Engineering and Control of Cold Molecules

N. P. Bigelow
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Why Cold (Polar) Molecules?
Why Cold (Polar) Molecules?

Talks from:
Shlyapnikov
Pfau
Burnett
Sengstock
Why Cold (Polar) Molecules?

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Shlyapnikov
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Sengstock

More Reasons Later in Talk
So what about cold molecules?

Hard to find cycling transitions

⇒ Laser cooling of molecules is hard if not impossible
Doyle approach - Thermalization with cold helium gas & magnetic trapping

Dilution Refrigerator

~mK
Paramagnetic molecules
Large samples
Meijer - time and spatially varying E fields
– decelerator –
(à la high-energy in reverse; H. Gould)
Meijer - decelerator
(à la high-energy; H. Gould)

decelerate
Here

“Build” the molecules from a gas of pre-cooled atoms via photoassociation
A Quick Review of Photoassociation
Starting with Homonuclears
Molecular potentials

Homonuclear

Singly excited state

Ground state
Photoassociation

- Singly excited state
- Ground state
- Photon
- 600-1000 Å
Ground state molecule formation?

Sp. emission

Excited state

photon

Ground state
These factors determine molecular production rates.
Ground state molecule formation

Poor Franck-Condon factors are discouraging

\[ \langle f | i \rangle \Rightarrow 0 \]
In 1997 Pierre Pillet and co-workers startled the community by showing the Cs$_2$ ground-state molecules were produced in a conventional optical trap.
Formation of Cold Cs$_2$ Molecules through Photoassociation

A. Fioretti, D. Comparat, A. Crubellier, O. Dulieu, F. Masnou-Seeuws, and P. Pillet

Laboratoire Aimé Cotton, CNRS II, Bâtiment 503, Campus d’Orsay, 91405 Orsay Cedex, France

(Received 10 November 1997)

We have performed photoassociative spectroscopy of cold Cs atoms in a vapor-cell magneto-optical trap, within 10 cm$^{-1}$ below the dissociation limit 6$^2$S$_{1/2}$ + 6$^2$P$_{3/2}$. Our detection method is based on pulsed-laser photoionization of Cs$_2$ molecules, selectively detected through a time-of-flight mass spectrometer. Temporal and spatial analysis of Cs$_2^+$ ions show for the first time the formation of translationally cold Cs$_2$ triplet ground state molecules, at a temperature $T \sim 300 \mu$K.

[S0031-9007(98)06137-7]
Observation of Ultracold Ground-State Potassium Molecules

University of Connecticut, Department of Physics, U-46, Storrs, Connecticut 06269
(Received 7 October 1998)

Ultracold potassium molecules are produced in the $X^1\Sigma_g^+$ electronic ground state by photoassociation in a magneto-optical trap. They are detected sensitively and selectively by use of two-color resonant ionization, followed by time-of-flight mass spectroscopy. We observe deeply bound molecules with $\nu \approx 36$, at a temperature of about 300 $\mu$K. Presently about 1000 molecules/sec are produced, and much higher production rates should be attainable in the future. [S0031-9007(08)08237-4]
Efficient Production of Ground-State Potassium Molecules at Sub-mK Temperatures by Two-Step Photoassociation


University of Connecticut, Department of Physics, U-3046, Storrs, Connecticut 06269
(Received 15 July 1999)

We have developed a two-step "R-transfer" method that efficiently produces translationally ultracold potassium molecules in the $X^1\Sigma_g^+$ electronic ground state. Laser-cooled atoms are initially photo-associated at large internuclear separation $R$ to form molecules in high vibrational levels of the $1^1\Pi_u$ state, which are in turn excited by an additional laser to shorter-range Rydberg states such as $5^1\Pi_u$ and $6^1\Pi_u$. Subsequent radiative decay produces ground-state molecules at rates up to $10^5$ molecules/second per vibrational level.

