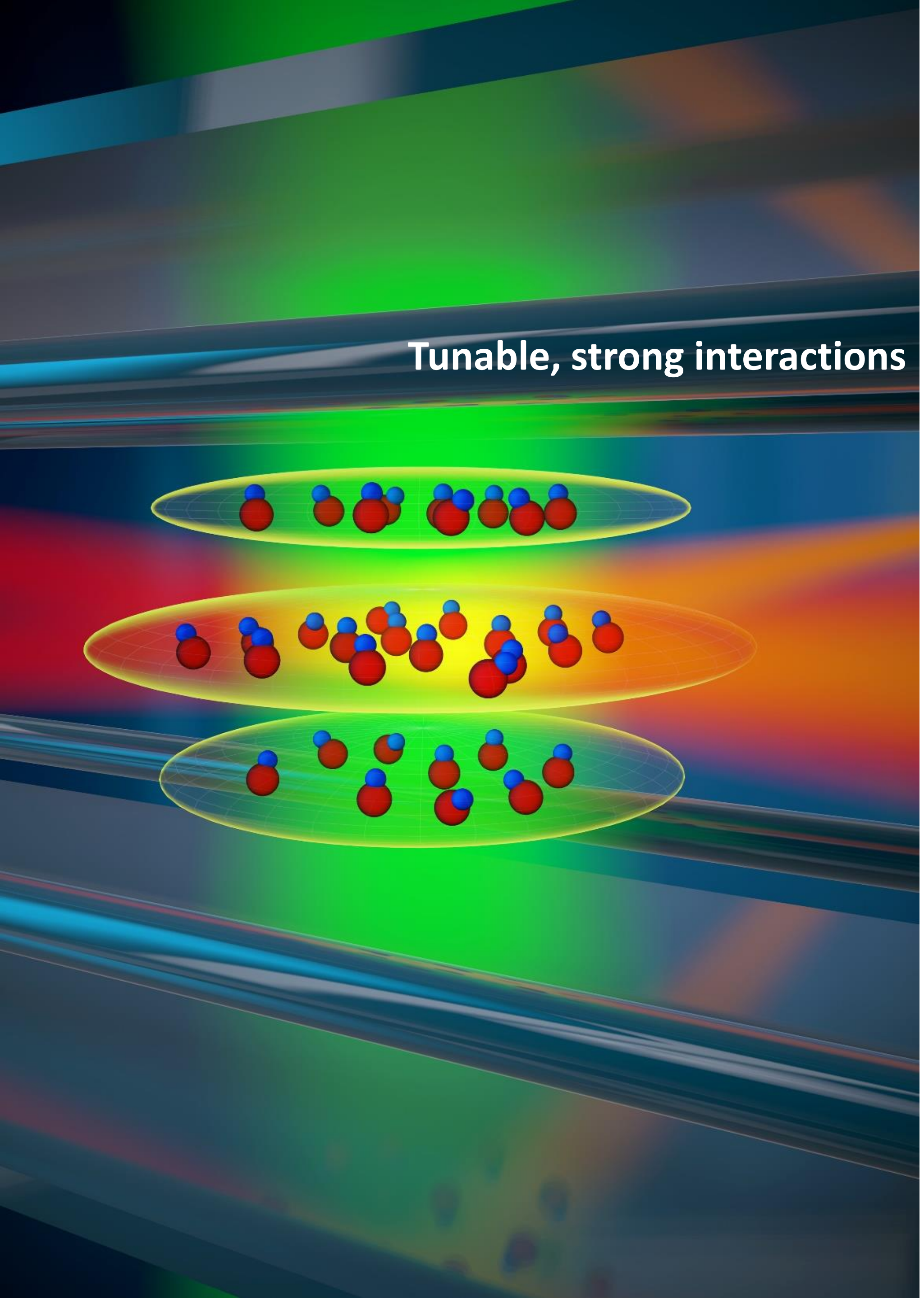
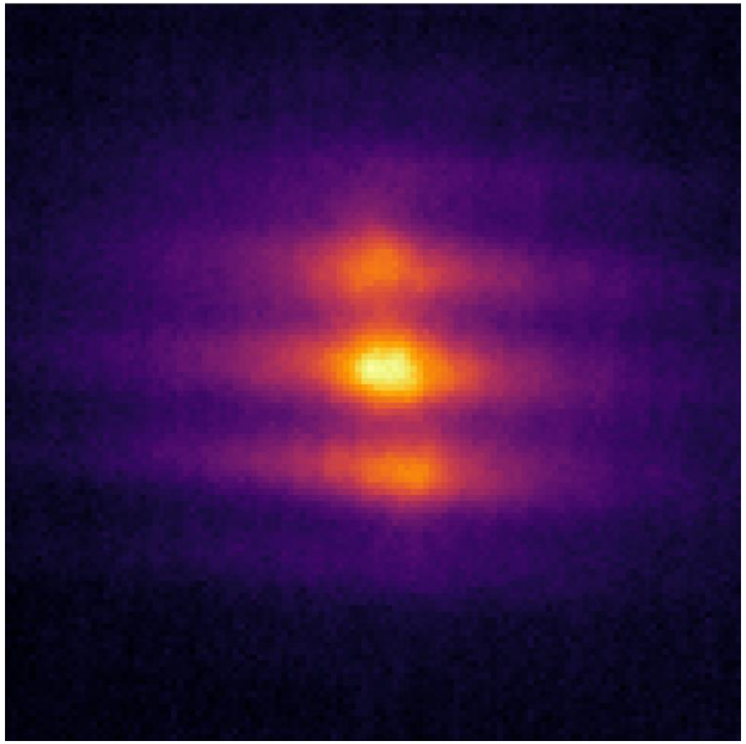


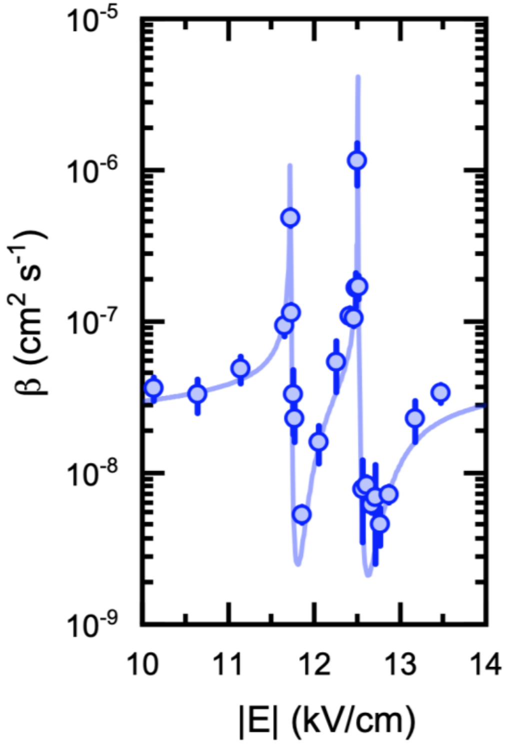
Quantum gas of molecules

Jun Ye

2021 Boulder Summer School for Condensed Matter and Materials Physics

July 5, 2021





Ultracold Atomic Matter

Precise control of quantum systems

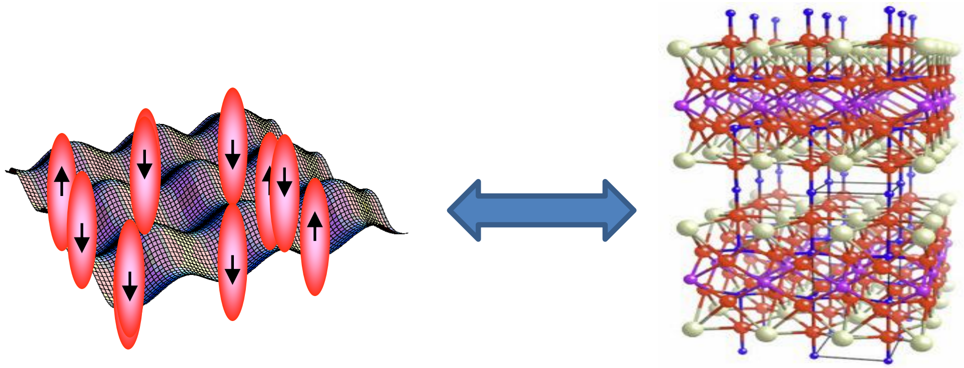
- Clocks, quantum information, sensors

Probe complexities & strong correlations

- Superconductivity & Superfluidity
- Quantum magnetism
- Quantum chemistry

Universality & scaling

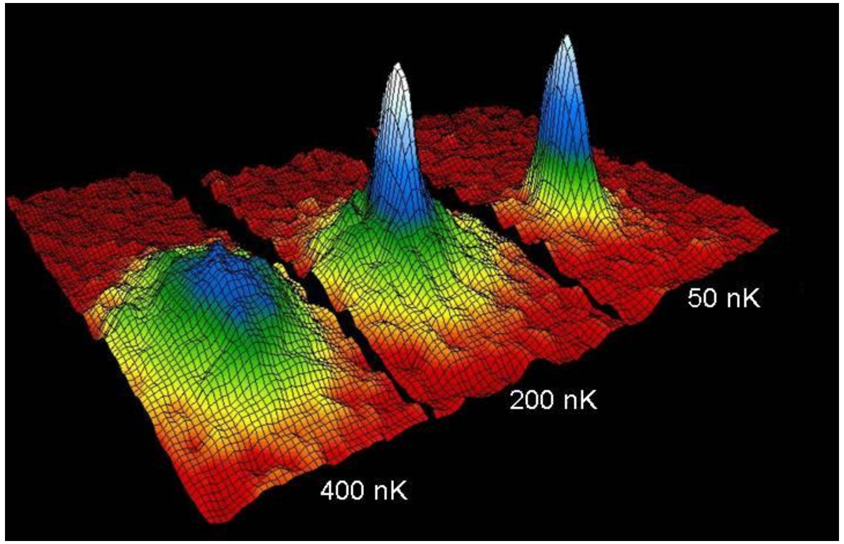
- Range of interactions
- Few to many-body



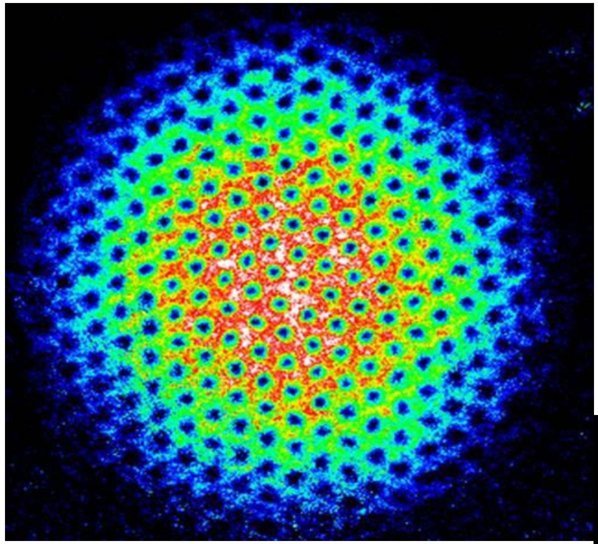
Atoms ↔ Electrons

Optical lattice ↔ Ionic Crystal

Quantum atomic gases



Bose-Einstein condensate



Superfluidity

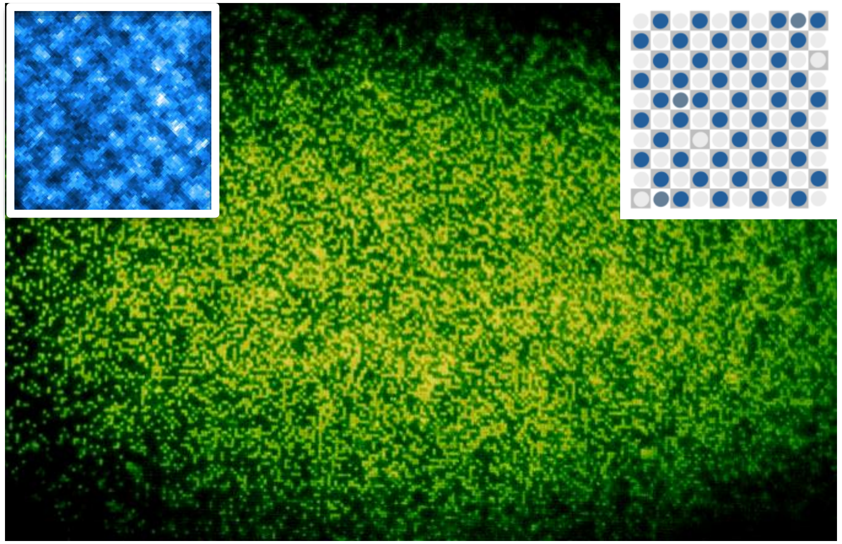
$T = 100$ nK

$N = 10^6$ atoms

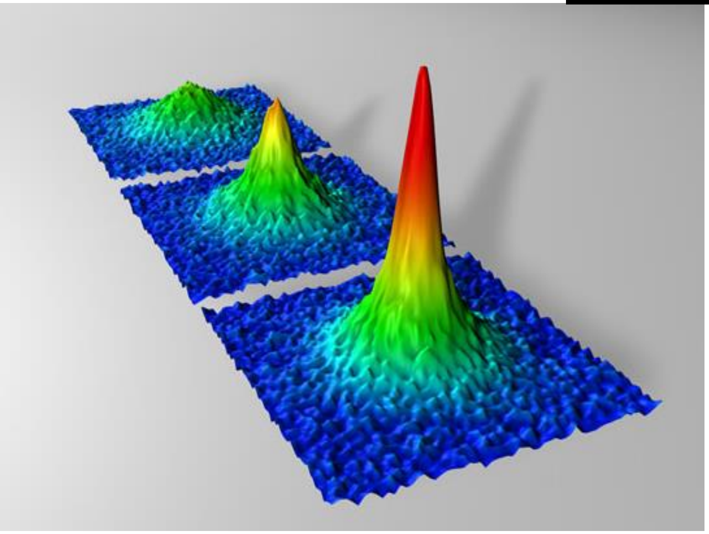
$n = 10^{13}$ cm^{-3}

Control atomic interaction

Control atomic motion



Magnetic ordering, transport



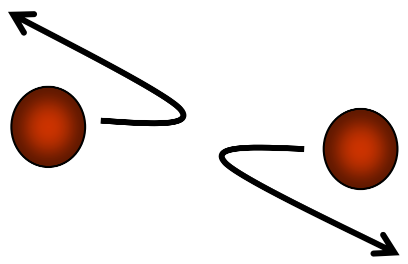
Fermi superfluidity

Why cold molecules?

- **Atoms:**

  contact interaction

  isotropic



- **Polar molecules:**

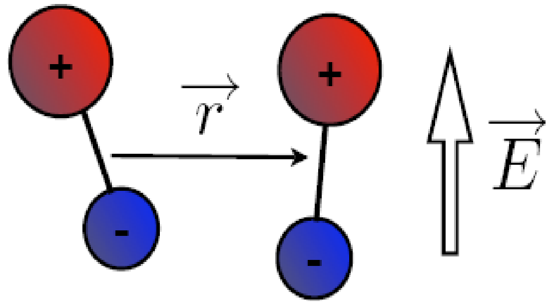
  dipole-dipole interaction

  tunable

  strong

  long-range

  anisotropic

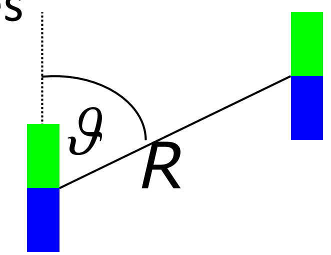


$$\frac{d^2}{r^3}\left(1-3\cos^2\theta\right)$$

Dipolar quantum systems

- Strongly correlated quantum material
- Dynamics with tunable, long-range interactions

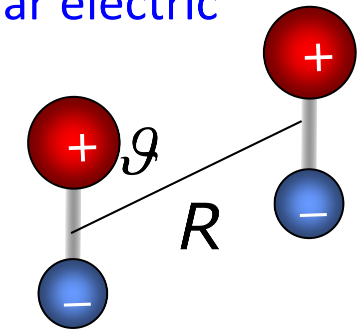
Atomic magnetic dipoles



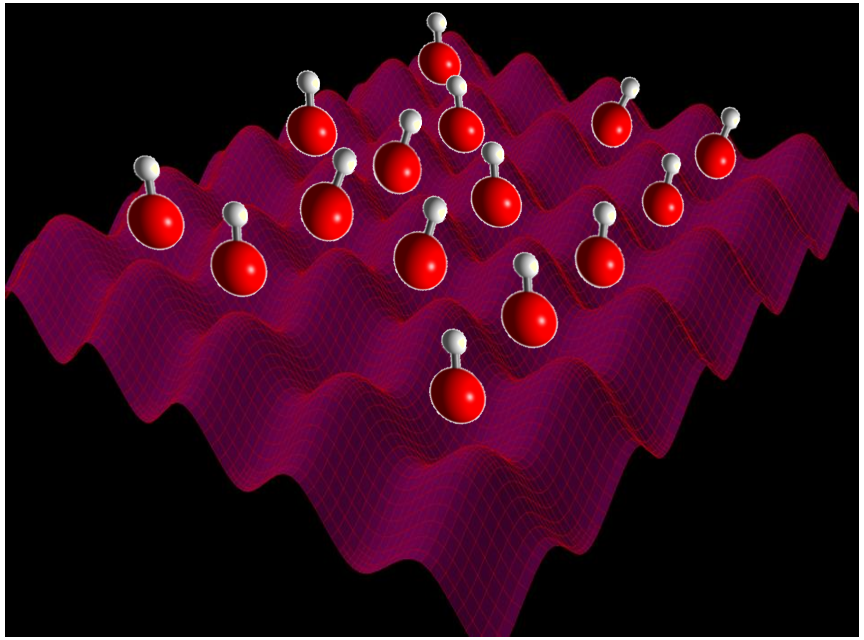
$$\frac{(\text{Debye})^2}{(\text{Bohr magneton})^2} \cdot c^2 = 10^4$$

Tunable by angle & strength (**E**)

Molecular electric dipoles



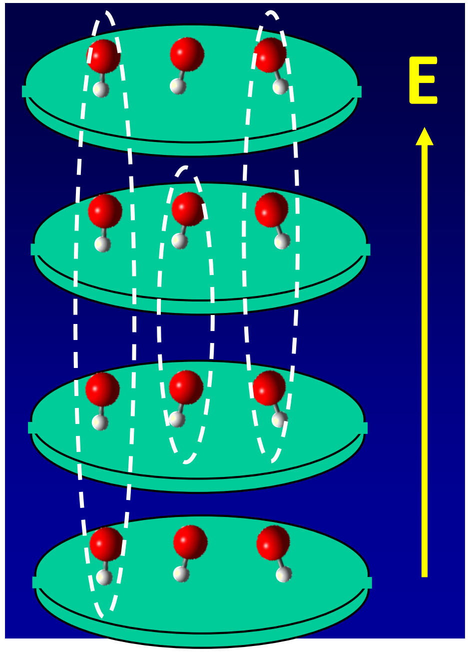
Stuttgart, Stanford, Innsbruck, Florence, Paris, …



Quantum metrology,

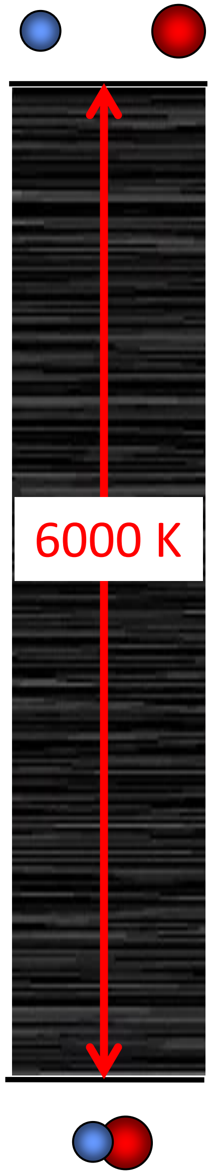
Quantum information

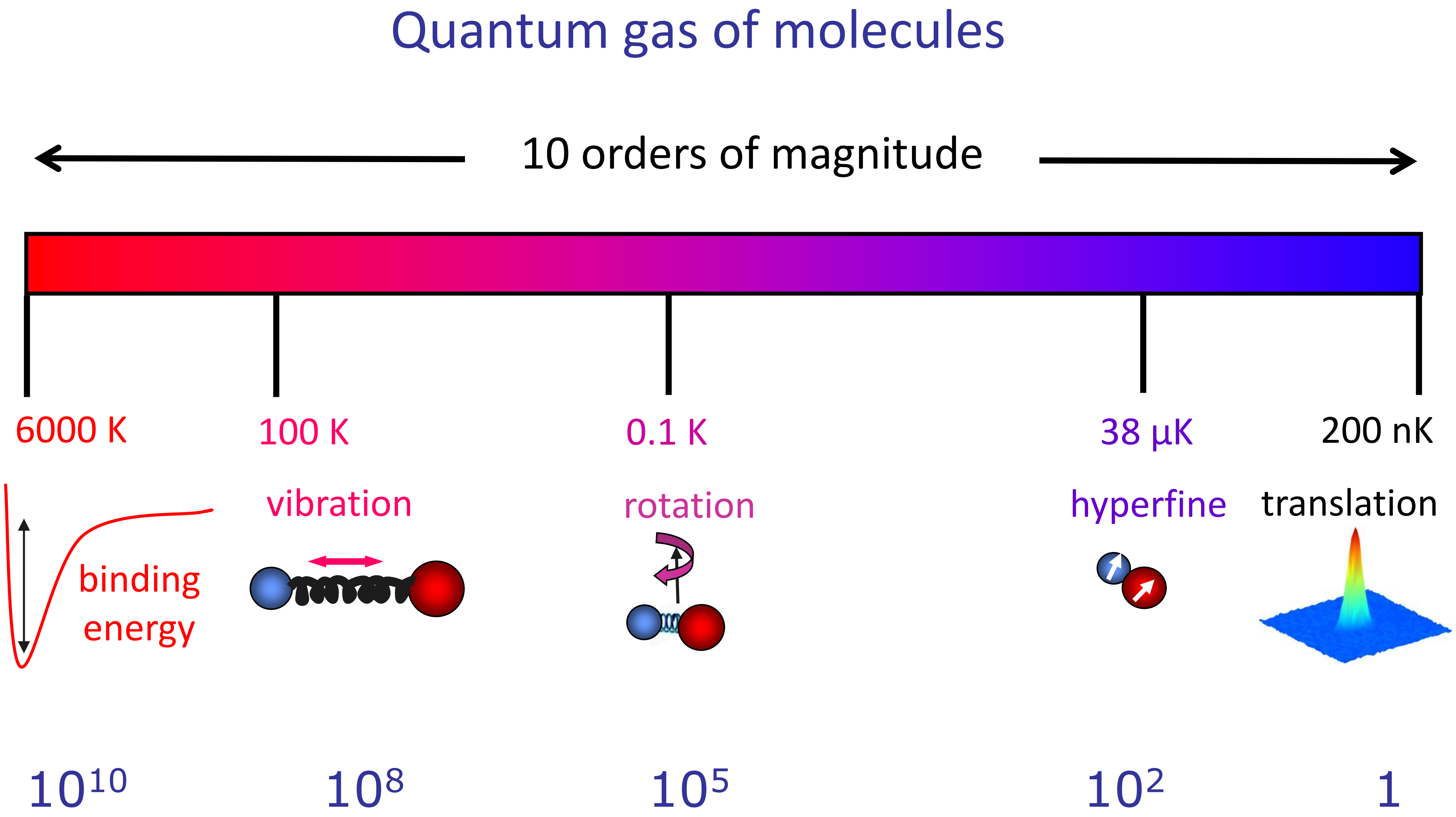
Chemistry



Molecules are complex

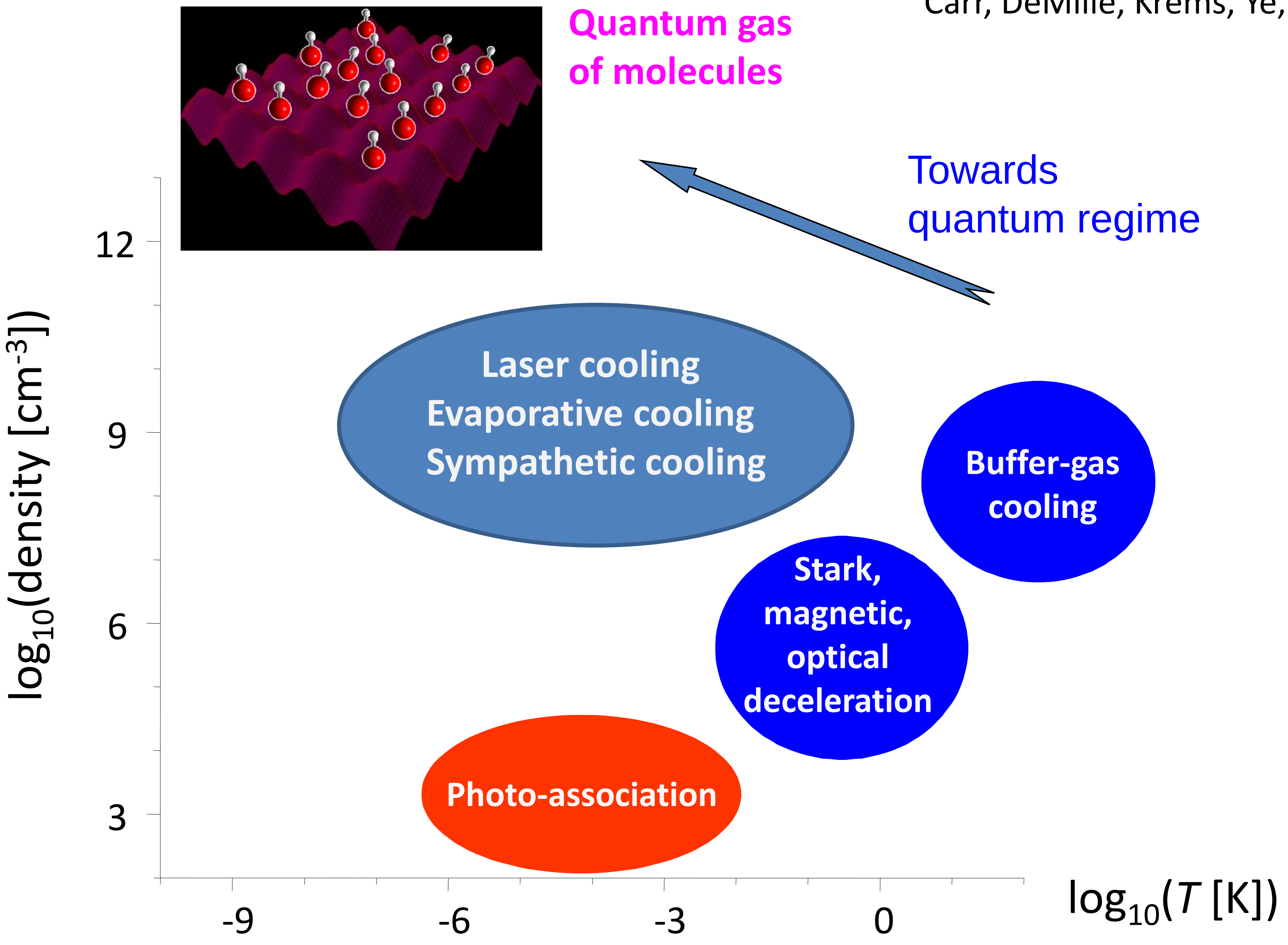
Jin & Ye, Physics Today **64**, 27 (2011).





Technology for making cold molecules

Carr, DeMille, Krems, Ye, New. J. Phys. 2009.

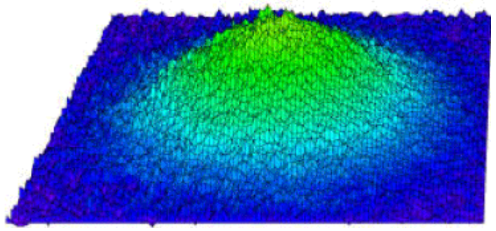


Ultracold polar molecules

Ni, Ospelkaus, de Miranda, Pe’er, Neyenhuis, Zirbel, Kotochigova, Julienne, Jin, & Ye,
Science **322**, 231 (2008).

