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
OUTLINE

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

\[ \lambda_{DB} \approx \text{distance between particles} \]

Quantum properties
Bose-Einstein gases
Quantum information

Control of collisions
Quantum chemistry

\[ \lambda_{DB} = \frac{h}{mv} \propto T^{-\frac{1}{2}} \text{ (quantum size)} \approx \text{classical size of particles (1nm)} \]

Precision measurements
Fundamental tests

Small velocity
MOTIVATIONS FOR COLD MOLECULES

• Precision measurements and fundamental tests

Improved measurement of the shape of the electron
They use cold YbF molecules

• Quantum information with polar systems

Quantum computation with trapped polar molecules.
A coherent all-electrical interface between polar molecules and mesoscopic supercond. resonators.

• Quantum gases, many-body physics

A toolbox for lattice-spin models with polar molecules.
(PROPOSAL with candidate polar molecules: CaF, CaCl and MgCl.)

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

Low-energy collisions of NH₃ and ND₃ with ultracold Rb atoms.

• ...
- Introduction. Why cold molecules?

- Introduction. Methods of production of cold molecules

- Optical pumping and vibrational cooling

- Rotational cooling

- Conclusions and perspectives
Laser cooling of atoms: many absorption-spontaneous emission cycles

Laser cooling of molecules?

A. Fioretti, seminario interno INO-CNR, U.O.S. «A. Gozzini», Pisa, 18/01/2013
COLD MOLECULE FORMATION 1

from molecules

- Cryogenic method: Buffer gas cooling + magnetic trapping
  CaH, PbO, O$_2$, ND$_3$, ... (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

- Velocity filtering by molecule guiding
  H$_2$CO, CH$_3$F, H$_2$O, ... (G. Rempe 2004)

- Translational temperature ~ K-mK
- Vibrational and rotational temperatures ~ cold
COLD MOLECULE FORMATION 2

from pre-cooled atoms
(Ca2, K2, Rb2, RbCs, KRb, LiCs, NaCs...)

- Magneto-association
  (Feshbach resonance)

- 3-Body Collision

- Photo-association

- Temperature (speed) ~ atomic temperature ~ ultracold (nK-μK)
- Vibrational temperature ~ HOT
- Rotational temperature ~ cold but several levels occupied
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~ 5 \cdot 10^7 \text{ atomes}, n \sim 10^{11} \text{ at/cm}^3, T\sim 100\mu \text{K}
EXPERIMENT

Work horse: a cesium magneto-optical trap MOT)
N~ 5 \cdot 10^7 atomes, n \sim 10^{11} at/cm^3, T\sim100\mu K

- MOT: diode lasers, cw, 852nm, 150mW, frequency stabilized <1MHz
- PA: Ti:Saphire cw, 852 nm, \sim1W, linewidth \leq 1MHz
- Detection (ionisation): pulsed laser 10 Hz-7ns, 5-10mJ/pulse

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

A. Fioretti, seminario interno INO-CNR, U.O.S. «A. Gozzini», Pisa, 18/01/2013
Photoassociation

Resonant absorption of one photon by 2 colliding cold atoms (T~100μK)

Emission

Trap losses

Cold molecules

Internuclear distance (a.u.)

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

PA: FORMATION in the $X^1\Sigma_g^+$ state
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?
- Introduction. Methods of production of cold molecules
- Optical pumping and vibrational cooling
- Optical pumping into a preselected level
- Rotational cooling
- Conclusions and perspectives
Very efficient to transfer molecules already in a single level (i.e. Feshbach molecules): Cs$_2$, KRb, Rb$_2$

Not efficient to transfer molecules distributed over many levels

Traslationally cold but Vibrationally HOT

Our aim

Traslationally and Vibrationally cold
VIBRATIONAL COOLING 2: optical pumping

- Broadband laser (femtosecond laser: 200 cm\(^{-1}\))
  - Excites all levels towards the target state B
  - Excitation probability:
    \[ \Gamma_{v_X \rightarrow v_B} \propto FC[v_X][v_B] (D[v_X][v_B])^2 I_{laser}[v_X][v_B] \]

