Ultracold dipolar atoms and molecules part I

Dajun Wang
Department of Physics
The Chinese University of Hong Kong

CROUCHER SUMMER COURSE
"ULTRACOLD ATOM PHYSICS"

May 20 – 24, 2024

Supported by RGC Hong Kong

Outline

Motivations

Interaction is the most important parameter in ultracold quantum gases

Atoms and molecules with dipole-dipole interactions (DDI) allow us to access physics beyond that provided by atoms with only short-range effective contact interaction

Dipoles and dipole-dipole interaction

Experiments with magnetic atoms

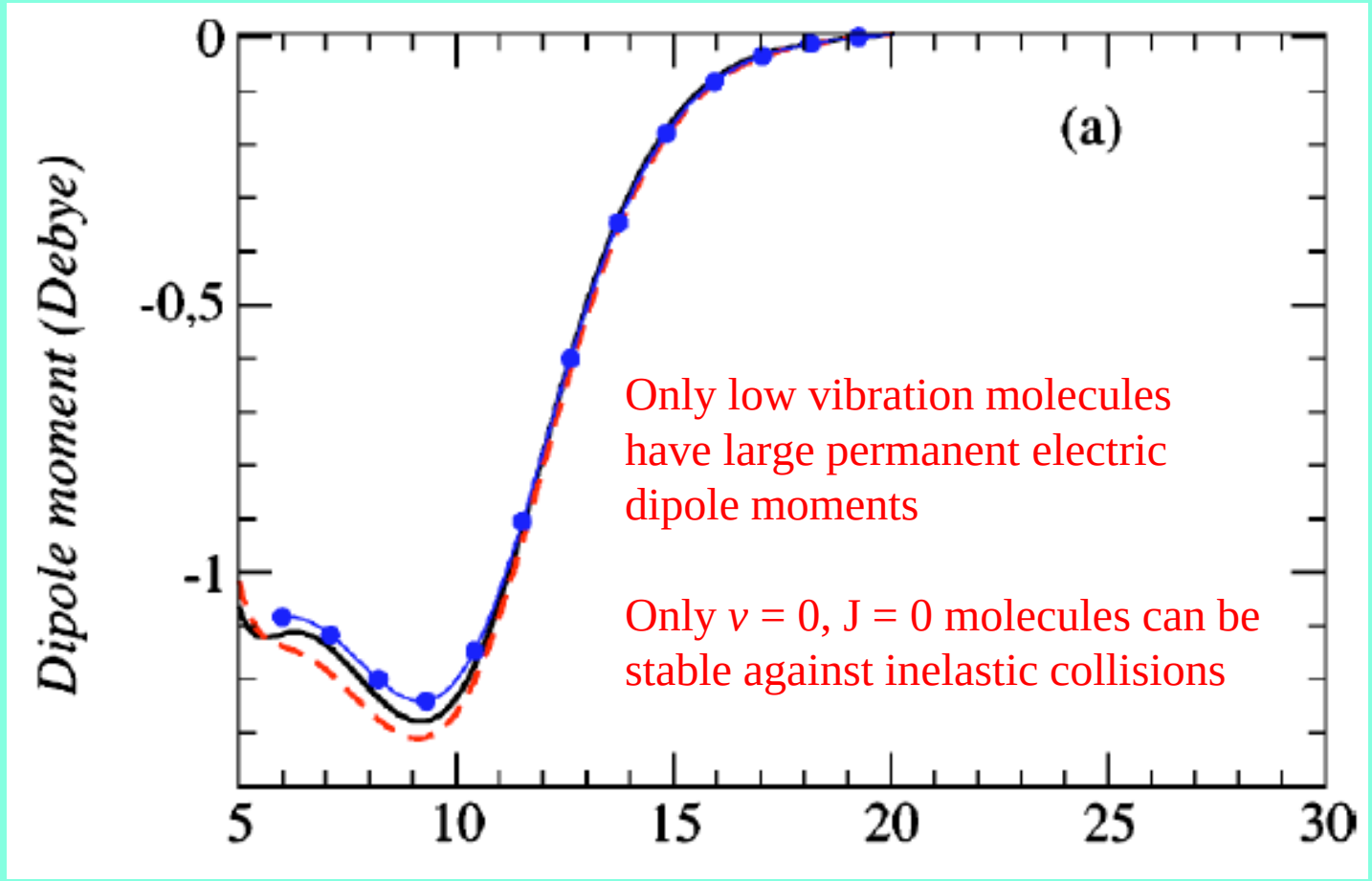
Ultracold polar molecules

Rep. Prog. Phys. ***72***, 126401 (2009)
Rep. Prog. Phys. ***86***, 026401 (2023)

Cold and Ultracold Molecules

Insight issue *Nature Physics (2024)*

Dipoles



Only low vibration molecules have large permanent electric dipole moments

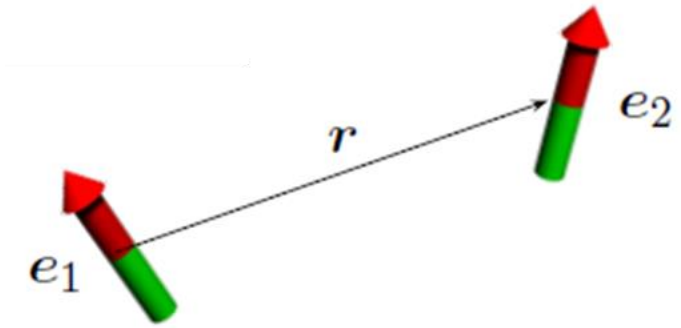
Only $v = 0$, J = 0 molecules can be stable against inelastic collisions

μ: Bohr magneton ($\mu_B = \frac{}{2m_e}$)

d: C·m; most often: atomic unit (ea_0) or Debye ($statC \cdot cm$)

$1D \simeq 3.3 \times 10^{-30}$C·m $\simeq 0.39ea_0$

Dipole-dipole interaction



In general:

$$U_{dd}(\mathbf{r}) = \frac{C_{dd}}{4\pi}\left(\frac{\hat{e}_1 \cdot \hat{e}_2 - 3(\hat{e}_1 \cdot \hat{\mathbf{r}})(\hat{e}_2 \cdot \hat{\mathbf{r}})}{r^3}\right)$$

$C_{dd} = \mu_0\mu^2$ for magnetic dipole moment μ

$C_{dd} = \frac{d^2}{\varepsilon_0}$ for electric dipole moment d

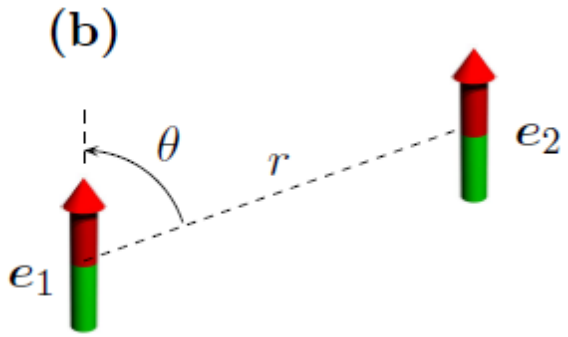
$\frac{\mu_0\mu_B^2}{(e\,a_0)^2/\varepsilon_0} = \alpha^2/4 = \left(\frac{1}{2\times 137}\right)^2$: DDI is much stronger in the electric case

Unit of dipole moment:

μ: Bohr magneton ($\mu_B = \frac{e\hbar}{2m_e}$)

d: C·m; most often:
atomic unit (ea_0)
or Debye ($statC \cdot cm$)
$1D \simeq 3.3 \times 10^{-30}$C·m $\simeq 0.39ea_0$

Dipole-dipole interaction: anisotropy



In general:

$$U_{dd}(\mathbf{r}) = \frac{C_{dd}}{4\pi}\left(\frac{\hat{e}_1 \cdot \hat{e}_2 - 3(\hat{e}_1 \cdot \hat{\mathbf{r}})(\hat{e}_2 \cdot \hat{\mathbf{r}})}{r^3}\right)$$

$C_{dd} = \mu_0 \mu^2$ for magnetic dipole moment μ

$C_{dd} = \frac{d^2}{\varepsilon_0}$ for electric dipole moment d

For polarized dipoles:

$$U_{dd}(\mathbf{r}) = \frac{C_{dd}}{4\pi}\left(\frac{1-3\cos^2\theta}{r^3}\right)$$

θ: 0 to $\pi/2$

$1-3\cos^2\theta$: -2 to 1

Long range: $1/r^3$
Anisotropic
Tunable with external (E or B) field and trap geometry

Rep. Prog. Phys. ***72***, 126401 (2009)
Rep. Prog. Phys. ***86***, 026401 (2023)

Dipole-dipole interaction



Can be realized with a pancake trap



Can be realized with a cigar trap

In general:

$$U_{dd}(\mathbf{r}) = \frac{C_{dd}}{4\pi}\left(\frac{\hat{e}_1 \cdot \hat{e}_2 - 3(\hat{e}_1 \cdot \hat{\mathbf{r}})(\hat{e}_2 \cdot \hat{\mathbf{r}})}{r^3}\right)$$

$C_{dd} = \mu_0\mu^2$ for magnetic dipole moment μ

$C_{dd} = \frac{d^2}{\varepsilon_0}$ for electric dipole moment d

For polarized dipoles:

$$U_{dd}(\mathbf{r}) = \frac{C_{dd}}{4\pi}\left(\frac{1 - 3\cos^2\theta}{r^3}\right)$$

Special cases:

side to side (θ=90^0) pure repulsive,

head to tail (θ=0^0) pure attractive

vanishes at the "magic angle" (θ=54.7^0)

Rep. Prog. Phys. ***72***, 126401 (2009)
Rep. Prog. Phys. ***86***, 026401 (2023)

Dipolar scattering

Ultracold collisions: the centrifugal barrier $\frac{2l(l+1)\hbar^2}{m\,r^2}$ prevents collisions via higher partial waves $l > 0$



For a $\frac{1}{r^n}$ potential, the scattering phase shift (with $k \to 0$ the relative momentum)

$$\delta_l(k) \propto \begin{cases} k^{2l+1} & \text{for } l < (n-3)/2 \\ k^{n-2} & \text{otherwise} \end{cases}$$

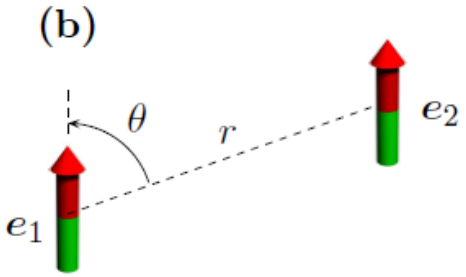
So, for $n \geq 4$, only s-wave and short-ranged.

However, for DDI with $n = 3$, $\delta_l(k) \propto k$ for all partial waves.

All partial waves can contribute to the scattering process. DDI is long-range and can be felt beyond the centrifugal barrier.

Example effect: identical fermionic dipolar gases can still thermalize despite the absence of s-wave interactions

Dipolar scattering

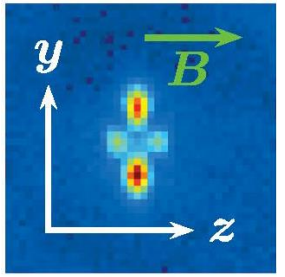


For polarized dipoles:

$$U_{dd}(\mathbf{r}) = \frac{C_{dd}}{4\pi}\left(\frac{1-3\cos^2\theta}{r^3}\right)$$

Angular symmetry: $Y_{20}(\cos\theta)$, i.e. d-wave;

Dipolar force is non-central.



T. Pfau group
PRL 101, 080401 (2008)

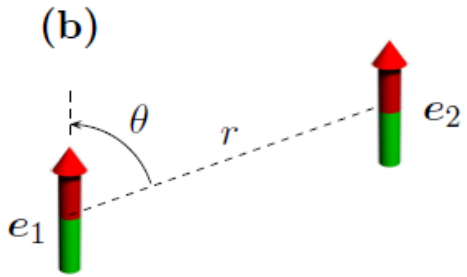
- ✓ Individual AM (l or s) is not conserved, but $j = l + s$ is: dipolar relaxation of spin
- ✓ U_{dd} can couple to higher partial waves

Scattering matrix $\sim \int Y_{l'm'} Y_{20} Y_{lm} d\Omega$

So $l' + l + 2$ must be even, m′=m;

$U_{dd}(\mathbf{r})$ can couple to even (for boson) and odd (for fermion) partial waves

Characterise DDI



For polarized dipoles:

$$U_{dd}(\mathbf{r}) = \frac{C_{dd}}{4\pi}\left(\frac{1-3\cos^2\theta}{r^3}\right)$$

Characteristic length (dipolar length):

$$a_{dd} = \frac{C_{dd}m}{12\pi\hbar^2}$$

Dipolar length:

$$\left(-\frac{\hbar^2}{m}\nabla^2 + U_{dd}(\boldsymbol{r})\right)\psi(\boldsymbol{r}) = E\psi(\boldsymbol{r})$$

$$\frac{\hbar^2}{m\,r_d^2} = \frac{d^2}{r_d^3} \Rightarrow a_{dd} = \frac{r_d}{3} = \frac{C_{dd}m}{12\pi\hbar^2}$$

Universal scattering cross section:

$$\sigma_d \propto a_{dd}^2 \propto (md^2)^2$$

For dipolar bosons:
Both the long-range and the short-range parts of the interaction.

The ratio between DDI to contact interaction:
$\varepsilon_{dd} = \frac{a_{dd}}{a}$ is another important characteristic quantity

For dipolar fermions:
Only by the long-range dipolar part of the interaction.

Rep. Prog. Phys. ***72***, 126401 (2009)
Rep. Prog. Phys. ***86***, 026401 (2023)

Characterise DDI

Characteristic length (dipole length):

$$a_{dd} = \frac{C_{dd} m}{12\pi\hbar^2}$$

Also, the ratio of DDI to contact interaction

$$\varepsilon_{dd} = \frac{a_{dd}}{a}$$

Species	Dipole moment	$a_{dd}(a_0)$	a (a_0)
^{87}Rb	1 μ_B	0.7	100
^{52}Cr	6 μ_B	16	~100
^{168}Er	7 μ_B	67	~175
^{164}Dy	10 μ_B	130	~100
KRb	0.6 D	3700	
NaK	2.7 D	43180	
NaRb	3.3 D	112700	~ 200
NaCs	4.7 D	324000	

Rep. Prog. Phys. ***72***, 126401 (2009)
Rep. Prog. Phys. ***86***, 026401 (2023)

Dipolar pseudo-potential: for bosons

For s−wave effective contact interaction,
the only parameter is the scattering length a

$$U_{contact}(\vec{r}) = \frac{4\pi\hbar^2 a}{m}\delta(\vec{r}) \qquad g = \frac{4\pi\hbar^2 a}{m}$$

- ✓ U_{dd} can couple to higher partial waves
- ✓ Modify the short-range s-wave $U_{contact}$

Pseudo−potential including DDI:

$$V(\vec{r}) = \frac{4\pi\hbar^2 a(d)}{m}\delta(\vec{r}) + \frac{C_{dd}}{4\pi\, r^3}(1 - 3\cos^2\theta)$$

$$g = \frac{4\pi\hbar^2 a(d)}{m}$$

The s-wave scattering could be modified by DDI for large ε_{dd}

Dipolar Gross−Pitaevskii equation (GPE)

$$i\hbar\frac{\partial\psi}{\partial t} = \left(-\frac{\hbar^2}{2m}\nabla^2 + U_{ext} + g|\psi|^2 + \int dr'\, U_{dd}(r - r')|\psi(r')|^2\right)\psi$$

S. Yi, L. You, *Phys. Rev. A*, **61**, 041604(2000).
A. Derevianko, *Phys. Rev. A* **67**, 033607(2003).
A. Derevianko, *Phys. Rev. A* , **72**, 039901(2005).
S. Ronen, D. Bortolotti, D. Blume, J.L. Bohn, *Phys. Rev. A*, **74**, 033611(2006).

Stability for a homogeneous dBEC

Bogoliubov spectrum $$\hbar\omega(\boldsymbol{k}) = \sqrt{\frac{\hbar^2k^2}{2m}\left(\frac{\hbar^2k^2}{2m} + 2gn(1 + \varepsilon_{dd}(3\cos^2\theta_k - 1))\right)}$$

s-wave Sound velocity: $c_0 = \sqrt{gn/m}$

Anisotropic sound velocity with DDI: $c(\theta_k) = c_0\sqrt{1 + \varepsilon_{dd}(3\cos^2\theta_k - 1)}$

$\theta_k = 0, along\ the\ dipole$: highest $c(\theta_k)$ (stiff modes)
$\theta_k = \pi/2\,,\ perpendicularto\ the\ dipole$: lowest $c(\theta_k)$ (softer modes)

For $\theta_k = \pi/2$, $c(\theta_k) = 0$ at $\varepsilon_{dd} = 1$

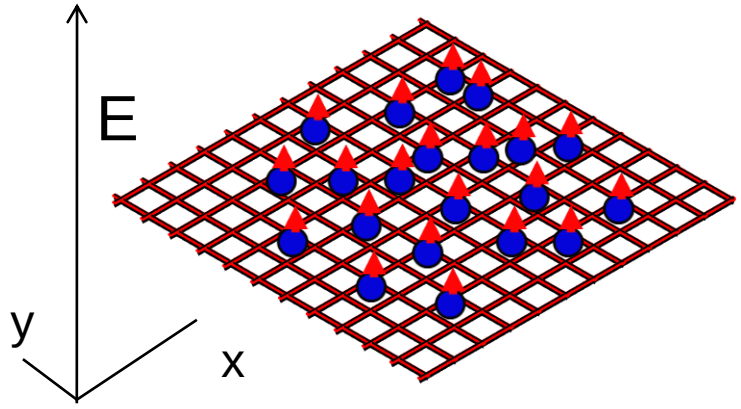
A homogeneous BEC is unstable for $$\varepsilon_{dd} = \frac{a_{dd}}{a} \geq 1$$

Phys. Rev. Lett. **85** 1791(2000)

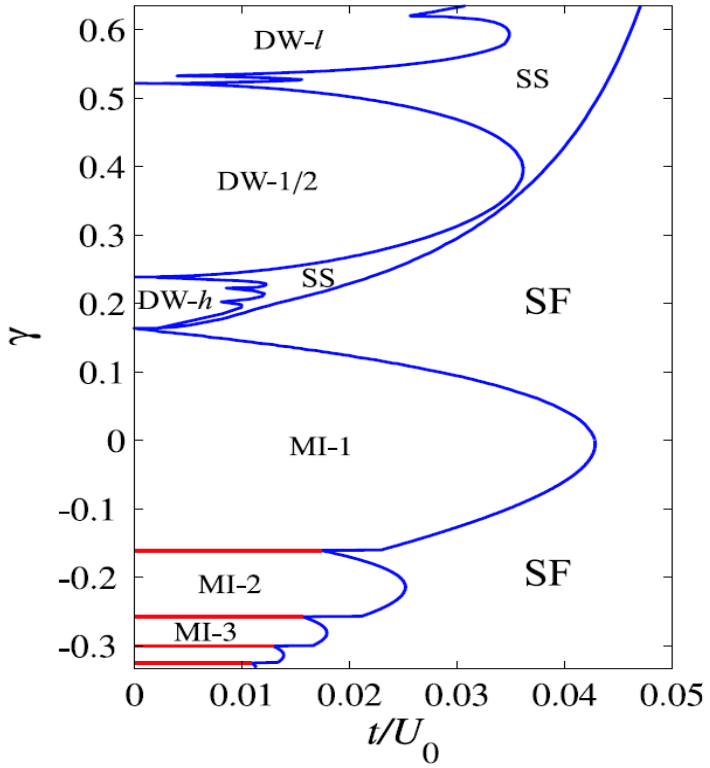
Theory work: many

Dipolar collisions
Dipolar quantum gas
Ultracold chemistry
Quantum information
Precision measurement
Dipolar quantum phase
Synthetic gauge field……

D. DeMille, T. Pfau, Li You, Su Yi, L. Santos, L. Carr, Lewenstein, Zoller, Büchler, Lukin, H. Pu, Baranov, Meystre, G. Shlyapnikov, Kurizki, Bohn, R. Krems, T.L. Ho, H. Zhai …



Long-term goal of our group



$\gamma \propto \varepsilon_{dd}$

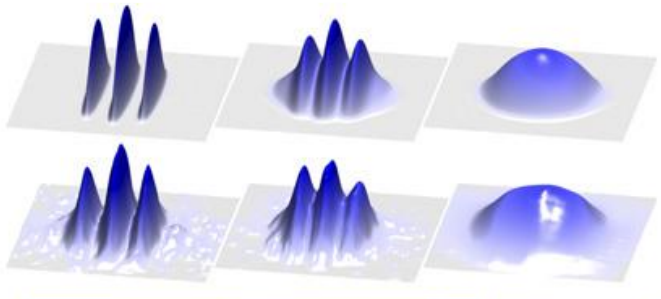
$\gamma > 0$ for electric dipole in DC field

S. Yi, C.P. Sun et al., PRL **98**, 260405 (2007)
Similar work: Capogrosso-Sansone, et al. PRL **104**, 125301(2010).

Ultracold dipolar systems

Magnetic atoms:

Magnetic dipole moment

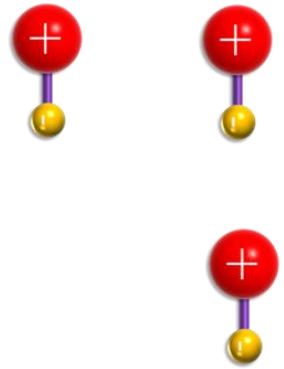


Rep. Prog. Phys. 84 012403 (2021)

- Cr (6 μ_B), Er (7 μ_B), Dy (10 μ_B)…
- Long lifetime
- BEC and DFG

Polar Molecules:

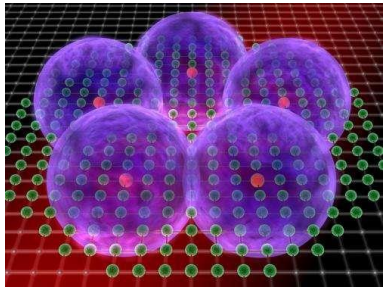
Electric dipole moment



- Many species available now: bi-alkali, CaF, SrF, YO…
- d ~ 1 to several Debye; tunable
- Long lifetime? Complicated by two-body interactions
- DFG 2019, BEC 2023

Rydberg atoms:

Giant electric dipole moment



Nature 491, 87 (2012)

- d ~ 1000 Debye
- Blockade mechanism
- Short lifetime: ~ 100 μs
- Tweezer array

Experiment: rapid progress

Magnetic atoms:

Cr, Dy, Er, Ho, Eu

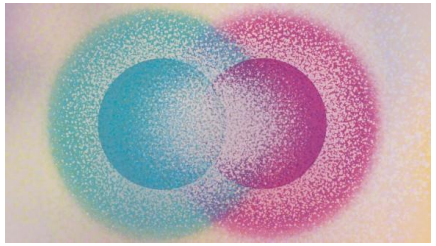
Rep. Prog. Phys. ***86***, 026401 (2023)

Polar molecules by cold atom association:

KRb, RbCs, NaK, NaRb, NaCs, LiK, LiNa…

Cold and Ultracold Molecules

Insight issue *Nature Physics (2024)*



Polar molecules by direct laser cooling:

SrF, CaF, YO, CaOH…

Outline

Dipole and dipole-dipole interaction

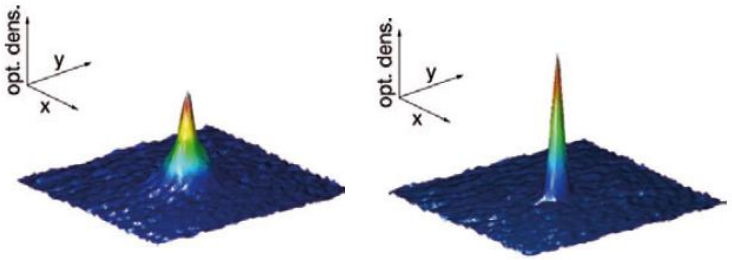
Experiments with magnetic atoms

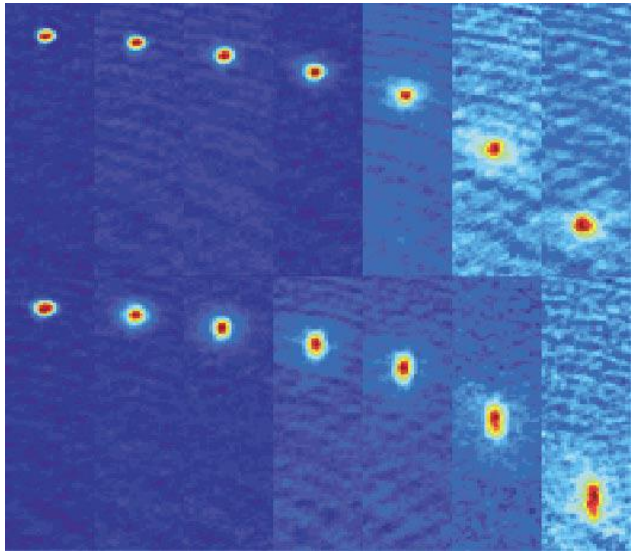
Ultracold polar molecules

^{52}Cr BEC: first ultracold dipolar quantum gas

A. Griesmaier et al., "Bose-Einstein condensation of chromium"
Phys. Rev. Lett. **94**, 160401 (2005)

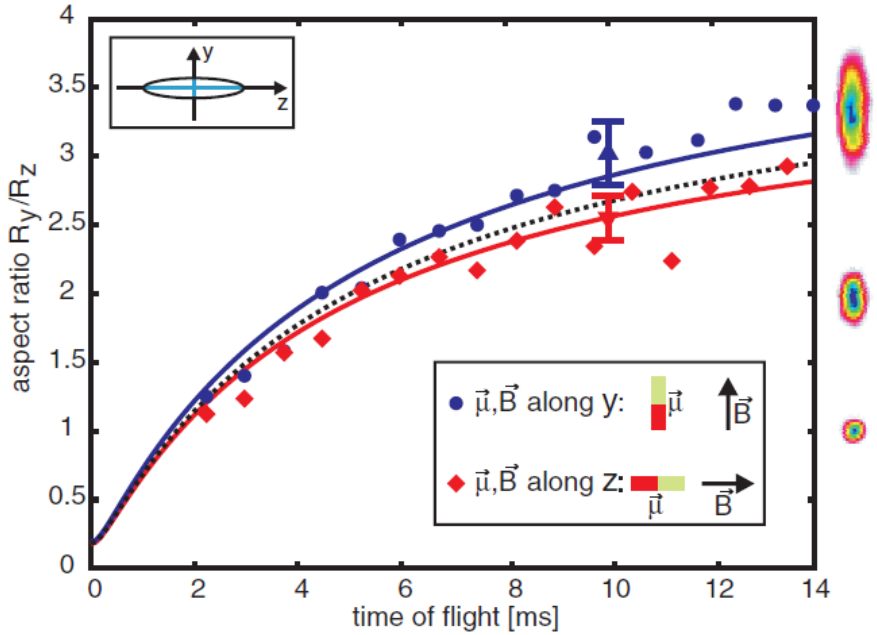
J. Stuhler, et al. "Observation of Dipole-Dipole Interaction in a Degenerate Quantum Gas"
Phys. Rev. Lett. **95**, 150406 (2005)







Tilman Pfau
in Stuttgart

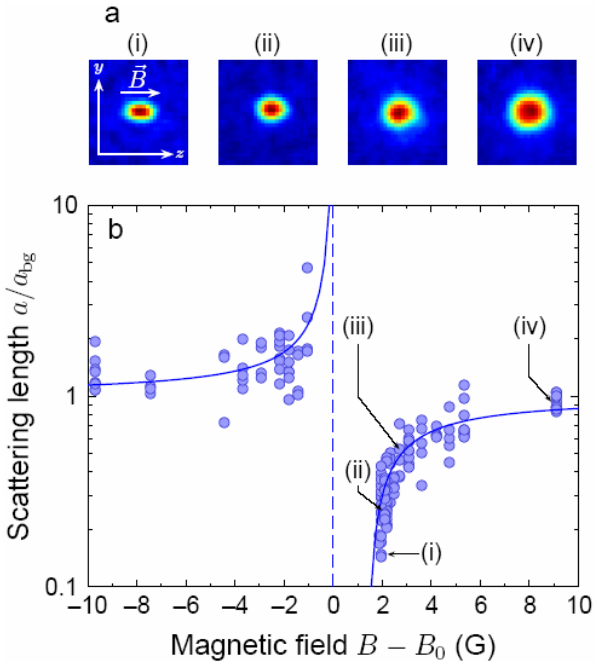


First evidence of DDI: Anisotropic expansion

^{52}Cr BEC: enhance the dipolar effect with Feshbach resonance

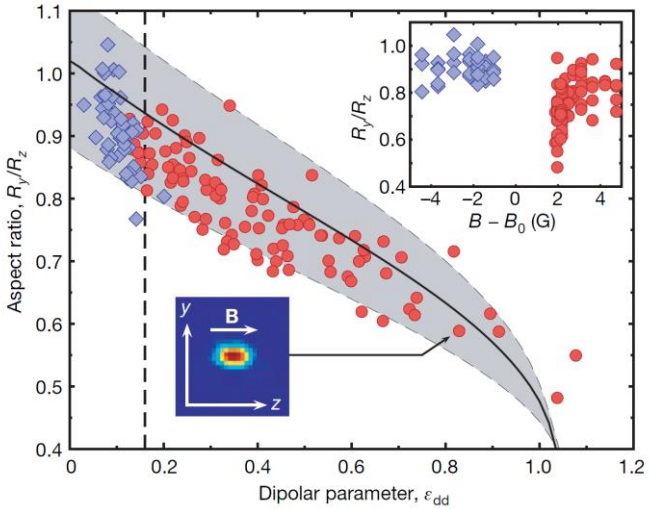
For Cr

$$\varepsilon_{dd} = \frac{a_{dd}}{a} \approx \frac{16a_0}{100a_0} = 0.16$$



Control ε_{dd} by tuning a

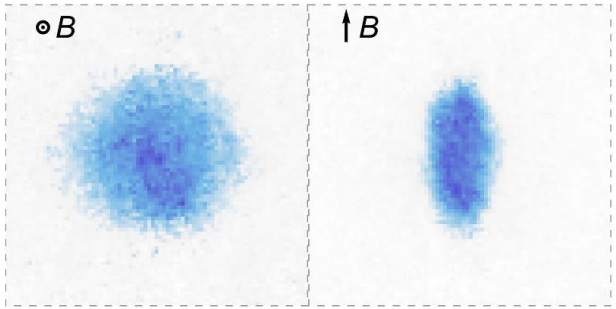
Tilman Pfau group
Nature **448**, 672 (2007)



Effect of increasing the dipolar parameter.

