

# Ultracold molecules - a new frontier for quantum & chemical physics

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JILA, NIST & CU, Boulder

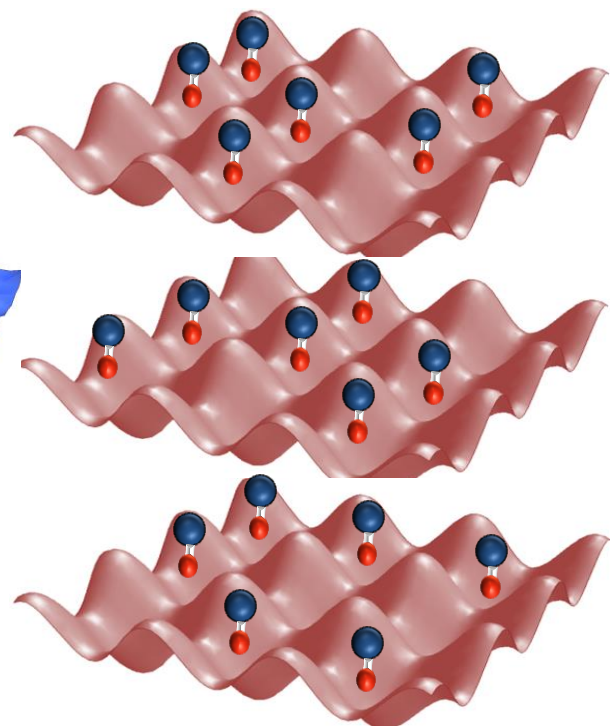
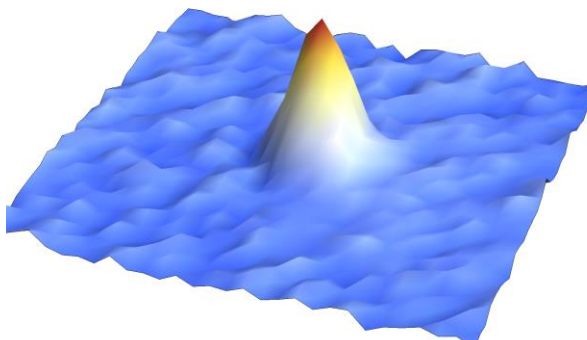
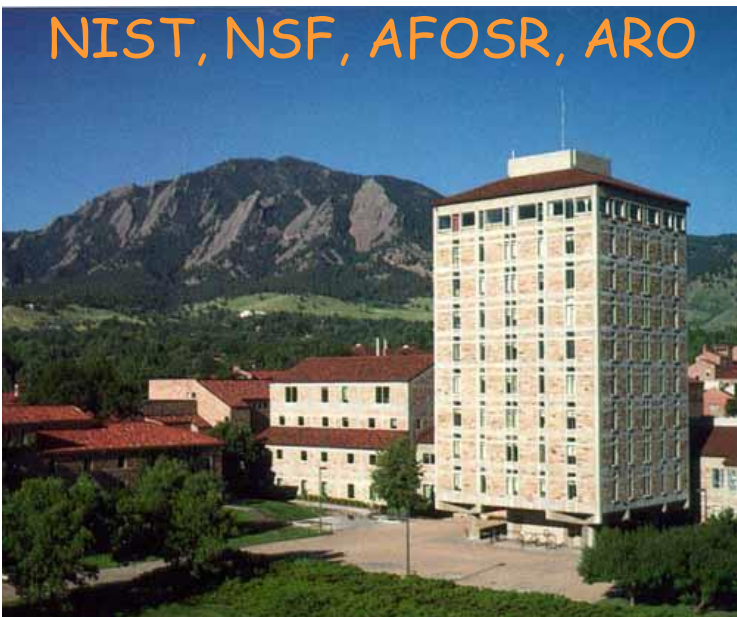
University of Virginia

April 24, 2015

Debbie Jin



NIST, NSF, AFOSR, ARO



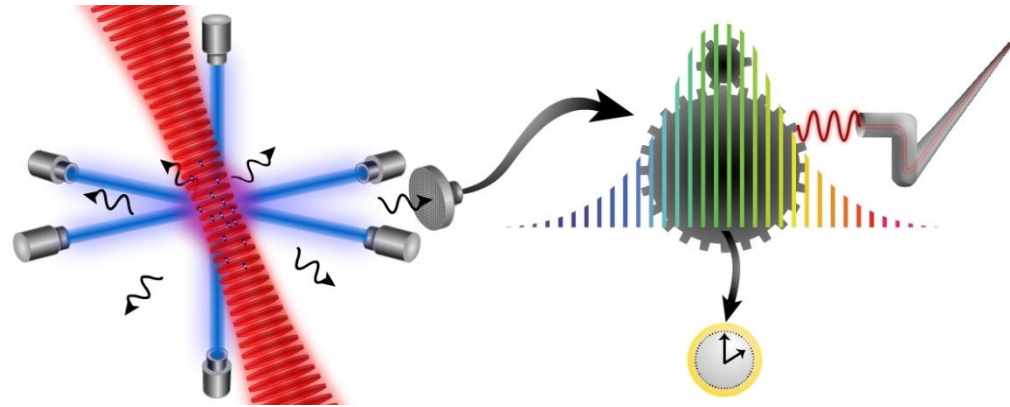
# Ultracold atomic matter

## Precise control of a quantum system

The most precise measurements, e.g., clock

Quantum information

Quantum sensors

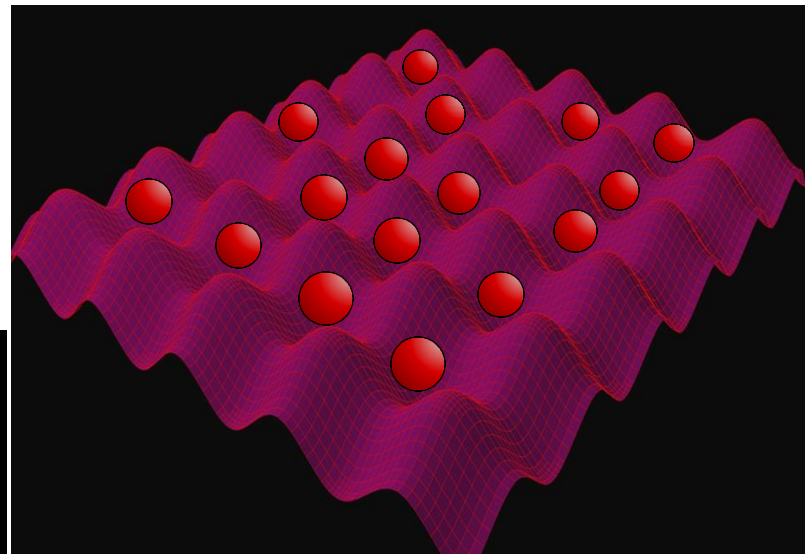


## Control: A tool for understanding complexity

Strongly correlated many-body quantum systems

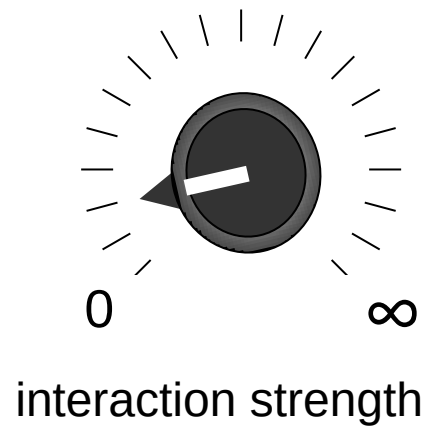
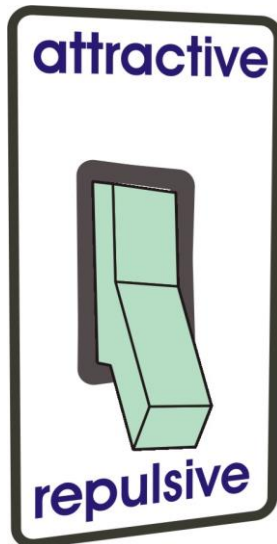
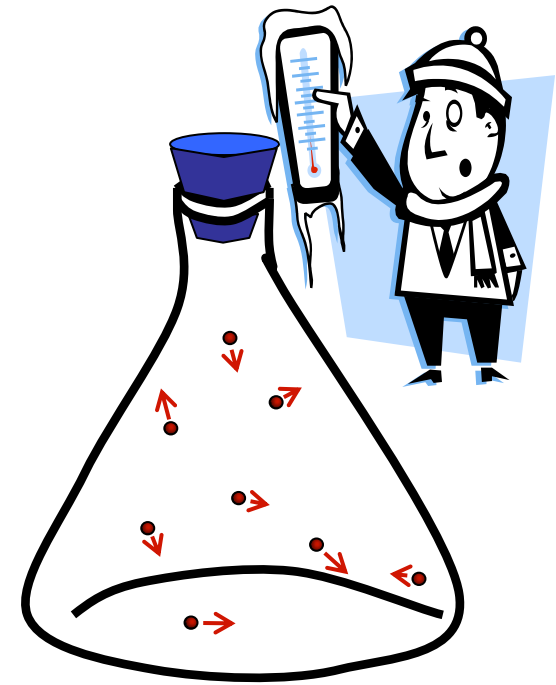
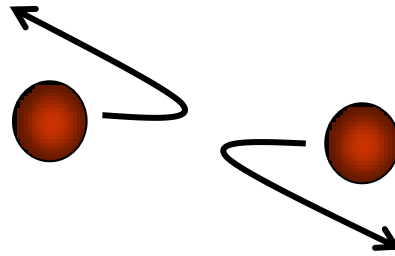
- Superfluidity & Superconductivity

**Control atomic interactions**  
**Regulate atomic motions**



# Atomic Interactions

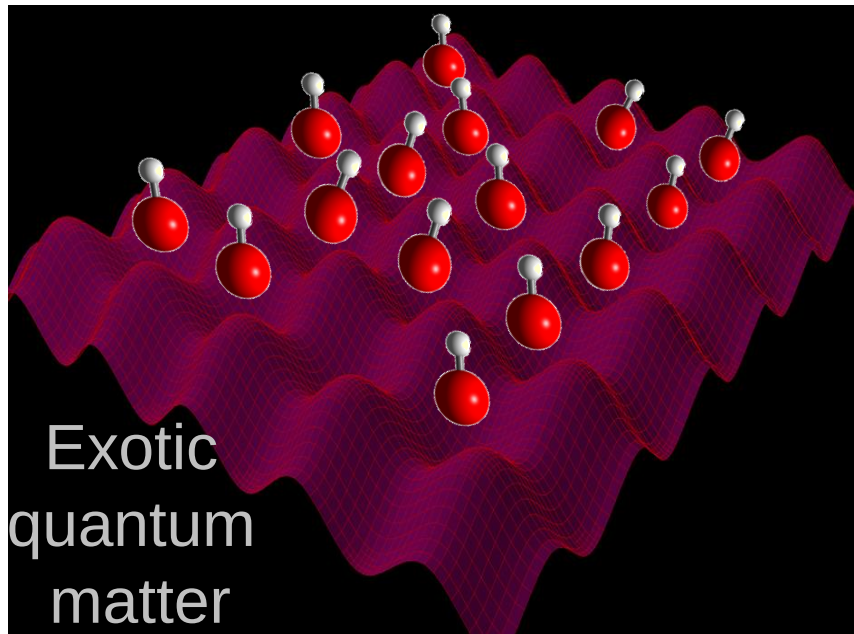
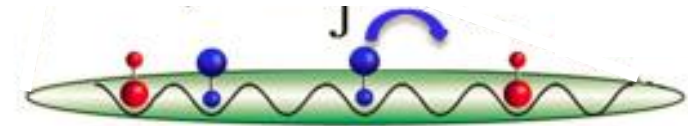
Short-range collisions



We can understand  
& control them!

# Quantum gas of polar molecules

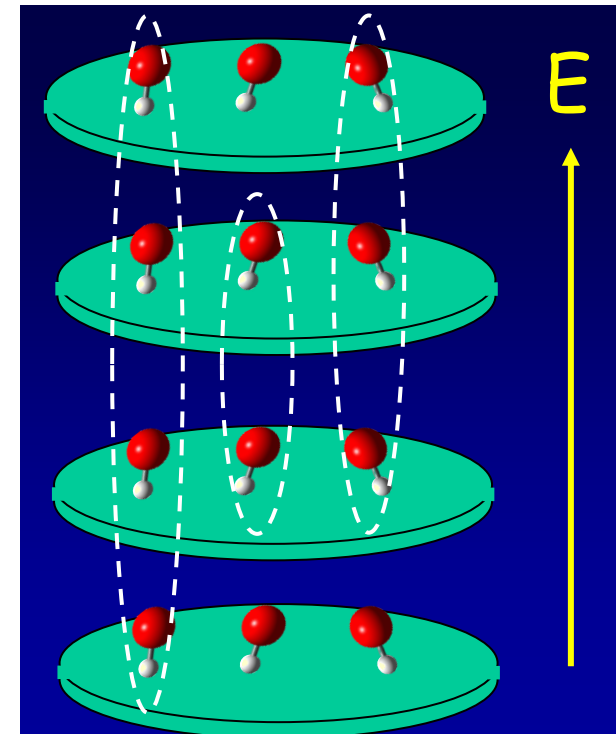
- Extend capability to control complex quantum systems
- Study frontier problems in strongly correlated quantum material, with
  - well-understood microscopics
  - tunable, long-range interactions
  - non-equilibrium quantum dynamics



