry and D. DeMille, Nature (7317), 820 (2010).

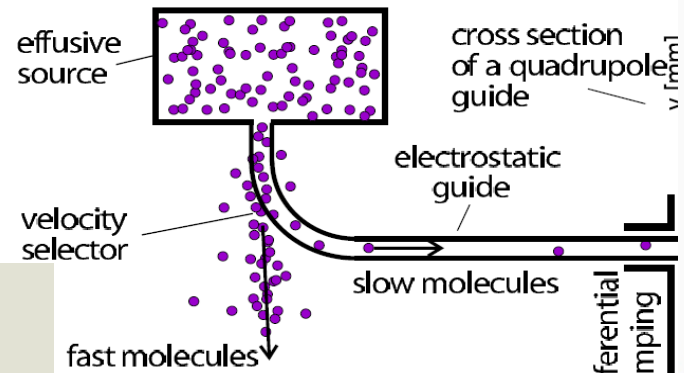
- **2-D magneto-optical trapping of YO**

M.T. Hummon, M. Yeo, B.K. Stuhl, A.L. Collopy, Y. Xia and J. Ye, 2012



- Velocity filtering by molecule guiding  
H<sub>2</sub>CO, CH<sub>3</sub>F, H<sub>2</sub>O, .... (G. Rempe 2004)

- **Translational** temperature ~ K-mK
- **Vibrational** and **rotational** temperatures ~ cold





## COLD MOLECULE FORMATION 2

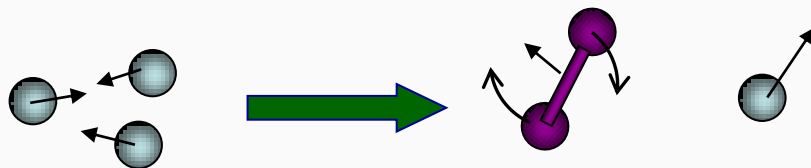
### from pre-cooled atoms

( $\text{Cs}_2$ ,  $\text{K}_2$ ,  $\text{Rb}_2$ ,  $\text{RbCs}$ ,  $\text{KRb}$ ,  $\text{LiCs}$ ,  $\text{NaCs}$ ...)

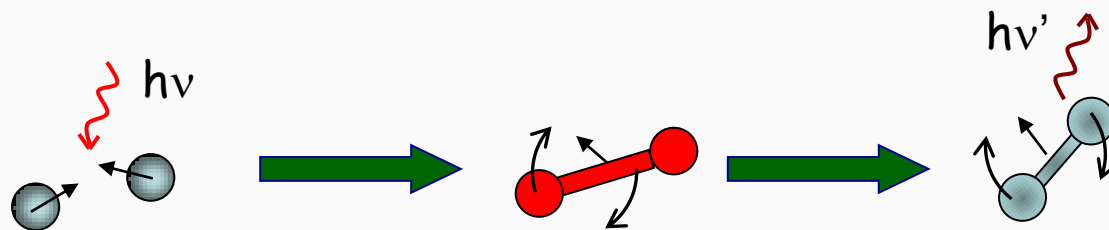
- Magneto-association  
(Feshbach resonance)



- 3-Body Collision



- Photo-association





- Temperature (*speed*)  $\sim$  atomic temperature  $\sim$  ultracold (nK- $\mu$ K)
- *Vibrational* temperature  $\sim$  **HOT**
- *rotational* temperature  $\sim$  cold but several levels occupied





# SUMMARY OF REQUIREMENTS

## From molecules

- **Motion**   
Temperatures ~ mK-K
- **Vibration and rotation:**   
In general cold but **many** levels  
are occupied

## From cold atoms

- **Motion**   
– Temperatures ~ nK-mK
- **Vibration:** very excited and/or  
many levels occupied 
- **Rotation:** low but several levels  
occupied

## AIM

**Control and cool all  
degrees of freedom**

**EXTERNAL:**

**Motion, position**

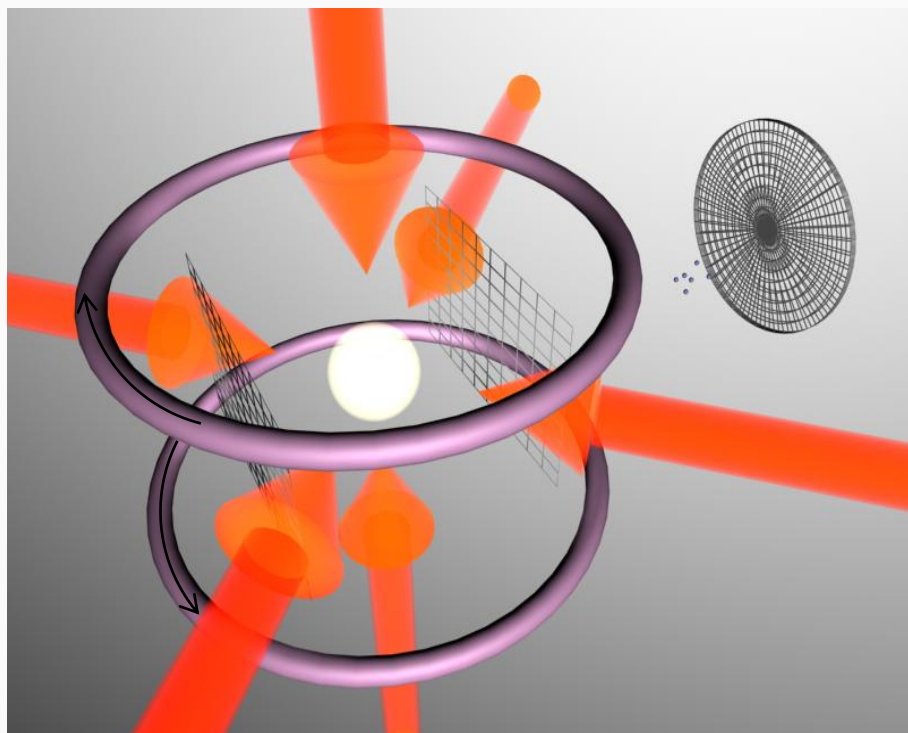
**INTERNAL:**

**electronic state,  
vibrational, rotational  
and hyperfine level**



Work horse: a cesium magneto-optical trap MOT

$N \sim 5 \cdot 10^7$  atomes,  $n \sim 10^{11}$  at/cm<sup>3</sup>,  $T \sim 100 \mu\text{K}$

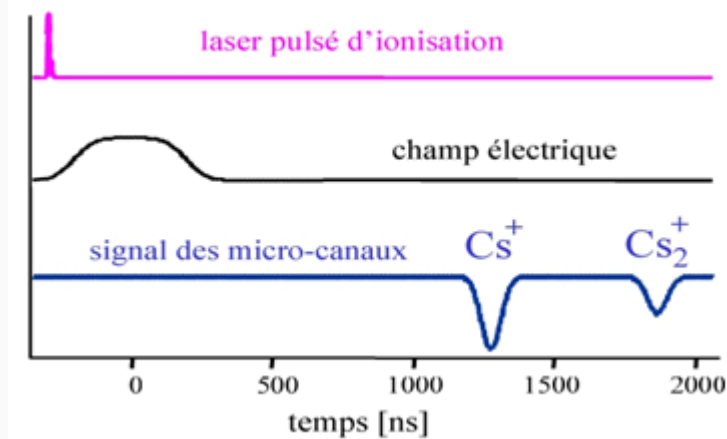
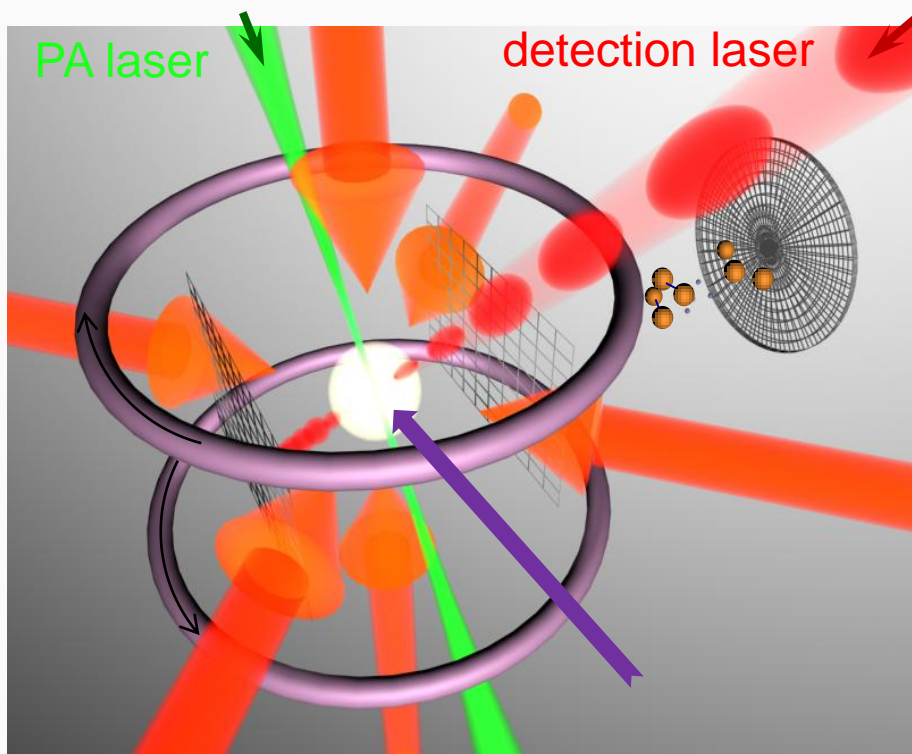




## EXPERIMENT

Work horse: a cesium magneto-optical trap (MOT)

$N \sim 5 \cdot 10^7$  atomes,  $n \sim 10^{11}$  at/cm<sup>3</sup>,  $T \sim 100 \mu\text{K}$

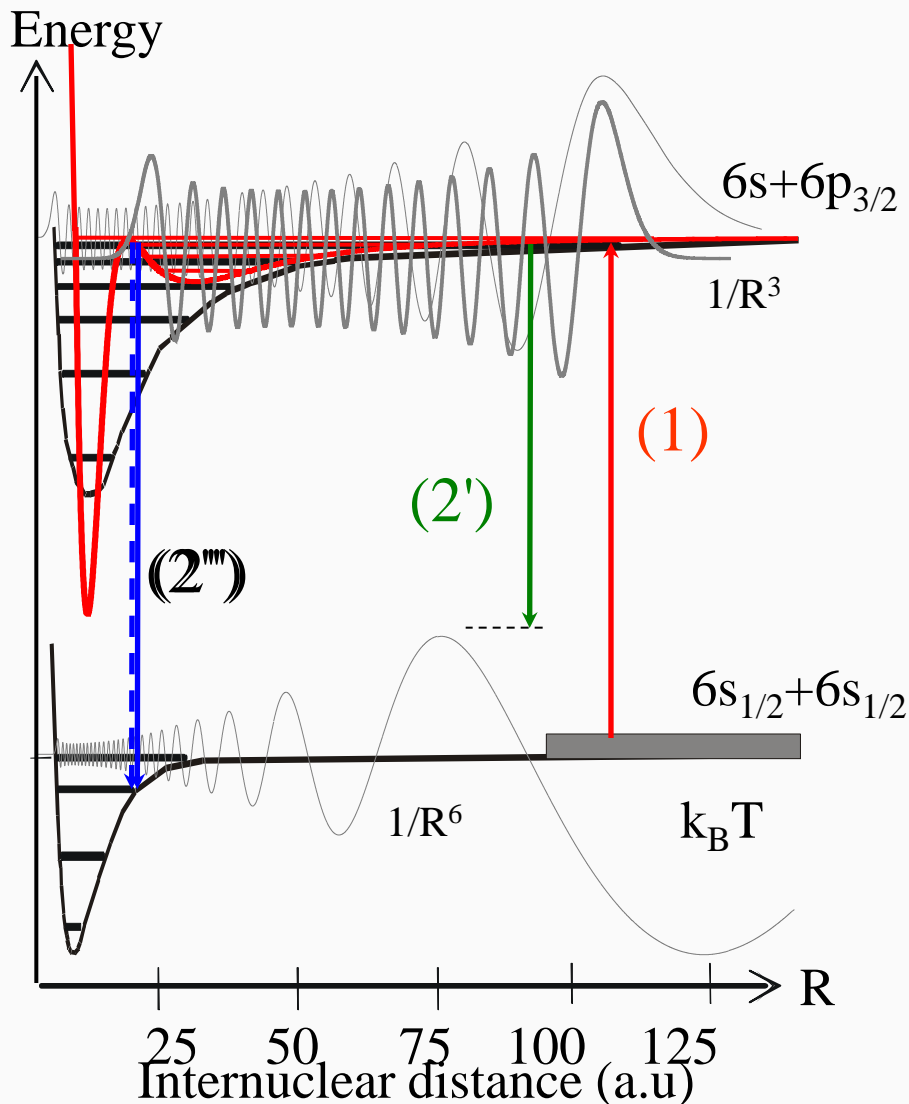


- MOT: diode lasers, cw, 852nm, 150mW, frequency stabilized <1MHz
- PA: Ti:Sapphire cw, 852 nm, ~1W, linewidth  $\leq 1$ MHz
- Detection (ionisation): pulsed laser 10 Hz-7ns, 5-10mJ/pulse

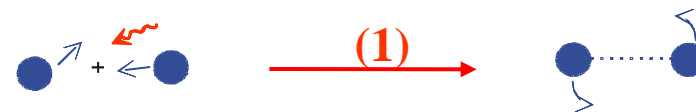
-Vibrational cooling: broadband (femtosecond) laser, 80MHz, 770 nm, 1W  
(but other choices are possible)



# PHOTOASSOCIATION

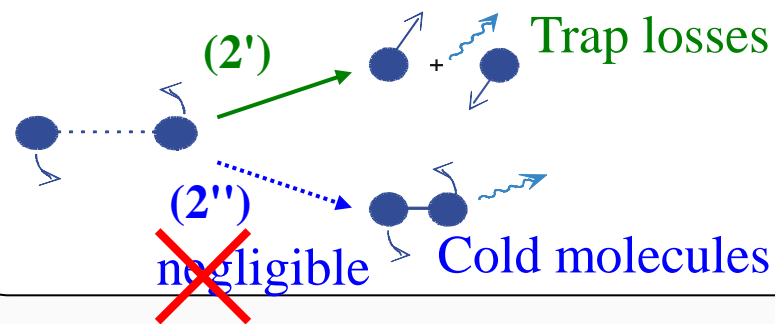


## Photoassociation



Resonant absorption of one photon  
by 2 colliding cold atoms ( $T \sim 100 \mu\text{K}$ )