For Dy

$$\varepsilon_{dd} = \frac{a_{dd}}{a} \approx \frac{130a_0}{100a_0} = 1.3$$



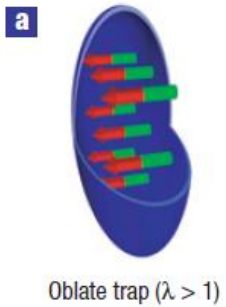
In situ imaging of magnetostriction of a 162Dy dBEC in a pancake-shaped trap.

^{52}Cr BEC: stability vs. trap geometry

$$\varepsilon_{dd} = \frac{a_{dd}}{a}$$

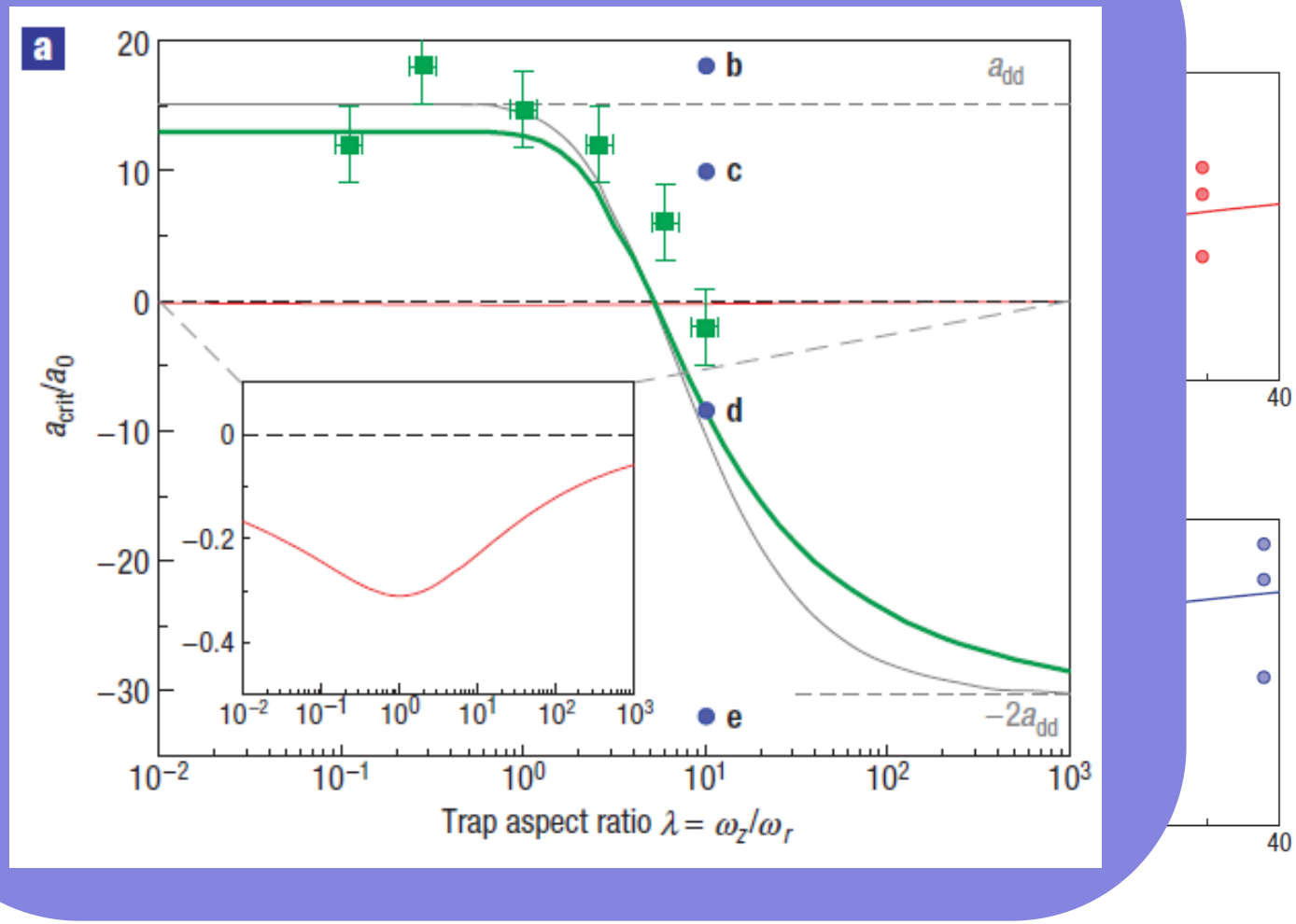
A homogeneous BEC is unstable

The stability also depends
the trap aspect ratio λ



Tilman Pfau group
Nature Physics **4**, 218 (2008)

$a_{critical}$ vs. λ



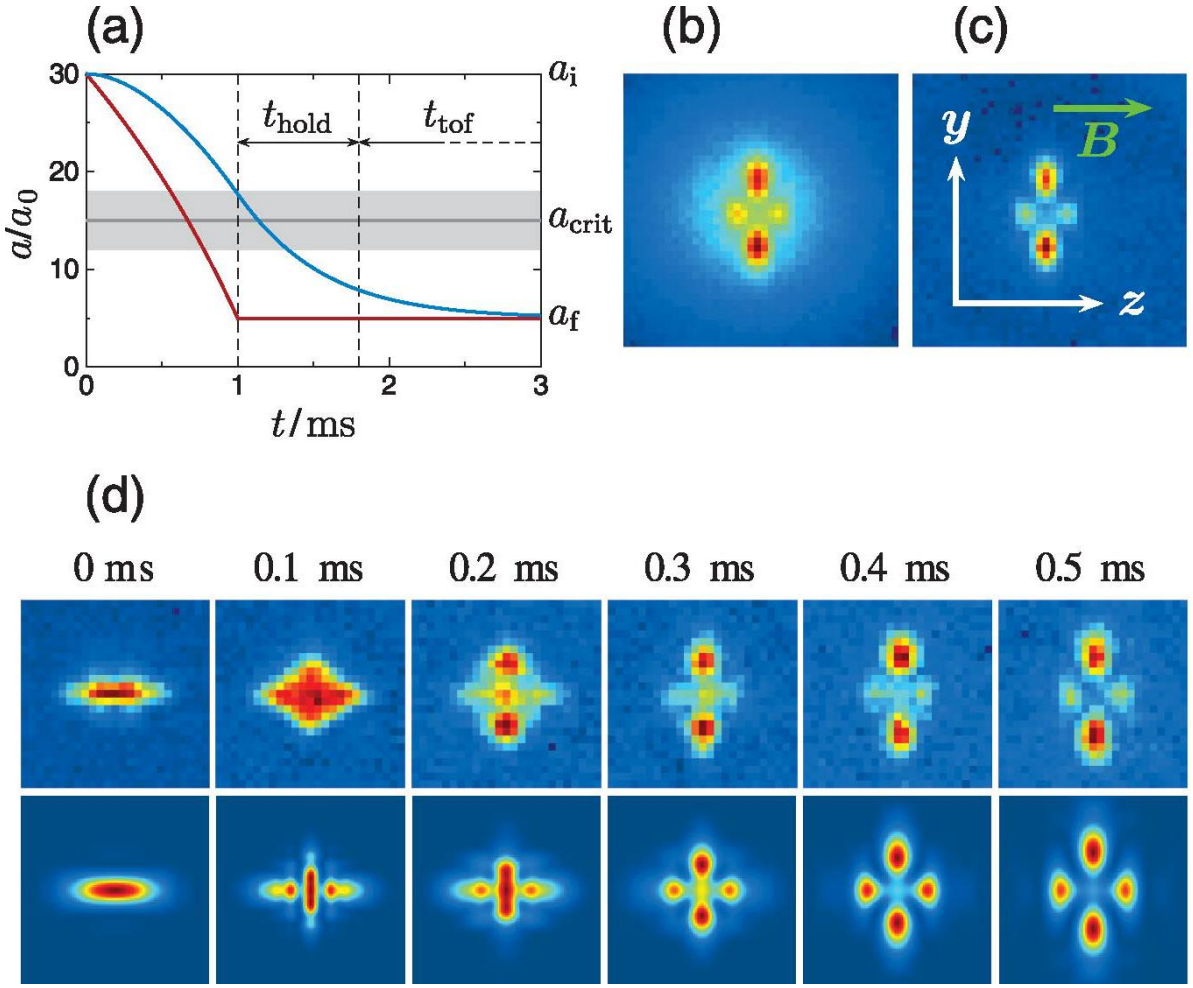
^{52}Cr BEC: d-wave collapse

For polarized dipoles:

$$U_{dd}(\mathbf{r}) \propto 1 - 3\cos^2\theta \propto Y_{20}$$

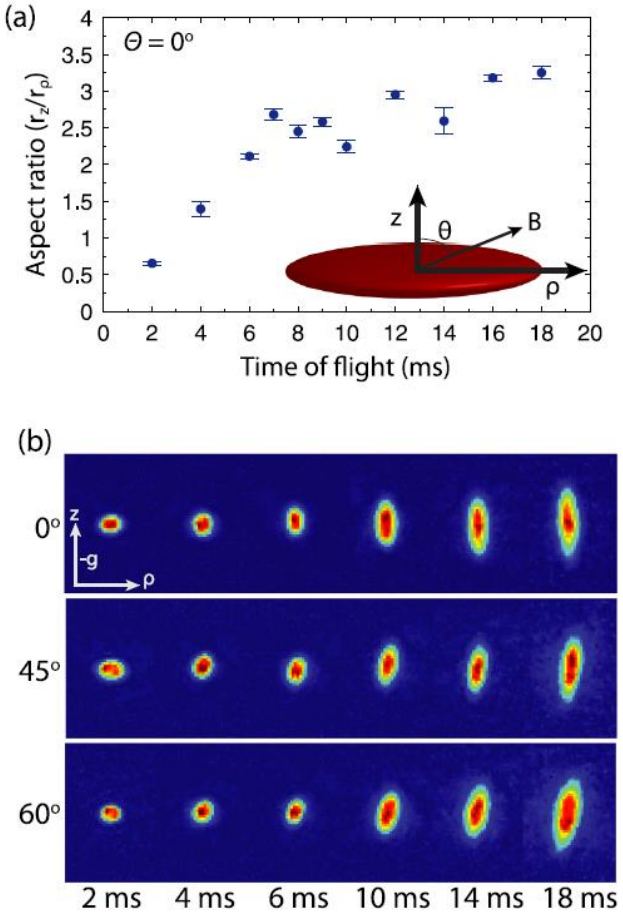


Pfau and Ueda et al.,
PRL **101**, 080401 (2008)

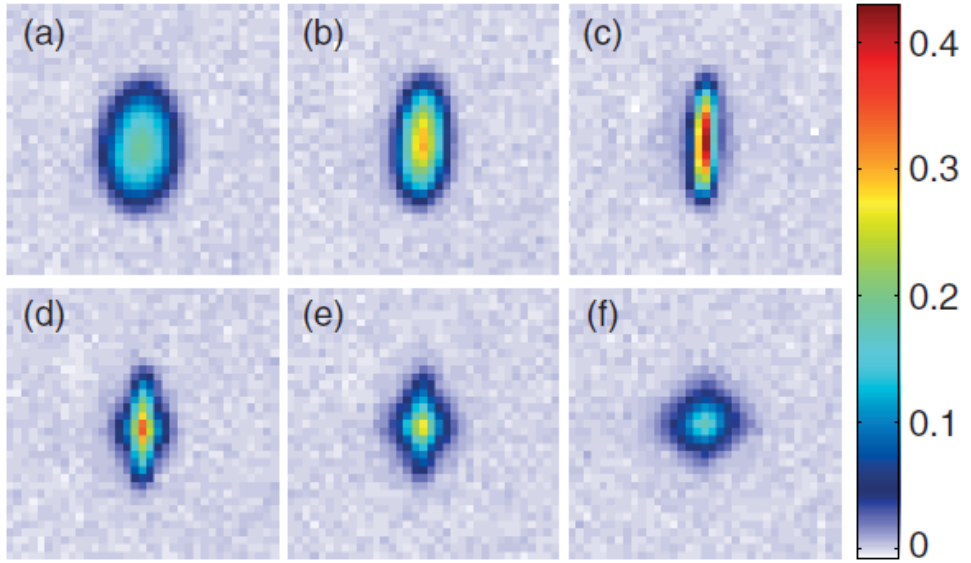


Also for Dy and Er BECs

^{52}Cr	6 μ_B	16	~100	0.16
Dy	10 μ_B	130	~100	1.3
Er	7 μ_B	67	~175	0.38



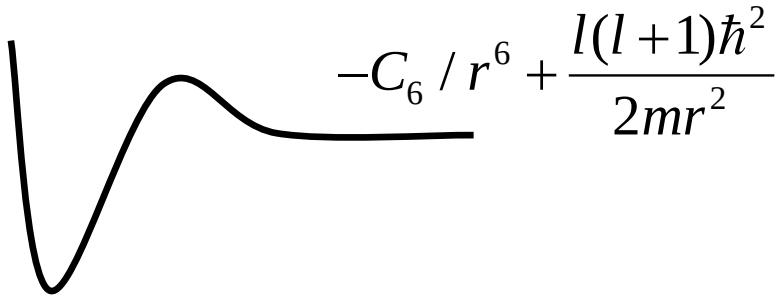
Stanford 2011: BEC of Dy
Anisotropic expansion and stability



Innsbruck 2012: BEC of Er
d-wave collapse vs. a

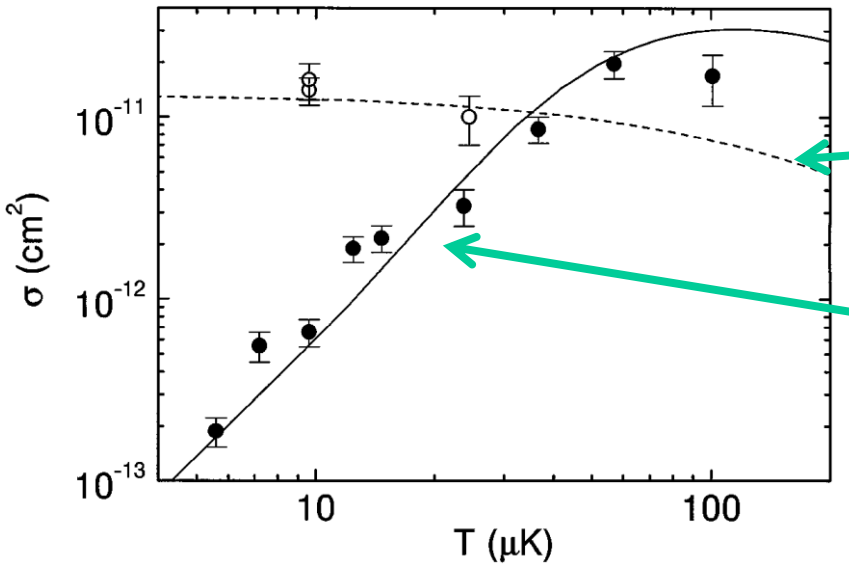
Dy and Er DFG: evaporative cooling via DDI

Identical ultracold fermions collide via p-wave (l = 1)



p-wave collisions are suppressed at ultralow temperatures

"Identical fermions do not collide with each other"



At ultracold T, p-wave collisions are slow

Two component

Single component

Threshold law:

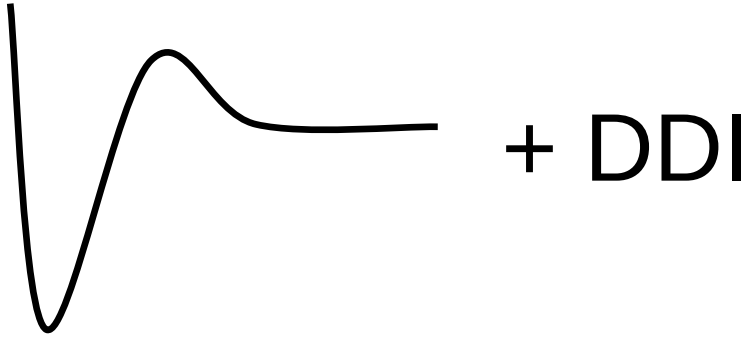
$\sigma_s \propto$ constant

$\sigma_p \propto T^2$

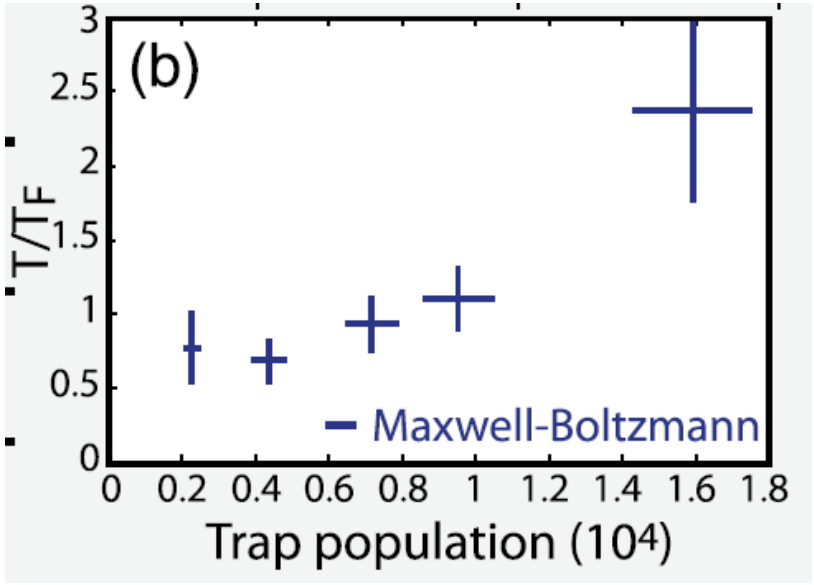
Measured with ultracold 40K by D.S.Jin's group, PRL (1999)

Dy and Er DFG: evaporative cooling via DDI

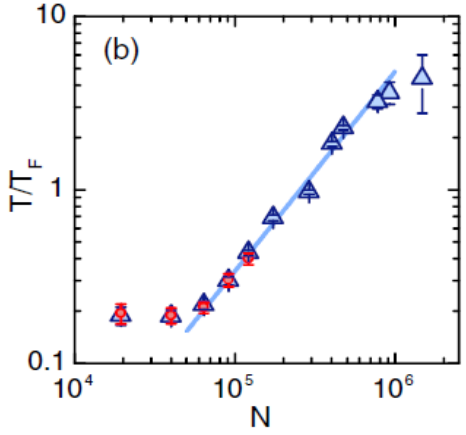
Achieved in Stanford (2012) and Innsbruck (2013), respectively

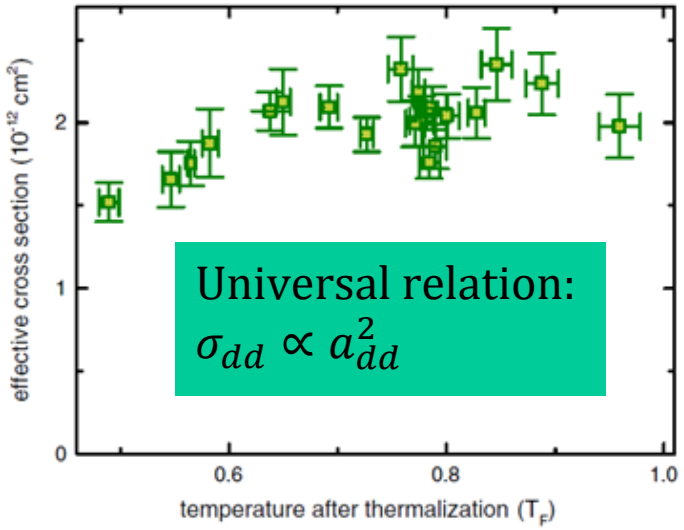


+ DDI



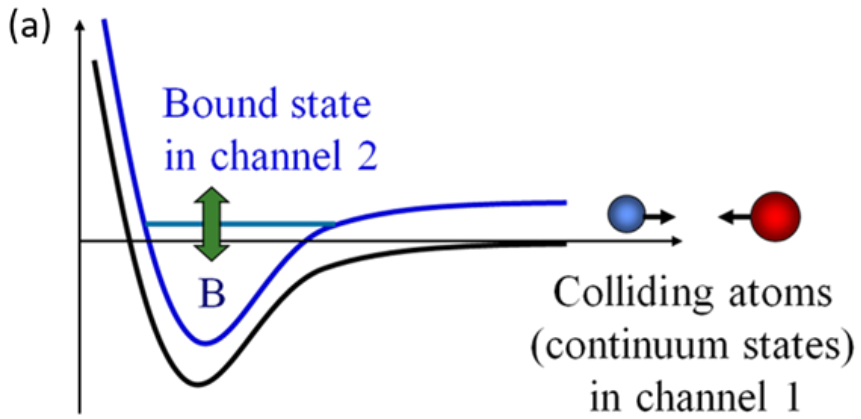
With single component fermion Dy, Lev group (2012)



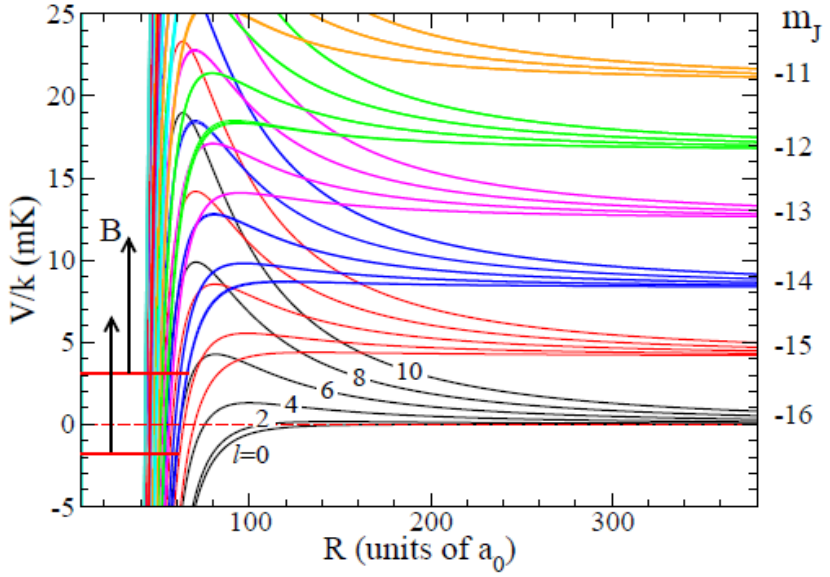


With Er, Innsbruck group (2014)

Dy and Er: chaotic Feshbach resonance



- ✓ Feshbach resonance for alkali atoms: single or a few partial waves; bound levels with large energy spacing
- ✓ Feshbach resonance for magnetic atoms: multi-partial waves; bound levels with large energy spacing
- ✓ Feshbach resonance (E field) for polar molecules: multi-partial wave; much denser bound states

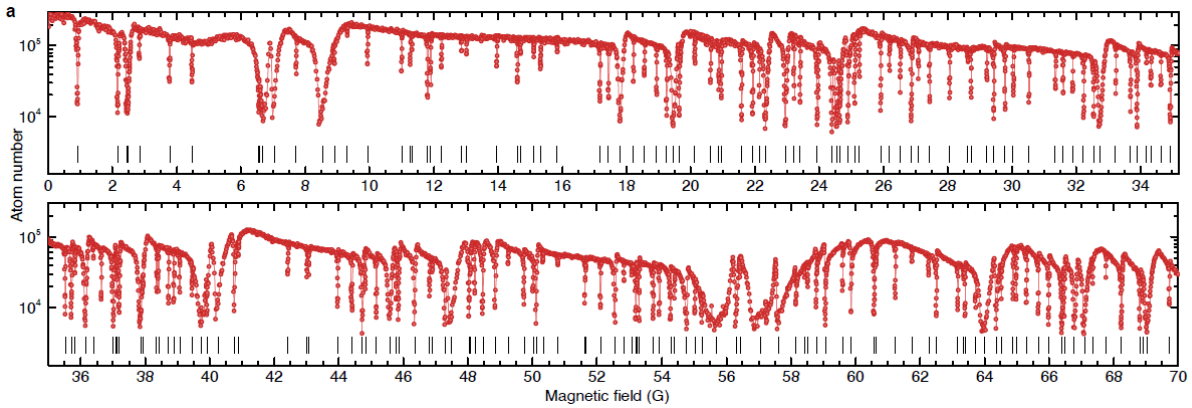


Example PEC for Dy+Dy

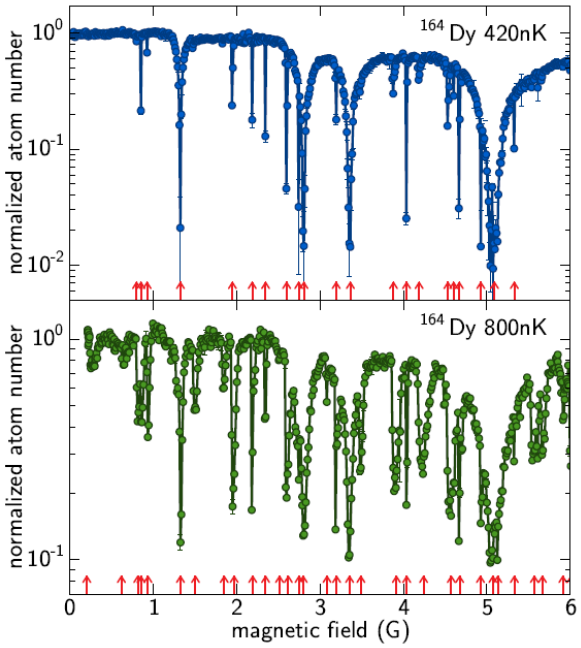
Phys. Rev. A 85, 062712 (2012).

Dy and Er: chaotic Feshbach resonance

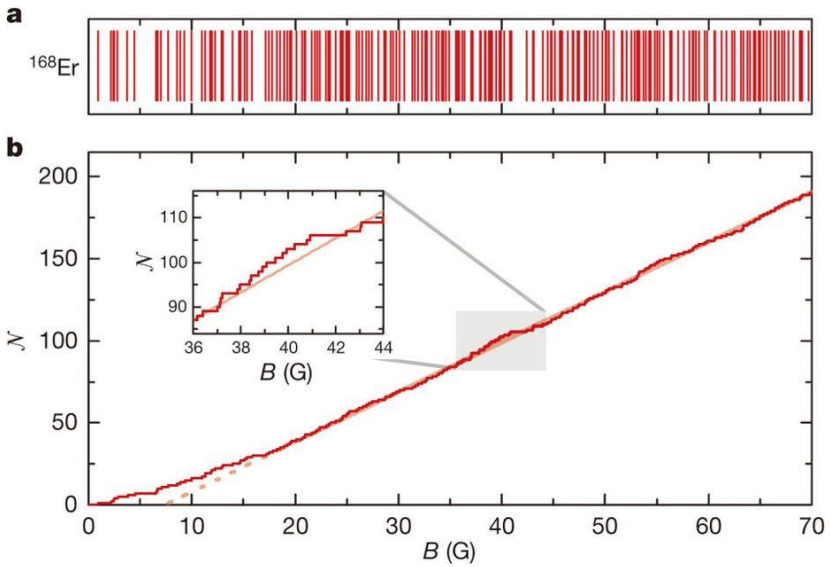
Dense Feshbach resonances in Er and Dy



Innsbruck
Stuttgart



Stanford

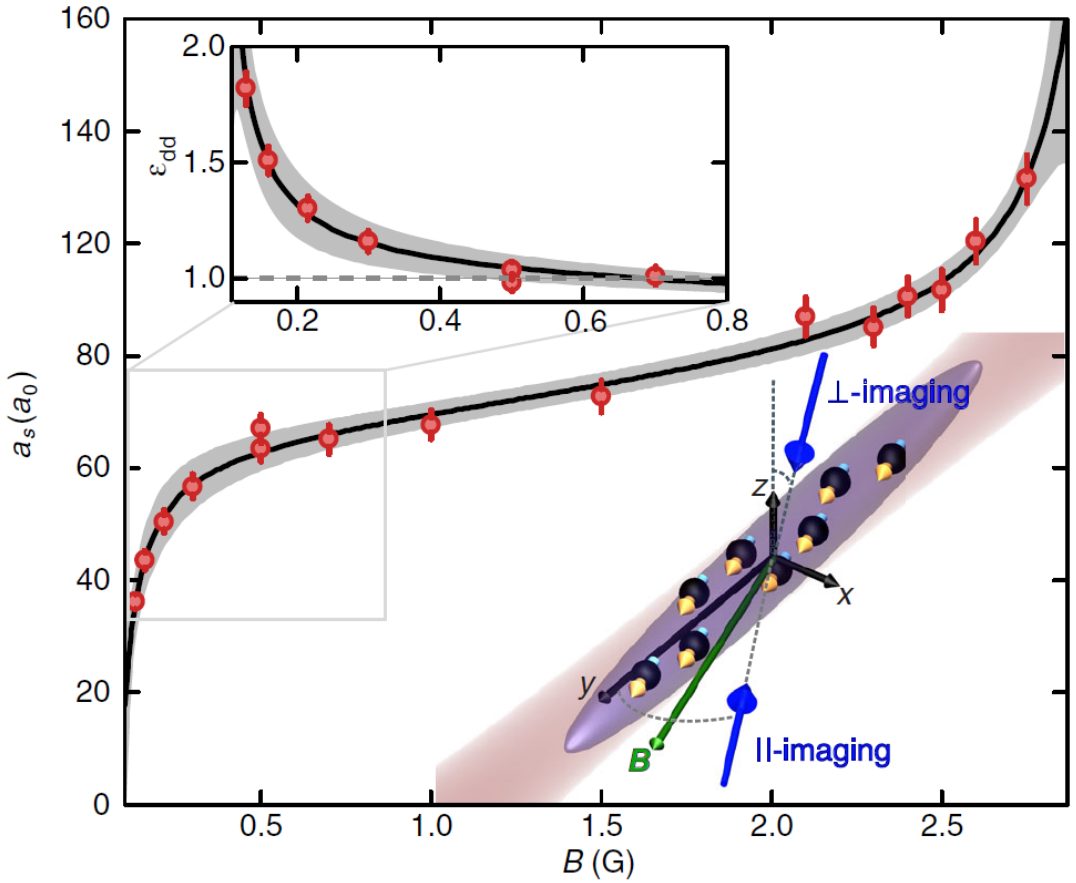


Nature **507**, 475–479 (2014)