Innsbruck, Durham, MIT, Hong Kong, USTC, MPQ, Harvard, Singapore, …

^{40}K Fermions

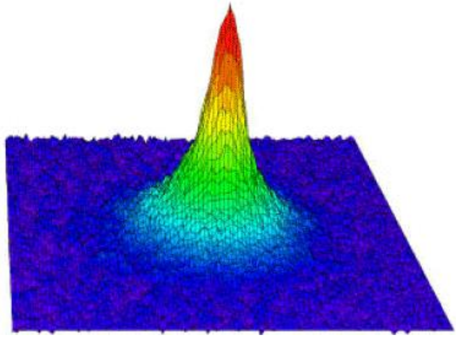


Temperature ~ 200 nK

Density ~10^{12}/cm^3

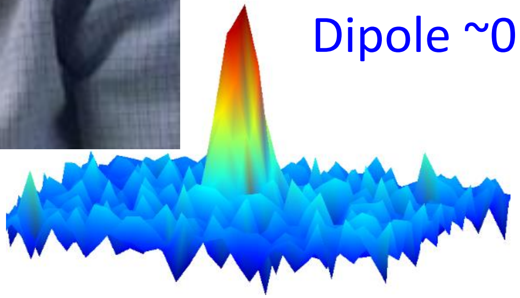
T/T_F ~ 1.4

^{87}Rb Bosons

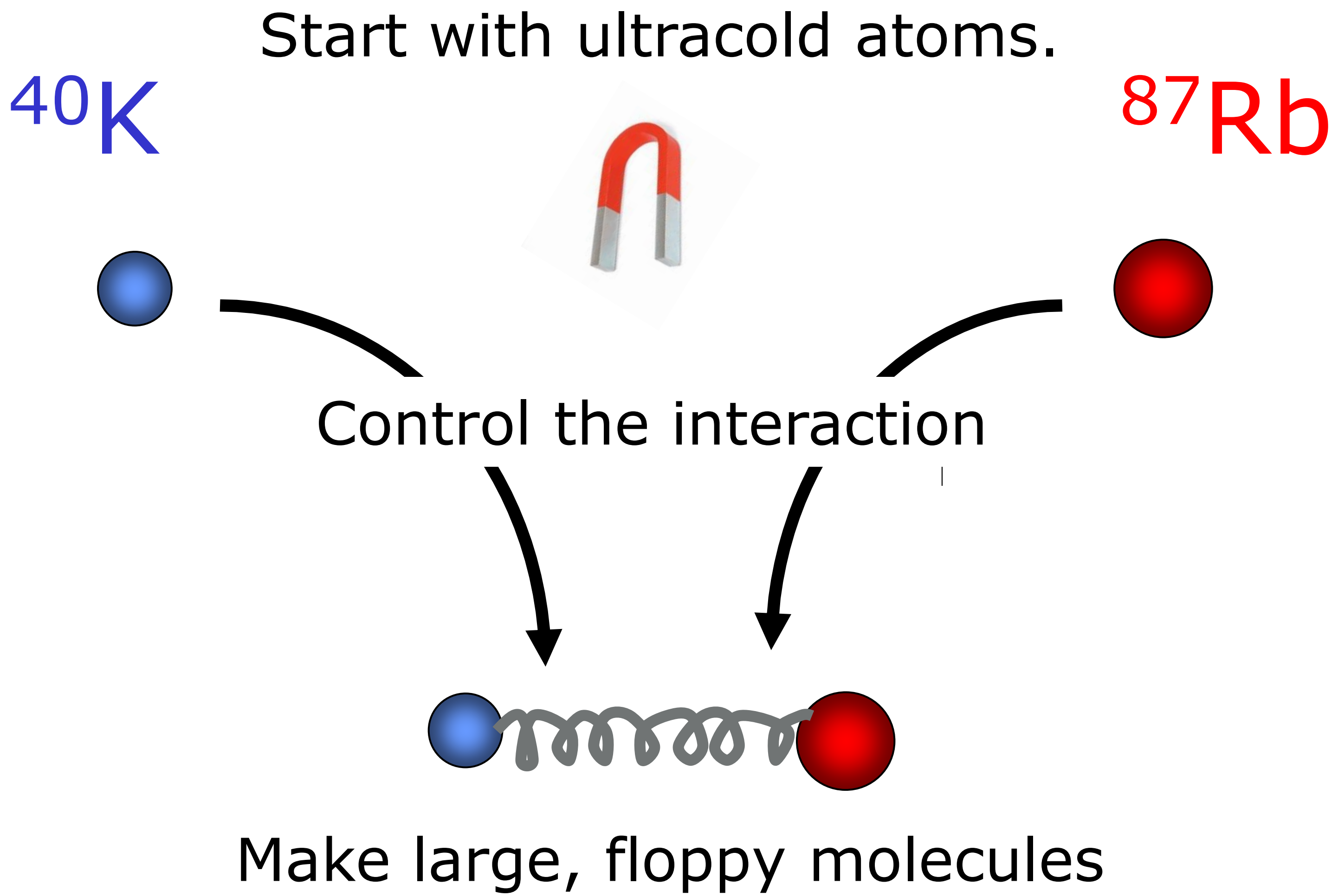


KRb molecules

Dipole ~0.5 Debye



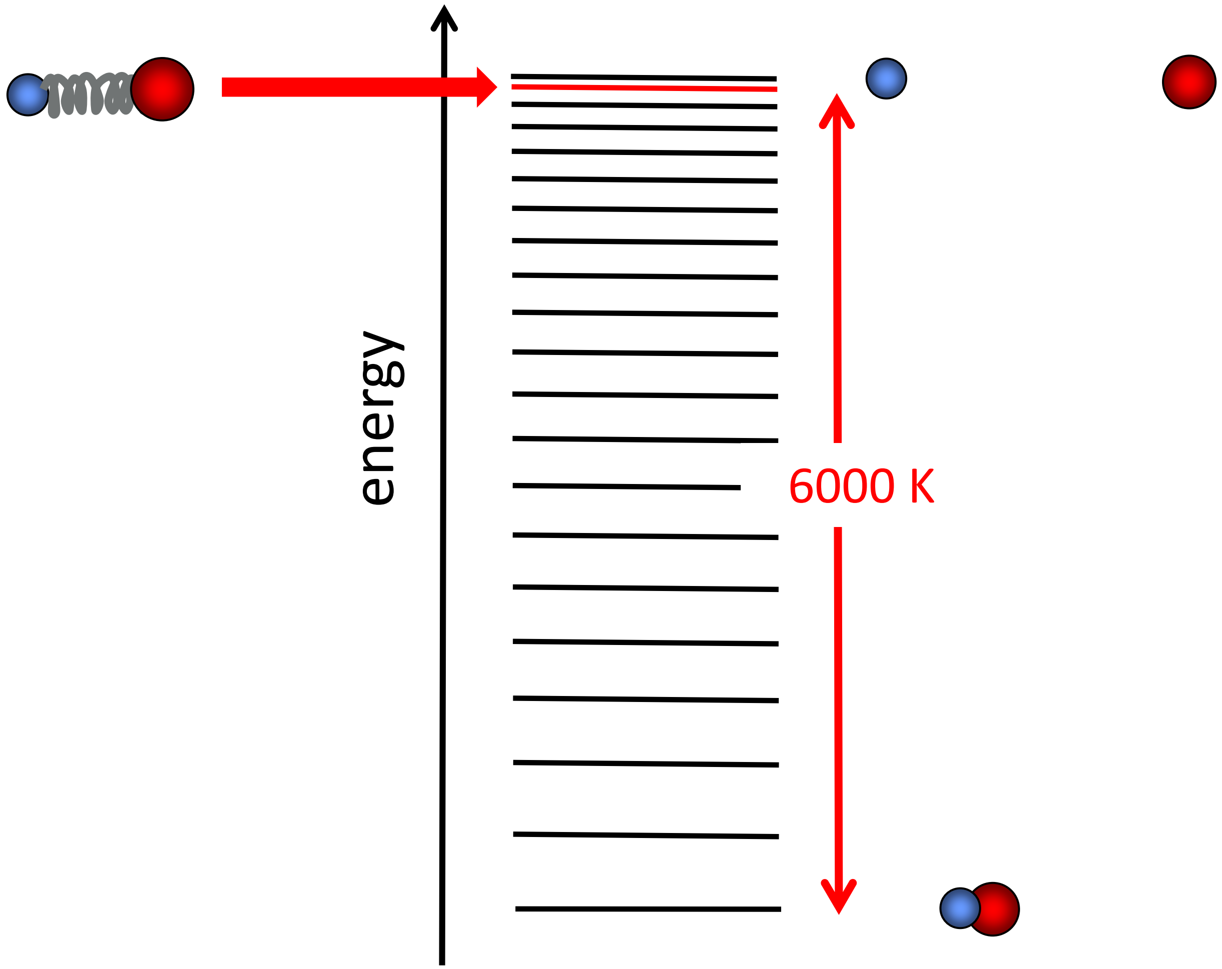
Feshbach molecules



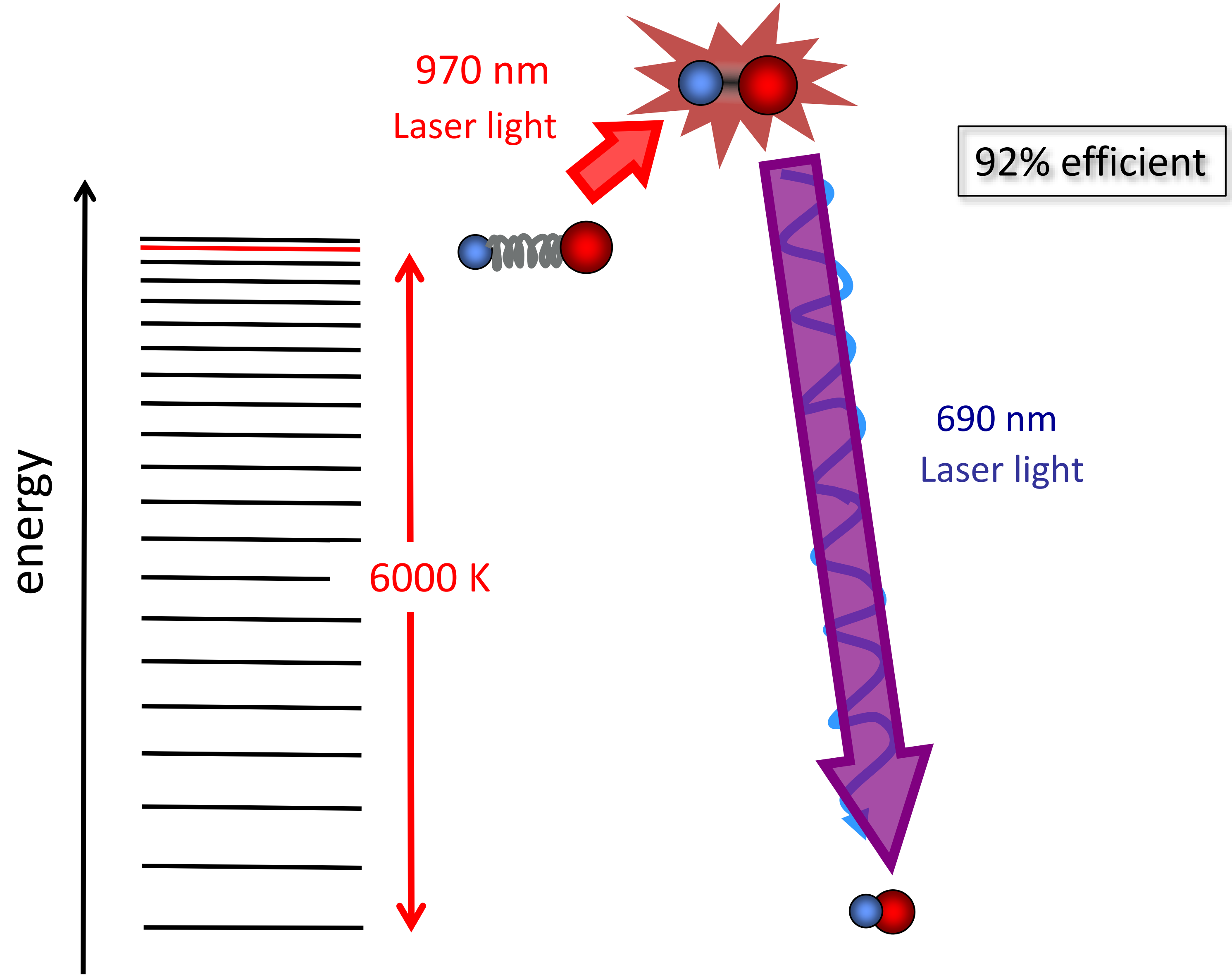
Weakly bound molecules

Zirbel et al., PRL **100**, 143201 (2008).

- no dipole moment
- losses

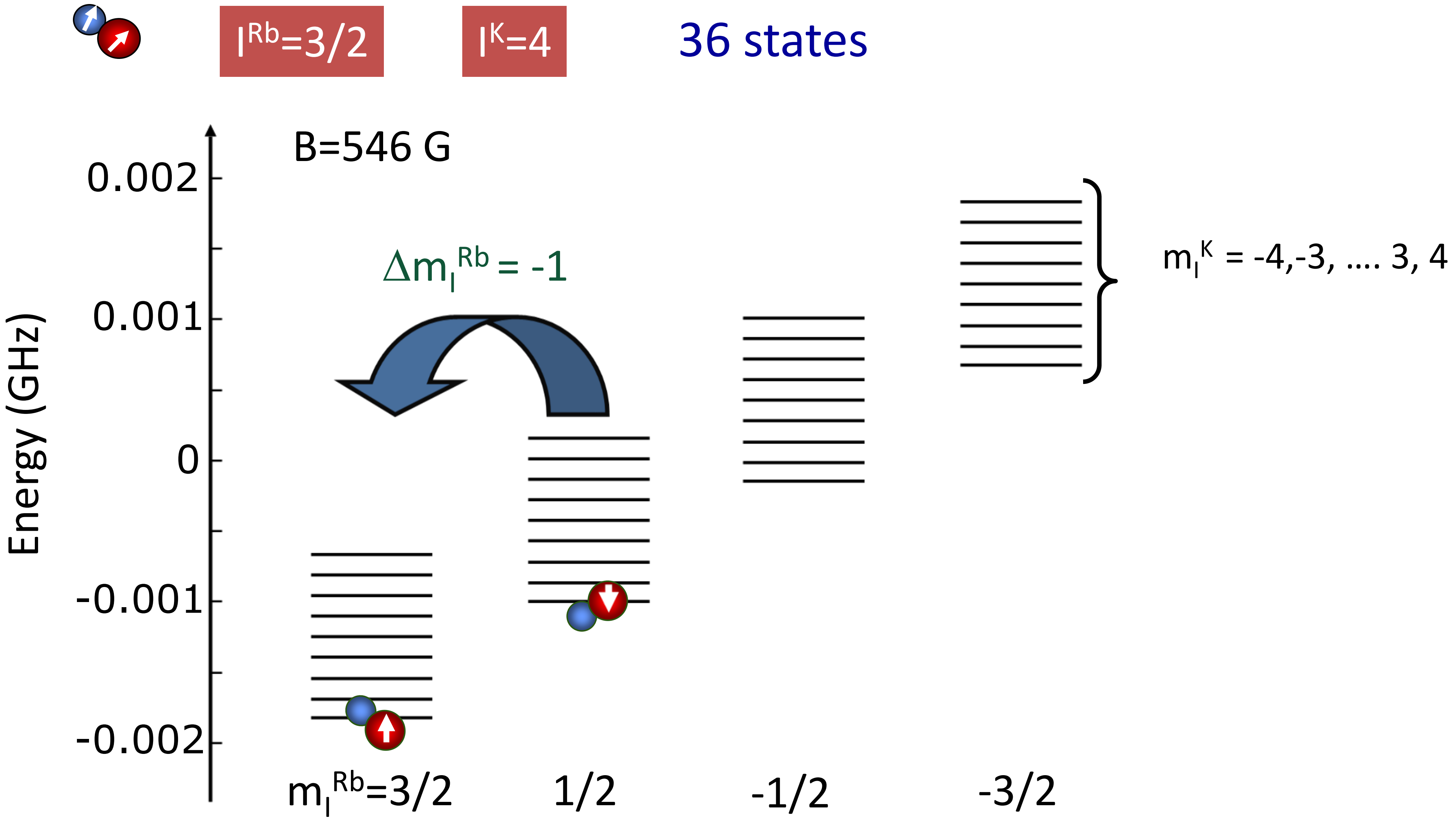


Transfer the molecules to the ground state



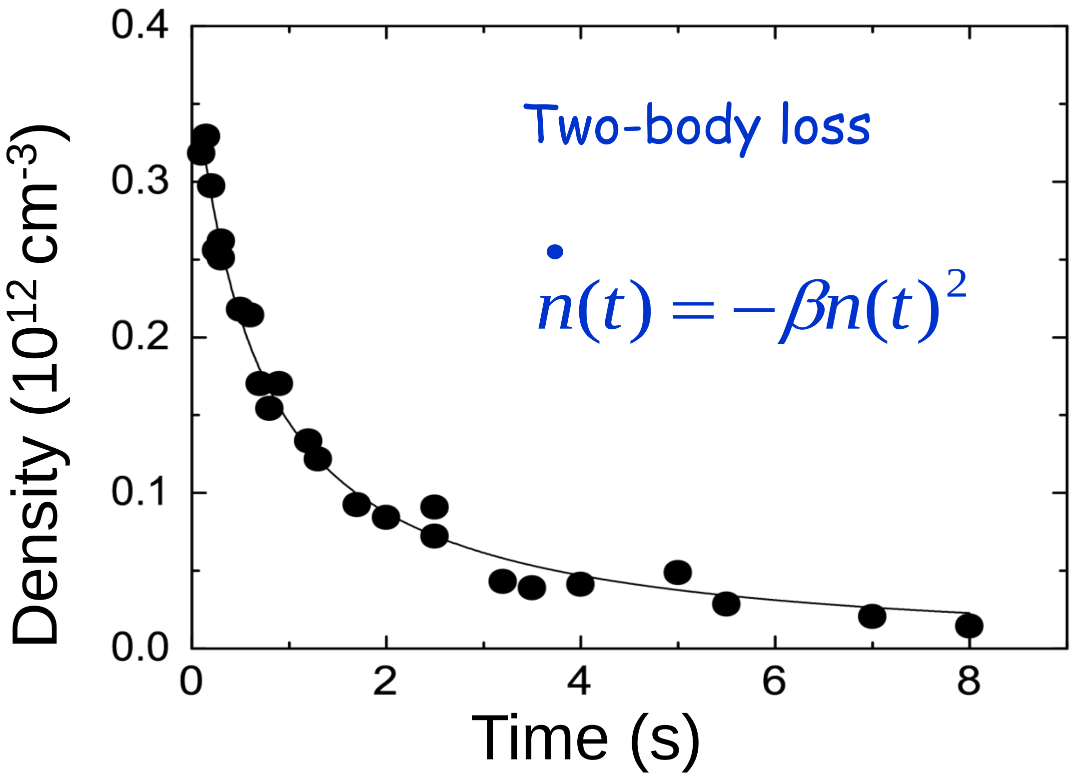
Light carries away the binding energy (& preserves the entropy)

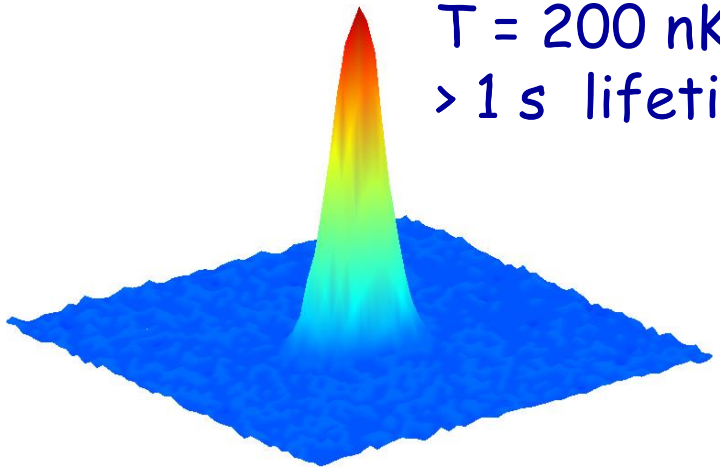
Hyperfine structure for $^1\Sigma$ (v=0, N=0)



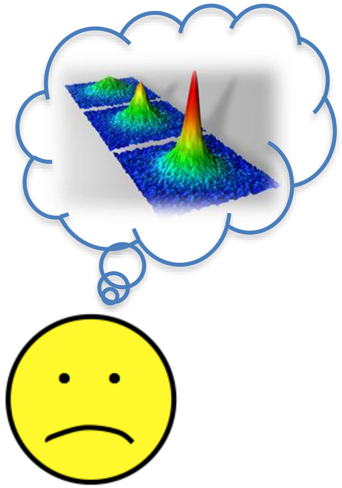
Chemistry near absolute zero

Molecules in the lowest energy state
(electronic, vibrational, rotational, hyperfine)





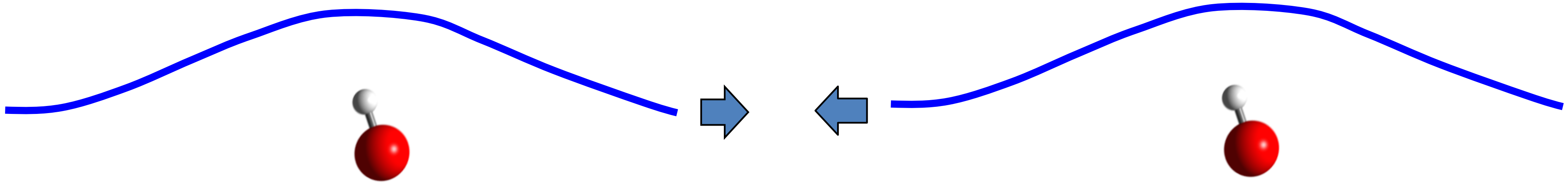
T = 200 nK
> 1 s lifetime



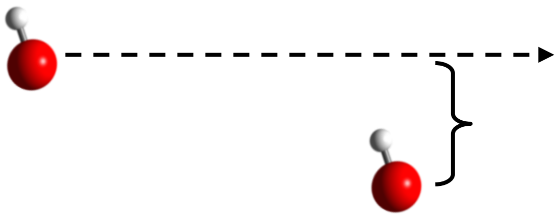
KRb + KRb → K_2 + Rb_2

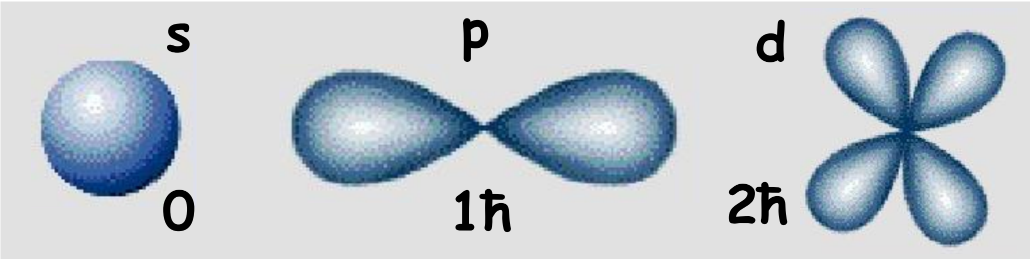
Cold collisions between identical Fermions

(1) Particles behave like waves

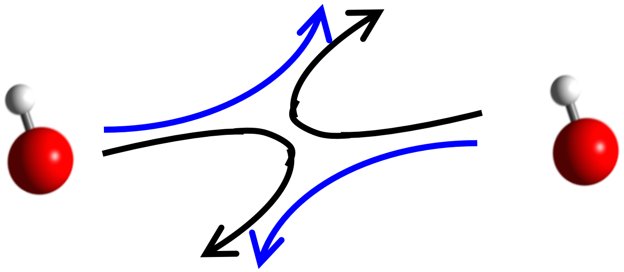


(2) Angular momentum is quantized

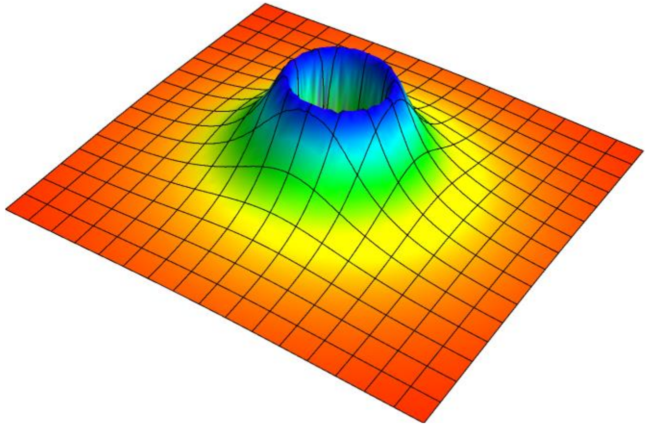




(3) Quantum statistics matter

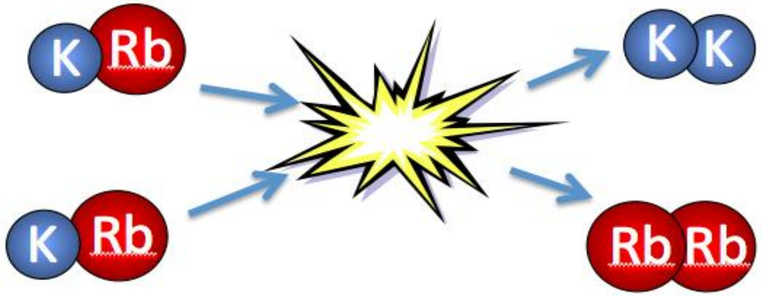


$$|\psi_0\rangle|\psi_1\rangle - |\psi_1\rangle|\psi_0\rangle$$



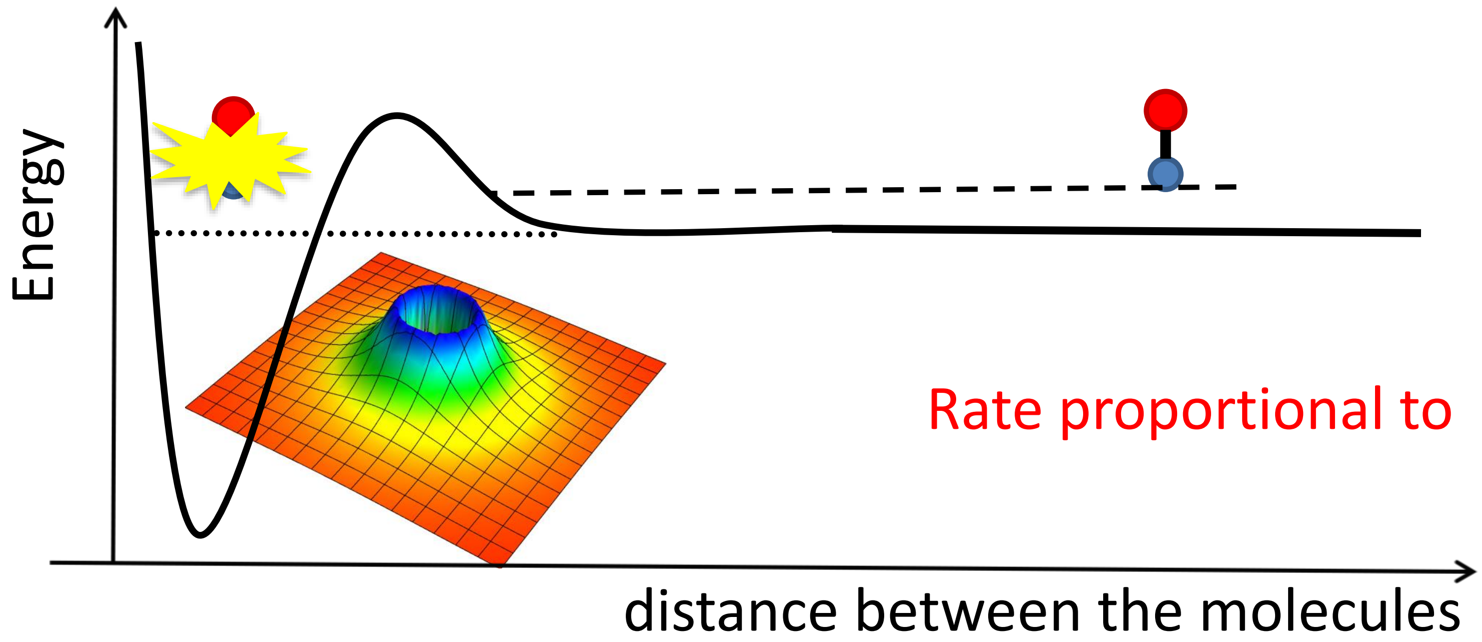
Fermions ⇨ $L = 1$, *p*-wave collisions

Ultracold quantum chemistry

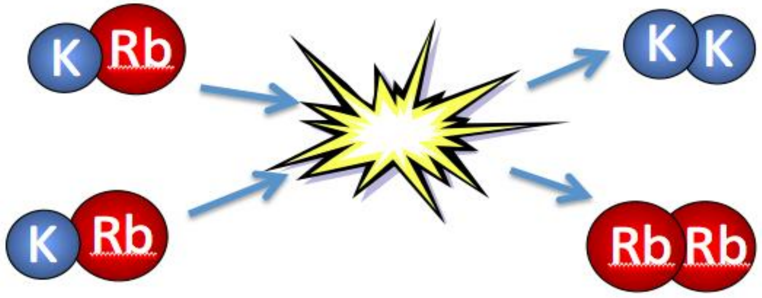


Ospelkaus *et al.*, Science **327**, 853 (2010).

At low T, the quantum statistics of fermionic molecules suppresses chemical reaction!