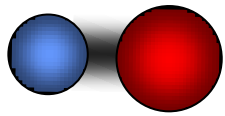
Quantum  
metrology,

Correlated  
material,

Chemistry



# Ultracold molecules: The challenge



KRb

energy ↑

$$E/k_B = 6000 \text{ K}$$

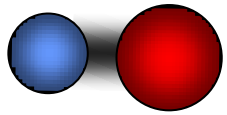


two atoms

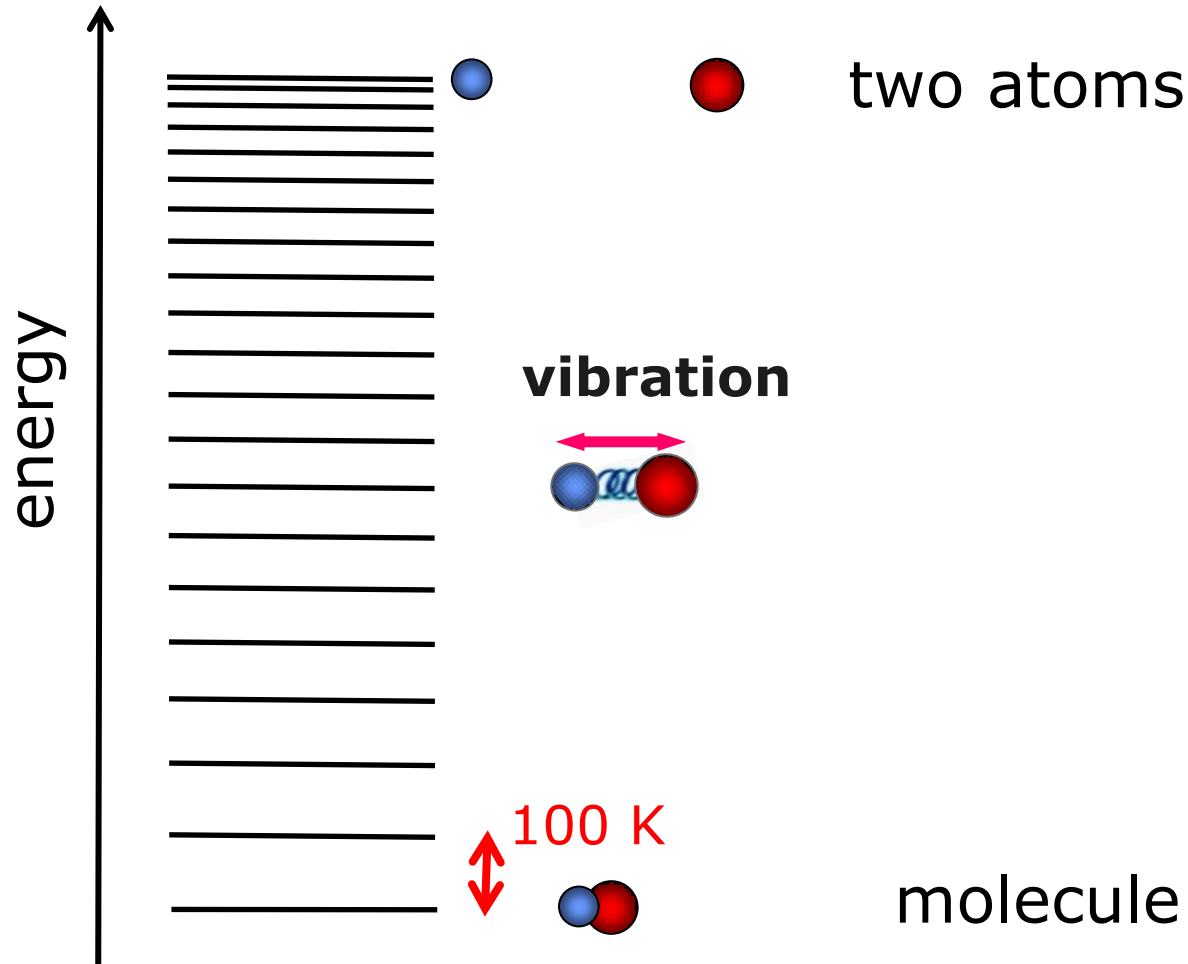


molecule

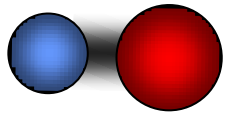
# Ultracold molecules: The challenge



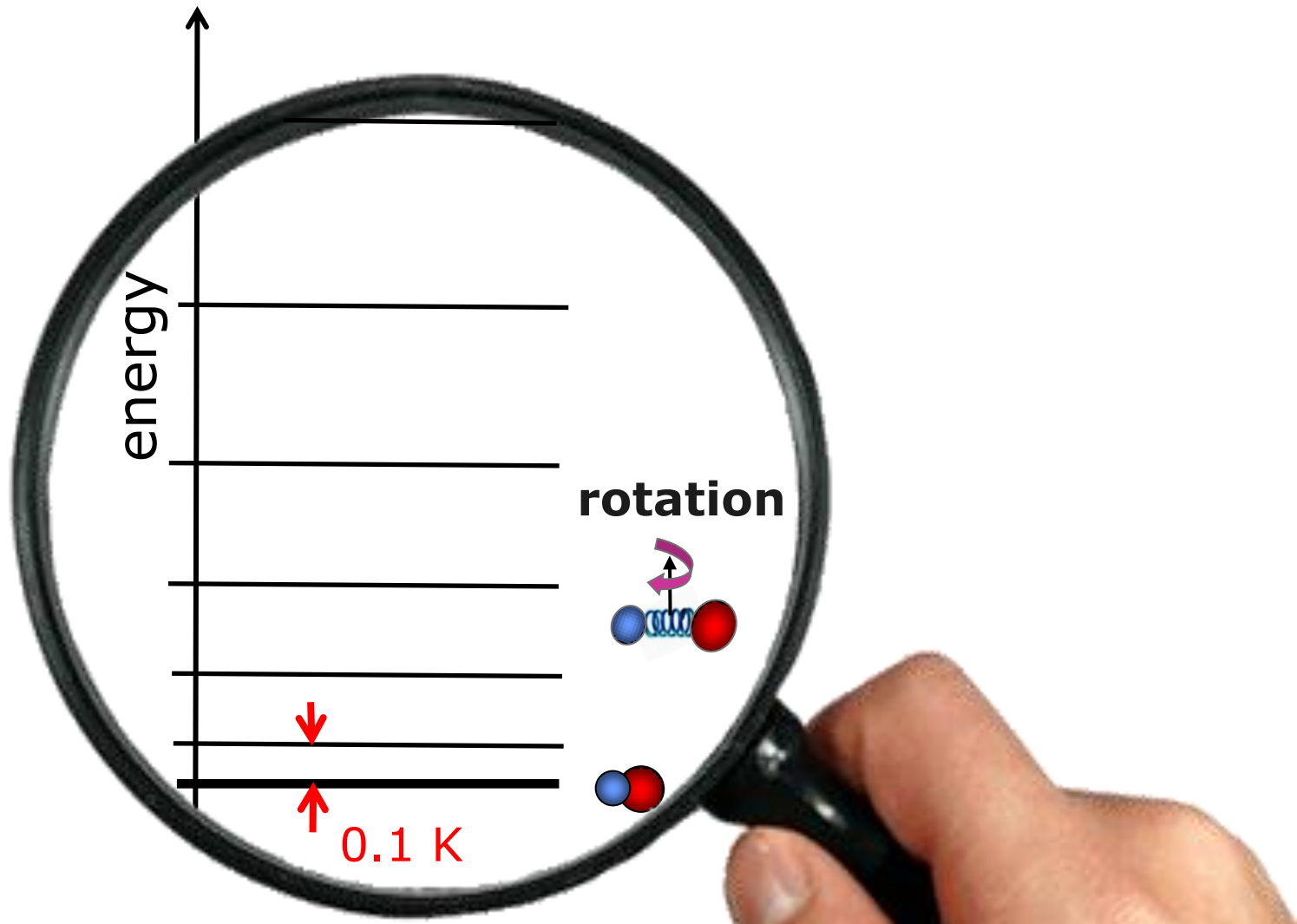
Molecules are complex!



# Ultracold molecules: The challenge

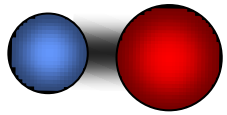


Molecules are complex!

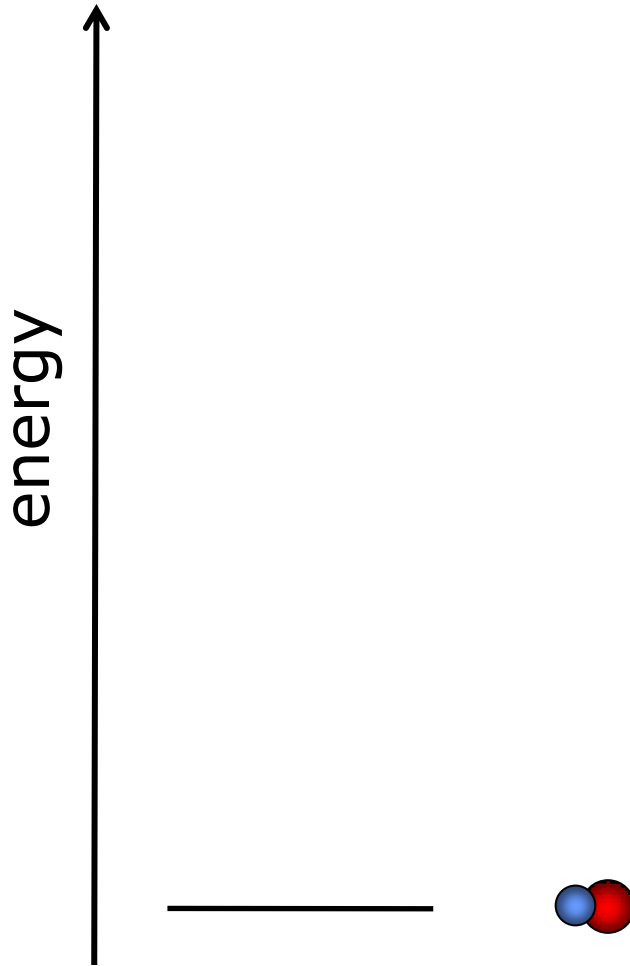




# Ultracold molecules: The challenge

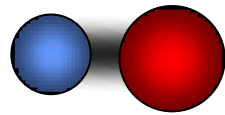


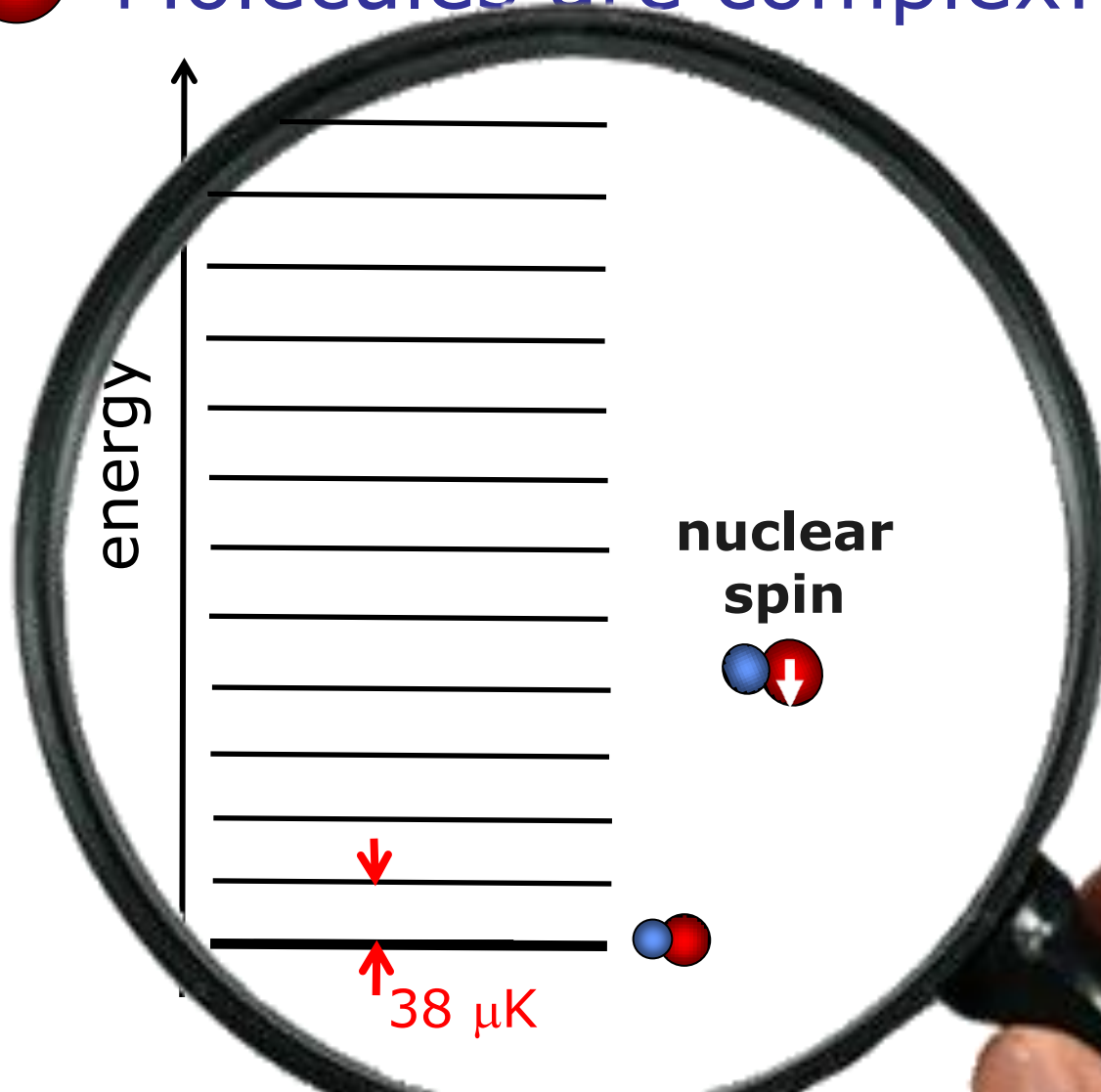
Molecules are complex!





# Ultracold molecules: The challenge

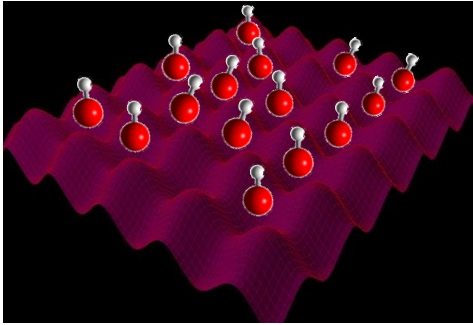
 Molecules are complex!