Dy and Er: chaotic Feshbach resonance

Some of these resonances can still be used for scattering length a tuning

$$\varepsilon_{dd} = \frac{a_{dd}}{a}$$



From Er experiment
PHYSICAL REVIEW X 6, 041039 (2016)

Dy and Er BEC: droplet

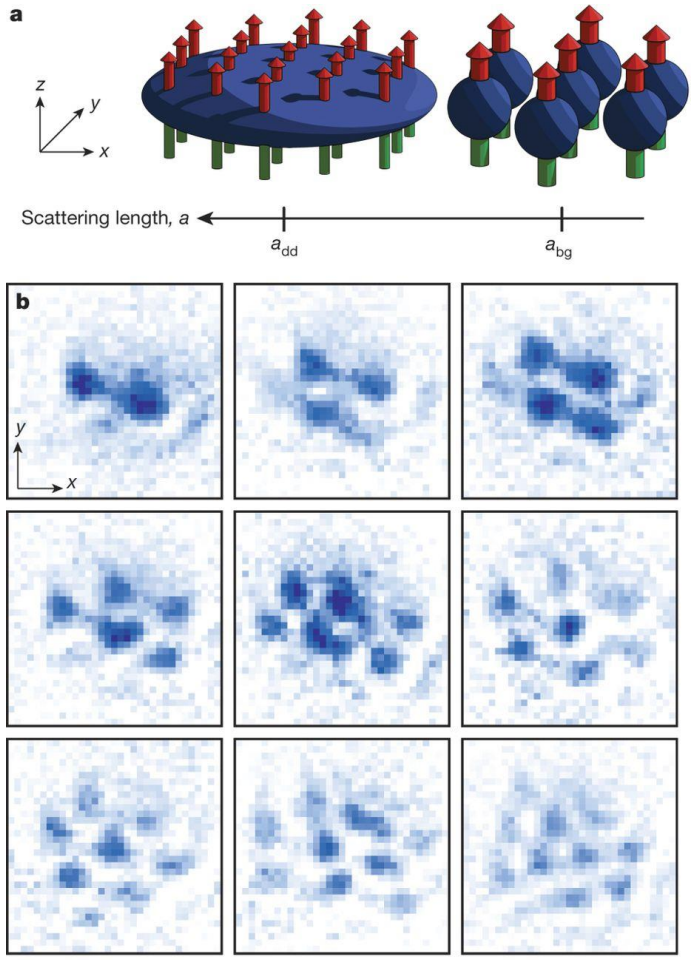
$\varepsilon_{dd} = \frac{a_{dd}}{a}$

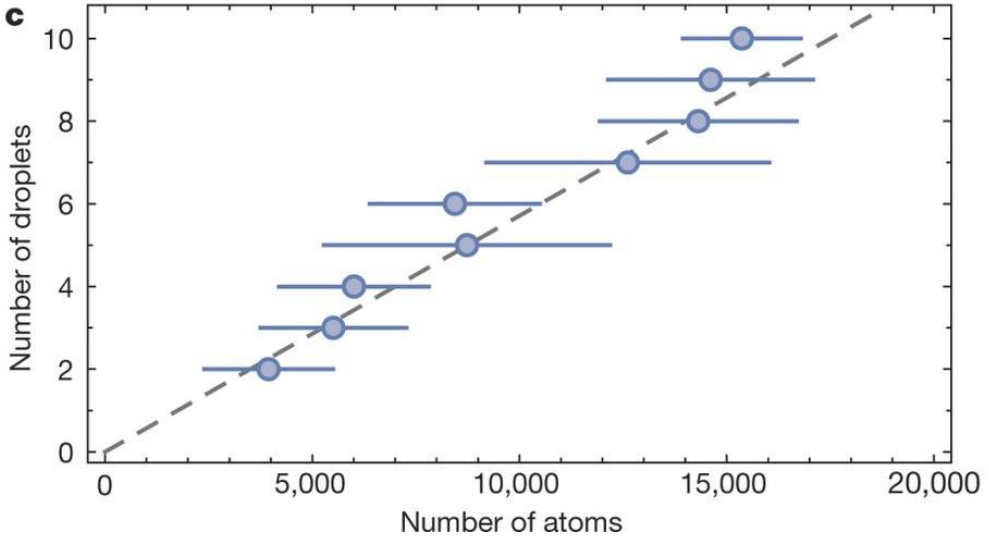
A homogeneous BEC is unstable if $a < a_{dd}$

Conclusion from mean-field theory

changing

$\varepsilon_{dd} = \frac{a_{dd}}{a}$

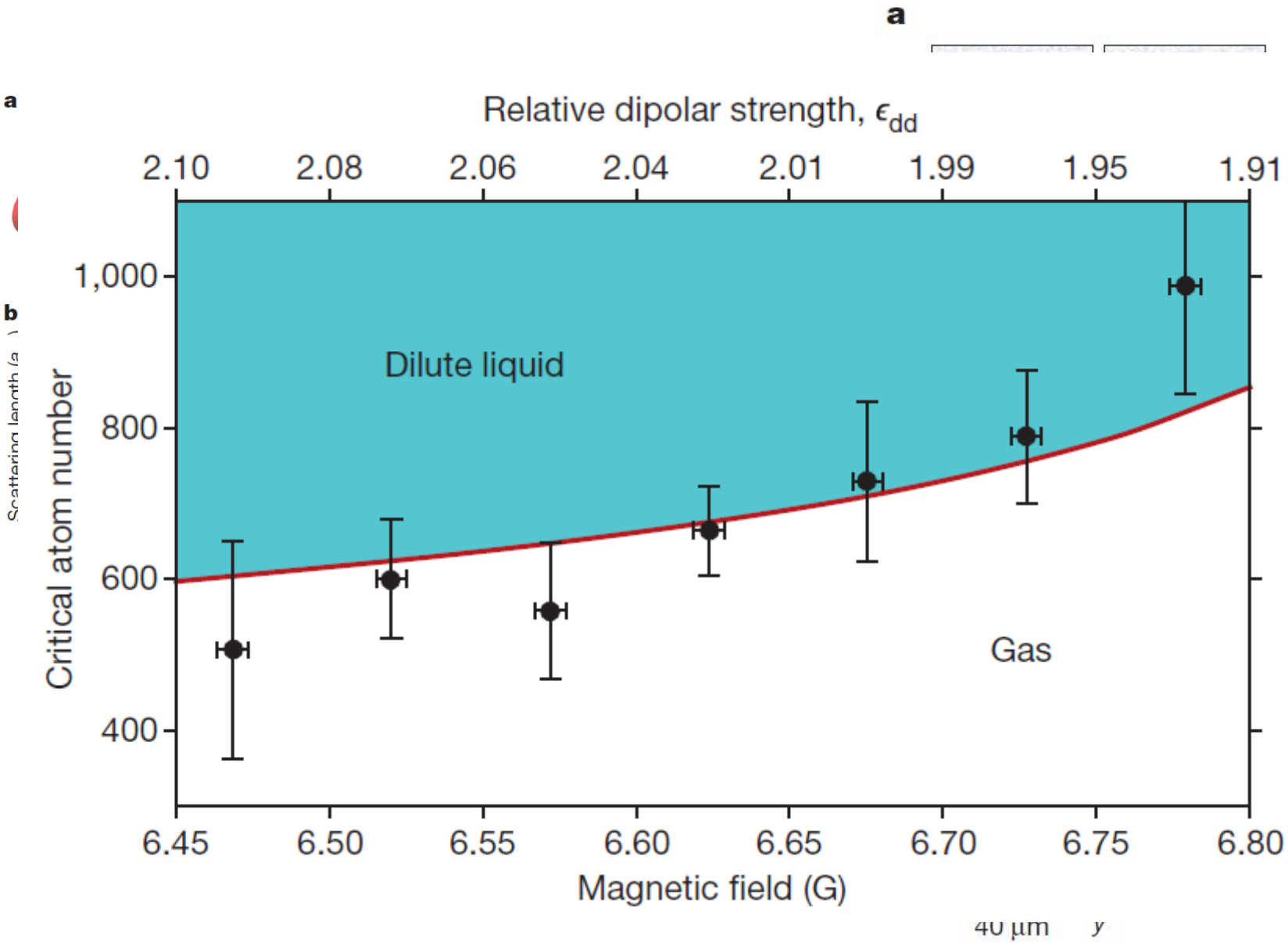




Nature 530, pages 194–197 (2016)

Dy and Er BEC: ground-state droplet

3D droplet



Dipolar droplet: beyond MF effect

$$E/V = (gn^2/2)(1 + 128\sqrt{na^3}/15\sqrt{\pi} + \cdots)$$

LHY correction due to quantum fluctuation

Phys. Rev. 106, 1135 (1957)

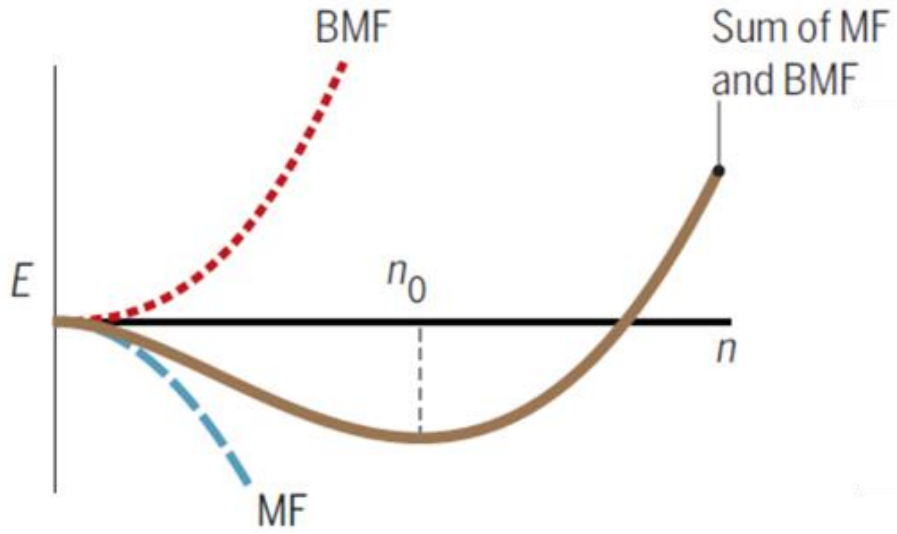
For dipolar BEC:

$$\frac{E_{\mathrm{MF}}}{N} = \frac{1}{2^{3/2}} \frac{g}{2} n_{\mathrm{c}} \left(1 - f_{\mathrm{dip}}(\kappa)\, \varepsilon_{\mathrm{dd}}\right),$$

$$\frac{E_{\mathrm{BMF}}}{N} = \left(\frac{2}{5}\right)^{3/2} \frac{g}{2} n_{\mathrm{c}} \frac{128}{15\sqrt{\pi}} \sqrt{n_{\mathrm{c}} a^3}\, Q_5(\varepsilon_{\mathrm{dd}}),$$

For $f_{dip}(\kappa)\varepsilon_{dd} > 1$:

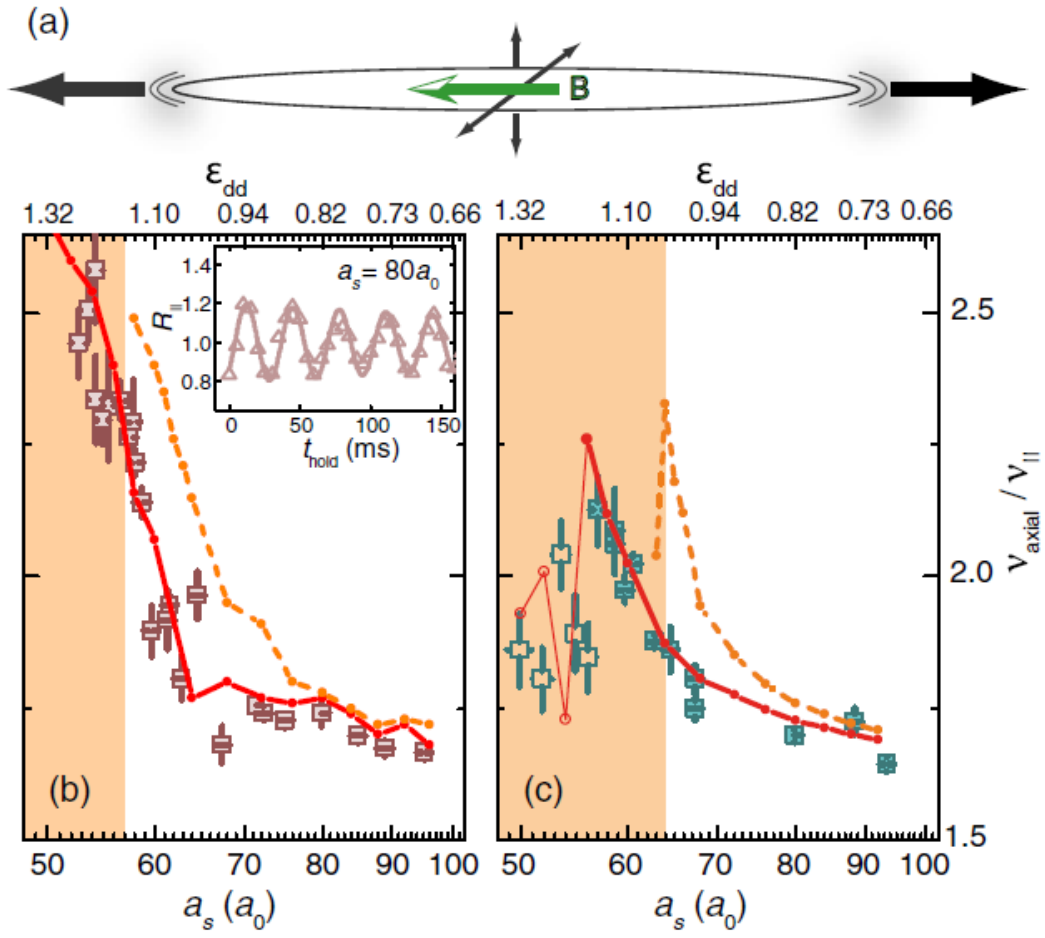
$E_{MF} < 0$, but
$E_{BMF} > 0$



E_{BMF} has a stronger dependency in the density

Dipolar droplet: beyond MF effect

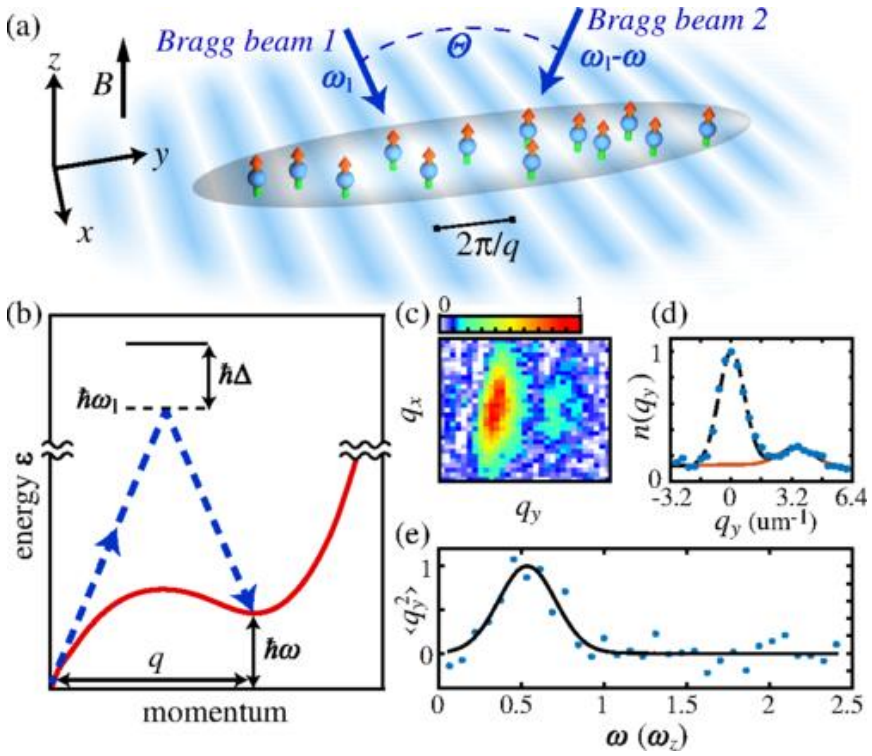
Also in Er BEC PHYSICAL REVIEW X 6, 041039 (2016)

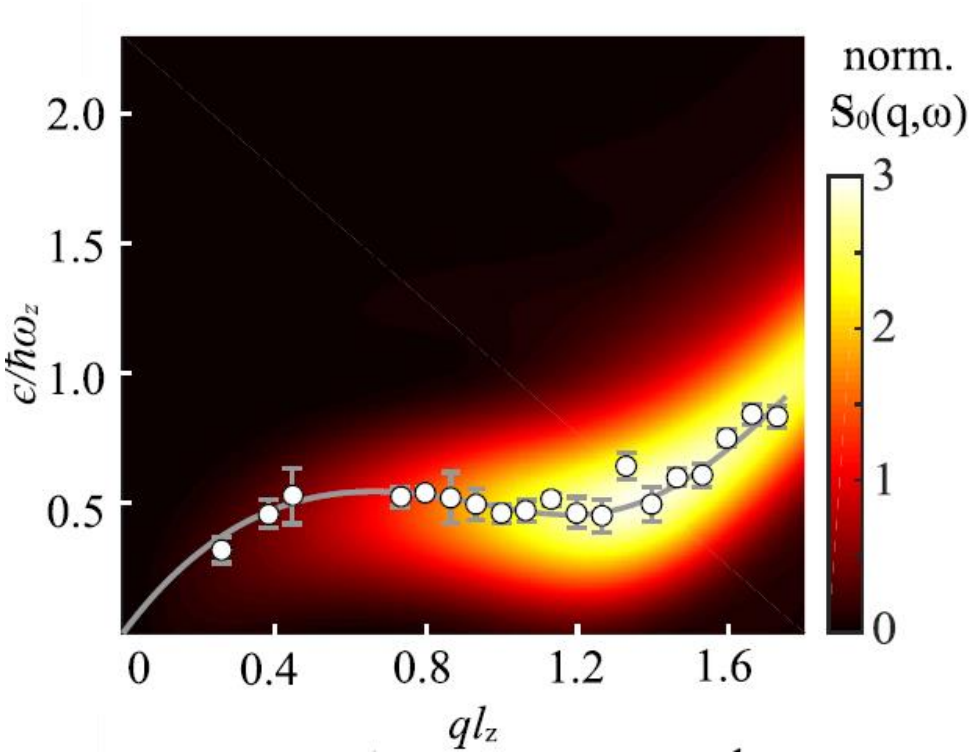


Detecting LHY effect from collective oscillation of Er BEC

Dipolar supersolid

Roton mode in dBECs confined in cigar-shaped geometries





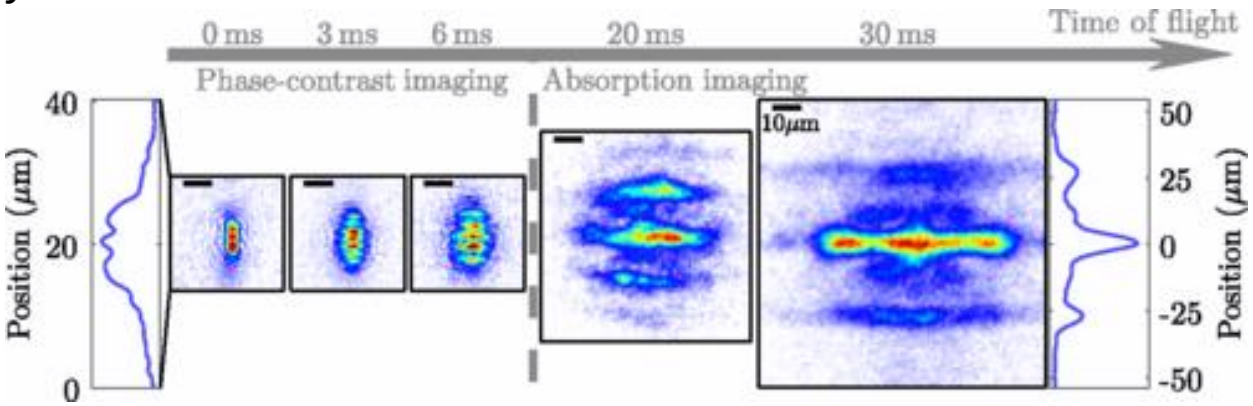
Roton minimum at $k \sim 1/lz$

Phys. Rev. Lett. 122, 183401 (2019)

Dipolar supersolid

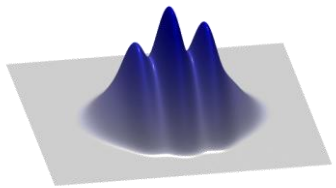
Two hallmarks of supersolidity:

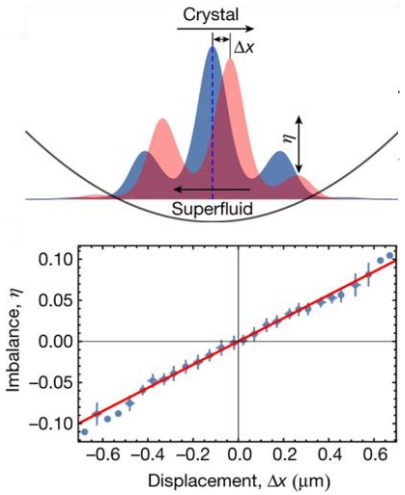
- spontaneously formed density modulation
- global phase coherence



Phys. Rev. X **9** 011051 (2019)

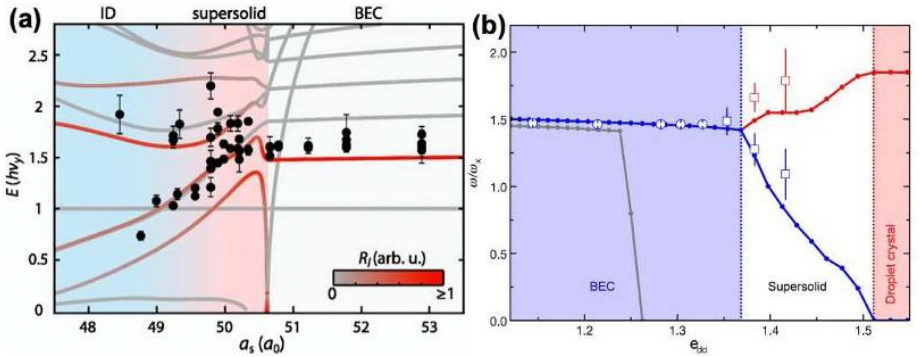
Dynamical response





Goldstone mode

Nature, 574,386 (2019)

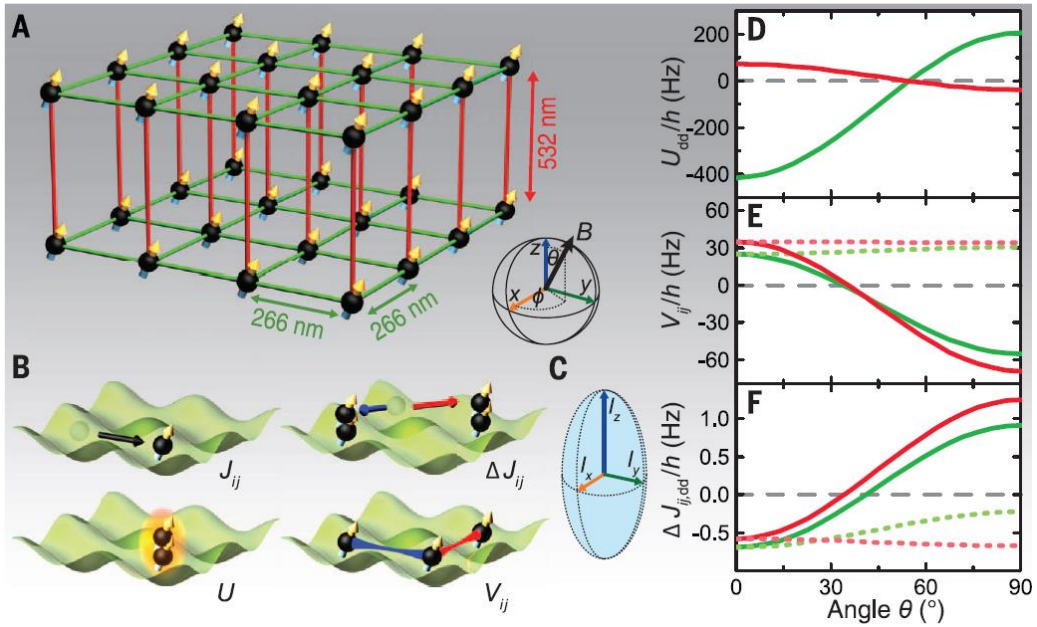


Two branches of the quadrupole mode

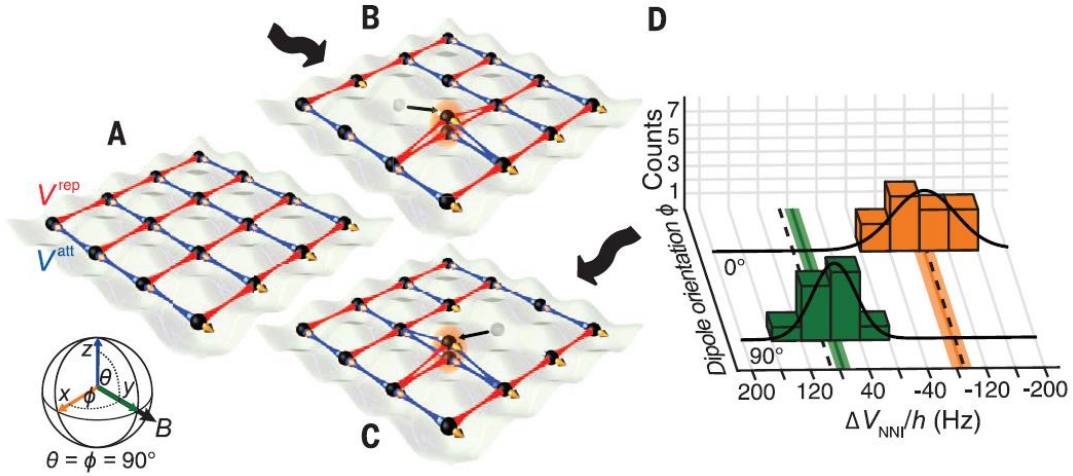
Nature 574 382 (2019)
PRL, 123 050402 (2019)

Magnetic atoms in optical lattices: extended Hubbard model

An early experiment



Science 352, 201-5 (2016)



Probe the nearest neighboring interaction

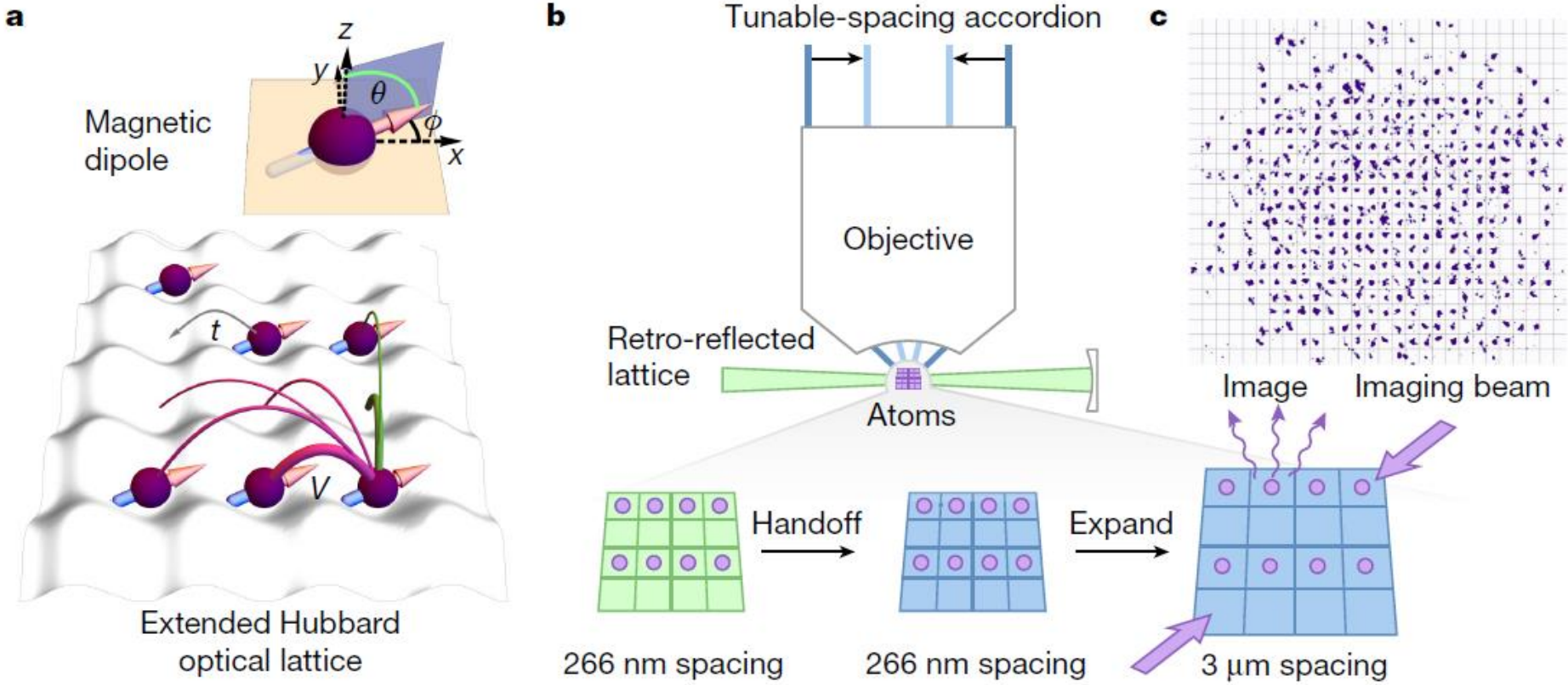
Extended Hubbard model: Dipolar quantum solids

266 nm lattice constant + Quantum microscope

On-site interaction > all other energy scales

Extended Hubbard model in the hard-core boson limit

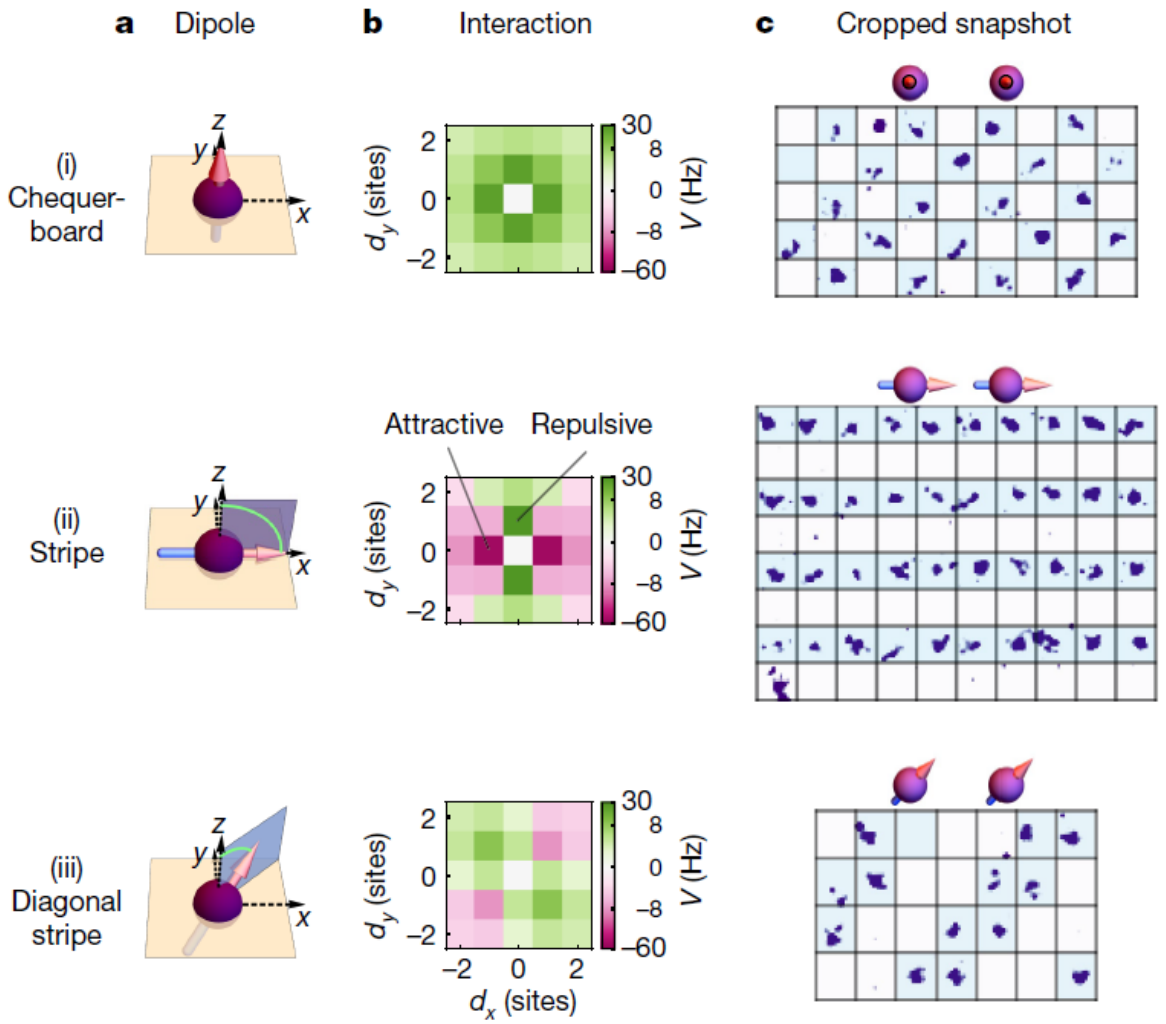
$$H = -t \sum_{\langle i,j \rangle} (\hat{a}_i^\dagger \hat{a}_j + \text{h.c.}) - \sum_i \mu_i \hat{n}_i + \sum_{i<j} V_{i,j} \hat{n}_i \hat{n}_j .$$



Nature 622, 754 (2023)

Extended Hubbard model: Dipolar quantum solids

266 nm lattice constant + Quantum microscope



Nature 622, 754 (2023)

Summary: magnetic atoms

- Degeneracy is easy to reach;
- DDI is not strong, but its effects can be enhanced by controlling contact interactions;
- DDI's anisotropic character can be explored;
- Strongly long-range many-body physics: smaller spacing.