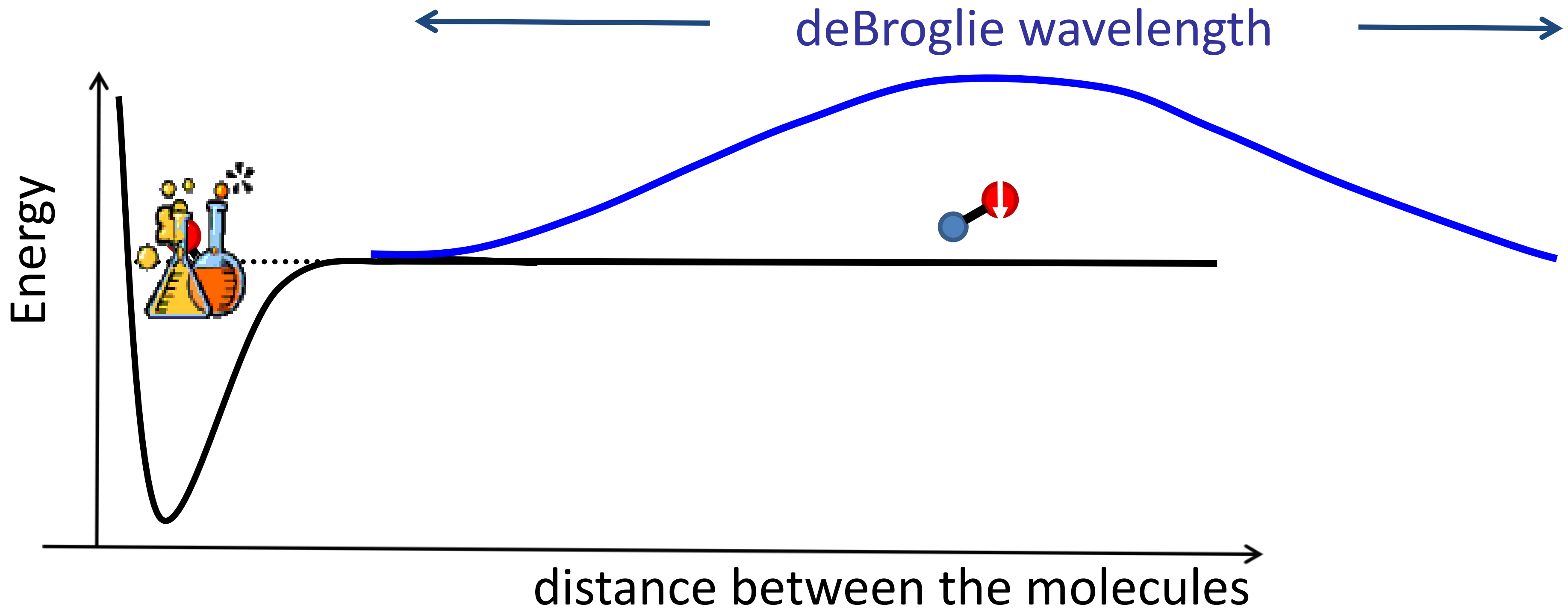


Ultracold quantum chemistry



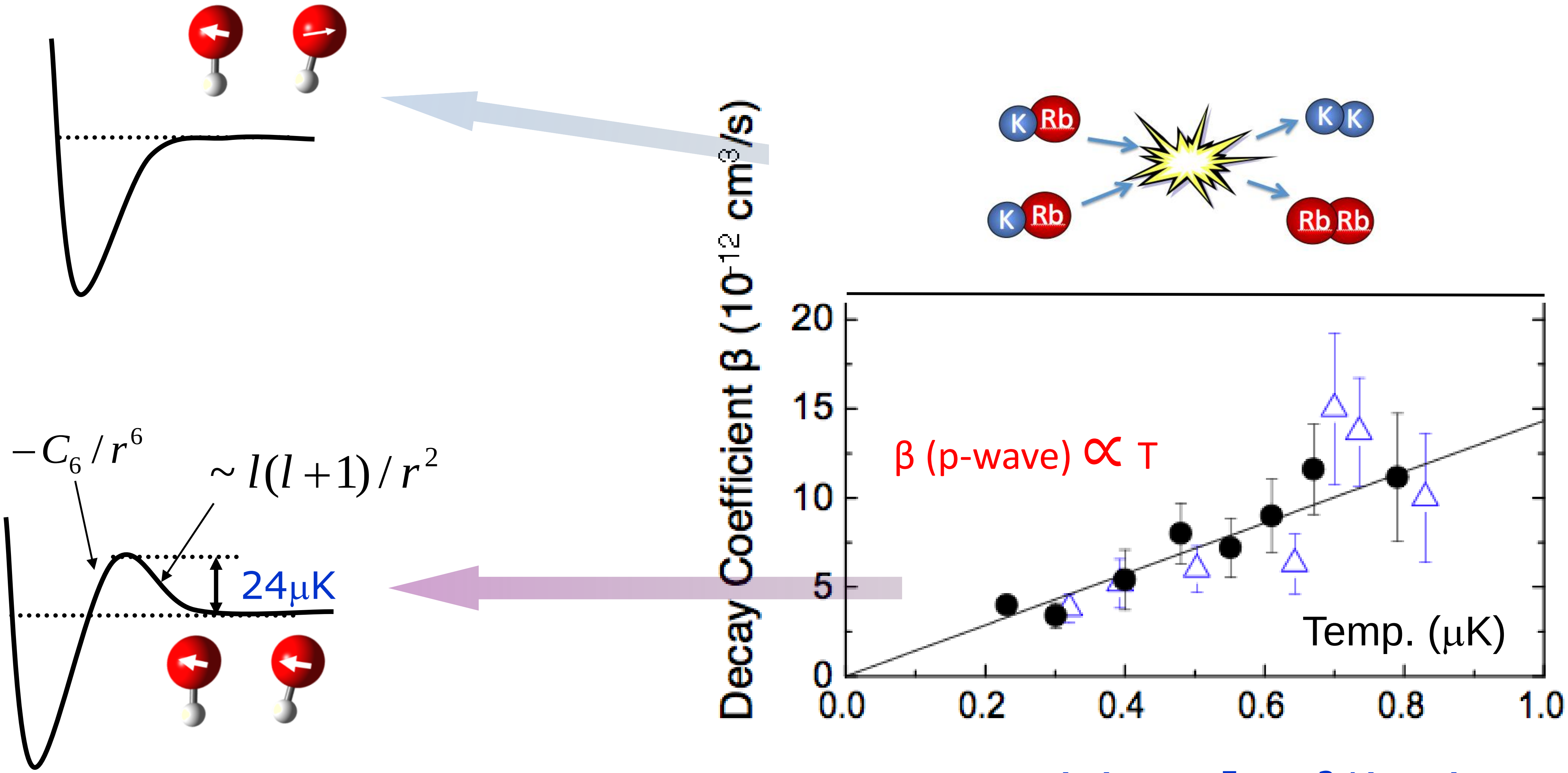
Ospelkaus *et al.*, Science **327**, 853 (2010).

Distinguishable molecules do not enjoy the suppression → rate is x 100 higher, independent of *T*



Bimolecular reactions: Wigner threshold law

Ospelkaus *et al.*, Science **327**, 853 (2010).

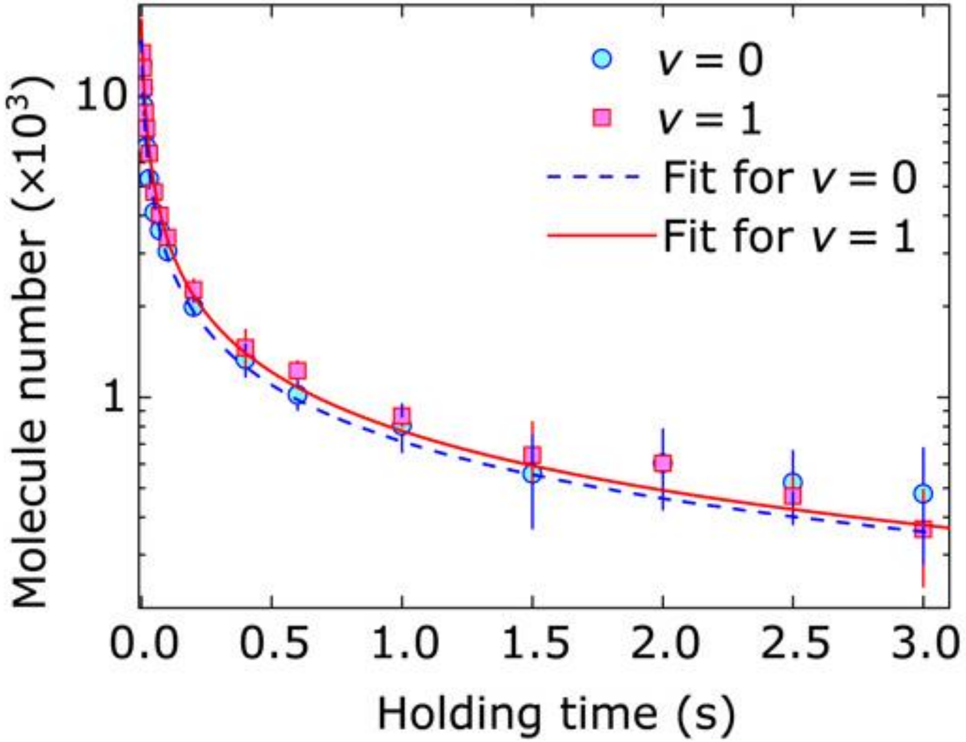


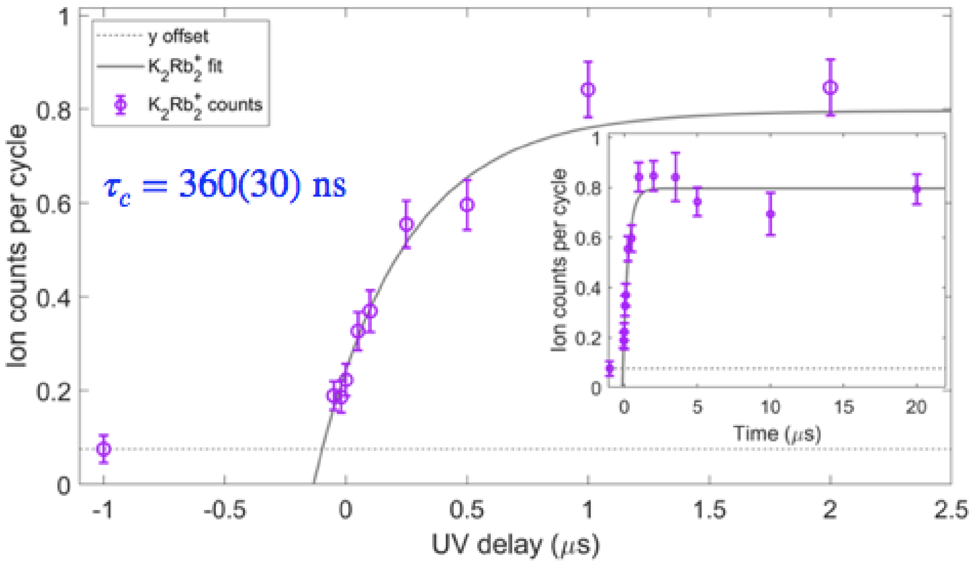
$\beta = 1.2(3)\text{x}10^{-5}\ cm^3/(s\ K)$

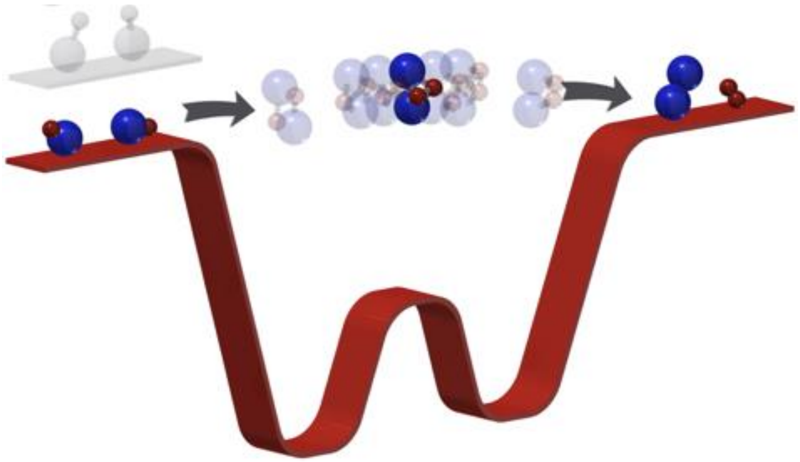
Molecular losses look universal

$NaRb + NaRb \rightarrow Na_2 + Rb_2$

Dajun Wang *et al.*, Sci. Adv., 4, eaaq0083(2018).







Direct detection of $K_2Rb_2^*$ intermediate

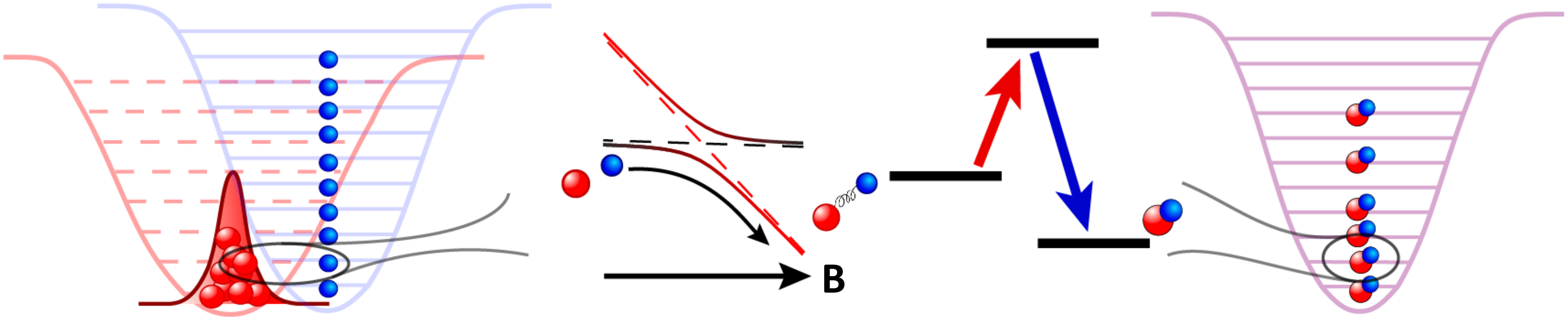
Kang-Kuen Ni *et al.*, Science 366, 1111 (2019); Nature Physics (2020).

A degenerate Fermi gas of molecules

Ni, Ospelkaus, de Miranda, Pe'er, Neyenhuis, Zirbel, Kotochigova, Julienne, Jin, & Ye, Science **322**, 231 (2008).

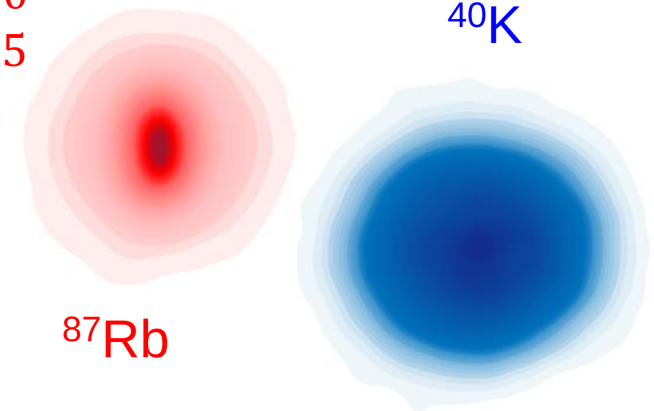
T/T_F ~ 1.4

Luigi De Marco *et al.*, Science **363**, 853 (2019).



Innsbruck, Durham, MIT, Hong Kong, USTC, MPQ, Harvard, Singapore, …

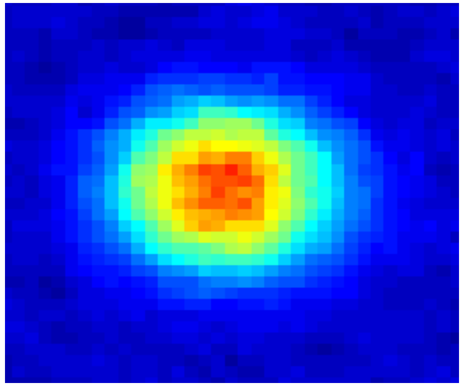
$N = 7 \times 10^4$
$T/T_c = 0.5$



$N = 5 \times 10^5$
$T/T_F = 0.1$

Atom – Molecule thermalization

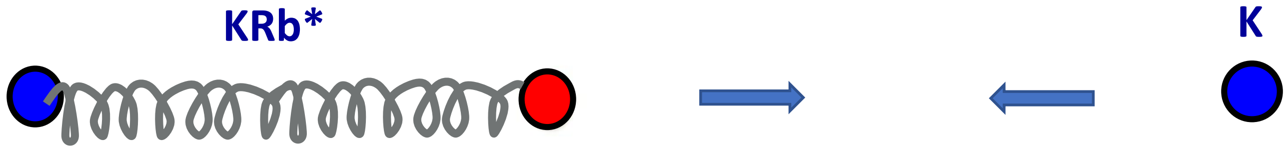
N = 3 x 10^4, T = 50 nK, T/T_F = 0.3



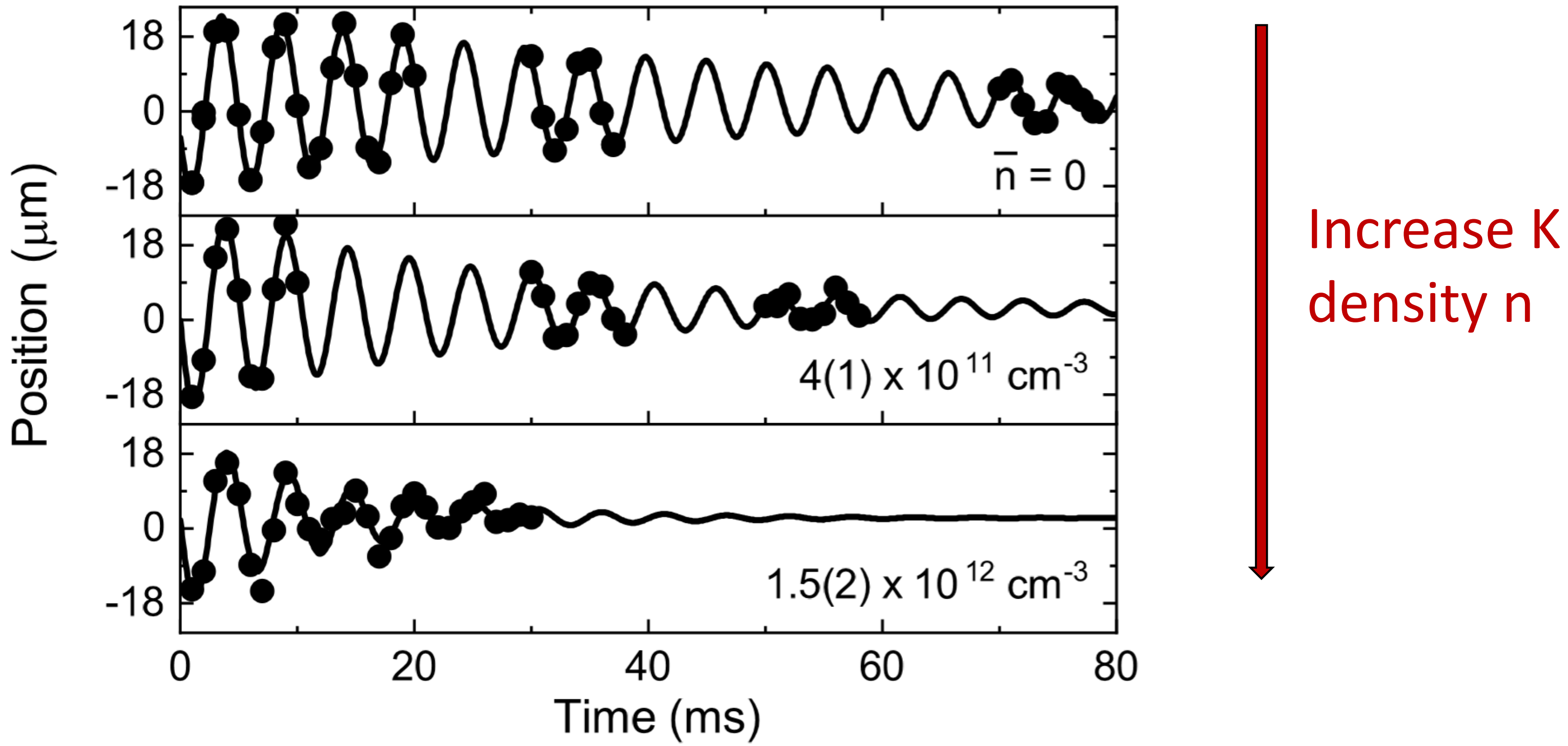
Atom – Molecule thermalization

W. Tobias *et al.*, Phys. Rev. Lett. **124**, 033401 (2020).

NaLi + Na:
Ketterle *et al*,
Nature **580**, 197 (2020).

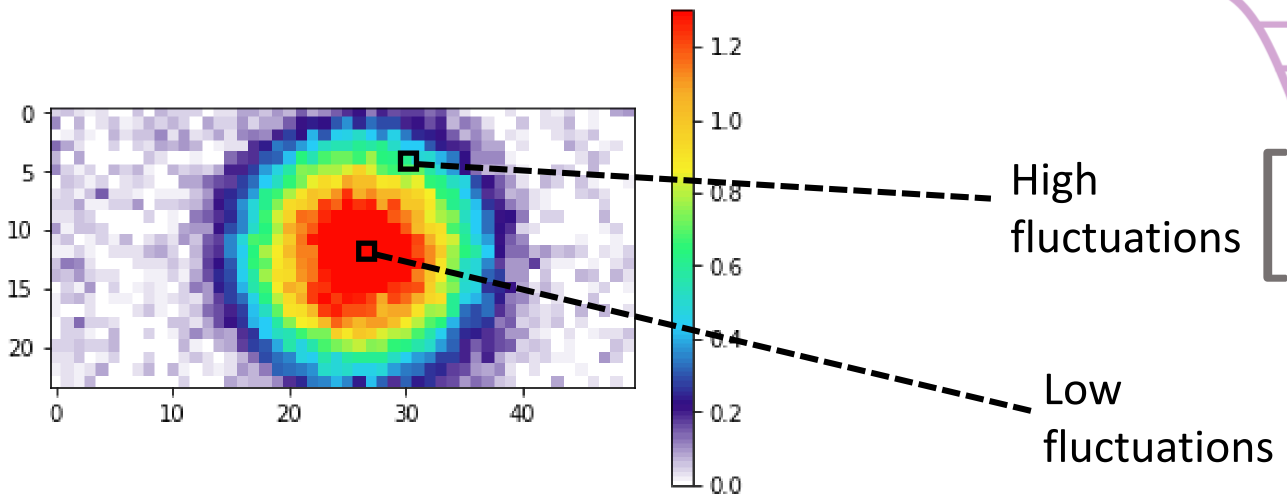


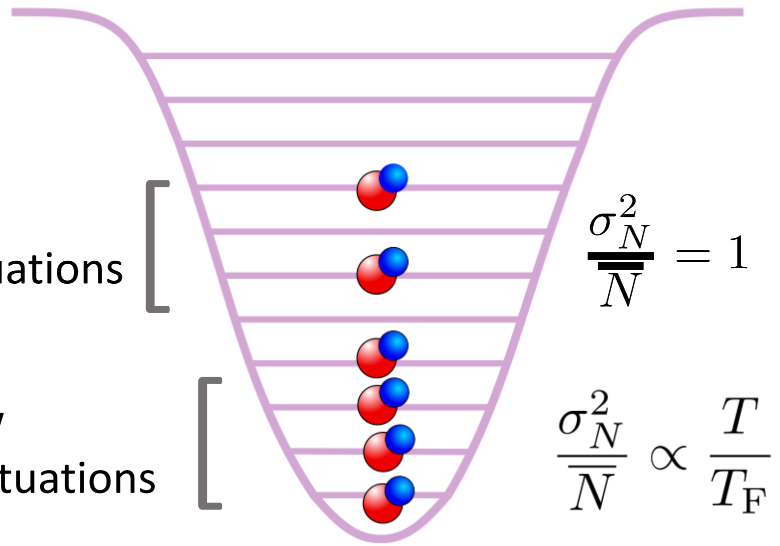
- Produce KRb*, remove remaining Rb
- Excite center of mass oscillation of KRb* but not K

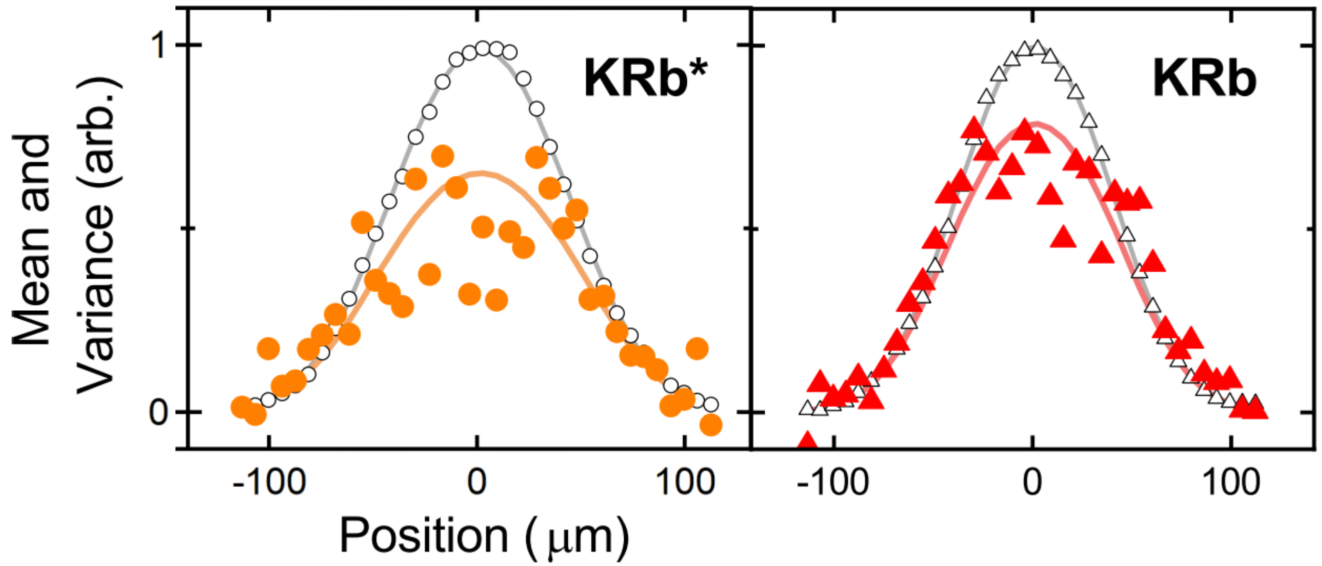


Fermi suppression of density fluctuations

W. Tobias *et al.*, Phys. Rev. Lett. **124**, 033401 (2020).



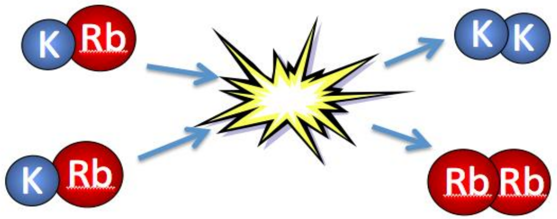




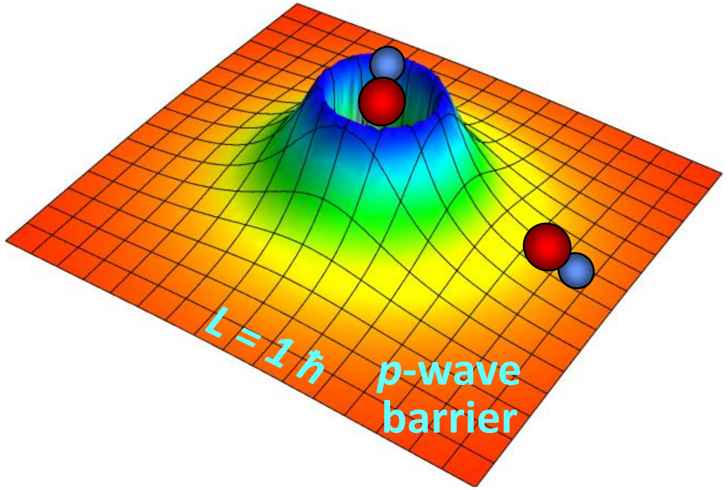
C. Sanner et al., PRL (2010);
T. Muller et al., PRL (2010).