R-transfer method
Frank-Condon Matching
Cold Rubidium Molecules Formed in a Magneto-Optical Trap

C. Gabbanini, A. Fioretti, A. Lucchesini, S. Gozzini, and M. Mazzoni

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4Unidad IFIM, Sez. A, Dipartimento di Fisica, Università di Firenze, Largo E. Fermi 2, 50125 Firenze, Italy
(Received 22 September 1999)

FIG. 1. Ion spectra for a Rb MOT as a function of the dye laser wavelength $\lambda_d$. The boxcar integrator is gated at the molecular (a, b) and at the atomic (c) times of flight, respectively. In case (b) a PA laser, red detuned by $\sim$10 GHz from the $F_a = 2 \rightarrow F_a = 3$ $^8$Rb resonance, is present. Peaks in (c) are due to atomic two-photon resonances to Rydberg states ($18D \rightarrow 13D$, from left to right); the peak labeled with (*) corresponds to the $5p_{3/2} \rightarrow 5F^+$ transition.
Observation of Optically Trapped Cold Cesium Molecules

T. Takekoshi, B. M. Patterson, and R. J. Knize
Laser and Optics Research Center, Department of Physics, United States Air Force Academy, Colorado 80840
(Received 30 June 1998)

We report the first observation of optically trapped cold neutral molecules. Cesium dimers in the electronic ground state are produced directly in a magneto-optical trap and transferred to a dipole trap formed at the focus of a CO$_2$ laser beam ($\lambda = 10.6 \mu$m). These neutral molecules were detected using photoionization and time-of-flight spectroscopy. Initial experiments indicate a cold molecule trap lifetime on the order of half a second. [S0031-9007(98)07817-X]
Why not heteronuclear molecules?
Molecular potentials

Far better Franck-Condon factors....

Singly excited state

Ground state
\[ \text{Energy (cm}^{-1}) \]

\[ \text{Na}(3^2P_{1/2}) + \text{Cs}(6^2S_{1/2}) \]

\[ \text{Na}(3^2S_{1/2}) + \text{Cs}(6^2S_{1/2}) \]

Internuclear Separation (Angstroms)
We might be CLOBBERED by the pair distribution function

\[ \frac{R_{\text{homo}}}{R_{\text{hetero}}} = \frac{950}{76} \sim 13 \]

\[ \left( \frac{R_{\text{homo}}}{R_{\text{hetero}}} = \frac{950}{76} \sim 1/10 \right)^2 \sim 100 ! \]

The interaction time is also smaller by a factor of \( \sim 13 \)!
Nevertheless, several groups built multi-species MOTs - including Rochester and Sao Carlos and
Kasevich  
Arimondo  
Ingusio  
Weidemuller  
Windholz  
DeMille  
Stwalley/Gould  
Wang/Buell  
Ruff  
*others*

\[ \begin{align*} 
\text{NaCs} & \\
\text{NaK} & \\
\text{NaRb} & \\
\text{RbCs} & \\
\text{KRb} & \\
\text{LiCs} & \\
\text{LiNa} & \\
? & \\
\end{align*} \]

Most “reactive”

Bose-Fermi&Isotopic: Jin, Salamon, Ertmer, Ketterle, Hulet, Sengstock…

Sukenik
We started with Na + Cs
Search for ground state molecules - the detection pathway

- Ionisation
- NaCs$^+$
- Na(3p)+Cs(6s)
- Na(3s)+Cs(6p)
- Na(3s)+Cs(6s)
- PA

20-200 µJ 9ns 580-589 nm 2 mm$^2$
NaCs formation and detection

100 µJ pulse @ 575-600 nm

Traps

Time (µs): -30 0 20 10^5
Choice of pulsed laser wavelength

575-600 nm was chosen

• To implement a resonant ionization pathway

• It has enough energy to 2-photon ionize NaCs

• It will NOT resonantly ionize Cs
Time of Flight

- Photons
- Cold Na
- Cold Cs
- Thermal Na
- Na$_2$
- Cold NaCs
- Cs$_2$
Is NaCs formed by the ionizing pulse?
Push–beam experiment

![Mass spectrum showing ion counts with and without Na-pushbeam](image)
How cold are the molecules?