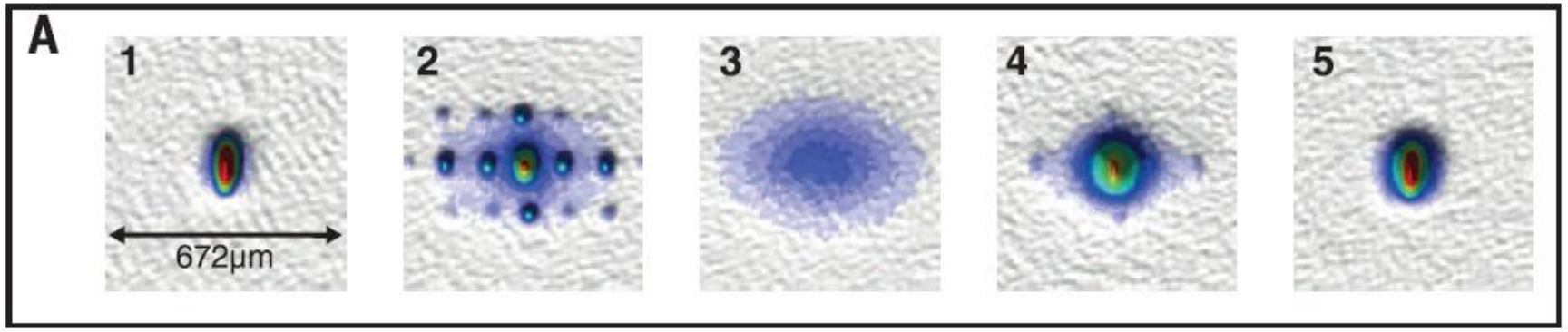
Lots of exciting results ahead: pay attention!

Not discussed:
Dipolar 1D system, 50 nm spacing system (MIT), Dipolar BKT (HKUST), Optical tweezer…

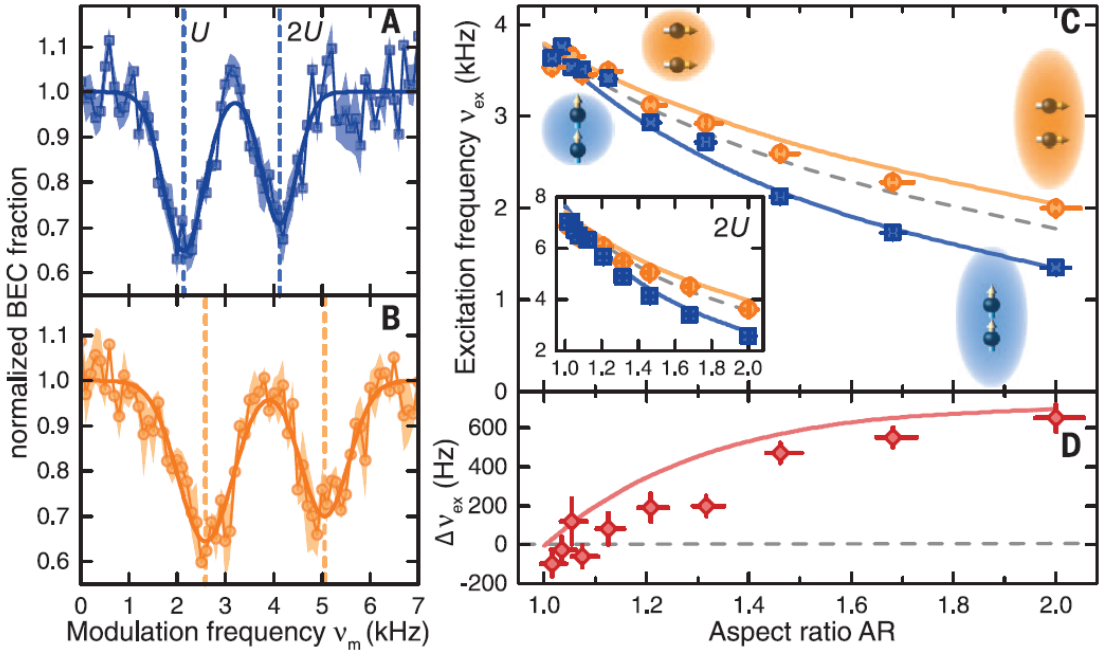
Thanks for your attention!



Magnetic atoms in optical lattices: extended Hubbard model



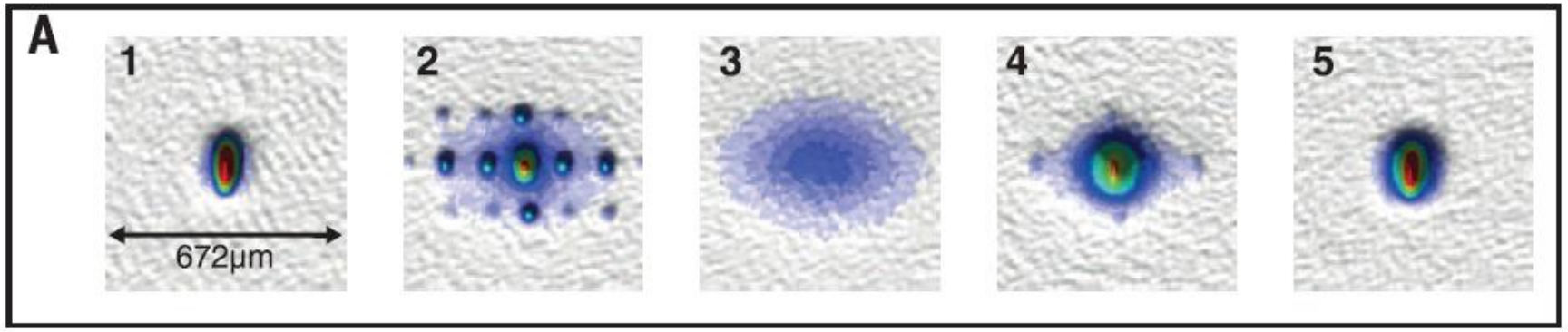
SF-to-MI transition



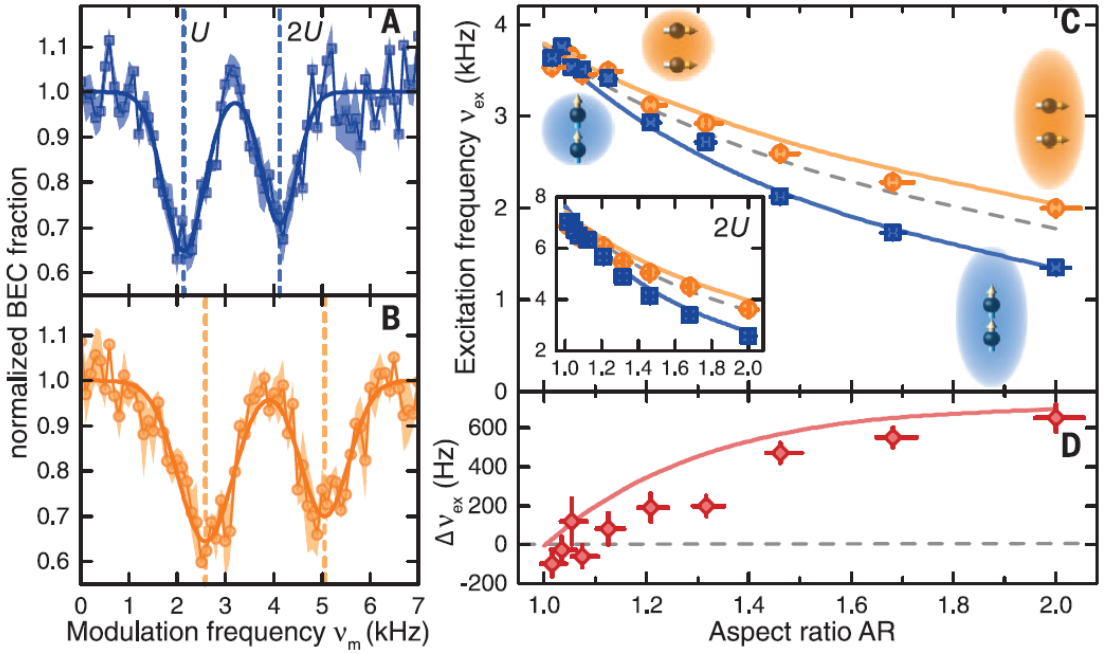
On site interaction

Science 352, 201-5 (2016)

Magnetic atoms in optical lattices: extended Hubbard model



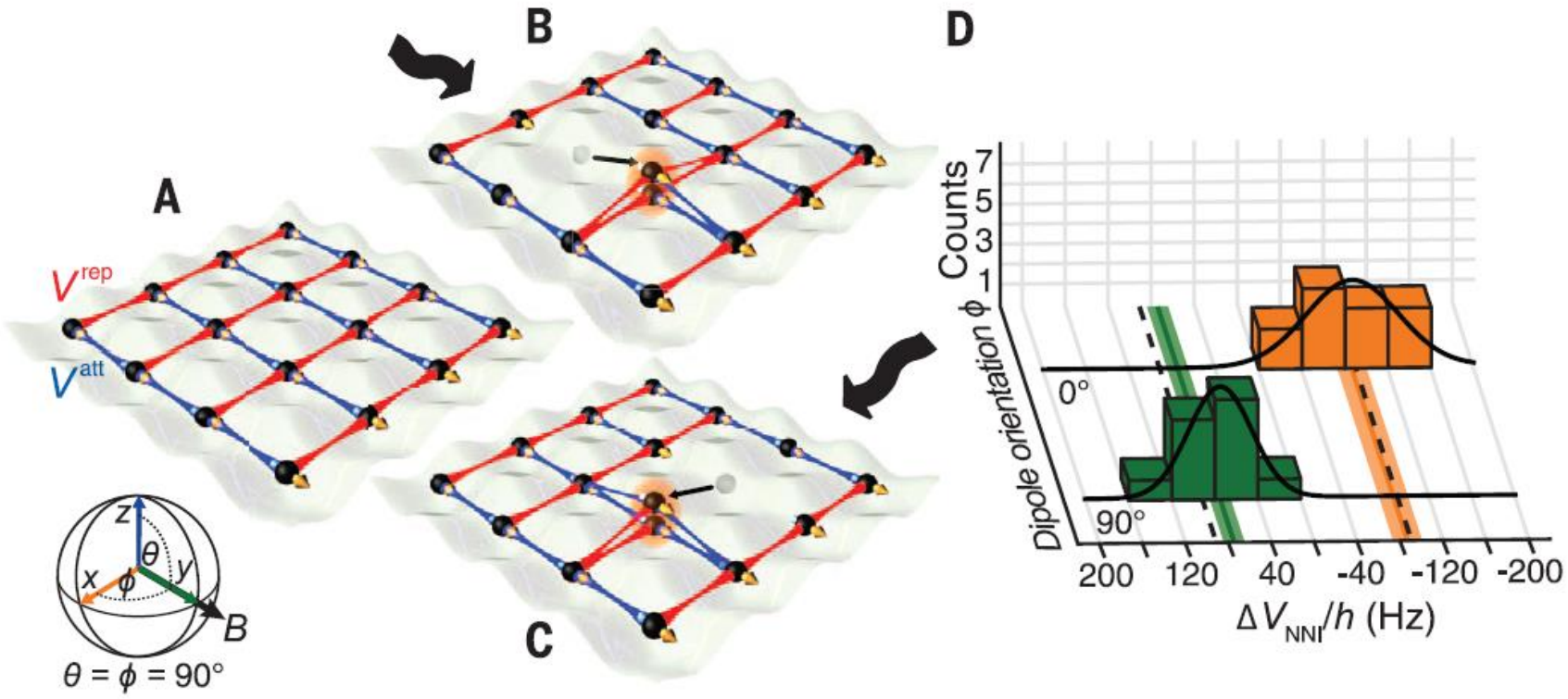
SF-to-MI transition



On site interaction

Science 352, 201-5 (2016)

Magnetic atoms in optical lattices: extended Hubbard model



Nearest neighboring interaction

Science 352, 201-5 (2016)

Ultracold dipolar atoms and molecules part II

Dajun Wang
Department of Physics
The Chinese University of Hong Kong

CROUCHER SUMMER COURSE
"ULTRACOLD ATOM PHYSICS"

May 20 – 24, 2024

Supported by RGC Hong Kong

Outline

Dipoles and dipole-dipole interaction

Experiments with magnetic atoms

Ultracold polar molecules

Rep. Prog. Phys. ***72***, 126401 (2009)
Rep. Prog. Phys. ***86***, 026401 (2023)

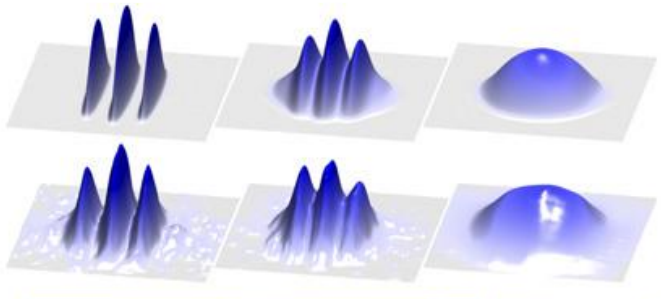
Cold and Ultracold Molecules

Insight issue *Nature Physics (2024)*

Ultracold dipolar systems

Magnetic atoms:

Magnetic dipole moment

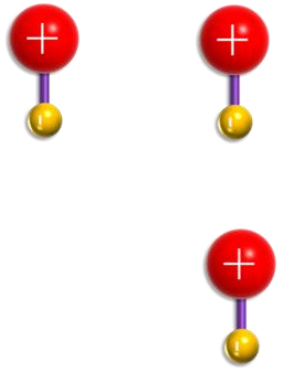


Rep. Prog. Phys. 84 012403 (2021)

- Cr (6 μ_B), Er (7 μ_B), Dy (10 μ_B)…
- Long lifetime
- BEC and DFG

Polar Molecules:

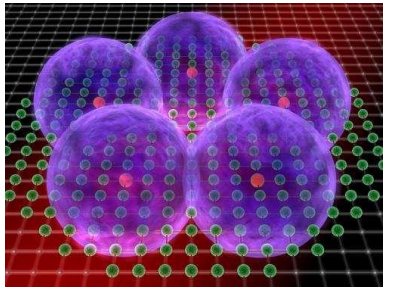
Electric dipole moment



- Many species available now: bi-alkali, CaF, SrF, YO…
- d ~ 1 to several Debye; tunable
- Long lifetime? Complicated by two-body interactions
- DFG 2019, BEC 2023

Rydberg atoms:

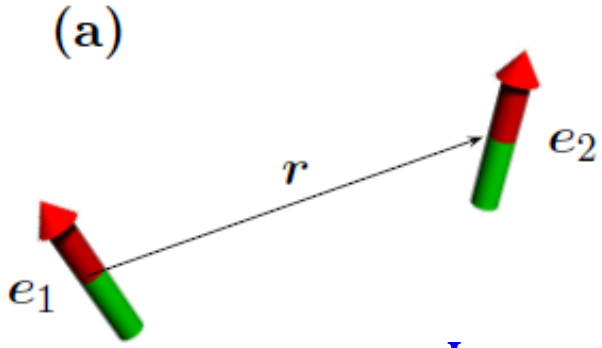
Giant electric dipole moment



Nature 491, 87 (2012)

- d ~ 1000 Debye
- Blockade mechanism
- Short lifetime: ~ 100 μs
- Tweezer array

Molecules can provide much larger DDIs



In general:

$$U_{dd}(\mathbf{r}) = \frac{C_{dd}}{4\pi}\left(\frac{\hat{e}_1 \cdot \hat{e}_2 - 3(\hat{e}_1 \cdot \hat{\mathbf{r}})(\hat{e}_2 \cdot \hat{\mathbf{r}})}{r^3}\right)$$

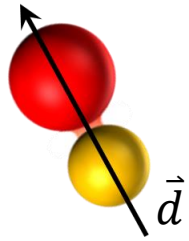
$C_{dd} = \mu_0 \mu^2$ for magnetic dipole moment μ

$C_{dd} = \dfrac{d^2}{\varepsilon_0}$ for electric dipole moment d

$$\frac{\mu_0 \mu_B^2}{(e\, a_0)^2/\varepsilon_0} \sim \alpha^2 = \left(\frac{1}{2 \times 137}\right)^2 : \text{DDI is much stronger in electric case}$$

T. Lahaye, et al, *Rep. Prog. Phys.* ***72***, *126401 (2009)*

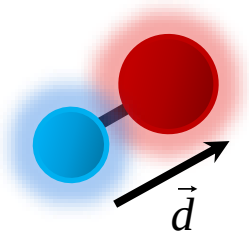
Polarize the polar molecule

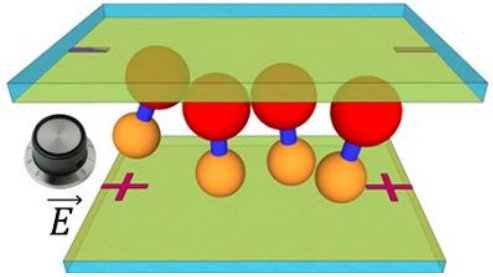


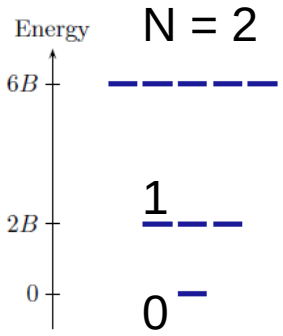
The electric dipole moment is along the axis of the molecule. Without external E field:

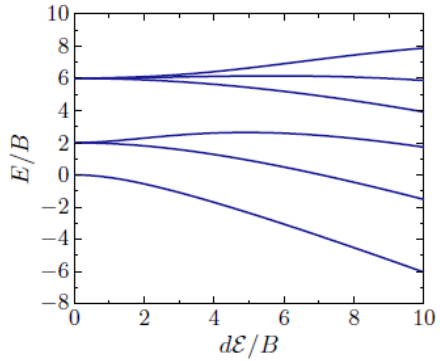
$$d_{eff} = \langle\psi|\hat{d}|\psi\rangle = e\langle\psi|\vec{r}|\psi\rangle = 0$$

Polarize with static DC electric fields

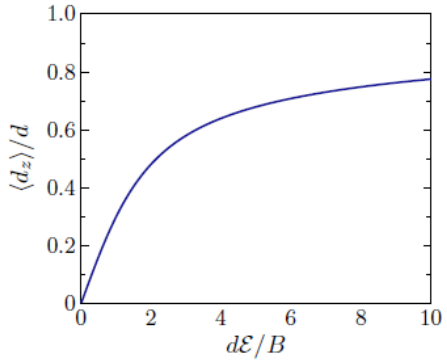






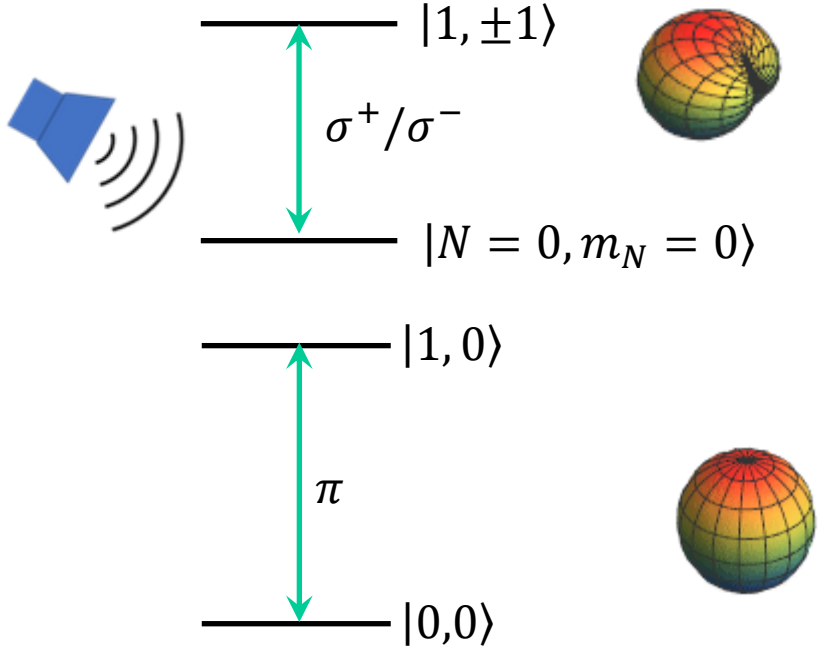


DC Stark shifts



Lab frame dipole d_{eff}

With microwave AC electric fields

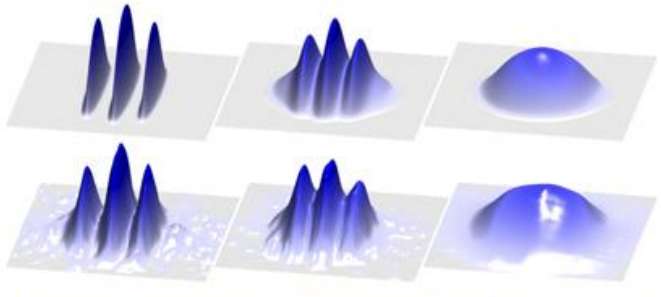


Effective dipole moment: $d_{eff} \propto \frac{d_0}{\sqrt{1+(\Delta/\Omega)^2}}$

Karman et al., PRA 105, 013321 (2022)

Dipolar droplet and supersolid

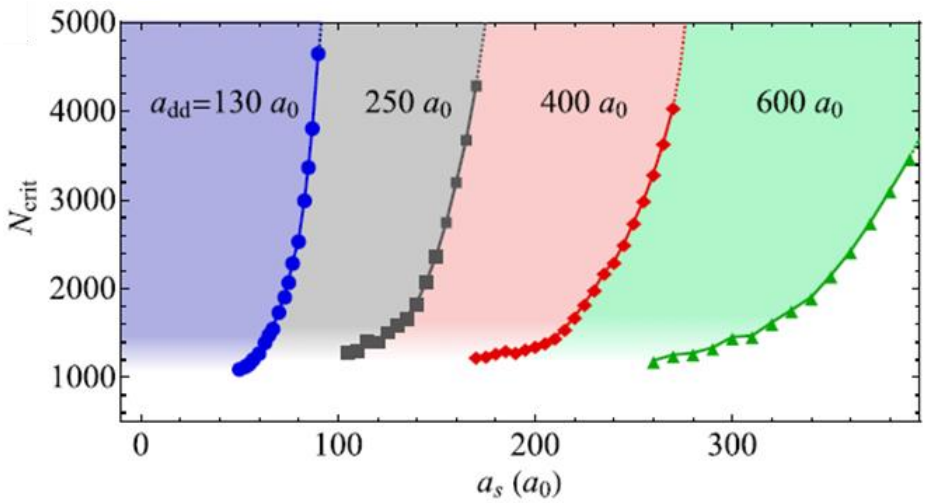
Droplet crystals with Er and Dy BEC

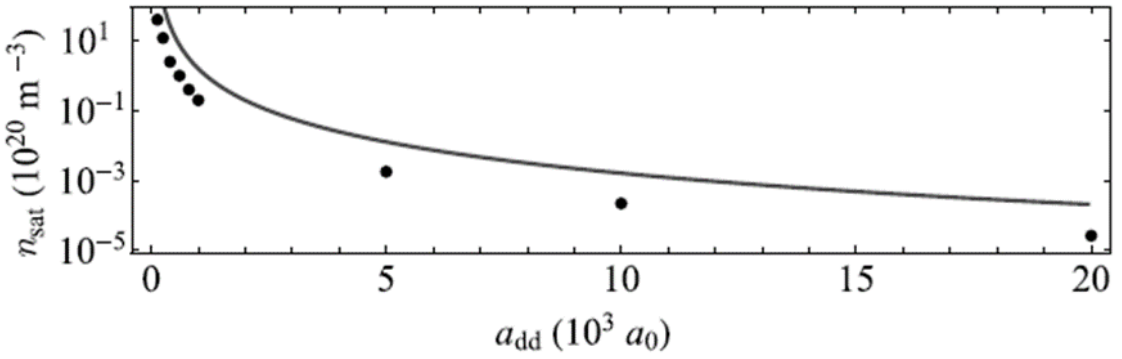


Rep. Prog. Phys. 84 012403 (2021)

Species	Dipole moment	$a_{dd}(a_0)$	a (a_0)
^{87}Rb	1 μ_B	0.7	100
^{52}Cr	6 μ_B	16	~100
^{168}Er	7 μ_B	67	~175
^{164}Dy	10 μ_B	130	~100
KRb	0.6 D	3700	
NaK	2.7 D	43180	
NaRb	3.3 D	112700	~ 200
NaCs	4.7 D	324000	

Larger a_{dd} : less particles are needed to form a droplet and to reach the saturated regime