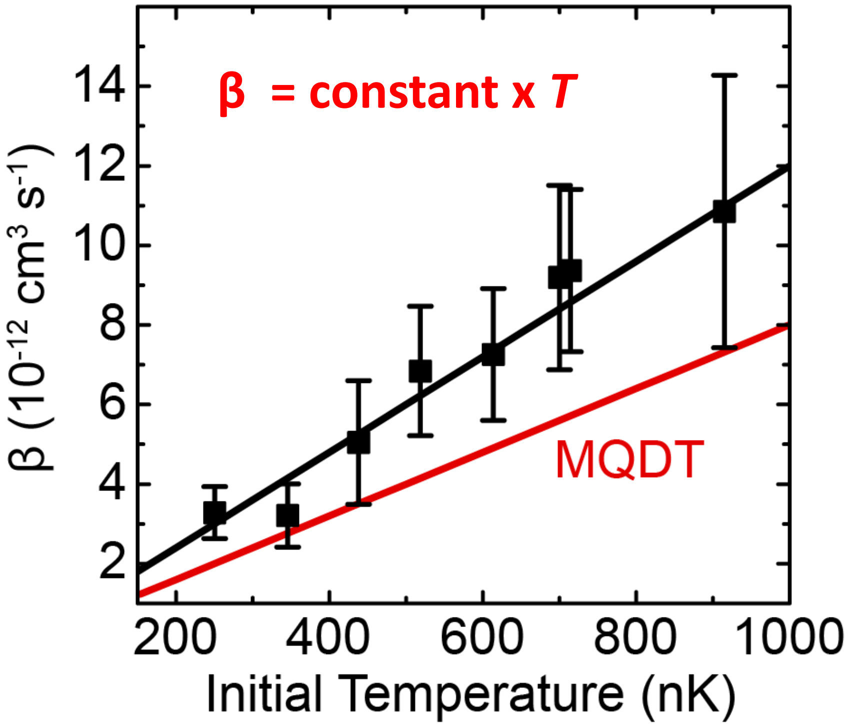
Chemistry near absolute zero

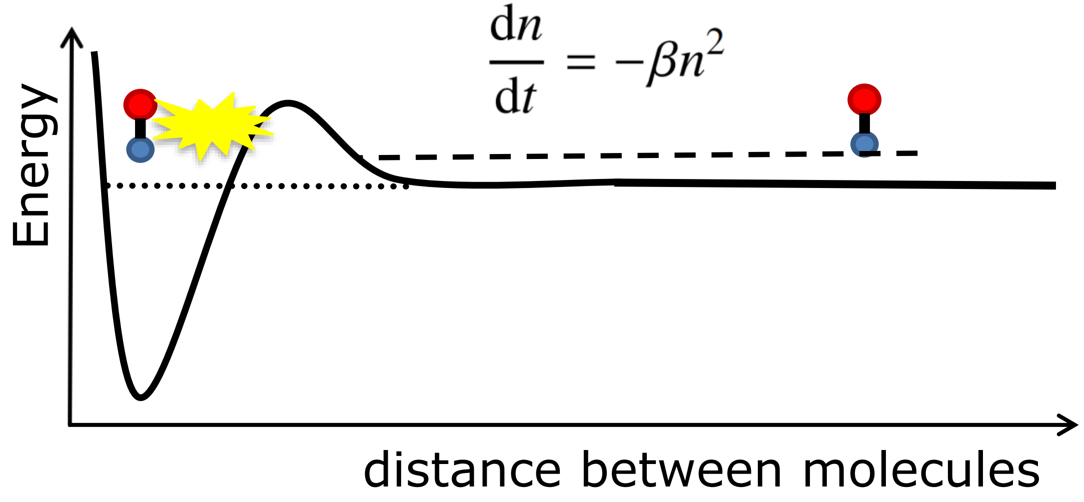
Ospelkaus *et al.*, Science **327**, 853 (2010): T/T_F ~1.4



- Quantum statistics dictate reaction rate
- Single collision partial wave

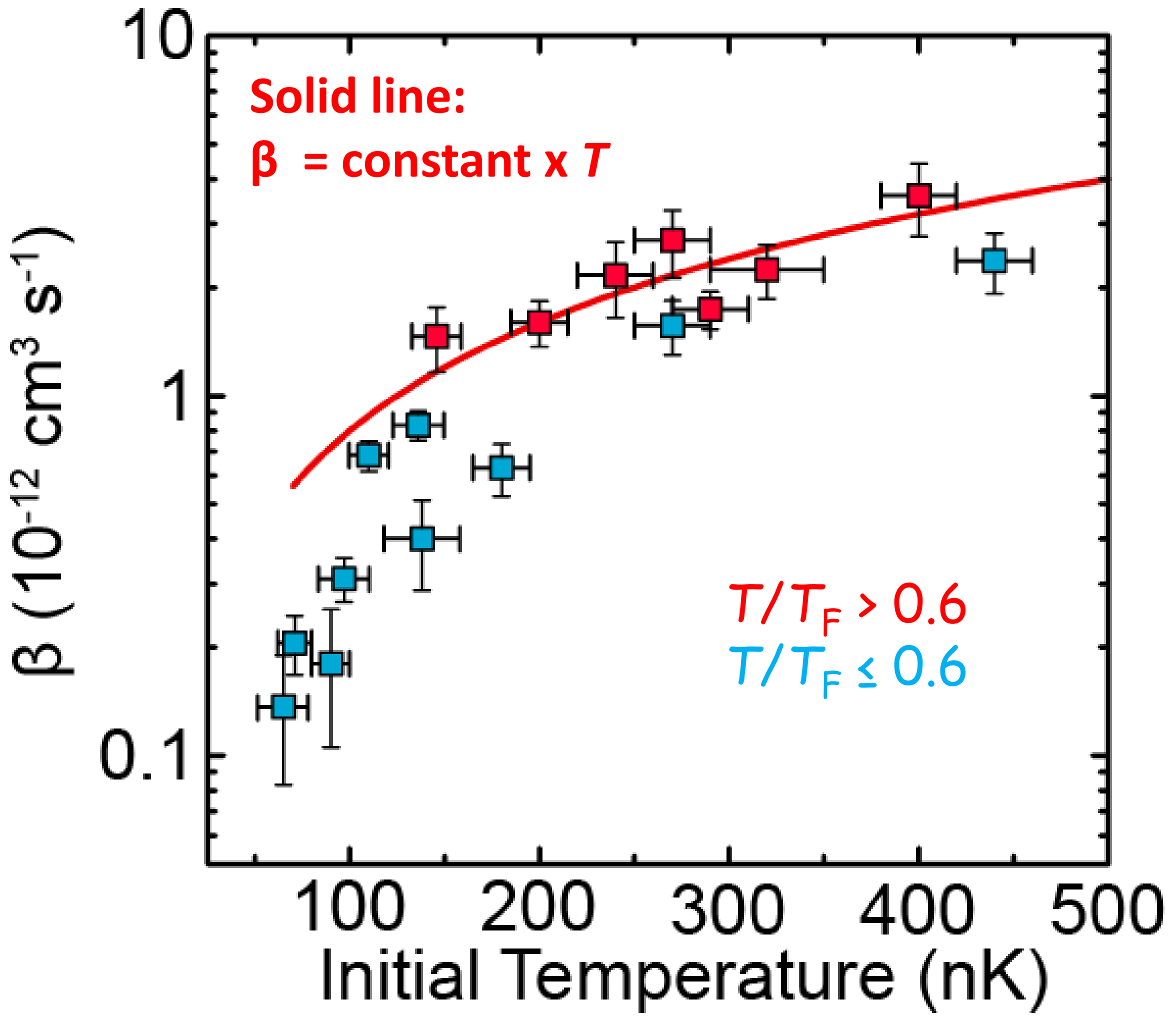






Reactions in Degeneracy

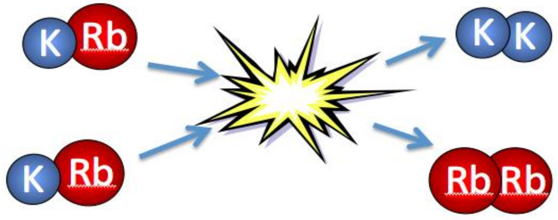
- At very low T, deviations from Bethe-Wigner threshold law
- For $T/T_F > 0.6$
  - $\beta/T = 0.84(6) \times 10^{-5}$ cm^3 s^{-1} K^{-1}
- For $T/T_F \leq 0.6$
  sub-linear behavior



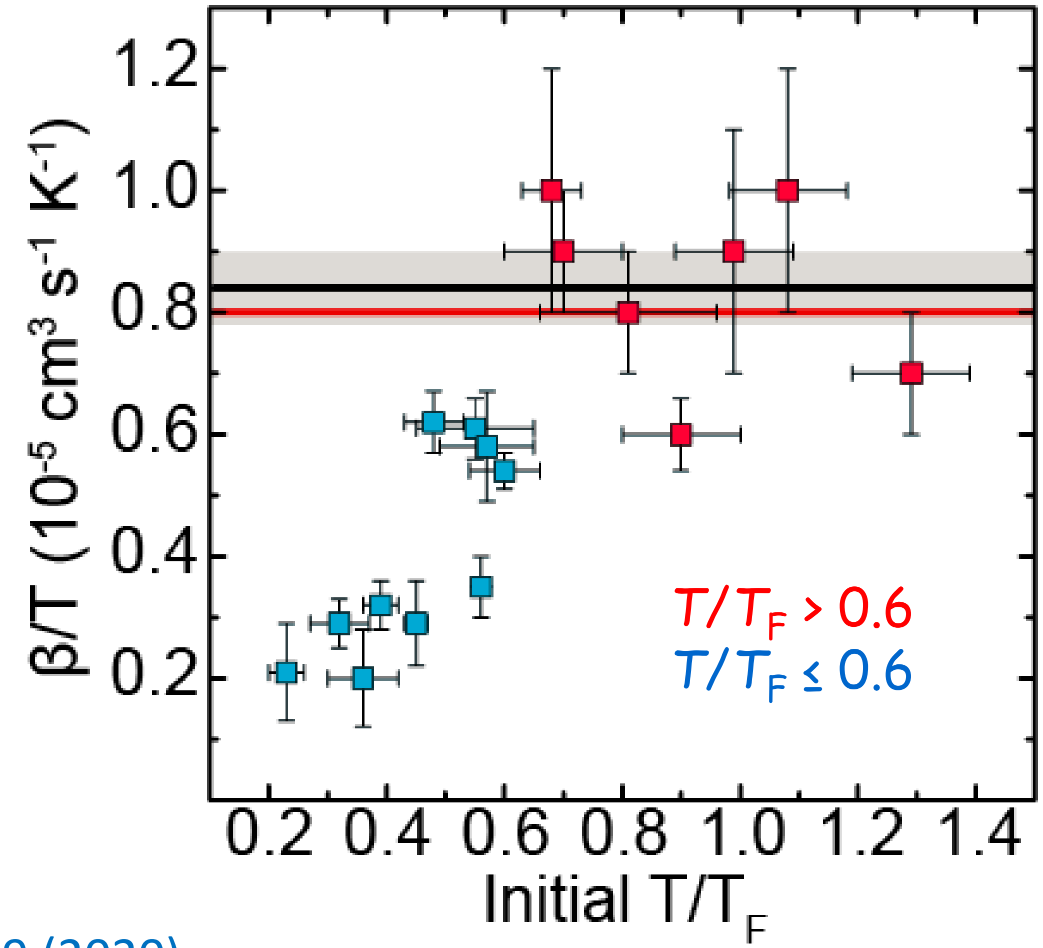
Chemistry in a degenerate Fermi gas

De Marco *et al.*, Science **363**, 853 (2019).

Rate coefficient β / T = constant (thermal gas, *p*-wave)



- For $T/T_F > 0.6$
  - $\beta/T = 0.84(6) \times 10^{-5}$ cm^3 s^{-1} K^{-1}
- For $T/T_F \leq 0.6$
  - β/T decreases



Qi Zhou group: *p*-wave contact, Science Advances **6**, eabd4699 (2020).

Ana Maria Rey group: Zeno effect via intermediates, PRA **102**, 063322 (2020).

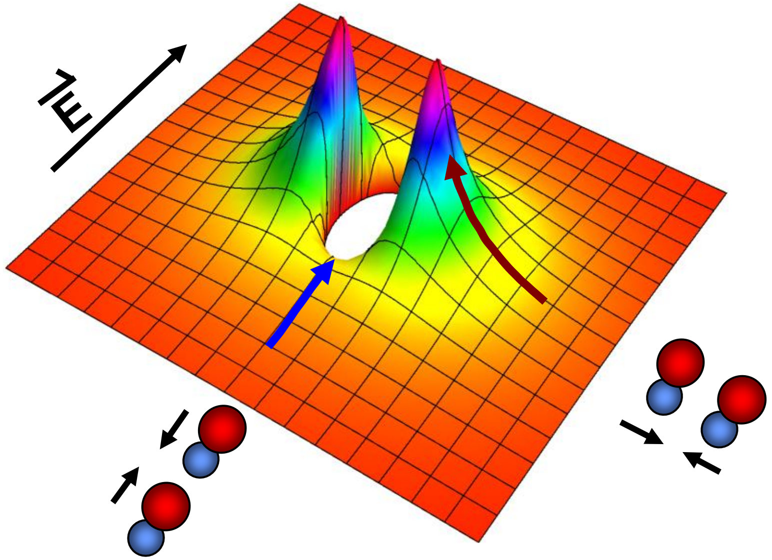
Inelastic collisions under E

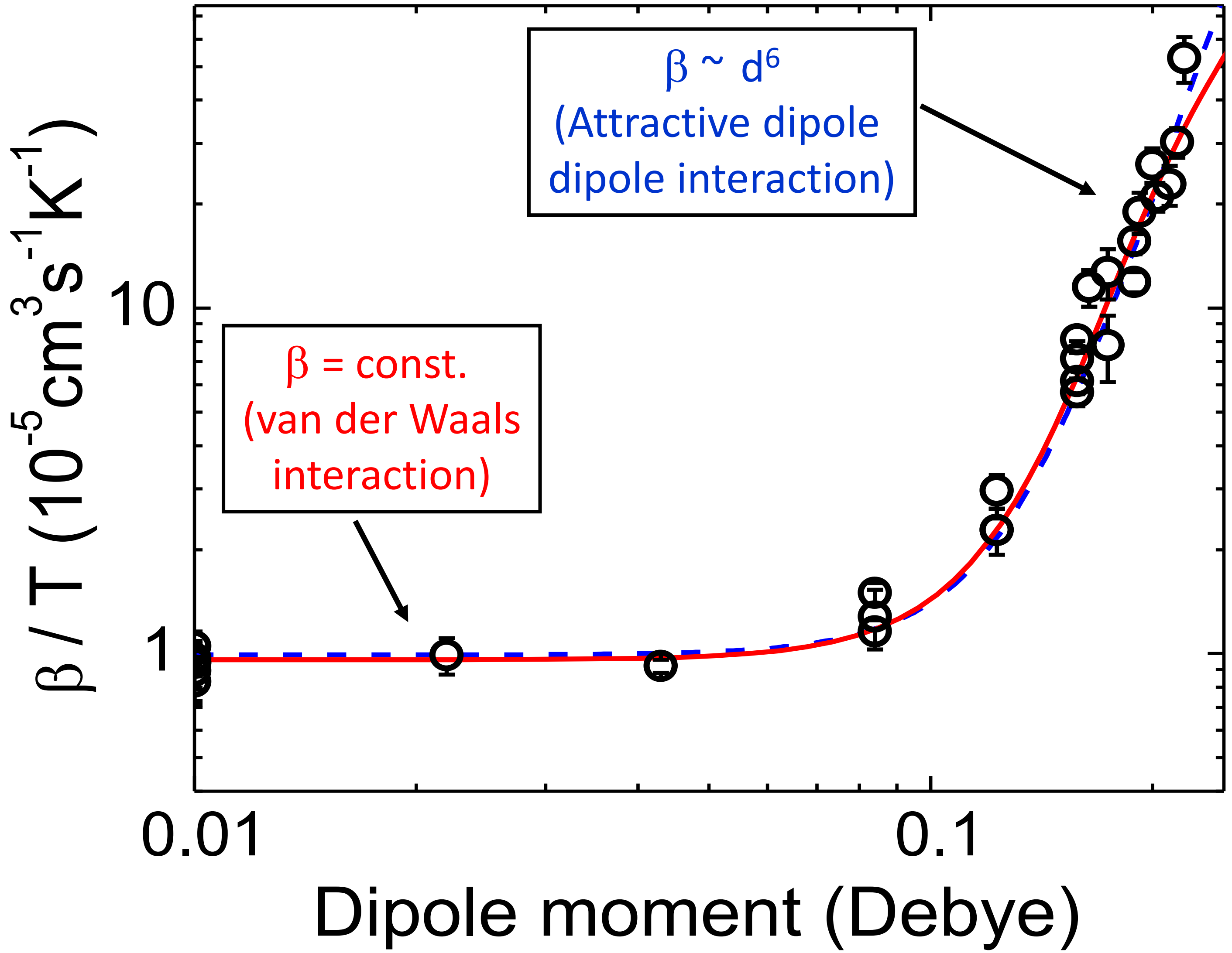
Ni *et al.*, Nature **464**, 1324 (2010).

- Dipolar interaction enhances reaction
- Stereo chemistry – geometry matters

Long-range potential

$$V(R) = \frac{\hbar^2 L(L+1)}{2\mu R^2} - \frac{C_6}{R^6} - \frac{C_3}{R^3}$$





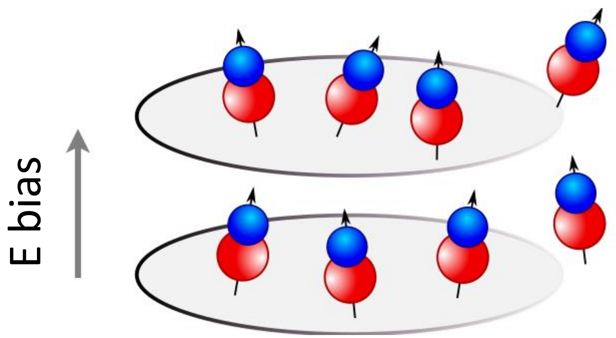
Dipolar physics in 2D

Büchler, Bohn, Bruun, Demler, Julienne, Lewenstein, Pupillo, Santos, Shlyapnikov, Baranov/Zoller, …

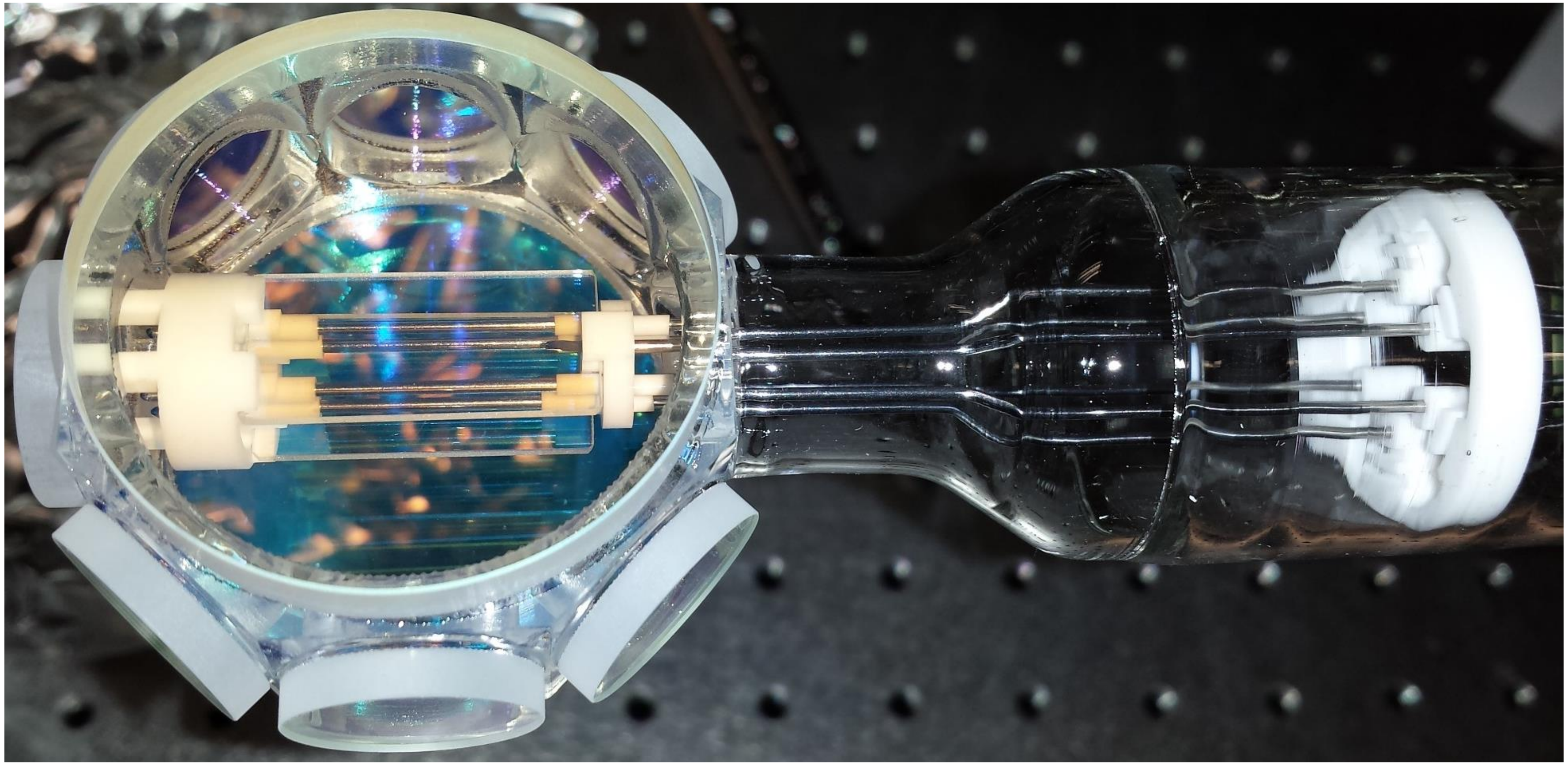
Büchler, Demler, Lukin, Micheli, Prokof'ev, Pupillo, & Zoller, PRL **98**, 060404 (2007).

- Suppression of chemical reaction
- Crossover from 3D regime to 2D
- Elastic interaction >> inelastic loss
- Dipolar evaporative cooling
- Dipole-mediated collision resonance

latest → Bulk gas in 3D
Dipolar exchanges



Control interactions in a molecular quantum gas



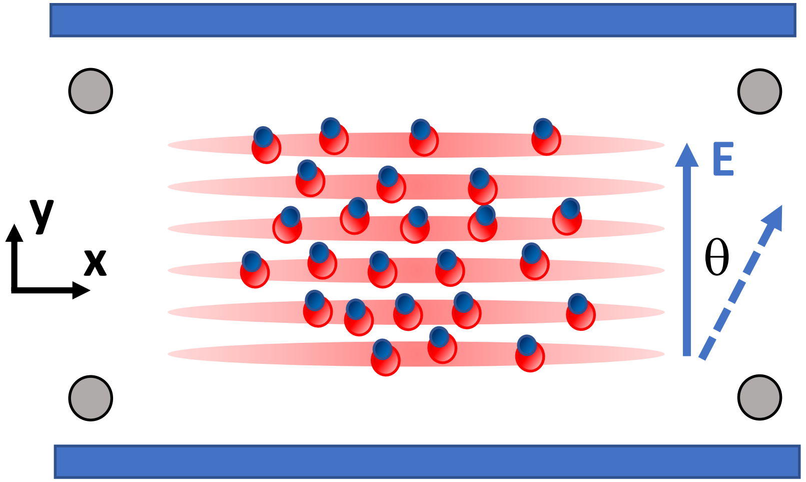
- All 6 electrodes are in vacuum

  15 kV/cm, arbitrary orientation, reconfigurable gradients

- Use E to suppress loss & tune dipolar interaction

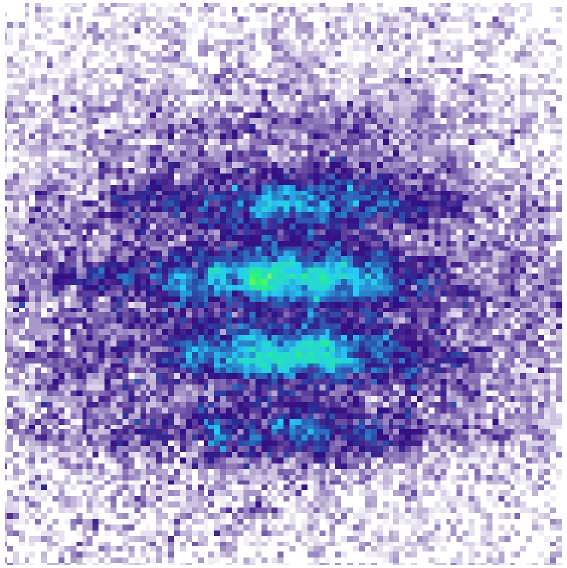
Polar molecules under precise E-field control

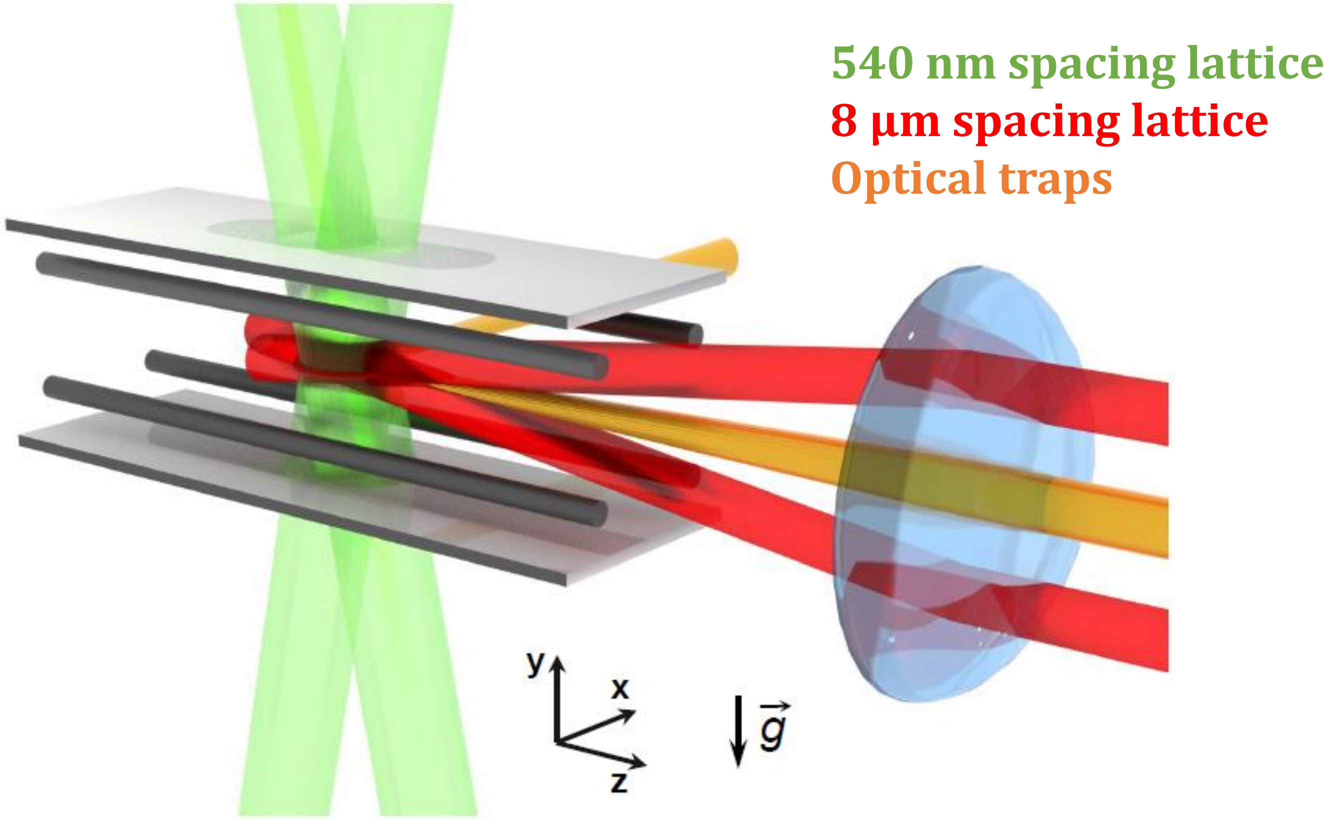
Valtolina *et al*., Nature **588**, 239 (2020).



