Physics and Chemistry with Diatomic Molecules Near Absolute Zero

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What is Ultracold?

- Laser cooling of atoms
Beyond Cold Atoms

Indirect molecule cooling

Direct molecule cooling

buffer gas (sympathetic) cooling

laser cooling

bond formation

optical or magnetic
Why Cold Molecules?

atomic H spectrum

molecular H\textsubscript{2} spectrum

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Ultracold Diatomic Molecules

Indirect molecule cooling

$10^{-6}$ K

200 m/s $\rightarrow$ 10 mm/s

No energy release
Tight Trapping: Optical Lattice Clocks

10^{-6} K

create $\text{Sr}_2$ molecules

standing wave of light

quantized motional states

trapping potential: ac Stark shift
Molecular Lattice Clock

G. Reinaudi et al., PRL 109, 115303 (2012)
Science with Cold and Ultracold Molecules

- Ultracold chemistry
- Molecular clocks
- Table-top particle physics
Ultracold Chemistry

Quantum-state selected reactants and products

Bimolecular collisions

\[ AB + AB \rightarrow A_2 + B_2 \]

Photoassociation

\[ A + A + \gamma \rightarrow A_2^* \]

Photodissociation

\[ A_2 + \gamma \rightarrow A + A^* \]
Ultracold Chemistry

Quantum-state selected reactants and products

Complete quantum state control of “reverse collision”

Photodissociation

$A_2 + \gamma \rightarrow A + A^*$
Ultracold Chemistry

Quantum-state selected reactants and products

Photodissociation

\[ \text{Sr}_2 + \gamma \rightarrow \text{Sr} + \text{Sr}^* \]

The “hydrogen atom” of ultracold chemistry

• Experiment → first-principles theory → comparison
Ultracold Photodissociation

Photofragment angular distribution

\[ J = 0, J = 2 \] \[ (J = 4; M = \pm 1) \]

Matter-wave interference \( \rightarrow \) \( \phi \) dependence!
Photofragment Angular Distributions

\[ |\Omega_i| = 1 \]

\[ m_i \]

\begin{array}{cccc}
0 & 1 & 2 & 3 & 4 \\
4 & & & & \\
3 & & & & \\
2 & & & & \\
1 & & & & \\
0 & & & & \\
\end{array}

\[ |\Omega_i| = 0 \]

\[ m_i \]

\begin{array}{cccc}
0 & 1 & 2 & 3 \\
3 & & & & \\
2 & & & & \\
1 & & & & \\
0 & & & & \\
\end{array}

M. McDonald et al., Nature 535, 122 (2016)
Photofragment Angular Distributions

$|\Omega_i| = 1$

$|Y^1_4 + e^{i\delta}Y^3_4|^2$

M. McDonald et al., Nature 535, 122 (2016)
Probing Reaction Barriers

\[ \text{Quantum tunneling?} \]

\[ |\Omega| = 1 \]

\[ |\Omega| = 0 \]

\[ ^1S + ^3P_1 \]
Probing Reaction Barriers

Continuum ($J = 1$)

$^1S + ^3P_1$
Probing Reaction Barriers

$I(\theta) \propto 1 + \beta_2 P_2(\cos \theta)$

Continuum energy (MHz)
Probing Reaction Barriers

Continuum energy (MHz)

$I(\theta) \propto 1 + \beta_2 P_2(\cos \theta)$
Field Control of Photodissociation

Comparable energies at ~ 1 mK:

- Kinetic
- Barrier
- Zeeman
Field Control of Photodissociation

Comparable energies at ~ 1 mK:

- Kinetic
- Barrier
- Zeeman

M. McDonald et al., PRL, accepted
Field Control of Photodissociation

\[ \text{Sr}_2 + \gamma \rightarrow \text{Sr} + \text{Sr}^* \]

Energy = 30 MHz = 1.5 mK

Key point: Mixing of partial waves in the continuum

M. McDonald et al., PRL, accepted
Science with Cold and Ultracold Molecules

- Ultracold chemistry
- Molecular clocks
- Table-top particle physics
Clocks

Electronic

Vibrational

Coherence time of $|1\rangle + |2\rangle$ superposition

- Intrinsic
- Trap & environment
Two-Body Quantum Optics

Identical nuclei → Inversion symmetry

superradiant
\[ |S\rangle |P\rangle + |P\rangle |S\rangle \]

subradiant
\[ |S\rangle |P\rangle - |P\rangle |S\rangle \]

odd \((u)\)

\[ E_1 \]

\[ 2\Gamma \]

even \((g)\)

\[ M_1 \]

\[ E_2 \]

\[ \neq 0! \]
Two-Body Quantum Optics

Subradiance

\[ \left( \frac{\mu_{M1}}{\mu_{E1}} \right)^2 \approx \left( \frac{R}{\lambda} \right)^2 \approx 10^{-4} \]

@ \( R = 100 \ a_0 \)

Need \( 10^4 \times \) suppression of E1!