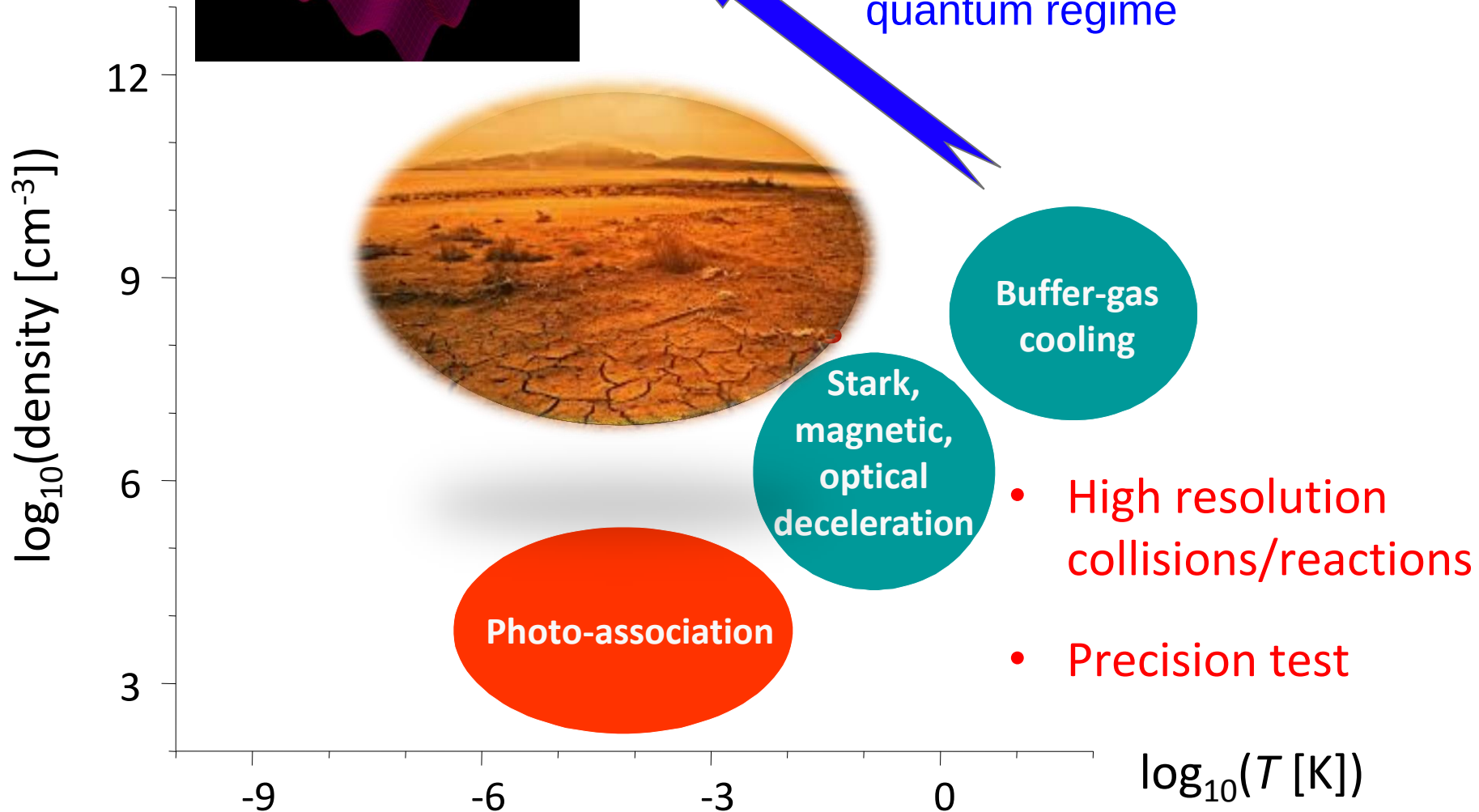
# Technology for making cold molecules

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

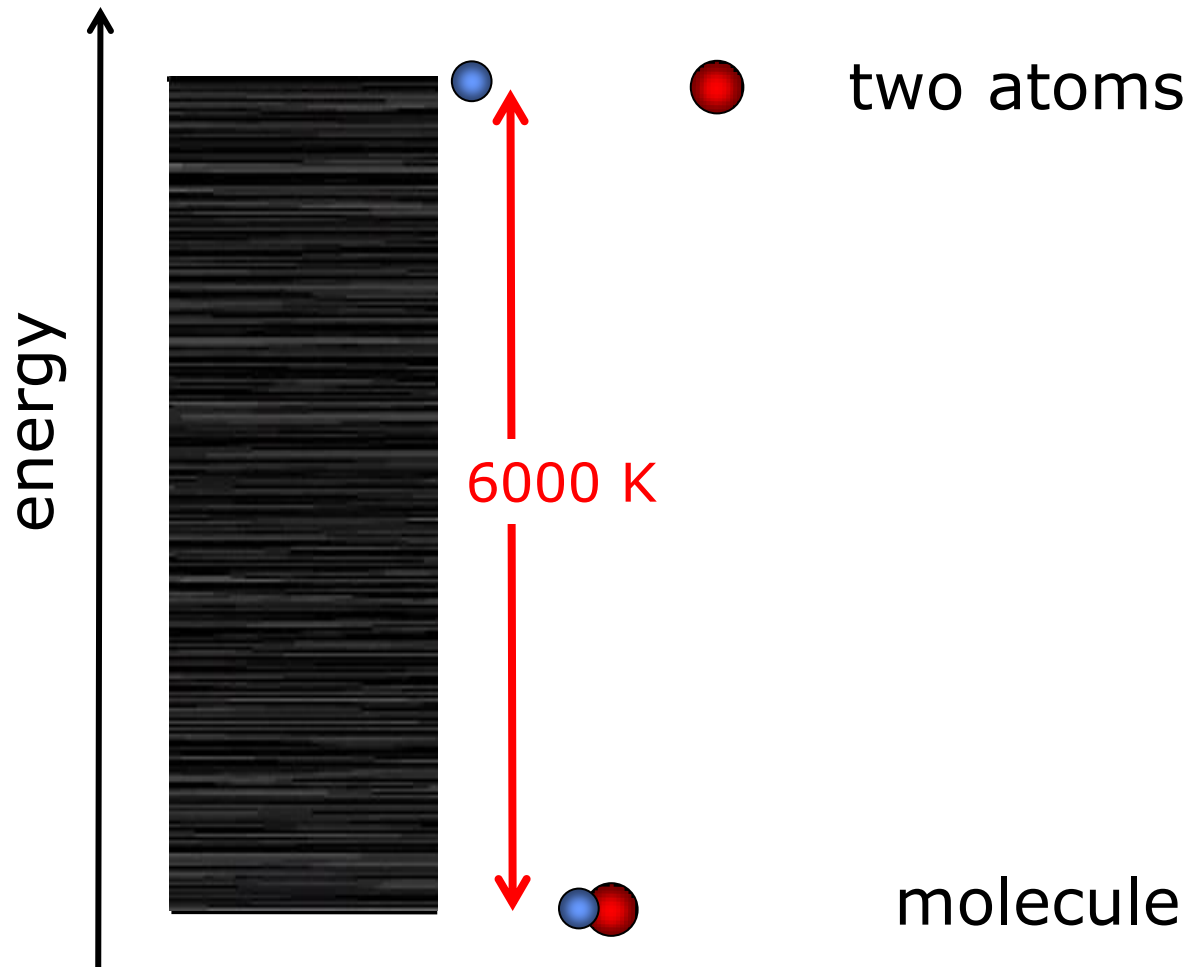
Quantum gas  
of molecules



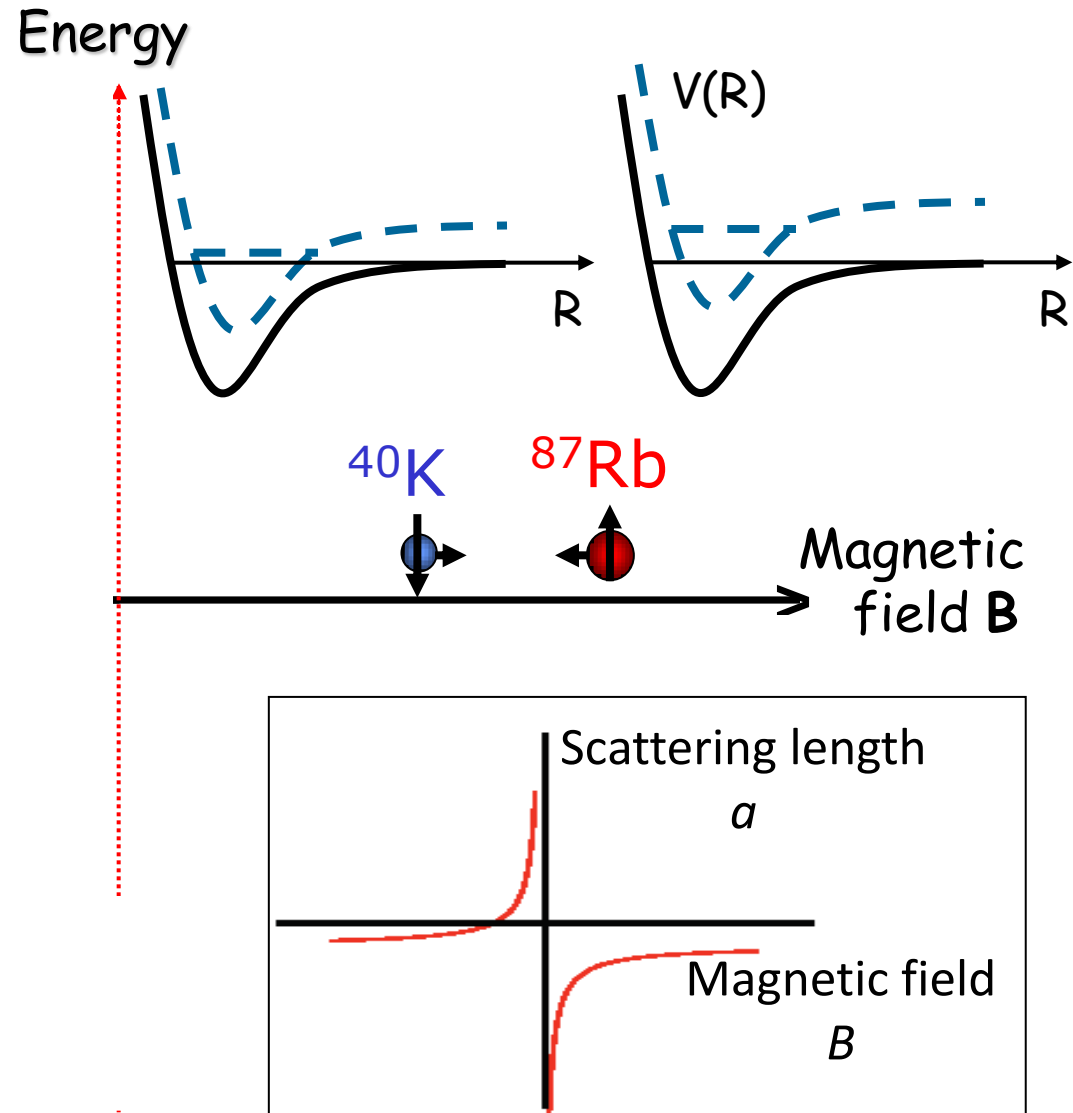
Towards  
quantum regime



# Associate ultracold atoms into molecules



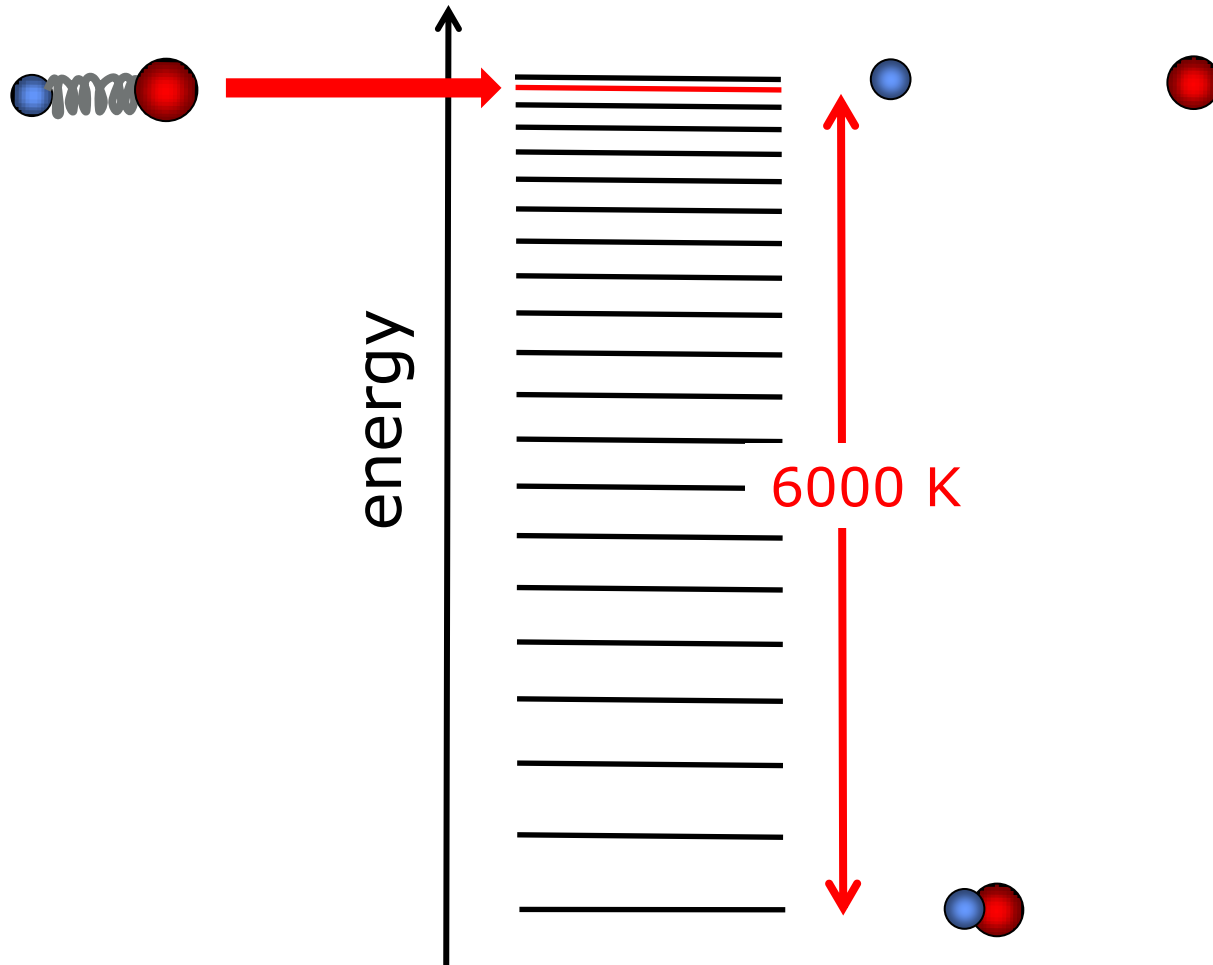
# Make Feshbach molecules



Interaction tuned by scattering resonance

# Weakly bound molecules

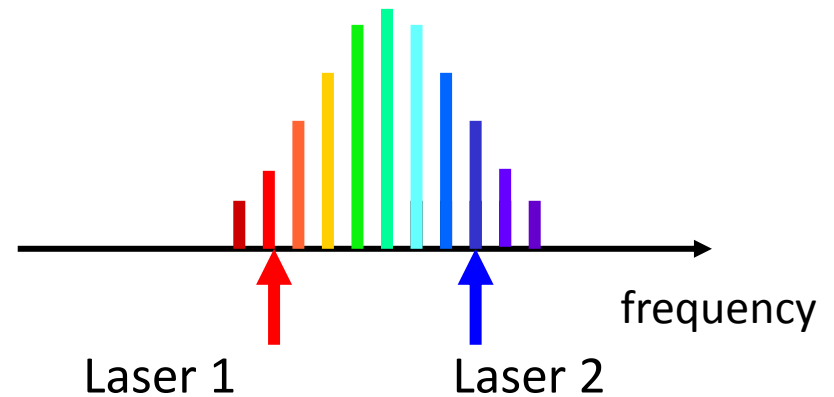
- no dipole moment
- losses



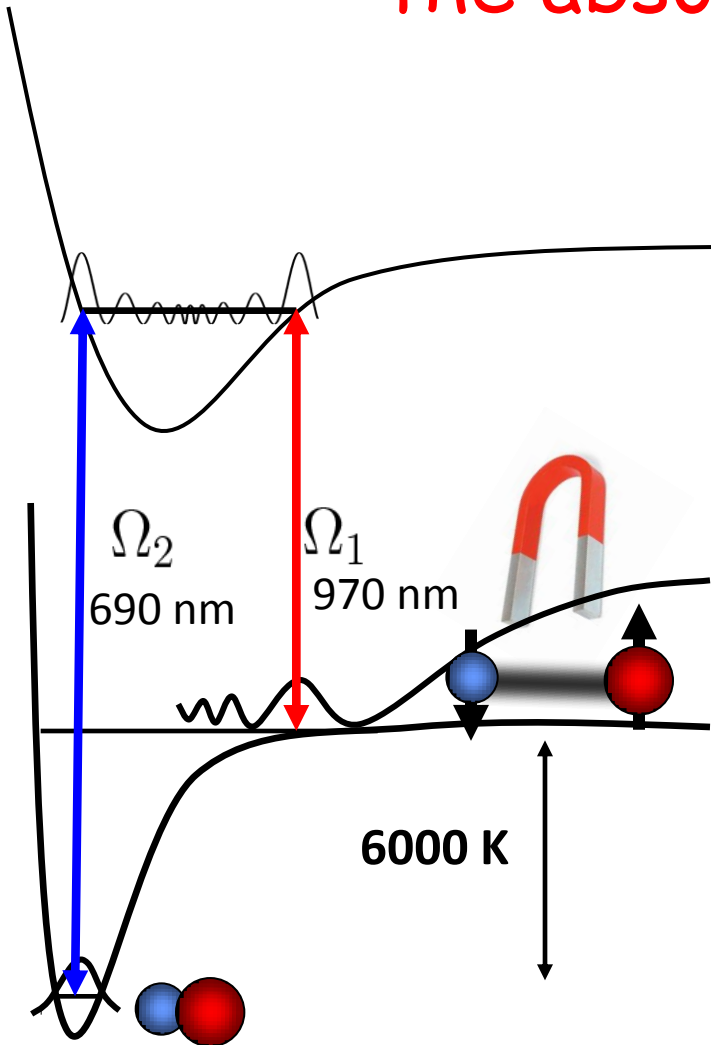
# Coherent two-photon transfer - the absolute ground state

(entropy-less chemistry)

Fully coherent, >90% efficiency



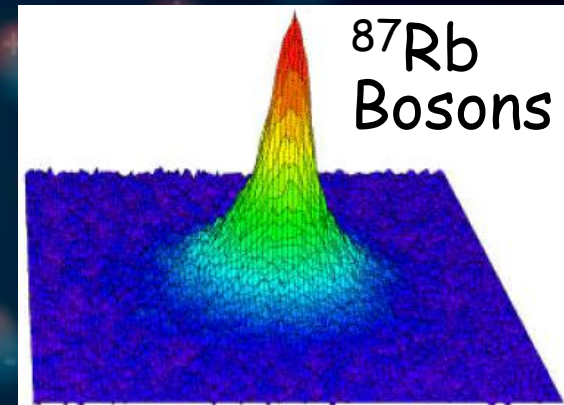
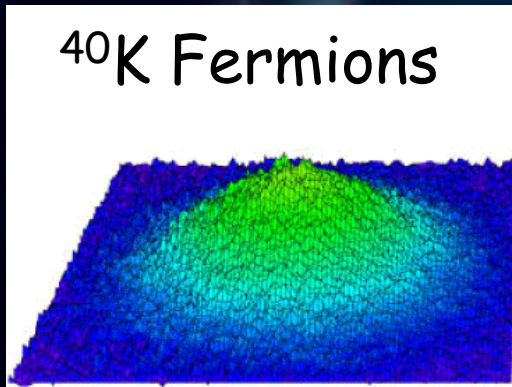
Beat note 125 THz



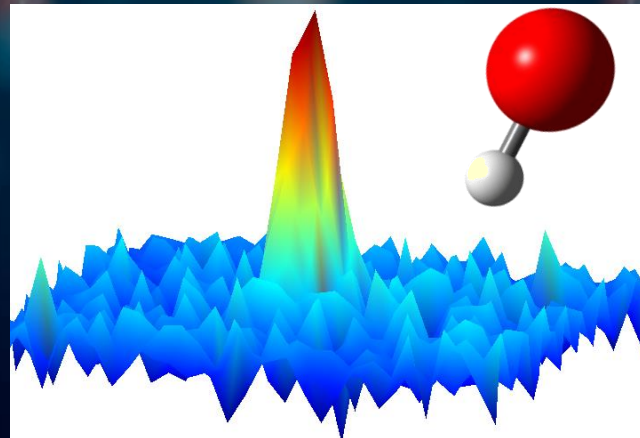
36 nuclear spin states: We populate & control single state  
S. Ospelkaus *et al.*, Phys. Rev. Lett. **104**, 030402 (2010).

# Polar molecules in the quantum regime

- Temperature  $\sim 100$  nK
- Density  $\sim 10^{12}/\text{cm}^3$
- $T/T_F \sim 1.3$



K.-K. Ni *et al.*,  
Science 322, 231 (2008).