We use time-of-flight from the interaction zone as our probe.
NaCs Temp

\[ T_{\text{NaCs}} \approx 260 \, \mu K \]

0.2 m/s
(kinetic temp)
**$K_{NaCs}$ : The rate of formation**

- We detect 500 molecules in 10,000 pulses
- Estimate ionization and detection efficiency $\sim$ 10%
- Kinetic model: 90% of NaCs drifts out of the trapping region between pulses.

$\Rightarrow$ Production rate $\sim$ 50 Hz

- $d [NaCs] / dt = K_{NaCs} [Na][Cs]$

$\Rightarrow$ $K_{NaCs} \sim 7.4 \times 10^{-15} \text{ cm}^3 \text{ s}^{-1}$
Measured depth of the triplet is 137-247 cm\(^{-1}\)

We have scanned 0 – 268 cm\(^{-1}\) and found strong ion signals
(includes 5% of singlet potential)

We have a mixture of singlets and triplets

C. Haimberger, J. Kleinert, npb PRA Rapid Comm.
70, 021402 (R) Aug 2004
Coherent Transfer - State Selective

Ro-vibrational ground state (lower dipole moment)  
or  
Single excited ro-vibrational state

Incoherent transfer results already at Yale
The most important part of this part of our work:

C. Haimberger
J. Kleinert

(M. Bhattacharya, J. Shaffer & W. Chalupczak)
Observation of Ultracold Ground-State Heteronuclear Molecules


Instituto de Física de São Carlos, Universidade de São Paulo, Caixa Postal 369, 13560-970, São Carlos-SP Brazil

(Received 13 August 2003; published 2 April 2004)

We report the observation of translationally ultracold heteronuclear ground-state molecules in a two-species magneto-optical trap containing $^{39}$K and $^{85}$Rb atoms. The KRb molecules are produced via photoassociation and detected by multiphoton ionization. We had characterized their temperature and measured their formation rate constant. We believe that the two-species trap could be used as a reliable source of ultracold molecules to be captured by electrostatic, magnetic, or optical traps. This possibility will certainly motivate further investigation of quantum collective effects as well as high-resolution spectroscopy of the rovibrational level structure of cold heteronuclear molecular systems.

DOI: 10.1103/PhysRevLett.92.135203
PACS numbers: 34.50.Rk, 34.10.+x, 32.80.Pj
Production and State-Selective Detection of Ultracold RbCs Molecules

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(Received 25 February 2004, published 12 April 2004)

Using resonance-enhanced two-photon ionization, we detect ultracold, metastable RbCs molecules formed in their lowest triplet state \( a^3\Sigma^+ \) via photoassociation in a laser-cooled mixture of \(^{85}\)Rb and \(^{133}\)Cs atoms. We obtain extensive bond-bound excitation spectra of these molecules, which provide detailed information about their vibrational distribution, as well as spectroscopic data on several RbCs molecular states including \( a^3\Sigma^+, (2)^3\Sigma^+, \) and \( (1)^3\Pi \). Analysis of this data allows us to predict strong transitions from observed levels to the absolute vibronic ground state of RbCs, potentially allowing the production of stable, ultracold polar molecules at rates in excess of \( 10^6 \) s\(^{-1}\).

RbCs
Photoassociative Production and Trapping of Ultracold KRb Molecules

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(Received 16 July 2004; published 7 December 2004)

KRb

40,000 molecules/sec !!
Franck-Condon Factors for hetero-pairs based on long range (dispersion) potentials

Cold (Polar) Molecules and Quantum Information
Polar Molecules Allow Coupling
Enabling Quantum Information Applications