→ Molecules ✓

Two-Body Subradiance

$H_{int} = -d \cdot E$

$E1$

$\Psi_e$

$\Psi_0$

$u$

$g$

$M1$

$E2$

$-\mu \cdot B$

$-\frac{1}{6} Q_{ij} \nabla_i E_j$

Normalized strength

$R$

$\nu'$ $-1$ $-1$

$\nu$ $-1$ $-2$

Theory

Expt.
Subradiant Lifetime

Two-Body Subradiance

Two-body subradiance

Predissociation $\propto \Delta E$

$\propto R^{-4}$

van der Waals $\propto R^{-2.5}$

dipole-dipole

$Q > 3 \times 10^{12}$

$\propto R^2$

Natural linewidth (Hz)

Bond length ($R/a_0$)
Trap-Insensitive Spectroscopy

“Magic” optical lattice trap

create molecules

optical probe

Equal light-induced frequency shifts!

$10^{-6}$ K
Trap-Insensitive Spectroscopy

“Magic” optical lattice trap

Coherent superposition of $|1\rangle + |2\rangle$
Trap-Insensitive Spectroscopy

‘Magic’-lattice optical absorption spectrum

red sideband  carrier  blue sideband

\[ \frac{\alpha'}{\alpha} = 0.98(2) \]
Trap-Insensitive Spectroscopy

\[ \frac{\alpha'}{\alpha} = 0.98(2) \]

\[ \frac{\alpha'}{\alpha} = 0.889(2) \]

\[ 2\nu_x \]
Trap-Insensitive Spectroscopy

“Magic” optical lattice trap

Nonresonant crossing:
Traditional choice; hard to find 😞

Resonant crossing:
Heating/loss

Resonant crossing:
* No heating/loss!
* Easy to find 😊

Lattice wavelength
Dynamic polarizability

>100 nm
Clock Based on Molecular Vibrations

<30 THz
Trap-Insensitive Spectroscopy

“Magic” optical lattice trap

Resonant crossing:
* No heating/loss!
* Easy to find

Lattice frequency (THz)

Lattice wavelength

Dynamic polarizability

narrow resonance

0.006 nm
Trap-Insensitive Spectroscopy

“Magic” optical lattice trap

- Line width (MHz): $600 \times$ coherence time
- 160 Hz
Trap-Insensitive Spectroscopy

“Magic” optical lattice trap

Lattice frequency (THz)

160 Hz

vibrational “clock” resonance

26 THz

Q = 2×10^{11}
(fiber limited)
Science with Cold and Ultracold Molecules

- Ultracold chemistry
- Molecular clocks
- Table-top particle physics
New Mass-Dependent Forces

\[ V = -\frac{GM^2}{r} \left(1 + Ae^{-r/\lambda}\right) \]

Yukawa

\[ A < 10^{21} \]

@ 1 nm!

→ Need state-of-the-art measurement of van der Waals interatomic force


M. Borkowski et al., arXiv:1612.03842
Molecular QED and 5\textsuperscript{th} force

Born-Oppenheimer approximation

\[ E_{\text{tot}} \approx E_{\text{el}} + E_{\text{vib}} + E_{\text{rot}} \]

Beyond B-O

- adiabatic
- nonadiabatic
- relativistic
- finite-nuclear-size

\[ \mu = \frac{m_e}{A m_p} \]

\[ \mu^2, \alpha^2 \mu, \alpha^3 \mu \]

\[ (r_c/a_0)^2 \]

higher-order \( \alpha^4 \mu < 1 \text{ Hz} \)

\( ^{84}\text{Sr}, \; ^{86}\text{Sr}, \; ^{88}\text{Sr} \) dimers (6 combinations): fit up to 5 \( \mu \)-dependent corrections
Molecular QED and 5\textsuperscript{th} Force

Neutron scattering 2006

Strength of non-1/r\textsuperscript{2} interaction

\[
\log |A| = \log \left( \frac{l}{m} \right)
\]

Van der Waals forces:

1-Hz Sr\textsubscript{2} spectroscopy projection

Casimir forces
Zlab

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