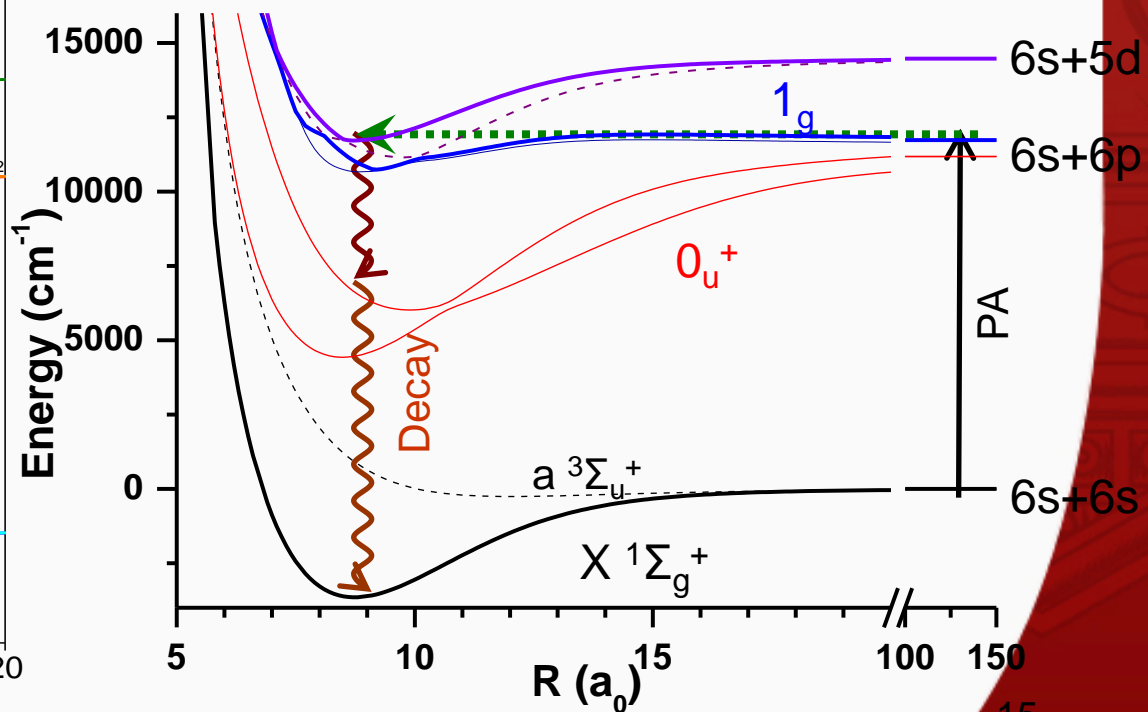
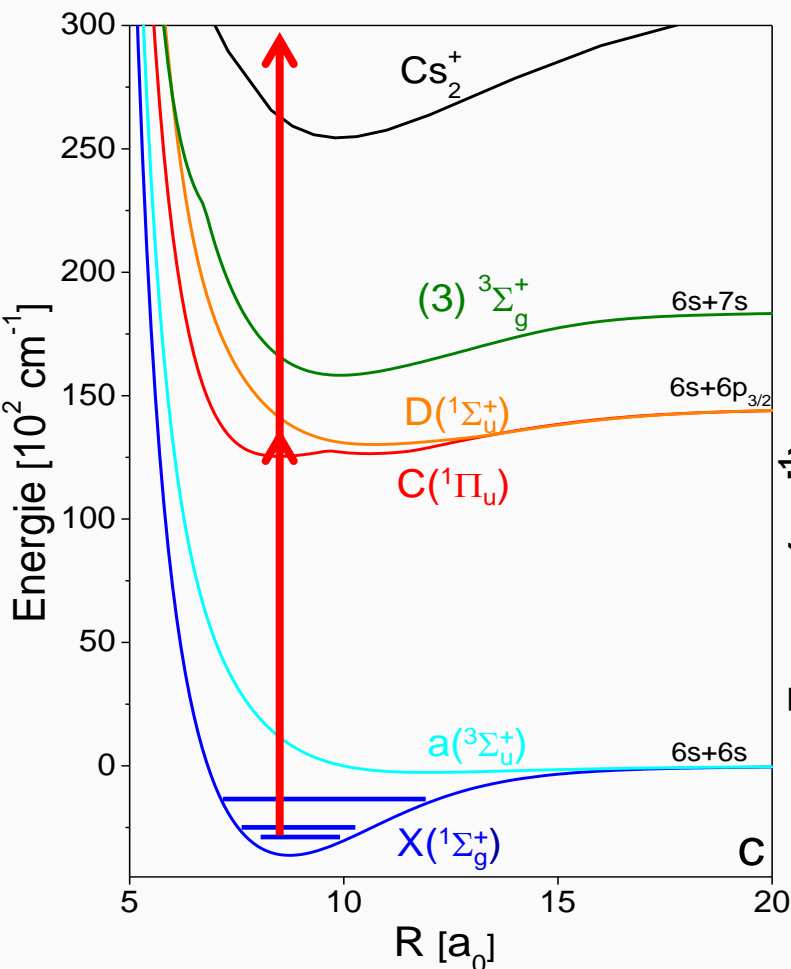
## Emission





# PA: FORMATION in the $X^1\Sigma_g^+$ state

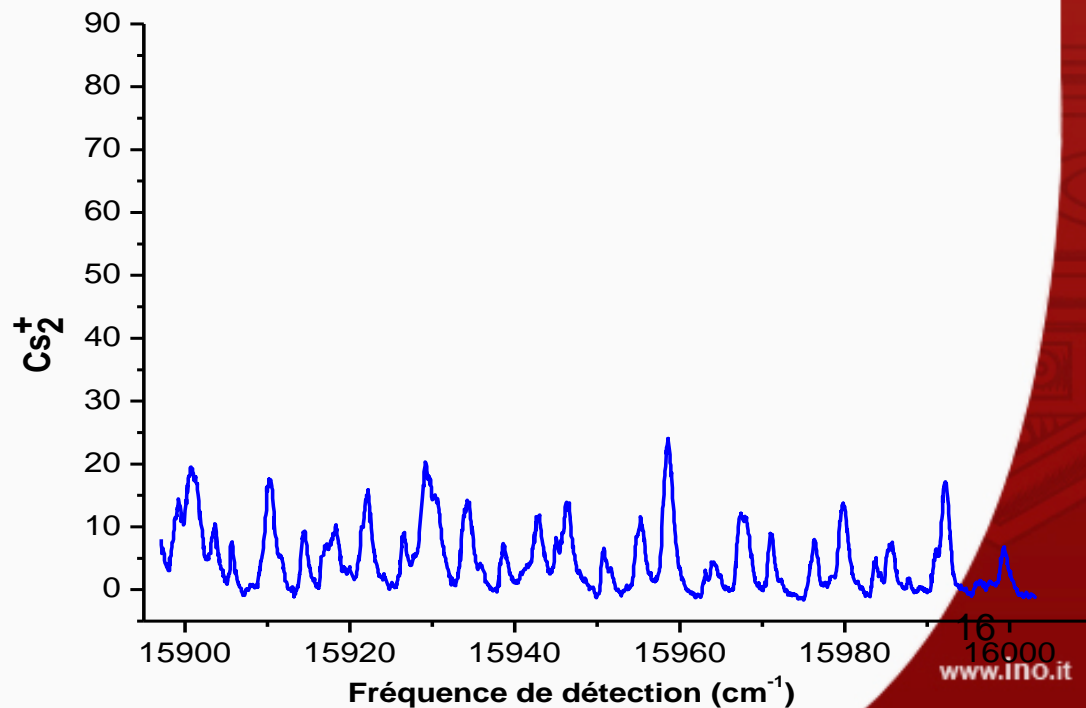
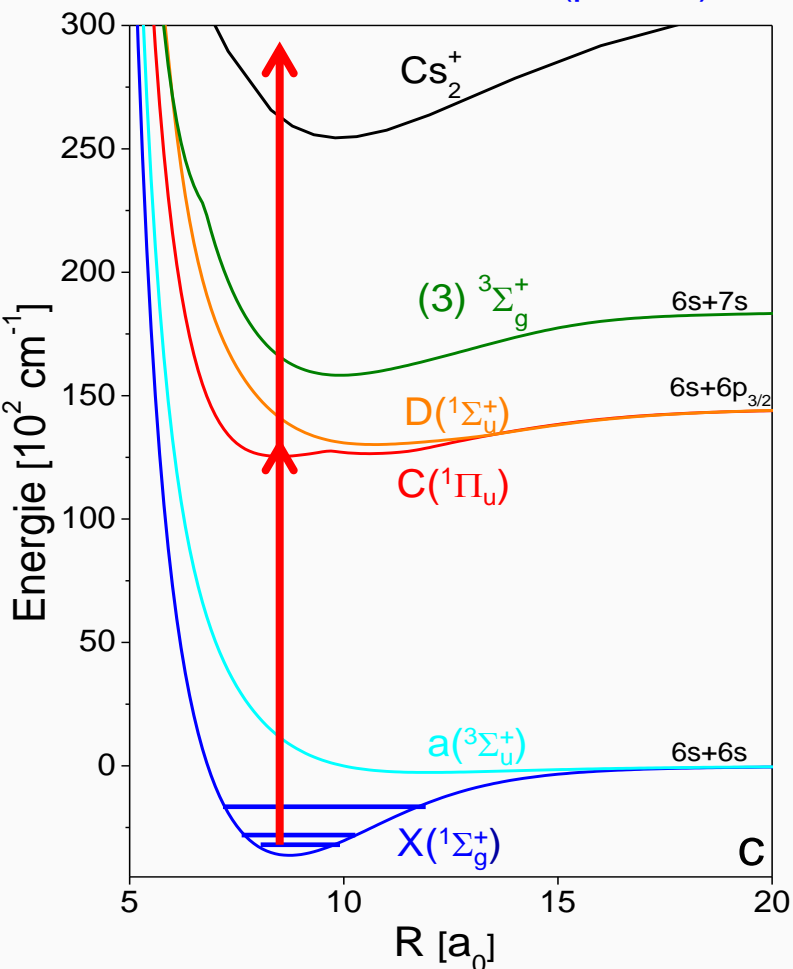
- Vibrationally selective detection: 2-photon ionization with resonant intermediate state (REMPI)





# PA: DETECTION in the $X^1\Sigma_g^+$ state

- **Vibrationally selective detection:** 2-photon ionization with resonant intermediate state (REMPI)
- Scan of detection laser (pulsed)

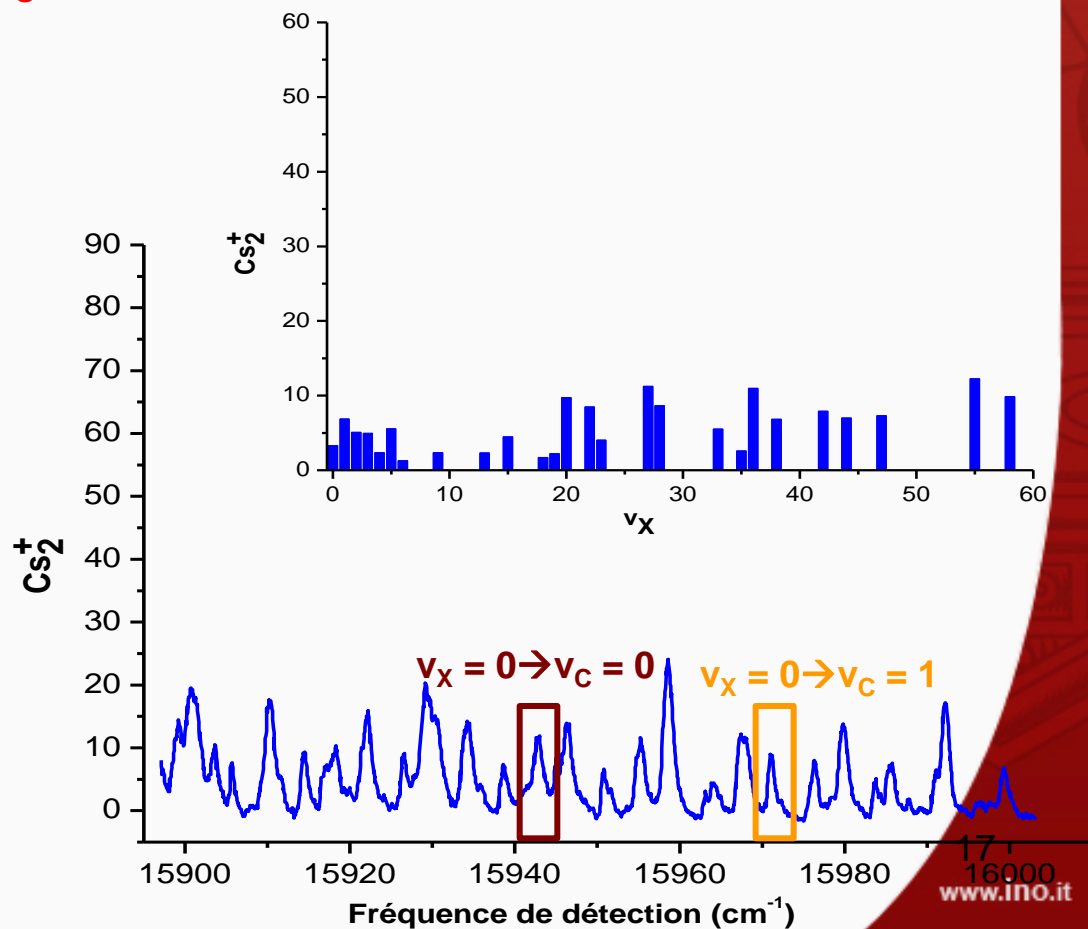
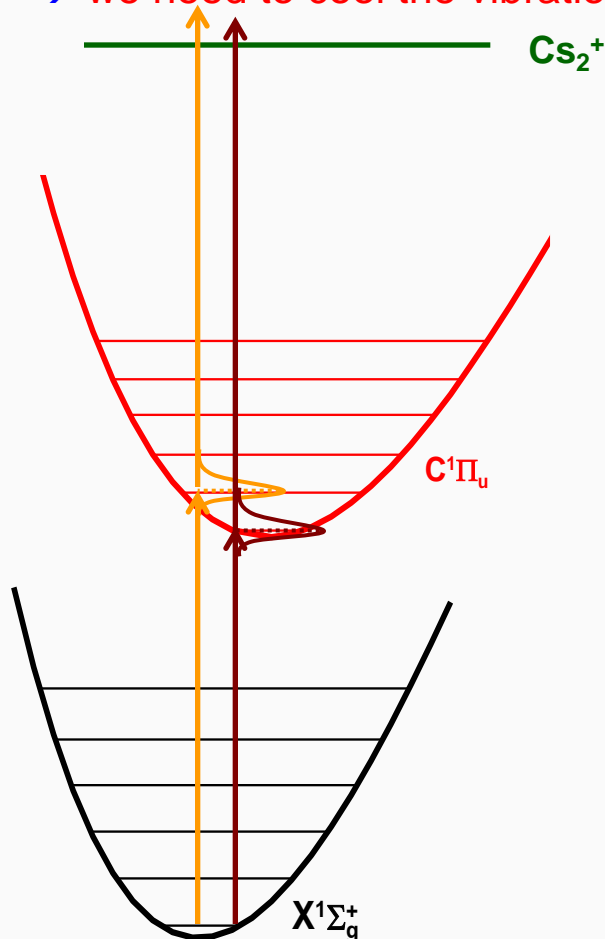






# PA: DETECTION in the $X^1\Sigma_g^+$ state

- several vibrational levels are populated
- we need to cool the vibrational degree of freedom





# OUTLINE

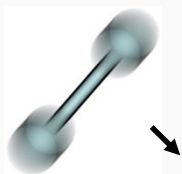
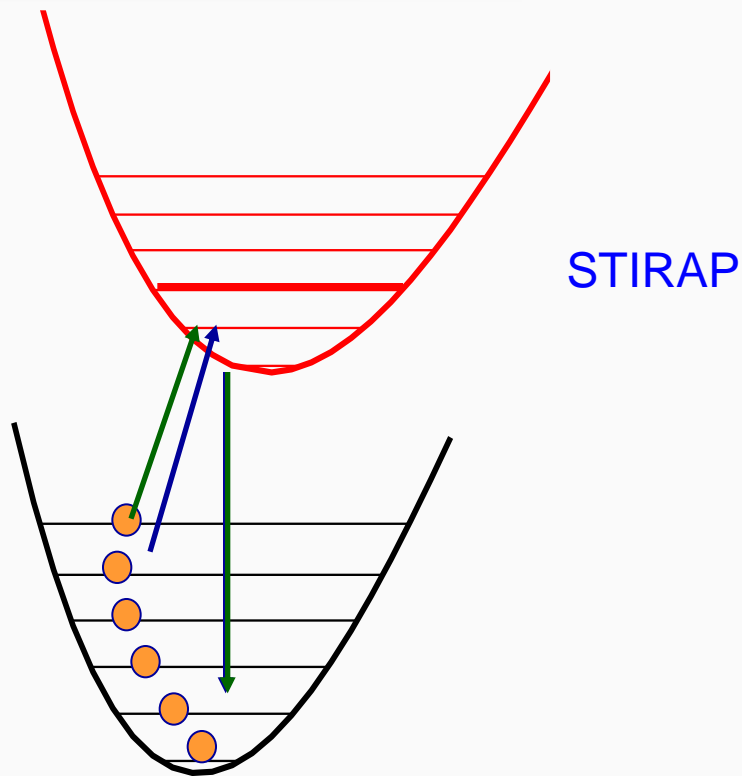
- Introduction. Why cold molecules?
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# VIBRATIONAL COOLING 1: STIRAP method?

