CREATION OF ULTRACOLD TETRATOMIC MOLECULES FROM A FERMI GAS OF MICROWAVE-SHIELD POLAR MOLECULES

Xin-Yu Luo

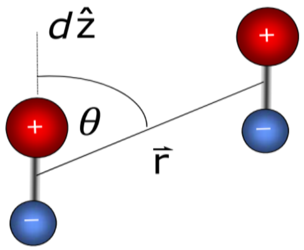
↘ *Max-Planck Institute of Quantum Optics*

The International Conference on Laser Spectroscopy 2023

Estes Park, 27 June 2023

ULTRACOLD POLAR MOLECULES

Dipolar Interaction Tuned by Electric Field



$$U_d(\vec{r}) = \frac{d^2}{4\pi\epsilon_0}\frac{1-3\cos^2\theta}{r^3}$$

Gadway & Yan J. Phys. B 49, 152002 (2016)

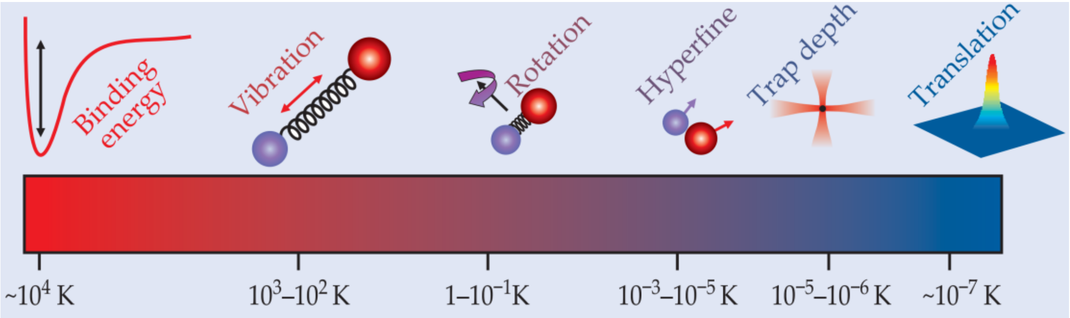
Applications:

- Cold collisions and chemistry
- Quantum many-body physics
- Quantum information
- Fundamental symmetries

…

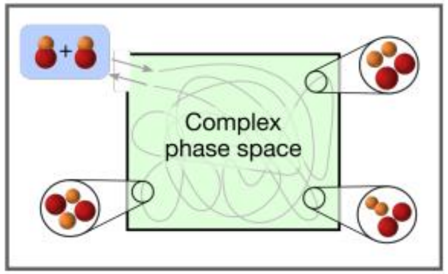
Direct cooling: 1 μK ~ 10 K
Cold-atom assembly: 20 nK ~ 10 μK

Rich Degrees of Freedom: Blessings and Curses



Jin & Ye Physics Today 64, 5, 27 (2011)

Rich (Complex and Unstable) Collisions



Collisional stable
Quantum degenerate
Scattering resonances

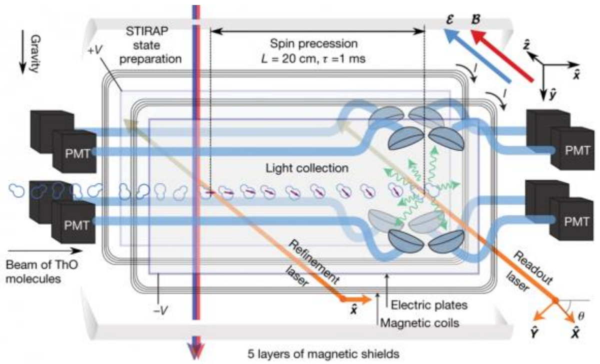
Bause et al., J. Phys. Chem. A 127, 729 (2023)

A BOOMING FIELD

Latest review: Langen et al., arXiv 2305.13445 (2023)

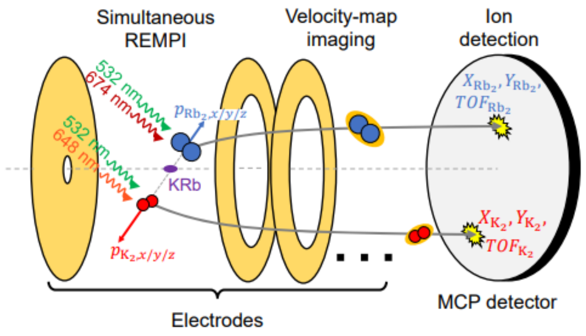
A glance of recent progresses

Testing Fundamental Symmetries



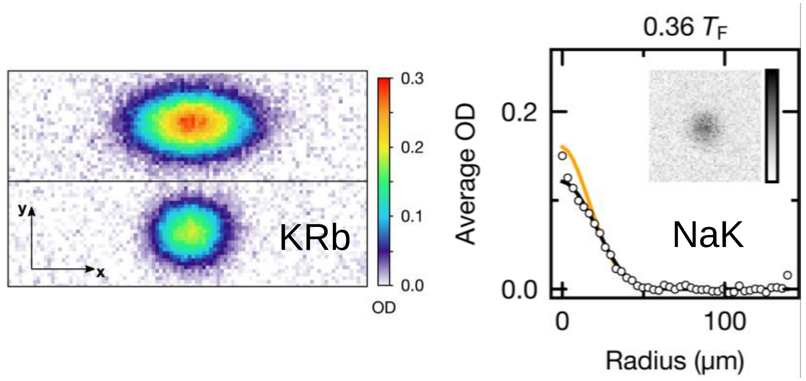
ACME III,JILA, ICL, Caltech, Columbia,…

Cold Chemistry



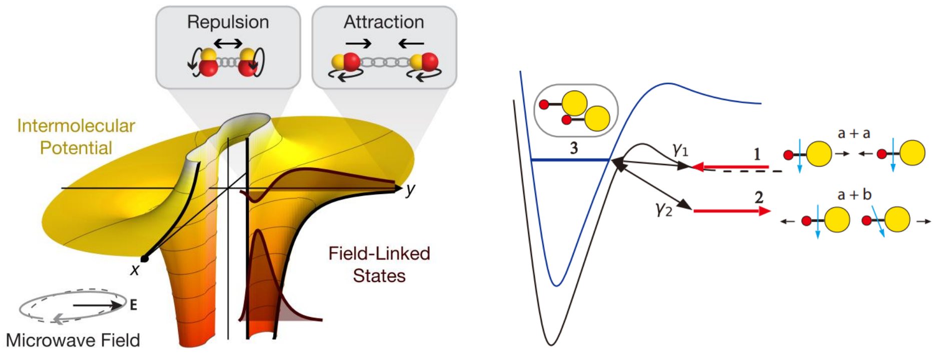
JILA, Harvard,…

Degenerate Quantum Gases



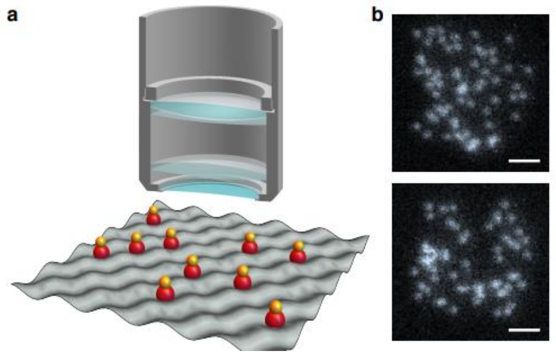
JILA, MPQ, USTC

Scattering Resonances



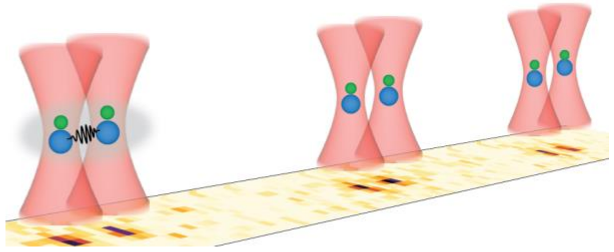
MIT, MPQ, USTC,…

Quantum Gas Microscope



Princeton

Tweezer Array

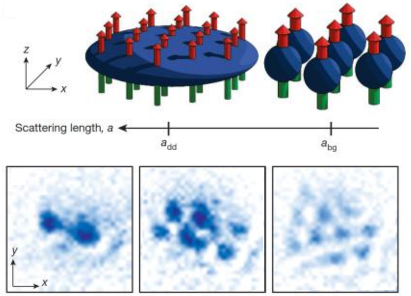


Harvard, Princeton, Durham,…

ULTRACOLD DIPOLAR MANY-BODY SYSTEMS

Magnetic Atoms

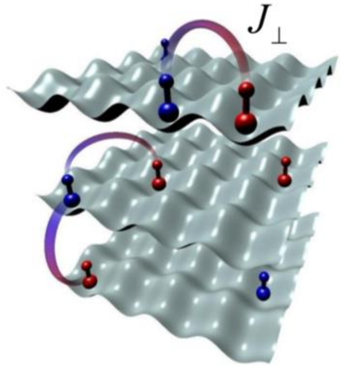
Weak dipoles ~ 10 μ_B, stable (10 s)



Kadau et al., Nature 530, 194 (2016)

Dipolar Molecules

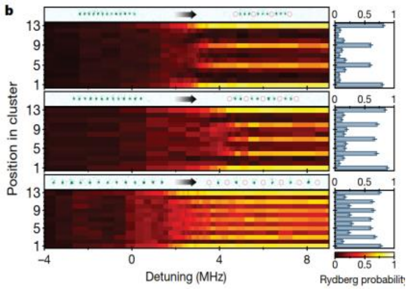
Medium dipoles ~ 3 Debye, alone stable (10 s)



Yan et al., Nature 501, 521 (2013)

Rydberg Atoms

Strong dipoles ~ 10^4 Debye, lifetime ~ 100 μs

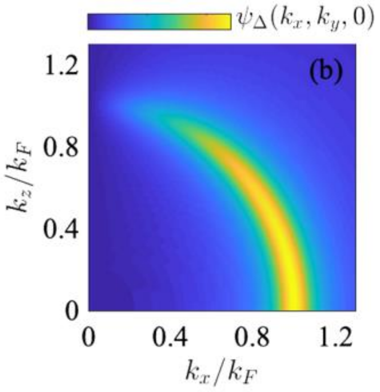


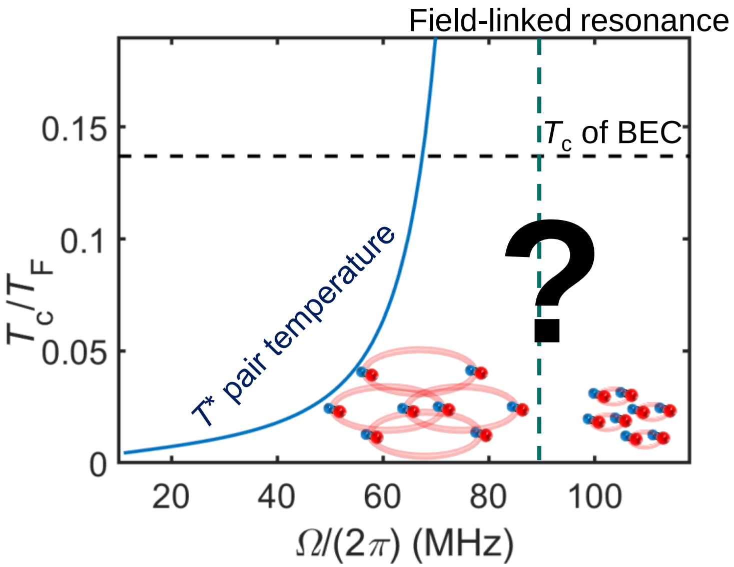
Bernien et al., Nature 551, 579 (2017)

Itinerating dipoles → **Fixed dipoles**

NEW POSSIBILITIES IN MOLECULAR QUANTUM GASES

Fermions: Dipolar BCS-BEC Crossover





Deng et al., PRL 130, 183001 (2023)

Interplay of contact interaction and strong dipolar interaction

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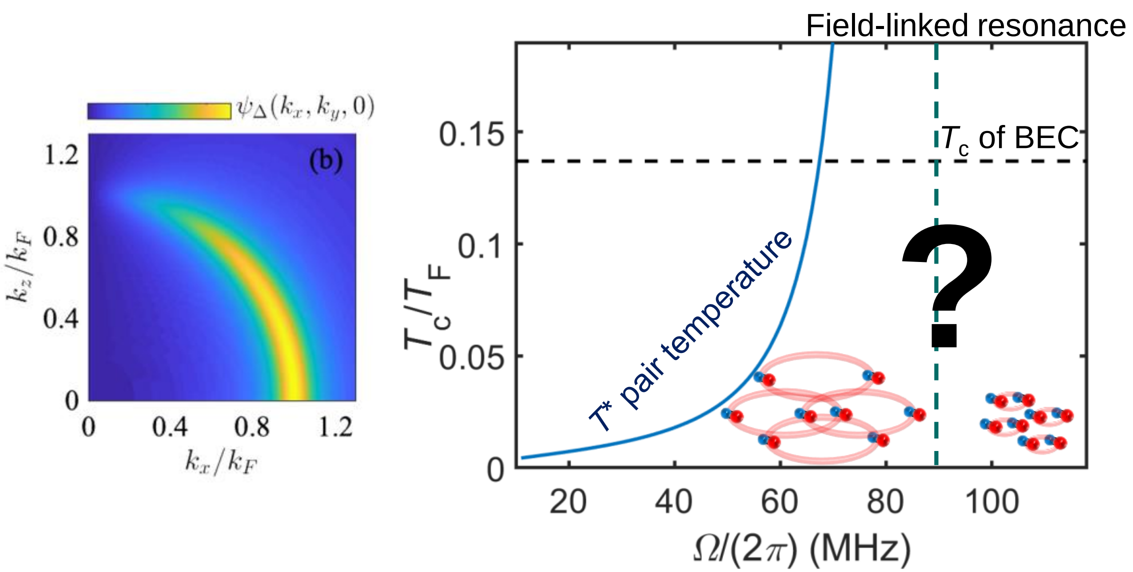
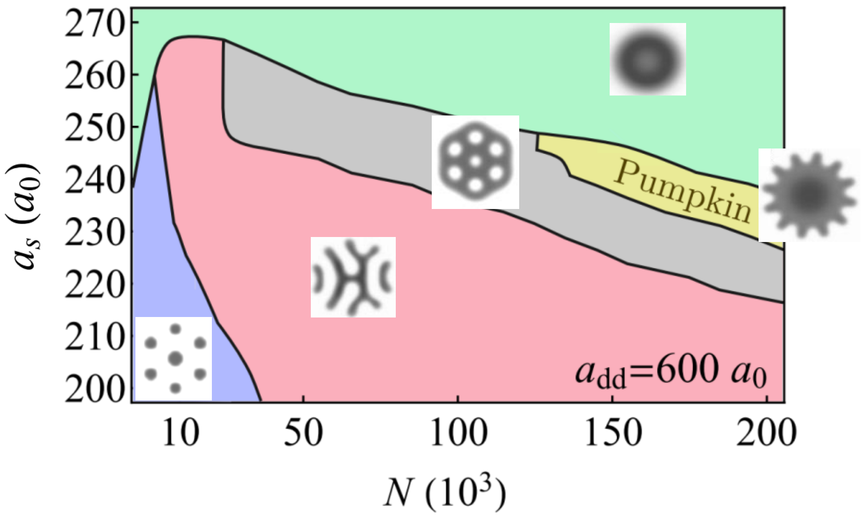