- E_{DC}: angle θ, 0 to 15 kV/cm (known to 10^{-4})
- Gradient (x, y): 0 to 8 kV/cm^2; Curvature
- RF for rotational transition (~ 100 kHz Rabi)

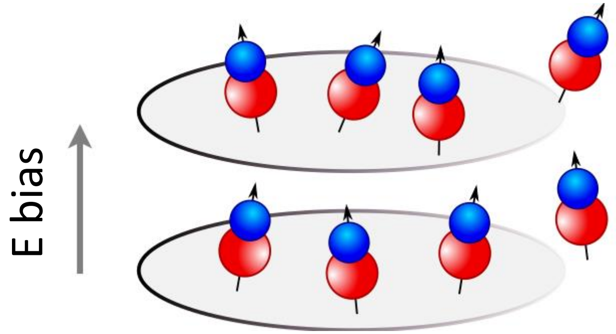
4 Layers

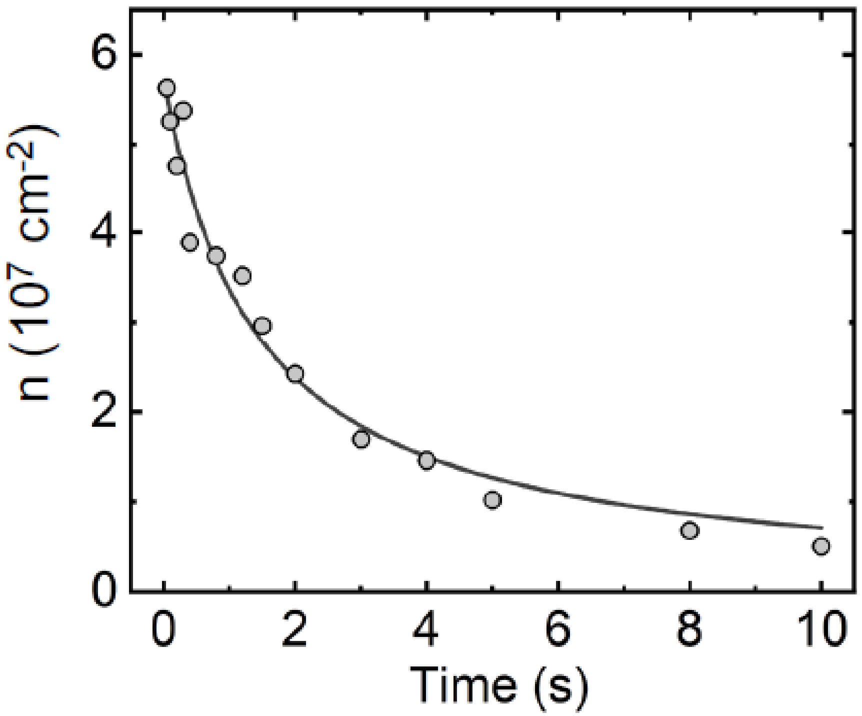


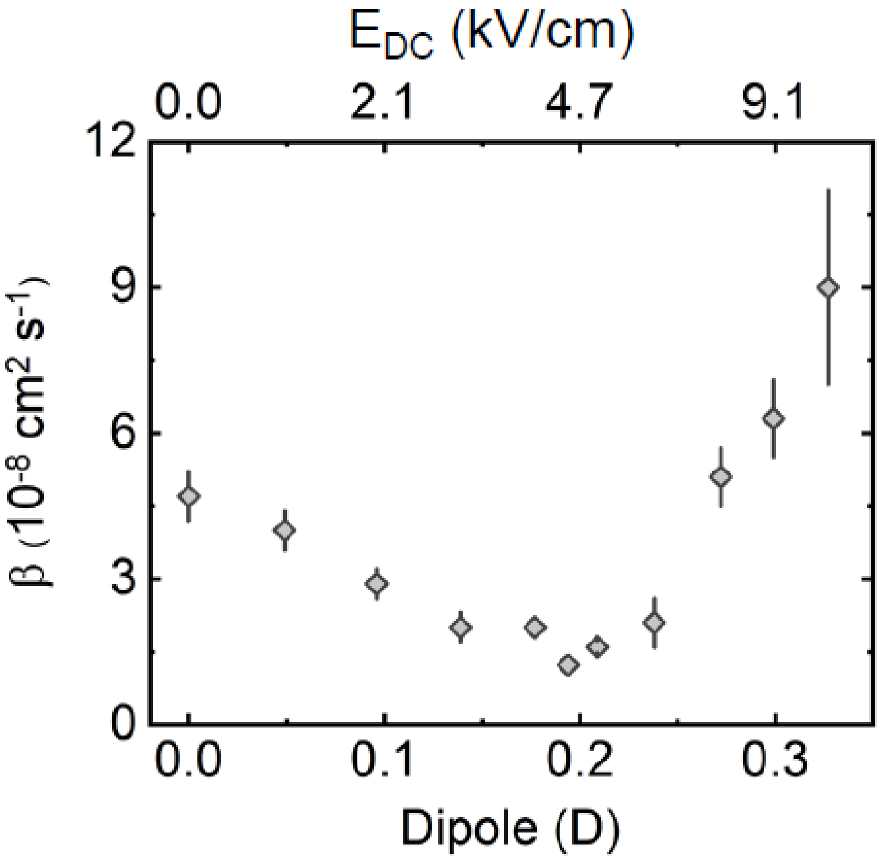


Suppression of inelastic loss in 2D

- For dipole moment of 0.2 D, elastic / inelastic ~200

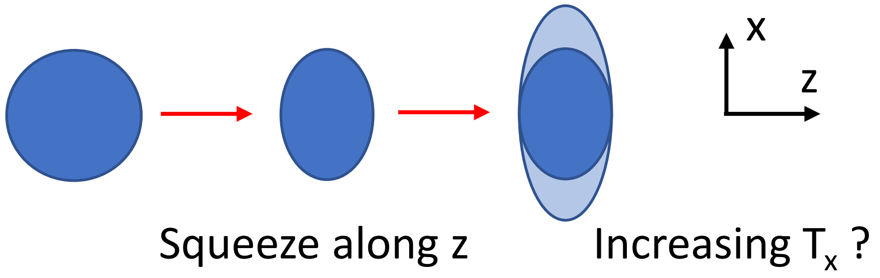


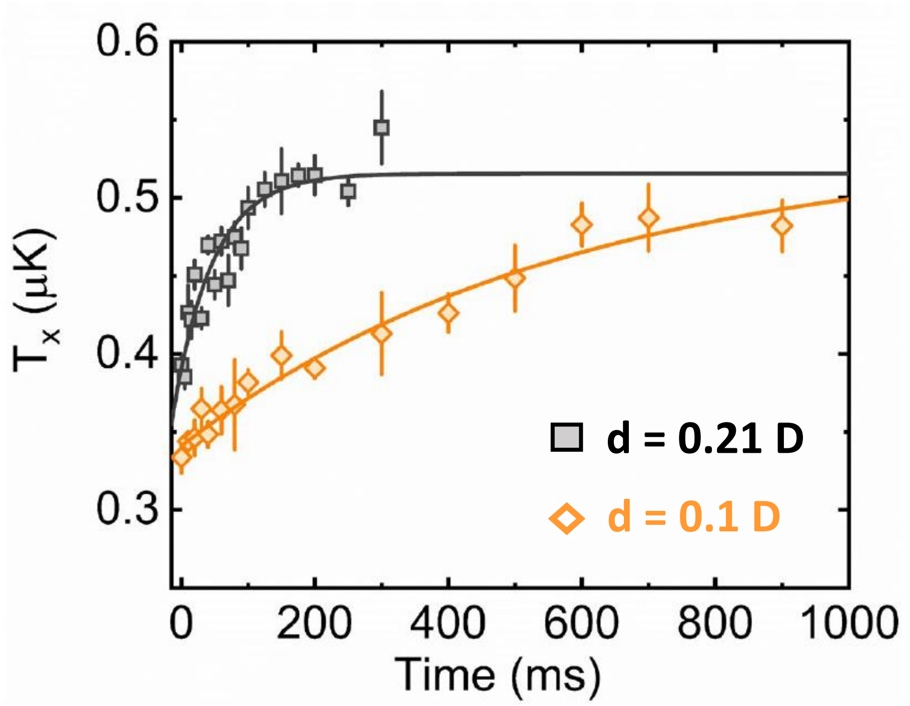


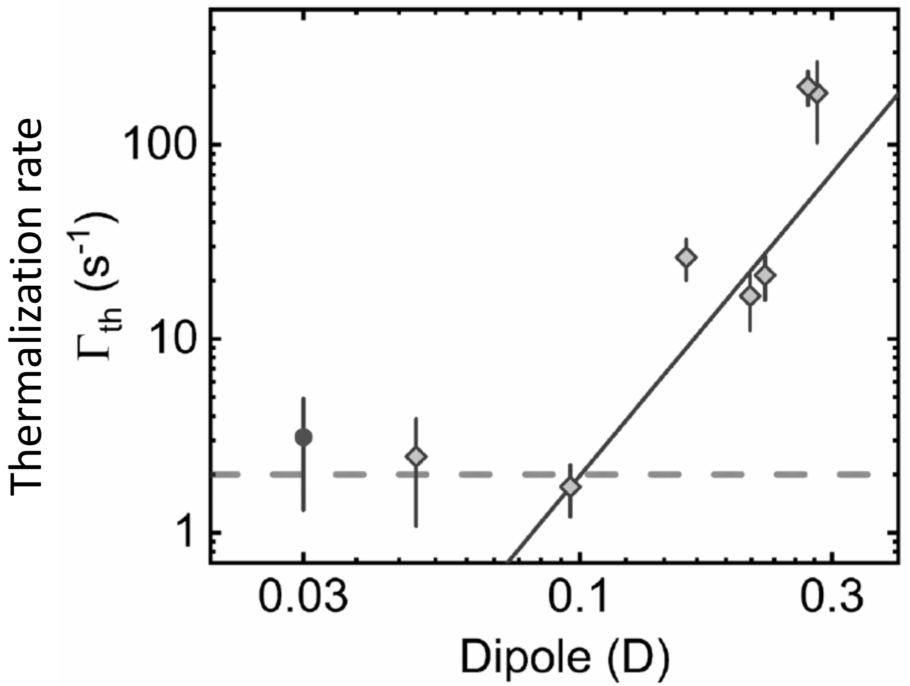


Dipolar thermalization

- Cross dimensional thermalization
- Determine dipolar collision cross-section







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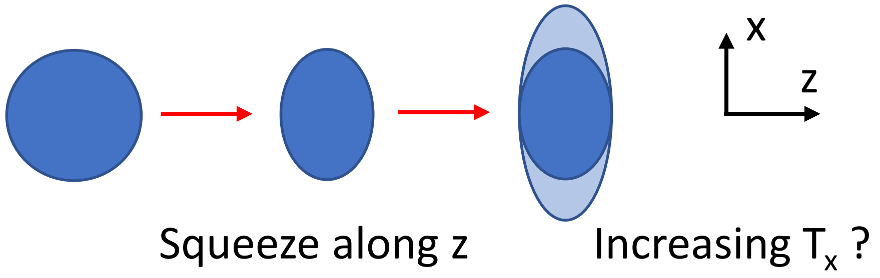
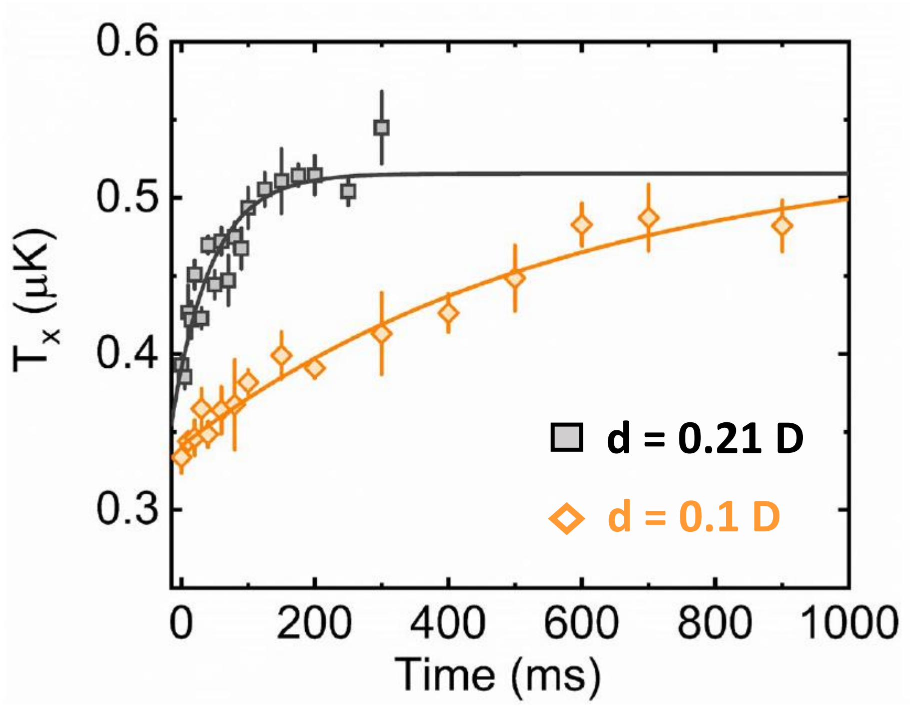
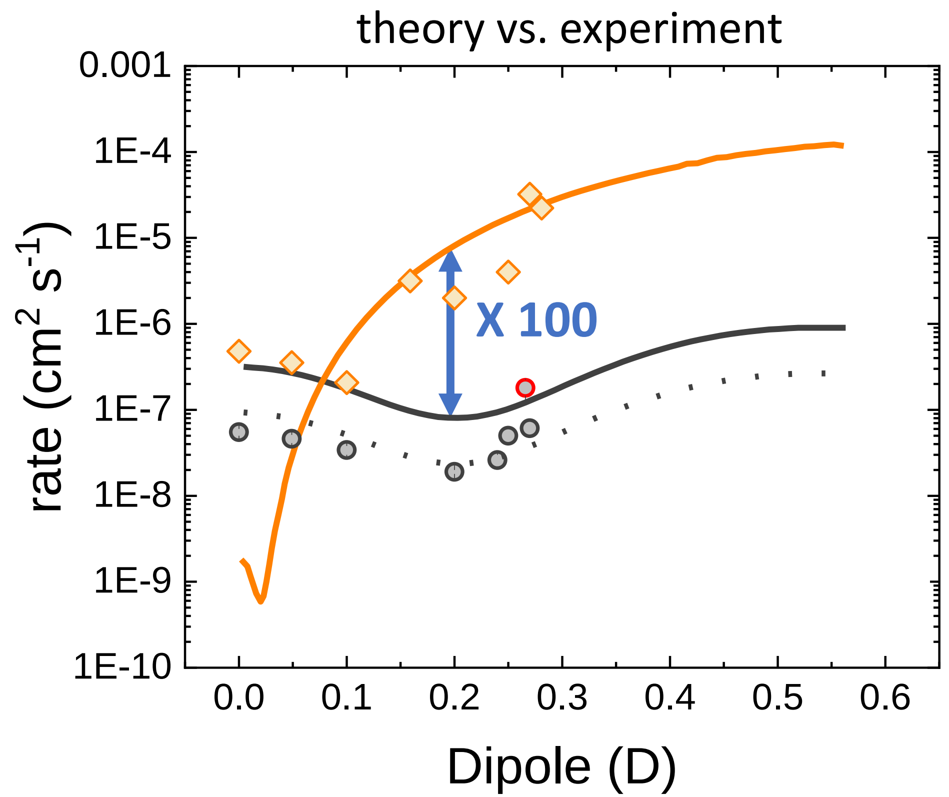
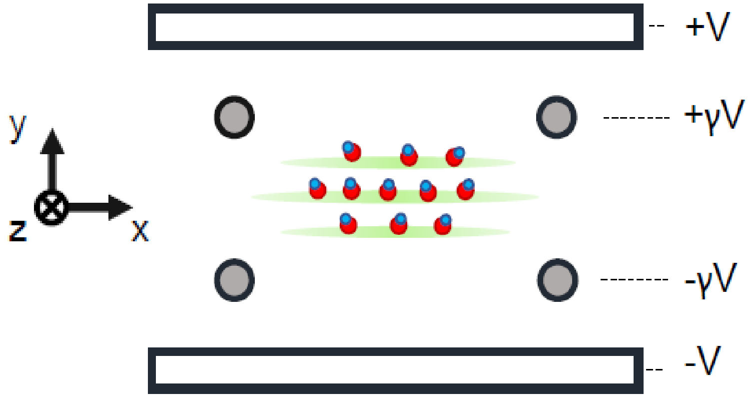




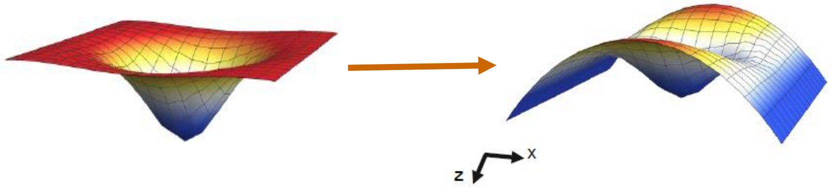


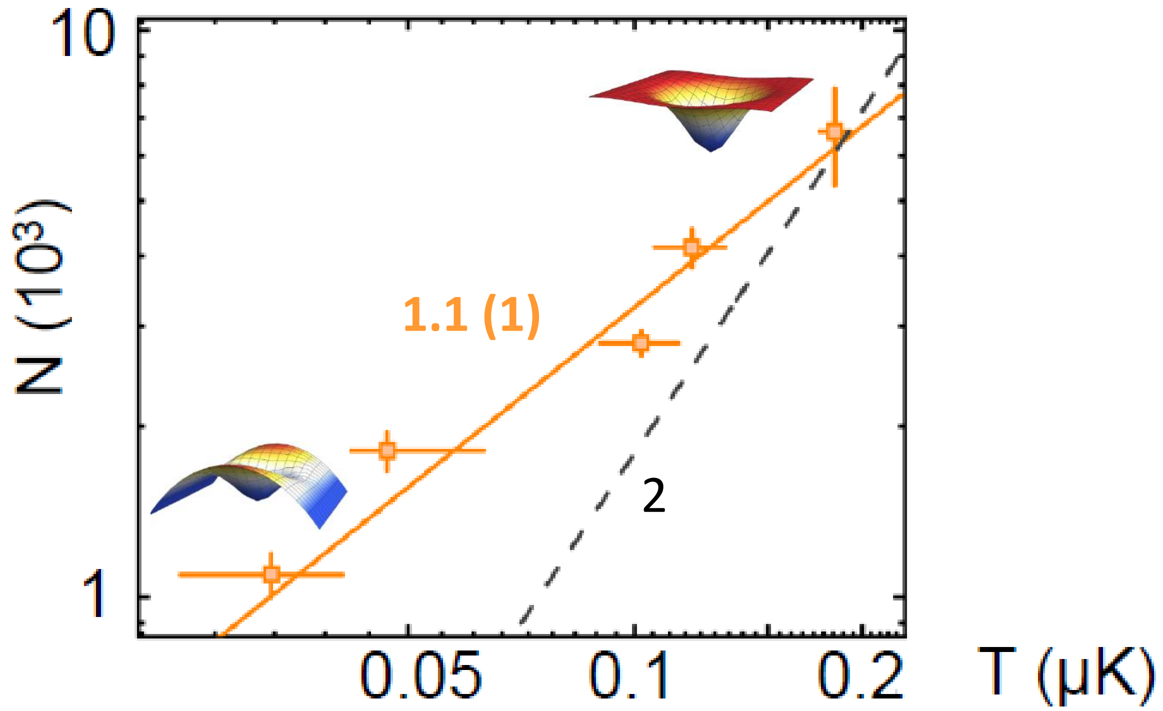
Dipole-assisted evaporation

Control trap potential with E & its gradient



Increase anti-curvature/decrease trap depth





Criterion for good evaporation:

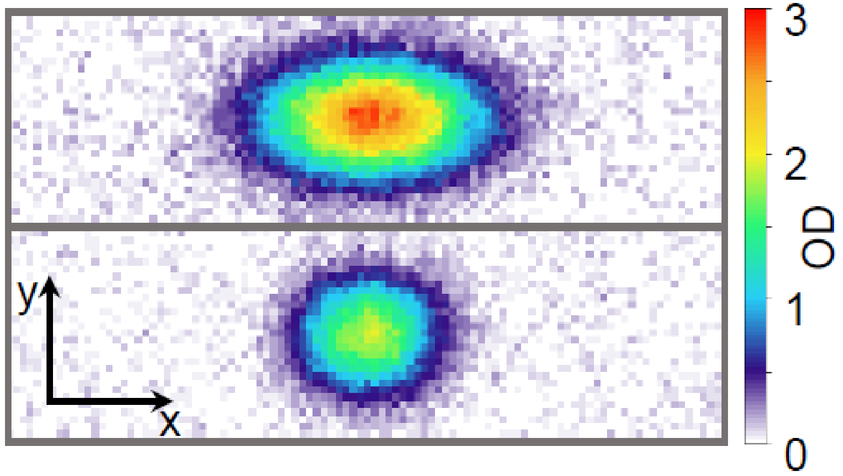
$$\frac{\partial \log N}{\partial \log T} < D = 2$$

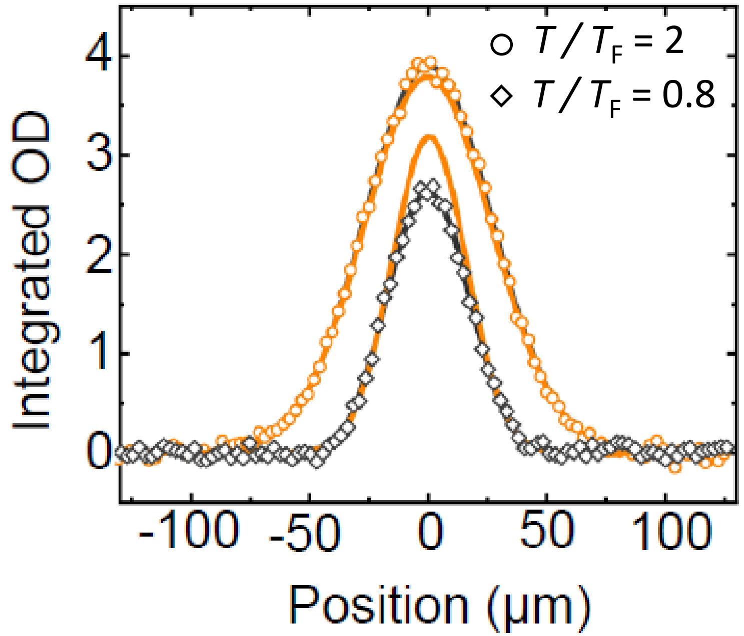
Dipole-assisted evaporation

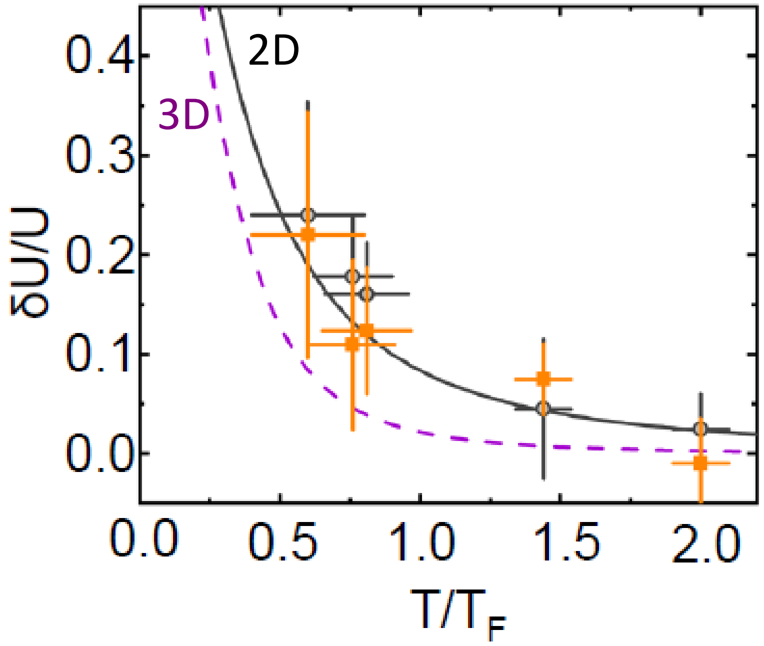
Valtolina *et al*., Nature **588**, 239 (2020).

Evaporation image

- thermal
- degenerate

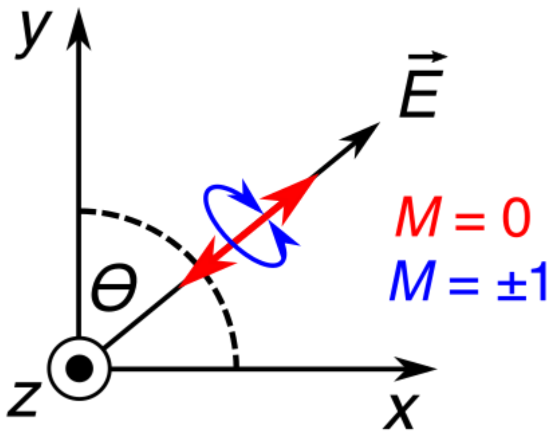


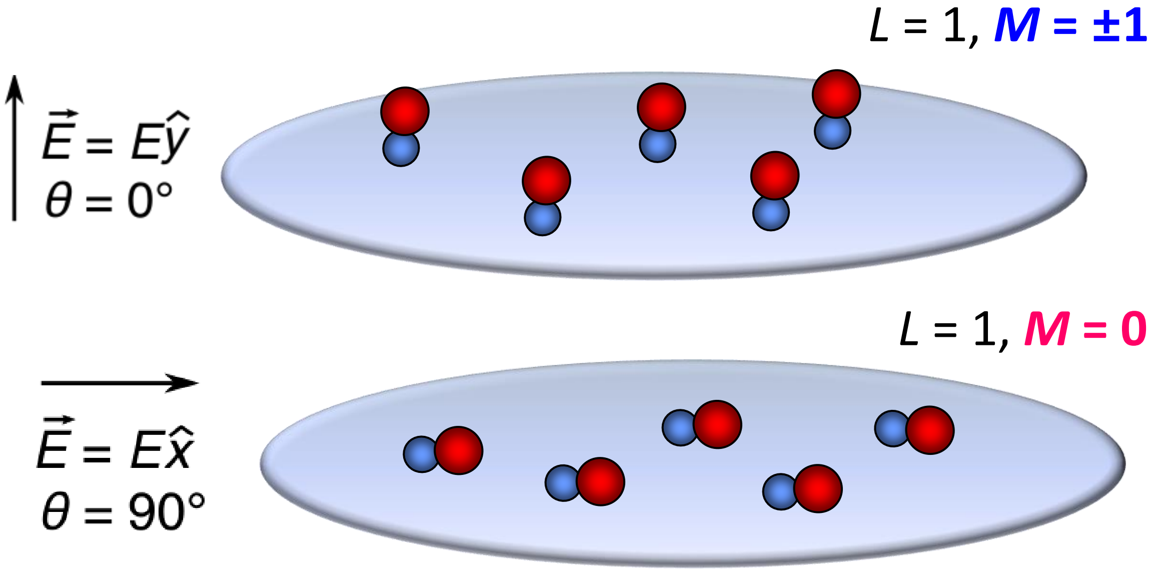


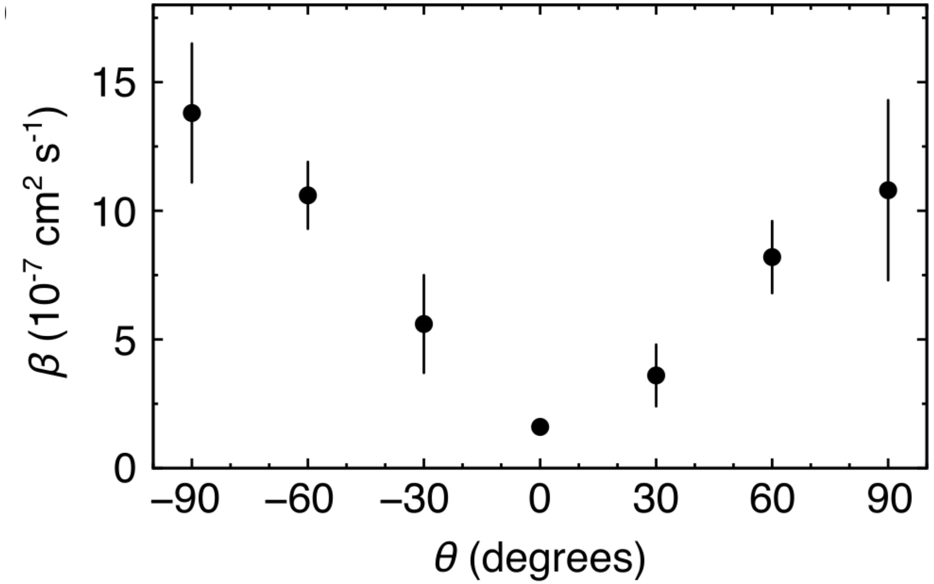


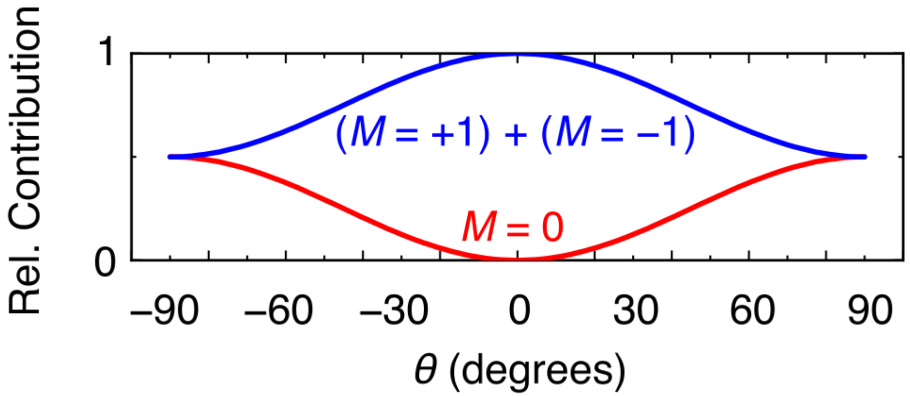
Control of reactions with **E** field orientation

Matsuda *et al*, Science **370**, 1324 (2020).