Very efficient to transfer molecules already in a single level (i.e. Feshbach molecules):  $\text{Cs}_2$ ,  $\text{KRb}$ ,  $\text{Rb}_2$

Not efficient to transfer molecules distributed over many levels



Traslationaly cold  
but  
Vibrationally **HOT**

Our aim

Traslationaly  
and  
Vibrationally  
**cold**





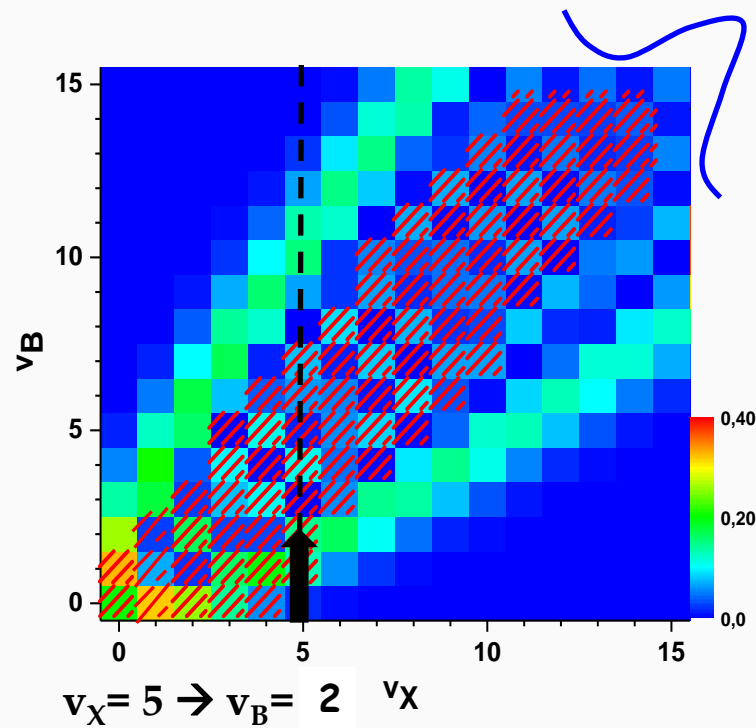
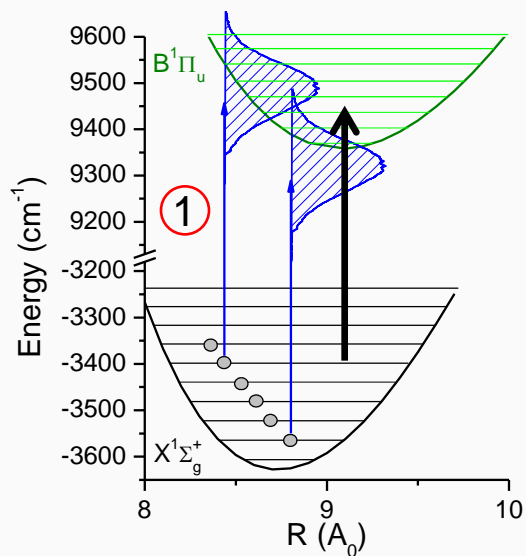
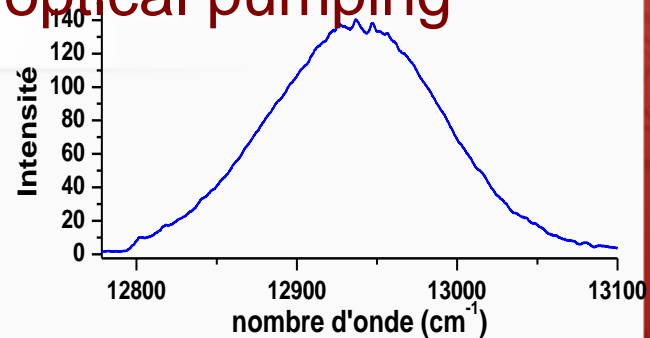
# VIBRATIONAL COOLING 2: optical pumping

➤ Broadband laser (femtosecond laser:  $200 \text{ cm}^{-1}$ )

→ Excites all levels towards the target state B

→ Excitation probability:

$$\Gamma_{v_X \rightarrow v_B} \propto FC[v_X][v_B] (D[v_X][v_B])^2 I_{laser}[v_X][v_B]$$



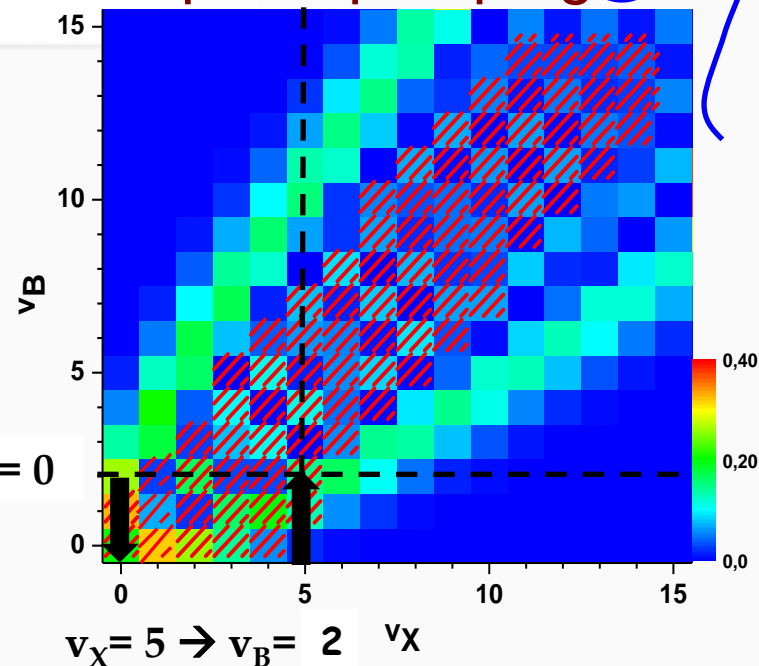


# VIBRATIONAL COOLING 2: optical pumping

➤ Emission spontanée → Redistribution des molécules dans l'état X

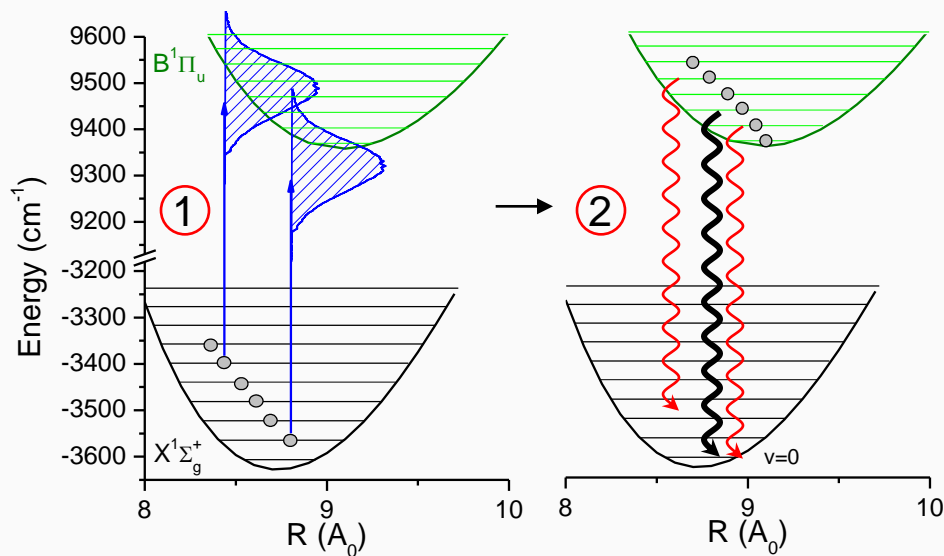
→ Probabilité de désexcitation :

$$\Gamma_{v_B \rightarrow v_X} \propto FC[v_X][v_B] (D[v_X][v_B])^2 \omega_{v_B \rightarrow v_X}^3$$



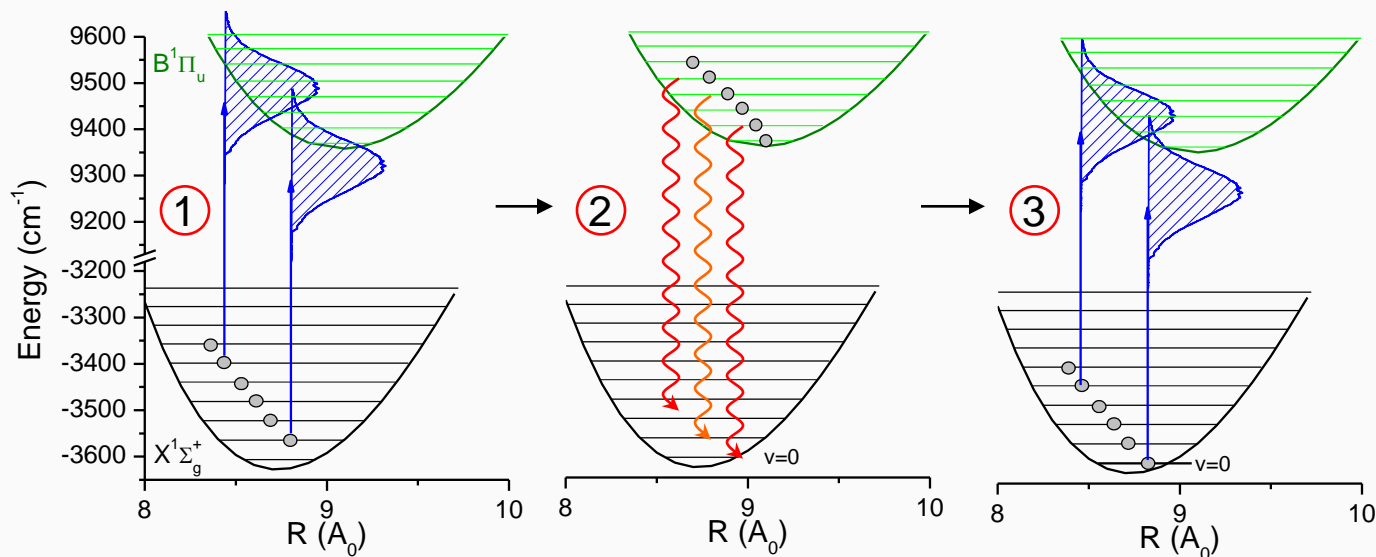
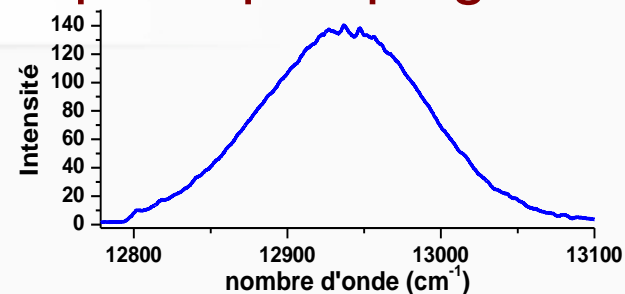
$v_B=2 \rightarrow v_X=0$

$v_X=5 \rightarrow v_B=2$





# VIBRATIONAL COOLING 2: optical pumping

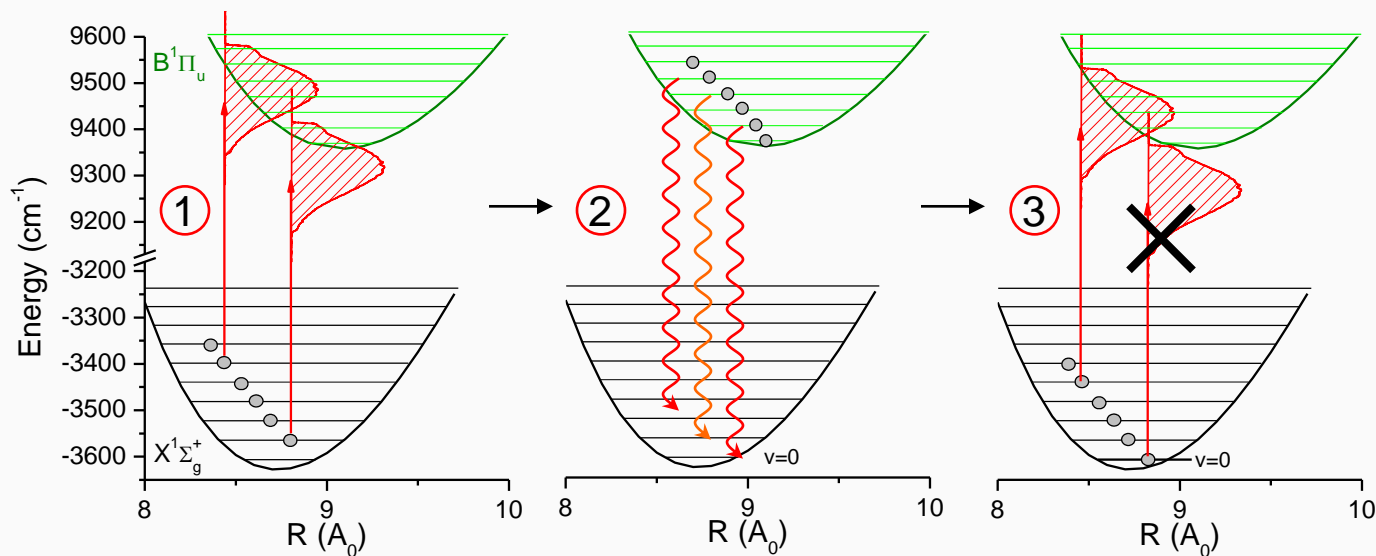
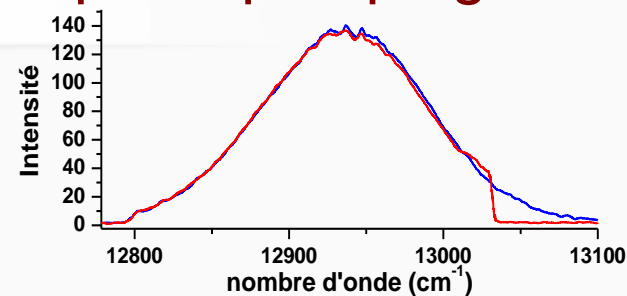




## VIBRATIONAL COOLING 2: optical pumping

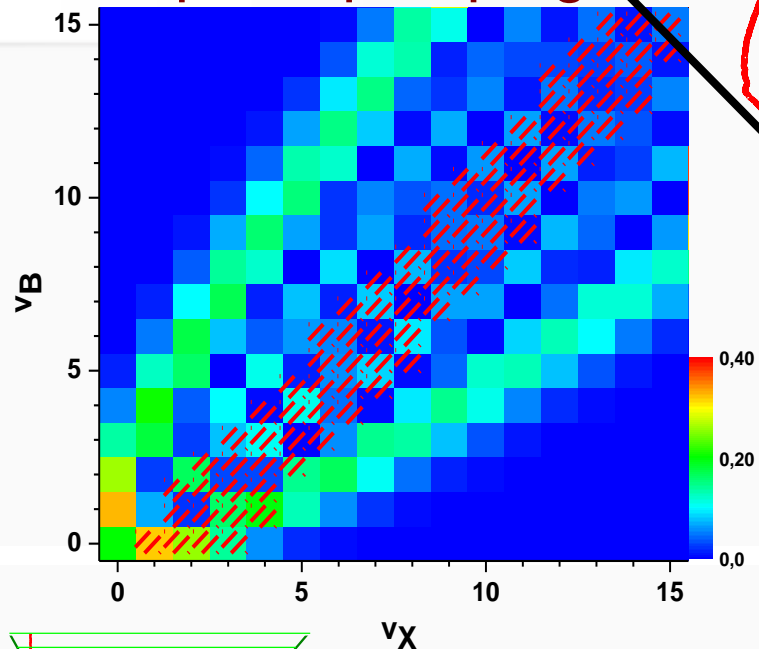
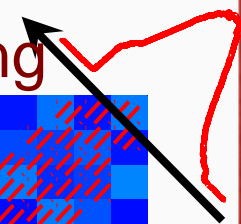
- Shaped broadband laser:  
no transition from  $v=0$