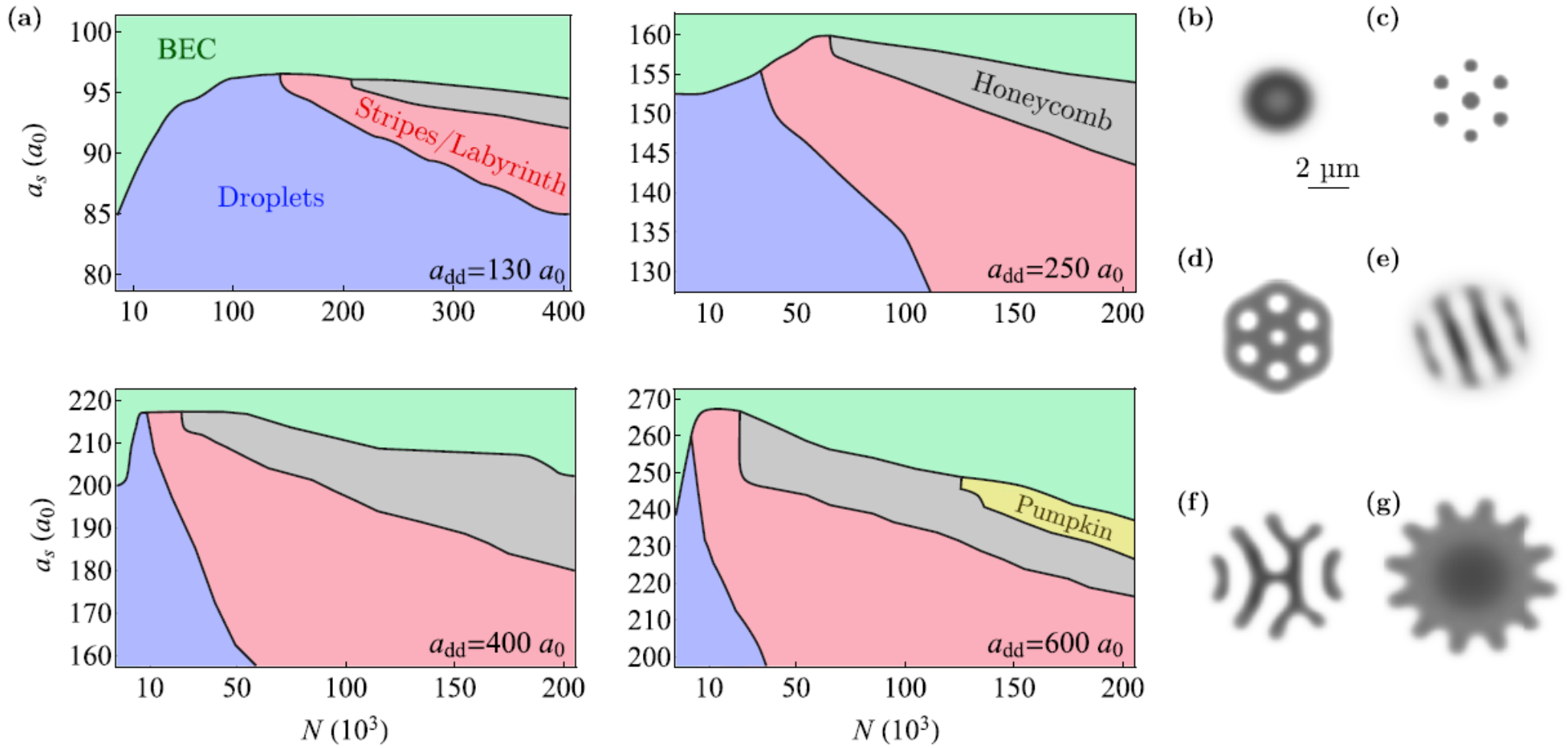




PRR **4**, 013235 (2022)

Dipolar droplet and supersolid

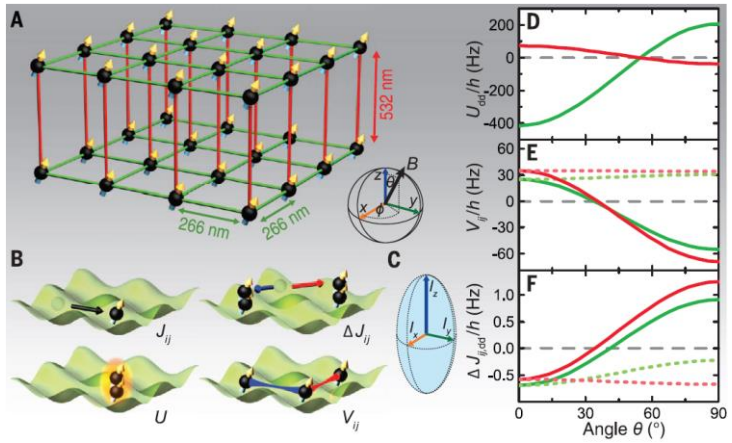
Larger a_{dd} : also richer supersolid states



PRR **4**, 013235 (2022)

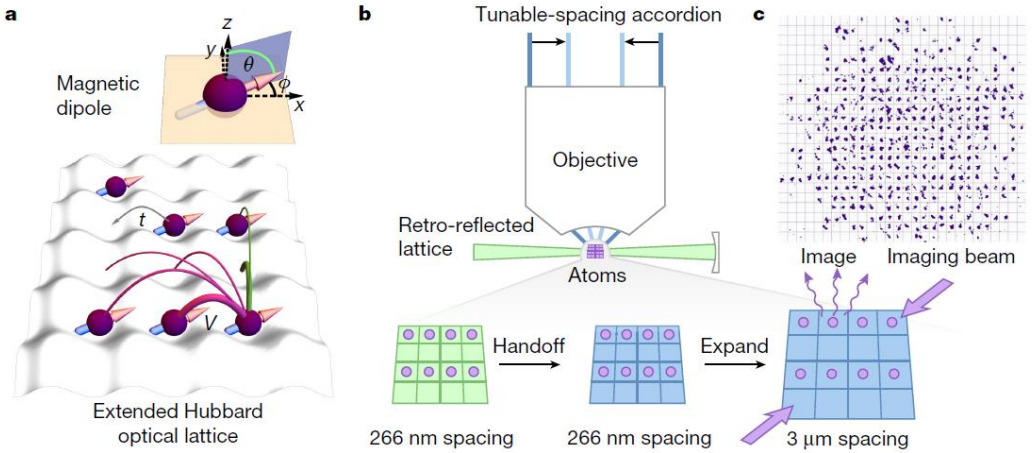
Extended Hubbard model

For magnetic atoms: needs small spacing



Science 352, 201-5 (2016)

for Er at 266 nm spacing, DDI~ 30 Hz



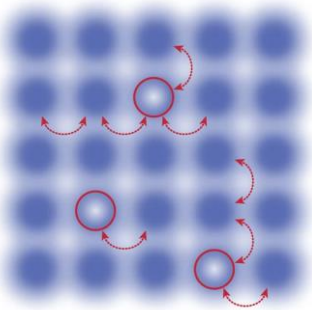
Nature 622, 754 (2023)

For polar molecules with a 1 Debye dipole moment:

At 532 nm spacing, DDI~ 1 kHz
At 266 nm spacing, DDI~ 8 kHz

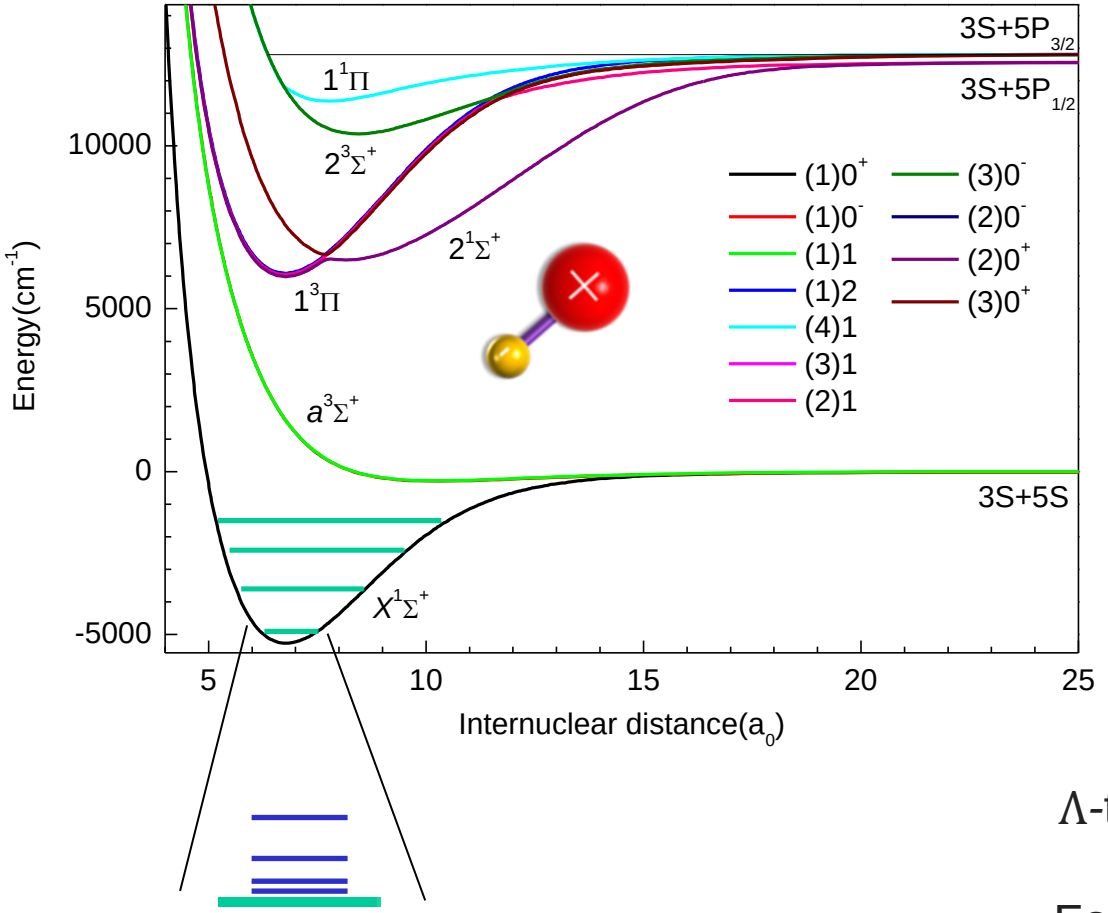
Richer dipolar quantum solid phases could be reached easily.

Make connection to the helium version of SS states?



Picture from:
Nature 464, 176 (2010)

More resources from molecules



Molecular states

Electronic: $^1\Sigma^+$, $^3\Pi$…
Fine structure: 0, 1, 2…
Hyperfine

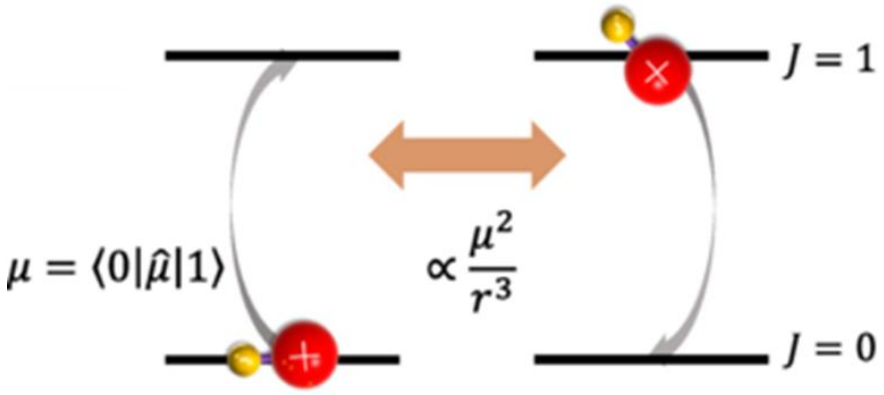
Vibration
Rotation

Λ-type *doubling?*

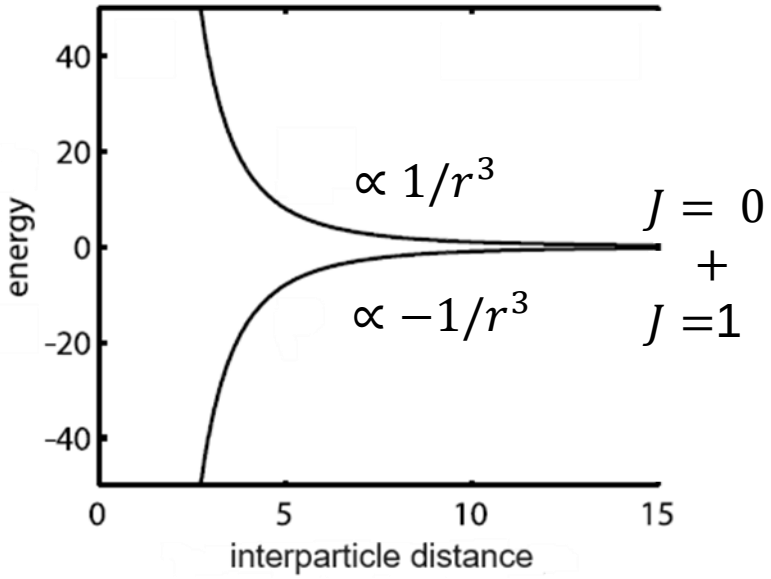
For non-Σ states.
Induced by coupling between nuclear rotation and electron orbital angular momentum

Rotational level: resonant dipolar interaction

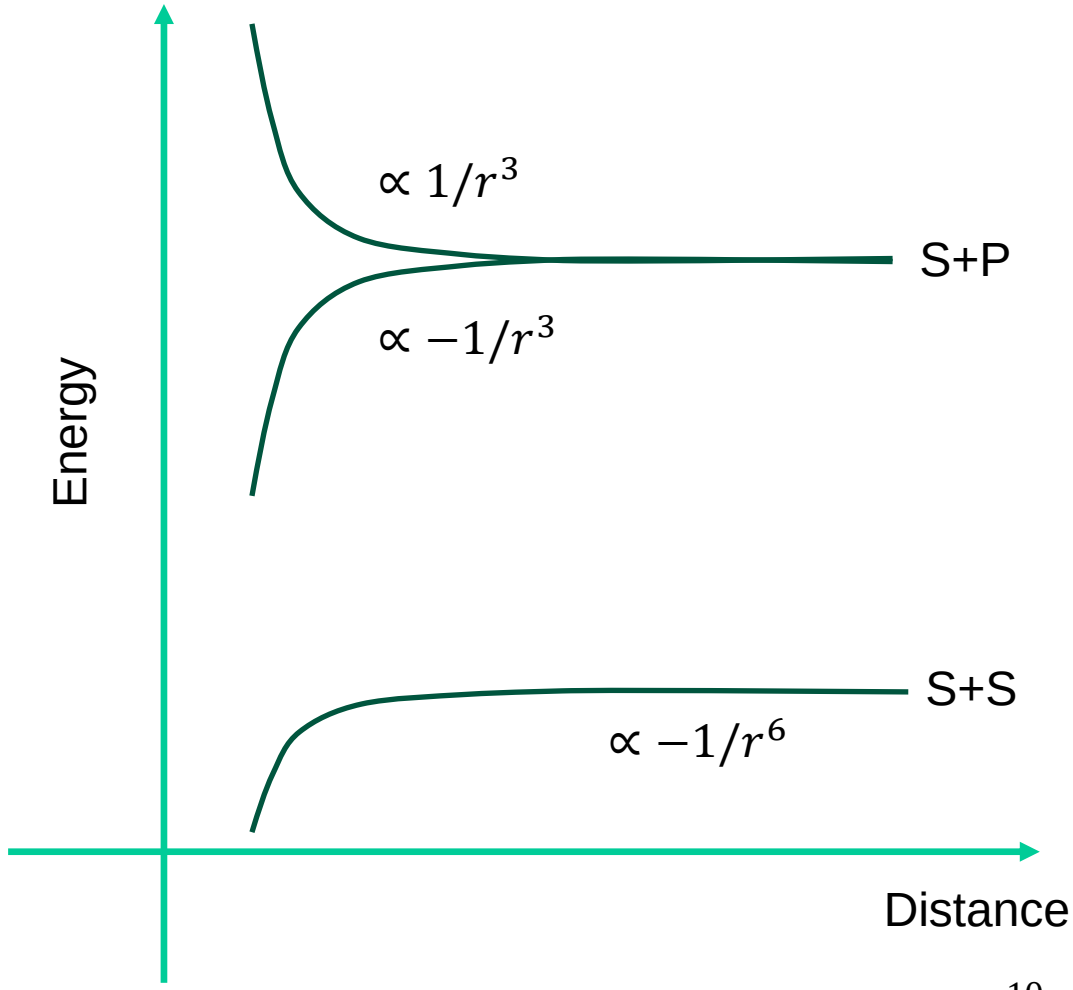
Two molecules in levels of opposite parities



Interaction potential

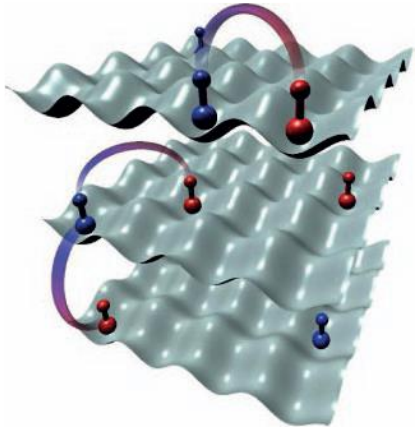


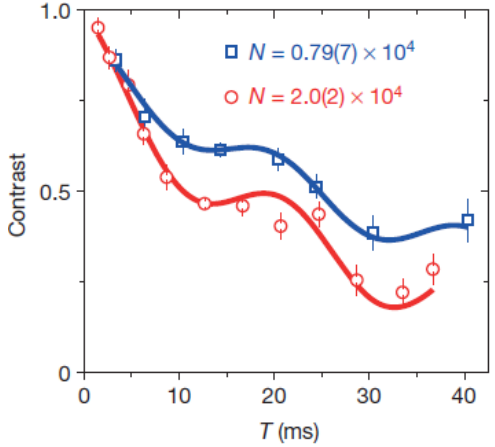
Same physics for two homonuclear atoms



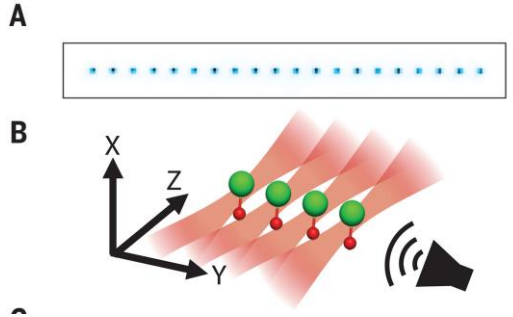
Dipolar spin exchange: spin model and entanglement

Resonant exchange of rotational excitations $H = \frac{J_\perp}{2} \sum_{i>j} V_{\mathrm{dd}}(\boldsymbol{r}_i - \boldsymbol{r}_j)\left(S_i^+ S_j^- + S_i^- S_j^+\right)$



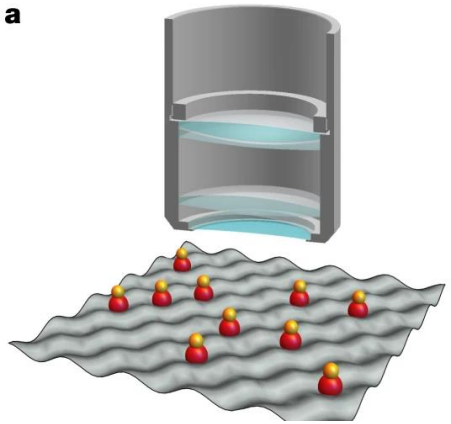


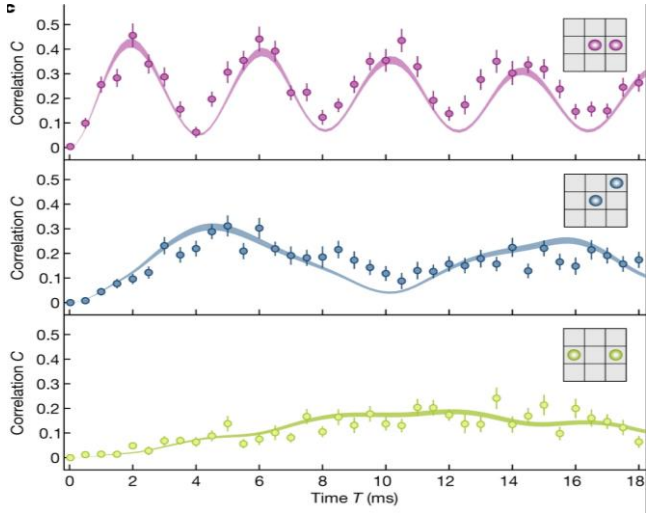
First experiment on JILA's KRb system, Nature 2013



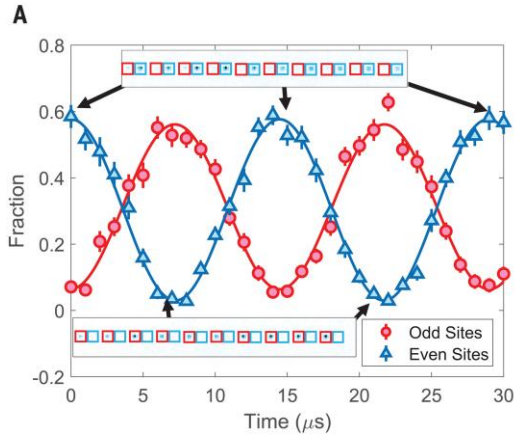
Directly laser cooled CaF molecules in OT

Doyle group, Harvard
Cheuk group, Princeton

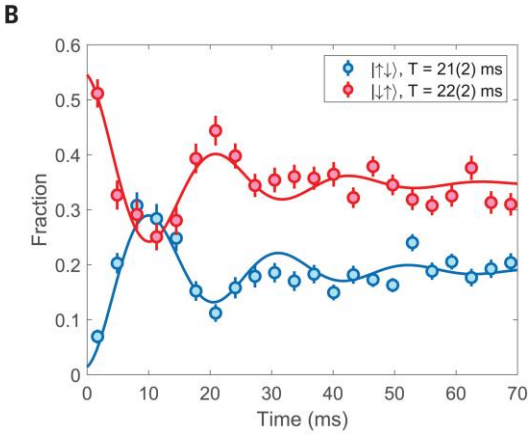




Quantum microscope, Bakr group, Nature 2023



Independent Rabi osc. at large spacing

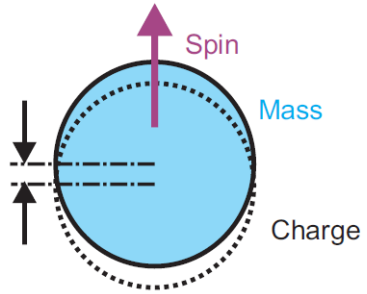


Dipolar spin exchange at small spacing

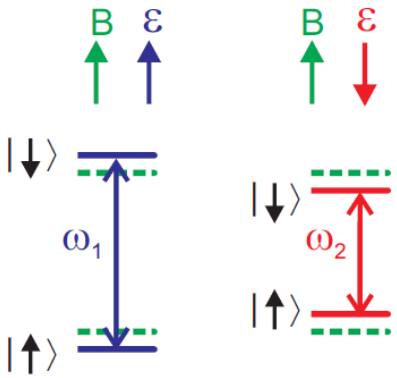
Fundamental physics with molecules

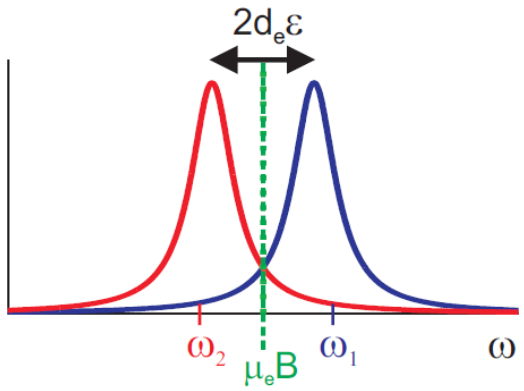
Example: Measurements of the electron electric dipole moment (eEDM) is a test of physics beyond standard model

Standard model: $d_e \sim 10^{-38}$ e cm



PhD thesis
Huanqian Loh

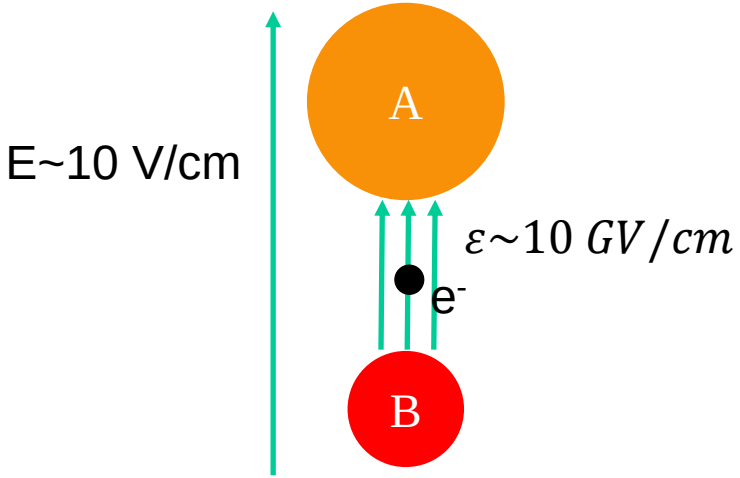




$$\delta d_e = \frac{\hbar}{2e\varepsilon\tau\sqrt{N}}$$

High ε enhances sensitivity

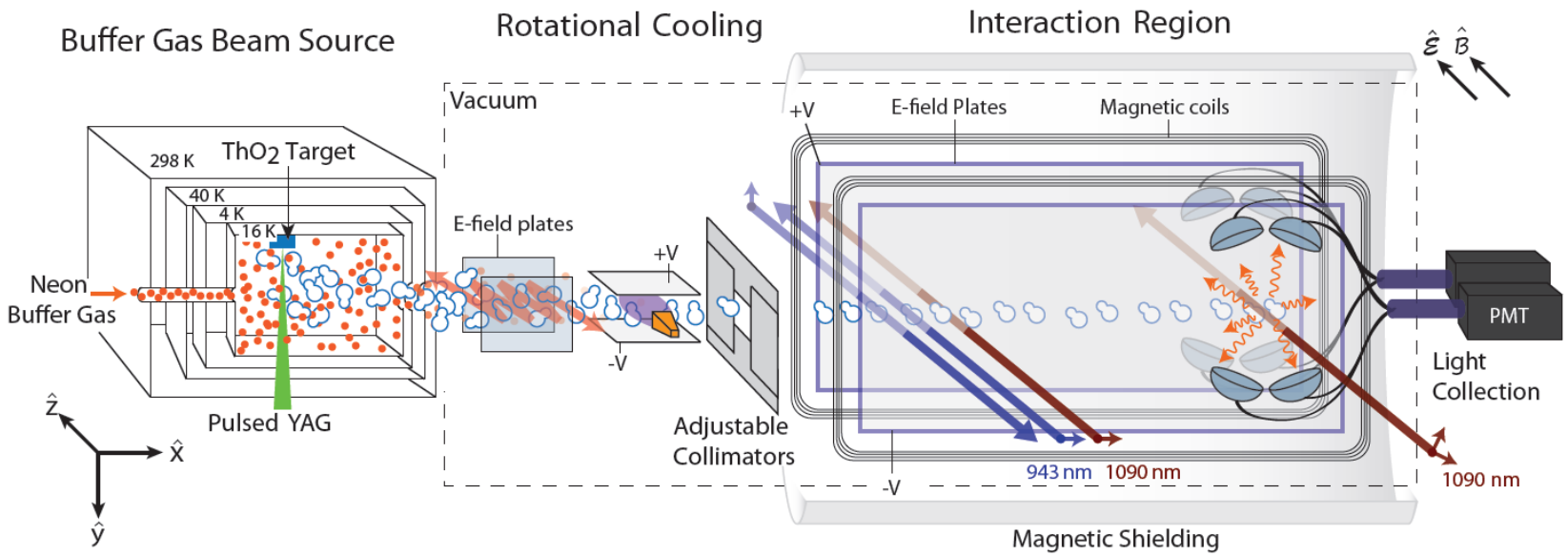
The internal E-field of polar molecules is very high



P. G. H. Sandars. 1975.

eEDM experiment with cold molecules

eEDM search with cold ThO molecule on the Ω-doublet structure of the $^3\Delta_1$ state



Science Vol. 343, 269 (2014)

ACME II result:

$d_e = (4.3 \pm 3.1_{stat} \pm 2.6_{syst}) \times 10^{-30}$

Nature 562, 355 (2018)

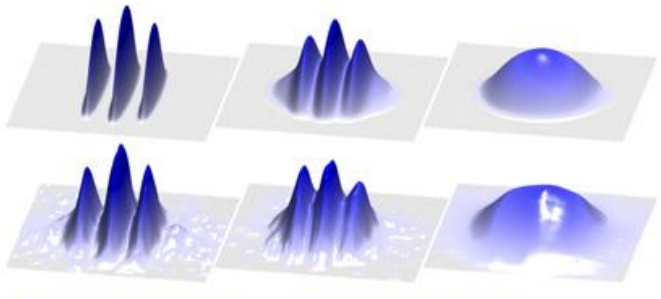
Lower T will further improve the limit by orders of magnitudes:
No Doppler effect;
Long interaction time…

$$\delta d_e = \frac{\hbar}{2e\varepsilon\tau\sqrt{N}}$$

Ultracold dipolar systems

Magnetic atoms:

Magnetic dipole moment

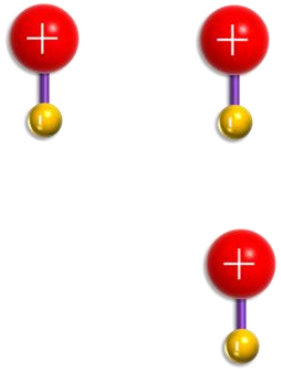


Rep. Prog. Phys. 84 012403 (2021)

- Cr (6 μ_B), Er (7 μ_B), Dy (10 μ_B)…
- Long lifetime
- BEC and DFG

Polar Molecules:

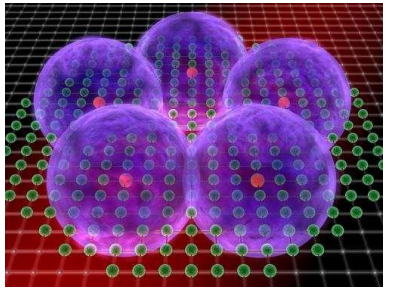
Electric dipole moment



- Many species available now: bi-alkali, CaF, SrF, YO…
- d ~ 1 to several Debye; tunable
- Long lifetime? Complicated by two-body interactions
- DFG 2019, BEC 2023

Rydberg atoms:

Giant electric dipole moment



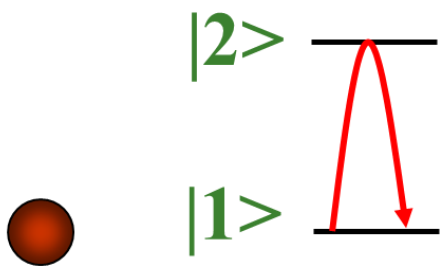
Nature 491, 87 (2012)

