Stable cold polar molecules

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JILA, NIST and University of Colorado

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$ Funding $

DOE, NIST, NSF
Why ultracold molecules?


Electric dipole moments: Orientation is a big deal!


Manifested at or below μK temperatures
Ultracold molecules: quantum physics

- **Quantum information**
  (strong dipolar interactions, long coherence time)

- **Quantum degeneracy (e.g. BEC)**
  (anisotropic interactions)

- **Dipolar phase transition**
  (Condensed matter system)

Ultracold molecules: Test fundamental principles

- Ultrahigh resolution spectroscopy
- Standards in wide spectral ranges
- Molecular interferometry
- Precision measurement

One system, two different fundamental forces!

Excited electronic state

Ground electronic state

Electronic $\sim \alpha$

Vibration $\sim m_e/m_p$

(mass on a spring)

Strong interactions

QED

$e^- e^- e^-$
Ultracold molecules: Precision Chemistry

- Molecules in single quantum states, under precise control, for internal & external motions
- Unprecedented study of fundamentally important reactions (Dial the rates):
  - $\text{OH} + \text{HBr} \rightarrow \text{H}_2\text{O} + \text{Br}$
  - $\text{OH} + \text{H}_2\text{CO}$, $\text{CN} + \text{O}_2$, $\text{OH} + \text{NO}$, $\text{OH} + \text{OH}$, $\text{CN} + \text{NH}_3$, $\text{OH} + \text{H}$
- Higher reaction rate at lower temperature (10 K, importance for interstellar chemistry)

**Controlled molecular collisions**

**Ultracold chemical reactions**

- Molecules in single quantum states, under precise control, for internal & external motions
- Unprecedented study of fundamentally important reactions (Dial the rates):
  - $\text{OH} + \text{HBr}$, $\text{OH} + \text{H}_2\text{CO}$, $\text{CN} + \text{O}_2$, $\text{OH} + \text{NO}$, $\text{OH} + \text{OH}$, $\text{CN} + \text{NH}_3$, $\text{OH} + \text{H}$
- Higher reaction rate at lower temperature (10 K, importance for interstellar chemistry)
Ways to make cold polar molecules

- Start from ultracold atoms & pair them (Magneto-Photo-association)
- Start from ground-state molecules (Direct cooling of molecules)
Cool Alkaline Earth – Strontium


$T \sim 0.5$ photon recoil
$\sim 220$ nK

Boyle-free
Potentials matched to $< 10^{-15}$
Extreme spectral resolution

$^3P_1 \rightarrow ^1S_0$

461 nm (32 MHz)
689 nm (7.4 kHz)
Ultracold Sr$_2$ Molecules in Lattice

- Narrow lines
  - All bound states are resolved
- Favorable decay to electronic ground state
- Control of atomic collision with minimum loss

Raman transition for ground state production

Theory:
P. Julienne
S. Kotochigova

Polar molecules KRb in collaboration with D. Jin
Photo-Association via Coherent Accumulation of Pulses

Pe’er, Shapiro, Stowe, Shapiro, Ye, Phys. Rev. Lett. 98, 113004 (2007).

The problem – overlap

Molecular wave-packet dynamics bridge the overlap mismatch

Coherent accumulations resolve single quantum state
Making cold molecules with comb

Why pulse-train?
Two inherent time scales:
molecular vibration & atomic coherence

Time Domain picture

Pulse shaping:
• Positive chirp: “one way ticket”
• Robust to energy fluctuations
• Avoid ionization/dissociation

Coherent accumulation:
Weak field interaction
for strong field effects

Coherent control of single pulses – Coarse resolution (vibration).
Coherent accumulation of pulses – Fine resolution (rotation, hyperfine)
Stark deceleration

Direct manipulation of ground state molecules

Initial cooling important
(supersonic jets: single internal quantum state; external temp. ~ 1 K in a moving frame)

Phase space selection (~ 10 mK)

Applicable to a large variety of molecules

Stark Decelerator

Slower electrodes

Stark energy

Position

Stark energy

Position
Mathematical description of slowing


\[
\frac{d^2 \Delta \phi}{dt^2} + \frac{\pi W}{mL^2} \left[ \sin(\phi_0 + \Delta \phi) - \sin \phi_0 \right] = 0
\]

Equilibrium position

\[
\frac{d^2 \theta}{dt^2} - \left[ \frac{g}{L} \sin(\theta) - \frac{T}{mL^2} \right] = 0
\]

Change in kinetic energy

Pendulum driven by constant torque
$\phi_0 = 0^\circ$

OH LIF Signal [arb.u.]