Deng et al., PRL 130, 183001 (2023)

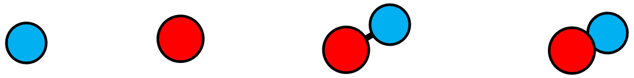
Bosons: Droplets, Supersolids, Crystals



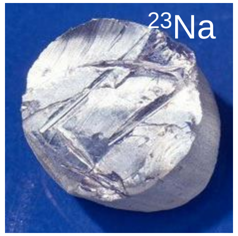
Schmidt et al., Phys. Rev. Research 4, 013235 (2021)

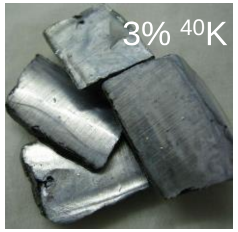
Interplay of contact interaction and strong dipolar interaction

ROAD TO A DEGENERATE FERMI GAS OF NAK



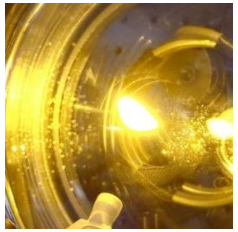
600/300 K

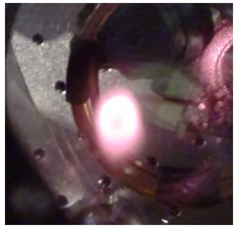




Laser cooling of atoms

100 μK





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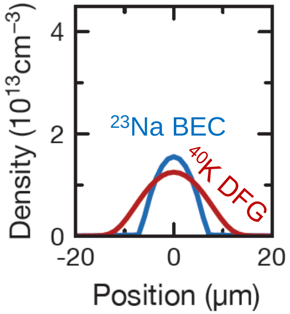
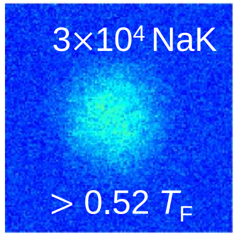


Assembly of atoms

100 nK

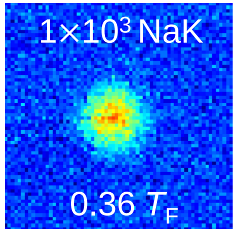
04/2021



Evaporation of molecules

20 nK

09/2021



Start at MPQ in 2010

Duda, Chen, Schindewolf, Bause, Milczewski, Schmidt, Bloch & Luo Nat. Phys. 19, 720 (2023)

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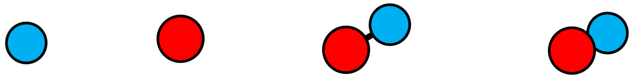

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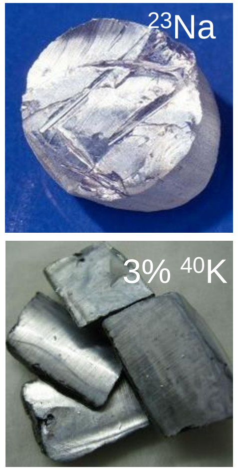

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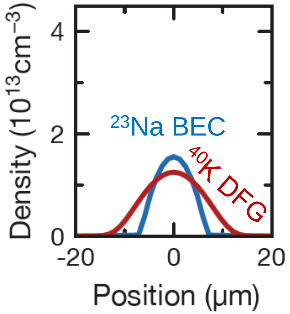

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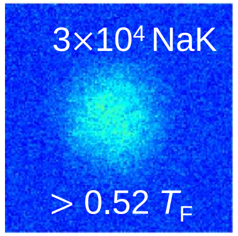

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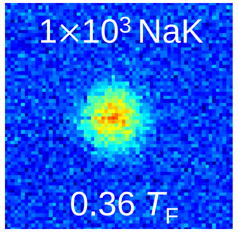
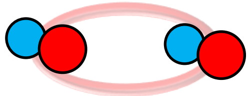


Assembly of molecules

And… even bigger molecules!



Start at MPQ in 2010

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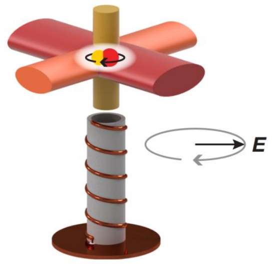
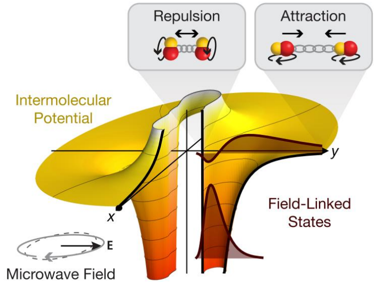
CONTENT

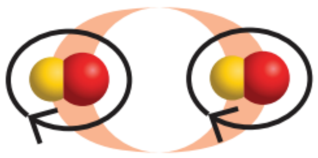
1. **Evaporation of microwave-shielded polar molecules**



2. Field-linked resonances of polar molecules



3. Ultracold field-linked tetratomic molecules



COLLISIONAL STABILITY OF ULTRACOLD MOLECULES

KRb + KRb → K_2 + Rb_2

NaK + NaK ↛ Na_2 + K_2

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(Near) universal two-body loss for all bi-alkali molecules

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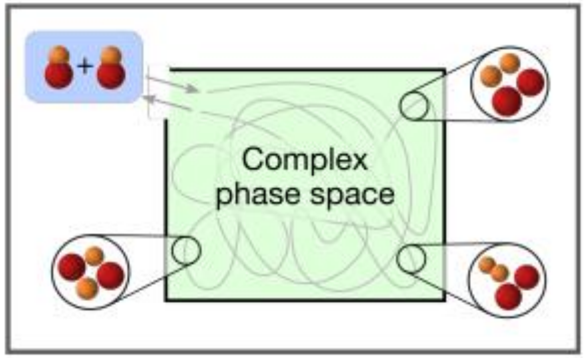
$$\tau_{\mathrm{RKKM}} = \frac{2\pi\hbar\rho}{N_{\mathrm{out}}}$$



Stick collisions: Mayle et al., PRA 87, 012709 (2013)

Bause, Christianen, Schindewolf, Bloch & Luo, J. Phys. Chem. A 127, 729 (2023)

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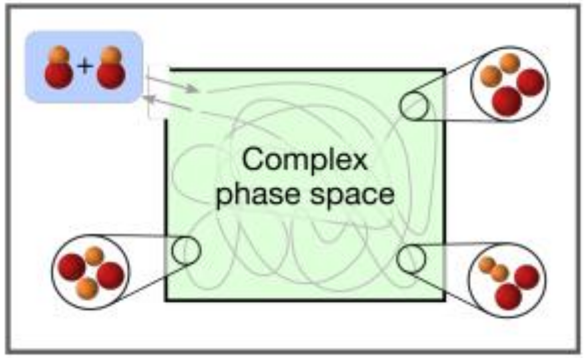

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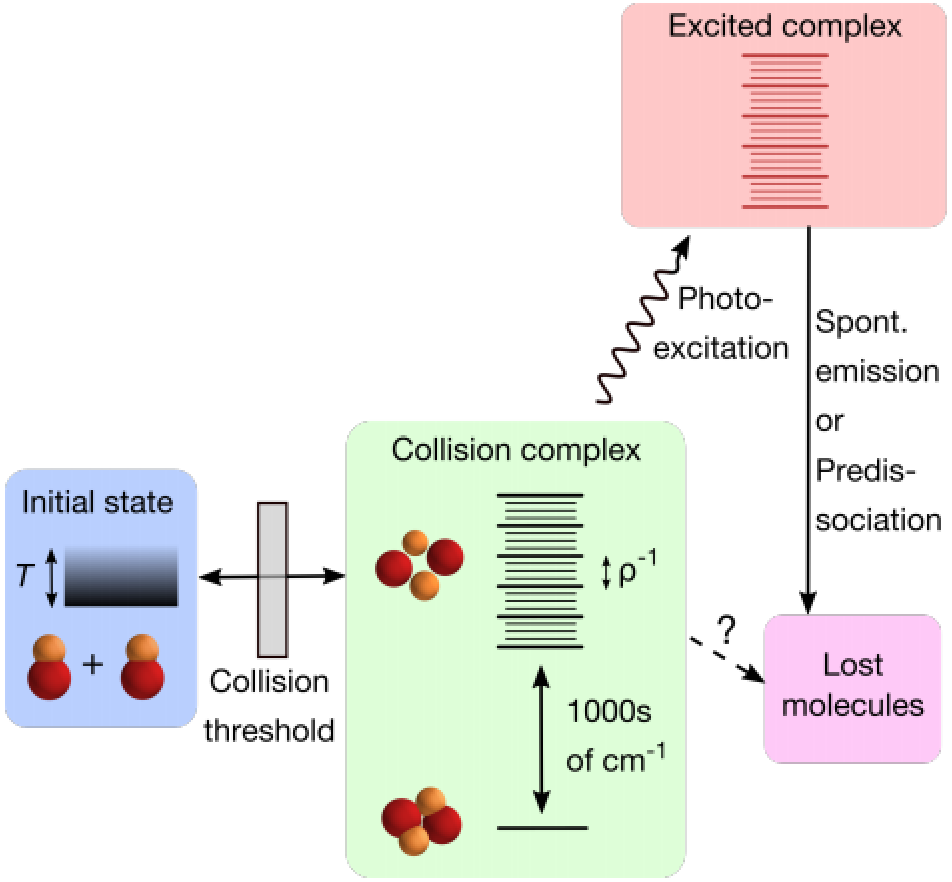
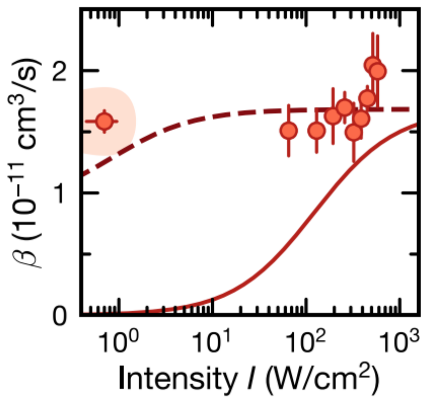


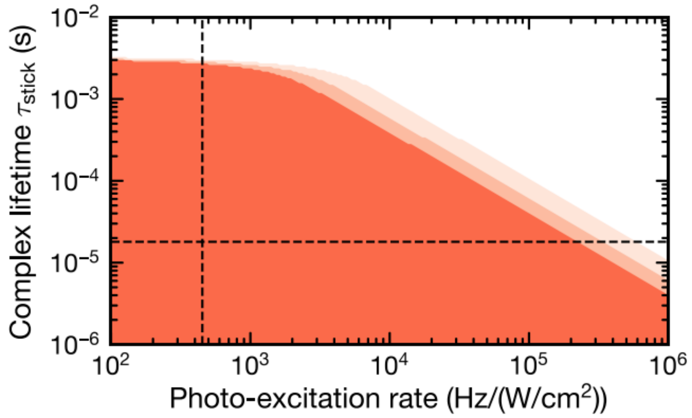
Photon induced loss: Christianen *et al.*, PRL 123, 123402 (2019)
Confirmed in RbCs and KRb molecules (2020)

Bause, Christianen, Schindewolf, Bloch & Luo, J. Phys. Chem. A 127, 729 (2023)

THE MYSTERY OF STICKY COLLISION

Save NaK molecules in dark? No.
The loss is independent of light intensity!





Similar results in NaRb and bosonic NaK

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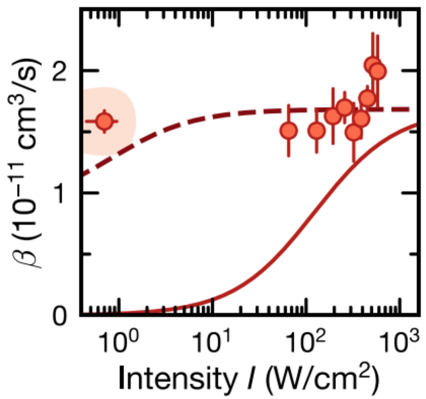
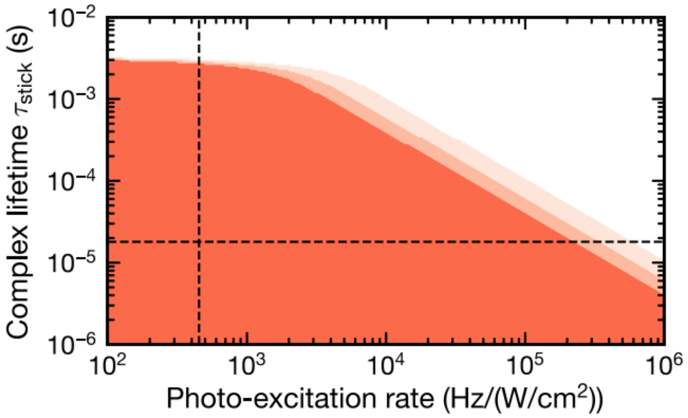




Similar results in NaRb and bosonic NaK

Short-range four-atom collisions is beyond modern quantum dynamics calculation!