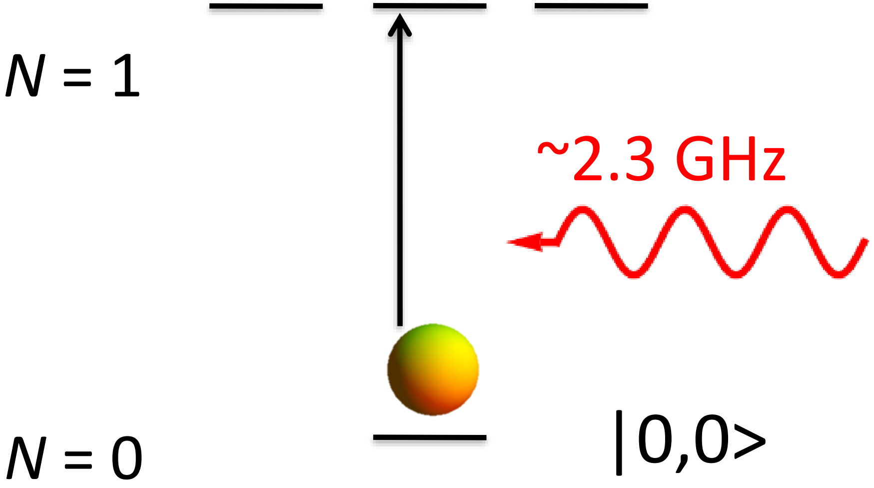


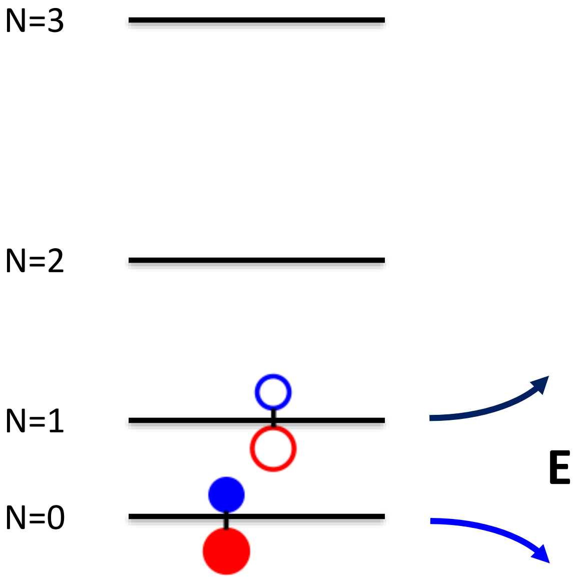




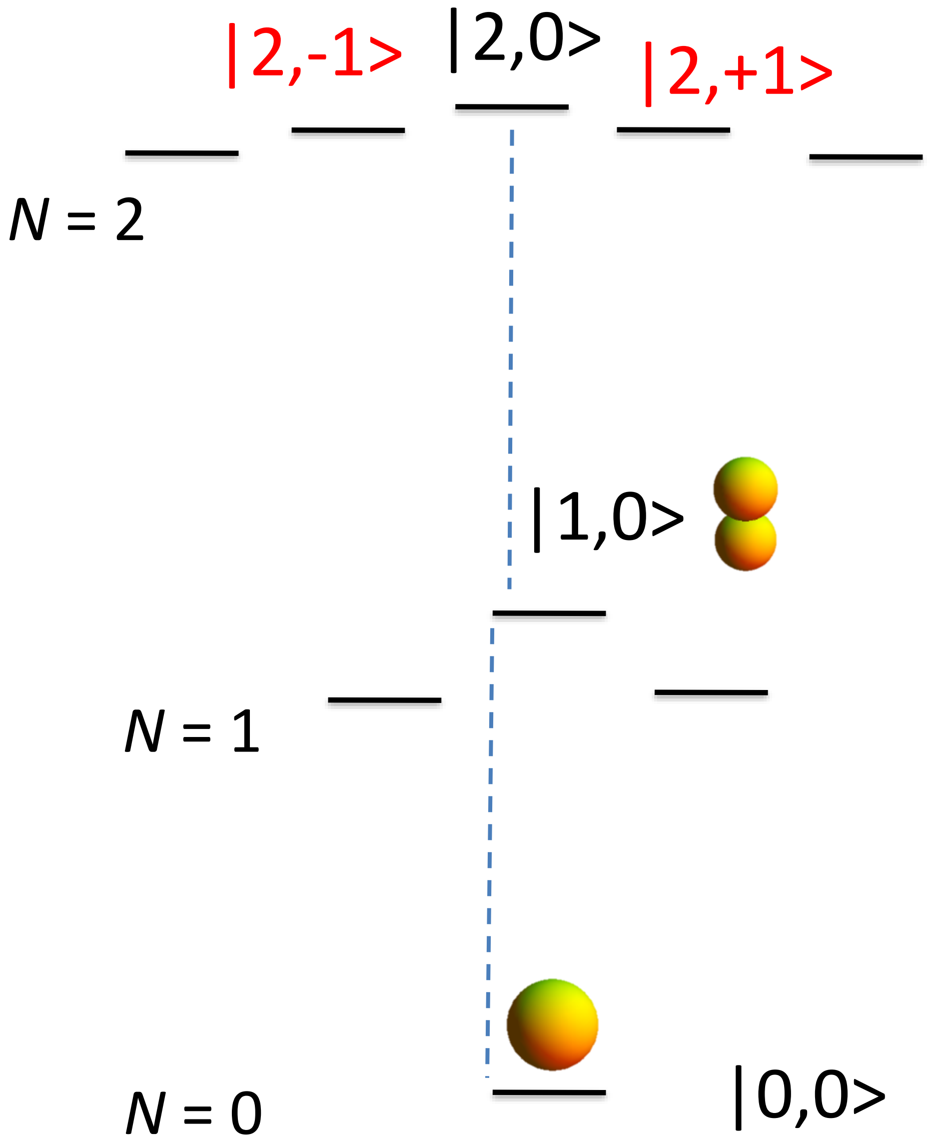


Molecular energy tuned with |E|





Molecular energy tuned with |E|

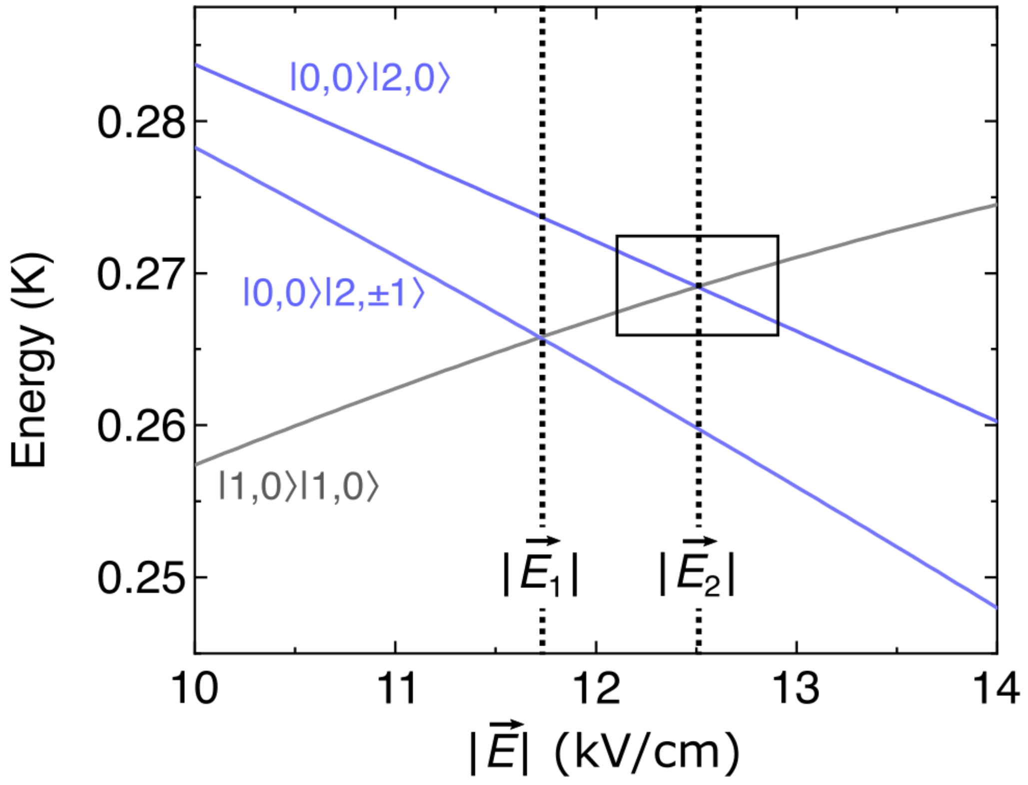


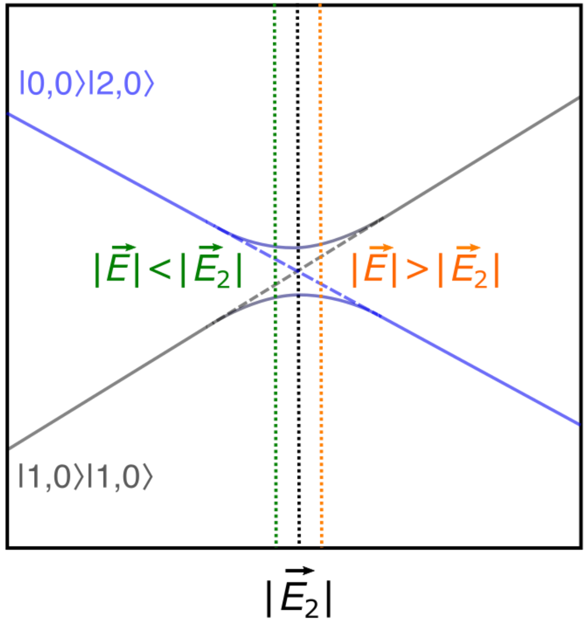
|1,0> + |1,0>
= |0,0> + |2,0>

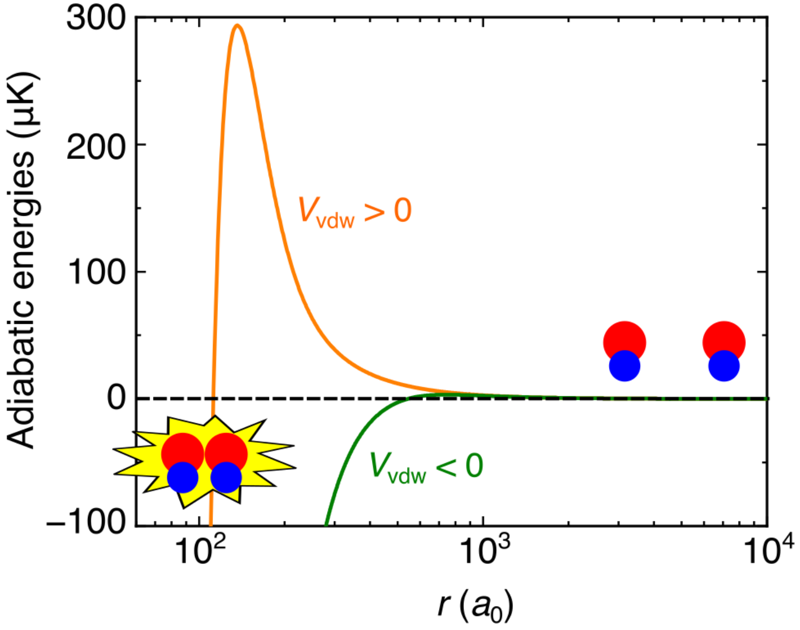
or,

|1,0> + |1,0>
= |0,0> + |2,±1>

E field induced resonance & shielding

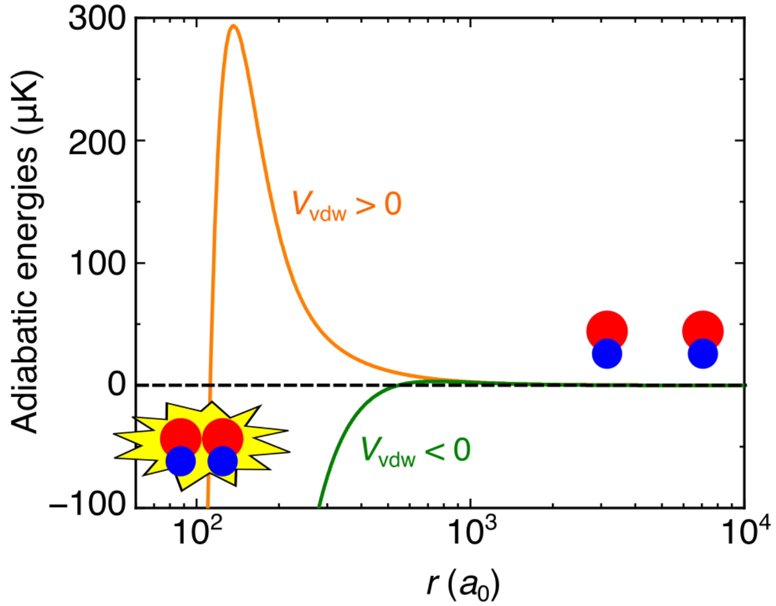


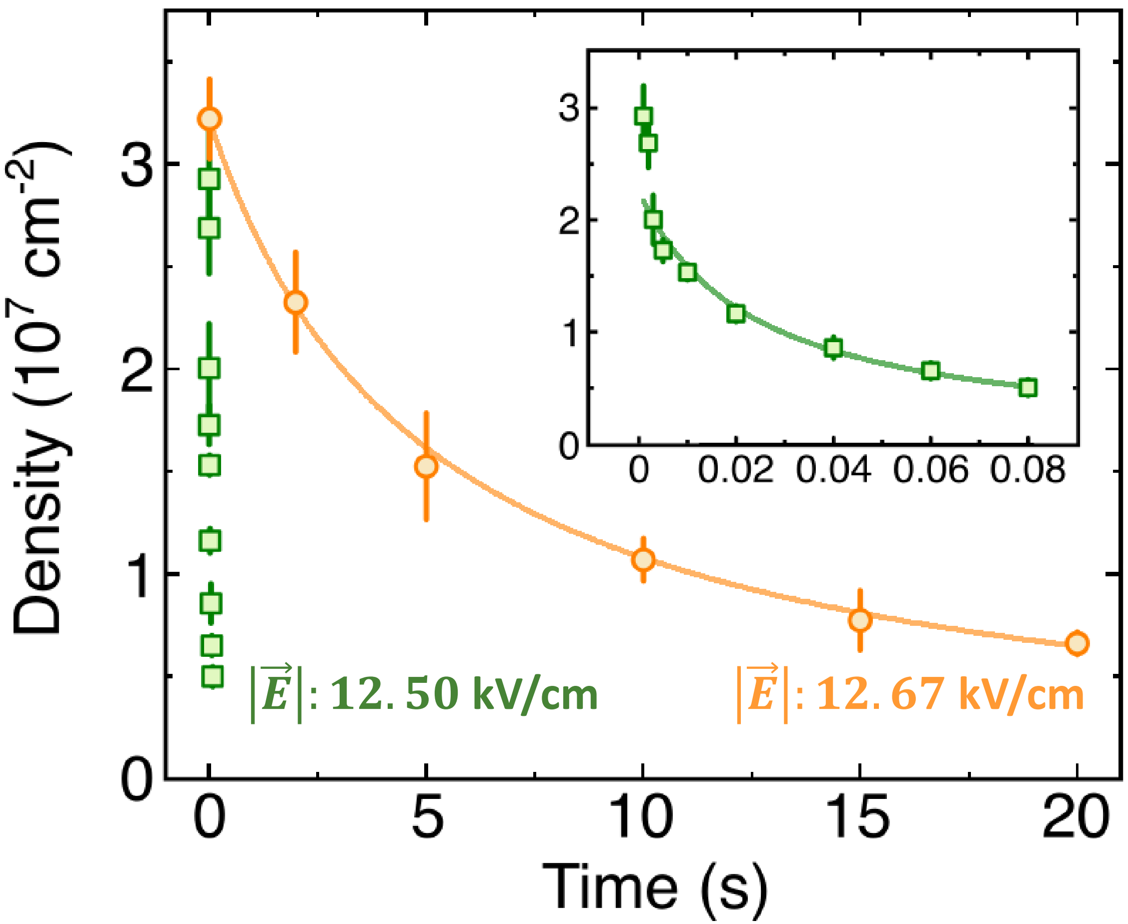




Resonant shielding & enhancement of reaction

Matsuda *et al*, Science **370**, 1324 (2020).

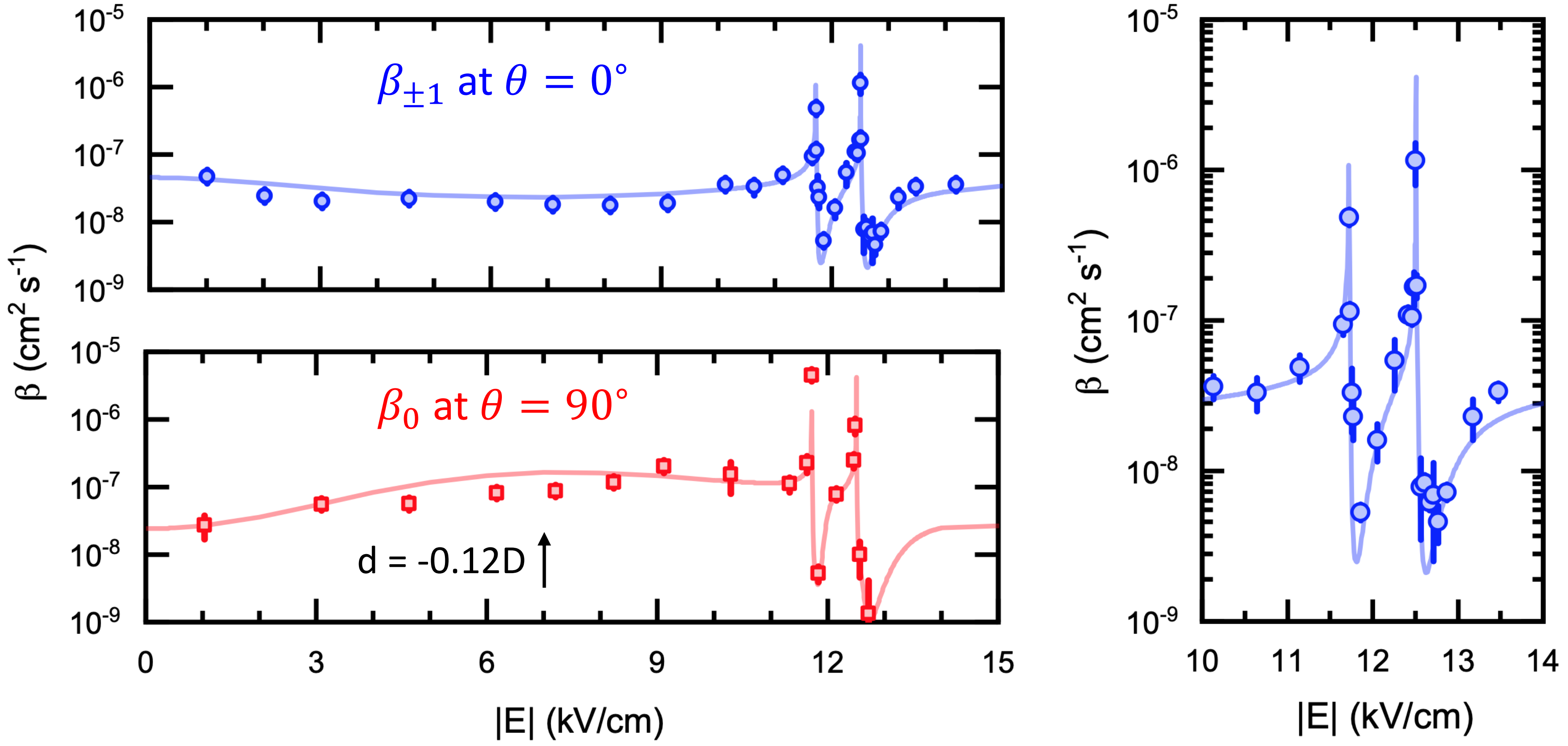




Characterization of resonant shielding

Matsuda *et al*, Science (Dec. 10, 2020).

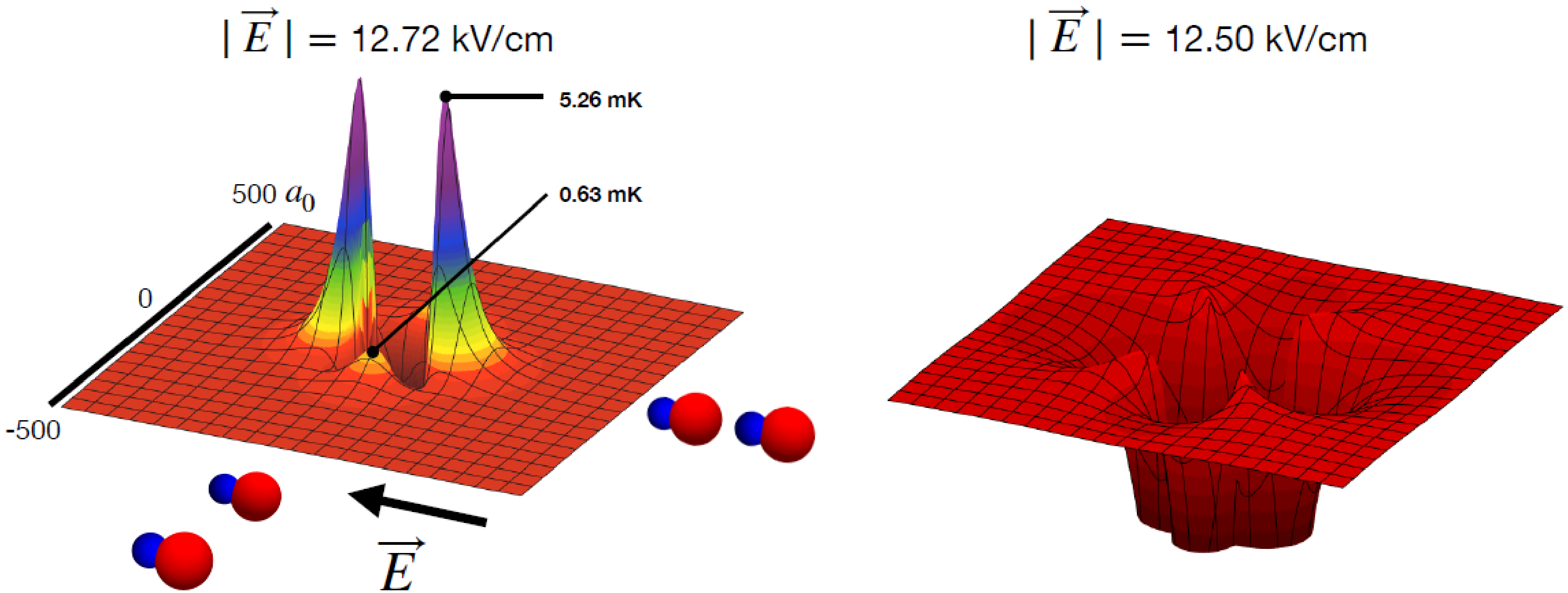
Solid lines: no free parameter scattering theory (Goulven Quéméner)



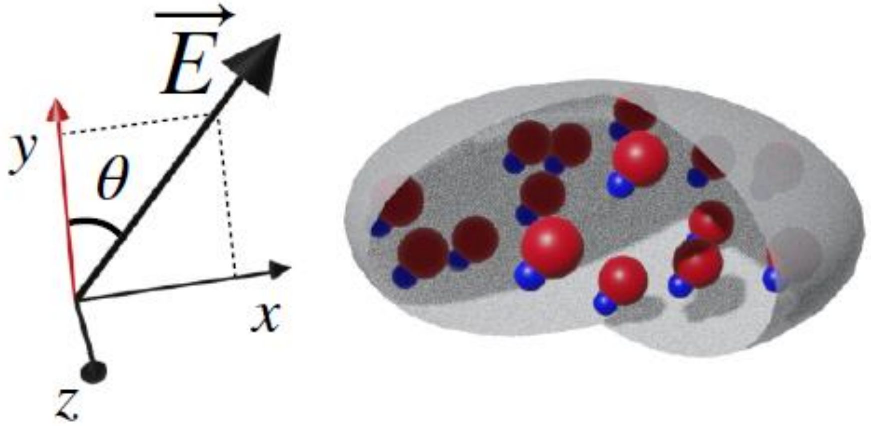
- Dipolar interaction controls collision resonances
- Shielding for both M = 0 & ±1 channels

Resonant shielding works in 3D !

J. Li *et al*, arXiv:2103.06246 (Nature Phys. 2021).

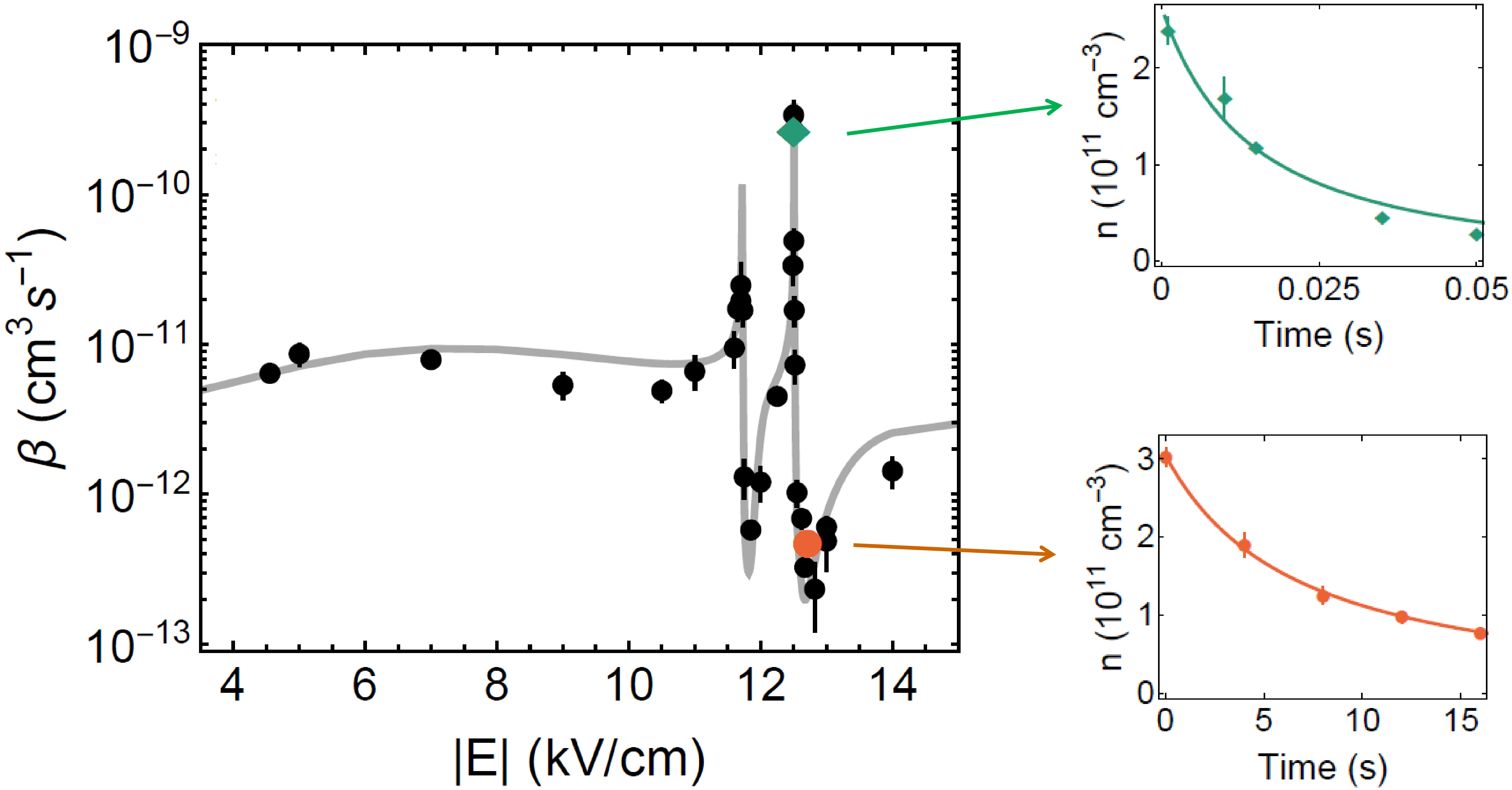


Collaboration: John Bohn, Goulven Quéméner

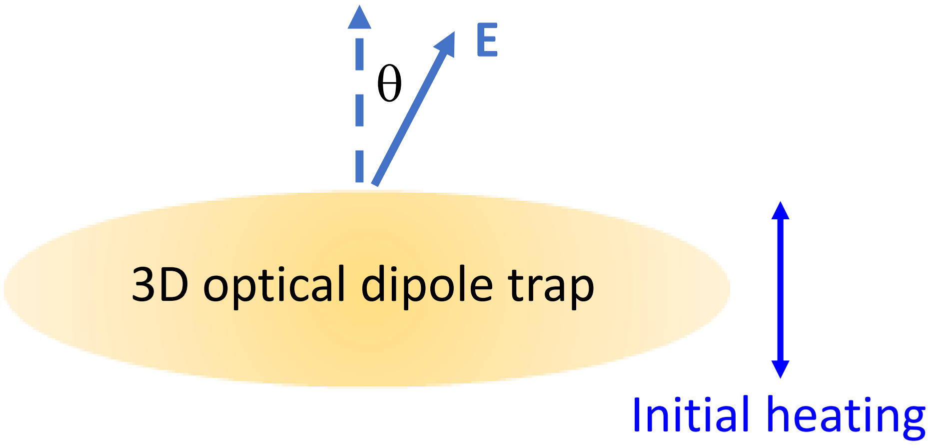


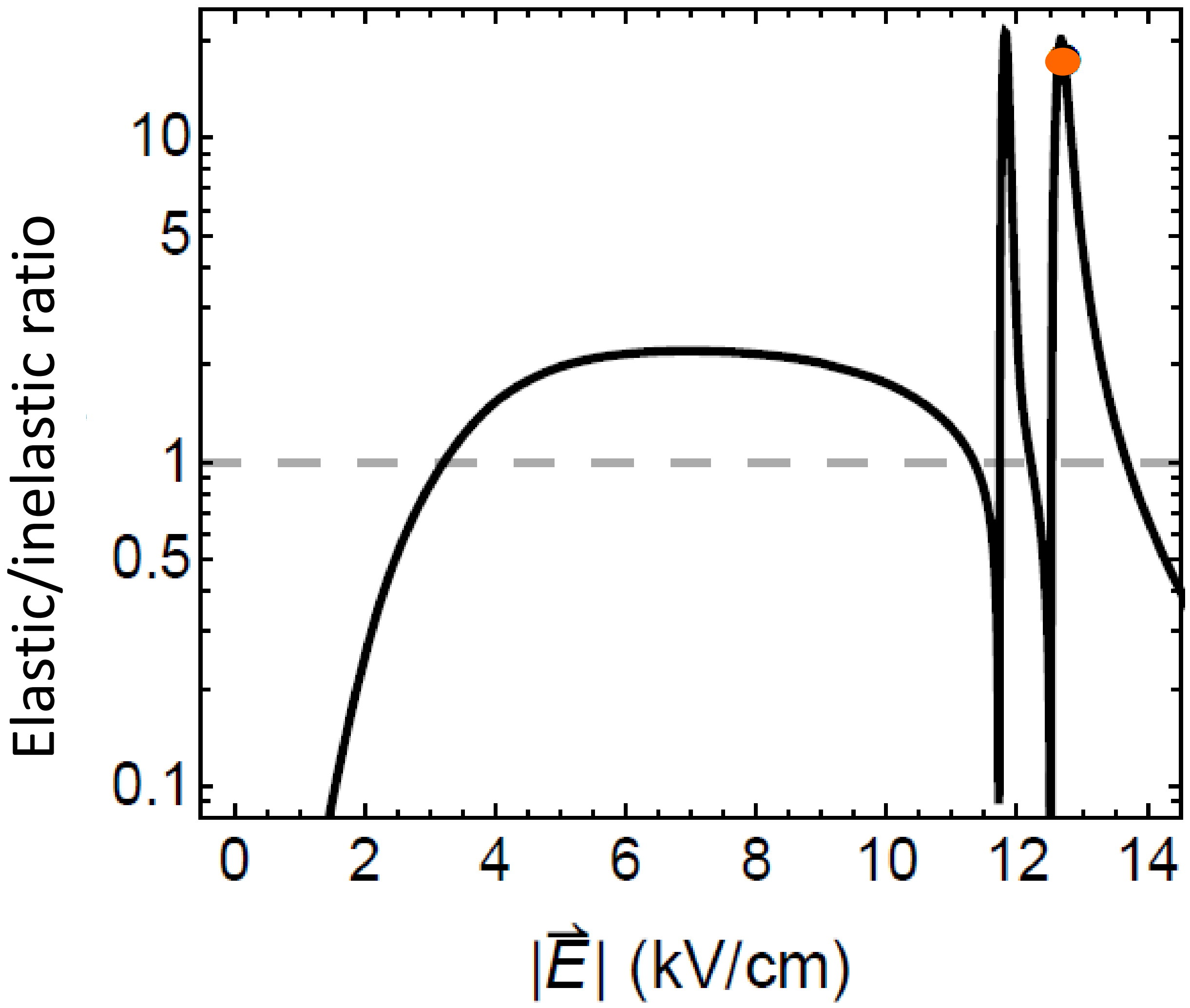
Trap frequencies: $(\omega_x, \omega_y, \omega_z) = 2\pi \times (40, 220, 40)$ Hz

Resonant shielding works in 3D !