→ **dark state**

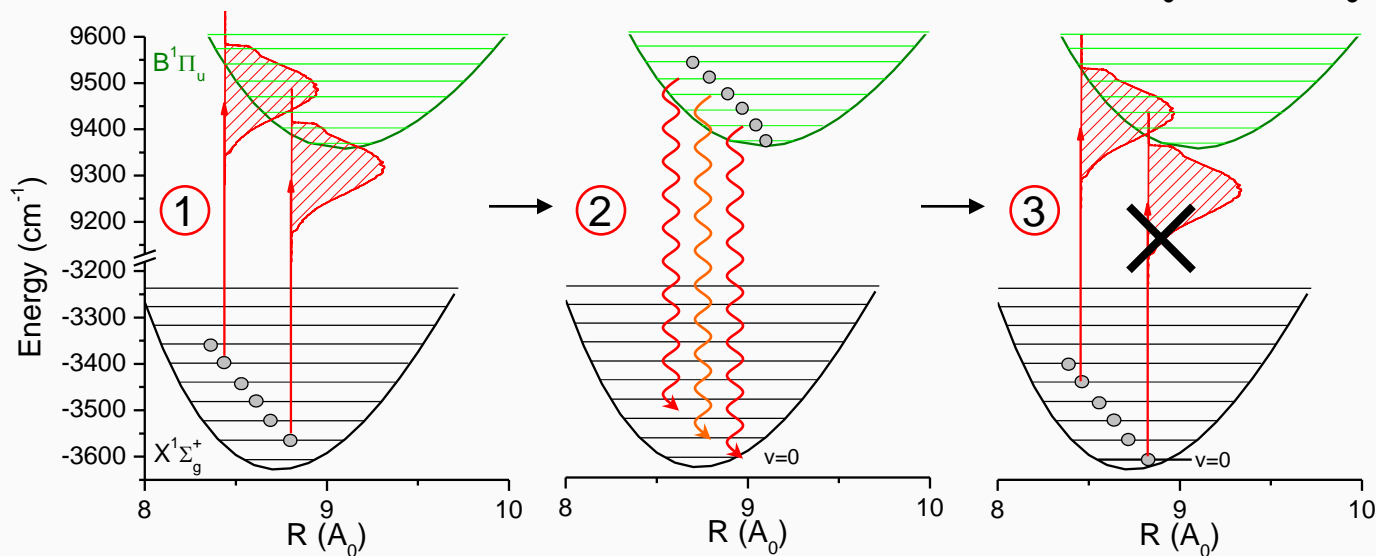




# VIBRATIONAL COOLING 2: optical pumping



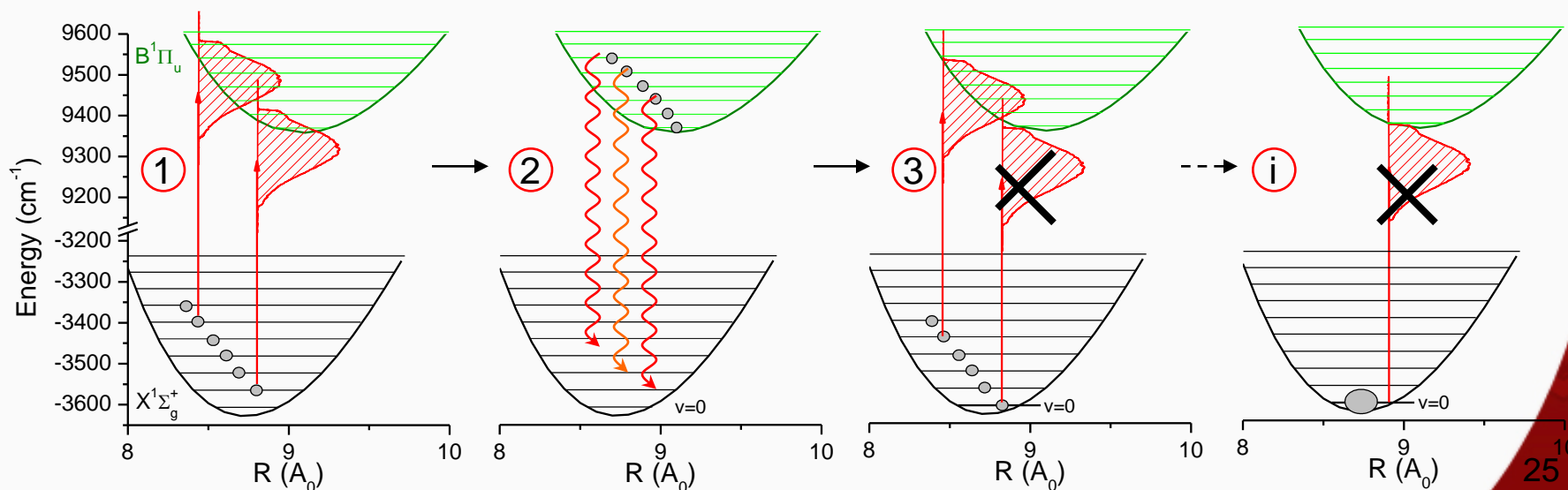
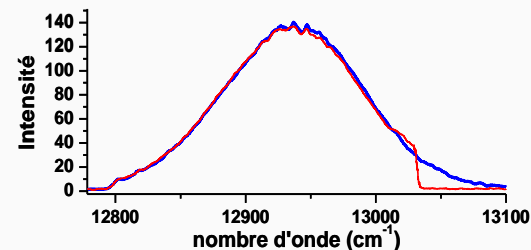
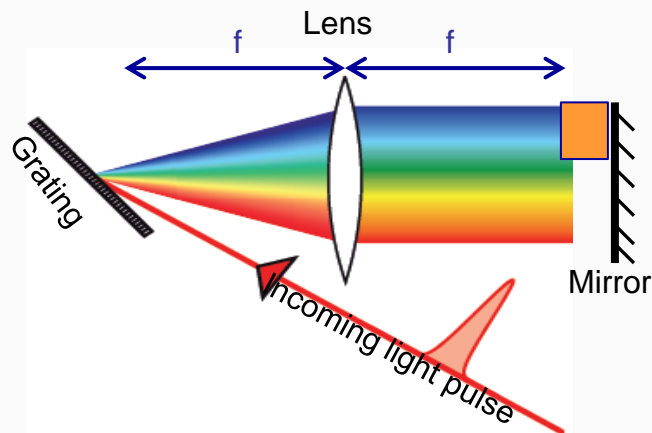
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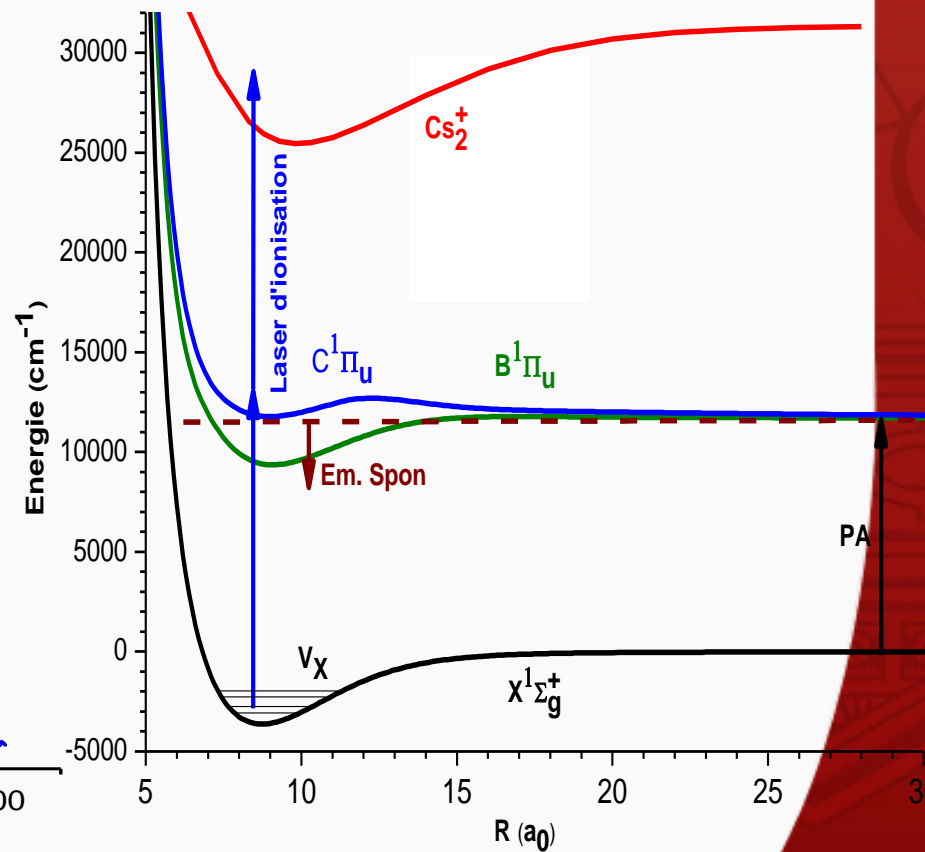
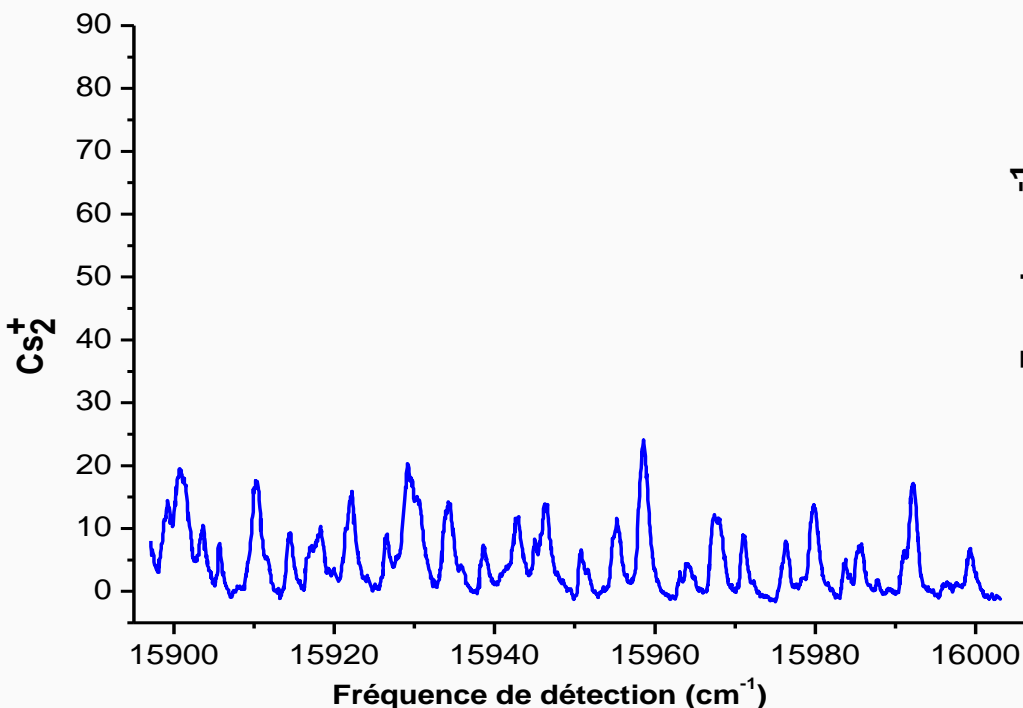
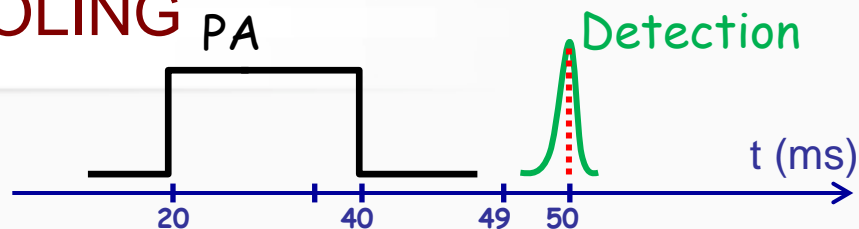




# BEFORE VIBR-COOLING PA

➤ Detection spectrum (via the C state)

→ Molecules distributed over many vibrational levels





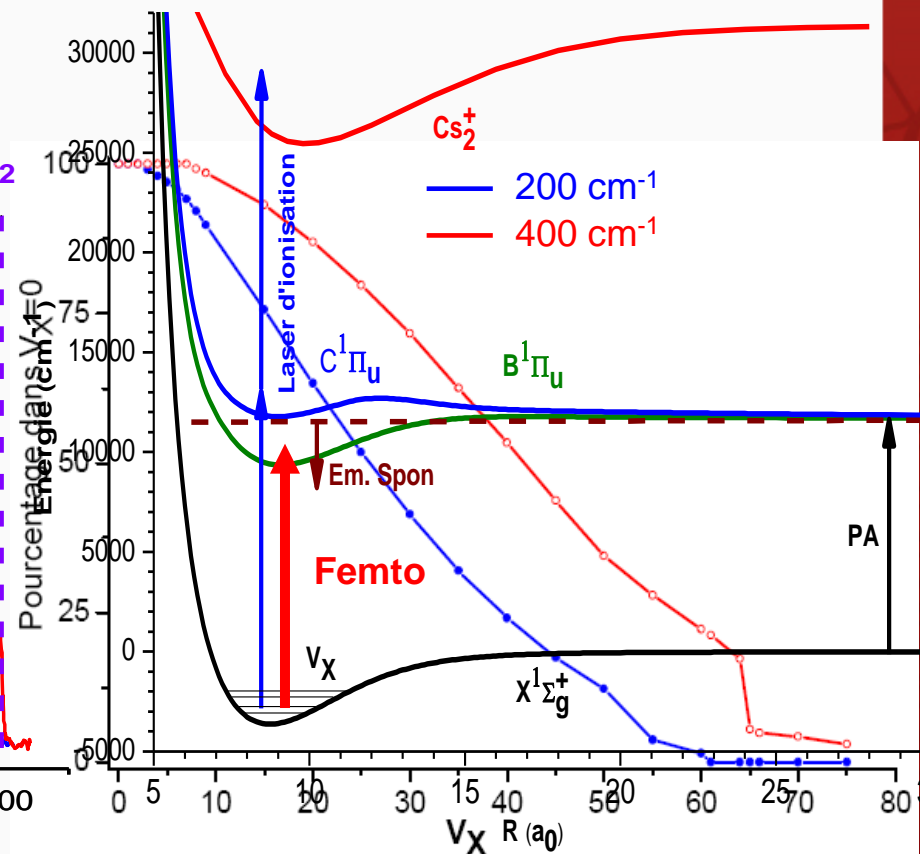
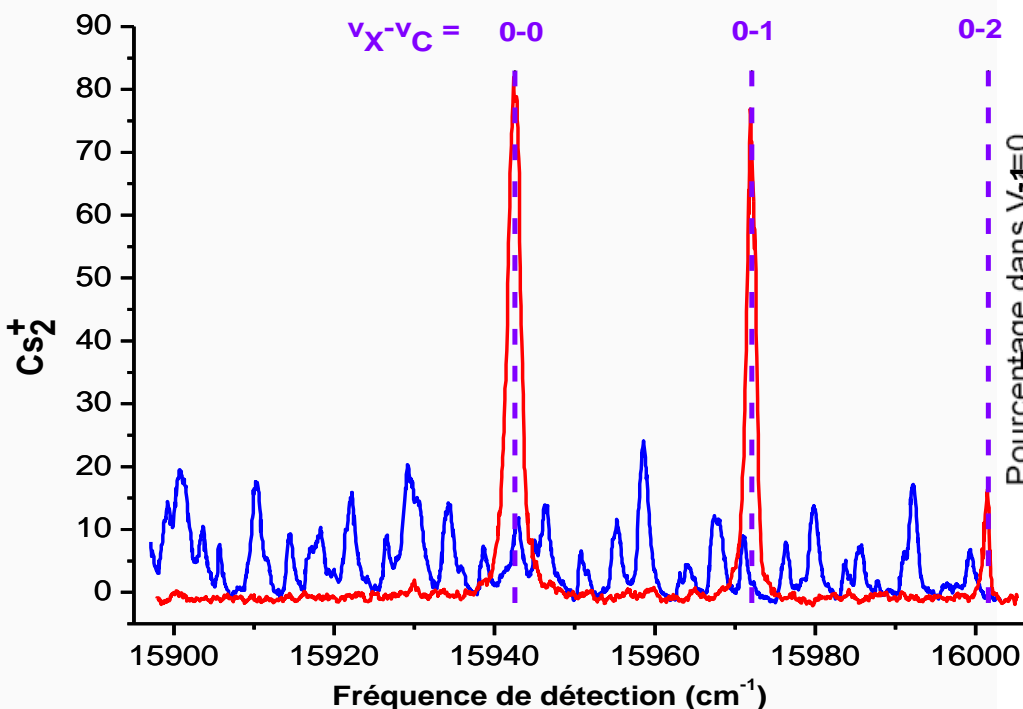
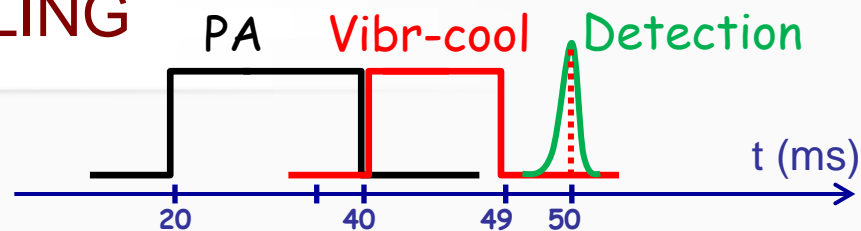
# AFTER VIBR-COOLING