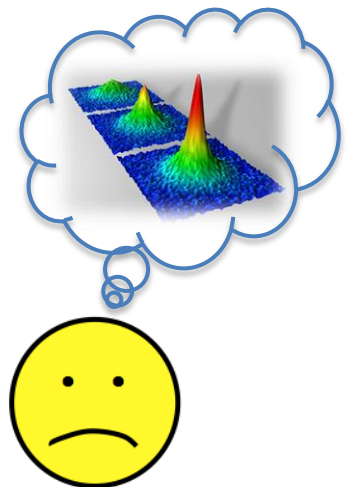
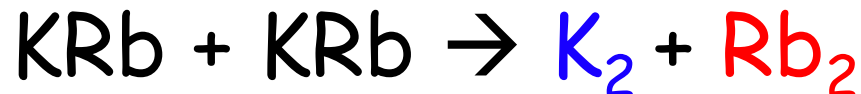
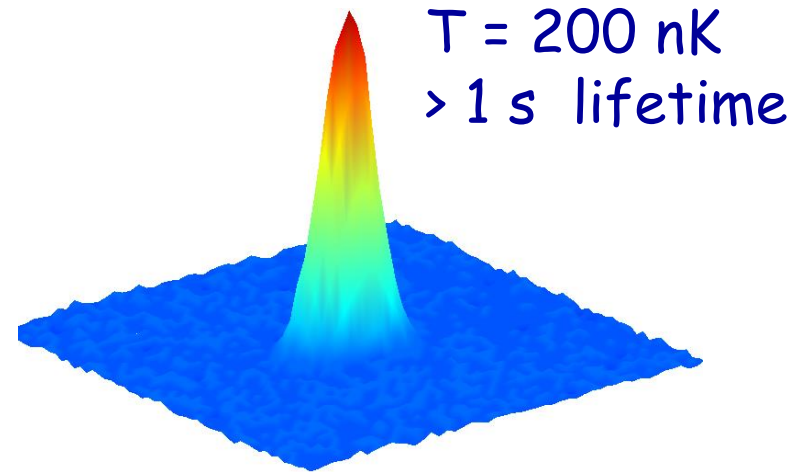
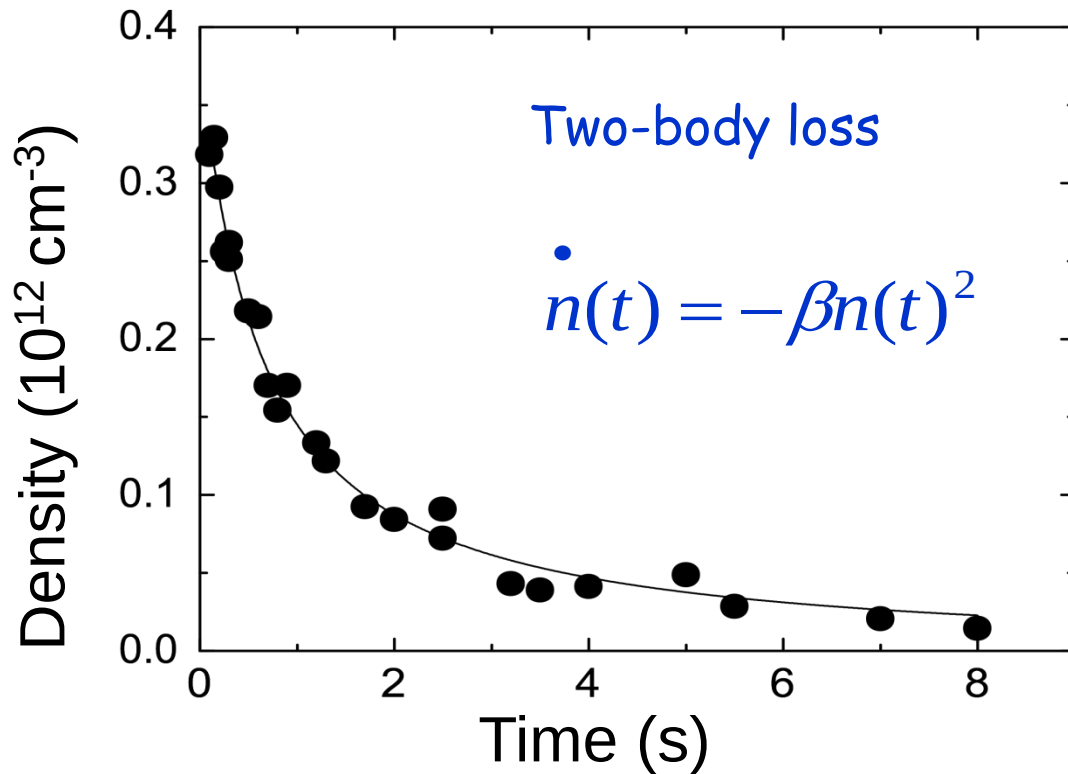


KRb molecules  
(Dipole  
 $\sim 0.5$  Debye)



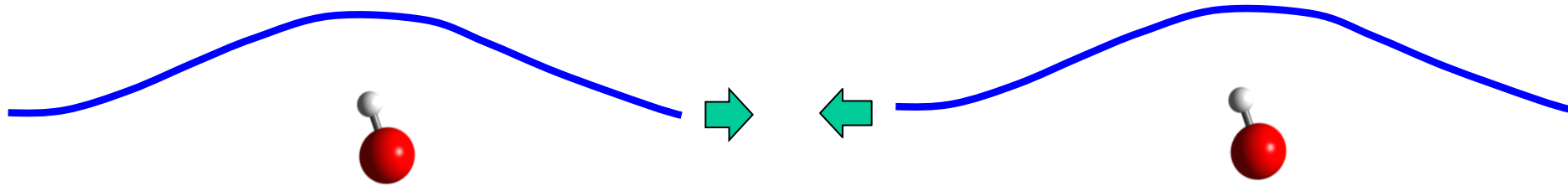
# Chemistry near absolute zero

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

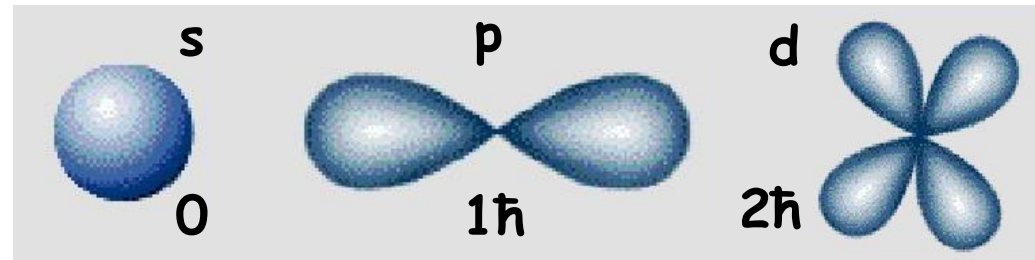
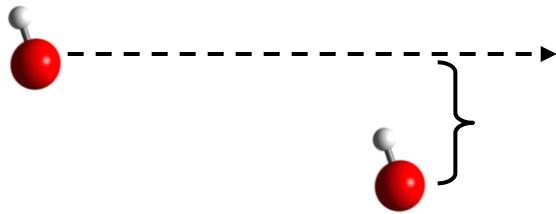


# Cold collisions between identical Fermions

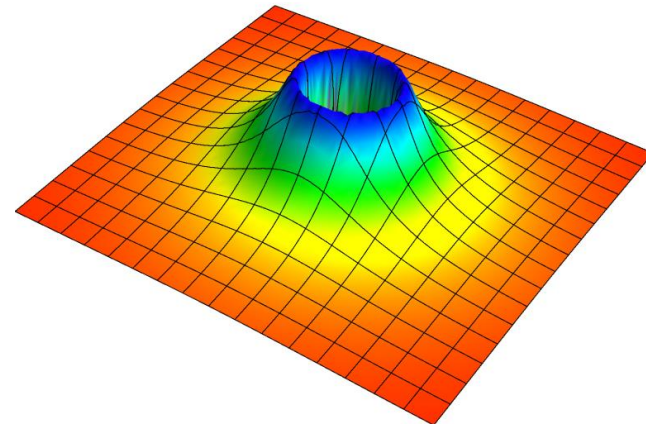
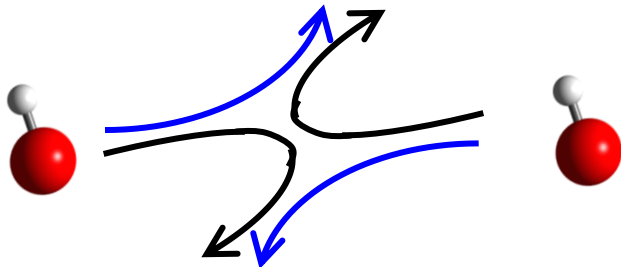
(1) Particles behave like waves



(2) Angular momentum is quantized



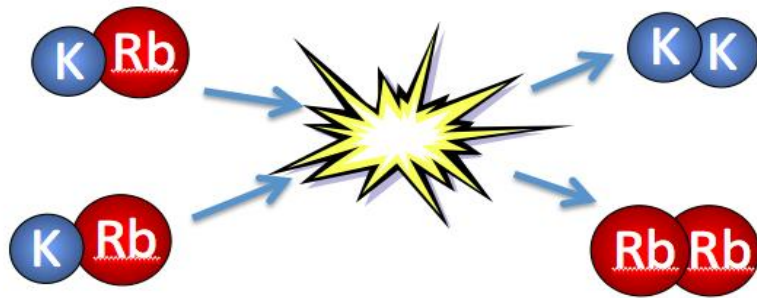
(3) Quantum statistics matter



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

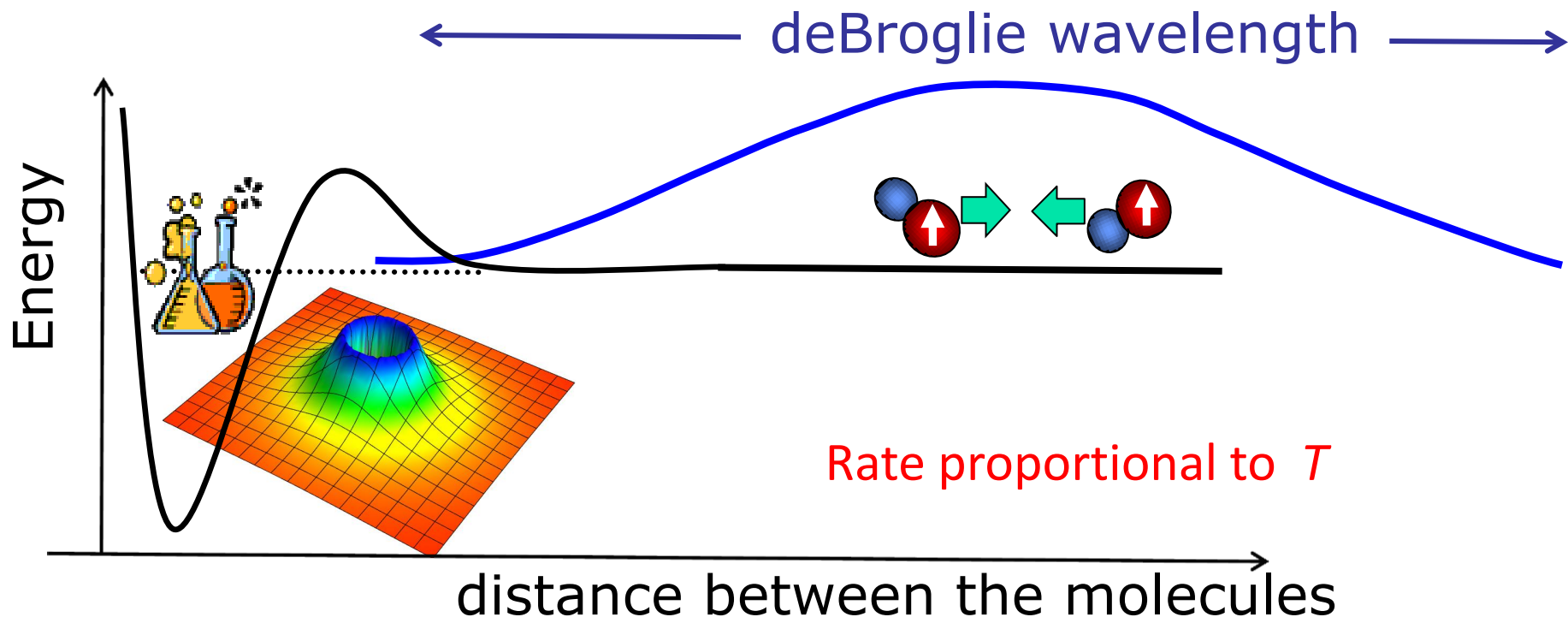
Fermions  $\Rightarrow L = 1$ ,  $p$ -wave collisions

# Ultracold **quantum** chemistry

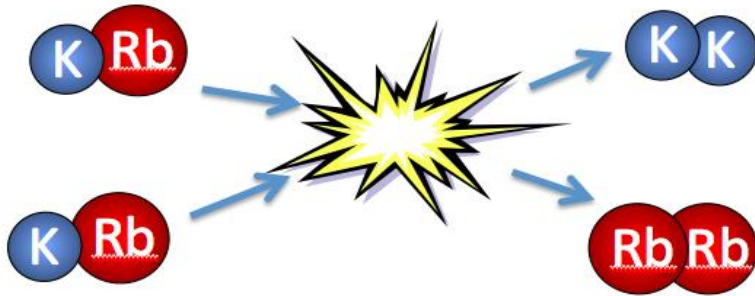


Ospelkaus *et al.*,  
*Science* **327**, 853 (2010).

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

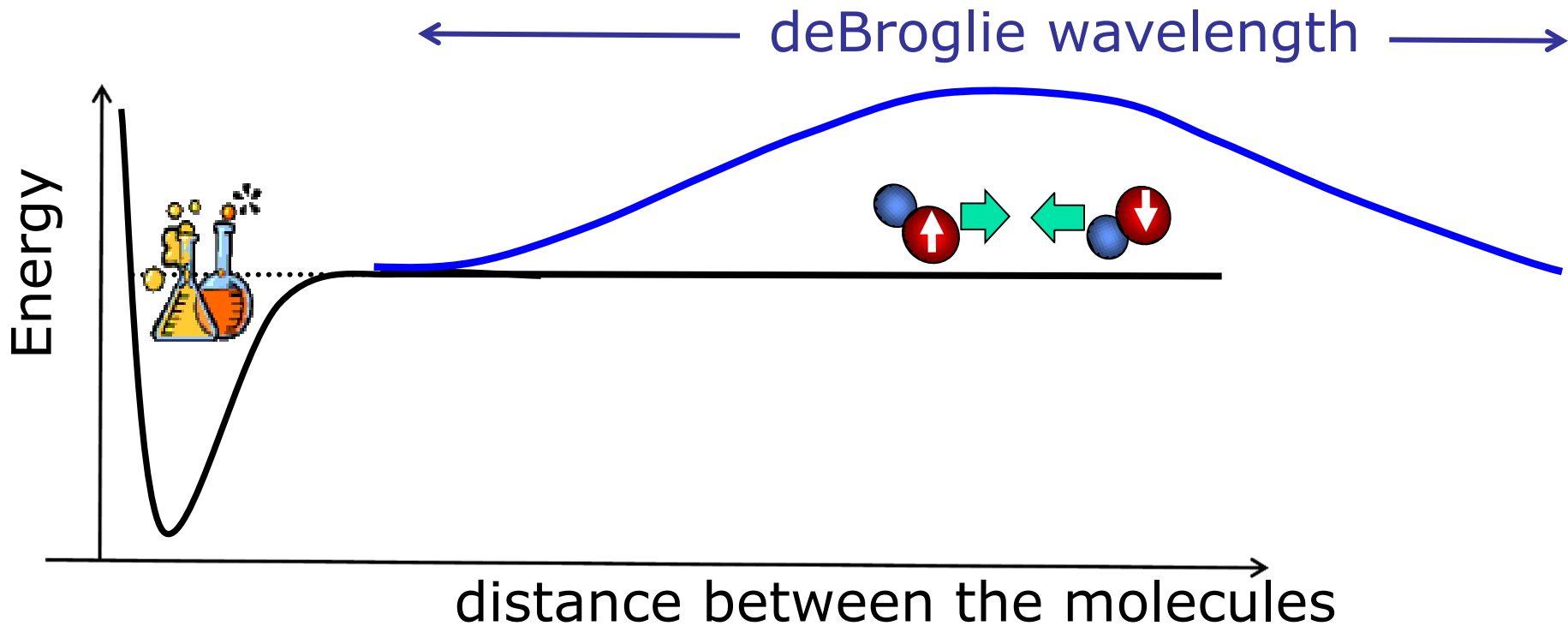


# Ultracold quantum chemistry



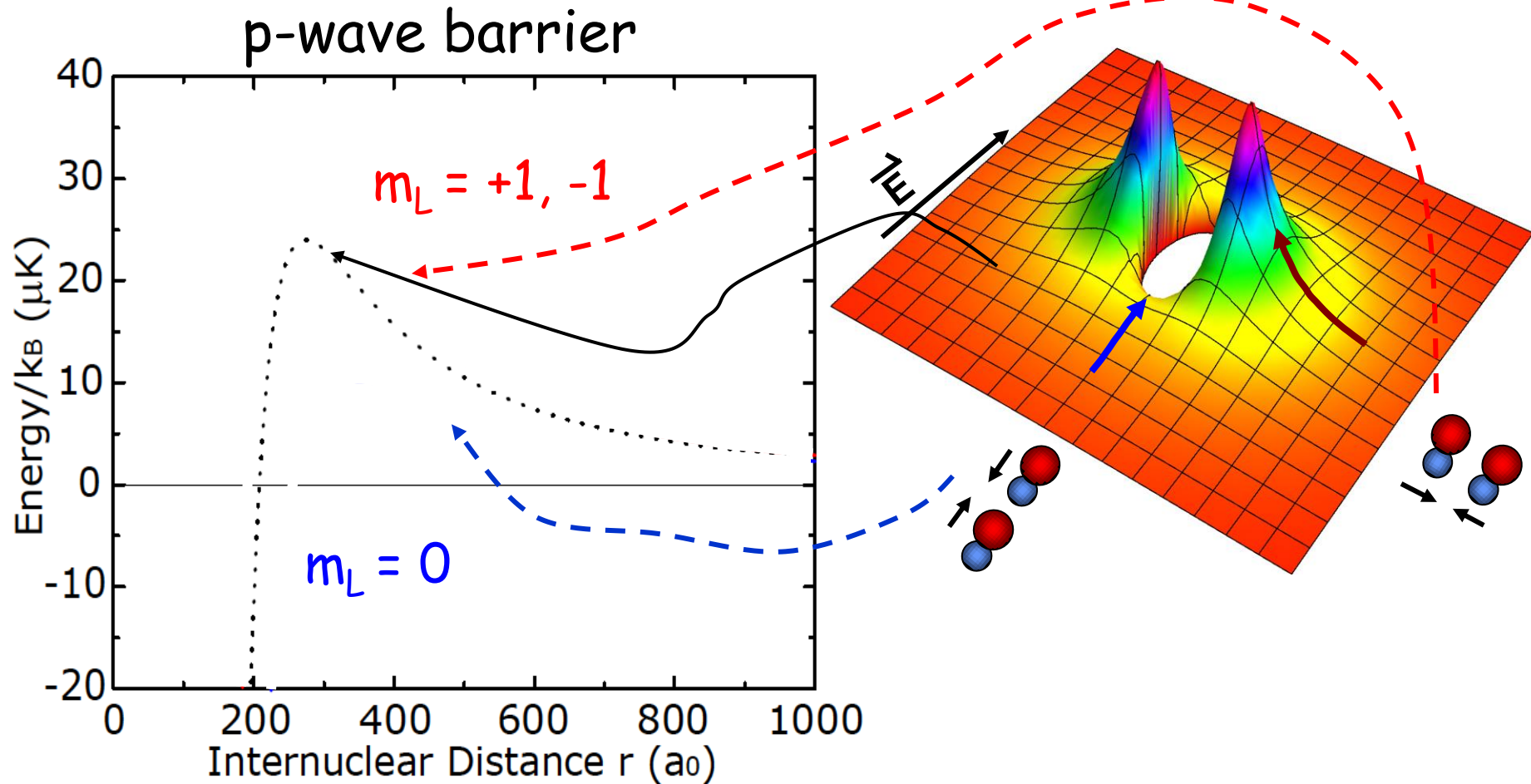
Ospelkaus *et al.*,  
Science **327**, 853 (2010).