Molecule	d_0/D	Nucl. spin	τ_{exp}	τ_{RRKM}
$^{23}Na^{39}K$	2.7	$\lvert -3/2, -1/2\rangle$	> 0.35 ms	6 μs
$^{23}Na^{40}K$	2.7	$\lvert 3/2, -4\rangle\dagger$	> 2.6 ms	18 μs (4.9 ms)
		$\lvert 3/2, -4\rangle\dagger$	> 1.4 ms	18 μs (4.9 ms)
		mixed	> 2.3 ms	18 μs (54 μs)
		mixed	> 133 μs	18 μs (54 μs)
$^{23}Na^{87}Rb$	3.2	$\lvert 3/2, 3/2\rangle * \dagger$	> 1.2 ms	19 μs
$^{40}K^{87}Rb$	0.6	$\lvert -4, 1/2\rangle$	360(30) ns	170(60) ns
$^{87}Rb^{133}Cs$	1.2	$\lvert 3/2, 7/2\rangle * \dagger$	0.53(6) ms	0.253 ms
		$\lvert 3/2, 7/2\rangle * \dagger$	0.8(3) ms	0.253 ms
		$\lvert 3/2, 5/2\rangle$	2.1(1.3) ms	0.253 ms
		$\lvert 1/2, 7/2\rangle$	> 3.3 ms	0.253 ms
$^{40}K^{87}Rb$ + ^{87}Rb			0.39 ms	1 ns

Ultracold sticky collisions: Theoretical and experimental status

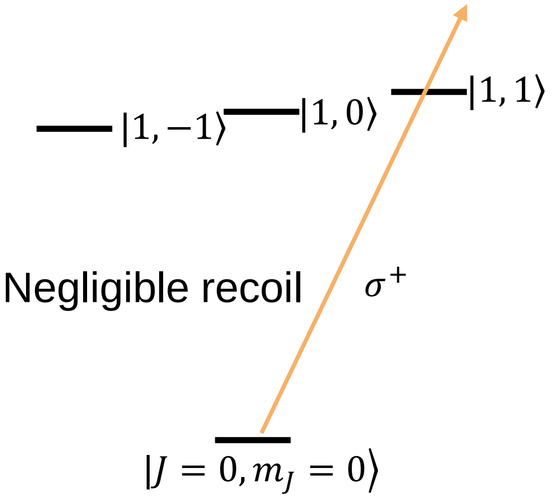
Roman Bause

Arthur Christianen

Bause, Christianen, Schindewolf, Bloch & Luo, J. Phys. Chem. A 127, 729 (2023)

MICROWAVE SHIELDING

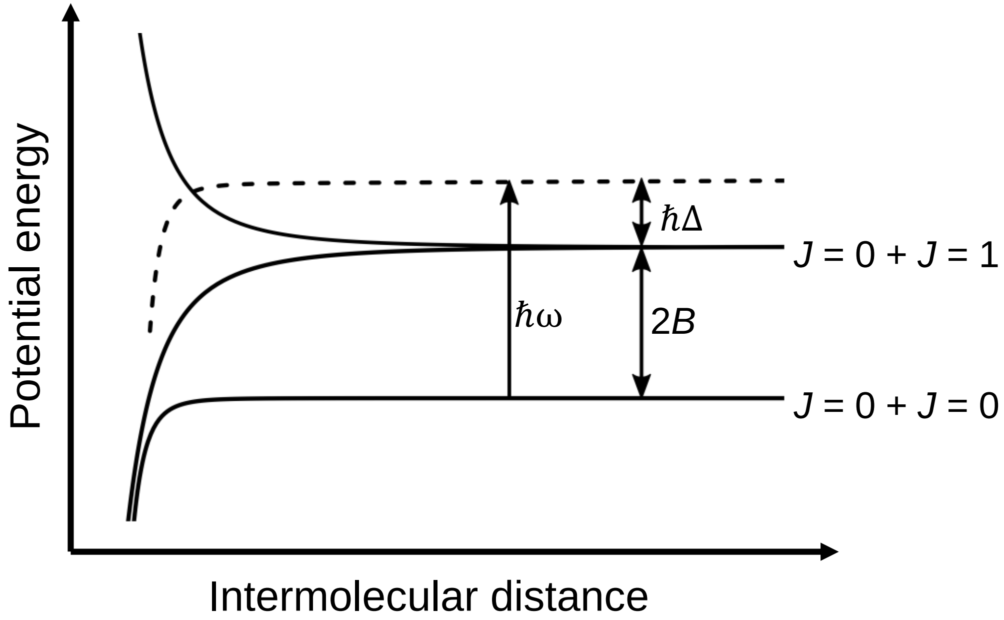
Excited state lifetime ~ 100 s



Tijs Karman

Goulven Quéméner

Intermolecular Potential



Back-to-back proposals:
Karman & Hutson, PRL **121**, 163401 (2018)
Lassablière & Quéméner, PRL **121**, 163402 (2018)
First experiment: Anderegg et al., Science **373**, 779 (2021).

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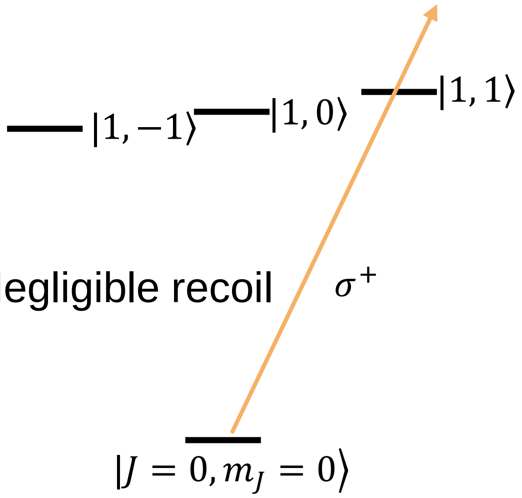

Negligible recoil

Tijs Karman

Goulven Quéméner

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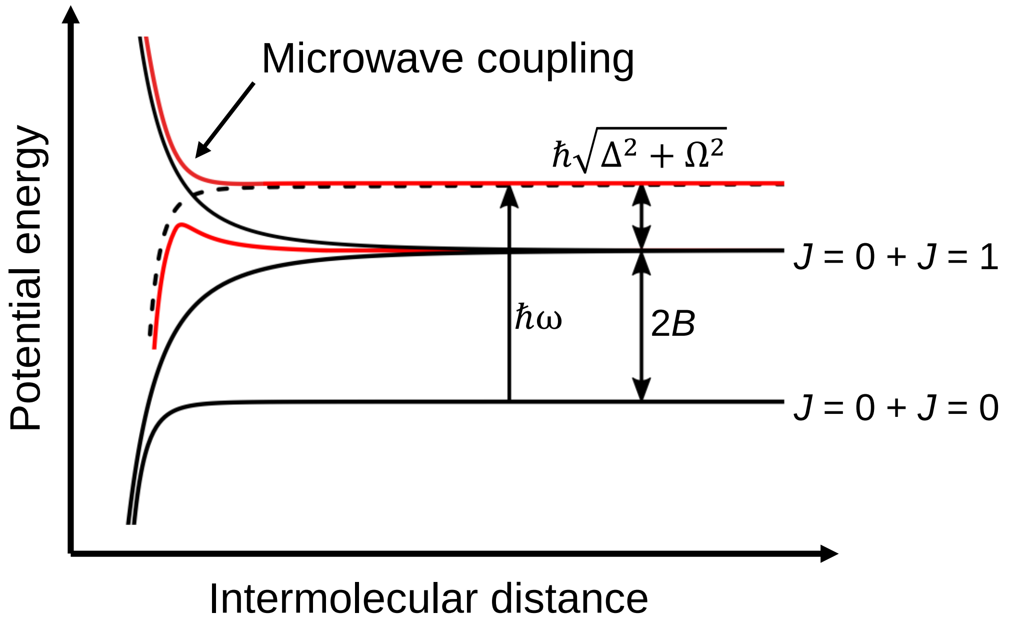

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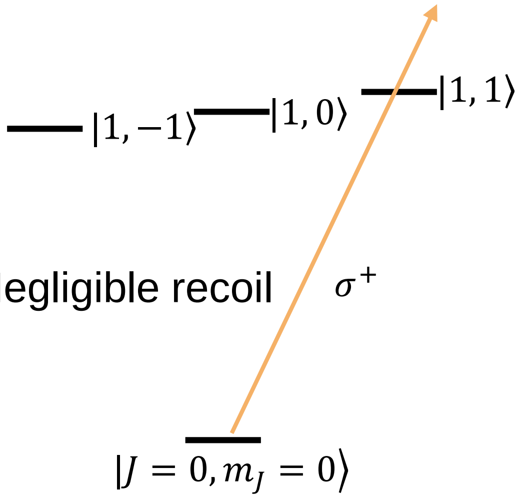

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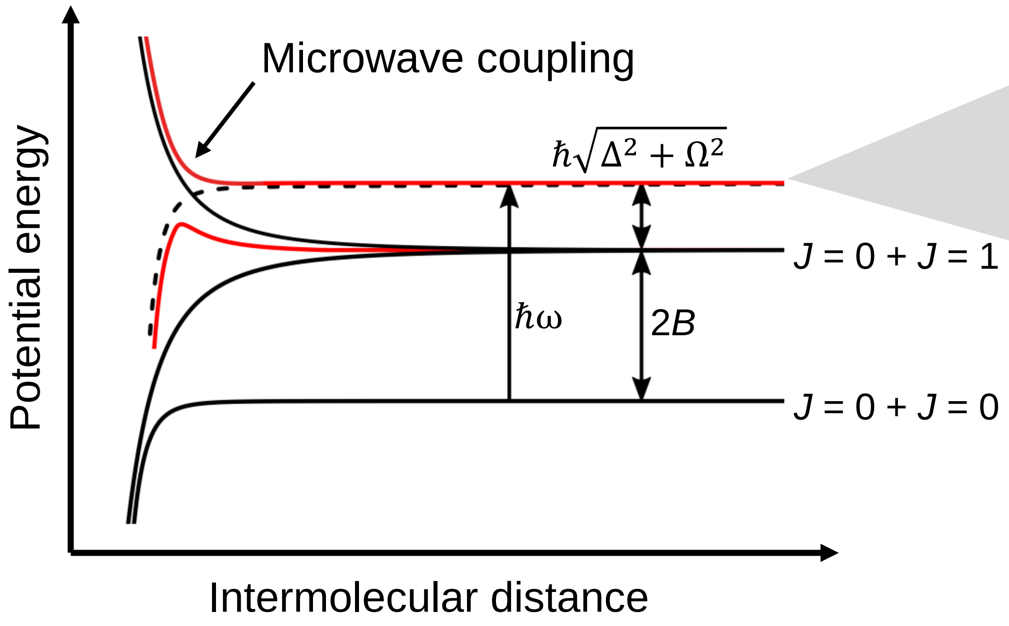
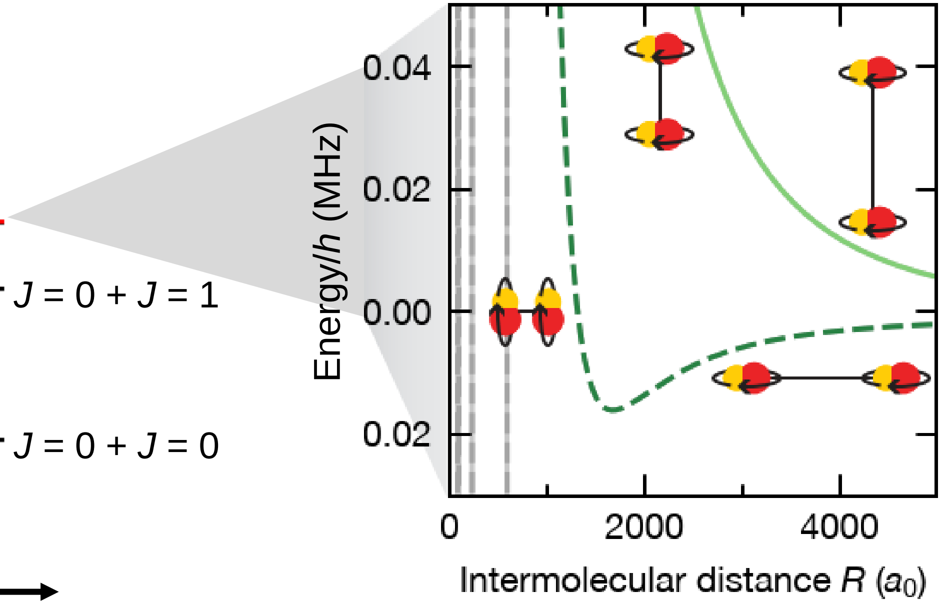
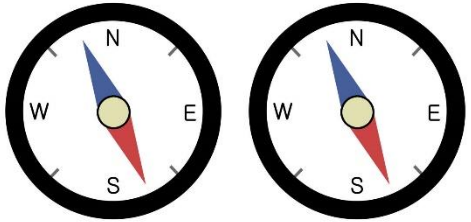




Classical analogy



HIGHLY TUNABLE INTERMOLECULAR POTENTIAL

Tuning knobs

- Rabi frequency Ω
- Detuning $\delta_r = \Delta/\Omega$
- Ellipticity ξ

van de Waals shielding core

$$V_{\text{eff}}(\mathbf{r}) = \frac{C_6}{r^6}\sin^2\theta\{1 - \mathcal{F}_\xi^2(\varphi) + [1 - \mathcal{F}_\xi(\varphi)]^2\cos^2\theta\}$$
$$+ \frac{C_3}{r^3}[3\cos^2\theta - 1 + 3\mathcal{F}_\xi(\varphi)\sin^2\theta],$$

Dipolar interaction

Tao Shi

Deng, Chen, Luo, Zhang, Yi & Shi PRL 130, 183001 (2023)

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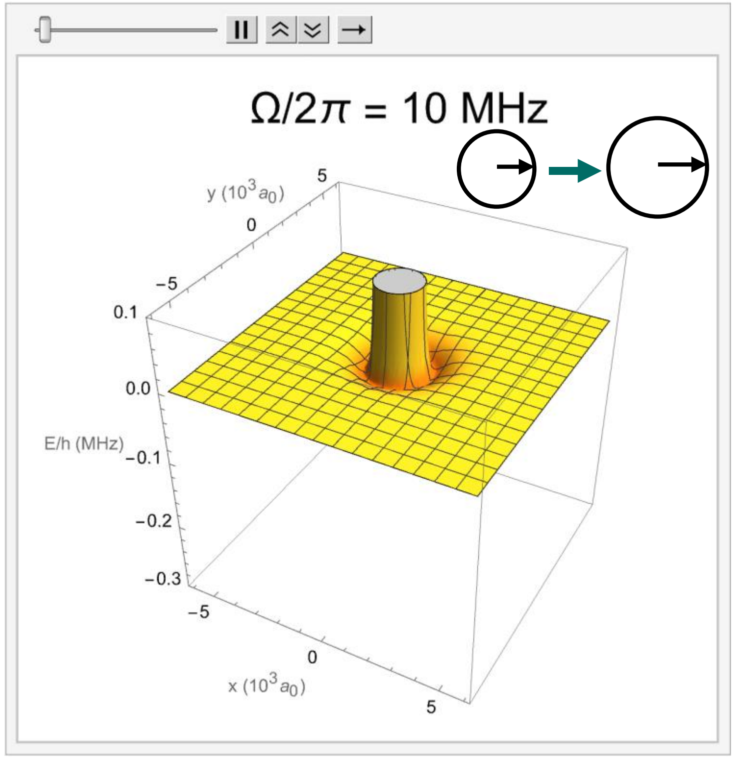
Dipolar interaction