Time from discharge [ms]

Experiment & Theory

data

simulations
Cold OH molecules


550 m/s to rest
1 K to 10 mK
$10^4 - 10^6$ molecules
Density: $10^5 - 10^7$ /cm$^3$
Cold molecule based precision spectroscopy

- Rabi or Ramsey interrogation
- High resolution and precision
- Systematic evaluations
Cold OH molecules to constrain $\dot{\alpha}$

$^2\Pi_{3/2}$

- $F' = 2$
- $F' = 1$

Hyperfine interactions $\sim \alpha^4$

 Lambda doubling $\sim \alpha^{0.4}$

OH megamasers

High redshift $z > 1$

Multiple transitions from the same gas cloud (Self check on systematics)

Precision measurement of OH ground structure

Measurement accuracy for all four lines: 4 – 10 Hz


• SUM (2 satellites) = SUM (2 main lines)

• Satellites calibrate B

• Observed satellites conjugate

\[ \Delta F = 0 \]

\[ \Delta F = \pm 1 \]

Kanekar et al., PRL 93, 051302 (2004).
Cold Chemical Reactions

Formaldehyde (H$_2$CO):
Bring this most general class of molecules to rest in laboratory

H$_2$CO - OH:
Fundamental reaction dynamics

Atmospheric chemistry
Combustion dynamics
Pollutant monitoring

H$_2$CO – near symmetric rotor:

$\mu_a = 2.34\text{D}$
OH + H₂CO reaction pathways

Barrier is likely negative, or very small (~ 60 K, within tuning range of Stark decelerator).

Decelerator allows probe of the reaction barrier at a resolution ≤ 10 K
External electric field tunes reaction barrier


External electric field adds:

$\Delta E \approx 1.8 \mu_{OH} |\bar{E}|$

Maximum experimentally attainable shift around 10 K

Control of cold chemical reactions; Unique dipolar interaction dynamics
A pressing requirement

Enhancement of the phase space density of cold molecules!
Sympathetic cooling in a trap

Molecules and atoms co-located in a trap.

Selectively remove highest energy Rb atoms.

Atoms and molecules rethermalize, through collisions, to a lower temperature.

Requires favorable collision rates

Jeremy Hutson’s talk (Friday)
Magnetic trapping of OH

Electric quadrupole

Magnetic trap coil

Imaging

Electrodes can be used or to apply uniform E field
OH Stark and Zeeman effects

\[ 2\Pi_{3/2} \]

\[ m_j = \frac{3}{2}, \frac{1}{2}, -\frac{1}{2}, -\frac{3}{2} \]

\[ f \]

\[ e \]

\[ \begin{align*}
&\text{Stark Shift [K]} \\
&\text{E-Field [kV/cm]} \\
&\text{B-Field [kG]} \\
\end{align*} \]

R. Krems' Talk Friday
Trapping Scheme

End view

Magnetic Quadrupole

Side view

0 A

20 m/s

2000 A
Trapping Scheme

End view

Side view

Magnetic Quadrupole

Uniform Electric Field

-1500 A

1500 A

z
Trap dynamics

\[ \text{LIF Signal [a.u.]} \]

\[ \text{Time from Decelerator Exit [ms]} \]

\[ \sim 30 \text{ mK, } 5 \times 10^3 \text{ cm}^{-3} \]
**OH - N₂ collisions**

**Collision-limited lifetimes**

- $\tau = 16 \pm 3$ ms
- $\tau = 98 \pm 5$ ms
- $1 \times 10^{-6}$ Torr N₂
- 20 ± 5 ms;
- $4 \times 10^{-8}$ Torr N₂
- 500 ± 100 ms
- **OH - N₂ cross section:**
  - $500 \pm 100$ Å²
Soon to study dipolar collisions!
Special thanks

http://jilawww.colorado.edu/YeLabs

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Collaborators

J. Bohn, D. Jin (JILA); E. Hudson, H. Lewandowski, J. Bocinski (former members)
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