Distinguishable molecules do not enjoy the suppression  
→ rate is x 100 higher !



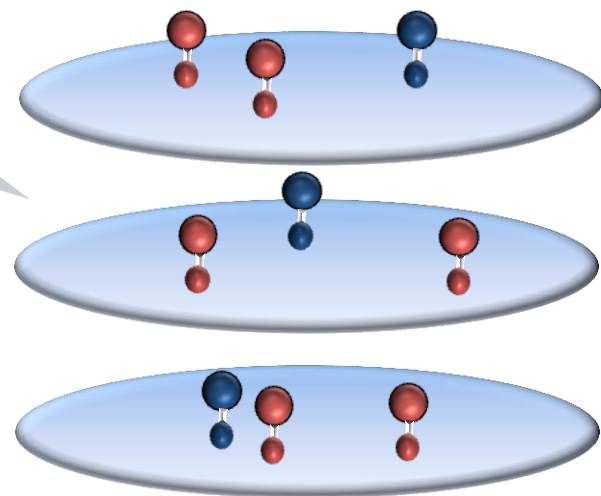
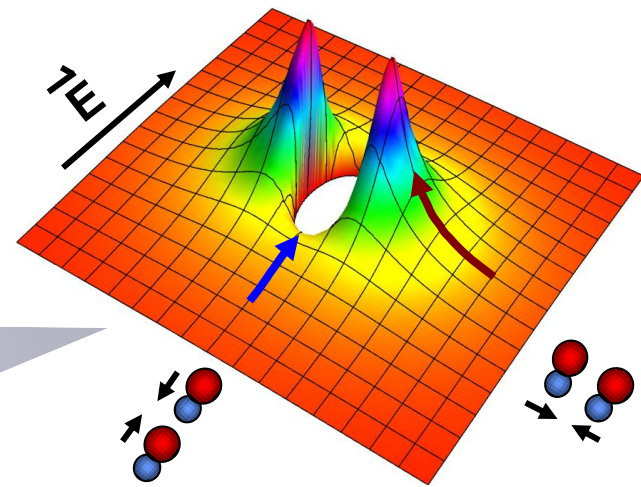
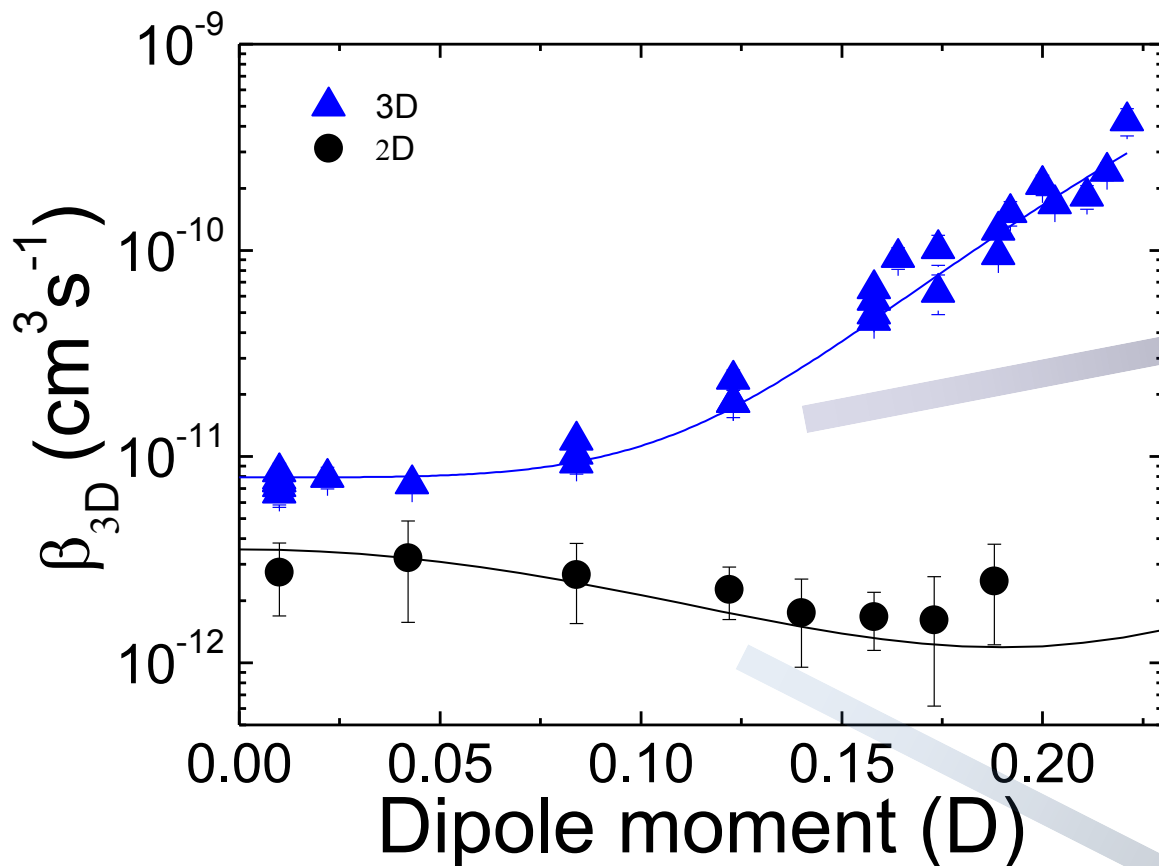
# Anisotropic dipolar collisions

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



Collisions under a single partial wave ( $L = 1\hbar$ ).

# 2D geometry - loss suppression



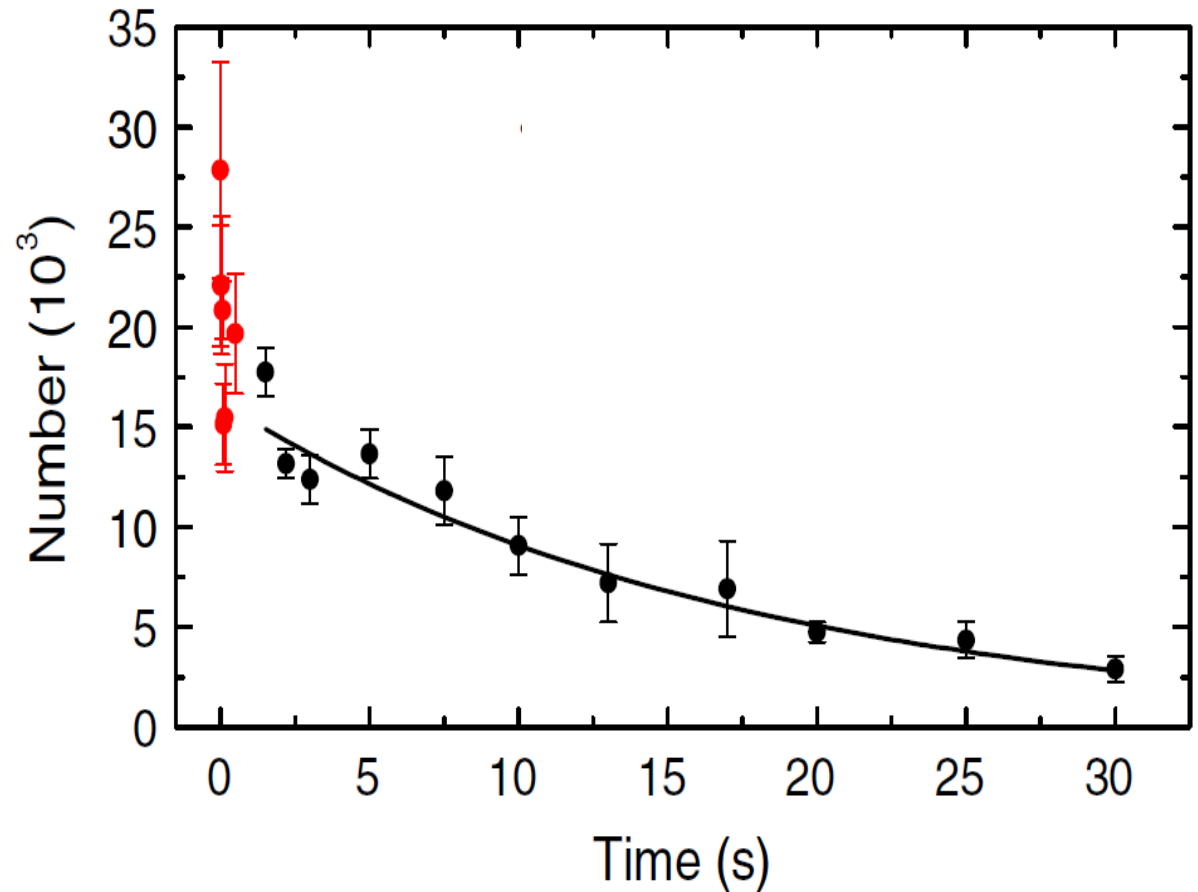
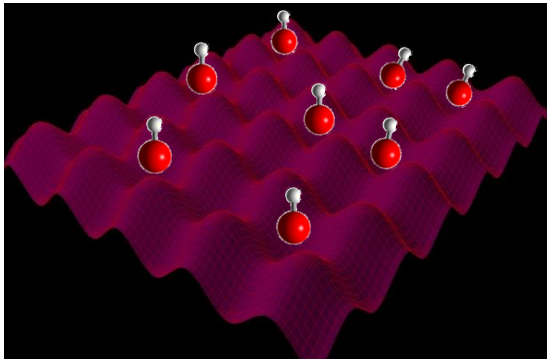
Theory: Büchler, Zoller, Bohn, Julienne

M. de Miranda, *et al.*,  
"Controlling the quantum stereodynamics  
of ultracold bimolecular reactions,"  
Nature Phys. **7**, 502 (2011).



# 3D optical lattice - suppressing chemical reaction with quantum Zeno effect

Lifetime  $\sim 20$  s  
Filling  $\sim 5\%$



A. Chotia *et al.*, Phys. Rev. Lett. **108**, 080405 (2012).

B. Zhu *et al.*, Phys. Rev. Lett. **112**, 070404 (2014).

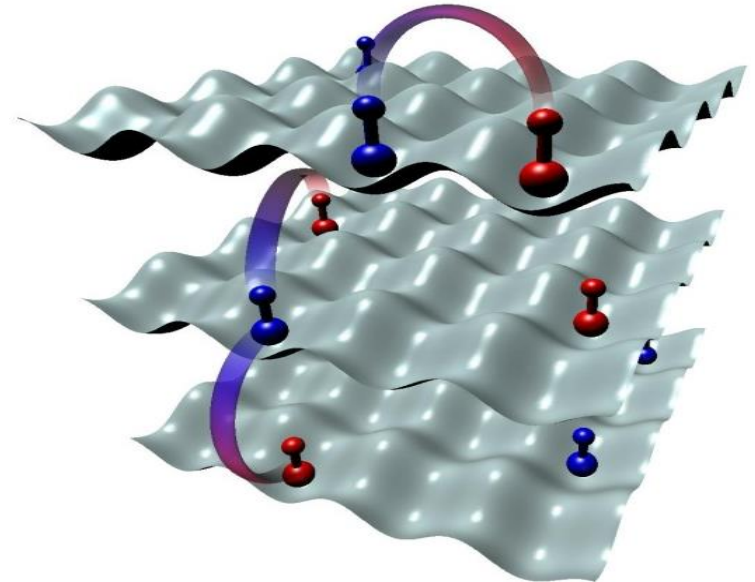
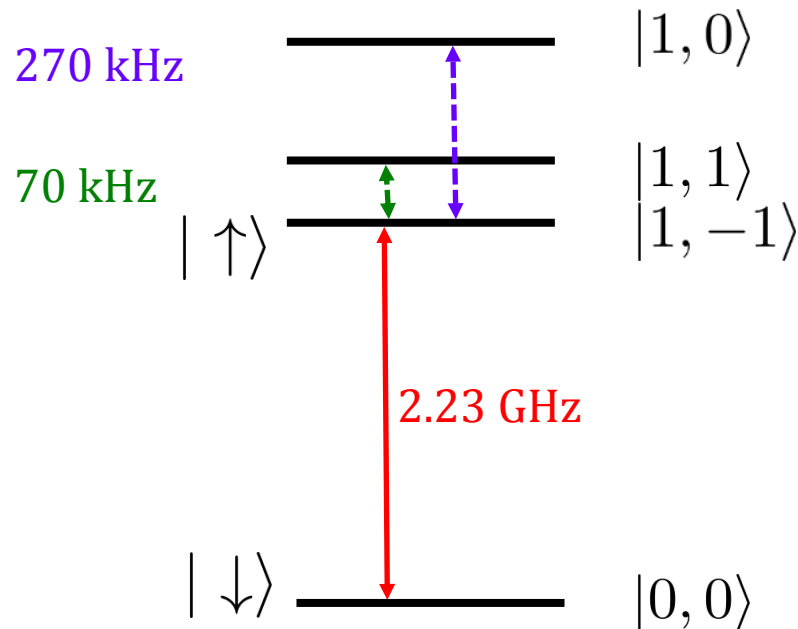
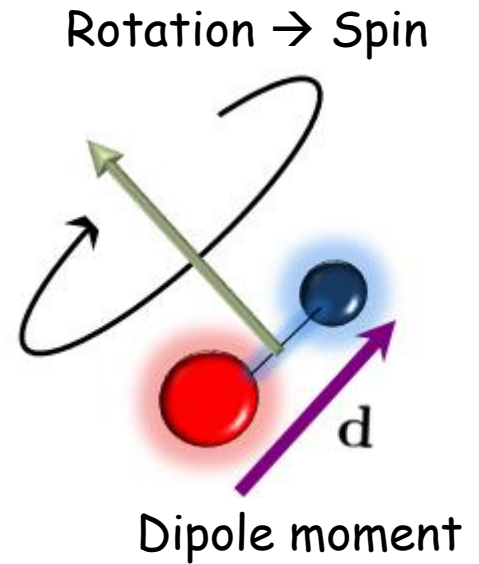
# Spin exchange in a lattice of molecules

Barnett *et al.*, Phys. Rev. Lett. **96**, 190401 (2006).

Micheli *et al.*, Nature Phys. **2**, 341 (2006).

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

- Long-range dipolar interactions for direct ( $\sim$ kHz) spin exchanges - motion & spin decoupled
- Fully tunable with electromagnetic fields

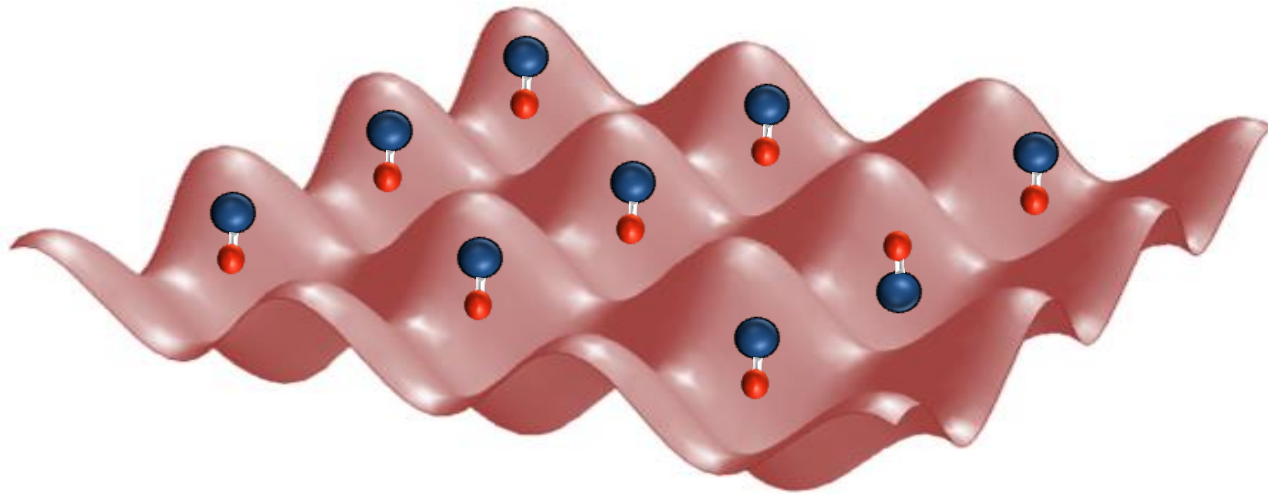




# A good system to study many-body quantum localization ?

D. Huse, G. Shlyapnikov, M. Lukin, E. Demler, ...

Molecules (material) are physically pinned down,  
but spins (excitations) can be exchanged and mobile !