Intermolecular potential in the plane of microwave field



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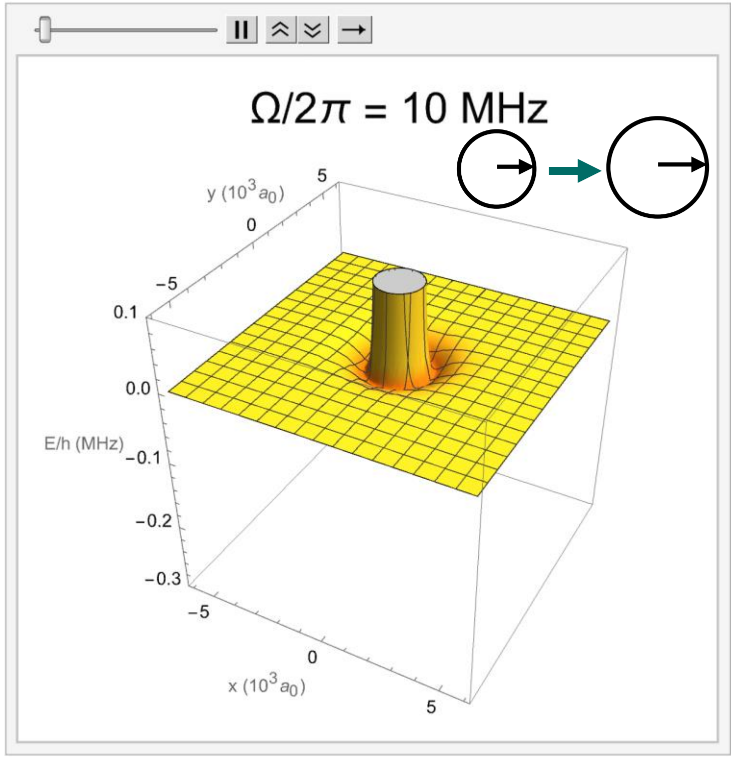
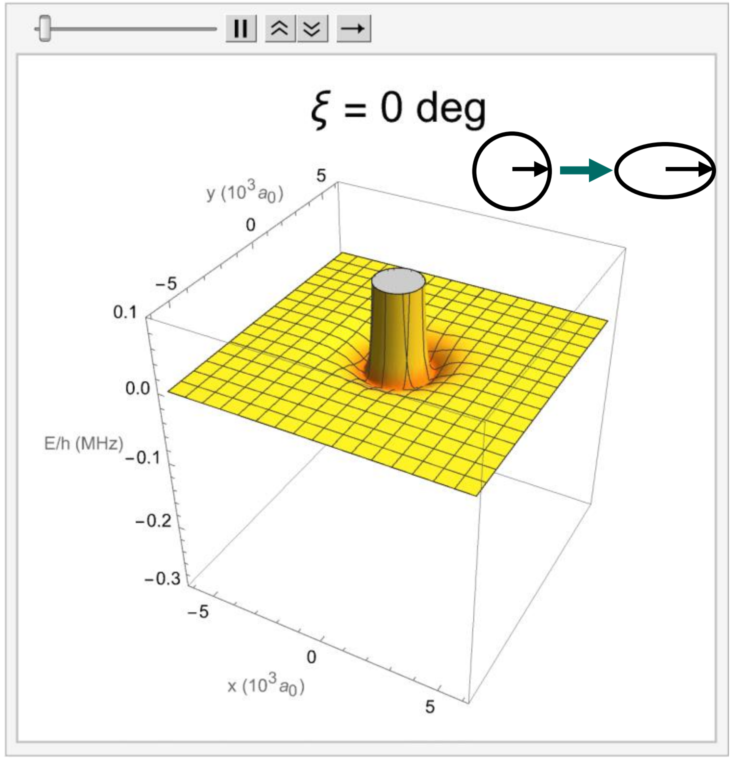
Dipolar interaction

Intermolecular potential in the plane of microwave field

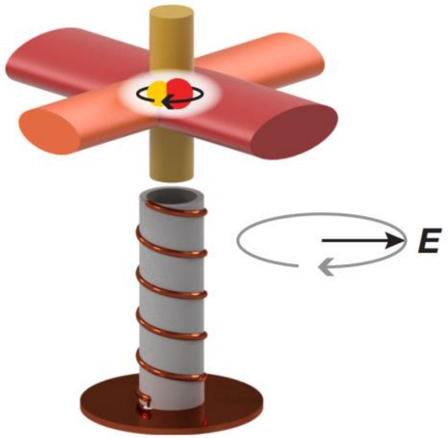




Tao Shi

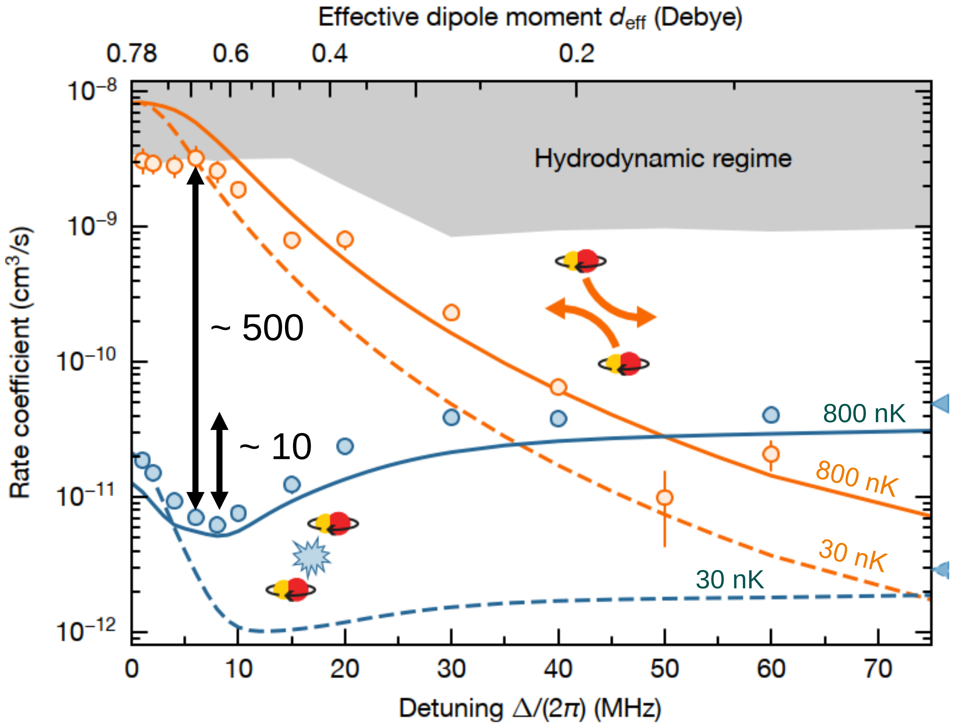
Deng, Chen, Luo, Zhang, Yi & Shi PRL 130, 183001 (2023)

STRONG DIPOLAR ELASTIC COLLISIONS AND LOW LOSS



$\Omega = 2\pi \times 11$ MHz

$\xi = 6°$



- **Thermalization rate saturated to trap frequency.**
- **Model predicts gamma ratio β_{el}/β_{inel} > 1000.**

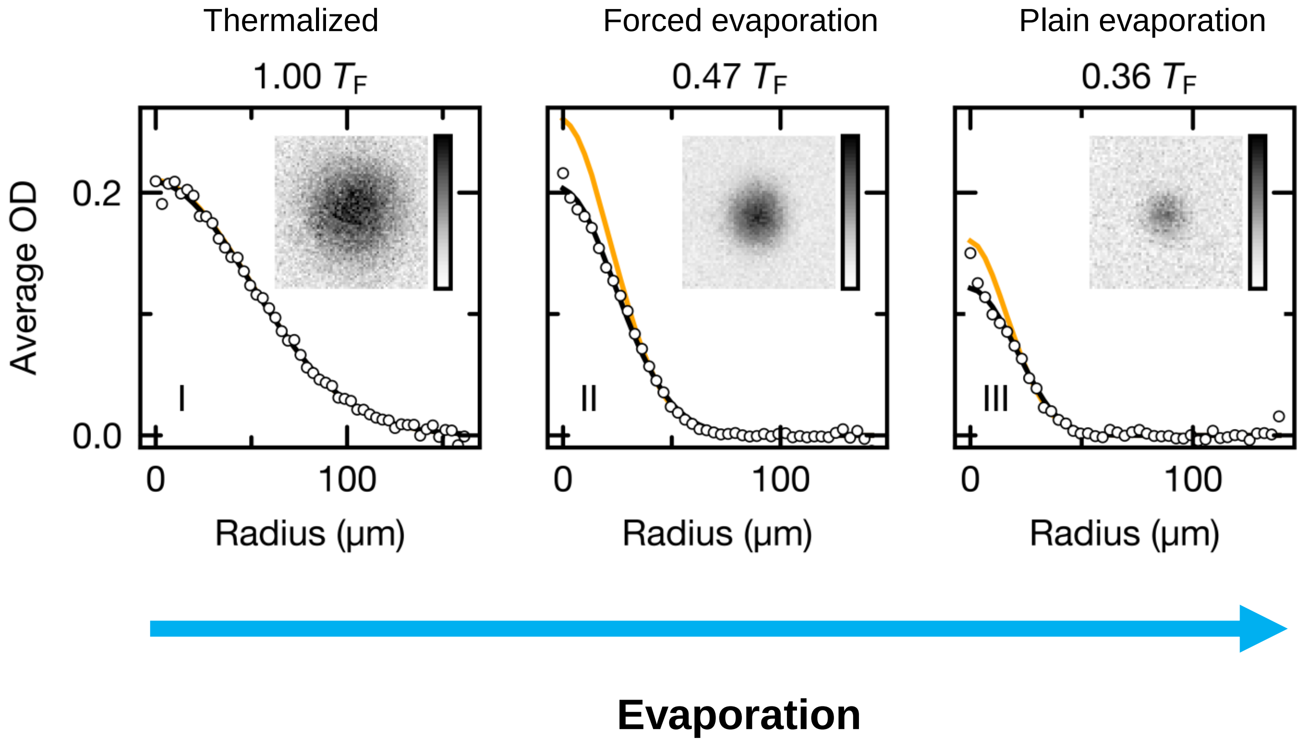
Andreas Schindewolf

Also demonstrated in bosonic NaCs (Will) and NaRb (Wang) molecules!

Schindewolf, Bause, Chen, Duda, Karmann, Bloch & Luo Nature 607, 677-681 (2022)

EVAPORATION TO QUANTUM DEGENERACY

21 nK – lowest temperature for polar molecules so far.



Andreas Schindewolf

Schindewolf, Bause, Chen, Duda, Karmann, Bloch & Luo Nature 607, 677-681 (2022)

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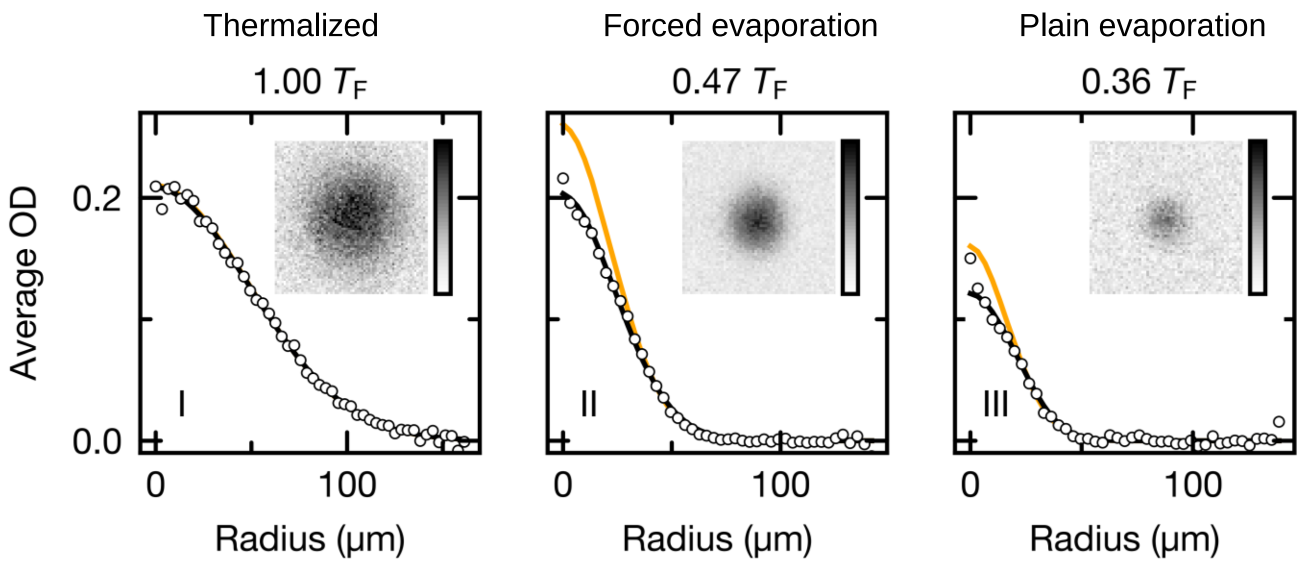


Evaporation

$U_{dd} \approx 0.05 E_F$

New possibilities:

- Fermi sea deformation and collapse
- Lattice spin models
- Electro association of tetramers

Shielding and evaporation to degeneracy in 2D (KRb): Valtolina et al., Nature **588**, 239 (2020).

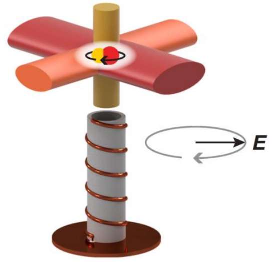
Forster resonance shileding (KRb): Matsuda et al., Science **370**, 1324 (2020).
Evaporation in 3D (KRb): Li et al., Nat. Phys. **17**, 1144 (2021).

Andreas Schindewolf

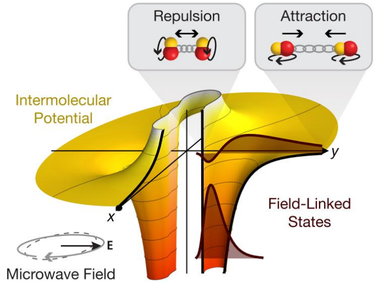
Schindewolf, Bause, Chen, Duda, Karmann, Bloch & Luo Nature 607, 677-681 (2022)

CONTENT

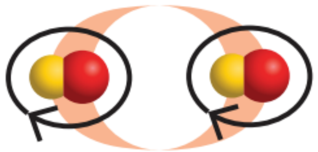
1. Evaporation of microwave-shielded polar molecules



2. **Field-linked resonances of polar molecules**

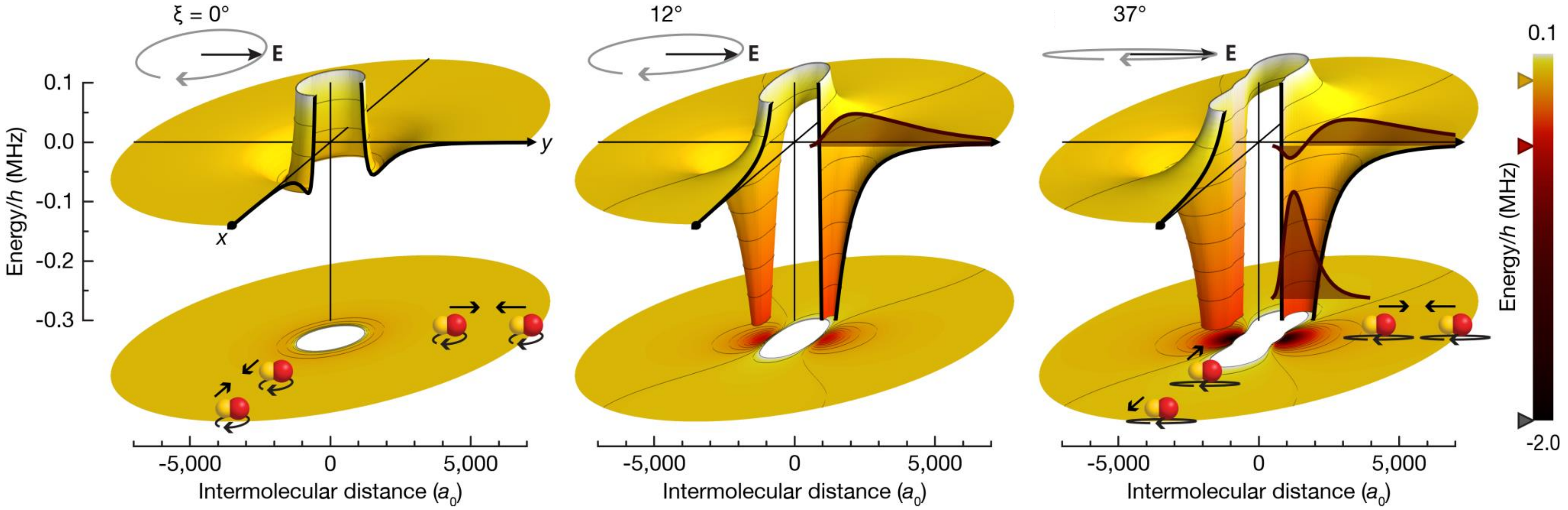


3. Ultracold field-linked tetratomic molecules



FIELD-LINKED STATES

Bound by Induced Dipolar Interaction



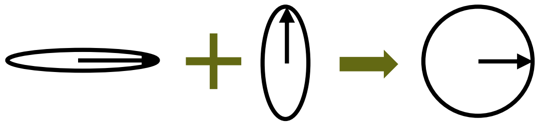
OH molecules (electric): Avdeenkov & Bohn, PRL **90**, 043006 (2003)
Bialkali (microwave): Lassablière & Quéméner, PRL **121**, 163402 (2018)