- Heteronuclear molecules are highly polarizable
- The dipole-dipole coupling of the molecules can be harnessed for quantum information applications (see protocols for EDMs in Quantum Dots - e.g. Barenco, *et al* PRL 82 1060)
Polar Molecules Allow Couplings Enabling Quantum Information Applications (based on scheme by Côté, Yellin [Lukin])
Polar Molecules Allow Coupling
Enabling Quantum Information Applications

\[ \begin{align*}
\text{Internuclear Separation (Å)} & \quad \text{Energy (10}^3 \text{ cm}^{-1}) \\
0 & \quad -6 \quad -5 \quad -4 \quad -3 \quad -2 \quad -1 \quad 0 \quad 1
\end{align*} \]

\[ \begin{align*}
\text{atoms} & \quad \text{bound} \quad \text{molecular} \quad \text{states}\n\end{align*} \]

- Start with two molecules in the same state
  \[ |A_i\rangle = |0\rangle \quad |B_i\rangle = |0\rangle \]

\[ \begin{align*}
|v'', j''\rangle & \equiv |v\rangle \\
|v', j'\rangle & \equiv |1\rangle \\
|v, j\rangle & \equiv |0\rangle
\end{align*} \]
Polar Molecules Allow Coupling
Enabling Quantum Information Applications

• Start with two molecules in same state

\[ |A_i\rangle = |0\rangle \quad |B_i\rangle = |0\rangle \]

• Use Raman pulses - create superposition

\[ |A\rangle \equiv a_0 |0\rangle + a_1 |1\rangle \quad |B\rangle \equiv b_0 |0\rangle + b_1 |1\rangle \]

\[ |v'', j''\rangle \equiv |v\rangle \]

\[ |v', j'\rangle \equiv |1\rangle \]

\[ |v, j\rangle \equiv |0\rangle \]
Polar Molecules Allow Coupling
Enabling Quantum Information Applications

Two-molecule state:

\[ |AB\rangle \equiv c_{00}|00\rangle + c_{11}|11\rangle + c_{01}|01\rangle + c_{10}|10\rangle \]
Polar Molecules Allow Coupling
Enabling Quantum Information Applications

Now excite $|1\rangle$ to the state $|\nu\rangle$

$|\nu^{\prime},j^{\prime}\rangle \equiv |1\rangle$

$|\nu^{\prime\prime},j^{\prime\prime}\rangle \equiv |\nu\rangle$

(large dipole moment)

$|\nu,j\rangle \equiv |0\rangle$
Polar Molecules Allow Coupling
Enabling Quantum Information Applications

Now excite $\vert 1 \rangle$ to the state $\vert \nu \rangle$, wait an
Interaction time $\tau$ then de-excite back to $\vert 1 \rangle$

$\vert AB \rangle \equiv c_{00} \vert 00 \rangle + e^{i\phi} c_{11} \vert 11 \rangle + c_{01} \vert 01 \rangle + c_{10} \vert 10 \rangle$

Phase gate, CNOT gates etc. possible

$\vert \nu'', j'' \rangle \equiv \vert \nu \rangle$

(large dipole moment)

$\vert \nu', j' \rangle \equiv \vert 1 \rangle$

$\vert v, j \rangle \equiv \vert 0 \rangle$

“Dipole blockade and quantum information processing using mesoscopic
Atomic ensembles”, Lukin, Fleischhauer, Côté, Duan, Jaksch, Cirac and Zoller
PRL 87 037901
A specific qubit platform

Each atom sees a local field that couples it to its neighbors and permits selective addressing

\[ \vec{E}_a = \vec{E}_{\text{ext}}(x_a) + \vec{E}_{\text{int}}(x_a) \]

The coupling cannot be switched off but instead can be compensated using echo-type refocussing techniques of NMR.