➤ With an optical pumping phase

Molecule pile up in the DARK state  $v=0$

→ Efficiency ~ 80 %

→ limited also by spectral bandwidth of the laser





# OUTLINE

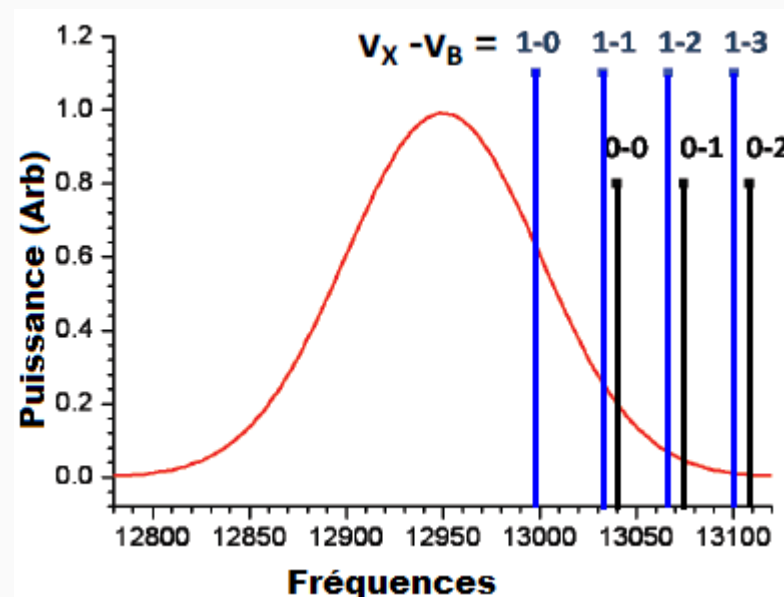
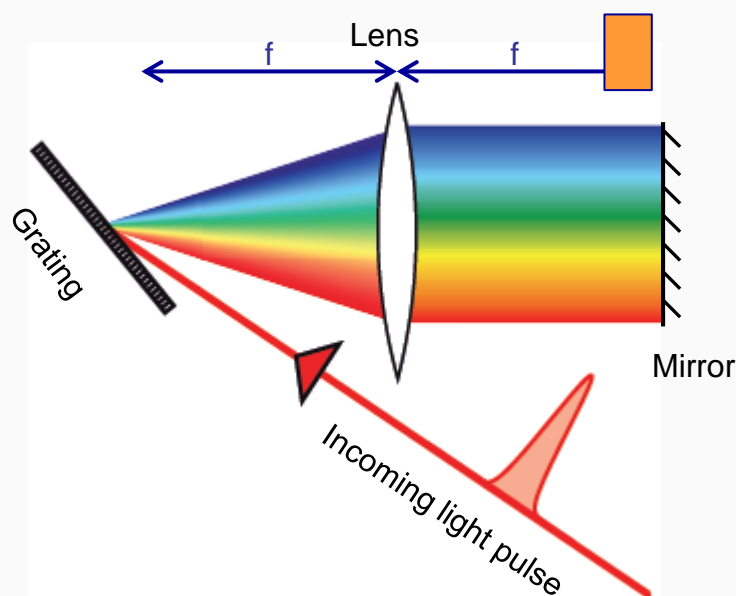
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## Pumping into a PRE-SELECTED LEVEL

How to accumulate molecules into another pre-selected level?

→ suppress ALL laser frequencies connecting the target level (example  $v=1$ ) to excited states

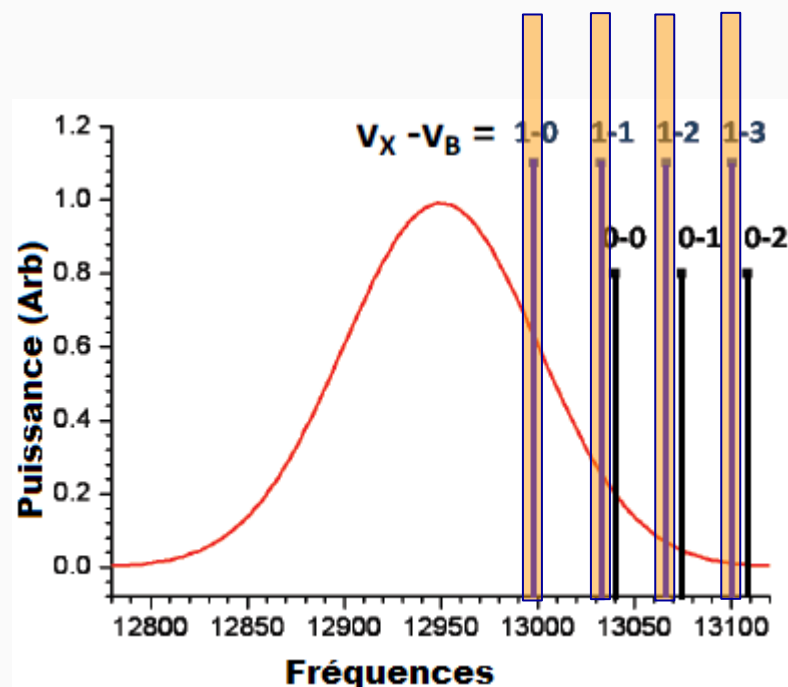
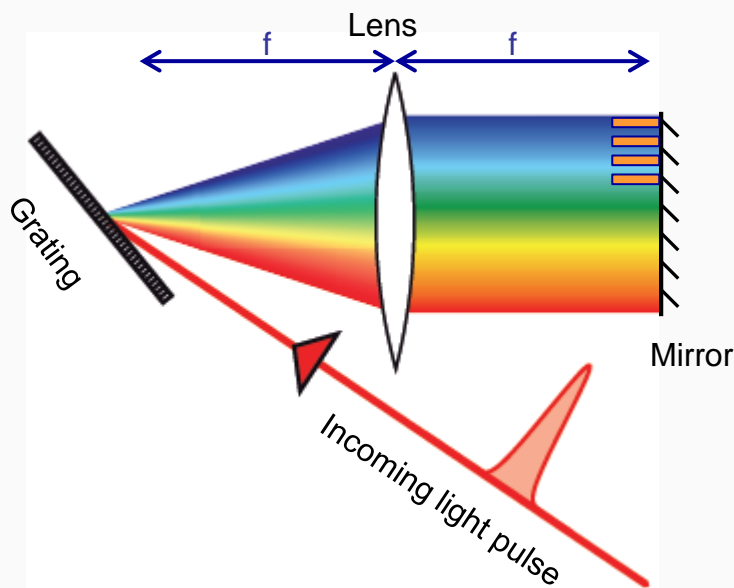




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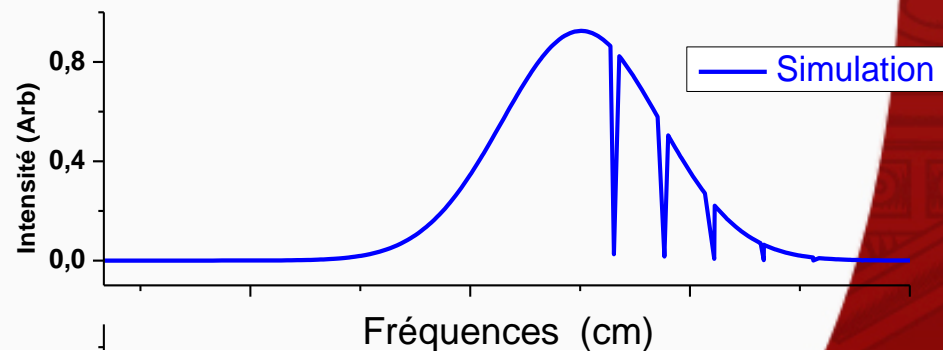
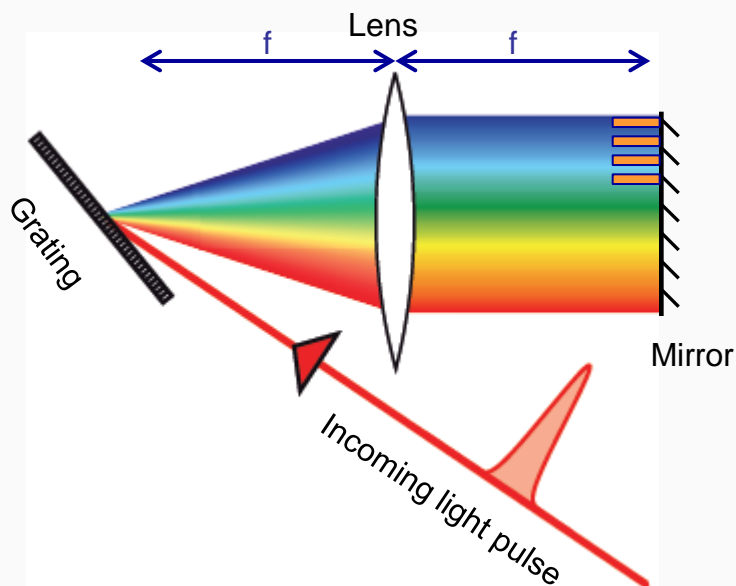




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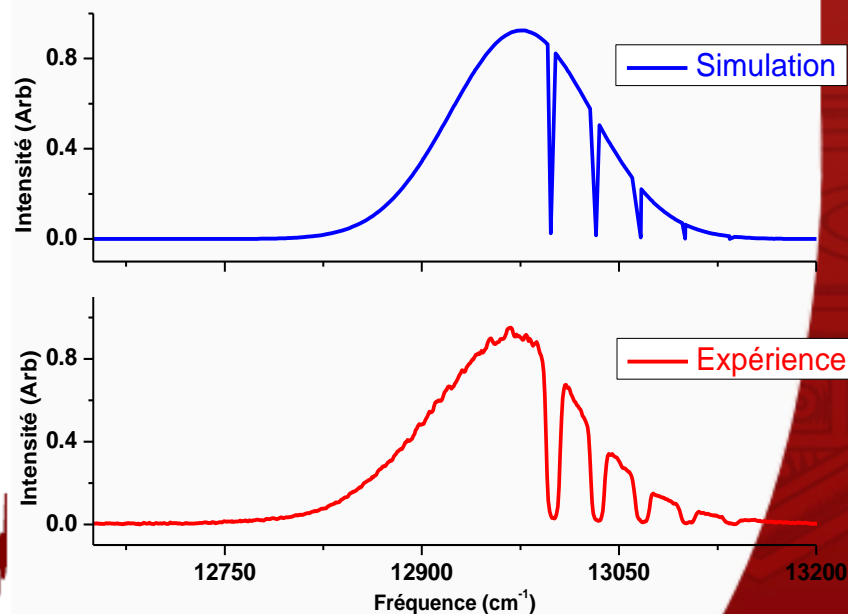
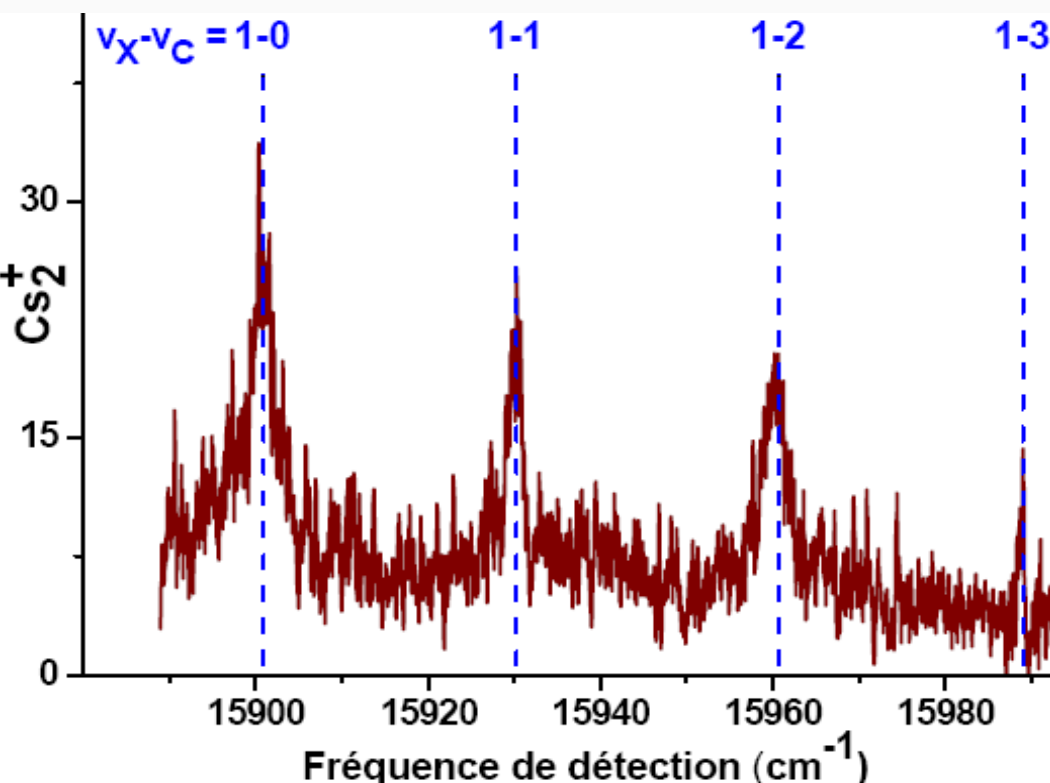
# Pumping into a PRE-SELECTED LEVEL

How to accumulate molecules into another pre-selected level?

→ suppress ALL laser frequencies connecting the target level (example  $v=1$ ) to excited states

→ different option to spectrally shape the laser

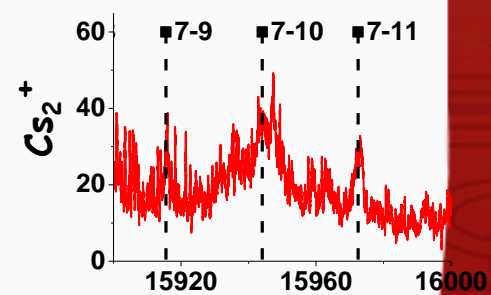
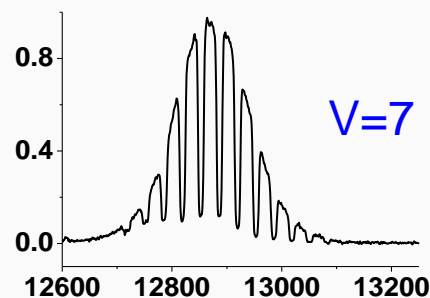
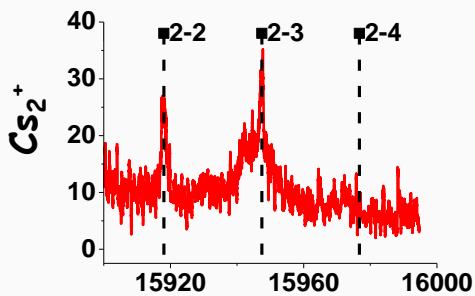
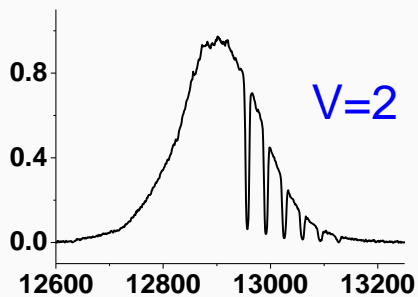
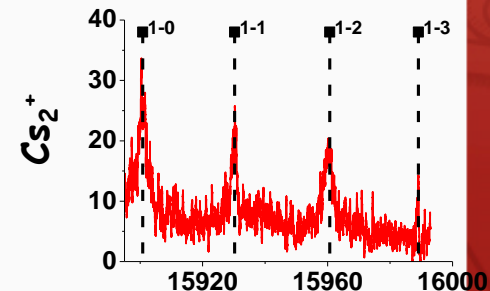
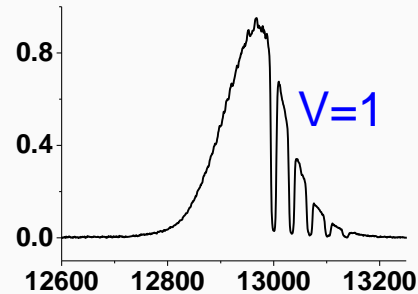
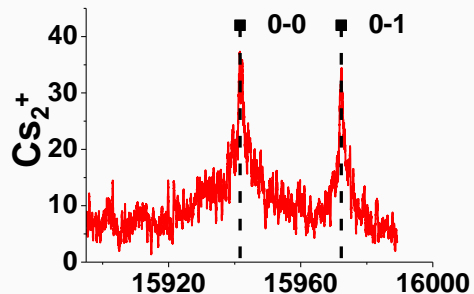
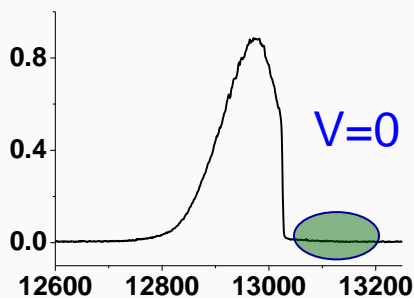
- 1) Liquid Crystal Spatial Light Modulator LC-SLM (collaboration with B. Chatel, Toulouse)
- 2) mechanical mask
- 3) micro-mirror array