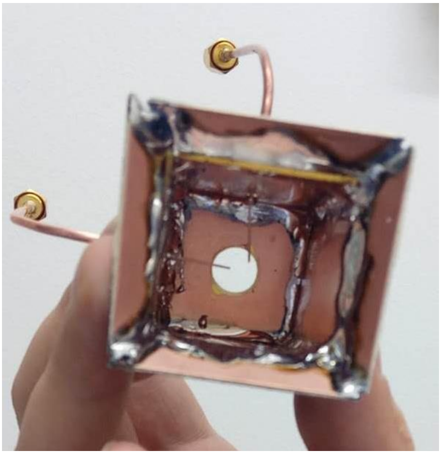
John Bohn

Goulven Quéméner

OBSERVATION OF FIELD-LINKED RESONANCES

Tuning Ellipticity by Dual-Feed Antenna





Xing-Yan Chen

Andreas Schindewolf

Chen, Schindewolf, Eppelt, Bause, Biswas, Duda, Karman, Hilker, Bloch & Luo Nature 614, 59 (2023)

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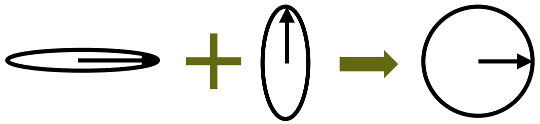
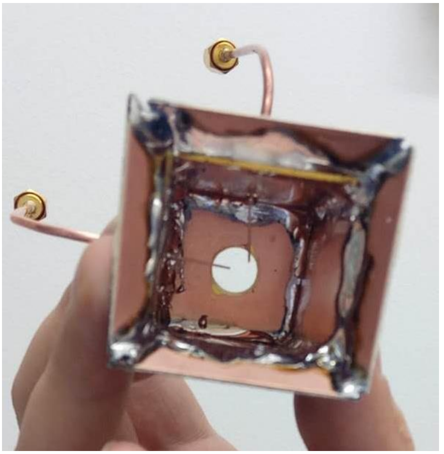
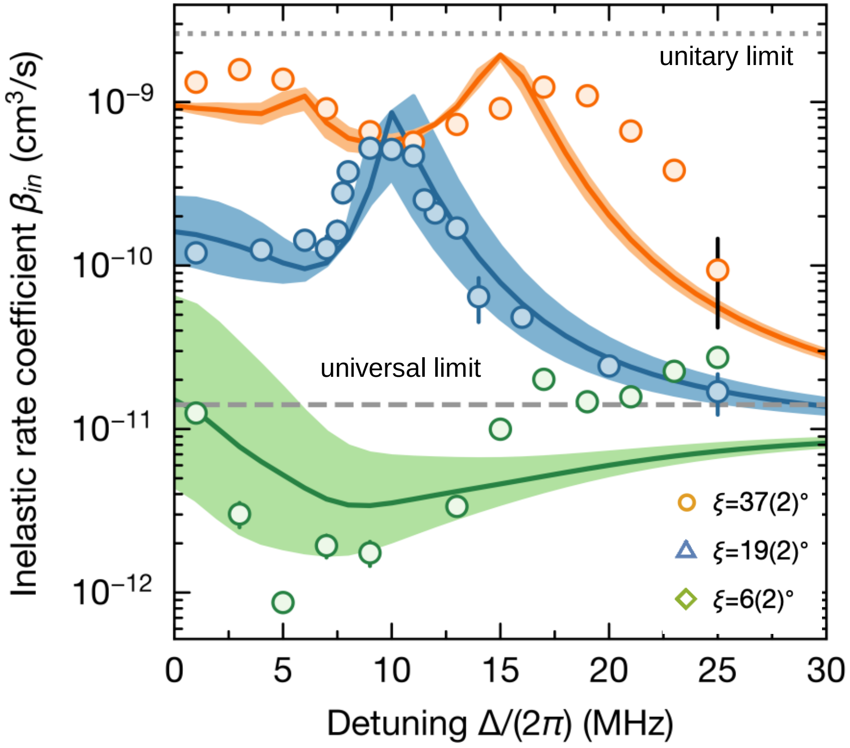




Enhanced Inelastic Collision Rate



$$\Omega = 2\pi \times 10\ \text{MHz}, T = 230\ \text{nK}$$

Xing-Yan Chen

Andreas Schindewolf
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Chen, Schindewolf, Eppelt, Bause, Biswas, Duda, Karman, Hilker, Bloch & Luo Nature 614, 59 (2023)

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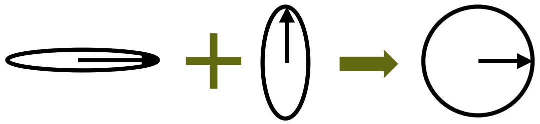

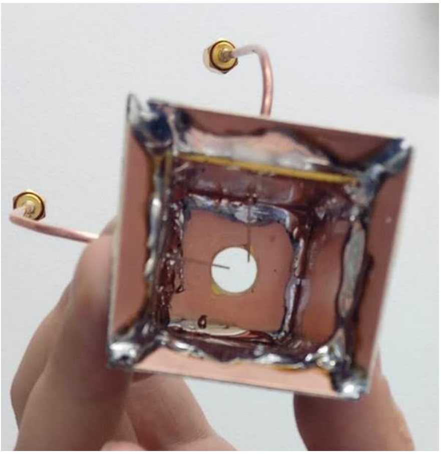

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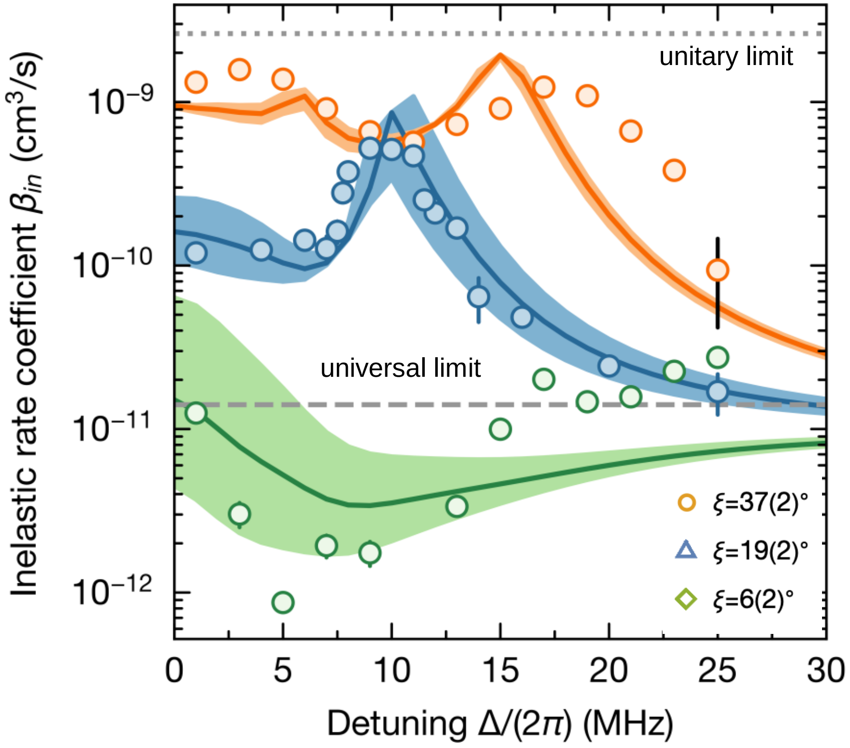
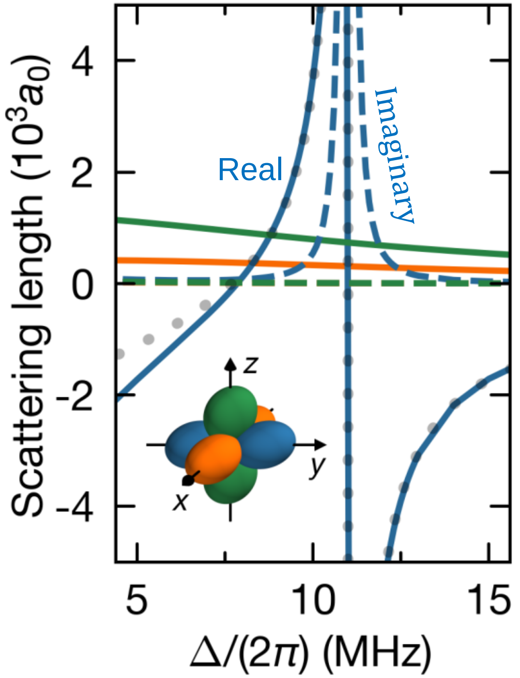


$$\Omega = 2\pi \times 10\ \text{MHz}, T = 230\ \text{nK}$$

Control of Scattering Length



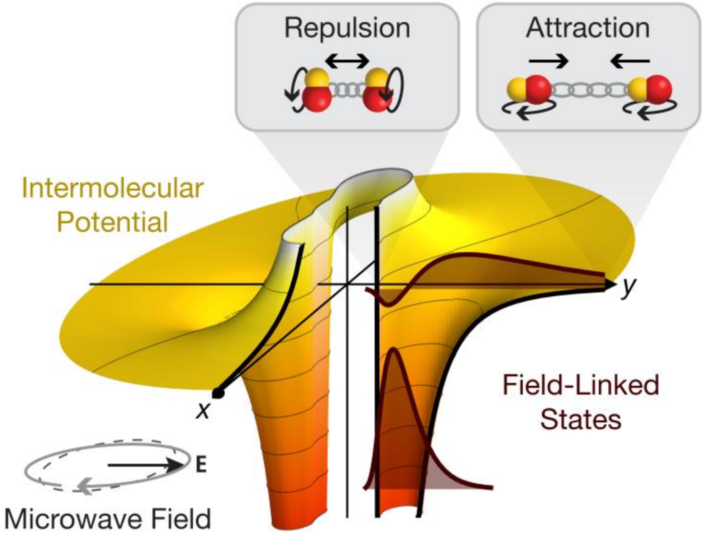
Xing-Yan Chen

Andreas Schindewolf

Chen, Schindewolf, Eppelt, Bause, Biswas, Duda, Karman, Hilker, Bloch & Luo Nature 614, 59 (2023)

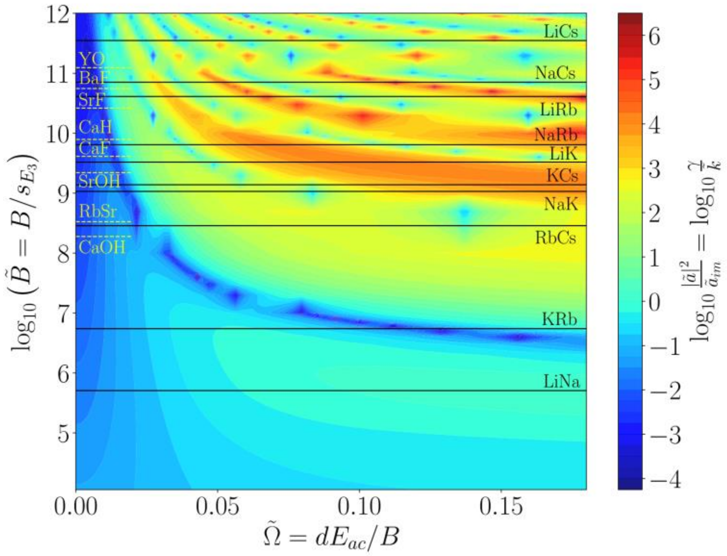
A NEW TYPE OF SCATTERING RESONANCE

Bound Induced by Electric field



Chen*, Schindewolf* et al., Nature **614**, 59 (2023)

Universal to Polar Molecules



Lassablière & Quéméner, PRL **121**, 163402 (2018)

Resonance	Field-linked	Feshbach
Tuning	Electric	Magnetic
Channel	Single	Two
Bound state size	~ 1000 a0	~ 40 a0
Bound state lifetime	Up to 100 ms	~ 1 μs
Dipole moment	~ 1.6 Debye	0 Debye
Apply to most UPMs?	Yes	No

Feshbach resonances (Ketterle):
Atoms: Nature 392, 151 (1998)
GS Molecules: Nature 614, 54 (2023)

CONTENT

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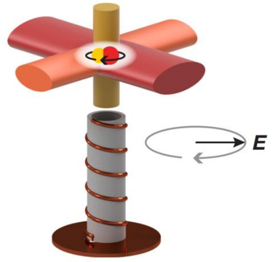

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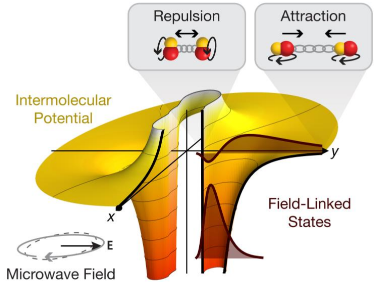
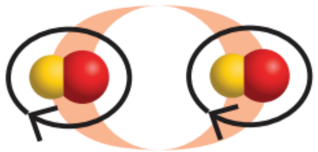


3. **Ultracold field-linked tetratomic molecules**



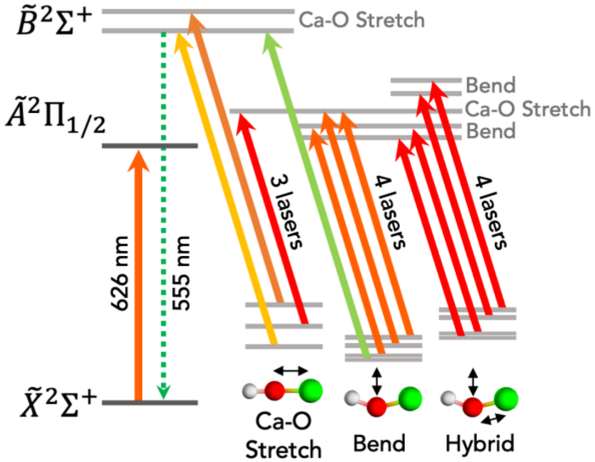
ULTRACOLD POLYATOMIC MOLECULES

Additional degrees of freedom for e.g., self-error-corrected qubit, edm…

However much more difficult to cool!