What is needed is a source of cold molecules

D. DeMille, PRL 88, 067901
Molecules and “Chips”
Atom Chip

\[ \sigma^+ - \sigma^- \text{ polarization} \]

\[ I_s \sim 1-3 \, \text{A} \]

\[ I_B \sim 15-20 \, \text{A} \]

\[ B = B_0 \]

\[ \mathbf{Y} \]

\[ \mathbf{X} \]

\[ \mathbf{Z} \]

\[ \text{SiO}_2(250\text{nm}) \]

\[ \text{Ag}(50\text{nm}) \]

\[ \text{Cu}(1\text{mm}) \]

\[ \text{Si}(1\text{mm}) \]

\[ Z_0 \]
Our 1\textsuperscript{st} Generation Atom Chips

Cross-section:
\[ h = 1 - 5 \, \mu m \]
\[ w = 250 \, \mu m \]

Current density:
\[ J \approx 10^6 \, A/cm^2 \]
Cold Atomic Mixtures-on-a-chip

Holmes, Tscherneck, Quinto-su and Bigelow
2nd Generation Chips


- Collaboration with Corning, Inc.
- <100 atom sensitivity
3rd Generation Chips: Molecule Chips

Creating, detecting and locating ultracold molecules in a surface trap
Department of Physics and Astronomy, and The Laboratory for Laser Energetics, The University of Rochester, Rochester, NY 14627
WHY OPTICS and MOLECULES ON THE CHIP?
WHY OPTICS and MOLECULES ON THE CHIP?
4th Generation Chips

Higher current Capability
Heroines and Heroes
of this part of the work:

Michaela Tschernebeck
Michael Holmes
Pedro Quinto-Su
C. Haimberger
J. Kleinert
Vortex Lattice in a Coupled Atom-Molecule Condensate (non-polar molecules)
Single component atomic BEC - Triangular vortex lattice

\[ n_v = \frac{\Omega M}{\pi \hbar} \]

\( \Omega \) is rotation frequency
**Atom-Molecule BEC (homo-nuclear)**

**Mean-field model**

- Three scattering constants $g_a$, $g_m$ and $g_{am}$
- Coherent constant $\chi$
- Two Gross-Pitaevskii equations

\[
egin{align*}
    i\hbar \frac{\partial}{\partial t} \psi_a &= -\frac{1}{2M_a} \nabla^2 \psi_a + \frac{1}{2} M_a \omega^2 r^2 \psi_a + g_{aa} |\psi_a|^2 \psi_a \\
    &\quad + g_{am} |\psi_m|^2 \psi_a + \chi \psi_a^* \psi_m, \\
    i\hbar \frac{\partial}{\partial t} \psi_m &= -\frac{1}{2M_m} \nabla^2 \psi_m + \frac{1}{2} M_m \omega^2 r^2 \psi_m + g_{mm} |\psi_m|^2 \psi_m \\
    &\quad + g_{am} |\psi_a|^2 \psi_m + \frac{1}{2} \chi \psi_a^2. \quad (1)
\end{align*}
\]
Atom-Molecule BEC – Energies

Two key types of interaction between atoms and molecules

- Coherent coupling

\[
E_C = \frac{1}{2} \chi \int \text{d}r \: \psi_a^* \psi_a^* \psi_m + \text{c.c.}
\]

\[
= \chi \int \text{d}r \: |\psi_a|^2 |\psi_m| \cos[\theta_m(\vec{r}) - 2\theta_a(\vec{r})]
\]

- Interspecies scattering

\[
E_S = g_{am} \int \text{d}r \: |\psi_a|^2 |\psi_m|^2
\]
Atom-Molecule BEC - rotating

\[ n_v = \frac{\Omega M}{\pi \hbar} \quad \quad M_m = 2M_a \]

\[(n_v)_m = 2(n_v)_a\]

Vortex density of molecular condensate is twice that of the atomic condensate
Vortex Lattice in a Coupled Atom-Molecule Condensate (non-polar molecules)

Sungjong Woo
Department of Physics & Astronomy
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Q-Han Park
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Korea University
Summary

We can create cold diatomic polar molecules

We can create them and manipulate them on “chips”

There are interesting new aspects of degenerate systems involving molecules
Summary

We can create cold diatomic polar molecules

We can create them and manipulate them on “chips”

There are interesting new aspects of degenerate systems involving molecules

Can you calculate your Archimedes factor?