Dipolar thermalization in 3D, with reactive molecules

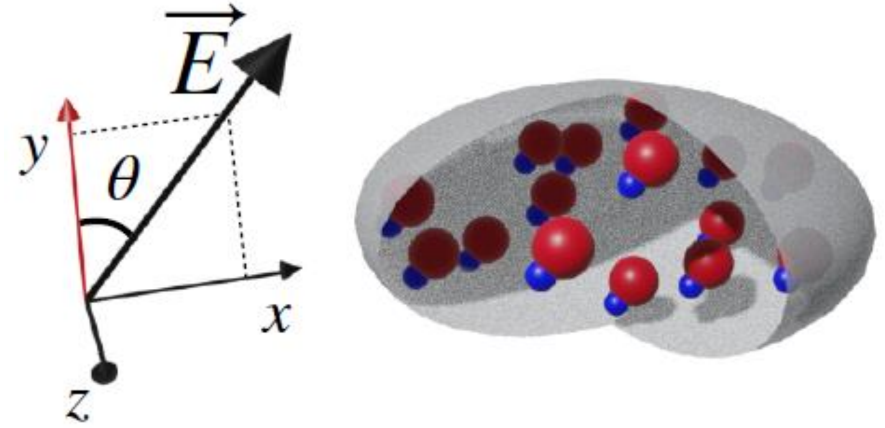




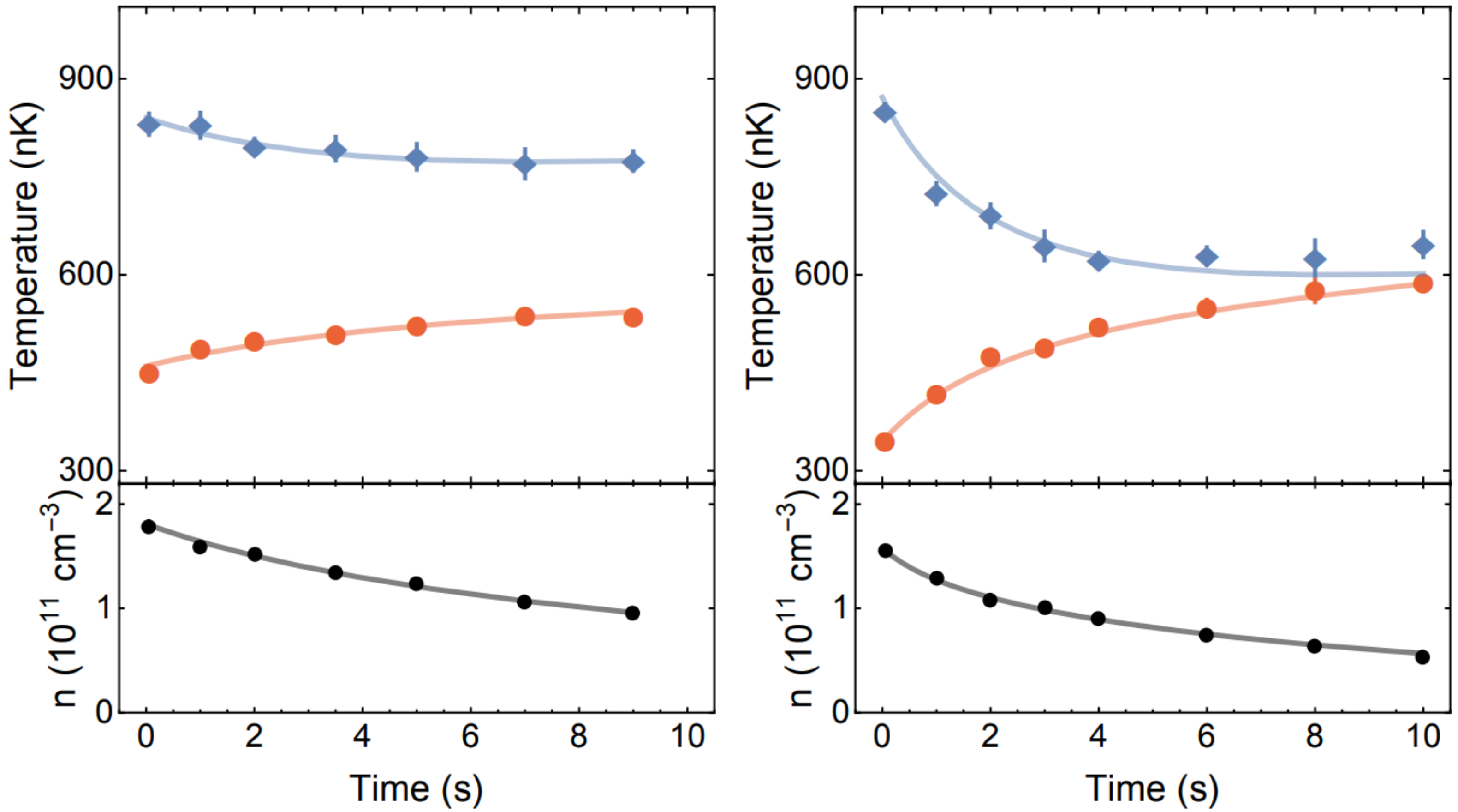
Dipolar thermalization in 3D

Anisotropic thermalization for magnetic atoms

(Ferlaino *et al.*, PRL 2014, Jin & Bohn PRA 2014)



$\theta = 90^\circ$ $\theta = 45^\circ$



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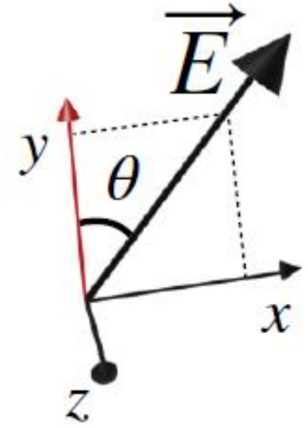
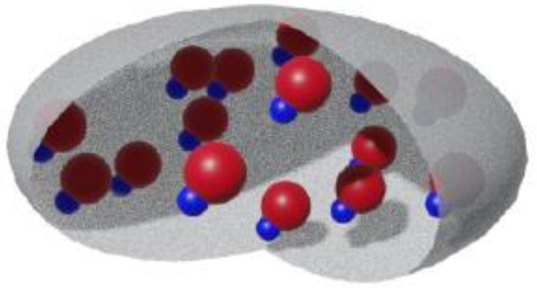
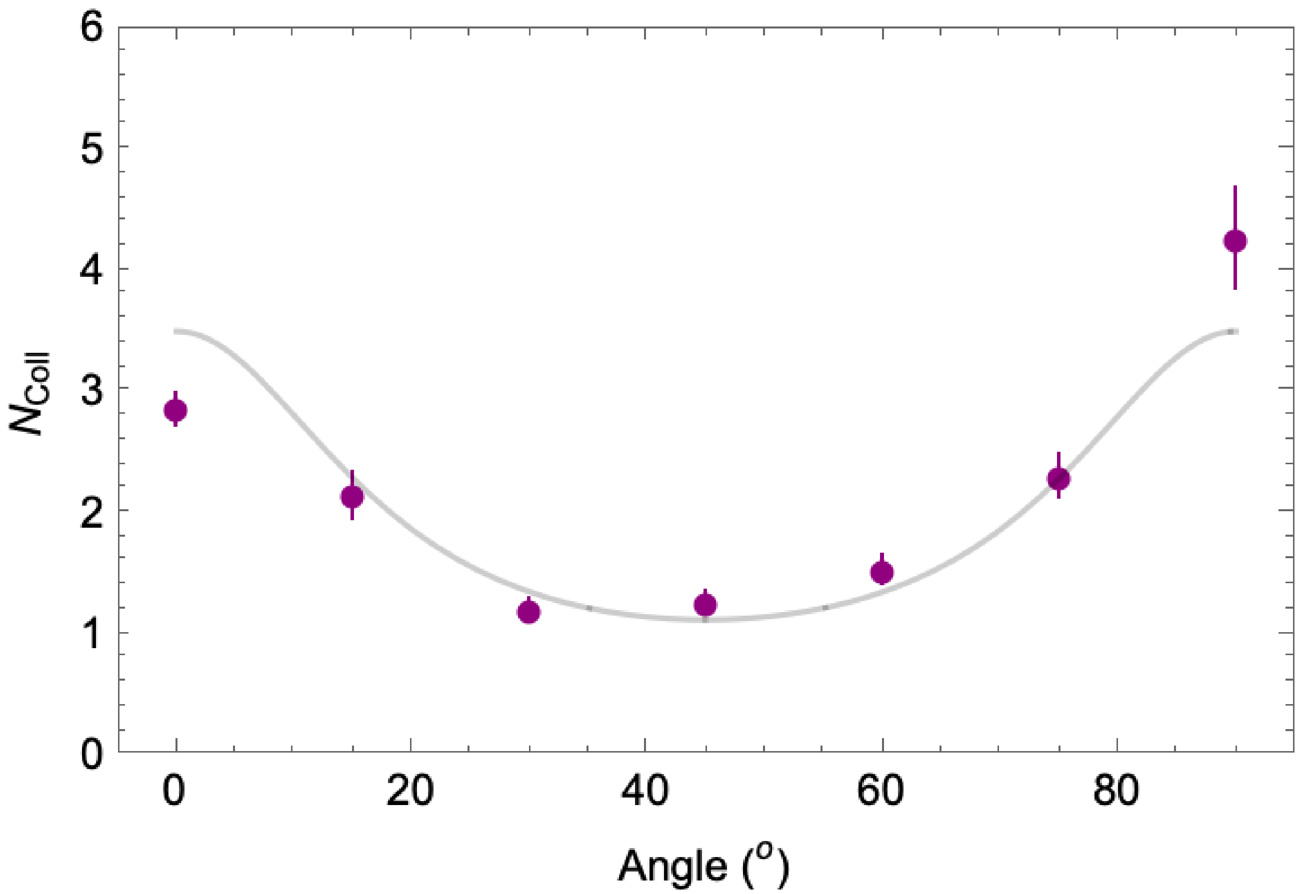
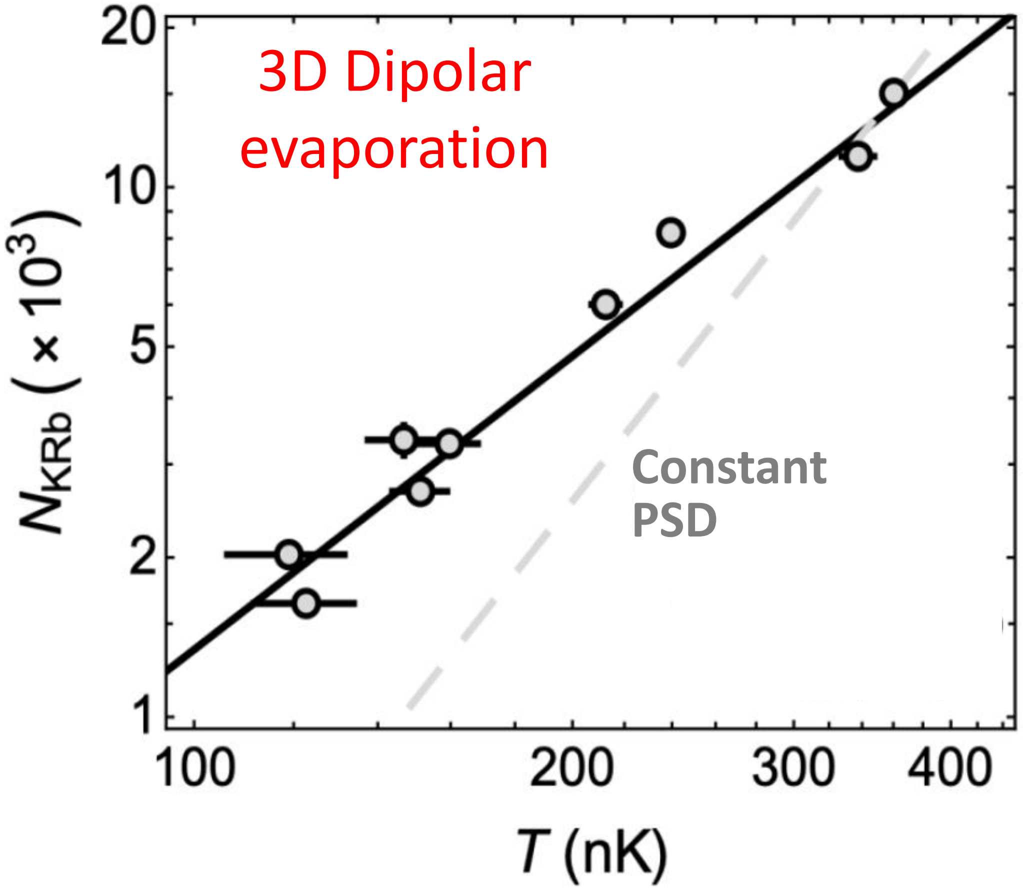


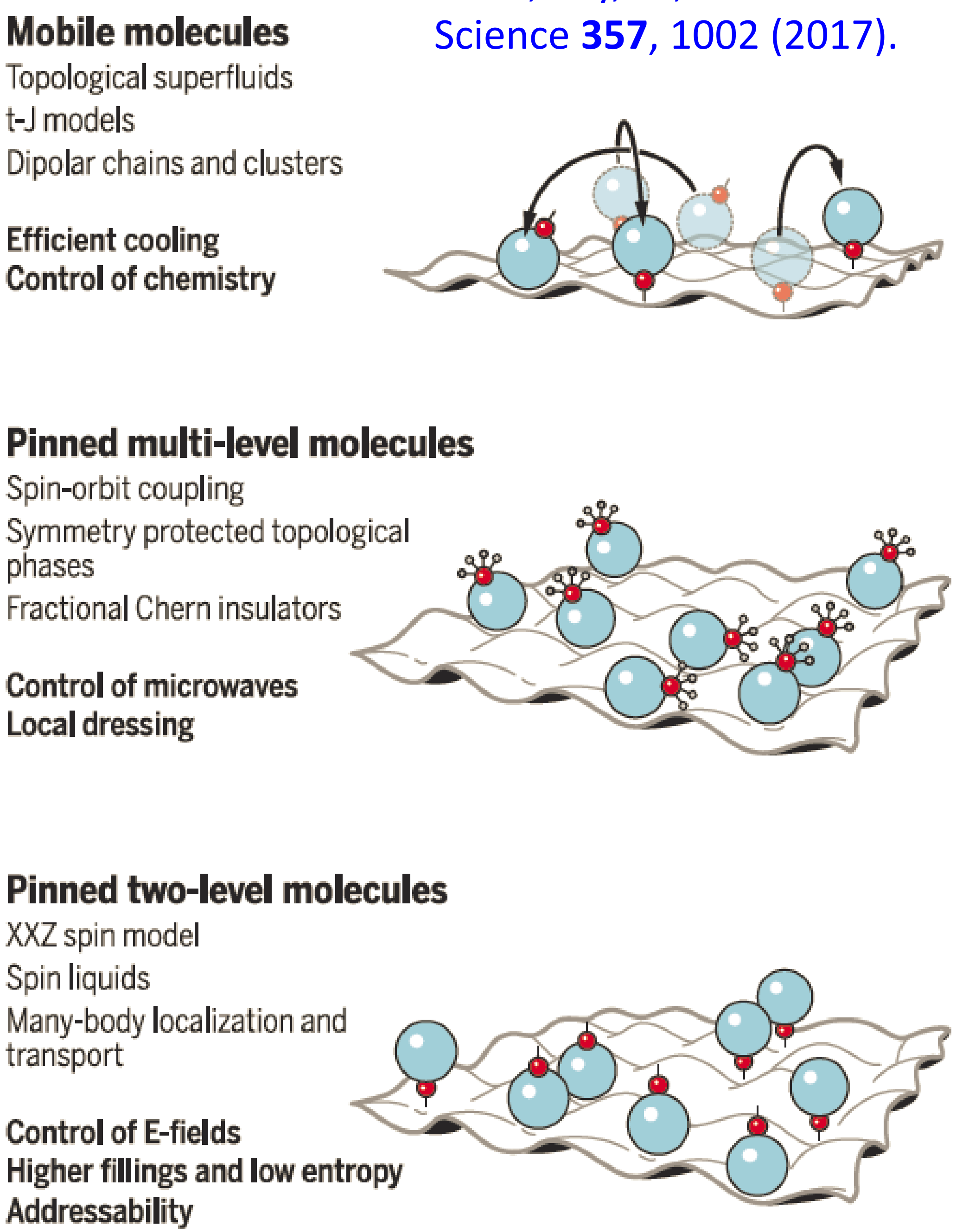






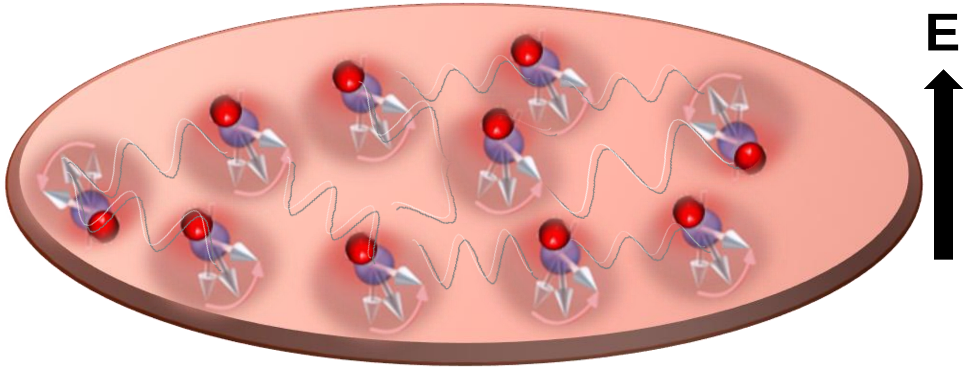
Many exciting explorations

Bohn, Rey, Ye,
Science **357**, 1002 (2017).

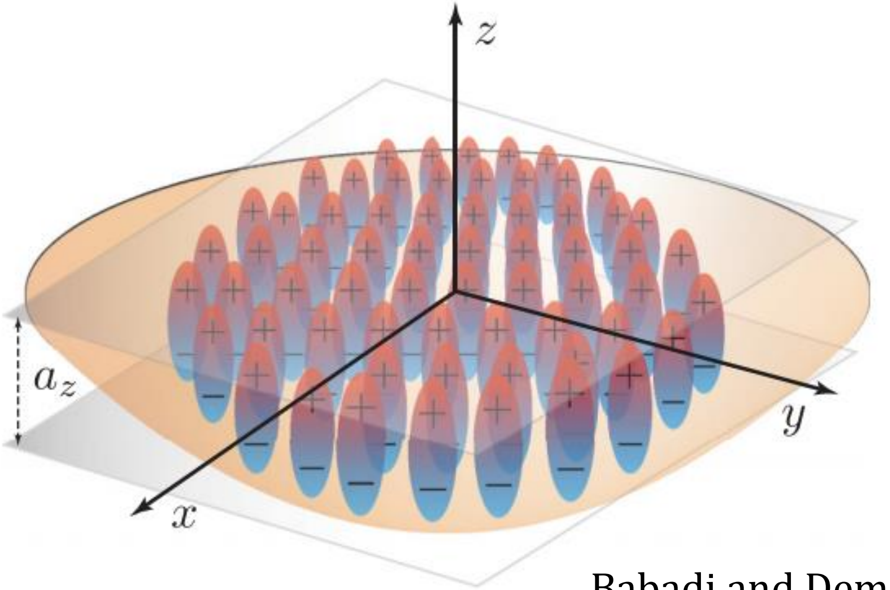


Collective spins in 2D plane (spin squeezing)

Bilitewski *et al.*, Phys. Rev. Lett. in press (2021).



Collective hydrodynamic modes of dipolar fermions

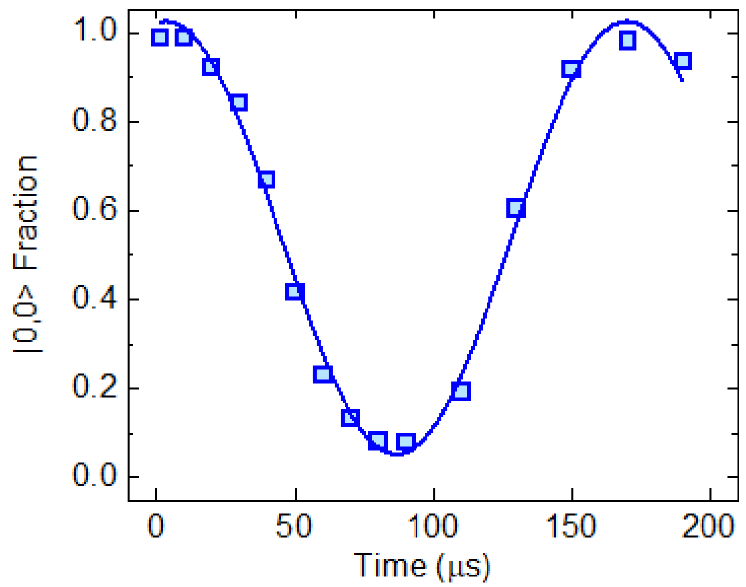


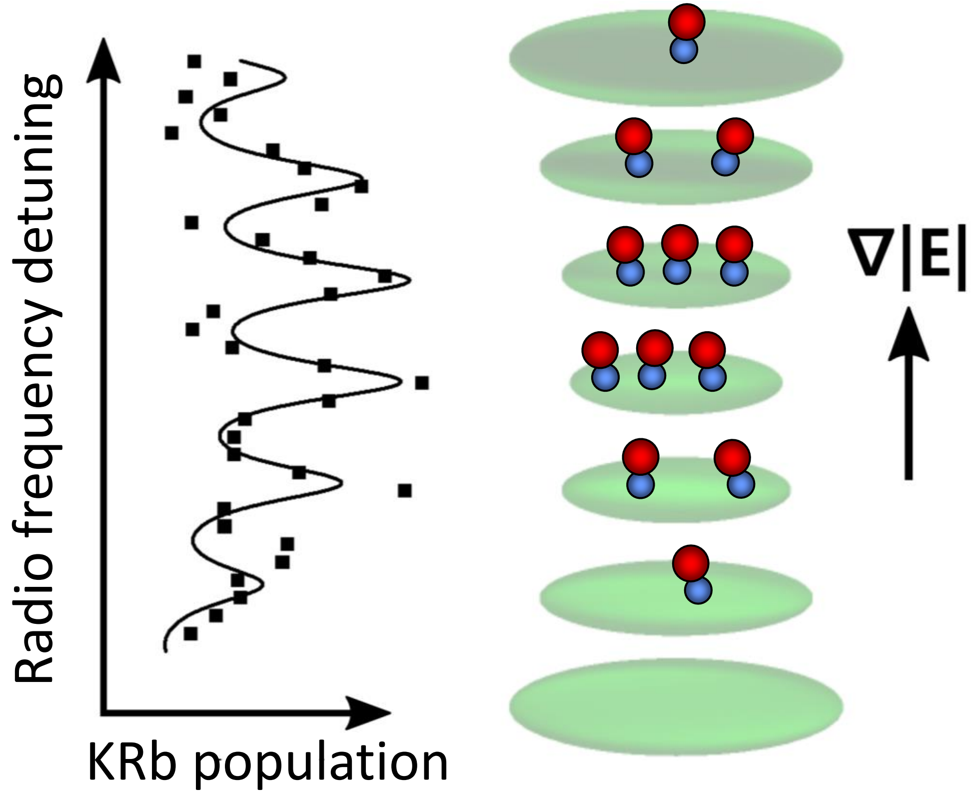
Babadi and Demler, PRA (2012)

Selecting a single layer of molecules

- Molecules confined in 1D optical lattice (540 nm spacing)
- Stabilize lattice phase relative to $\boldsymbol{E}$; Apply $\nabla \boldsymbol{E}$ along lattice
- Single-site address of the rotational transition

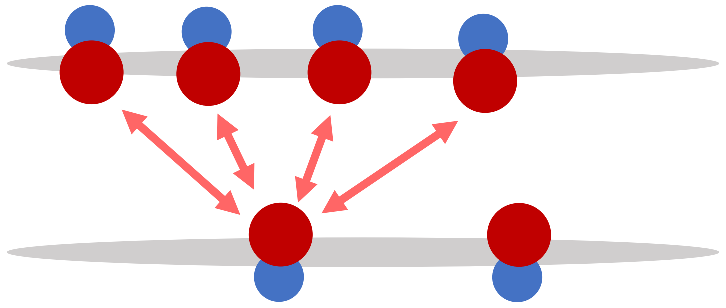
Single-site Rabi spectroscopy:

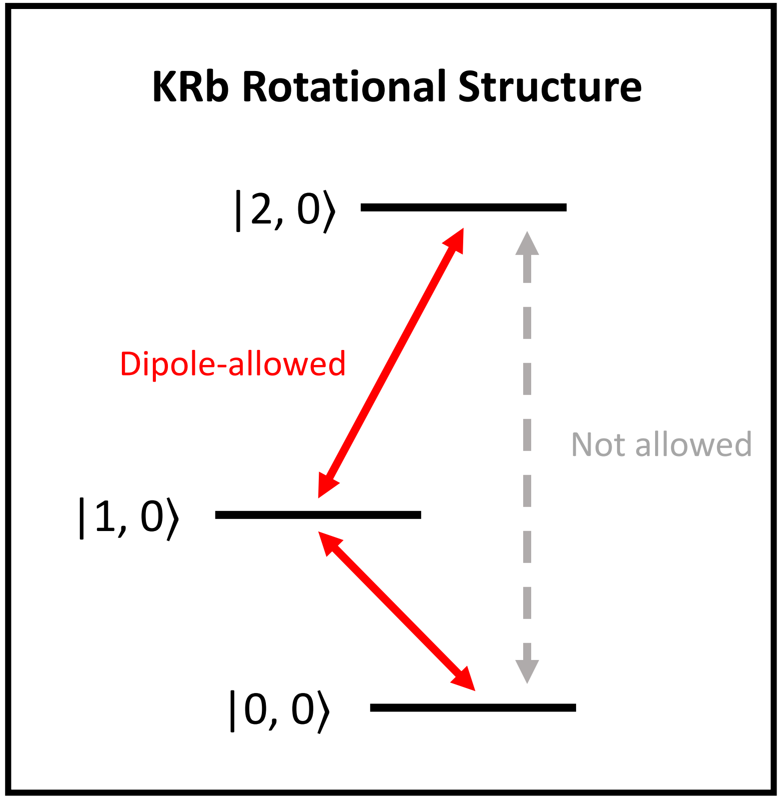




Interlayer spin exchange

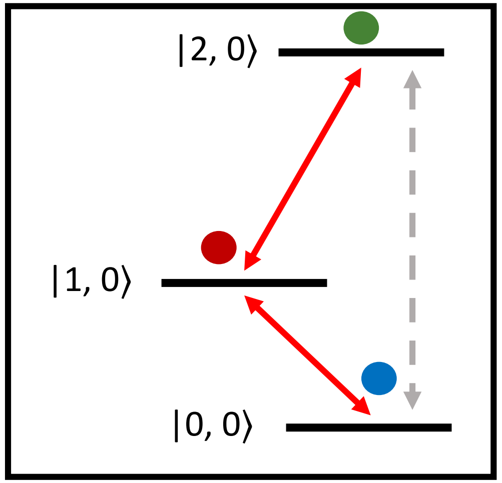
- Preparing 2D layers in selected rotational states $|N, m_N\rangle$
- Spin exchange between layers allowed for $\Delta N = 1$
- Many-body effects may modify the spin exchange rate

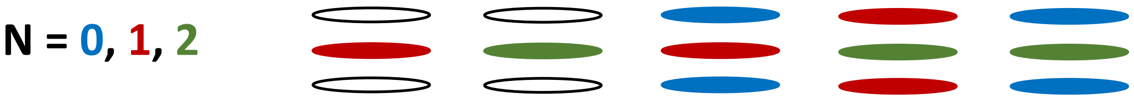




Interlayer spin exchange

- Preparing single layer with different "spin environments" (rotational states of neighboring layers)
- Measuring loss rate of molecules from single layer
- Three conditions
  1. No neighboring layers (slow loss)
  2. Neighbors in ΔN = 1 rotational state (fast loss)
  3. Neighbors in ΔN = 2 rotational state (slow loss)

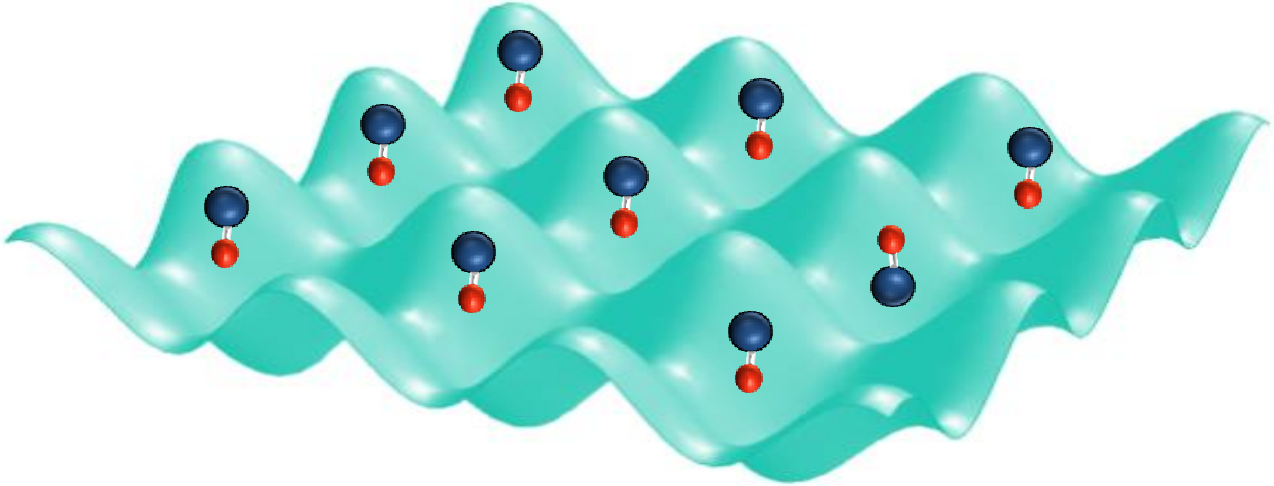




Molecules as dipolar spins

Gorshkov *et al.*, Phys. Rev. Lett. **107**, 115301 (2011).