Laser Cooled Triatomic Molecules

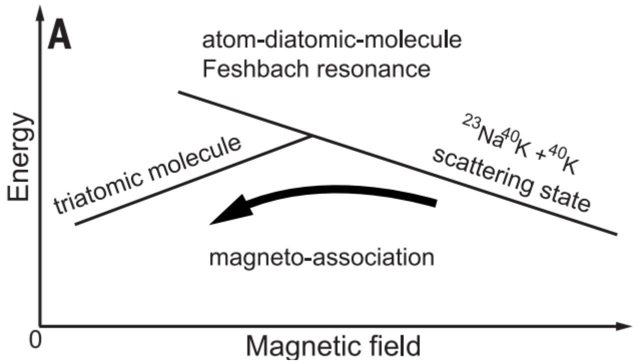
CaOH (20 μK), Doyle group



Vilas et al., Nature 606, 70 (2022)

Feshbach Triatomic Molecules

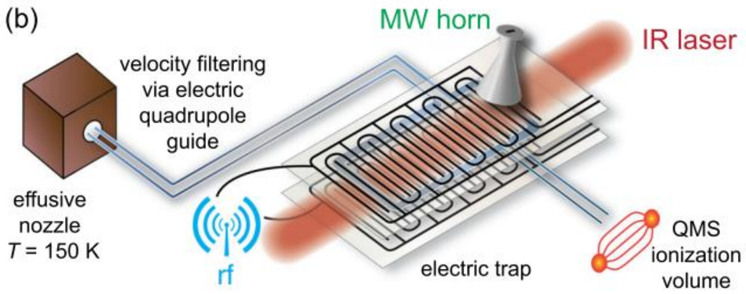
Na_2K (250 nK), Pan/Zhao group



Yang et al., Science 378, 1009 (2022)

Electro-Optic Cooling

H_2CO (420 μK), Rempe group



Prehn et al., PRL 116, 063005 (2016)

Direct cooling techniques to Kelvin regime

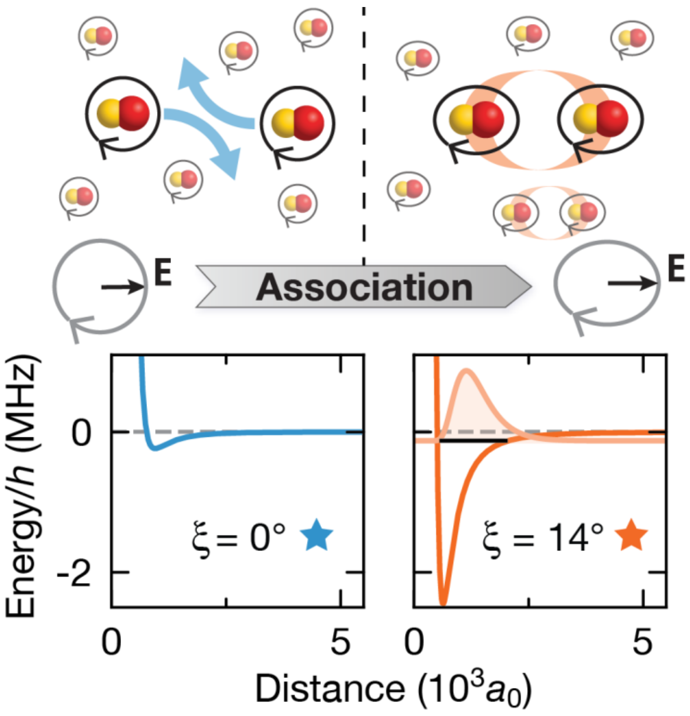
- **Buffer gas cooling**
- **Stark deceleration**
- **Cryofuge deceleration**

…

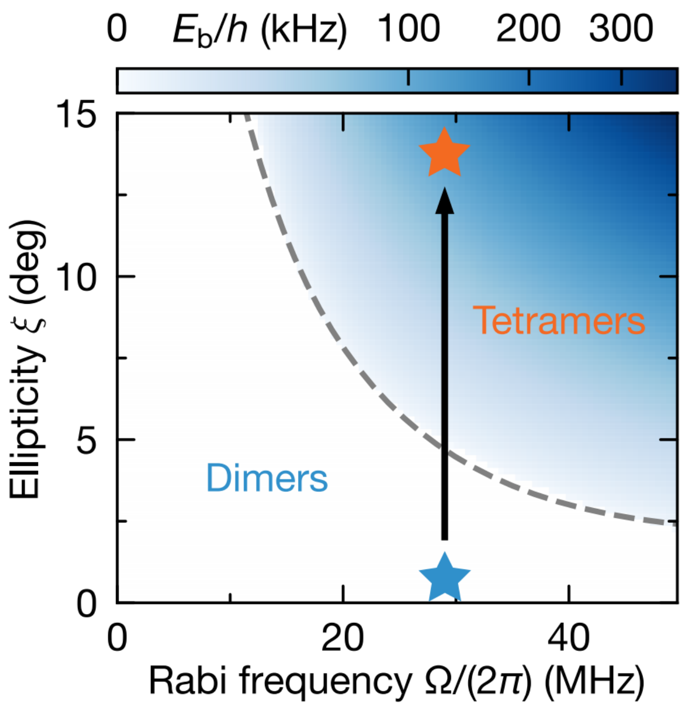
Ultracold Polyatomic Molecules for Quantum Science and Precision Measurements
Doyle et al., JPS Conf. Proc. 37, 011004 (2022)

ELECTROASSOCIATION OF THE FL TETRAMERS

Ramp the Ellipticity



Association Path



Theory:

Quéméner, Bohn & Croft, arXiv:2304.09525 (2023)

Deng et al., Formation and dissociation of field-linked tetramers (in preparation)

Chen, Biswas, Eppelt, Schindewolf, Deng, Shi, Yi, Hilker, Bloch & Luo arXiv:2306.00962 (2023)

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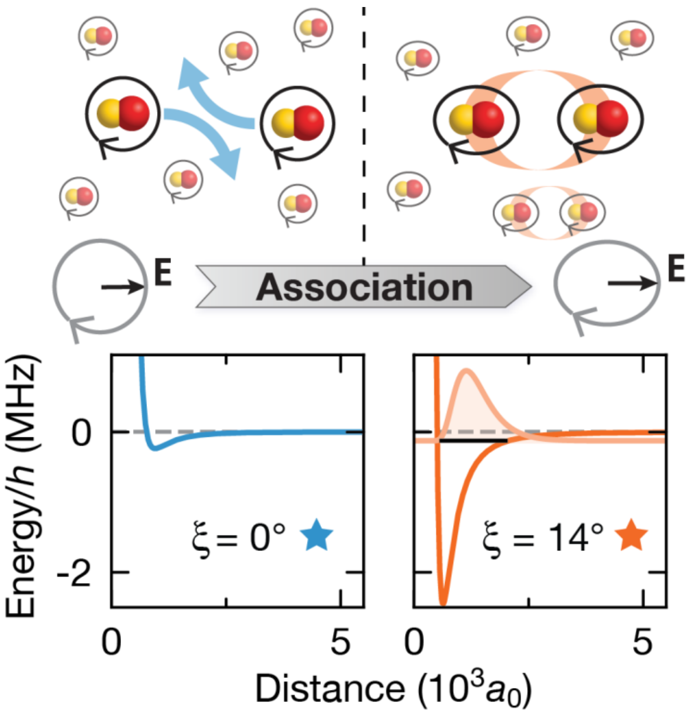

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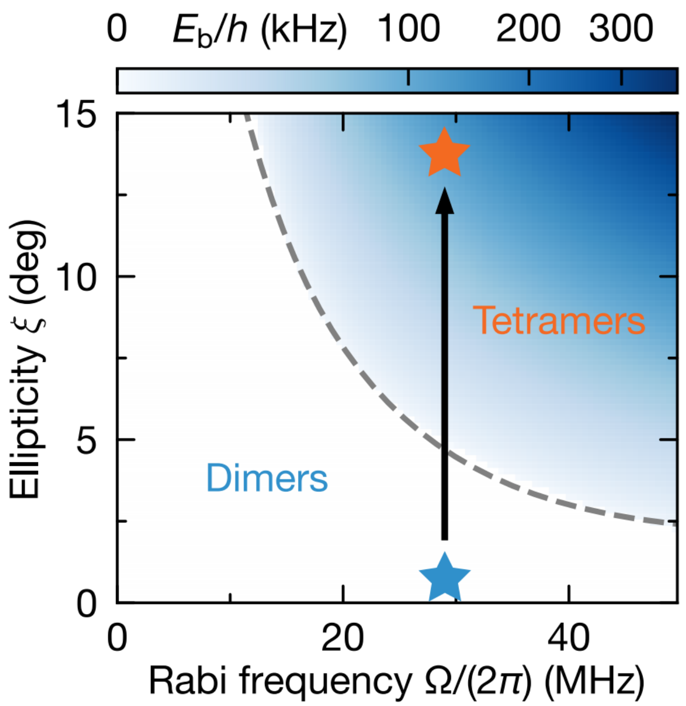
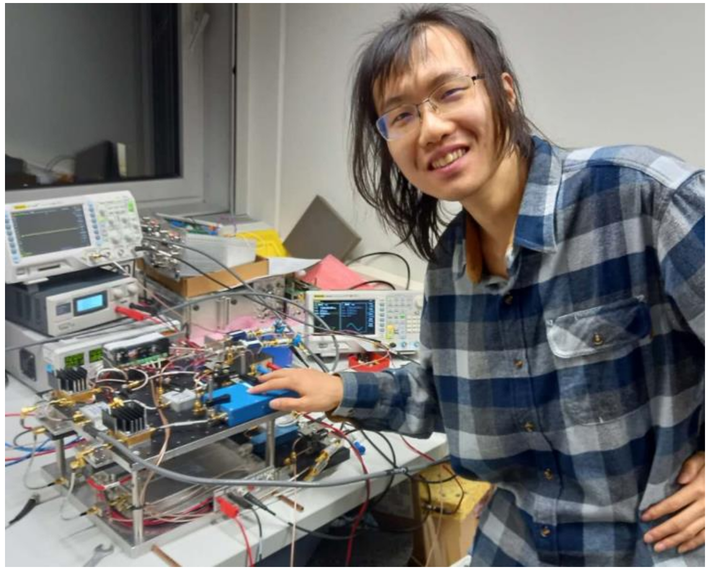


Xing-Yan's 2×100 Watt steampunk!

×10 more power!



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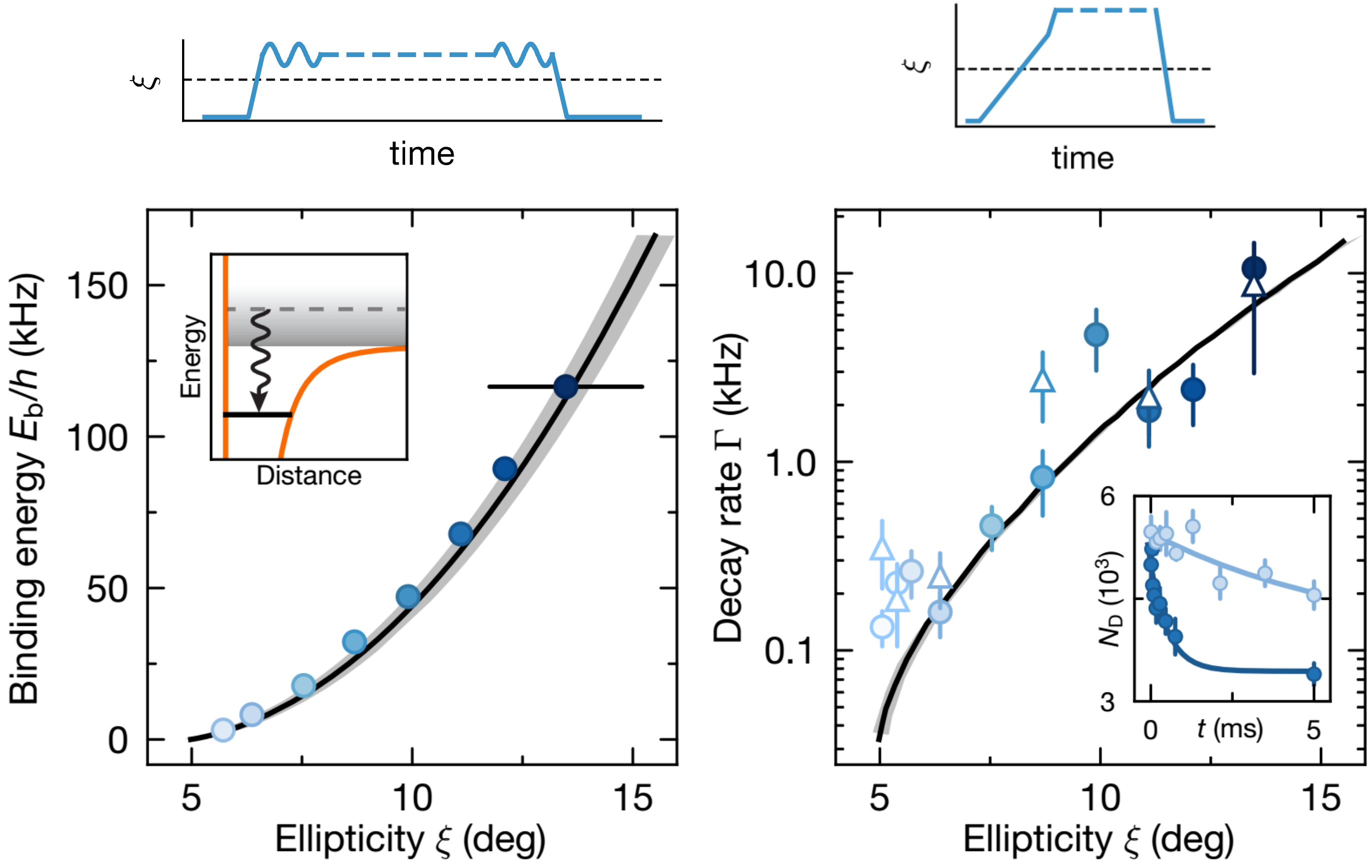
BINDING ENERGY AND LIFETIME OF TETRAMERS