- d ~ 1000 Debye
- Blockade mechanism
- Short lifetime: ~ 100 μs
- Tweezer array

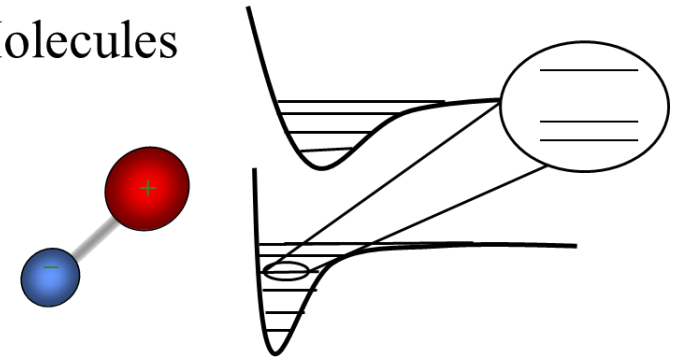
Roadblocks for molecules

1. More challenging to prepare (and detect)

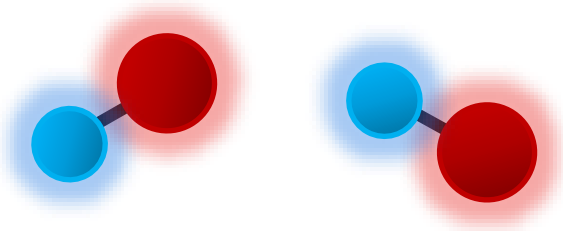
Atoms



Molecules



2. More complicated interactions



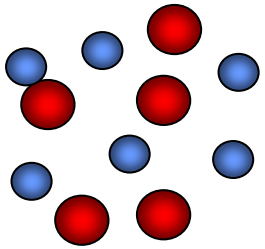
A four-atom system, with more degrees of freedom:

beyond the current computational capability

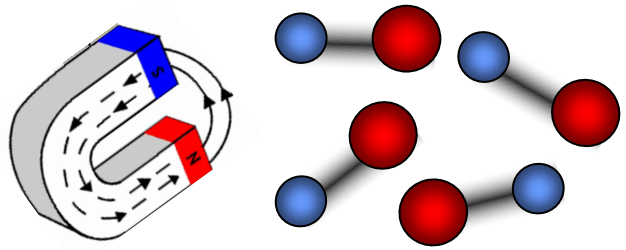
Two main production techniques

Cold atom association: KRb, RbCs, NaK, NaRb, NaCs, LiK, LiNa…

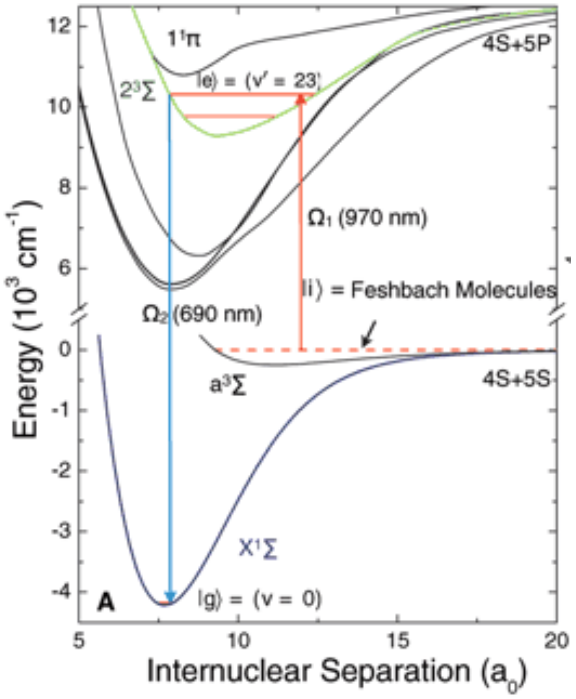
1. Prepare ultracold (< μK) mixture



2. Form heteronuclear Feshbach molecules near degeneracy

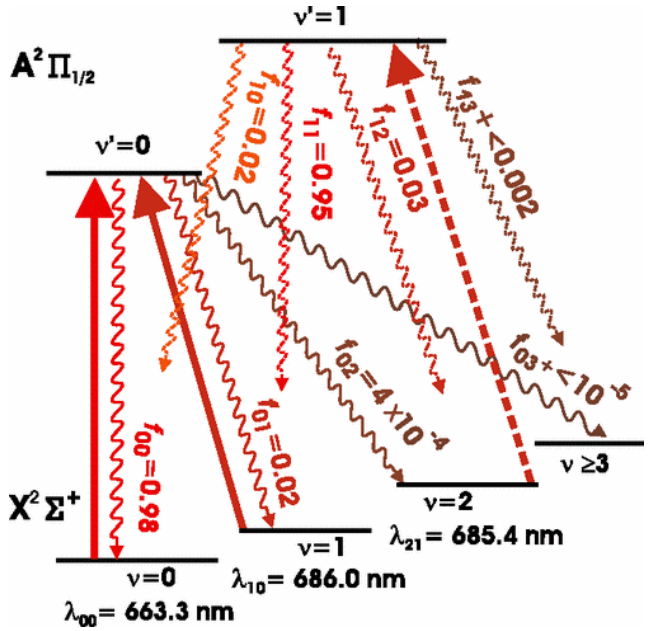


3. Transfer to ground state



Direct laser cooling:

SrF, CaF, YO, BaF, CaOH…

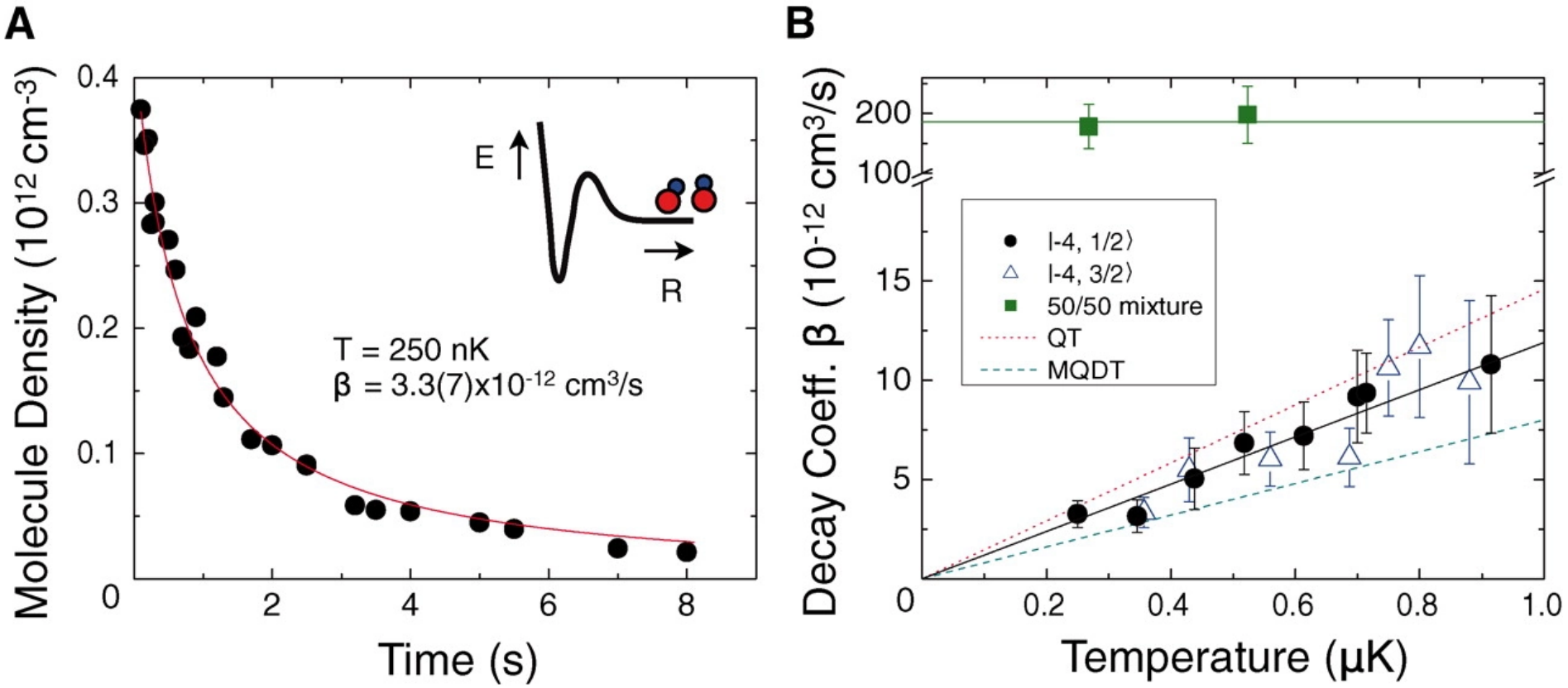


Cold and Ultracold Molecules

Insight issue *Nature Physics (2024)*

Two-body interaction between molecules

$$KRb + KRb \rightarrow K_2Rb_2 \rightarrow K_2 + Rb_2$$

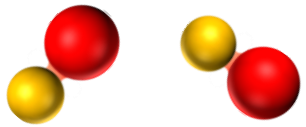


JILA, 2010

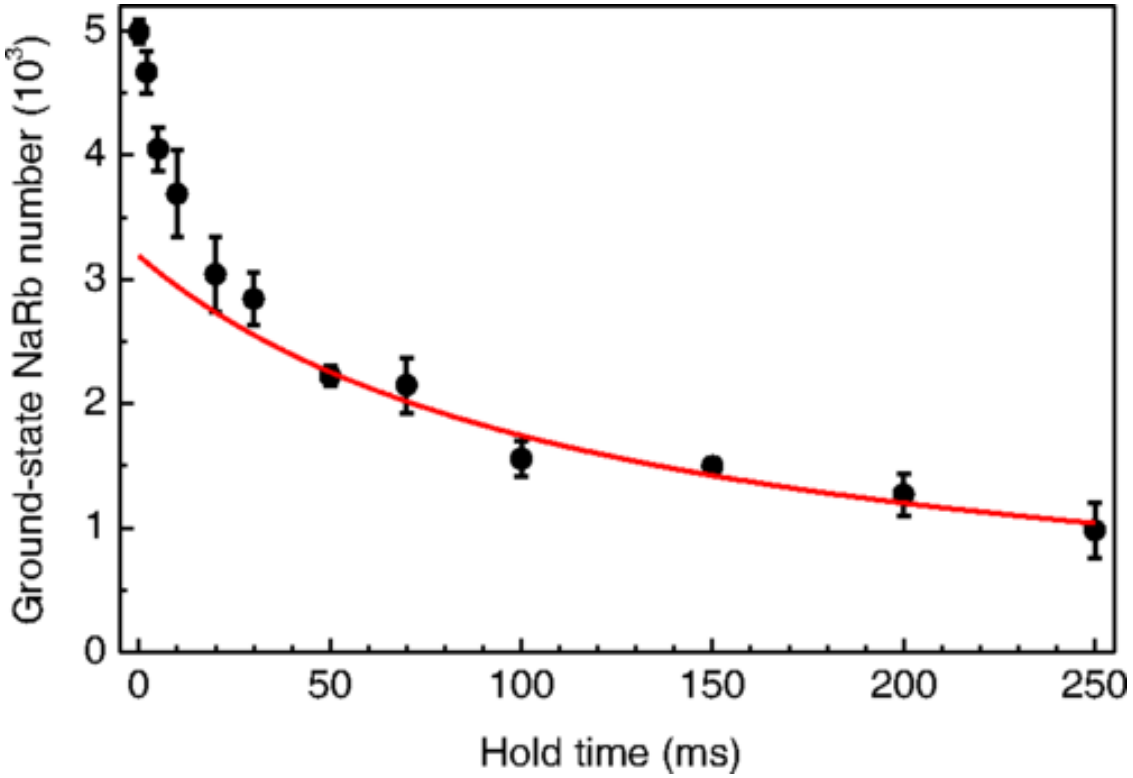
Two-body interaction between molecules

For $\mathrm{NaRb}(v = 0)$

$$NaRb + NaRb \not\rightarrow Na_2 + Rb_2$$



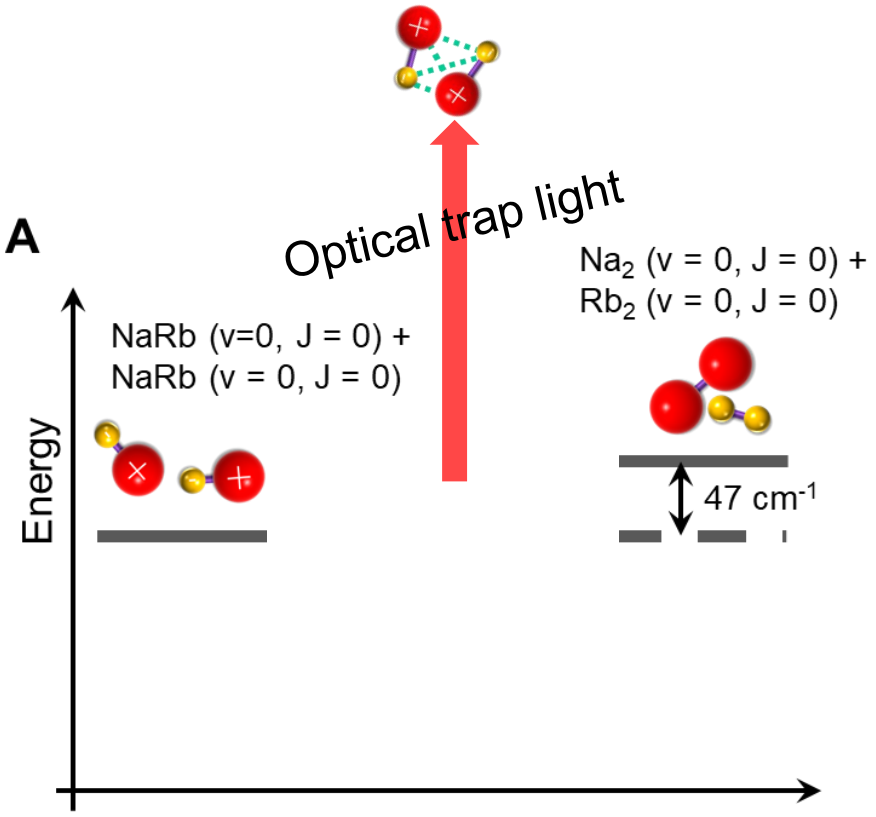
Fast loss is still observed!



M. Guo et al. Phys. Rev. Lett. **116**, 205303 (2016)

Also in RbCs, NaK, NaCs…

Complex-mediated collisions



Excitation of the complex by trapping light causes loss regardless of the chemical reactivity.

Molecular reaction dynamics, Levine, R. D. (2009).

Complex lifetime predicting by RRKM theorem

$$\tau_c = \frac{h\rho(E)}{N_0(E)}$$

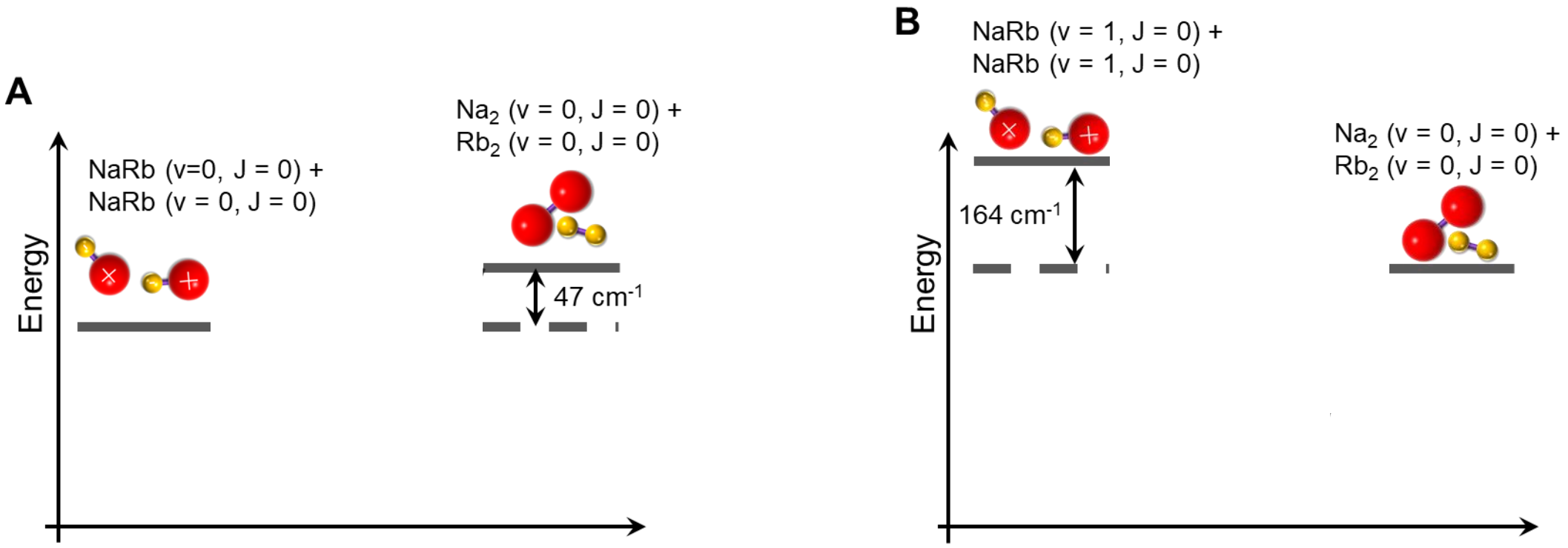
ρ: DOS of the complex at the incident energy
N_0: number of dissociation channel for complex

Ultracold ground-state molecules (non-reactive):

- High energy resolution (~100 nK)
- Well-defined reactants [both internal and external states (single partial wave)]
- $\boldsymbol{N_0 = 1}$ (back to the reactants)

Chemical reactivity control with vibrational excitation

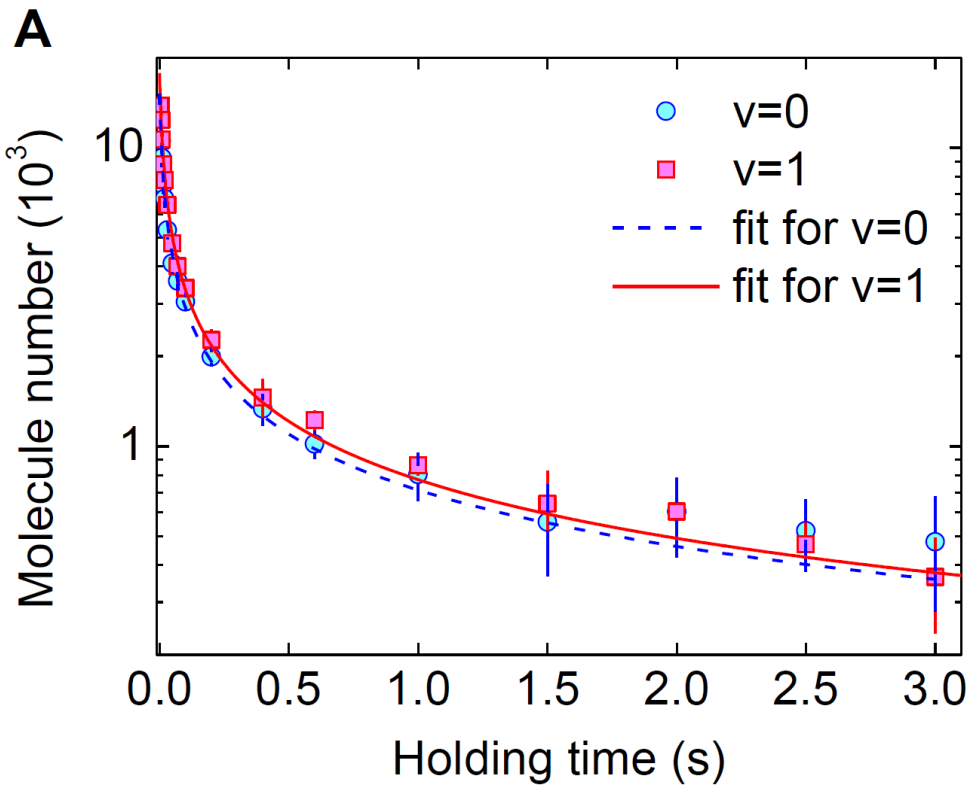
$$NaRb + NaRb \rightarrow Na_2 + Rb_2$$

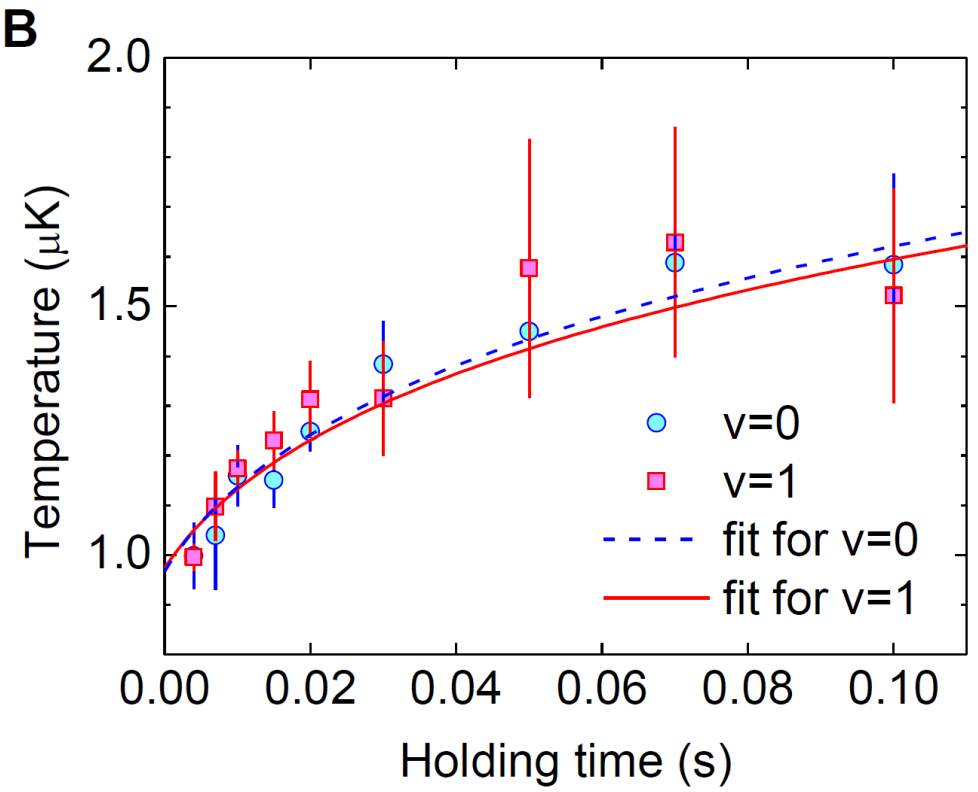


X. Ye, et al.,
Sci. Adv. 4, eaaq0083(2018)

Similar situation in NaK, NaCs and RbCs

Reactive vs. Non-reactive loss





Coupled rate equations for two-body inelastic loss

$$\frac{dN_m}{dt} = -\frac{A\beta N_m^2}{T^{3/2}}$$

$$\frac{dT}{dt} = \frac{A\beta N_m}{4T^{1/2}}$$

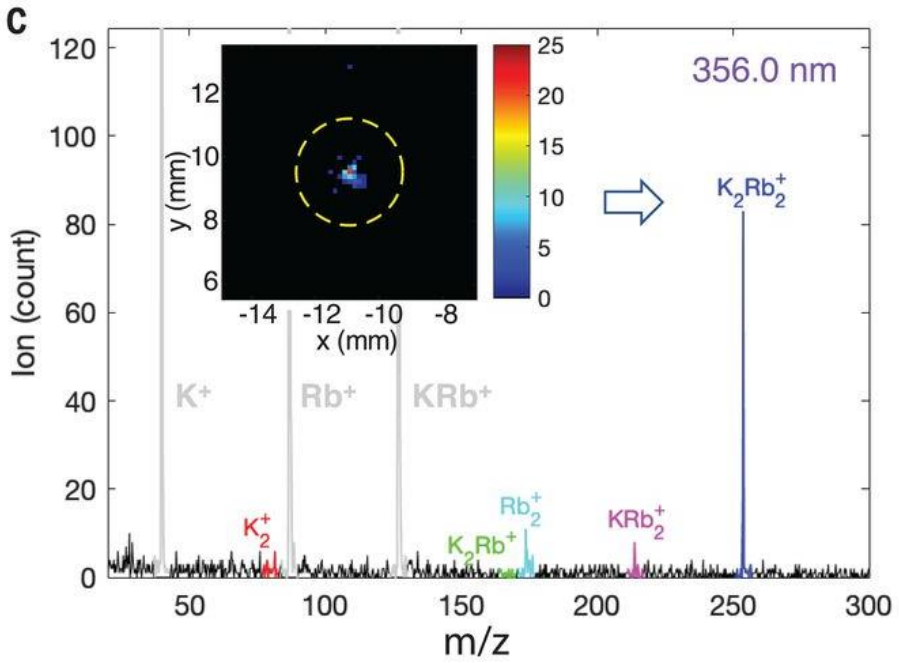
$$A = \left(\frac{m\bar{\omega}^2}{4\pi k_B}\right)^{3/2}$$

X. Ye, et al.,
Sci. Adv. 4, eaaq0083(2018)

Direct detection of the complex by ionization

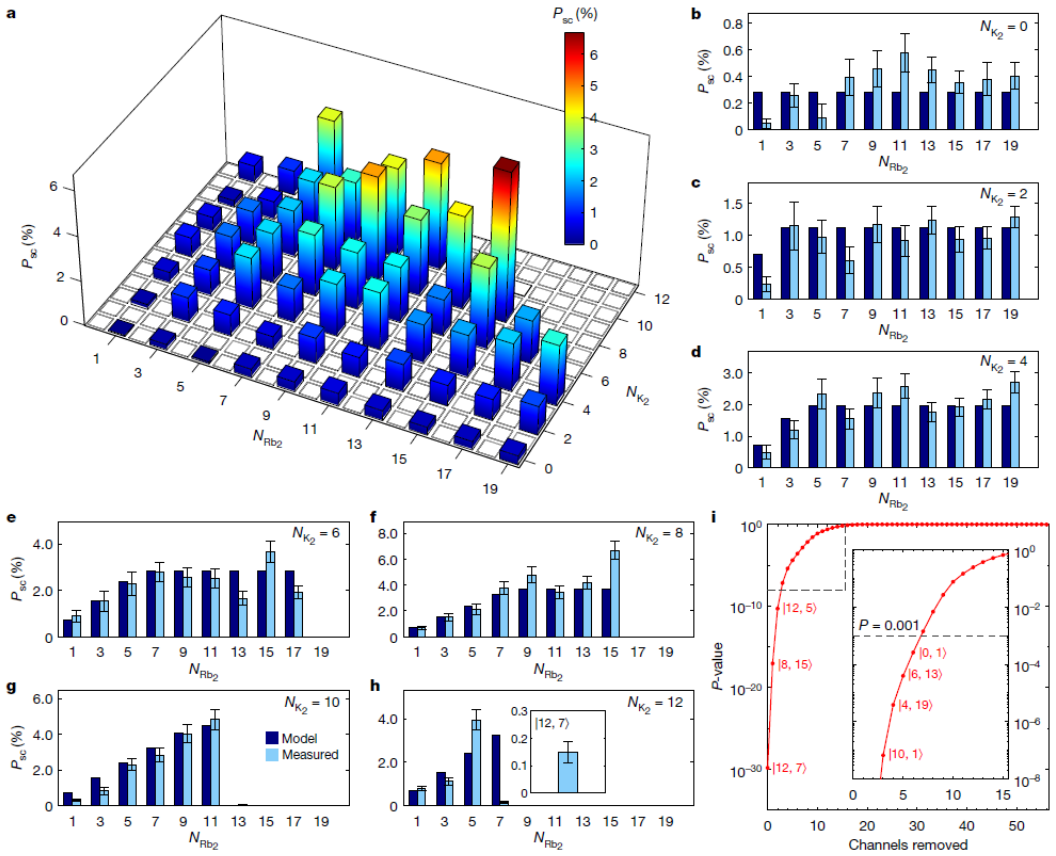
Ni group at Harvard

with ultracold $^{40}K^{87}Rb$ molecules (reactive)



Observation of the complex

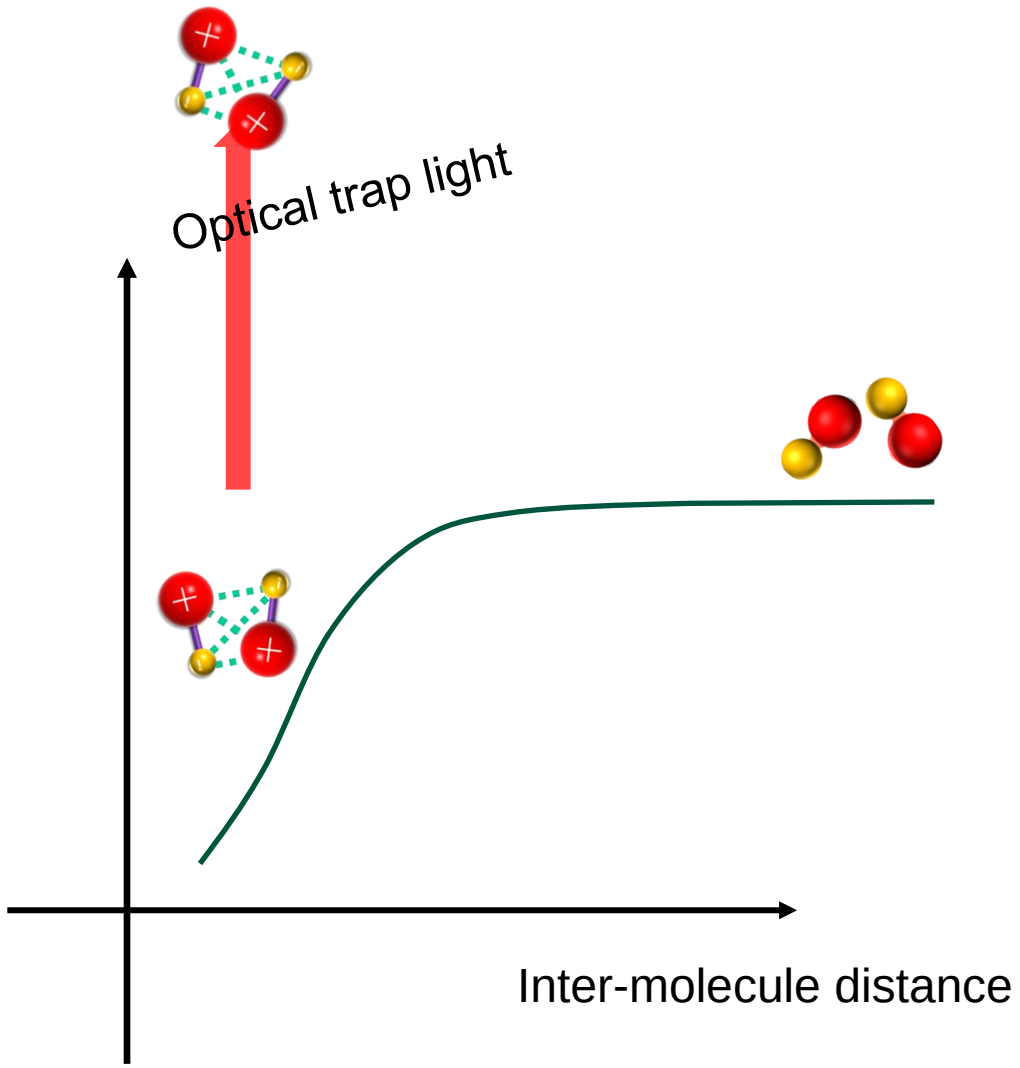
Science 366, 1111 (2019)

