High-Precision Physics and Chemistry with Ultracold Molecules

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Quantum science with molecules

Fundamental chemistry

Quantum materials

Fundamental physics

Why ultracold?

Quantum science with molecules

- Can small molecules contribute to our understanding of fundamental forces ("old" and "new" physics)?

- Is it possible to control these molecules across frequency regimes, and to decouple their internal dynamics from the environment?

- Is there a metrological advantage for molecular systems?
Producing ultracold molecules

direct cooling

assembly
Ultracold molecule photo-assembly

$10^{-6}$ K

$200 \text{ m/s} \rightarrow 10 \text{ mm/s}$

No kinetic energy release:
Light carries away excess energy
Optical lattice trap

Standing wave of light holds atoms or molecules in bright microtraps

$10^{-6}$ K

Graphics: S. Blatt
Making ultracold molecules

G. Reinaudi et al., PRL 109, 115303 (2012)
Ultracold van der Waals dimers

- Singlet–triplet narrow transitions: quantum control
- Spinless ground state: precise metrology & ab initio

Ab initio (BO) & experimental potential
Narrow-line control of molecules

Identical nuclei $\rightarrow$ Inversion symmetry

$$|S\rangle|P\rangle + |P\rangle|S\rangle$$

superradiant

$$|S\rangle|P\rangle - |P\rangle|S\rangle$$

subradiant

~10 ms

odd ($u$)

2\Gamma

E1

~10 \mu s

even ($g$)

0 \neq 0!

M1

E2
Narrow-line control of molecules

Subradiant states

Molecule–light coherence

5.5 ms lifetime

Narrow-line control of molecules

red sideband  carrier  blue sideband

Precise measurements of sidebands & temperature
Narrow-line control of molecules

State-resolved photochemistry

$J=1 \rightarrow ^1S_0+^3P_1$

$J=0 \rightarrow 0_g(^1S_0+^1S_0)$

Side-view camera

Line-of-sight camera: quantum interference

M. McDonald et al., Nature 535, 122 (2016)
B. McGuyer et al., NJP 17, 055004 (2015)
Creation of Ultracold Exotic Gases via Laser Cooling and Precise Dissociation of Molecules

Creating exotic ultracold gases

Step 1: Direct laser cooling

Step 2: Precise molecular dissociation

Step 3: Probe exotic ultracold gases

I. C. Lane, *PRA* 92, 022511 (2015)
Studying novel ultracold gases

Columbia – Harvard, W. M. Keck

Step 3a: Ultracold chemistry

Step 3b: Fundamental physics
Ultracold photochemistry

Photodissociation

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

- Quantum-state selected reactants and products
- Reactions described by matter waves
- High-precision studies of electronic continua
Ultracold Photodissociation

Photofragment angular distribution
Ultracold Photodissociation

Matter-wave interference

\[ J = 4; \quad M = \pm 1 \]

\[ J = 4; \quad M = 1,3 \]

M. McDonald et al., Nature 535, 122 (2016)
Ultracold Photodissociation

Matter-wave interference

\[ J = 4; \ M = \pm 1 \]

\[ J = 4; \ M = 1,3 \]

"Classical" fragmentation of diatomic molecule

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

\[ |\Omega_i| = 1 \]

\[ m_i \]

\[ |\Omega_i| = 0 \]

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

\[ 1S + 3P_1 \]

Quantum tunneling?
Probing Reaction Barriers

Continuum \((J = 1)\)

\(^1S + ^3P_1\)
Probing Reaction Barriers

Continuum energy (MHz)

$J_\parallel = 0$

$J_\perp = 2$

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

$\beta_2$

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

Anisotropy parameter: QM info
Probing Reaction Barriers

\[ \beta_2 \propto 1 + \beta_2 P_2(\cos \theta) \]

\[ I(\theta) \propto 1 + \beta_2 P_2(\cos \theta) \]

Continuum energy (MHz)

barrier

QC theory
Field Control of Photodissociation

Comparable energies at ~ 1 mK:
- Kinetic
- Barrier
- Zeeman

M. McDonald et al., PRL 120, 033201 (2018)
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 120, 033201 (2018)
Molecular lattice clock

Maintain long-term coherence between quantum states!

New science with molecular clocks

Interatomic force sensing

Ultrashort-range gravity

\[ V(R) \]

Sub-ppt sensitivity

\[ V = 1 \]

Also THz metrology, \( \frac{d}{dt} \left( \frac{m_e}{m_p} \right) \), ...

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

A < \( 10^{22} \)!

Y. Kamiya et al., PRL 114, 161101 (2015)
Creation of Sr$_2$ in absolute ground state

Stimulated Raman adiabatic passage (STIRAP)

Probe detuning (MHz)

EIT spectrum across full potential
Creation of $\text{Sr}_2$ in absolute ground state

Stimulated Raman adiabatic passage (STIRAP)

100% in $\nu = -1$

Up to 90% one-way efficiency

$\nu = 0$

$\nu = -1$
Molecular lattice clock

$|1\rangle \rightarrow |2\rangle$

Magic-wavelength lattice
Molecular lattice clock

Coherence in magic lattice

Energy of \( |2\rangle \)

Clock resonance linewidth

Molecular lattice clock

Coherence in magic lattice: ×3,000!

\[ Q = 8 \times 10^{11} \quad \text{and} \quad Q \text{ (intrinsic)} > 10^{26} \]
Molecular lattice clock

Coherence in magic lattice

Quantum Rabi oscillations

\[ |0\rangle + \sqrt{V = 4} |\ldots\rangle \]
\[ \sqrt{V = 62} |\ldots\rangle \]
Molecular lattice clock

Coherence in magic lattice

K. H. Leung et al., PRL 125, 153001 (2020)
Two-body loss of $\text{Sr}_2$ molecules

$\text{Sr}_2 + \text{Sr}_2 \rightarrow \text{Sr}_4 \rightarrow \text{Sr}_3 + \text{Sr} + 3,000(400) \text{ cm}^{-1}$

- “Universal” two-body loss rate
- No dependence on vibrational state $\nu$
- Unknown if $\text{Sr}_4$ complexes live long enough to experience trap-light excitation
- 3D optical lattice can protect molecules from collisions

Ultracold Sr$_2$ reactive collisions

Molecule loss rates are independent of vibrational state
High-precision vibrational spectrum

Autler-Townes spectroscopy of van der Waals force

Selected bound states: $>10^4\times$ higher precision than prior work

A. Stein et al., *EPJD* 57, 171 (2010)
Summary

- Ultracold van der Waals molecules
- Photochemistry in the quantum regime
- Field control of reactions
- Molecular lattice clock: $Q \sim 10^{12}$
- ppt interatomic force measurements
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