# Pumping into a PRE-SELECTED LEVEL



Efficiency  $\sim 50\%$  limited by :

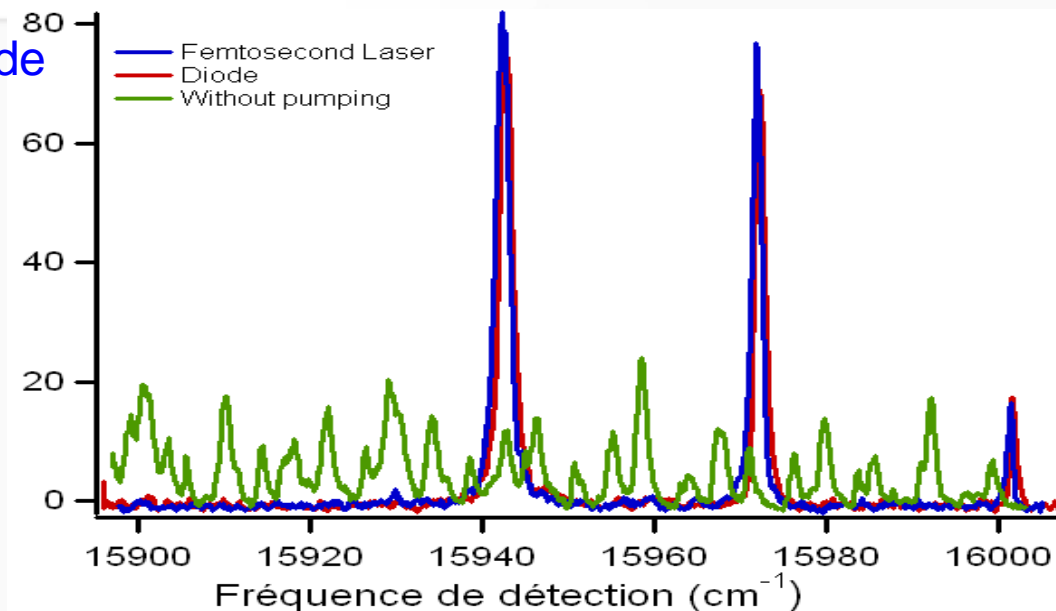
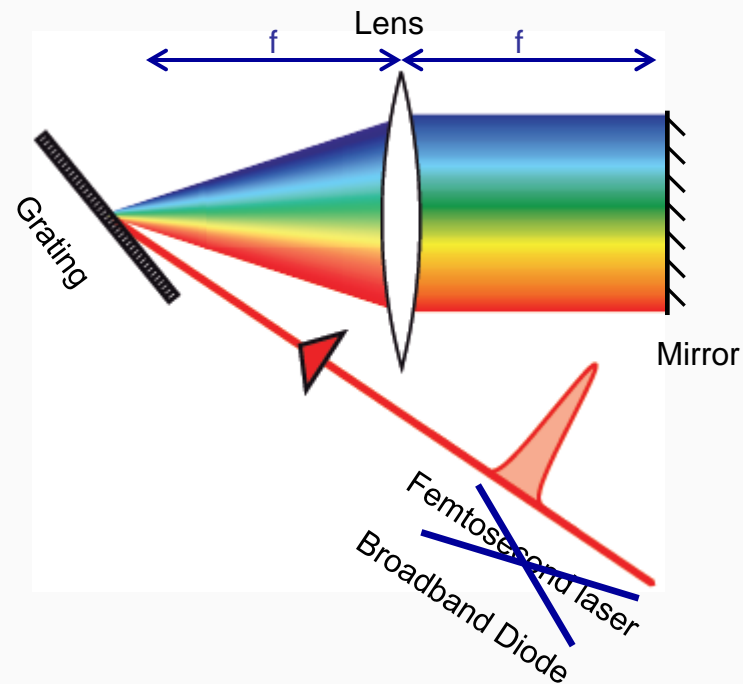
- Laser bandwidth
- SLM extinction ratio  $\sim 97\%$

SLM resolution  $\sim 0,06\text{nm} = 0,8\text{cm}^{-1}$  @ 852nm  
Vibrational spacing  $\sim 40\text{cm}^{-1}$



# VIBR-COOLING WITH INCOHERENT LIGHT

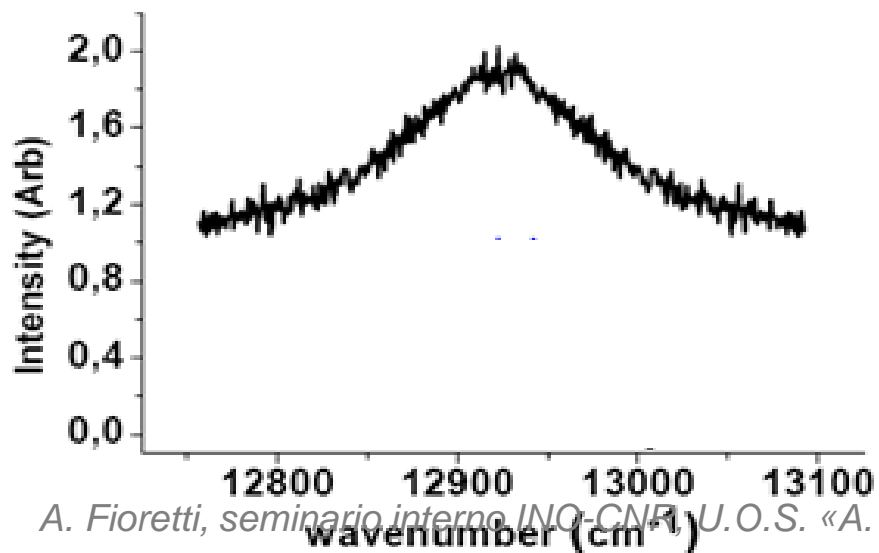
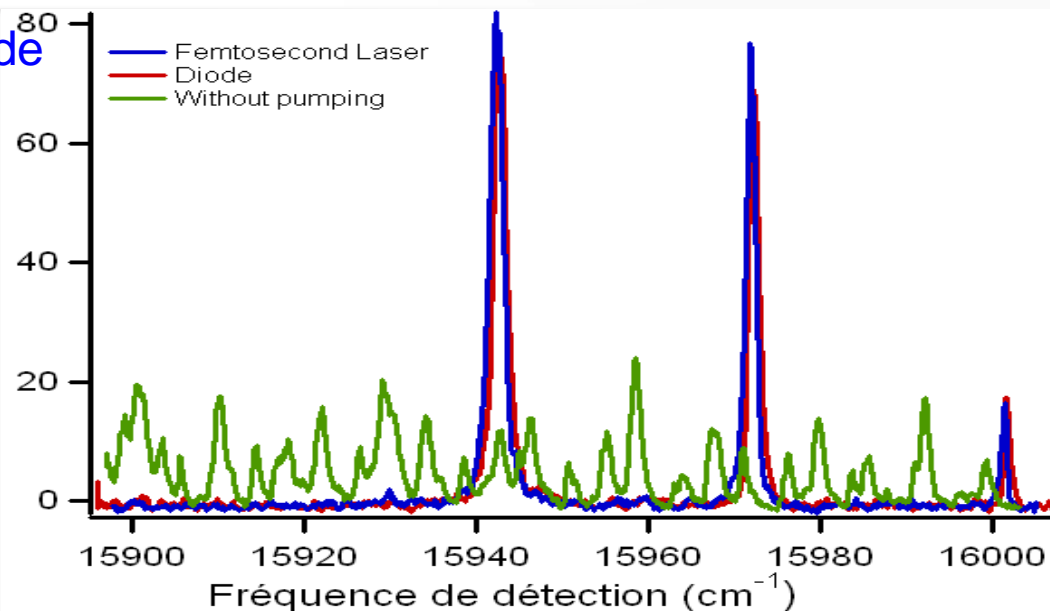
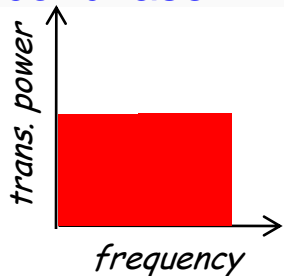
femtosecond laser → broadband diode





# VIBR-COOLING WITH INCOHERENT LIGHT

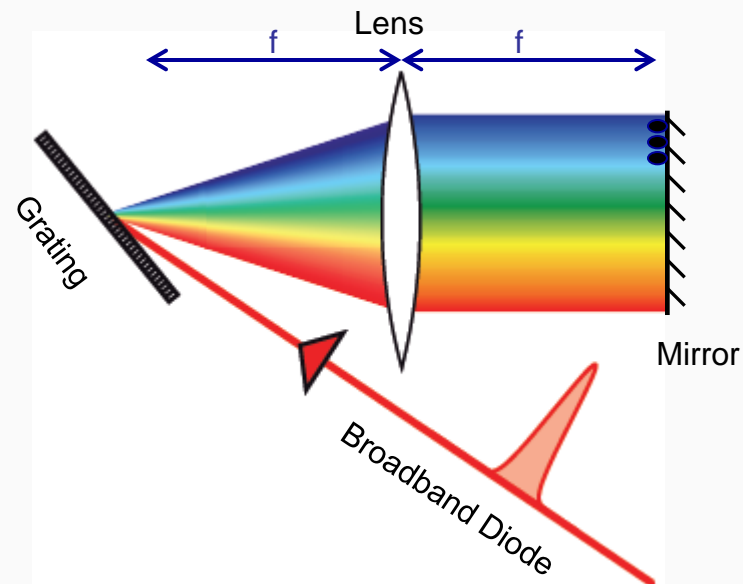
femtosecond laser → broadband diode



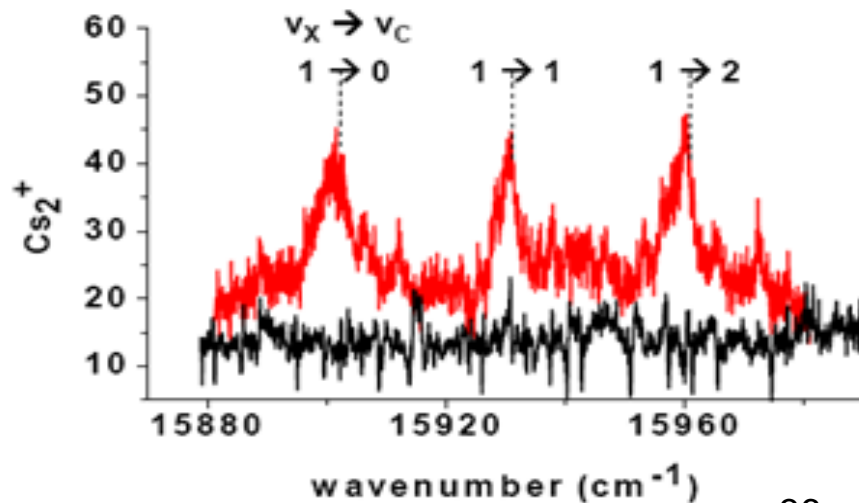
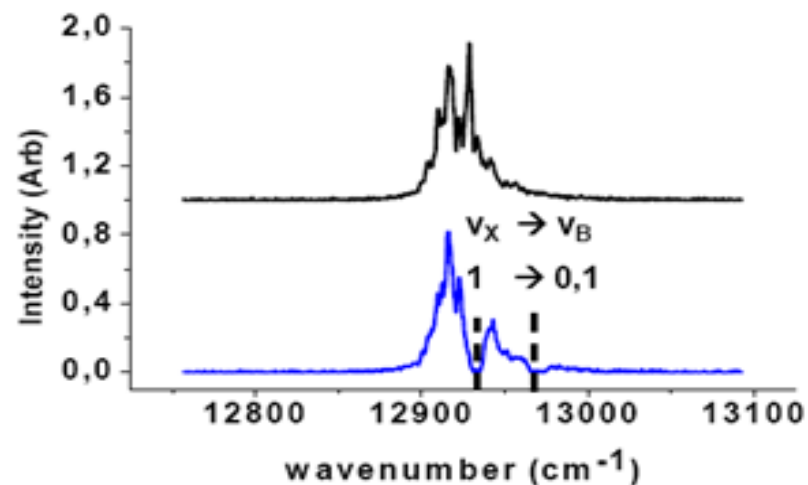


# VIBR-COOLING WITH INCOHERENT LIGHT

femtosecond laser  $\rightarrow$  broadband diode



Mechanical mask for  $v=1$   
with extinction ratio 100%





# OUTLINE

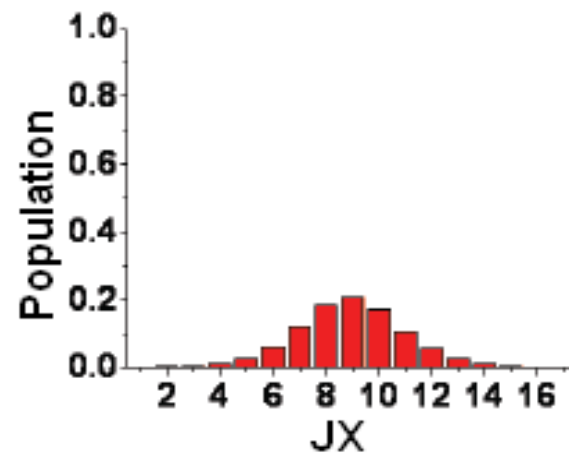
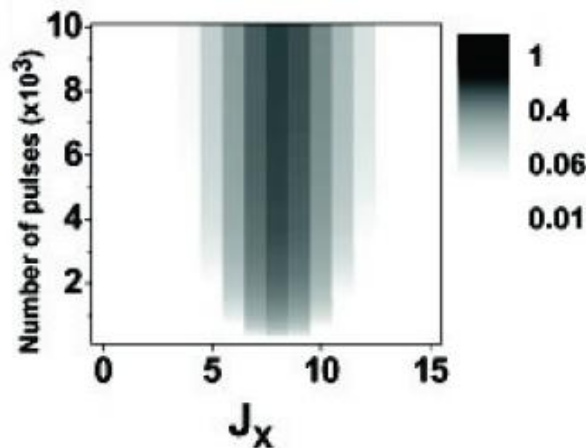
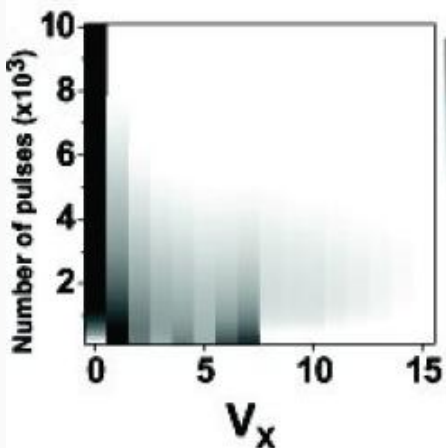
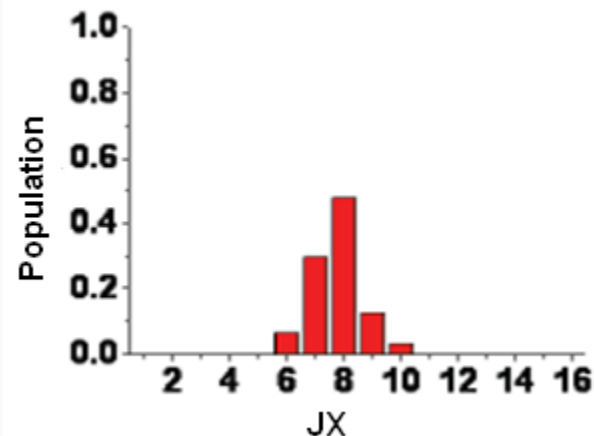
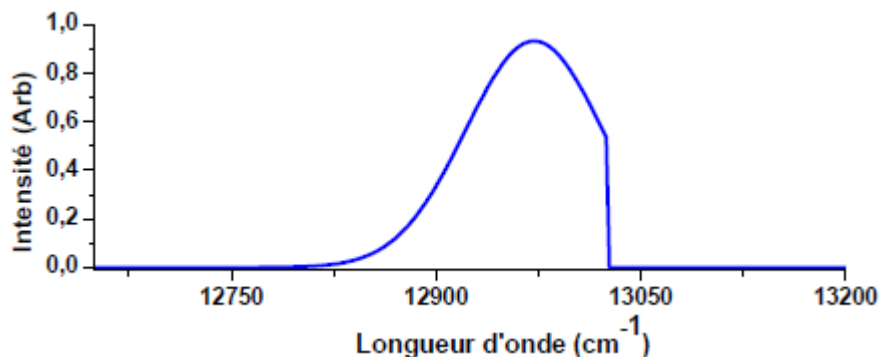
- Introduction. Why cold molecules?
- Introduction. Methods of production of cold molecules
- Optical pumping and vibrational cooling
- Optical pumping into a preselected level
- **Rotational cooling**
- Conclusions and perspectives