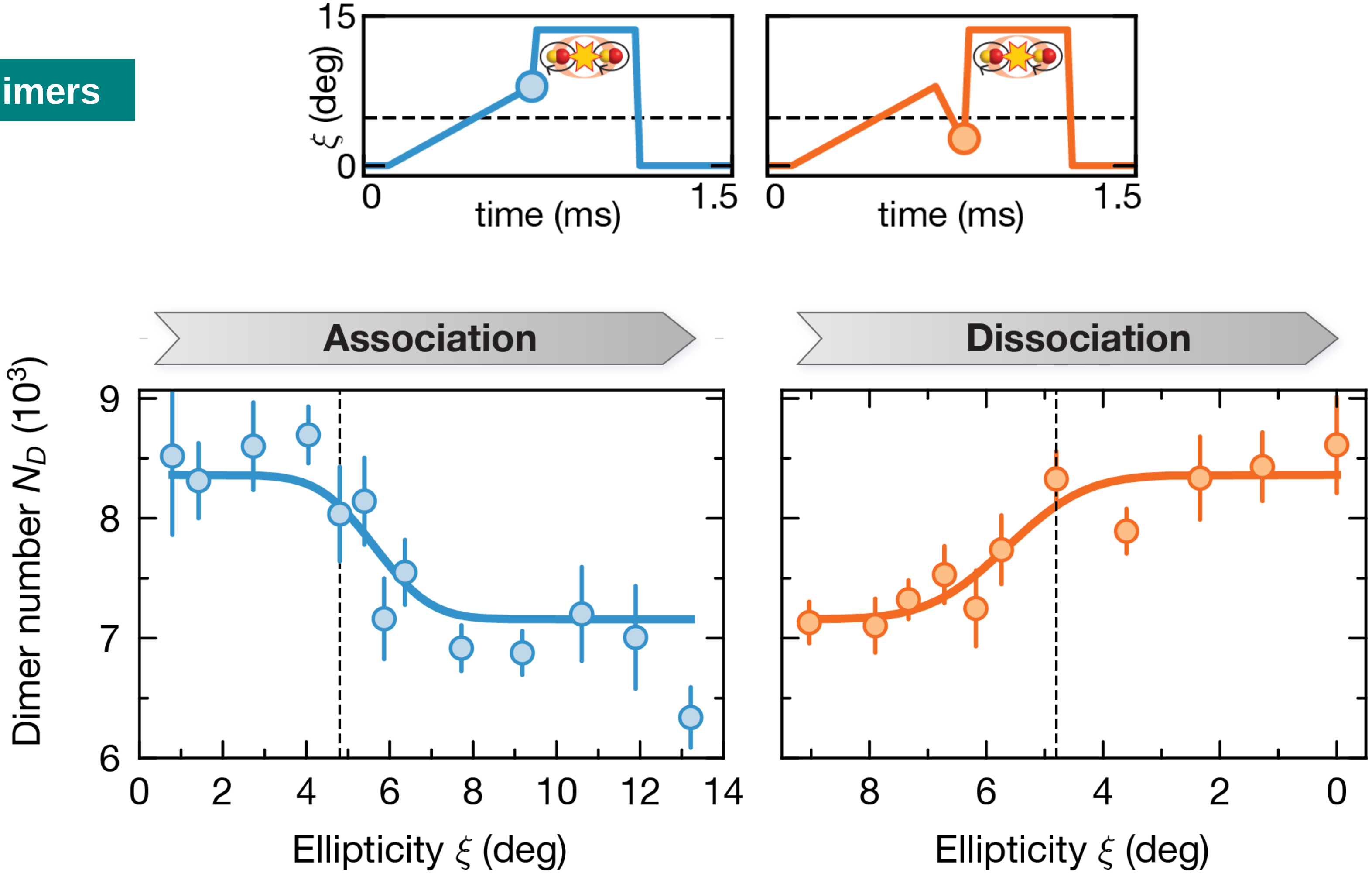
- Spontaneous dissociation
- Suggest collisionally stable
- 8(2) ms near threshold
- Expect >100 ms lifetime at circular polarization and 90 MHz Rabi frequency

Chen, Biswas, Eppelt, Schindewolf, Deng, Shi, Yi, Hilker, Bloch & Luo arXiv:2306.00962 (2023)

ASSOCIATION AND DISSOCIATION PROCESSES

Detect unpaired dimers

Loss & revival

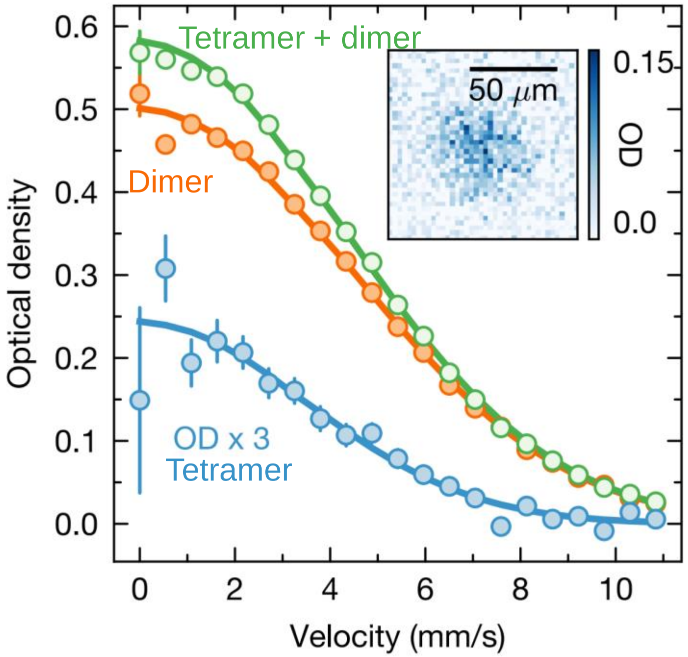


Chen, Biswas, Eppelt, Schindewolf, Deng, Shi, Yi, Hilker, Bloch & Luo arXiv:2306.00962 (2023)

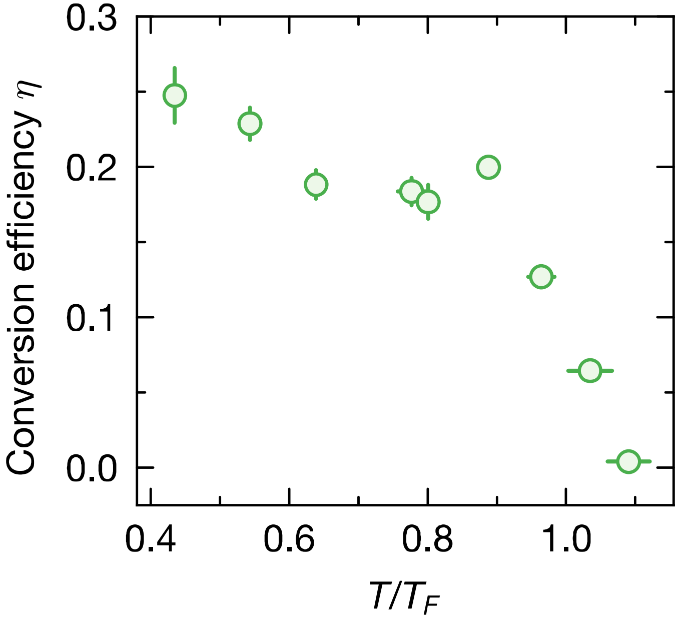
TEMPERATURE AND PHASE SPACE DENSITY

3000 times colder. PSD $\times 10^{11}$

T = 134(3) nK，　PSD = 0.040(3)



25% conversion efficiency at 0.44 T_F

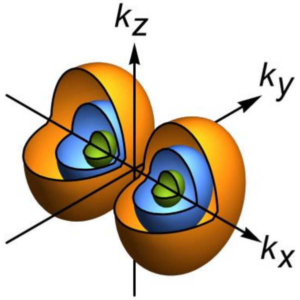


Chen, Biswas, Eppelt, Schindewolf, Deng, Shi, Yi, Hilker, Bloch & Luo arXiv:2306.00962 (2023)

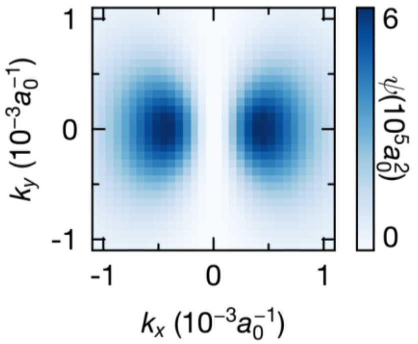
ANGULAR DISTRIBUTION OF TETRAMER WAVEFUNCTION

Tetramer Wavefunction in Momentum Space

3D distribution



Top view



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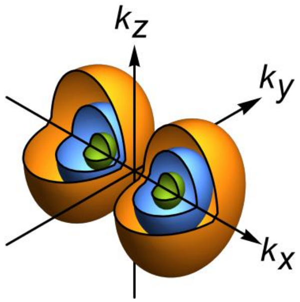

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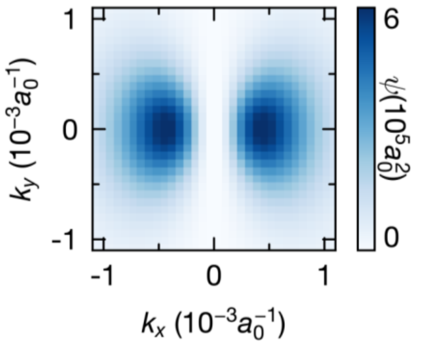
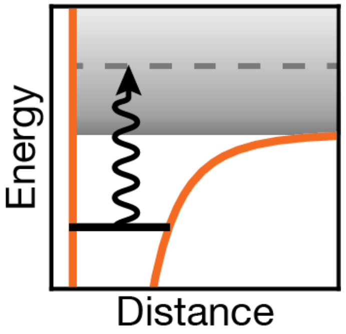


Bound-to-Free Transition



Inspired by

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

Chen, Biswas, Eppelt, Schindewolf, Deng, Shi, Yi, Hilker, Bloch & Luo arXiv:2306.00962 (2023)

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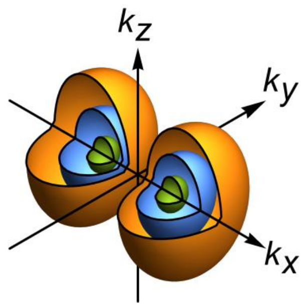
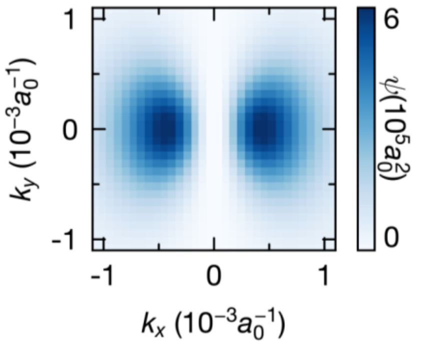


Top view



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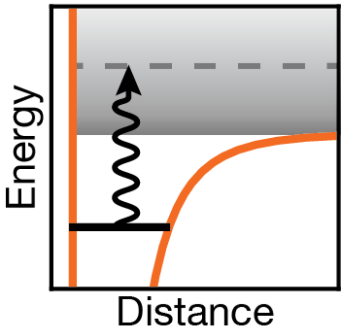
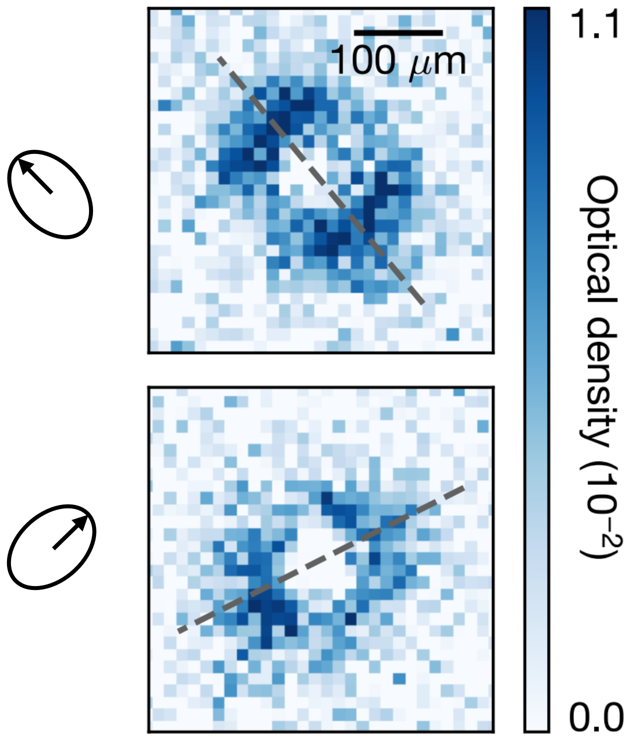


Inspired by

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

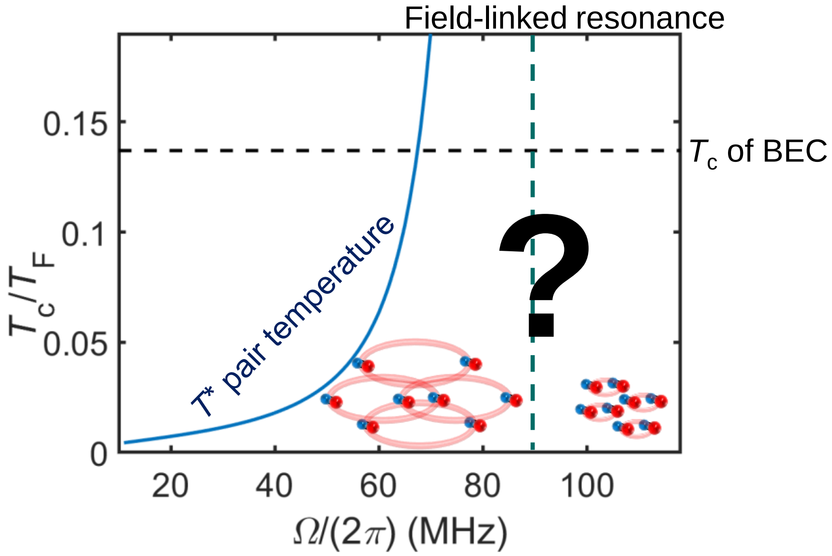
Momentum Distribution of Dissociated Tetramers



Chen, Biswas, Eppelt, Schindewolf, Deng, Shi, Yi, Hilker, Bloch & Luo arXiv:2306.00962 (2023)

OUTLOOK

1. Dipolar BCS-BEC Crossover



Deng et al., PRL 130, 183001 (2023)

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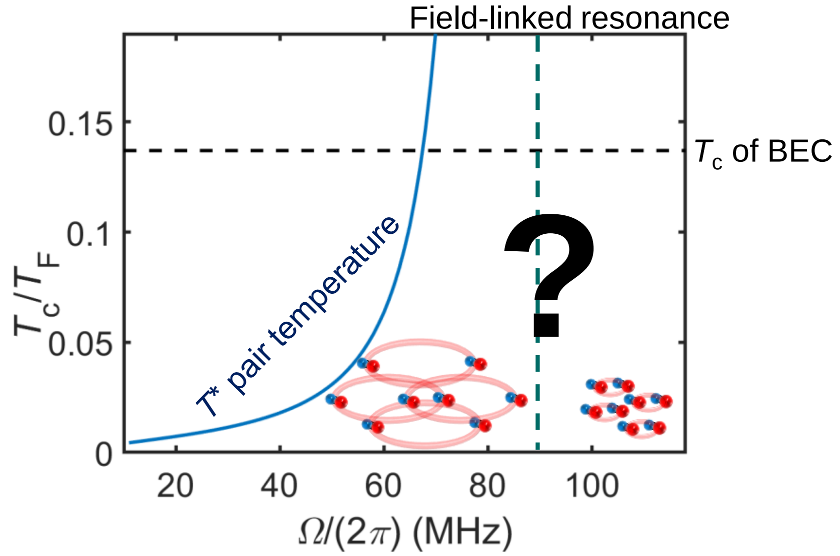
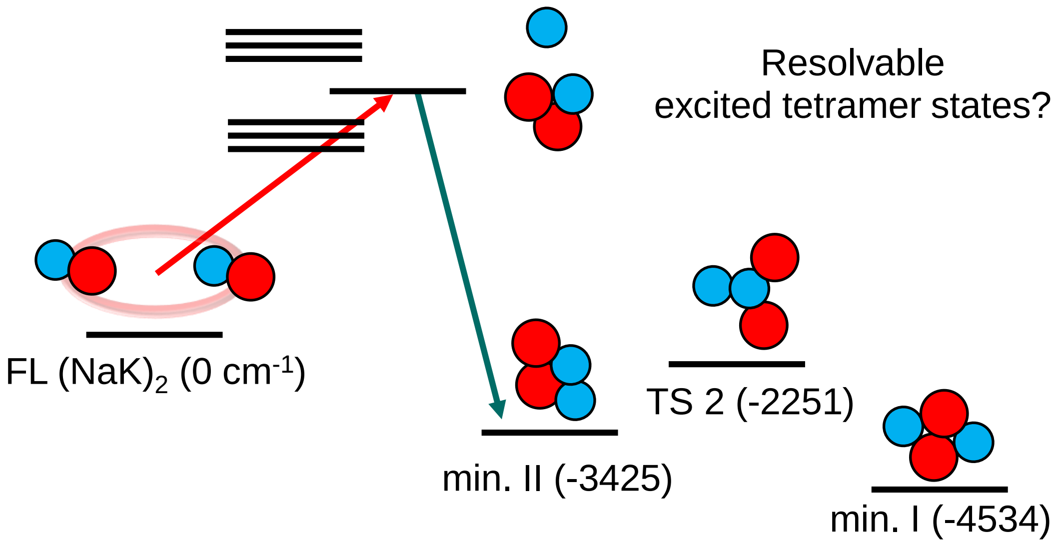