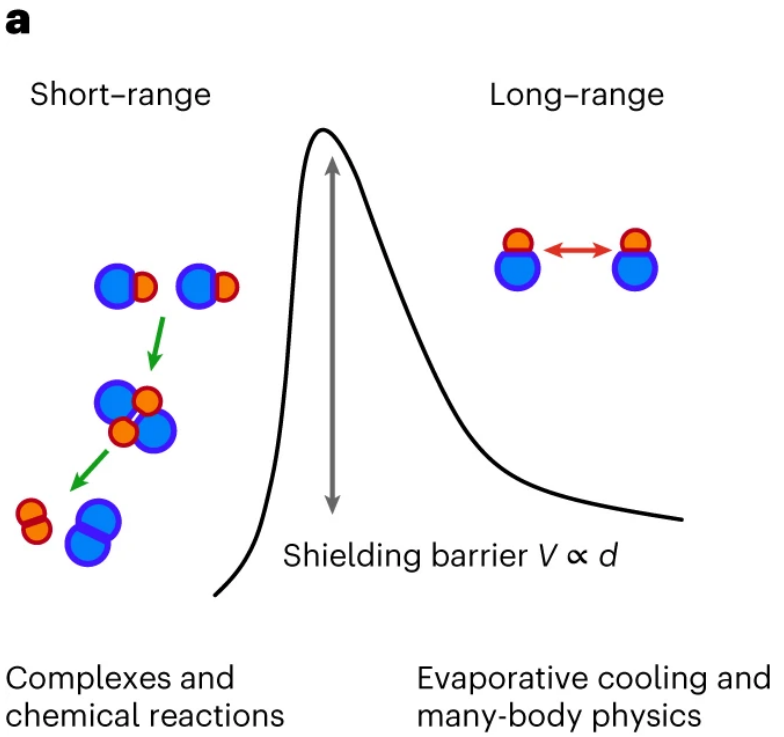


Measured product state distribution and comparison to statistical theory

Nature 593, 379 (2021)

Loss suppression by engineered long-range barrier





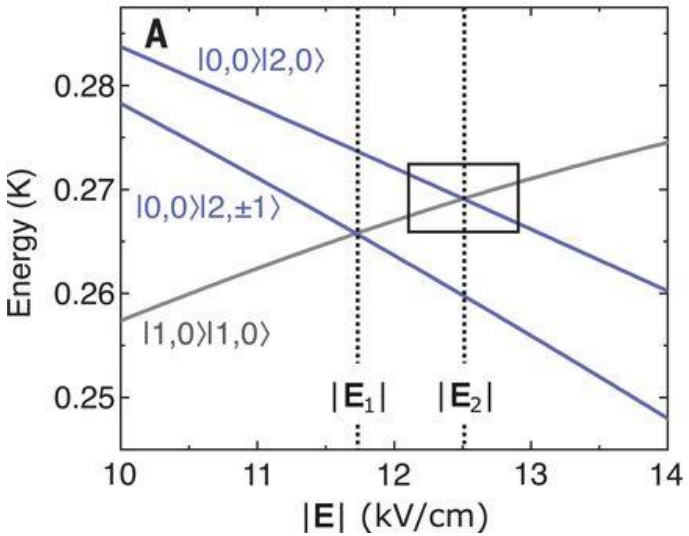
Nature Physics (2024)

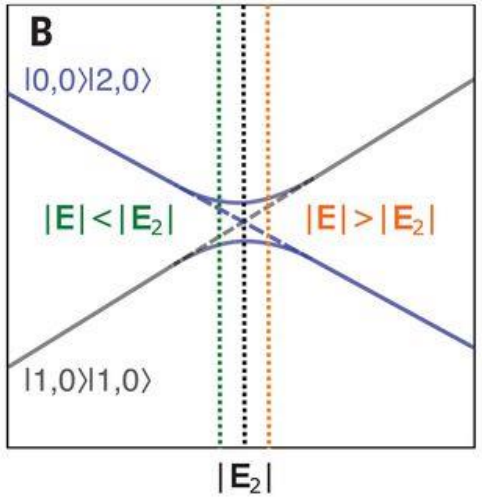
DC field induced rotational Förster resonance

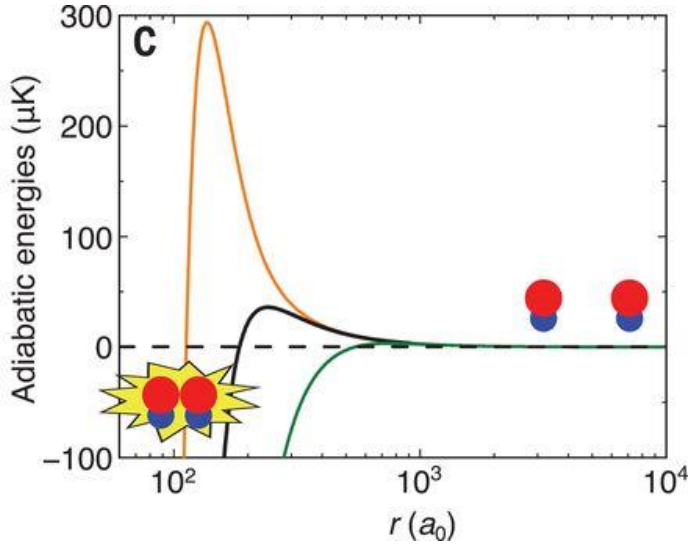
DC Stark shift of two KRb molecules

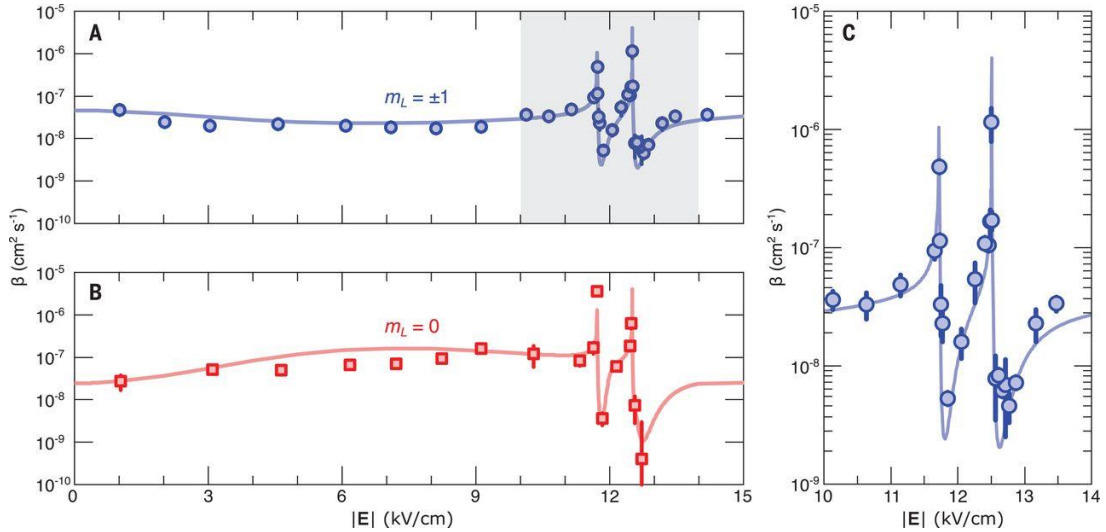
Förster resonance between $|1,0\rangle|1,0\rangle$ *and* $|0,0\rangle|2,0\rangle$

Barrier in the long range







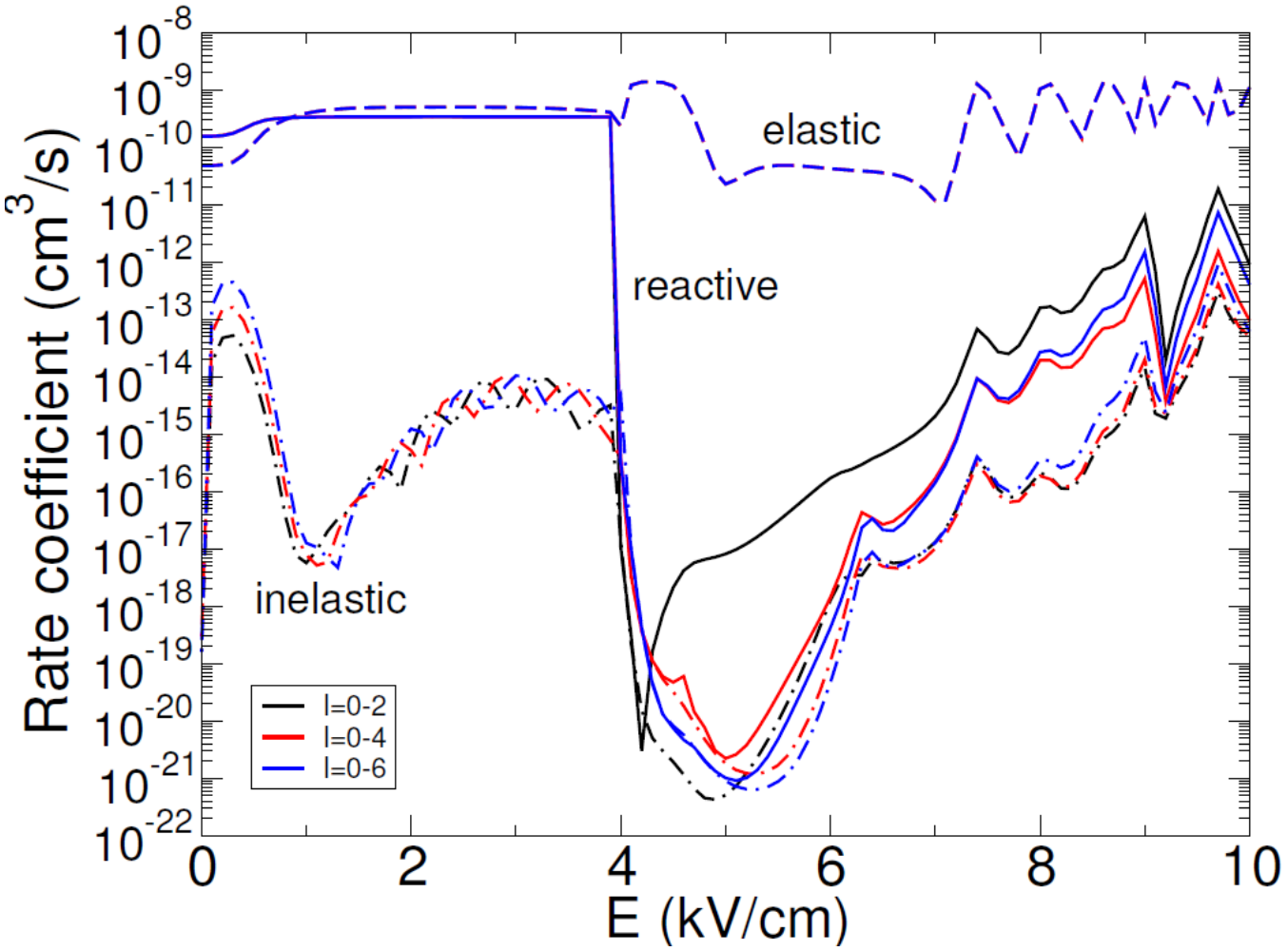


Resonant collisional shielding of loss

Science, 2020

DC field induced rotational Förster resonance

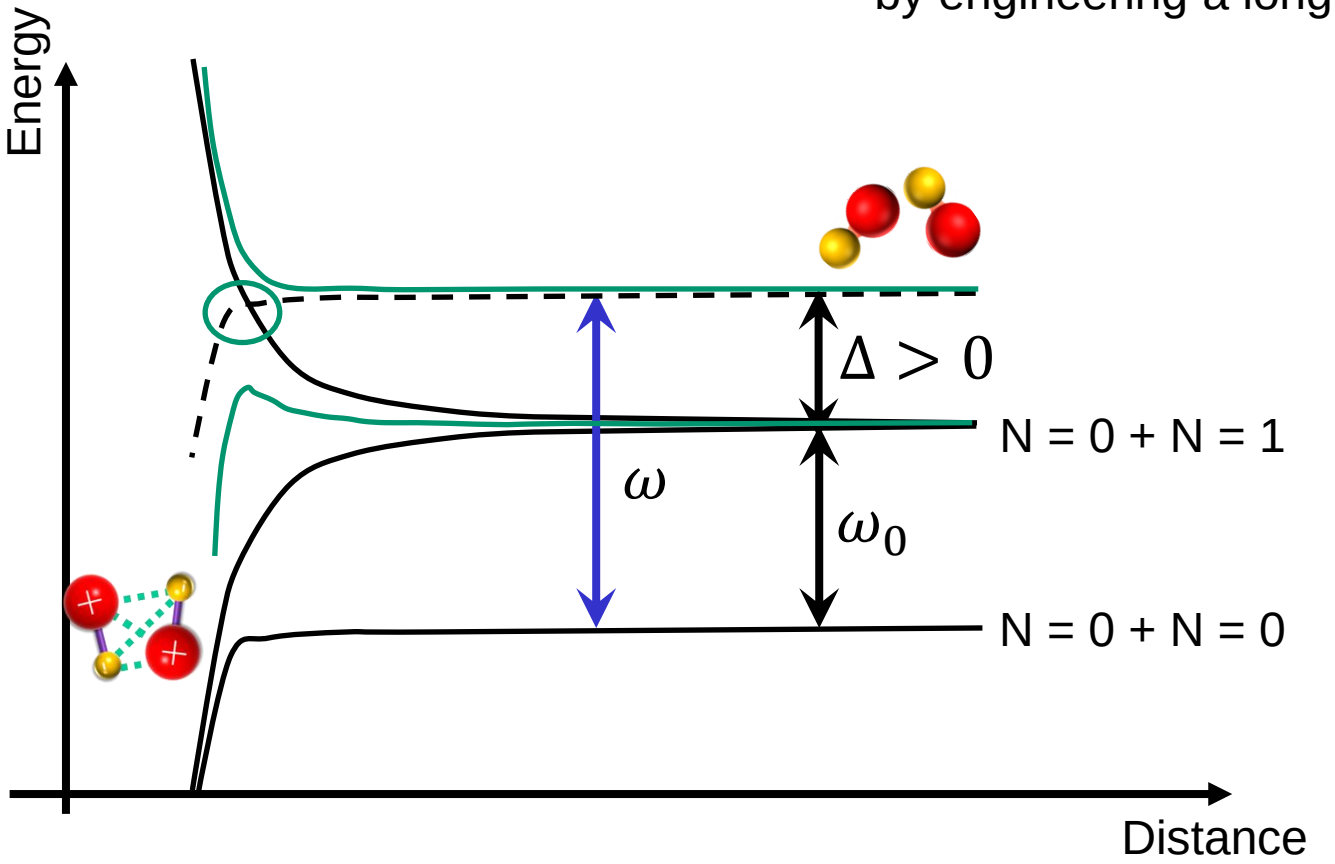
Alao works for other molecules, including NaRb

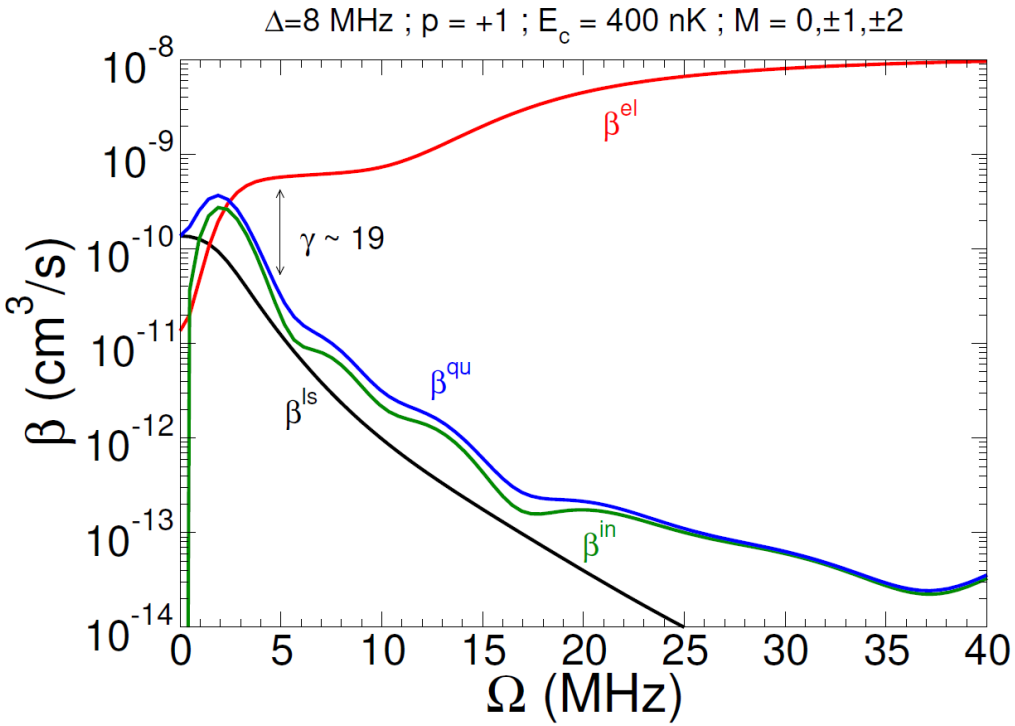


From Goulven Quemener

Microwave shielding

Microwave loss suppression
by engineering a long-range barrier



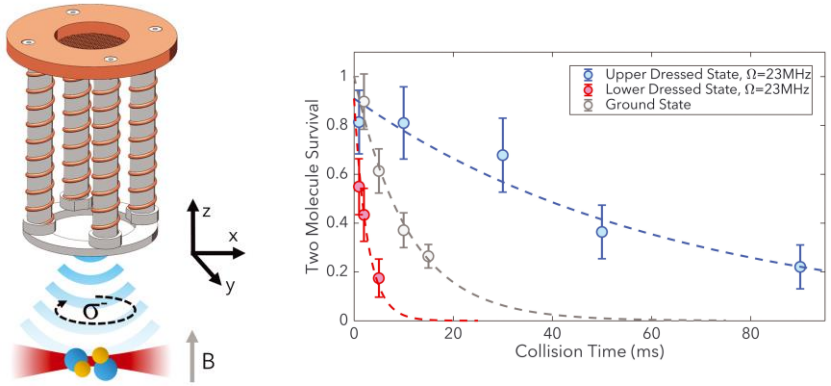


By courtesy of G. Quemener

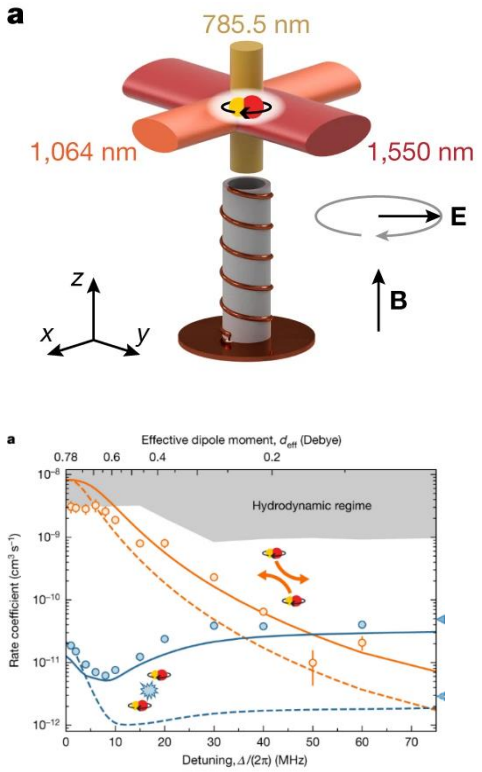
G. Quemener et al., PRL, 2018
Karman et al., PRL, 2018

Need high Rabi frequency, high purity circular polarization

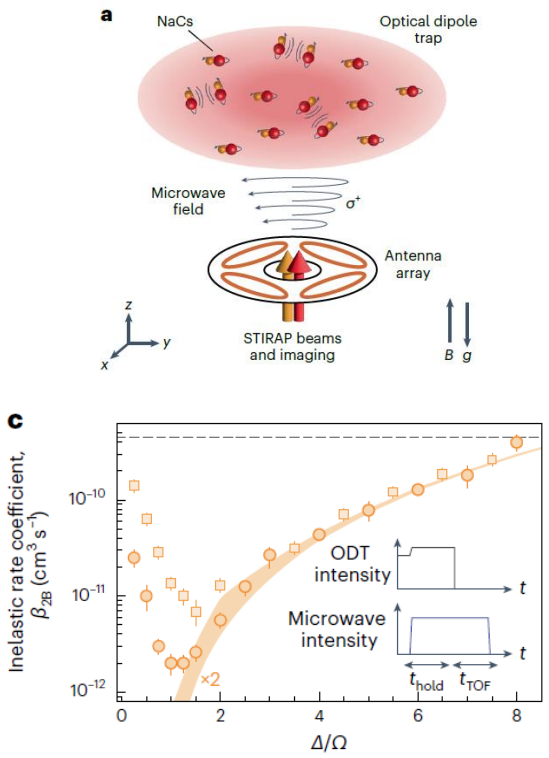
Experiments on microwave loss suppression



CaF in optical tweezer (Harvard and MIT)
Science, 2021

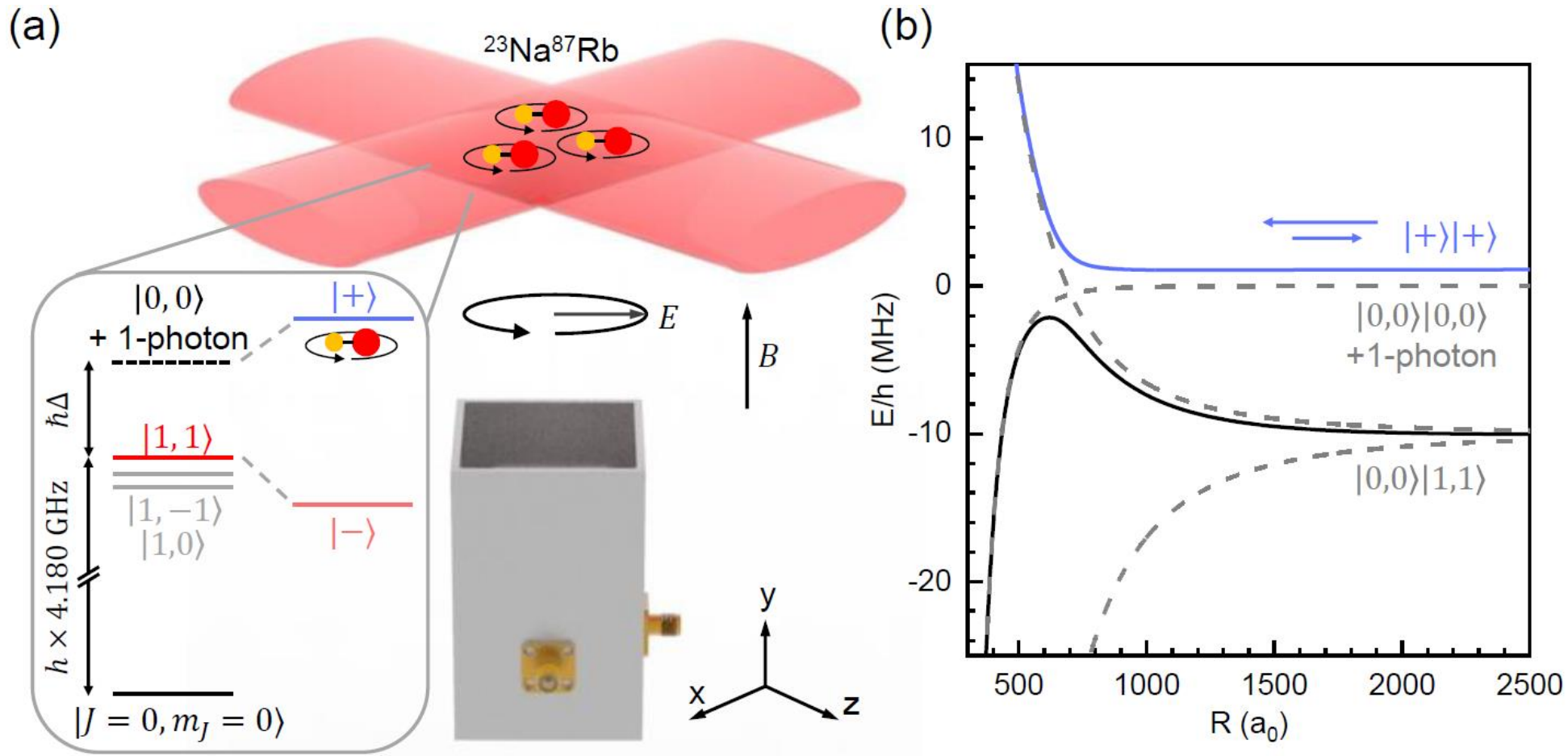


$Na^{40}K$ evaporative cooling to DFG
Nature, 2022
Field-linked state, Nature 2023
(MPQ)



NaCs loss suppression
Nat. Phys. 2023
(Columbia)

NaRb molecule with microwave dressing

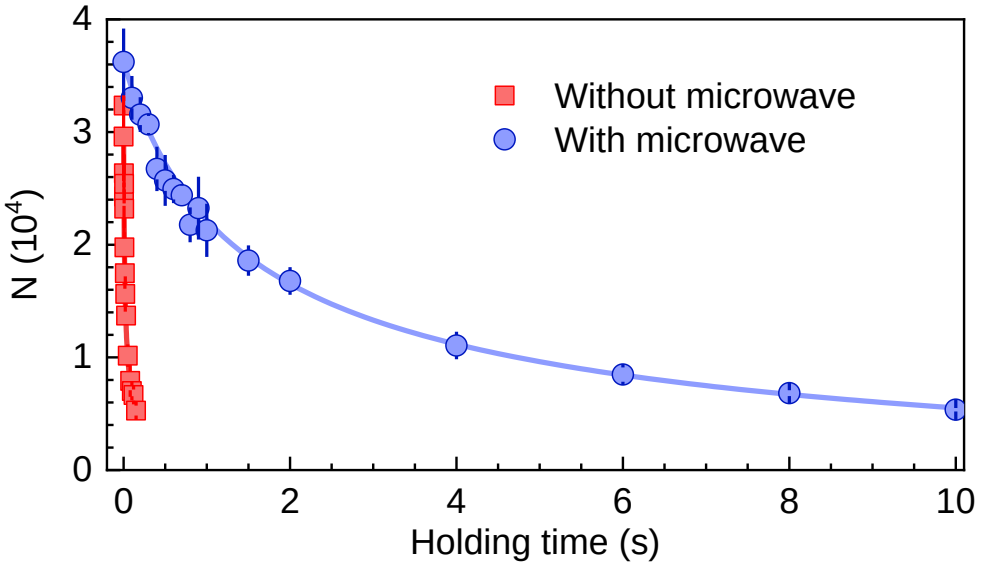


Need high Rabi frequency, high purity circular polarization

Loss suppression

Density: $3 \times 10^{11}\ cm^{-3}$
Temperature: 600 nK

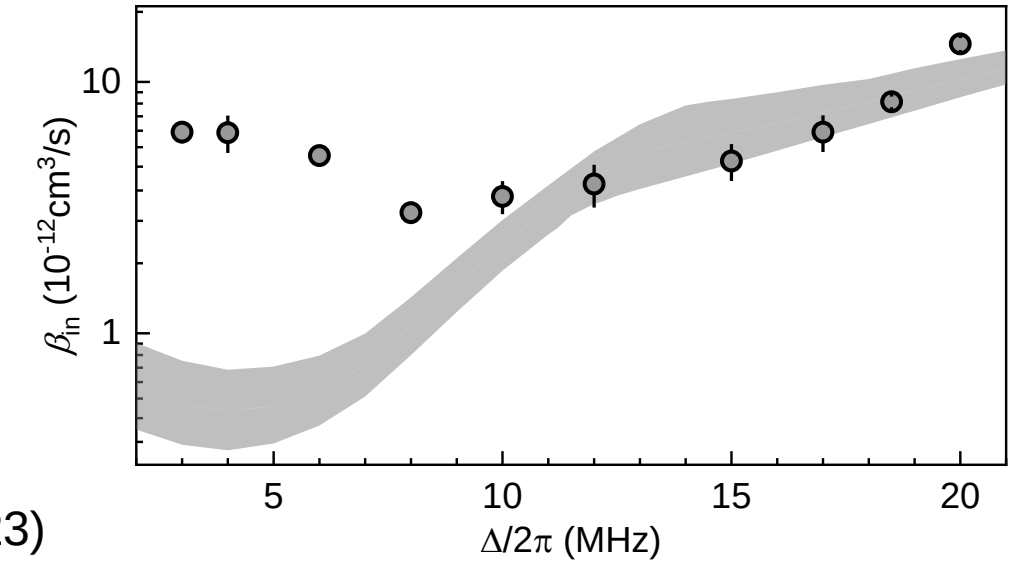
$\Omega = 2\pi \times 10$ MHz
$\Delta = 2\pi \times 7$ MHz



$$\frac{dn}{dt} = -\beta_{in} n^2 - \Gamma n$$

Bare sample:
$\beta_{in} \sim 3.4 \times 10^{-10} cm^3/s$

Best with MW:
$\beta_{in} \sim 3.0 \times 10^{-12} cm^3/s$

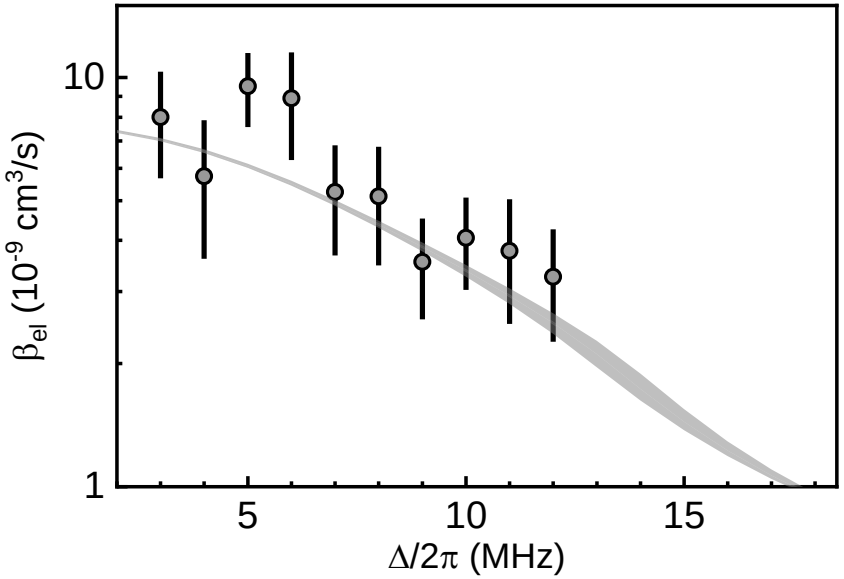


Theory:
coupled-channel scattering calculation

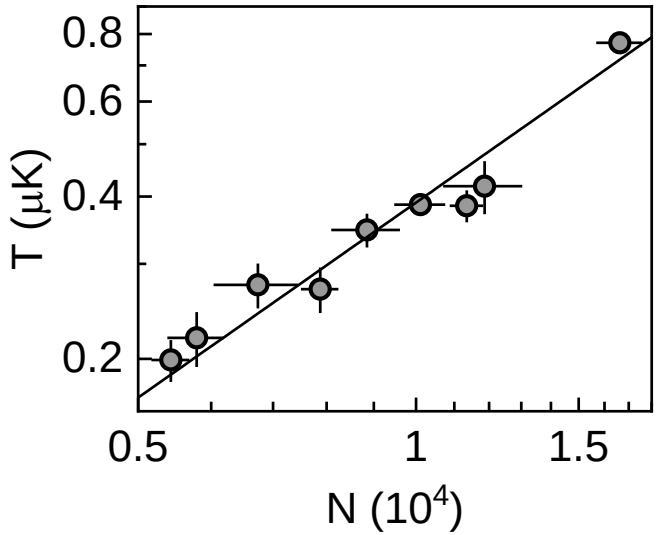
Junyu Lin et al. PRX (2023)