# ROTATIONAL COOLING 1: simulation

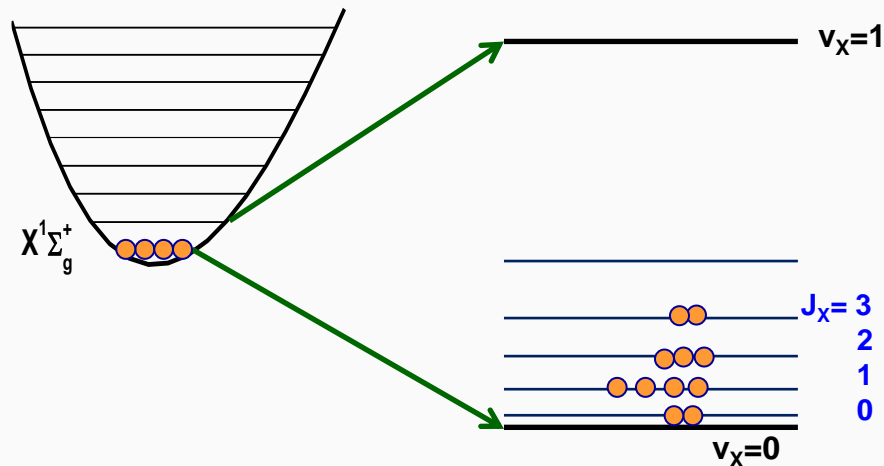
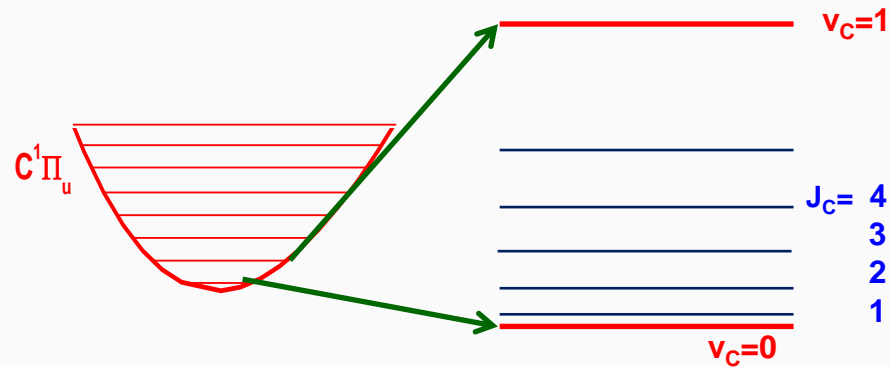
➤ Rotational distribution:

- after photoassociation
- after vibrational cooling





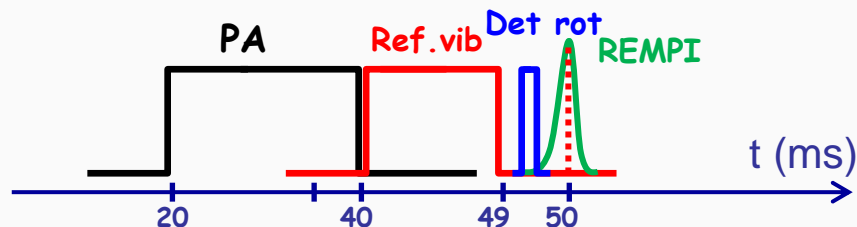
## ROTATIONAL COOLING 2: detection



- for each vibrational level, several rotational levels are populated
- In  $Cs_2$  (X state), the rotational separation ( $\sim 600$  MHz) is less than the detection laser linewidth used for the REMPI ( $> 5$  GHz)
- A narrow bandwidth laser is required for detection

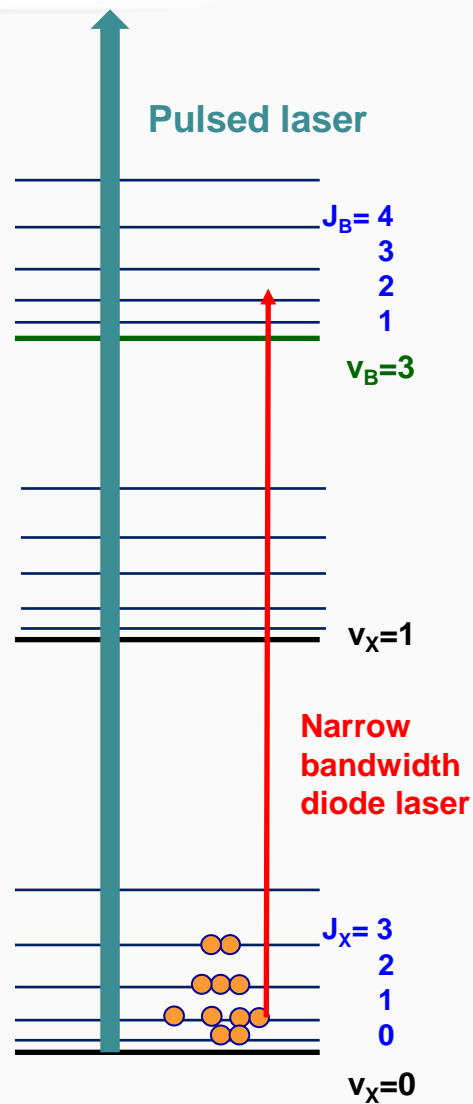
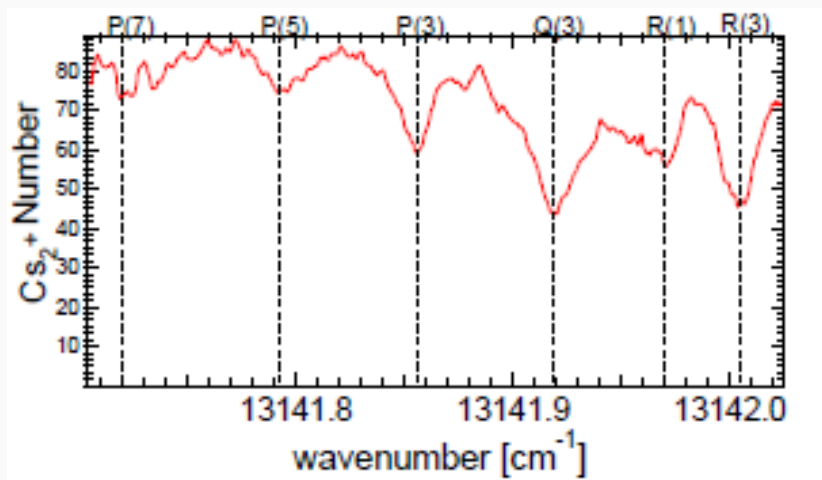


# ROTATIONAL COOLING 2: detection



Two possibilities:

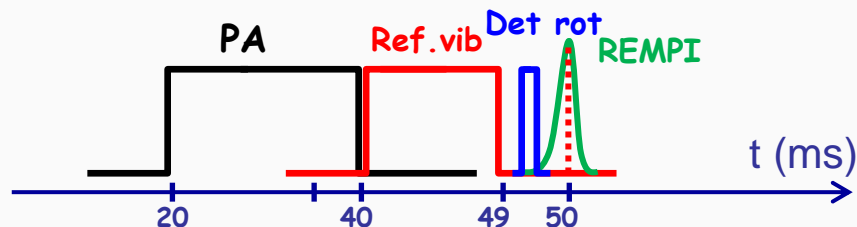
- 1) Depletion spectroscopy
- 2) Spontaneous-decay-induced double resonance







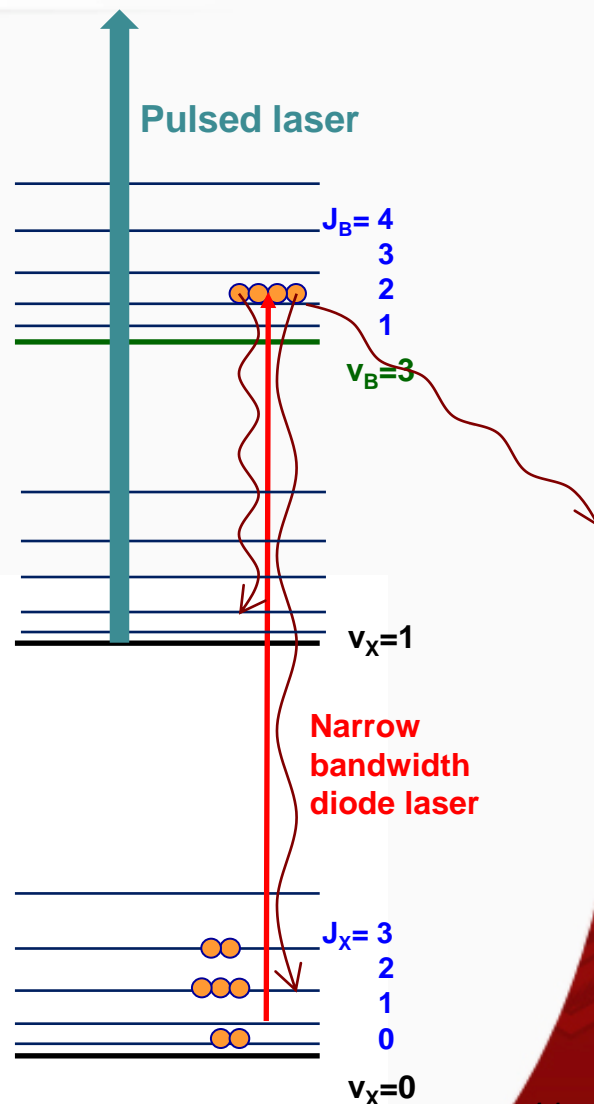
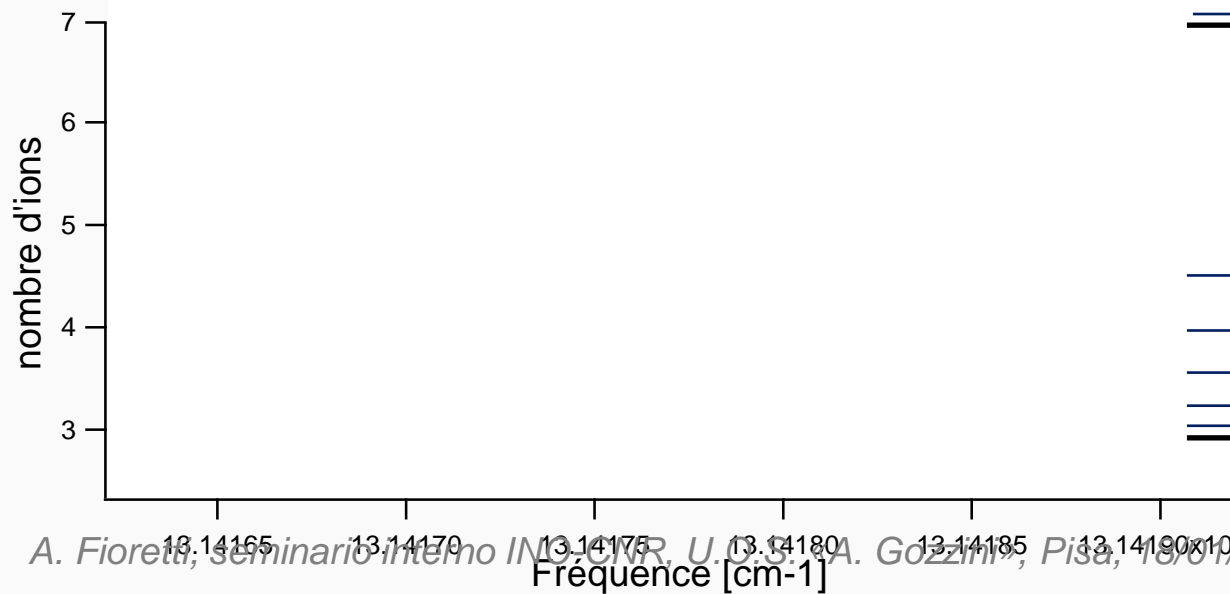
# ROTATIONAL COOLING 2: detection



Two possibilities:

1) Depletion spectroscopy

2) Spontaneous-decay-induced double resonance





## ROTATIONAL COOLING 3: cooling

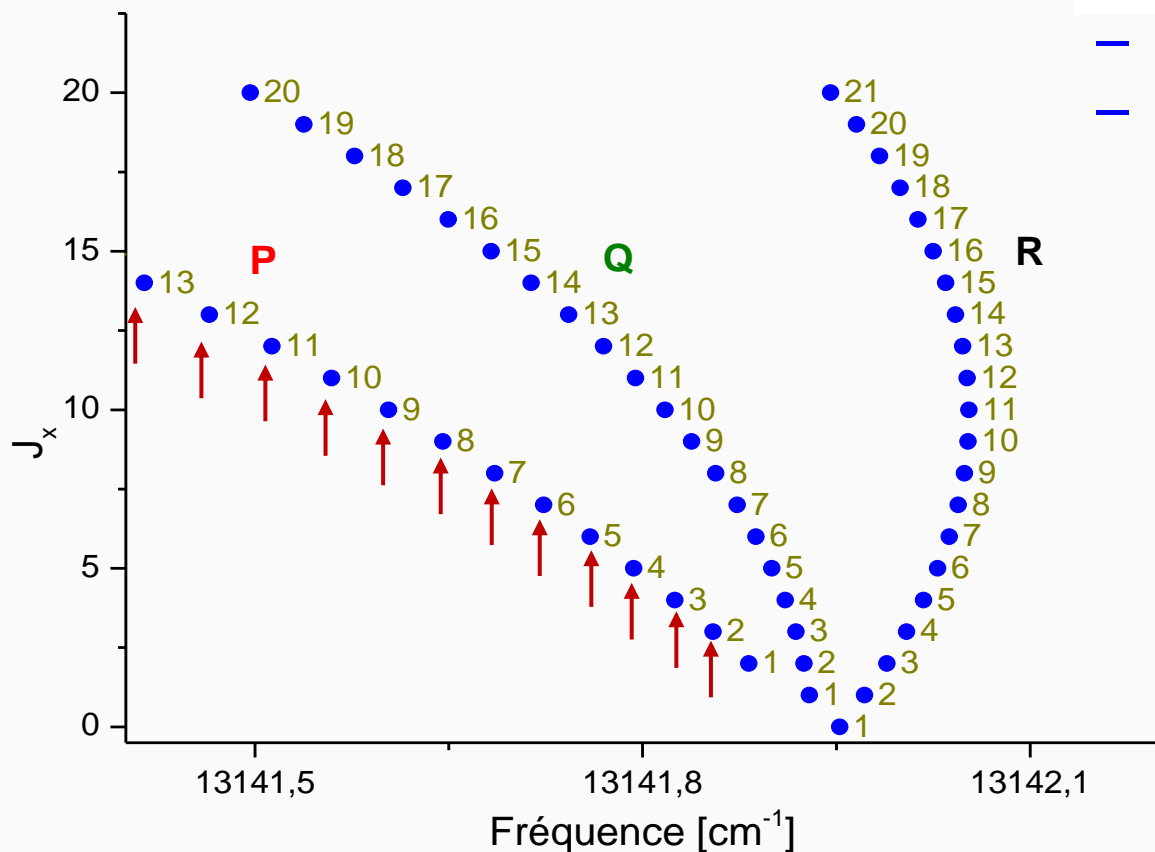
- Selection rules:  $\Delta J=0, \pm 1$  and parity

- **P** : J lowering transitions
- ~~Q : J constant transitions~~
- ~~R : J raising transitions~~

The cesium rotational structure is too narrow for shaping a broadband laser with a grating

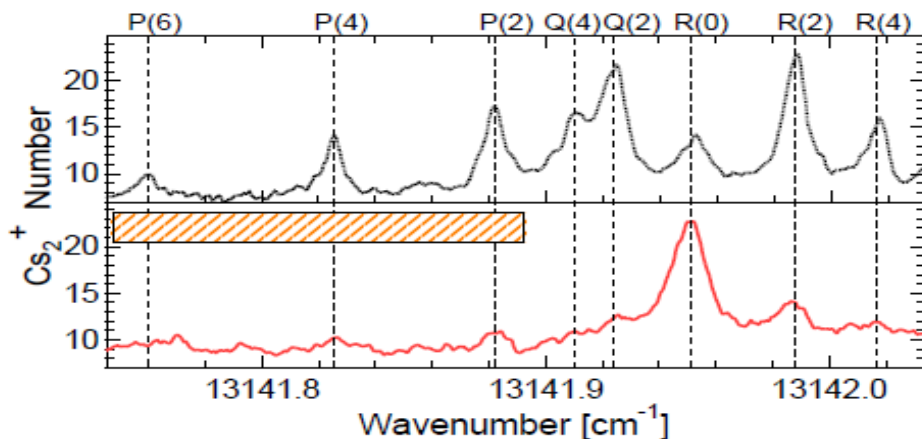
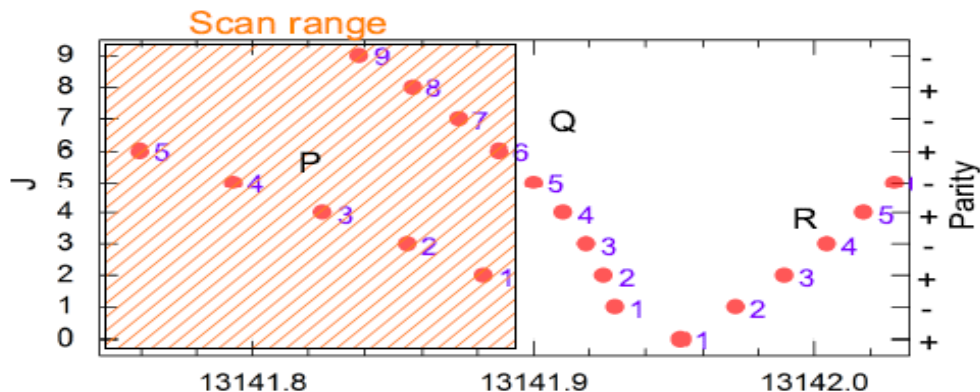
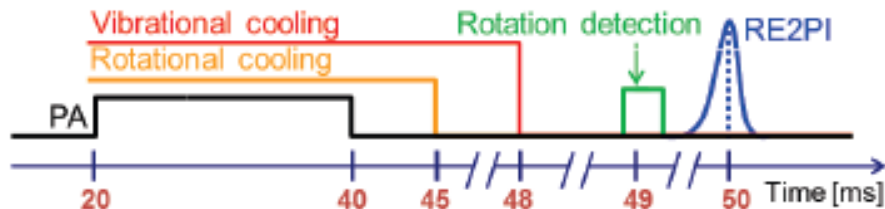
Any rotational pumping will affect also the vibration

⇒ A narrow linewidth laser is required!