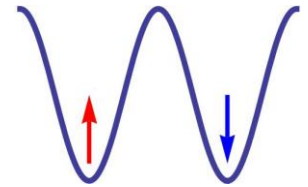
Energy flow in a macro-molecule !

# Interaction strength for quantum magnetism

$$\langle \downarrow | \hat{d} | \downarrow \rangle = \langle \uparrow | \hat{d} | \uparrow \rangle = 0$$

$$\langle \downarrow | \hat{d} | \uparrow \rangle \neq 0$$

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

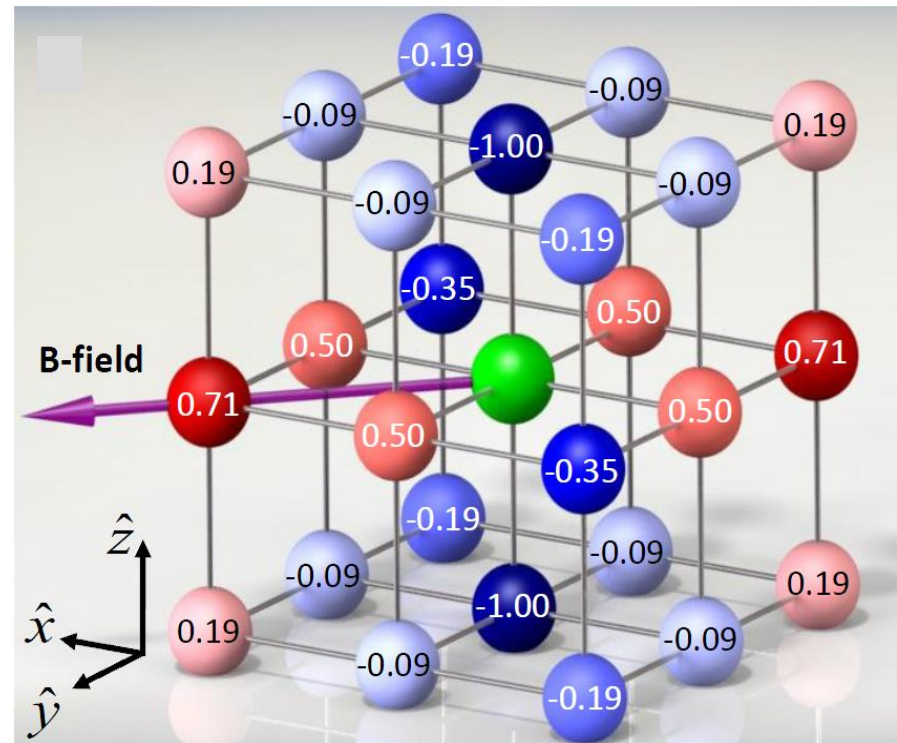


$$= S_i^x S_j^x + S_i^y S_j^y \quad \text{Flip-flop term}$$

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

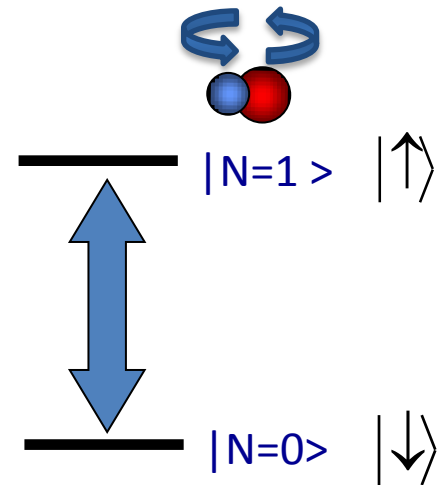
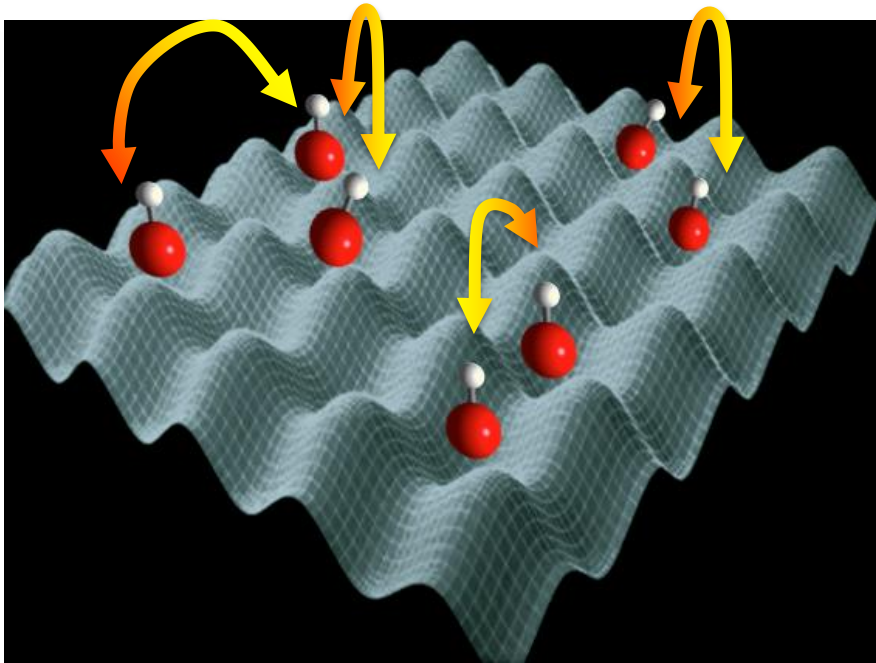
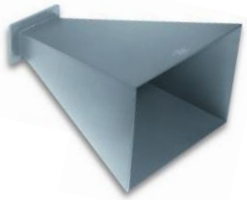
$$\frac{J_{\perp}}{h} = \frac{d_{\uparrow\downarrow}^2}{4\pi\epsilon_0 \hbar a_{\text{lat}}^3}$$

The oscillation frequency for a pair of molecules is  $J_{\perp}/2\hbar$ .

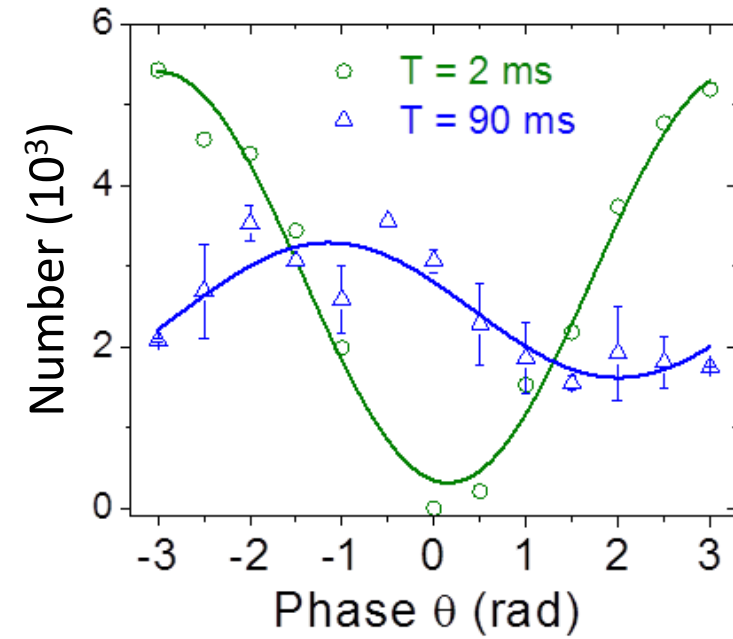


# A Dipolar Spin-Lattice Model

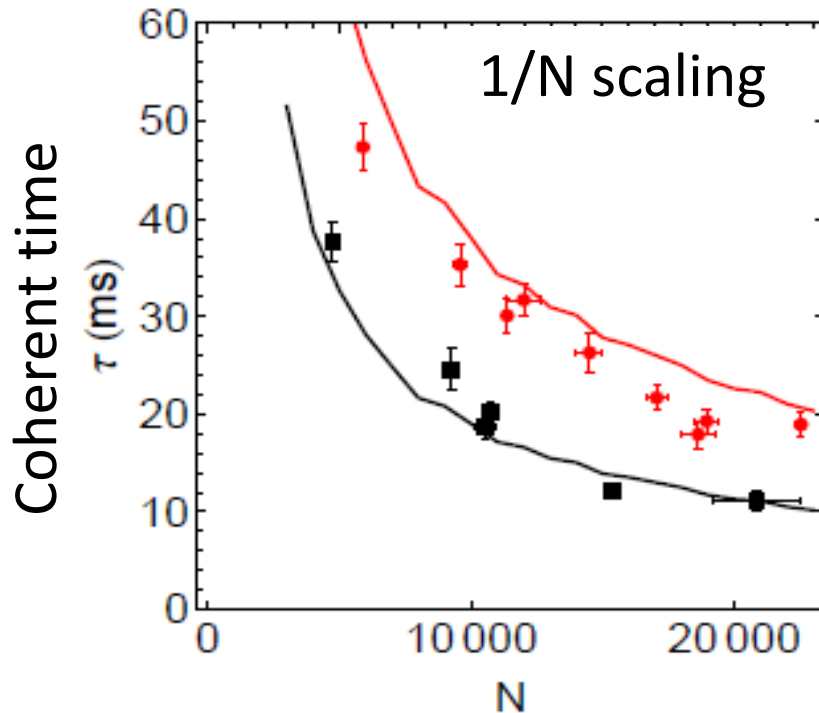
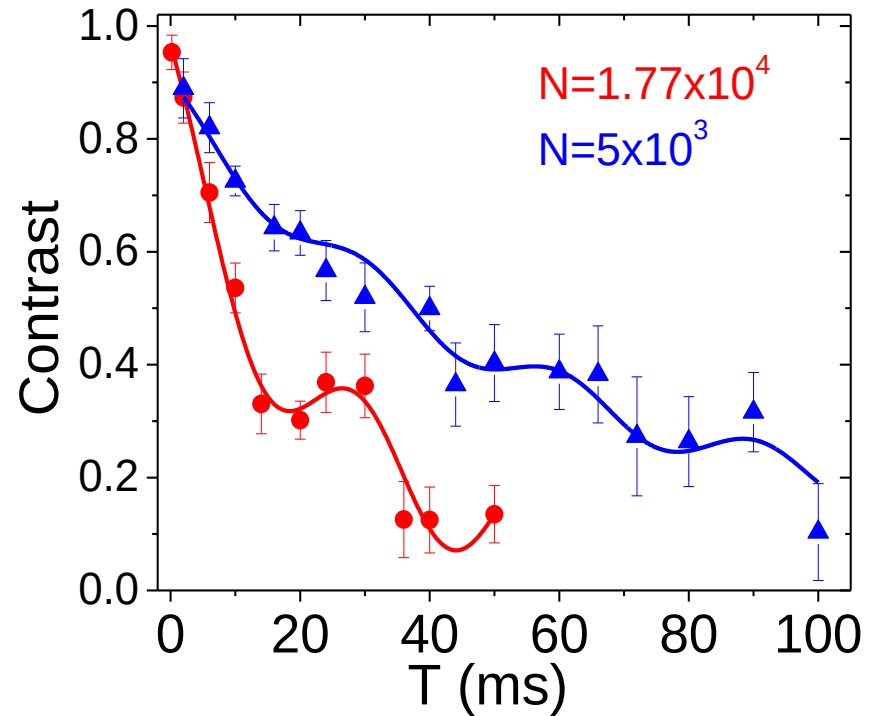
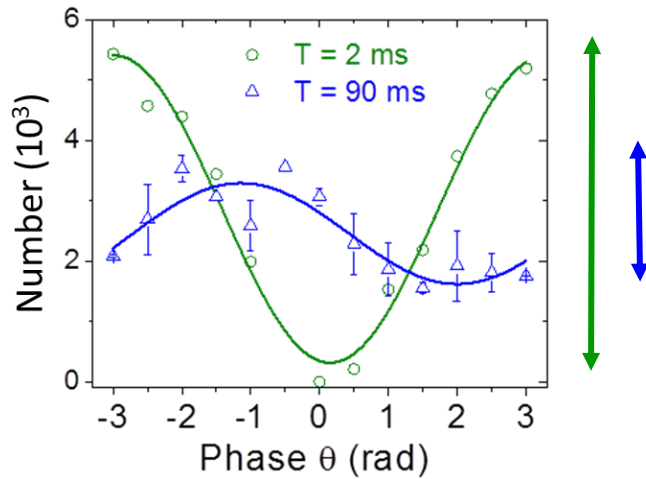
B. Yan *et al.*, Nature **501**, 521 (2013).



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



# Oscillations due to dipolar interactions



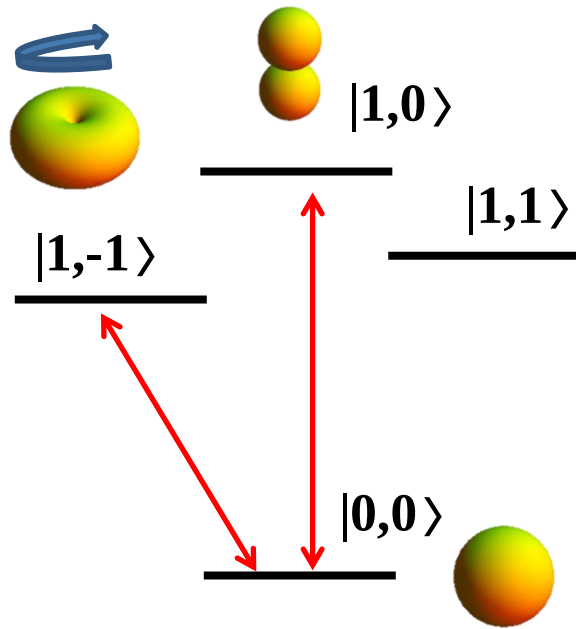
$|1, -1\rangle \leftrightarrow |0, 0\rangle$

$|1, 0\rangle \leftrightarrow |0, 0\rangle$

Oscillation frequencies  
& decay time  
both depend on  
rotational states

# Control dipolar interaction

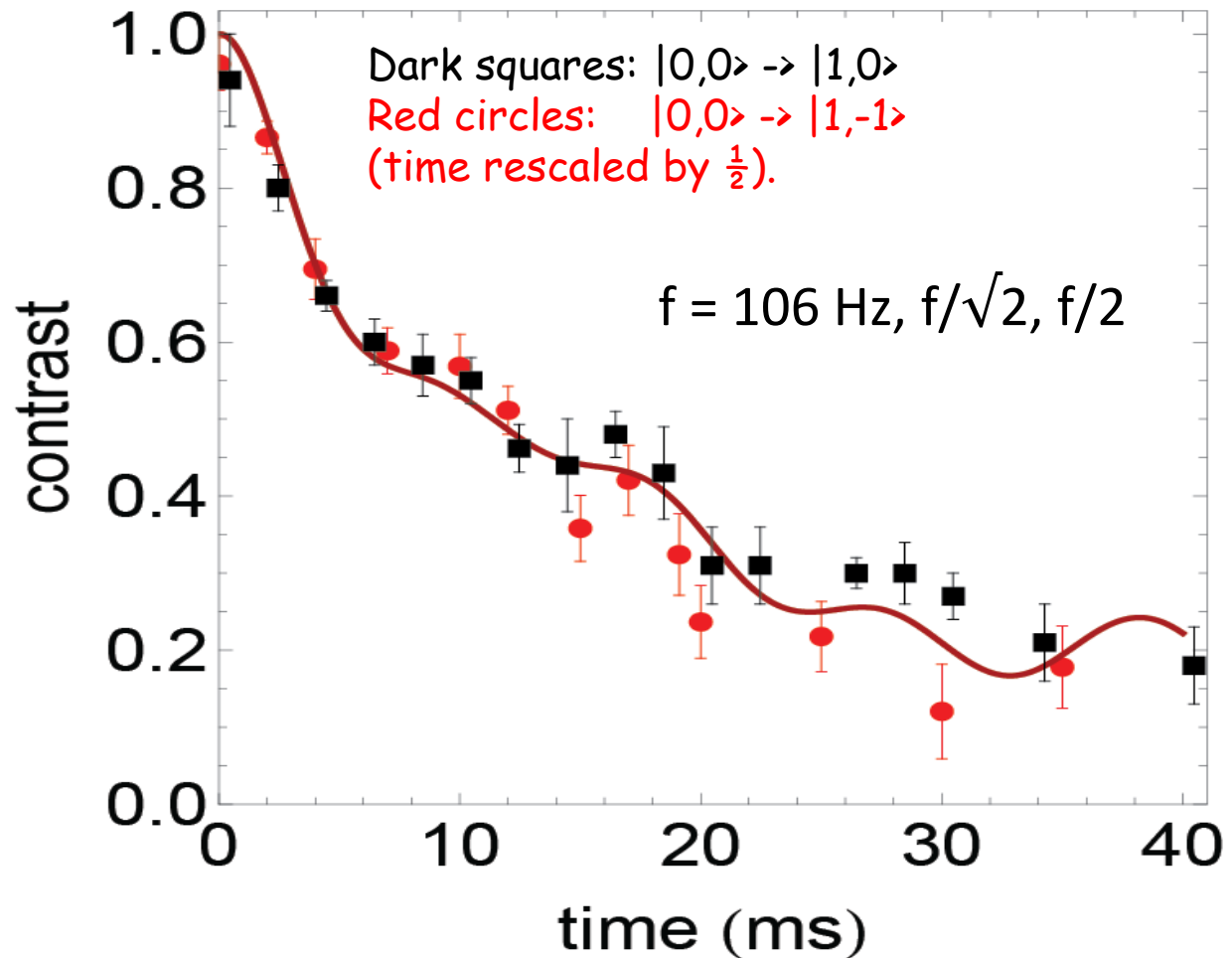
K. Hazzard *et al.*, Phys. Rev. Lett. **113**, 195302 (2014).