Molecules are physically pinned down,
but spins are exchanged & mobile

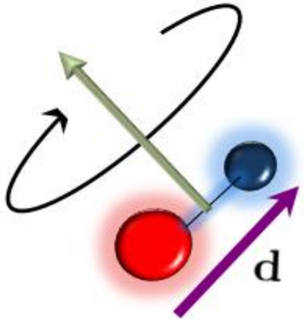


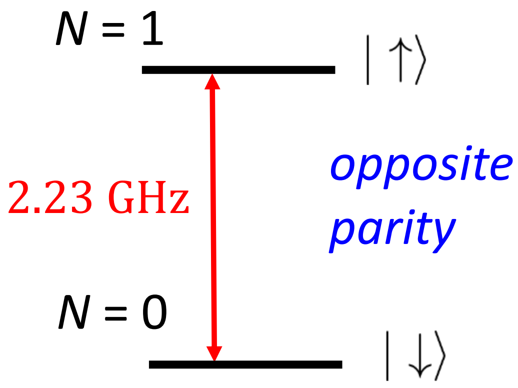
$$H = \sum_{i>j} V_{dd}(\mathbf{r}_i - \mathbf{r}_j) \left[J_z S_i^z S_j^z + \frac{J_\perp}{2} \left(S_i^+ S_j^- + S_i^- S_j^+ \right) \right]$$

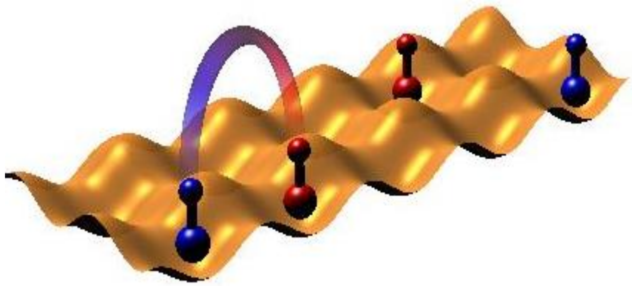
$$\frac{1 - 3\cos^2\theta_{ij}}{|\mathbf{r}_i - \mathbf{r}_j|^3}$$

Ising $J_z \sim (d_\uparrow - d_\downarrow)^2$

Spin exchange $J_\perp \sim d_{\uparrow\downarrow}^2$ ~ 100 Hz

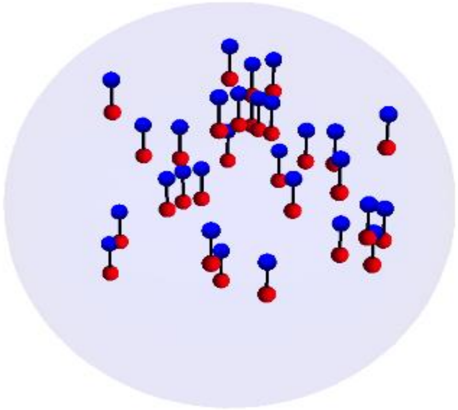


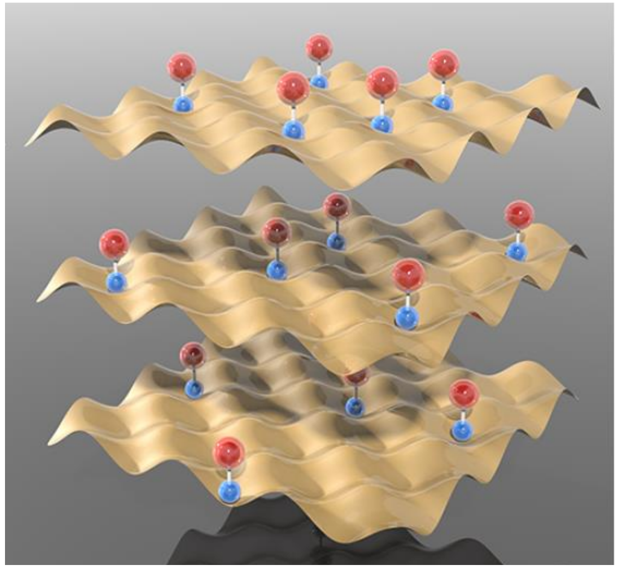


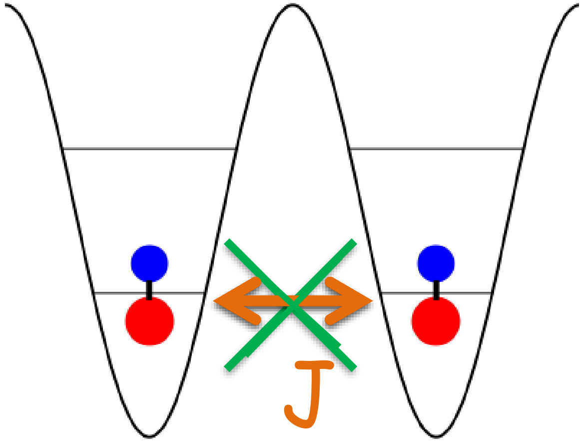


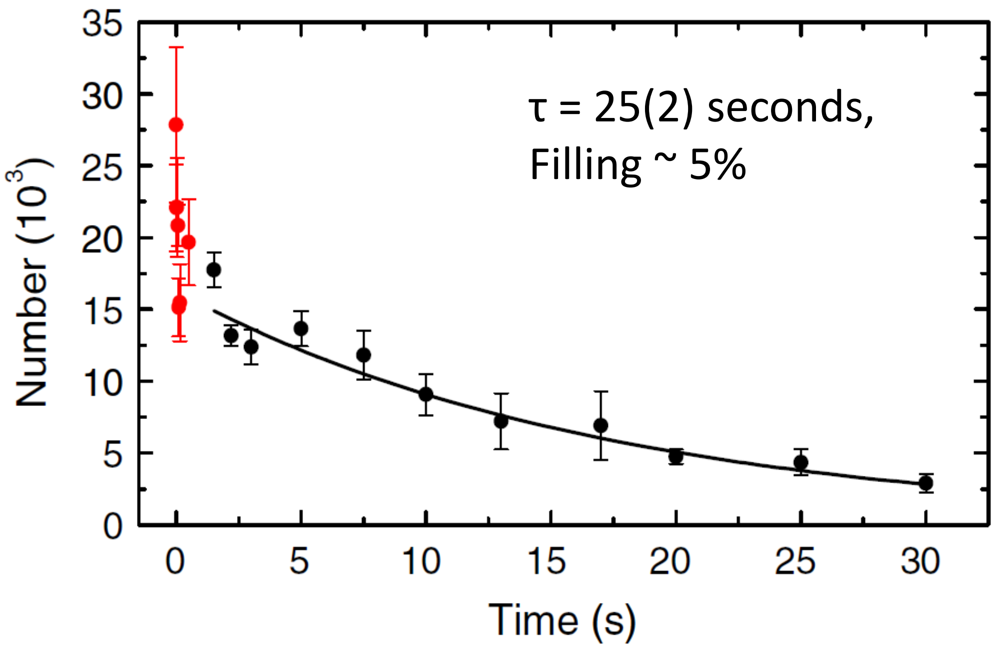
Putting dipoles in a 3D lattice

Chotia *et al.*, PRL **108**, 080405 (2012); Zhu *et al.*, PRL **112**, 070404 (2014).







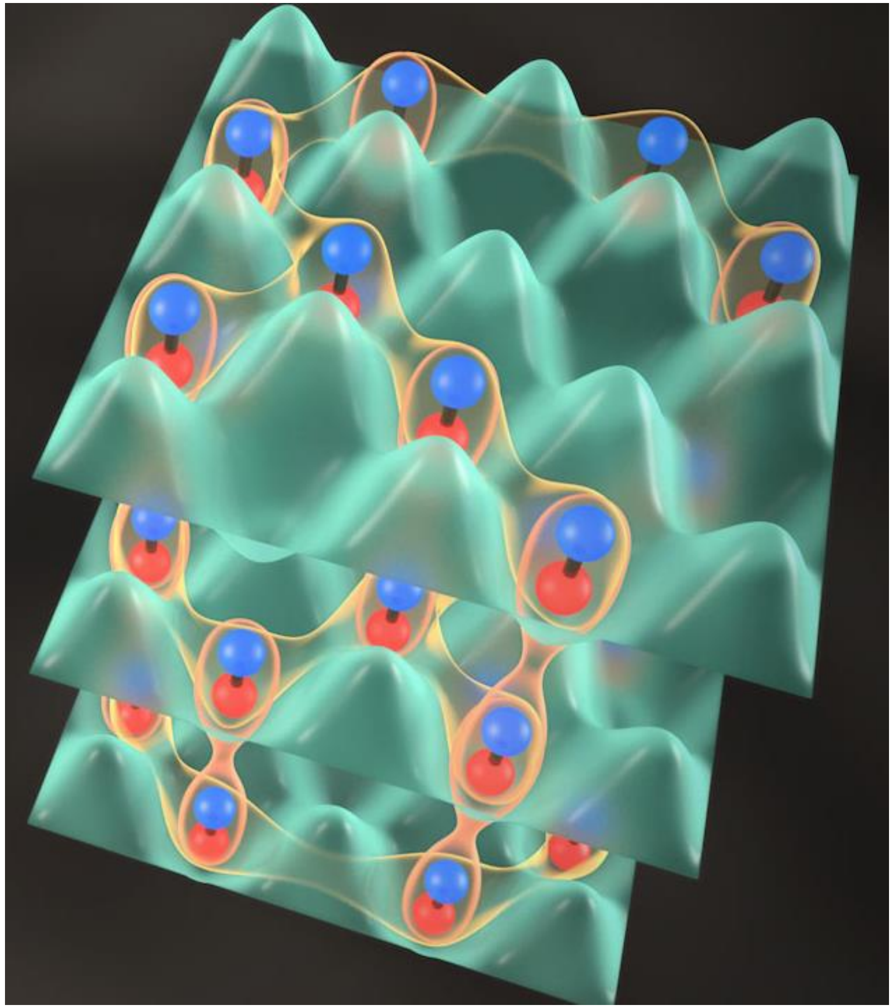


Pauli exclusion
Interaction blockade

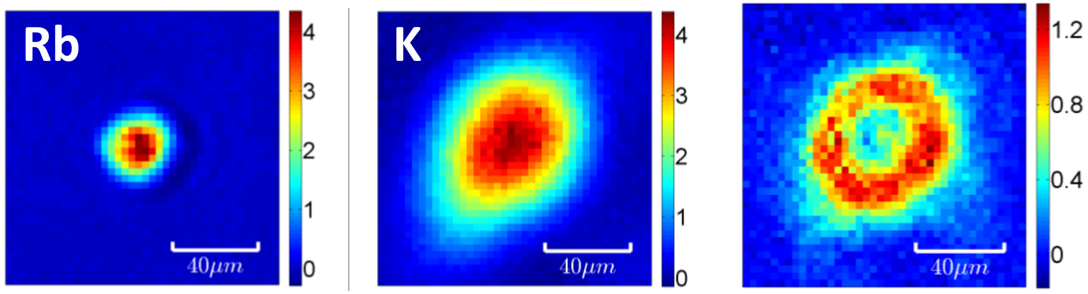
Synthesis of molecular matter

Moses *et al.*, Science **350**, 659 (2015).

KRb: 1.5 x 10^3, ~25% filling



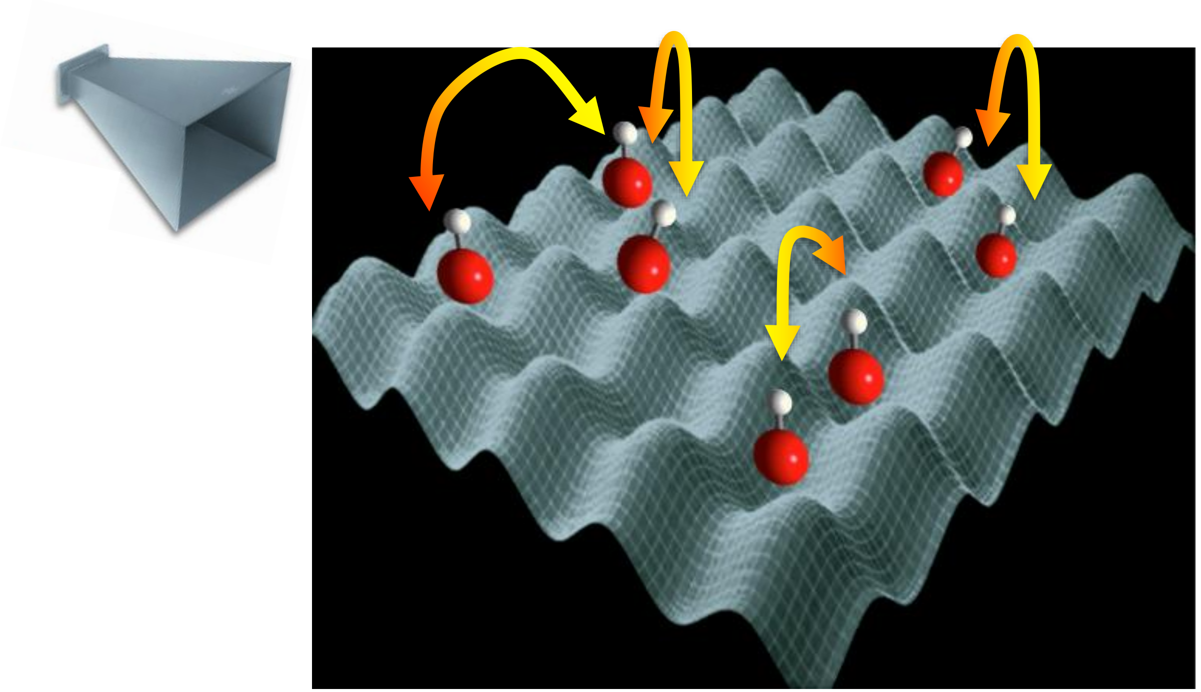
Dual Mott (Rb) / band (K) insulator

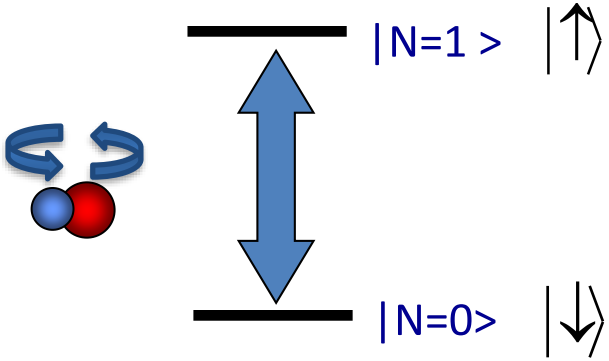


Sites with one atom of each species

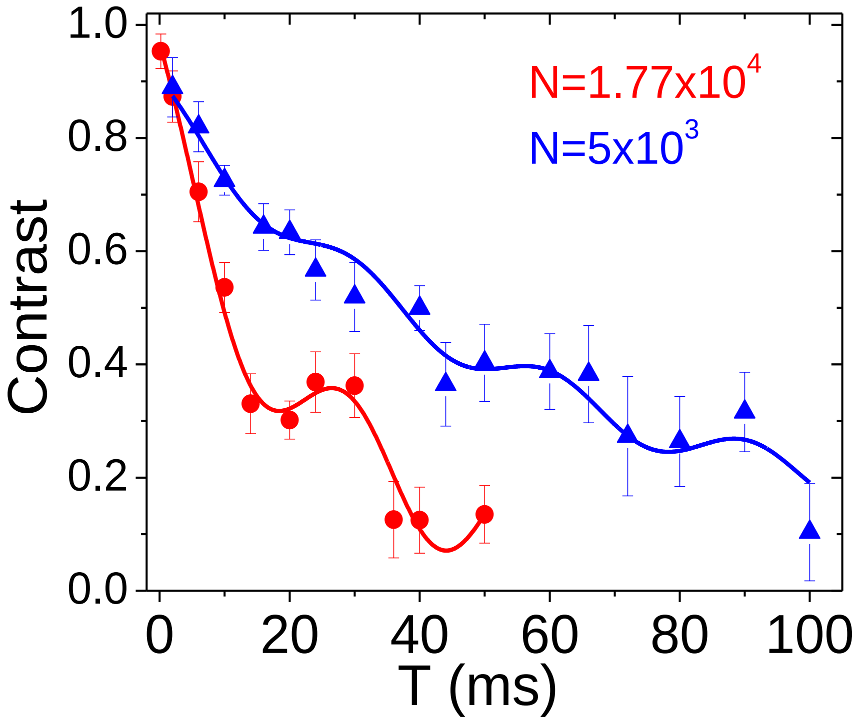
A Dipolar Spin Lattice

Yan *et al.*, Nature **501**, 521 (2013); Hazzard *et al.*, Phys. Rev. Lett. **113**, 195302 (2014).



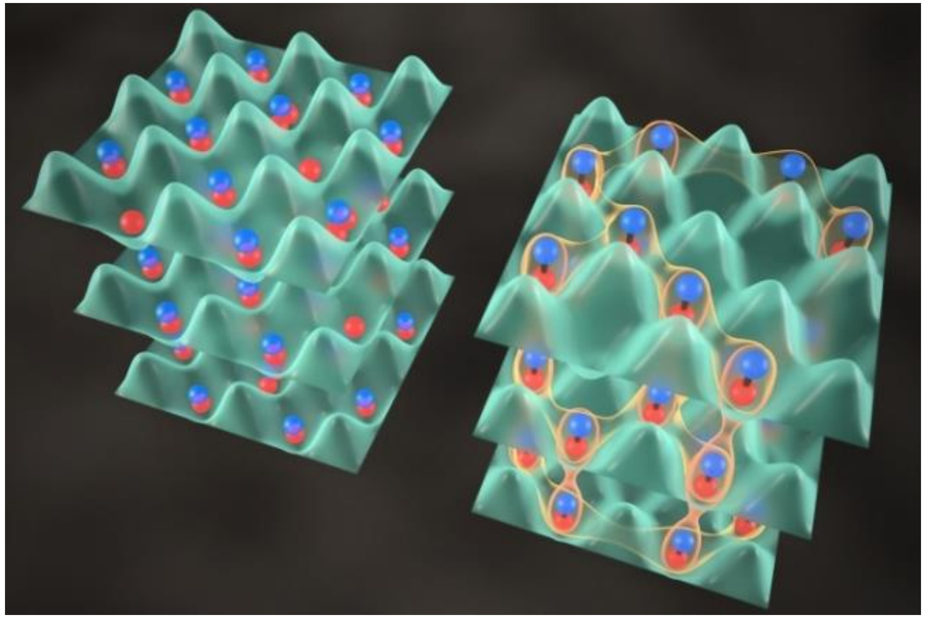


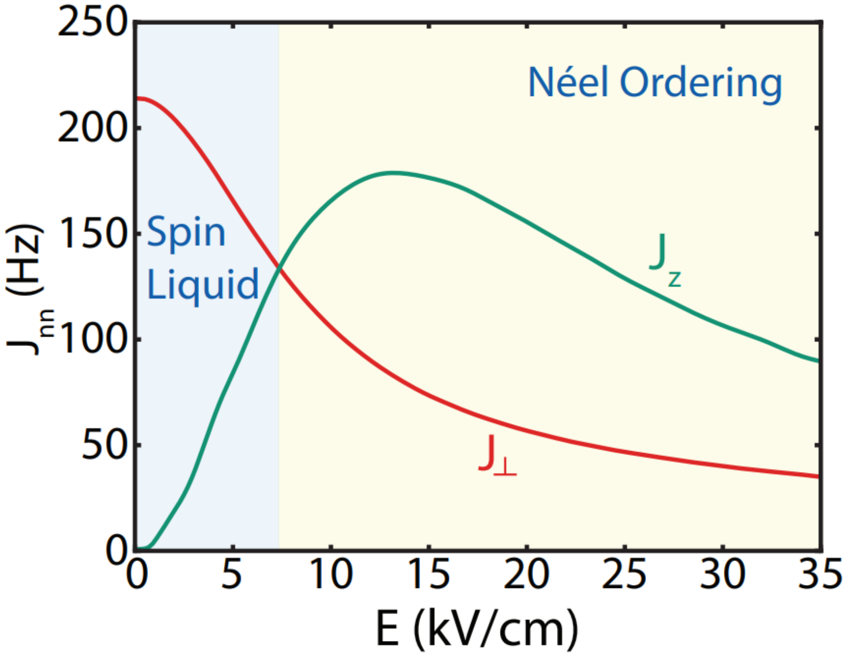
- Start with N=0. $|\downarrow\rangle$
- Drive a coherent spin superposition. $\frac{1}{\sqrt{2}}\left(|\uparrow\rangle+|\downarrow\rangle\right)$
- Probe spin coherence at T. (Ramsey spectroscopy)



A Dipolar Spin Lattice

$$H = \frac{1}{2}\sum_{i\neq j} V_{dd}(i,j)\left[J_z S_i^z S_j^z + \frac{J_\perp}{2}(S_i^+ S_j^- + \text{H.c.})\right]$$

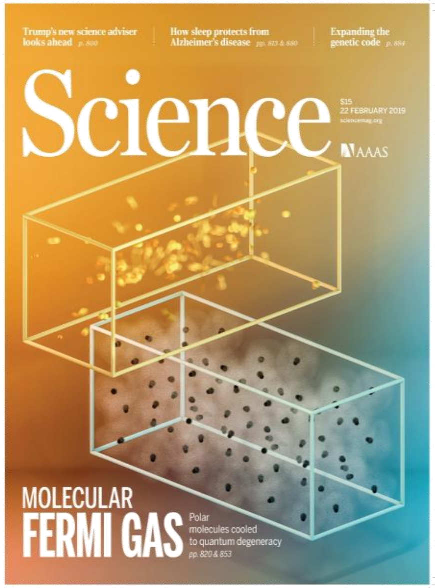


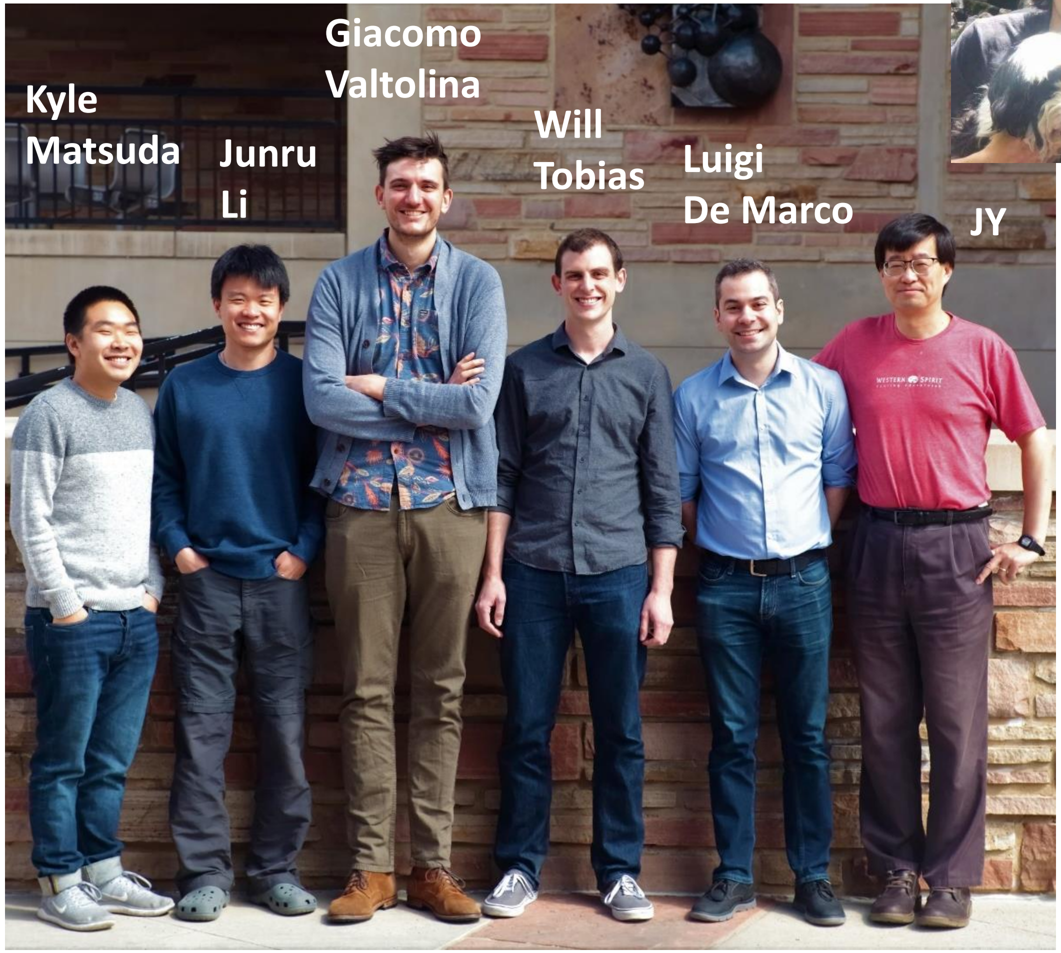


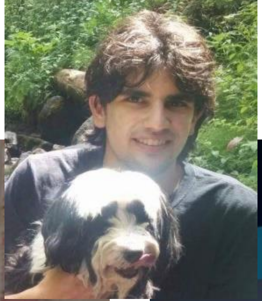
- Exploring phase transition ($J_\perp/J_z$)
- Transport and localization
- Exotic lattice geometries (long-range interactions + frustration)

- Spin-orbit coupling
- Pair production using shielding resonance ($|1,0\rangle|1,0\rangle \rightarrow |0,0\rangle|2,0\rangle$)
- Microwave dressing & Floquet dynamics

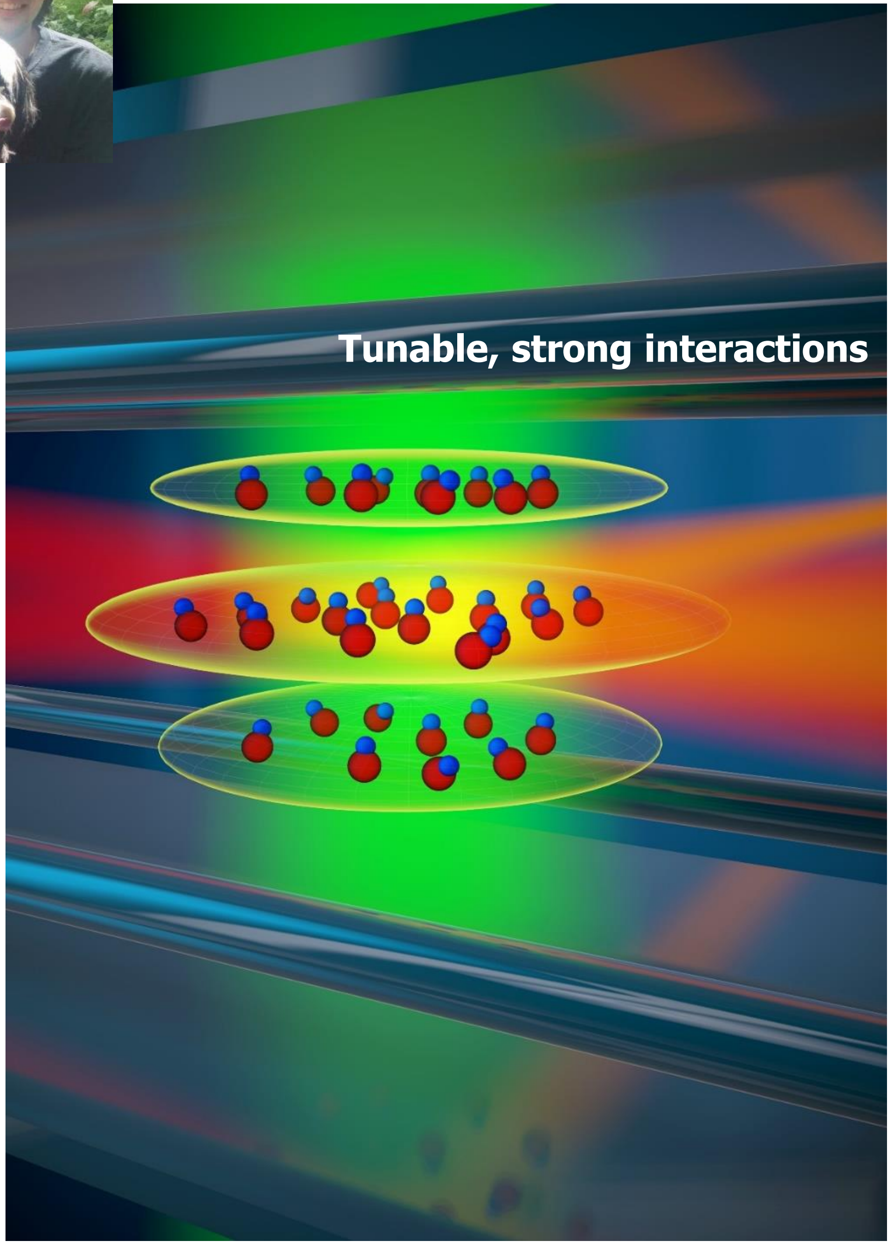
Future plans







Calder Miller



<u>Theory Collaborations</u>

John Bohn, Goulven Quéméner (Dipolar collisions)
Ana Maria Rey (Spin squeezing, lattice spin model)
Eugene Demler (Dipole-mediated hydrodynamics)
Svetlana Kotochigova (Coherence)
Norm Yao, Baranov/Peter Zoller (Many-body, exotic quantum phenomena)