# ROTATIONAL COOLING 3: cooling in $v=0, J=0$ !



**V=0**  
**J=0**

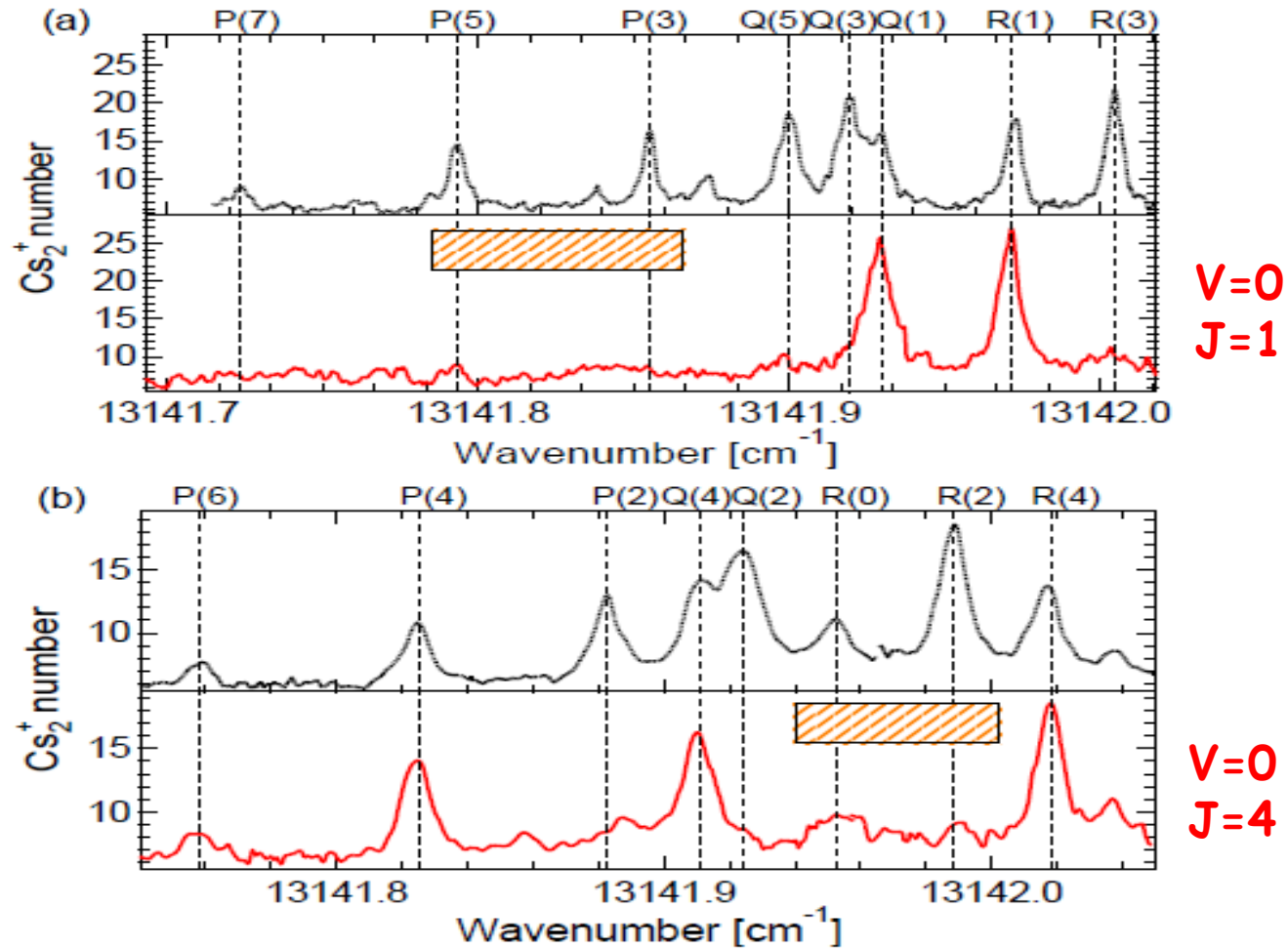
Rotational cooling obtained by the frequency scanning of a diode laser ( $\Delta t \approx 100\mu\text{s}$ )

Efficiency of rotational pumping  $\approx 40\%$

Optimization is possible



# ROTATIONAL COOLING 4: cooling in $v=0$ , $J \neq 0$ !





# OUTLINE

- Introduction. Why cold molecules?
- Introduction. Methods of production of cold molecules
- Optical pumping and vibrational cooling
- Optical pumping into a preselected level
- Rotational cooling
- **Conclusions and perspectives**



## CONCLUSIONS

- Optical manipulation of the internal degrees of freedom (**electronic**, **vibrational** and **rotational**) of the cesium dimer has been obtained.

- Laser cooling into the absolute ( $v=0$ ,  $J=0$ ) level as well as into other pre-selected levels have been obtained

Vibrational cooling: M. Viteau *et al.*, *Science* **321**, 232 (2008)

Vibr. cooling into a pre-selected level: D. Sofikitis *et al.*, *New Journal of Physics*, **11** (2009)

Vibr. cooling with laser diodes: D. Sofikitis *et al.*, *Phys. Rev. A* **80**, 051401, (2009)

State transfer: R. Horchani *et al.*, *Phys Rev. A* **85**, 030502 (2012)

Rotational cooling: I. Manai *et al.*, *Phys. Rev. Lett.* **109**, 183001 (2012)

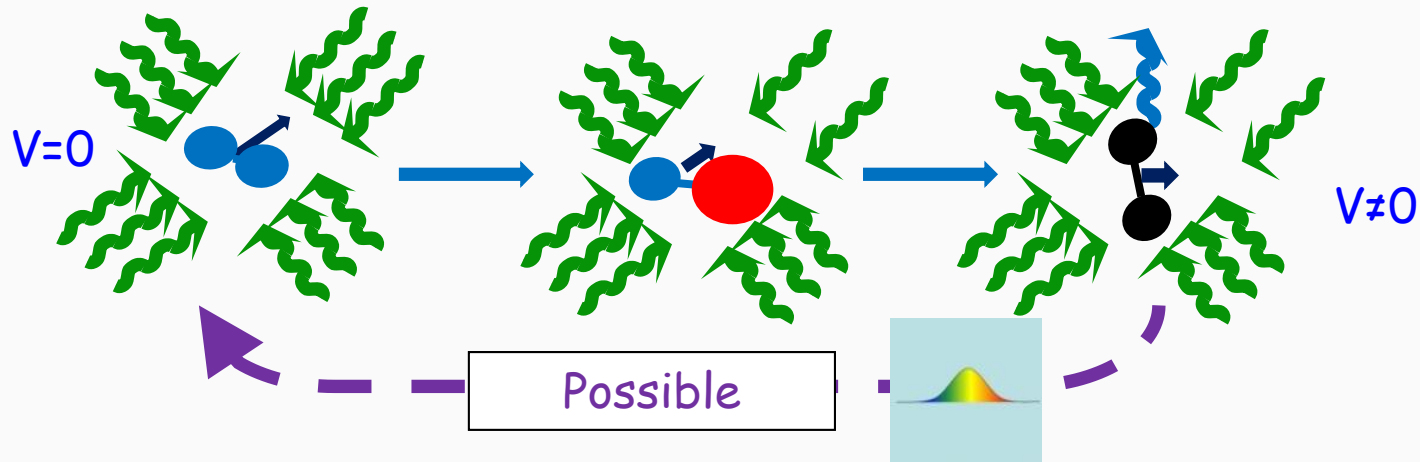
- Optical (incoherent) pumping offers an alternative approach to coherent transfer (STIRAP) towards the attainment complete control of external and internal degrees of freedom in simple molecules from laser cooled atoms

*An ultracold high-density sample of rovibronic ground-state molecules in an optical lattice*  
J. G. Danzl, M. J. Mark, E. Haller, M. Gustavsson, R. Hart, J. Aldegunde, J. M. Hutson, and H.-C. Nägerl, *Nature Physics* **6**, 265 (2010)



# PERSPECTIVES

- 1) This method can be extended to more general (and interesting) molecules:  
**laser sources** (diodes, supercontinuum) and **spectroscopic knowledge needed!**
  - Example: **NaCs** (N. Bigelow group, *Optics Express*, Vol. 20, No. 14, (2012))
  - **RbCs** possible (INO-PISA)
- 2) State distillation of molecular samples is possible
- 3) Direct laser cooling/trapping of molecules could be extended

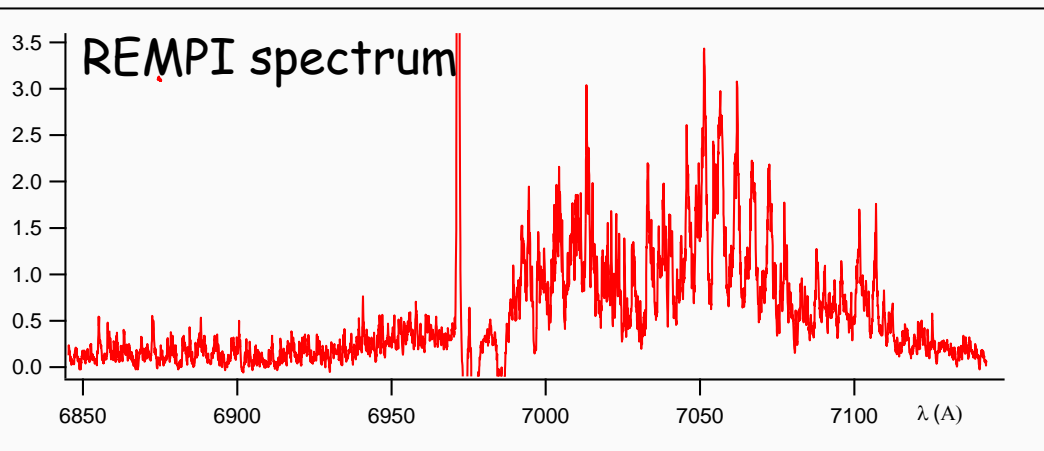
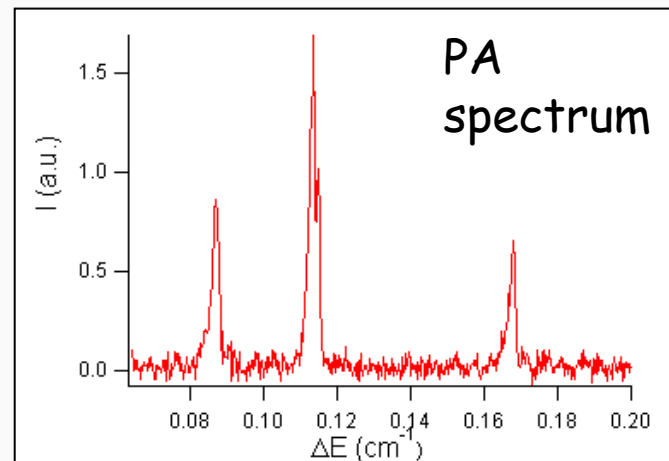
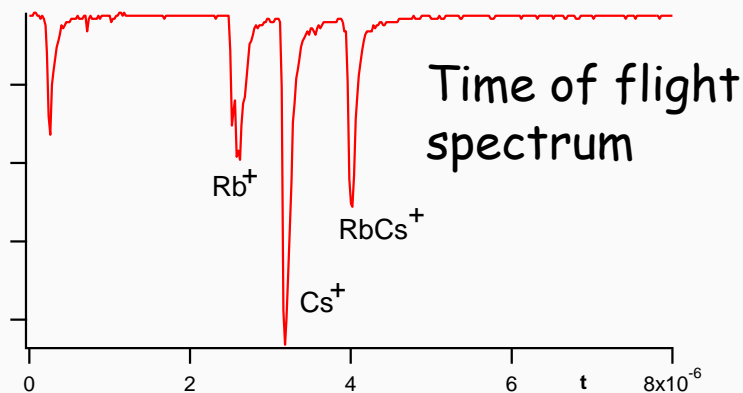




## PERSPECTIVES at INO-Pisa

RbCs double MOT in operation, PA and cold molecule production,  
PA spectroscopy under way, vibrational cooling should be possible.

More laser sources needed!



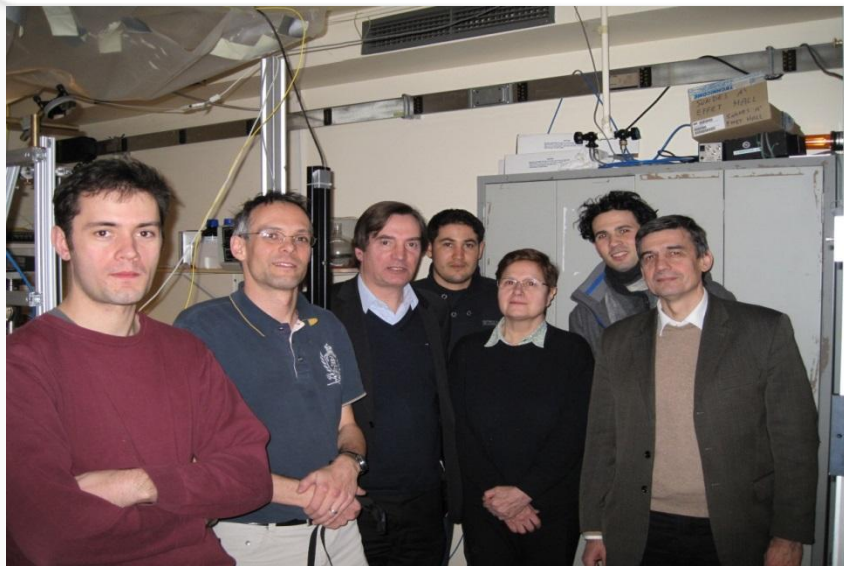
$$\Delta_{\text{PA}} = -8.1 \text{ cm}^{-1}$$

$$\lambda_{\text{dye}} \sim 700\text{-}712 \text{ nm}$$





## THE CREW



## Experiment LAC

- Isam Manai
- Ridha Horchani
- Hans Lignier
- Daniel Comparat
- Pierre Pillet
- + former PhD students

## Theory LAC

- Nadia Bouloufa
- Olivier Dulieu

## Visitors

- Marin Pichler
- Maria Allegrini
- Andrea Fioretti
- Goran Pichler
- Emiliya Dimova
- Lirong Wang
- + others

**Thank you for  
your attention !**

## Collaboration

- Béatrice Chatel
- Sébastien Weber  
(LCAM, Toulouse, France)