$$\frac{J_{\perp}}{2} \sim 102 \text{ Hz}$$

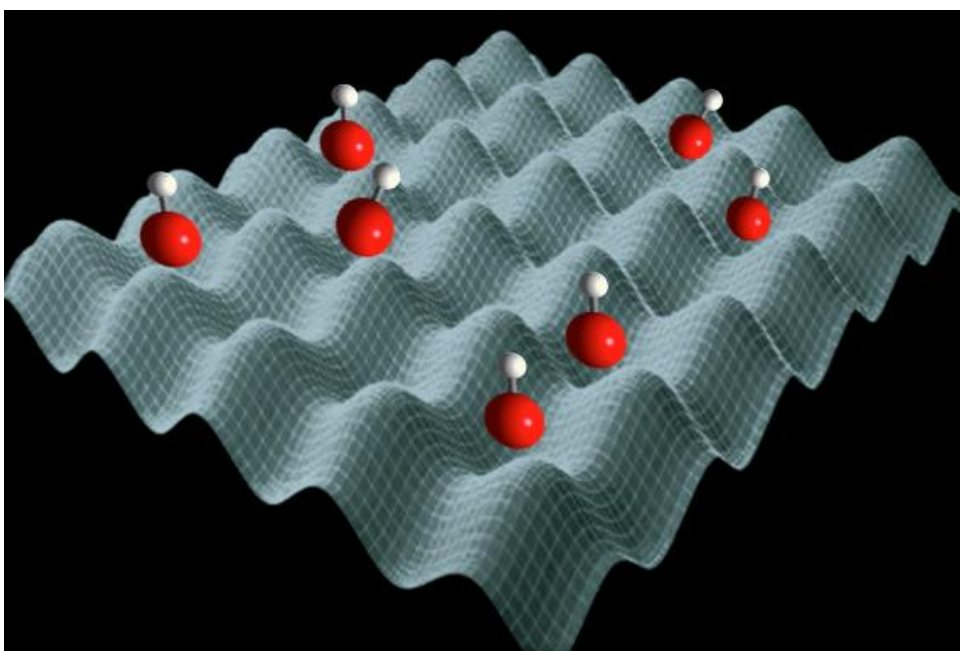
for  $|0,0\rangle$  to  $|1,0\rangle$

$$\left(\frac{J_{\perp}}{2} \sim 51 \text{ Hz for } |0,0\rangle \text{ to } |1,-1\rangle\right)$$

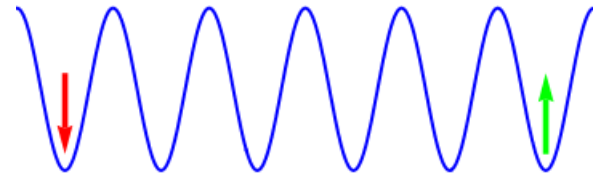


One fitting parameter (filling 5-10%) reproduces the experiment

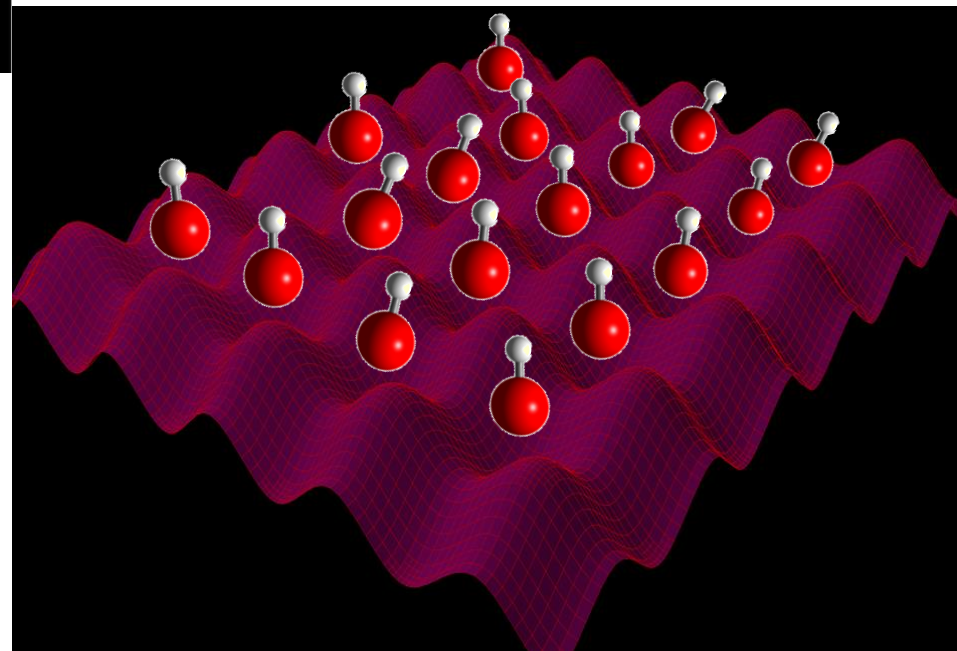
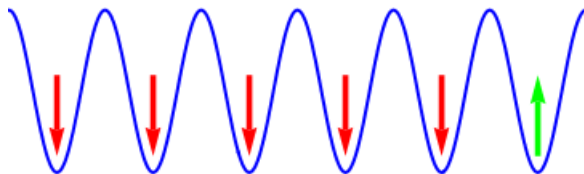
# Highly filled optical lattice



~5% filling in a 3D lattice

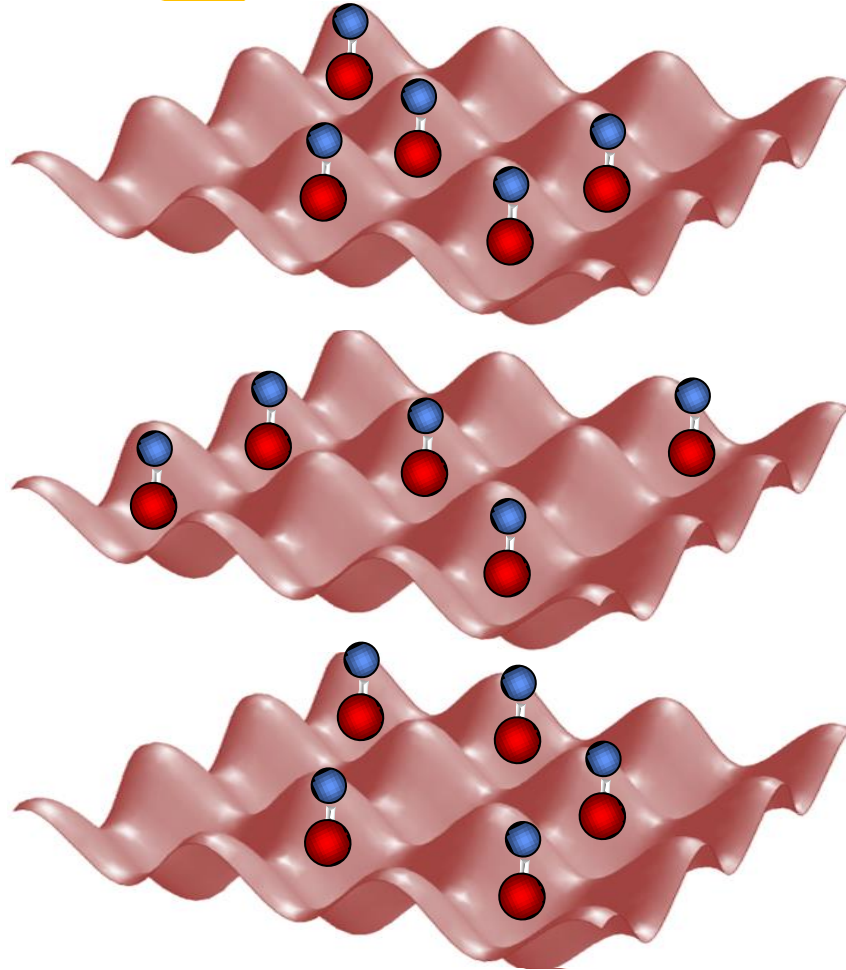


Goal:  
A near zero entropy 3D  
lattice

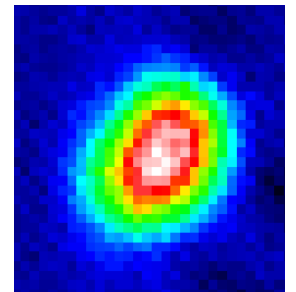
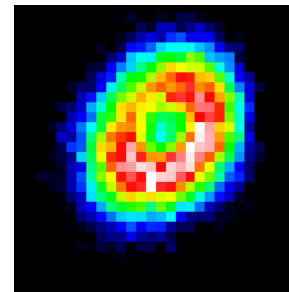
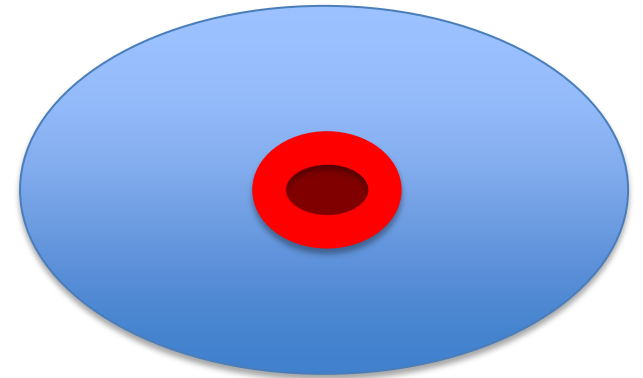




# Creating molecules in a 3D lattice



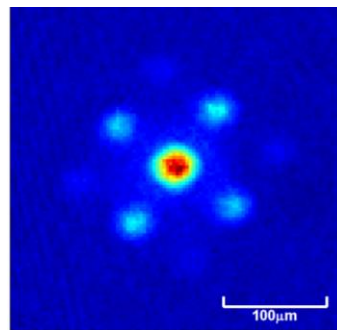
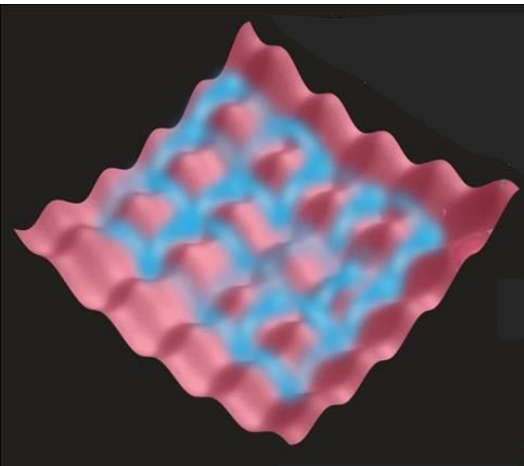
1. Rb MOTT insulator
2. Add lots of K atoms  
(tune Rb - K  
interaction energy)
3. Magnetic association  
& Raman transfer



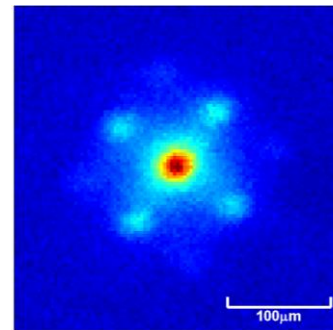


# Rb Mott insulator imaged *in-situ*

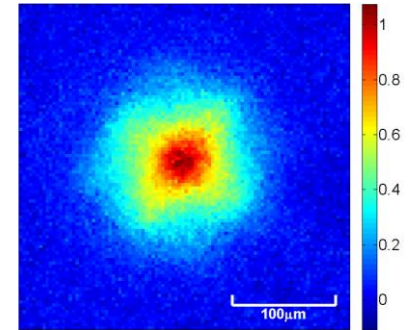
A superfluid BEC phase transition to a MOTT insulator



$12 E_{\text{rec}}$

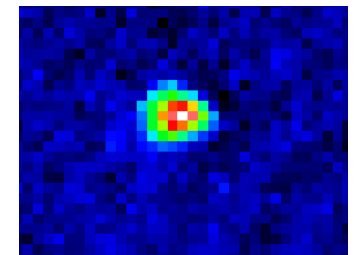
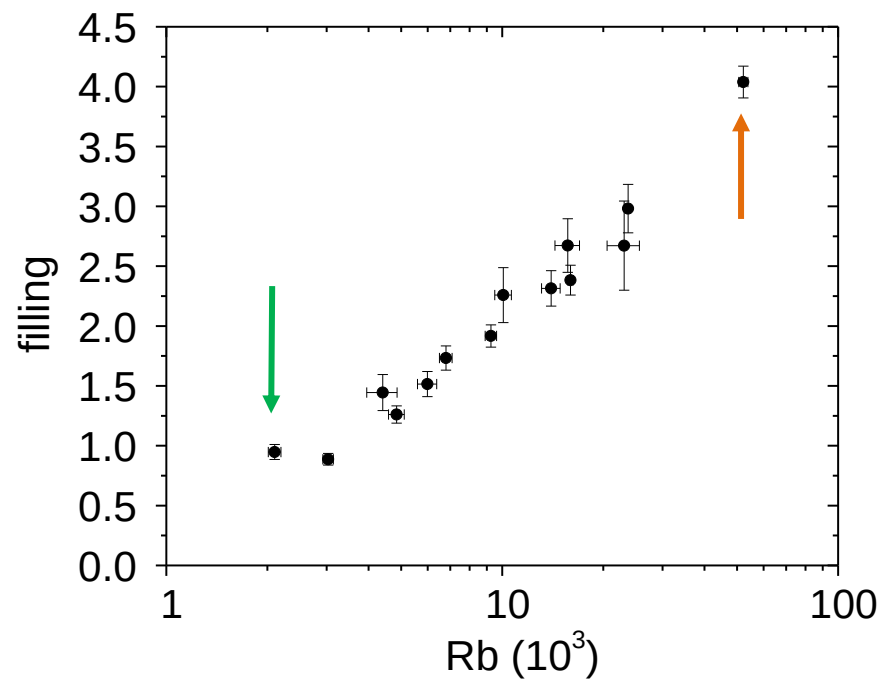