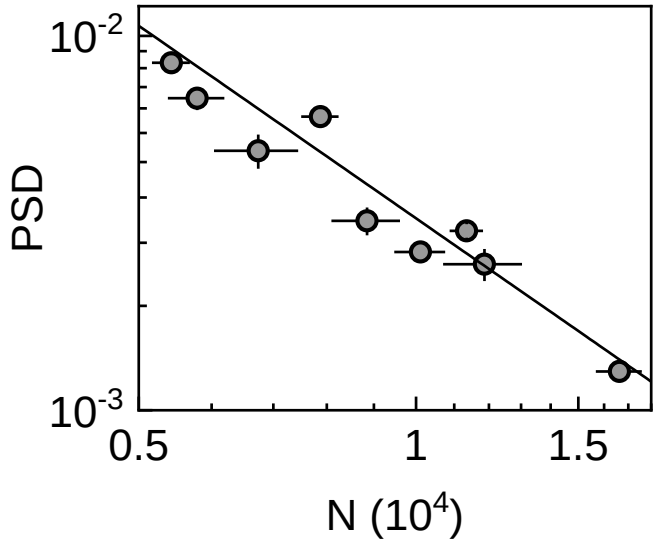
Elastic collision and evaporative cooling

Junyu Lin et al. PRX (2023)



DDI results in very large β_{el}



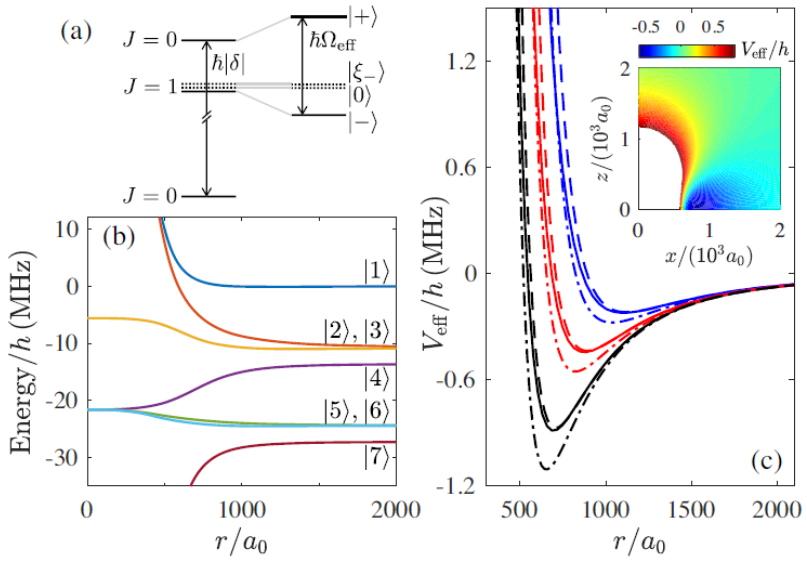


Temperature decrease and phase-space density increase are observed

Influence of filed-linked tetramer state

Problem at small detuning: 3-body loss of molecules due to field-linked states

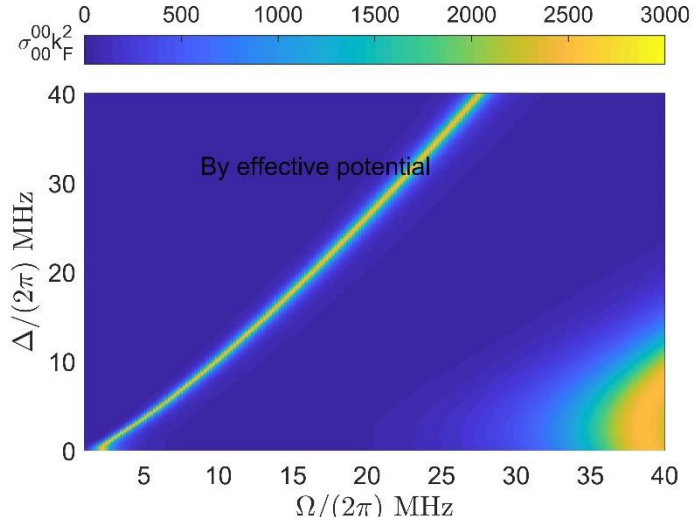
John Bohn: RRL, 2008

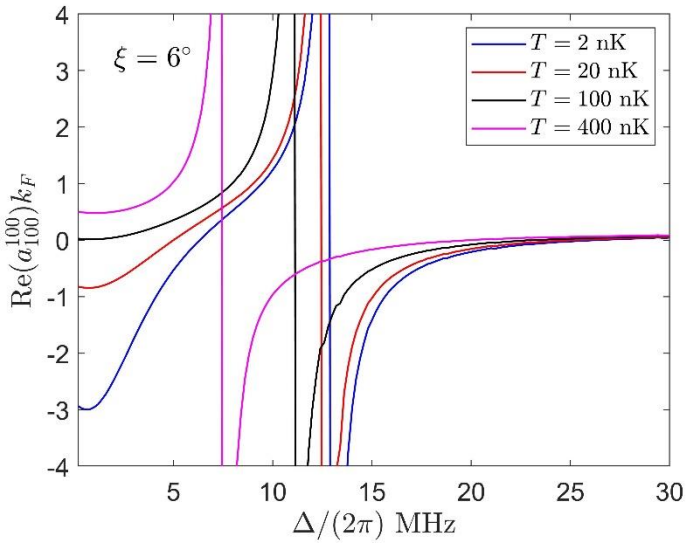


$$V_{\rm eff}(\mathbf{r}) = \frac{C_6}{r^6}\sin^2\theta\{1-\mathcal{F}_\xi^2(\varphi)+[1-\mathcal{F}_\xi(\varphi)]^2\cos^2\theta\}$$
$$+\frac{C_3}{r^3}[3\cos^2\theta-1+3\mathcal{F}_\xi(\varphi)\sin^2\theta],$$

Tao Shi, Su Yi et al. (2023)

Experimental observation by Xinyu Luo et al in MPQ, Nature 2024





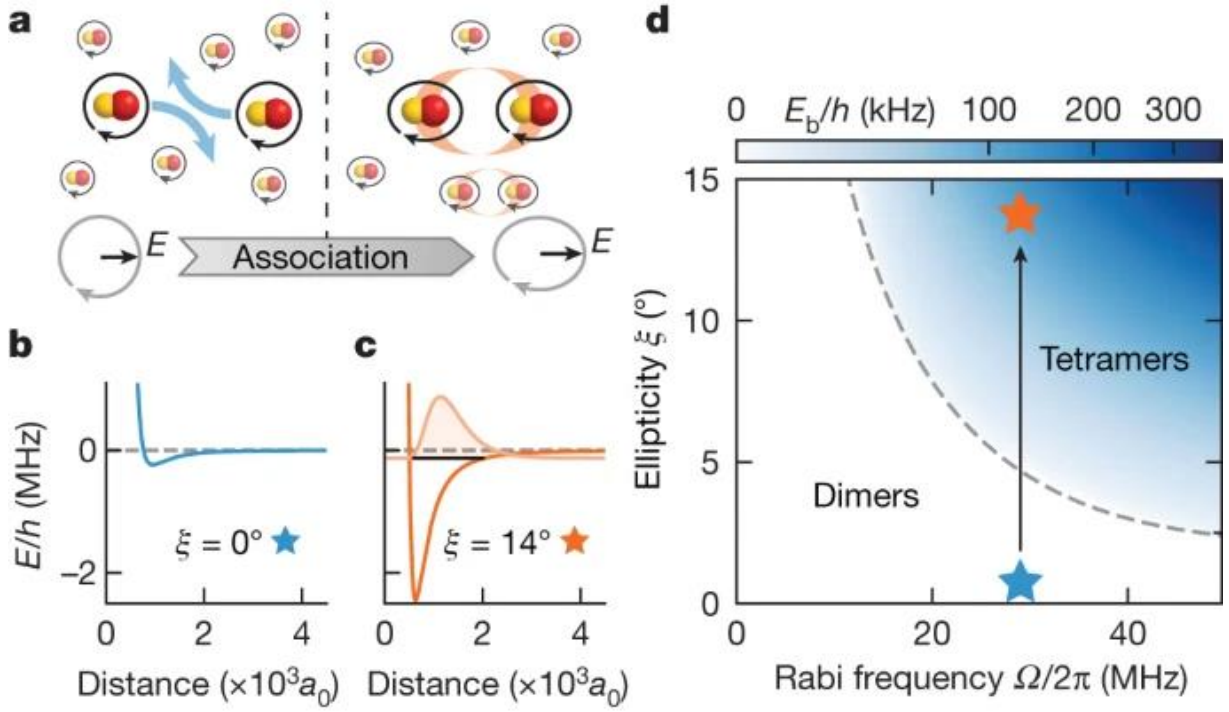
For NaRb

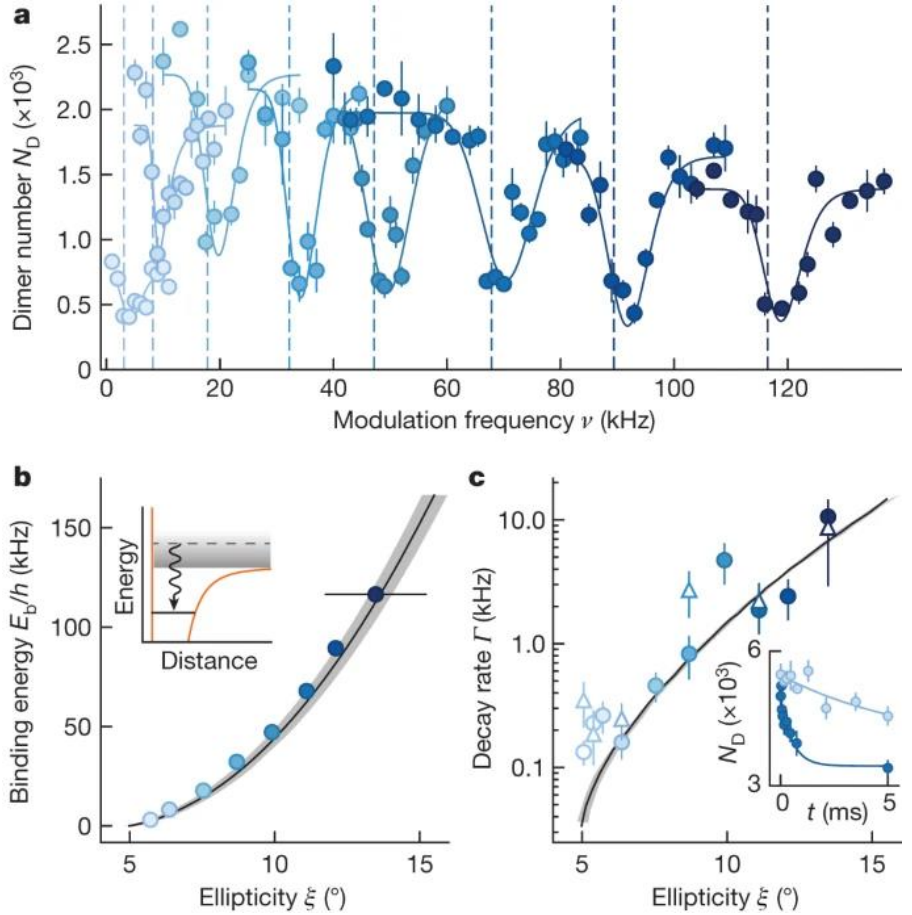
by Tao Shi

Filed-linked tetramer state

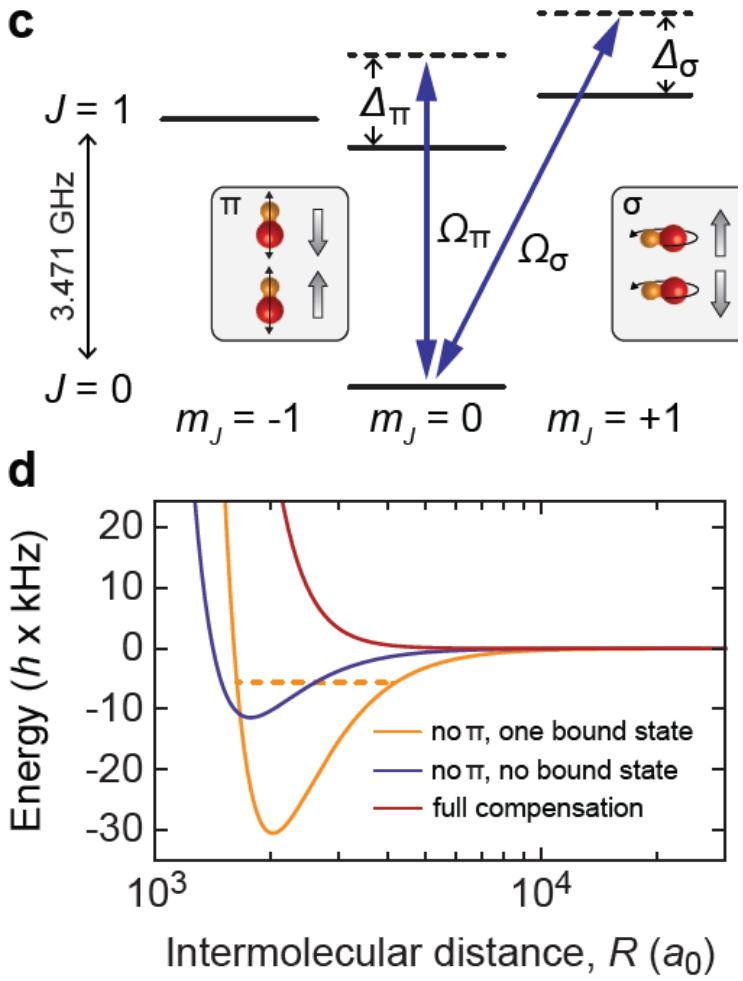
Experimental observation by Xinyu Luo et al in MPQ, Nature 2024

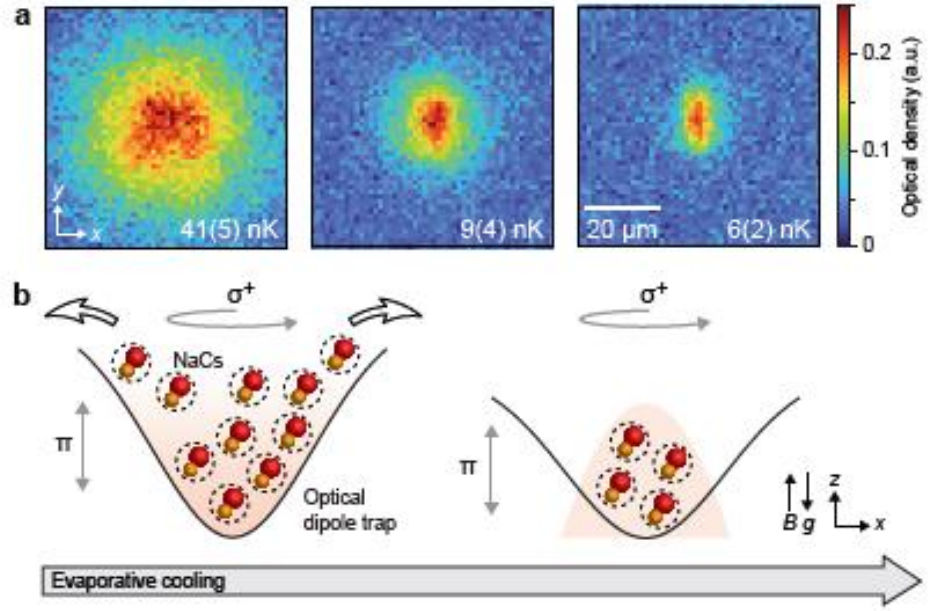
With microwave dressed $Na^{40}K$ molecules

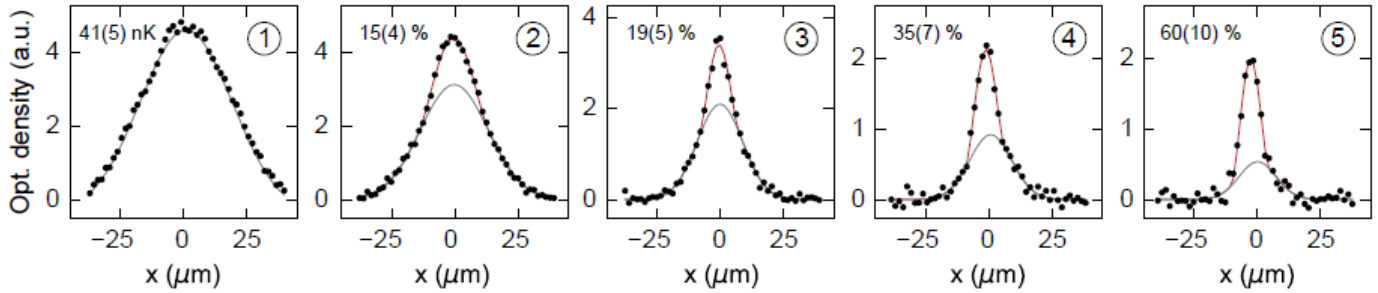




BEC of polar molecules with double MW dressing







arXiv: 2312.10965

NaCs molecule, Will group at Columbia

Summary and outlook for polar molecules

- Both association of cold atoms and direct cooling work well for creating ultracold ground-state polar molecules
- The loss has deep connections with chemical reaction
- Dipolar interaction can be used for loss suppression and interaction engineering
- With the achievements of DFG and BEC, more and more interesting results are expected
- Quantum microscope and optical tweezers offer even more possibilities

Only a small portion of the recent results are included in this talk

Cold molecule groups

Association

Molecule	PI	location
KRb	Ye	JILA
RbCs/AgFr	Demille	Chicago
RbCs/KCs	Naegerl	Innsbruck
RbCs/CsYb	Cornish	Durham
NaK	Zwierlein	MIT
NaRb	Wang	CUHK
NaLi	Ketterle	MIT
NaK	Zhao/Pan	USTC
NaK	Luo/Bloch	MPQ
NaK	Ospelkaus	Hannover
KRb/NaCs	Ni	Harvard
NaCs	Will	Columbia
NaCs	Bigelow	Rochester
LiCs	Weidemuller	Heidelberg
LiK	Dieckmann	NUS
NaRb	Bakr	Princeton
NaK	Park	Postech
YbLi	Gupta	Washington
SrRb	Schreck	Amsterdam
NaCs	Jia	Shanxi

Direct laser cooling

Molecule	PI	location
SrF	Demille	Chicago
CaF/CaOH	Doyle	Harvard
CaF/YbF	Tarbutt	ICL
CaF	Ospelkaus	Hannover
CaF	Cheuk	Princton
YO	Ye	JILA
BaF	Yan	Zhejiang
BaF	Langen	Stuttgart
AlCl	McCarron	Uconn
MgF	Yin	ECN Shanghai
BaH	Zelevinsky	Columbia
YbOH	Hutzler	Caltech

There are also other methods (not included)

Theory

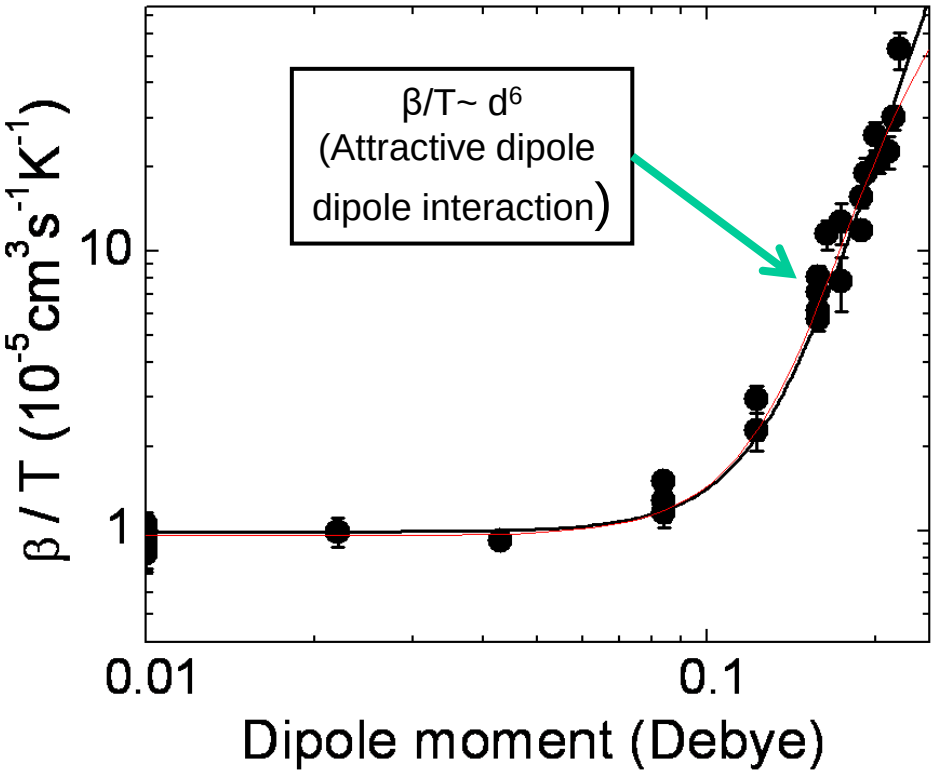
PI	location
Dulieu	LAC
Quemener	LAC
Bohn, Rey	JILA
Karman	Radboud
Hutson	Durham
Tomza	Warsaw
Idziaszek	Warsaw
Żuchowski	UMK, Poland
Cong/Wang	Dalian
Zhang	Dalian
Shi, Yi	IOP, CAS



Thanks for your attention!

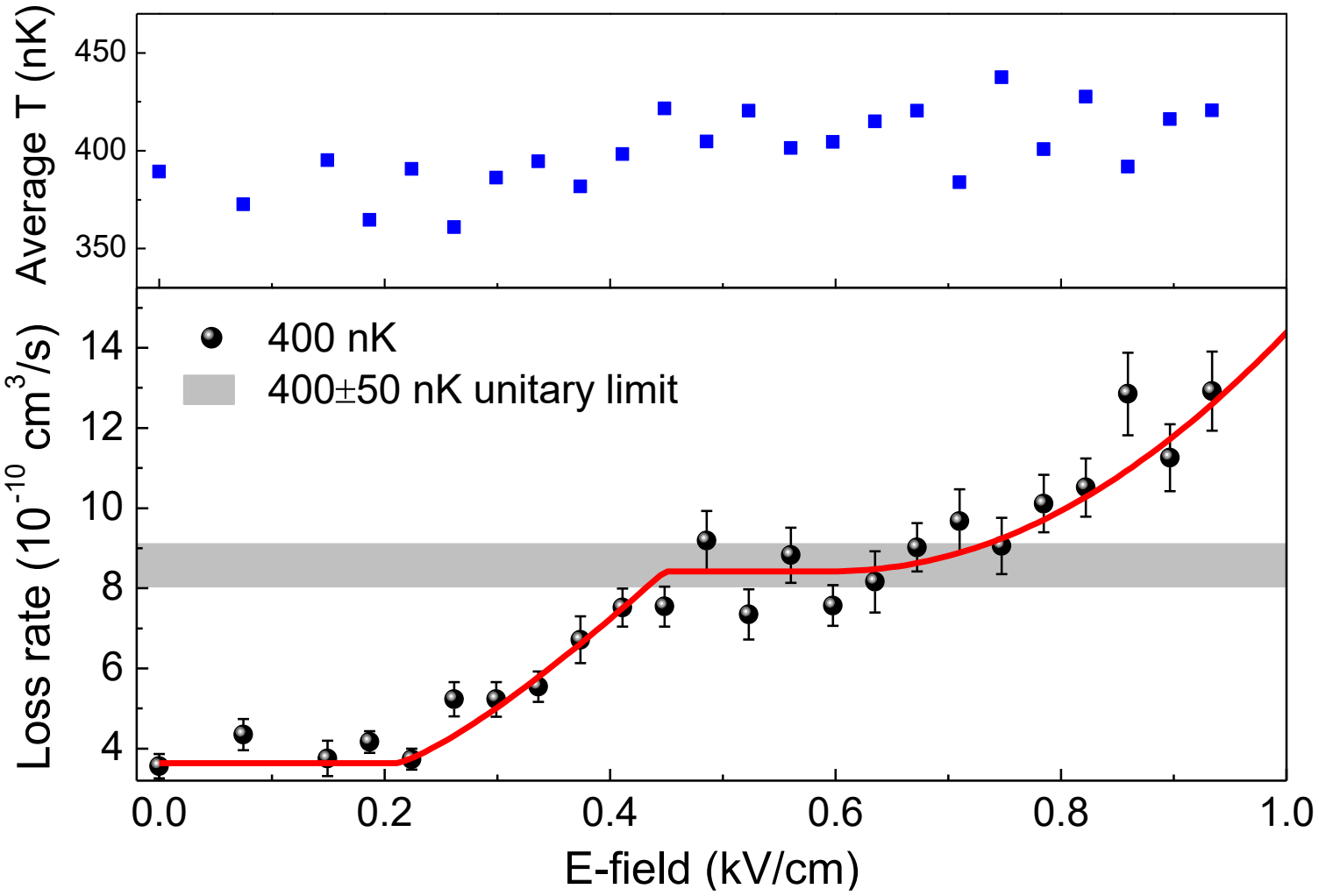
Dipolar collision of $^{40}K^{87}Rb$

For bulk sample, dipolar collisions accelerate the chemical reaction



Ni, et al. Nature(2010)

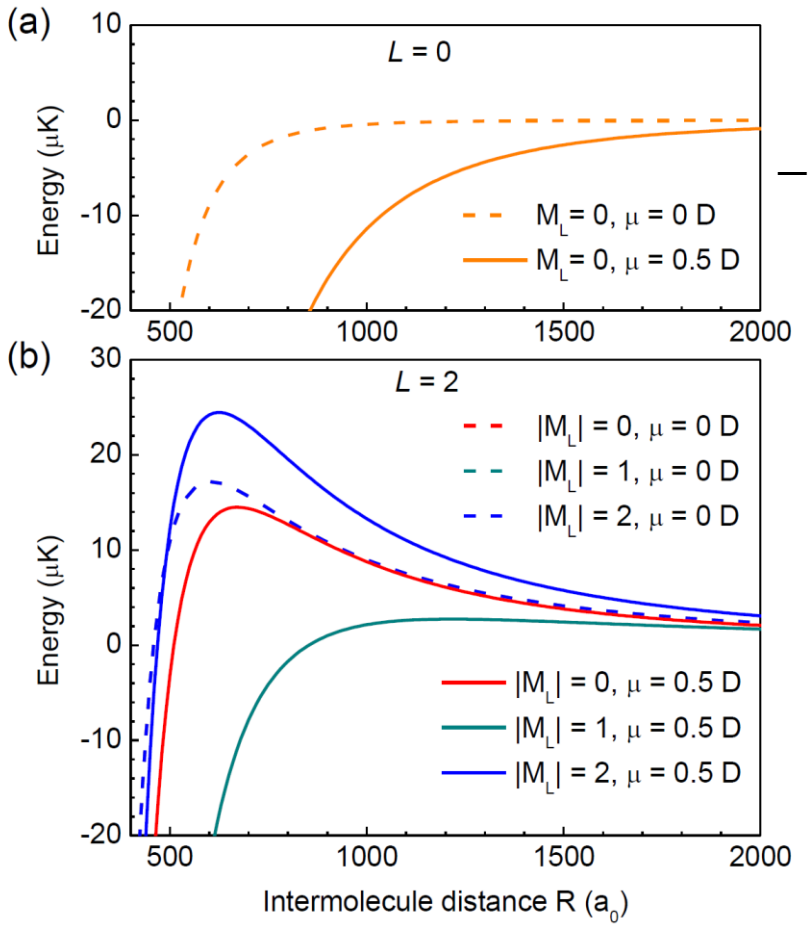
Dipolar collisions of NaRb



Dipolar collision for NaRb

- Effective potential:

$$\langle LM_L|\,V(R)\,|L'M'_L\rangle = \left\{\frac{\hbar^2 L(L+1)}{mR^2} - \frac{C_6}{R^6}\right\}\delta_{L,L'}\delta_{M_L,M'_L} - \frac{C_3(L,L';M_L)}{R^3}\delta_{M_L,M'_L},$$



$-\frac{C_\alpha}{r^\alpha}$

- Modify the "s-wave" potential shape

Char. length $r_\alpha = \left(\frac{mC_\alpha}{\hbar^2}\right)^{1/(\alpha-2)}$

Char. energy $E_\alpha = \frac{\hbar^2}{mr_\alpha^2}$

- Modify the "*d*-wave" barriers: anisotropic nature of DDI

Analysis by partial wave