Deng et al., PRL 130, 183001 (2023)

2. Deeply Bound Tetramer States



Christianen, et al., J. Chem. Phys. 150, 064106 (2019)

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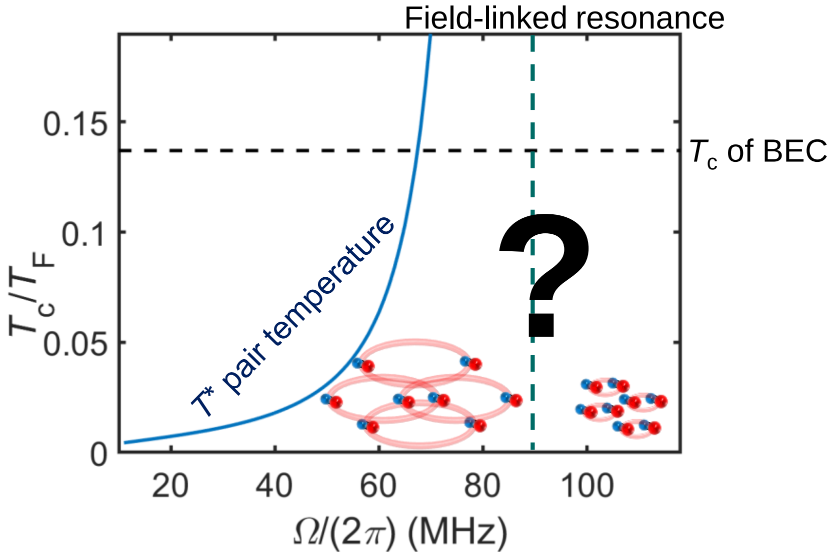

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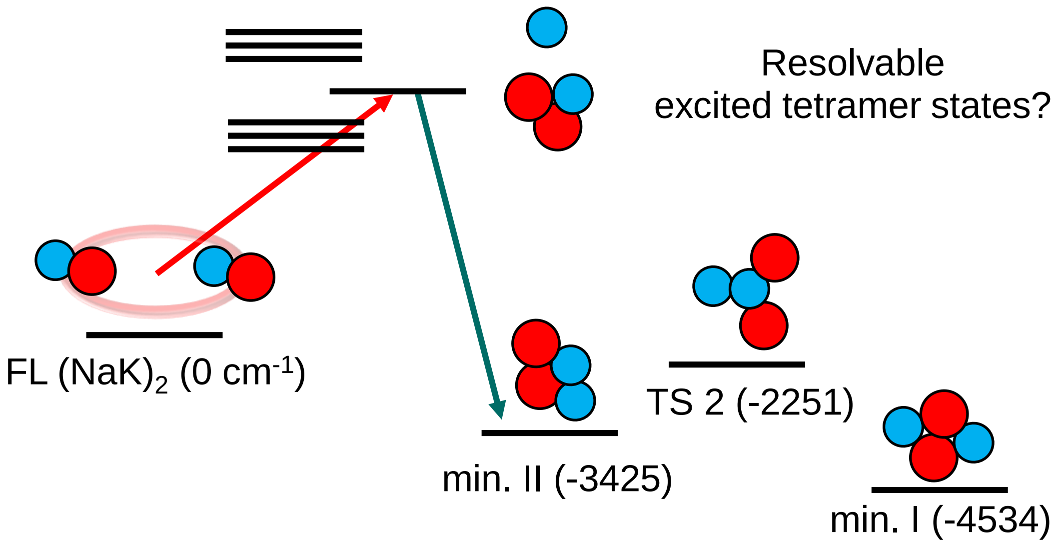
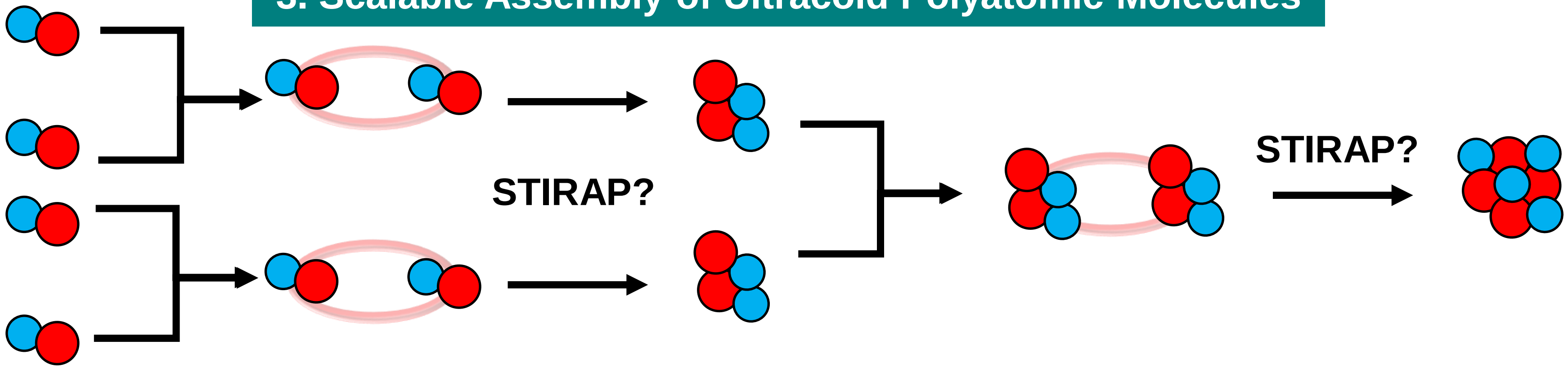


Christianen, et al., J. Chem. Phys. 150, 064106 (2019)

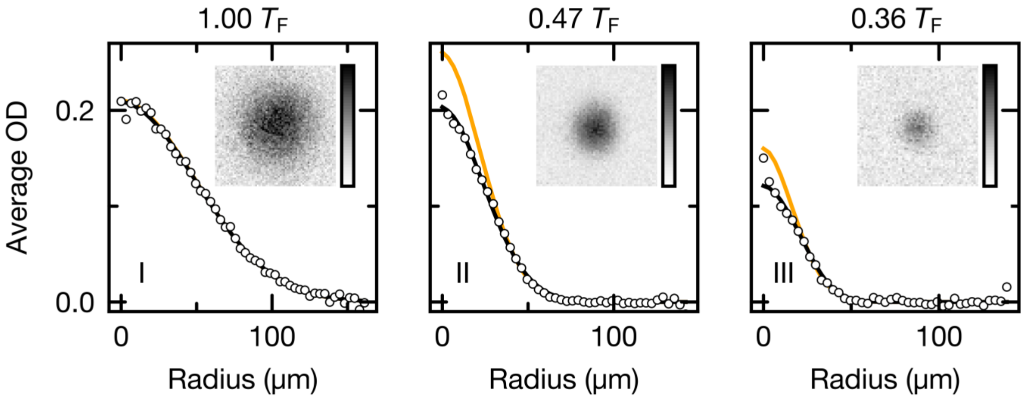
3. Scalable Assembly of Ultracold Polyatomic Molecules



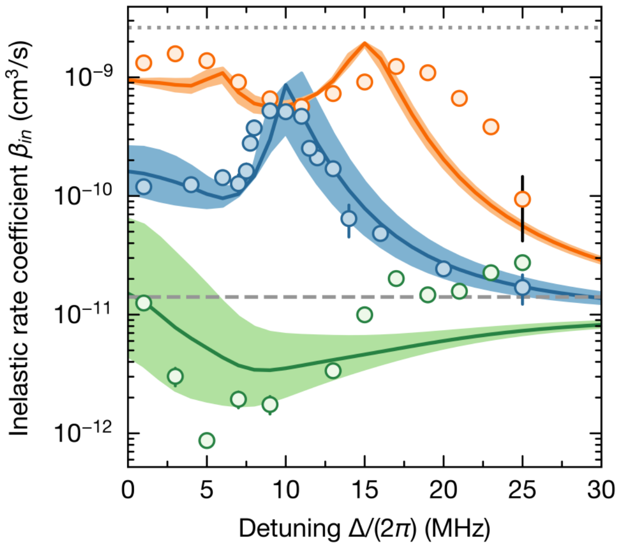
SUMMARY

- ✓ Stable dipolar Fermi gases of NaK molecules
- ✓ Resonant tuning of molecular interactions
- ✓ Assembled bosonic tetratomic molecules $(NaK)_2$
- ☐ Dipolar *p*-wave superfluid – Tetramer BEC?
- ☐ Scalable assembly?
- ☐ Spin models and extended Hubbard models

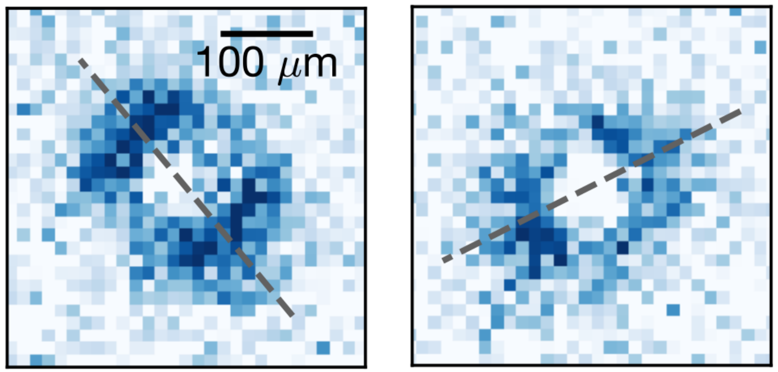
…



Schindewolf et al., Nature **607**, 677-681 (2022)



Chen*, Schindewolf*, et al., Nature **614**, 59 (2023)



Chen et al., arXiv:2306.00962 (2023)

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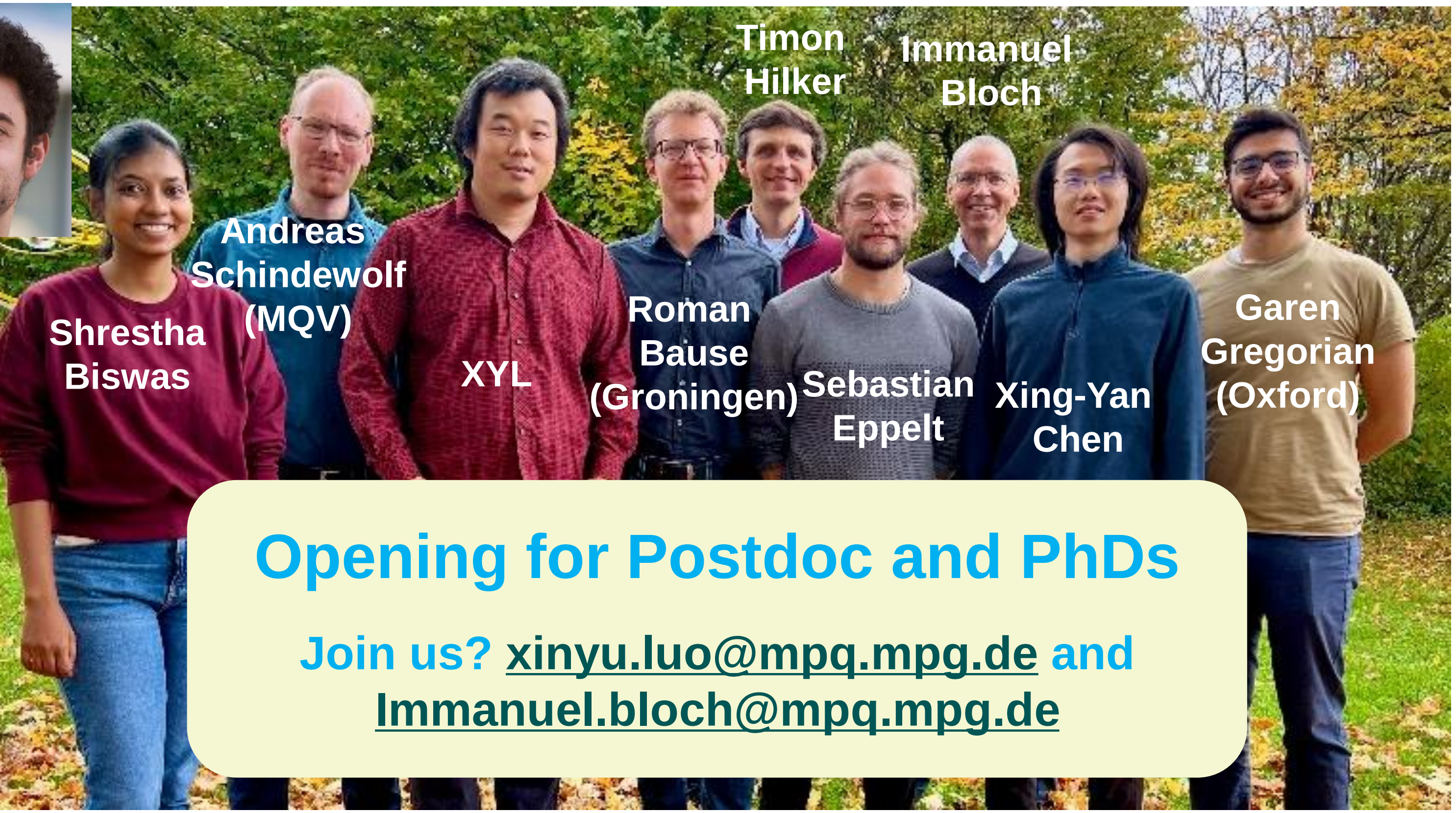

Former members:
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Svetlana Kotochigowa
Ming Li

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Richard Schmidt

Jonas von Milczewski

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Tijs Karman

Arthur Christianen

Goulven Quéméner

Field-linked tetramers

Tao Shi

Fulin Deng

Su Yi