$18 E_{\text{rec}}$

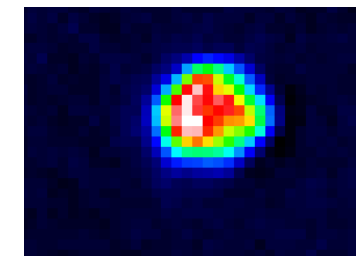


$24 E_{\text{rec}}$

Lattice depth

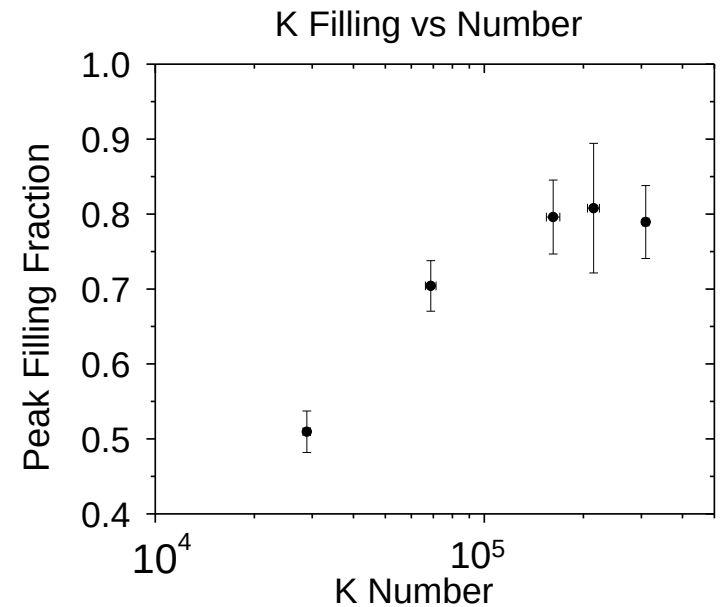
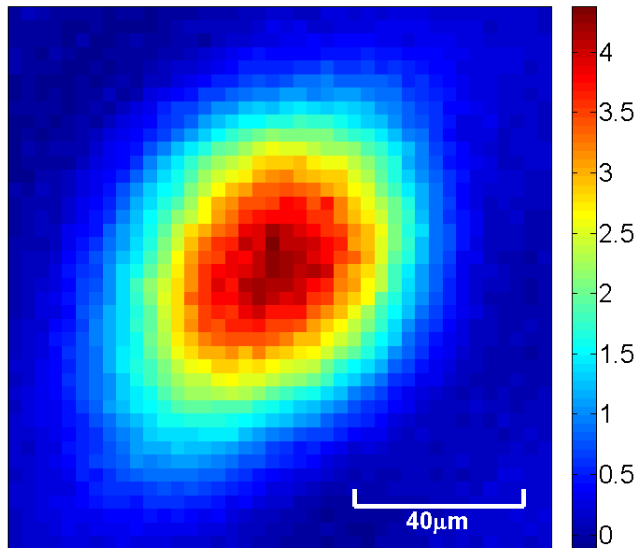
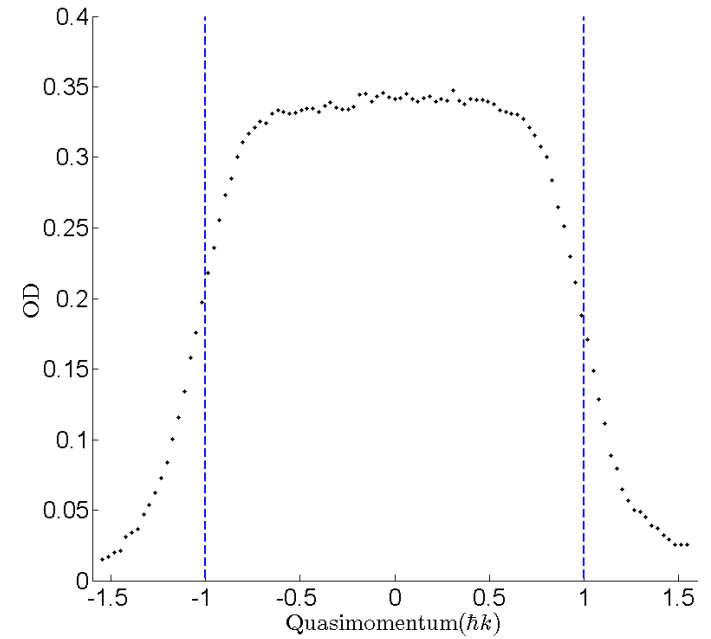
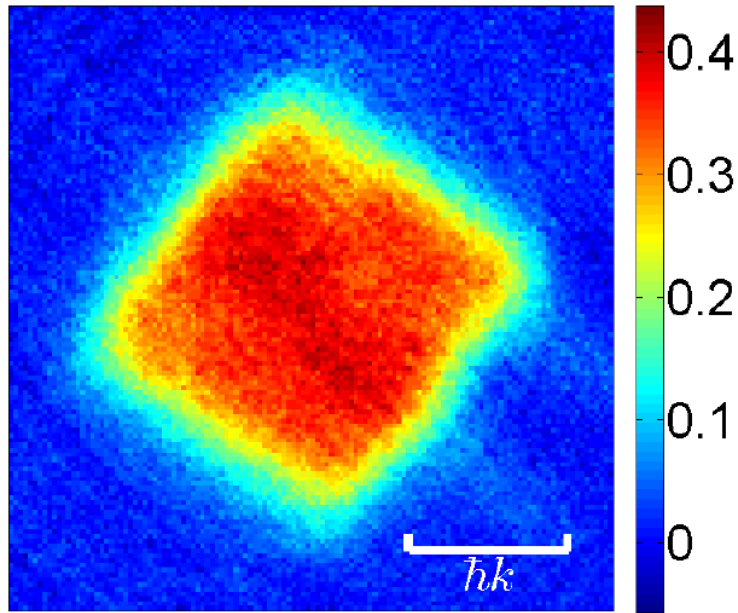


2000 Rb

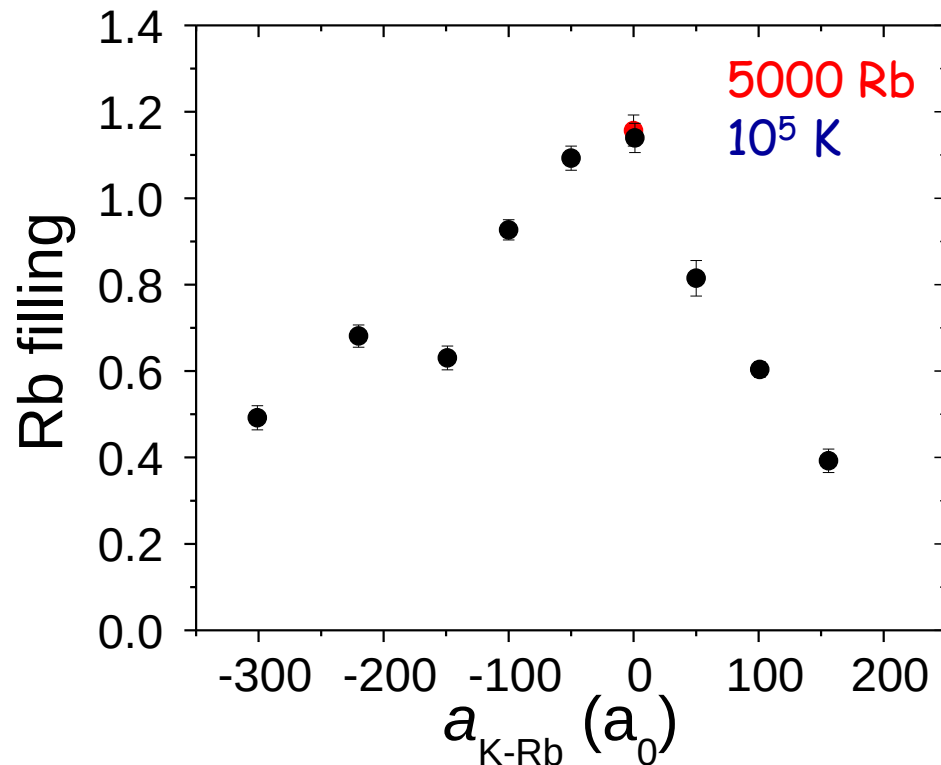
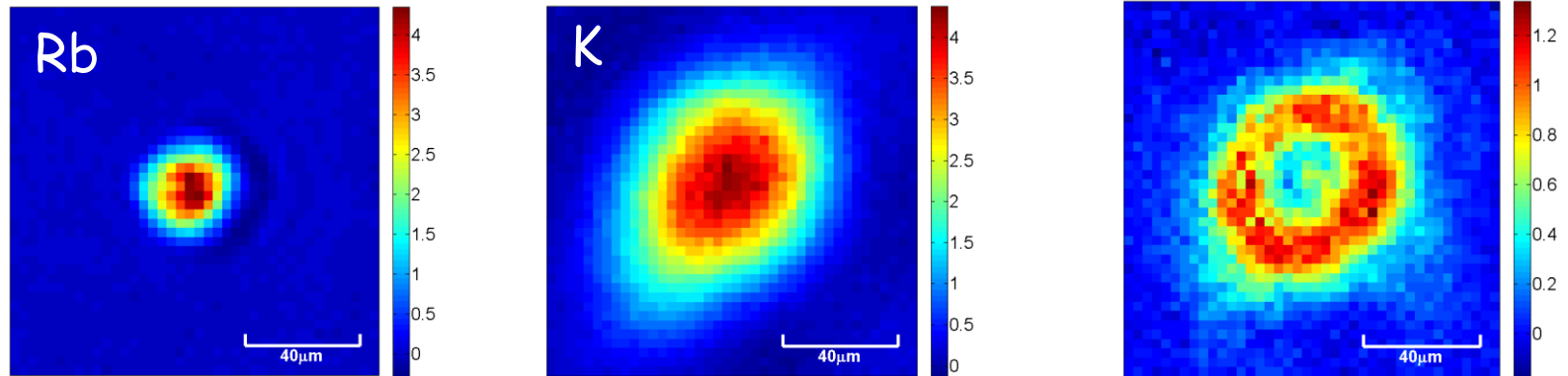


50,000 Rb

# Imaging of K in momentum & real space



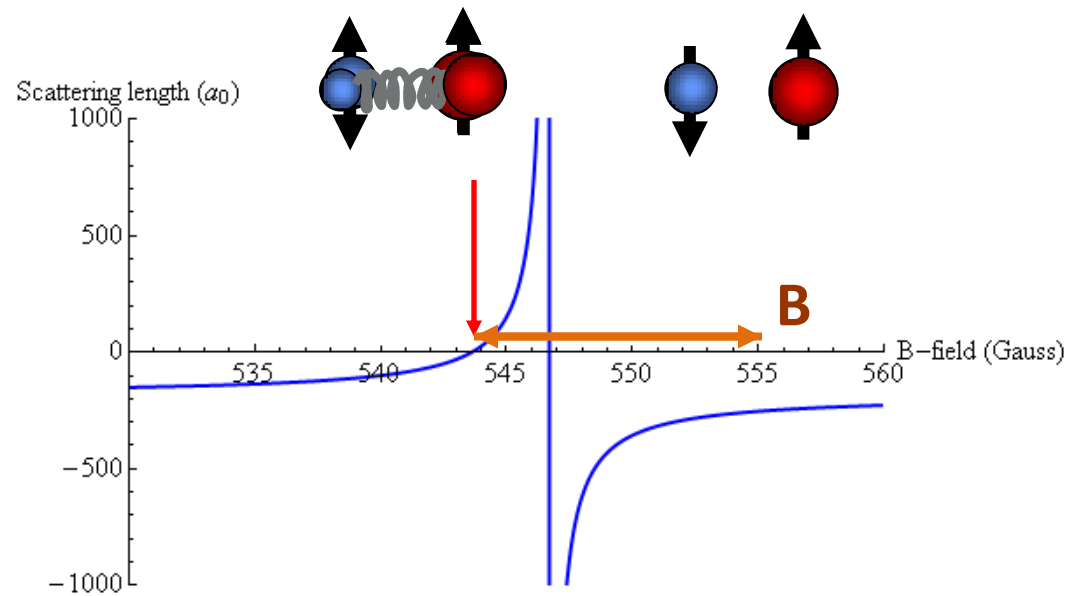
# Overlap of Rb and K



- Vary  $a_{\text{KRb}}$  before loading lattice
- At  $a_{\text{KRb}} = 0$ , Rb MI unaffected by K

# Pairing up K-Rb in lattice without heating

- Load the lattice at  $a_{\text{KRB}} = 0$
- Jumping across the resonance dilutes the filling (populating higher bands)
- Flip to a noninteracting spin state ( $|9/2, -7/2\rangle$ ) to avoid resonance when ramping B
- Flip back to  $|9/2, -9/2\rangle$ , then proceed with Feshbach association



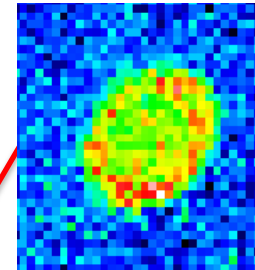
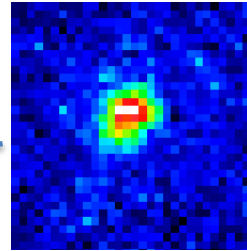
K:  $|F = 9/2, m_F = -9/2\rangle$  + Rb:  $|F = 1, m_F = 1\rangle$



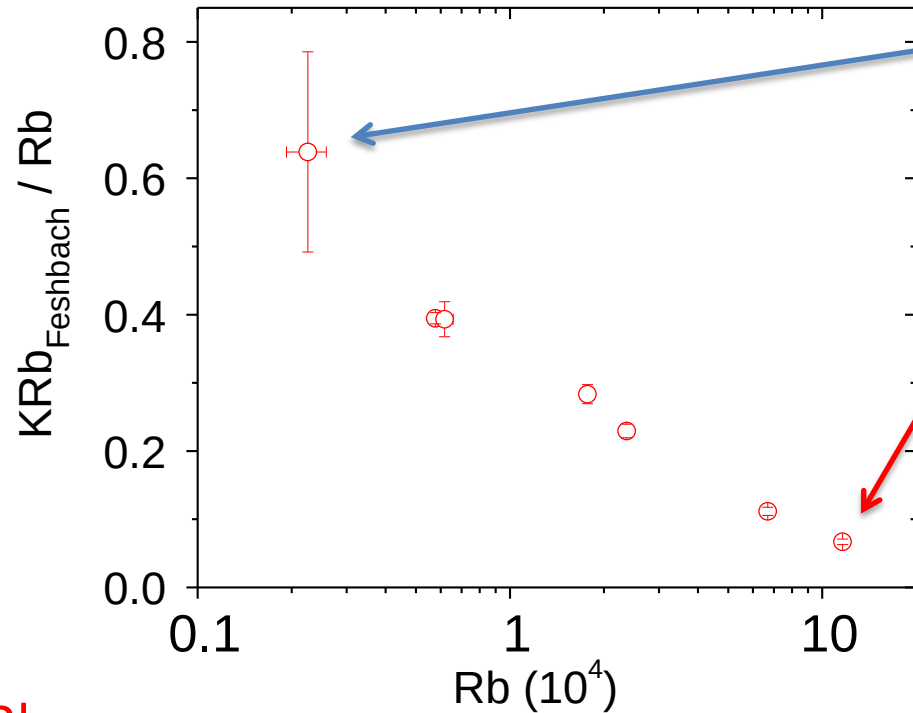
# A low entropy lattice of molecules

Convert > 60% of Rb MOTT Insulator to KRb

Image of KRb

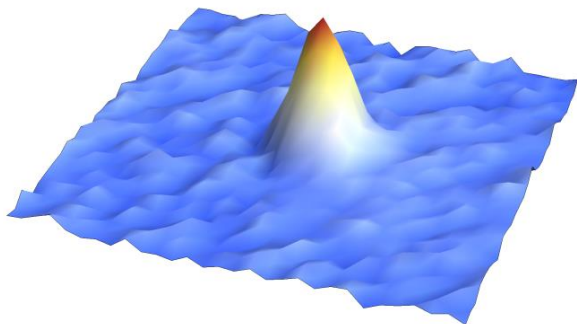


~5% filling

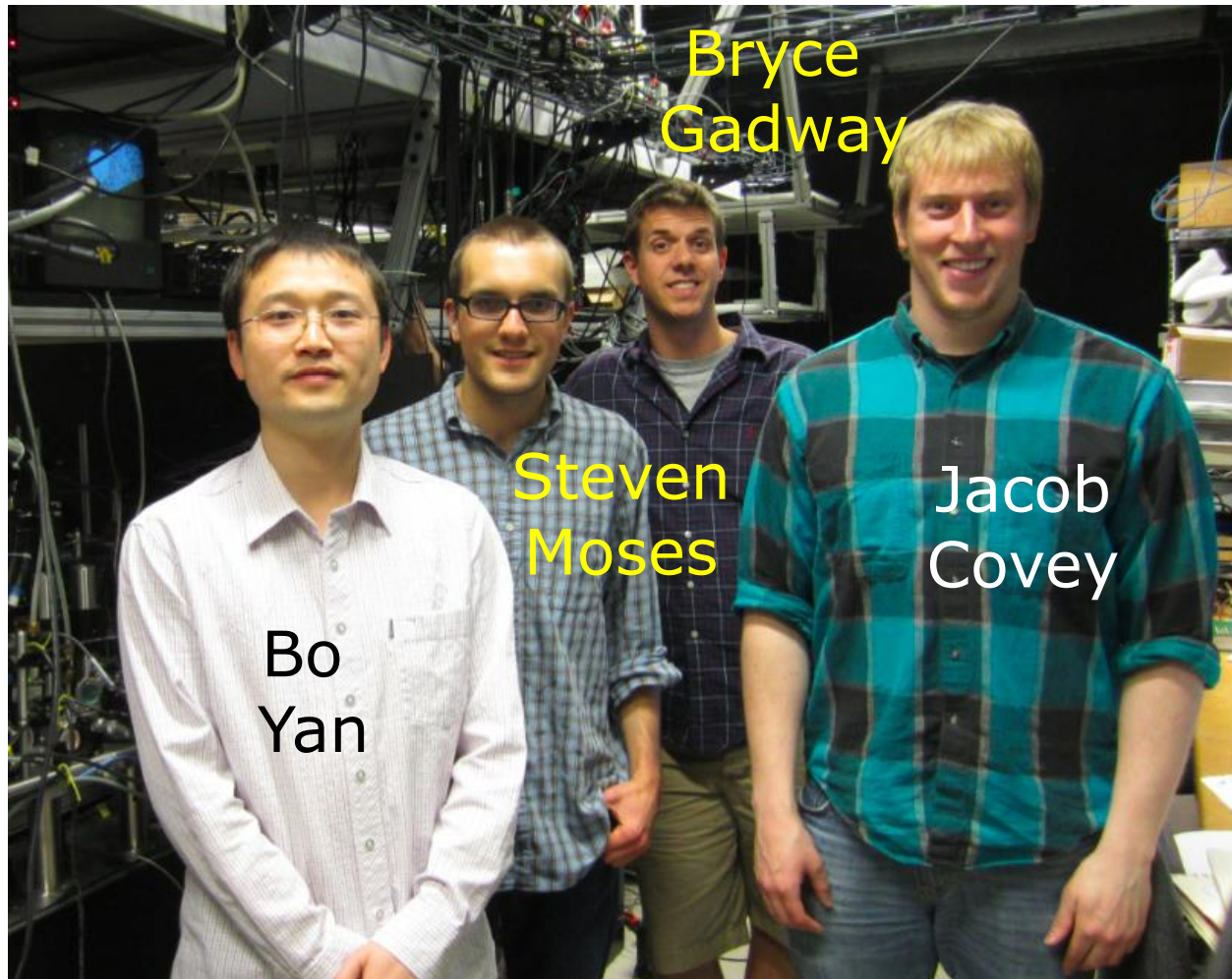


Ground state KRb,  
~ 40% filling in 3D lattice

(entropy/molecule ~ 1.7  $k_B$ )



# Special Thanks (KRb team):



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Amodsen Chotia  
Marcio de Miranda  
Dajun Wang  
Silke Ospelkaus  
Kang-Kuen Ni  
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