A. Fioretti, seminario interno INO-CNR, U.O.S. «A. Gozzini», Pisa, 18/01/2013
Emission spontanée → Redistribution des molécules dans l’état X

→ Probabilité de désexcitation :

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

VIBRATIONAL COOLING 2: optical pumping
VIBRATIONAL COOLING 2: optical pumping

Energy (cm$^{-1}$)

\begin{align*}
\text{B}^1\Pi_u & \quad \text{X}^1\Sigma_g^+ \quad \text{R} (\text{A}_0)
\end{align*}
VIBRATIONAL COOLING 2: optical pumping

- Shaped broadband laser:
  no transition from \(v=0\)
  → dark state
Shaped broadband laser: no transition from \(v=0\) → dark state
VIBRATIONAL COOLING 2: optical pumping

A. Fioretti, seminario interno INO-CNR, U.O.S. «A. Gozzini», Pisa, 18/01/2013
Before VIBR-Cooling

- Detection spectrum (via the C state)
  - Molecules distributed over many vibrational levels

Detection spectrum (via the C state)

- Molecules distributed over many vibrational levels

A. Fioretti, seminario interno INO-CNR, U.O.S. «A. Gozzini», Pisa, 18/01/2013
AFTER VIBR-COOLING

- With an optical pumping phase

Molecule pile up in the DARK state \( v=0 \)

\[ \rightarrow \text{Efficiency } \sim 80 \% \]

\[ \rightarrow \text{limited also by spectral bandwidth of the laser} \]
- 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
How to accumulate molecules into another pre-selected level?

- suppress ALL laser frequencies connecting the target level (example v=1) to excited states.
How to accumulate molecules into another pre-selected level?

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

$\nu_X - \nu_C = 1-0$

Fréquence de détection (cm$^{-1}$)

Intensité (Arb)

Fréquence (cm$^{-1}$)

Simulation

Expérience

A. Fioretti, seminario interno INO-CNR, U.O.S. «A. Gozzini», Pisa, 18/01/2013
Pumping into a PRE-SELECTED LEVEL

Efficiency ~ 50 % limited by:
- Laser bandwidth
- SLM extinction ratio ~ 97%

SLM resolution ~ 0.06nm = 0.8cm⁻¹ @852nm
Vibrational spacing ~ 40cm⁻¹

A. Fioretti, seminario interno INO-CNR, U.O.S. «A. Gozzini», Pisa, 18/01/2013
VIBR-COOLING WITH INCOHERENT LIGHT

femtosecond laser → broadband diode

Grating

Mirror

Femtosecond Laser
Diode
Without pumping

Fréquence de détection (cm⁻¹)

15900 15920 15940 15960 15980 16000

80 60 40 20 0
VIBR-COOING WITH INCOHERENT LIGHT

femtosecond laser → broadband diode

Fréquence de détection (cm⁻¹)

Intensity (Arb)

A. Fioretti, seminario interno INO-CNR, U.O.S. «A. Gozzini», Pisa, 18/01/2013
VIBR-COOLING WITH INCOHERENT LIGHT

femtosecond laser → broadband diode

Mehanical mask for v=1 with extiction 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 distribution:
- after photoassociation
- after vibrational cooling
• for each vibrational level, several rotational levels are populated

• In Cs₂ (X state), the rotational separation (~ 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

Pulsed laser

P(9)
P(5)
P(6)
P(7)
P(8)
Q(9)
Q(7)
Q(8)
Q(6)
Q(10)

Fréquence [cm⁻¹]

PA
Ref. vib
Det rot
REMPI

20 40 49 50

J
B
= 4
3
2
1

v
B
= 3

v
X
= 1

Narrow bandwidth diode laser

J
X
= 3
2
1
0

v
X
= 0
Selection rules: $\Delta J=0$, ± 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 obtained by the frequency scanning of a diode laser ($\Delta t \approx 100\mu s$)

Efficiency of rotational pumping $\approx 40\%$

Optimization is possible

$V=0$

$J=0$
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
  Vibr. cooling into a pre-selected level: D. Sofikitis et al., New Journal of Physics, 11 (2009)

- 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
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!

![Time of flight spectrum](image1)

![REMPE spectrum](image2)

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

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