Ultracold molecules – New frontiers in quantum & chemical physics

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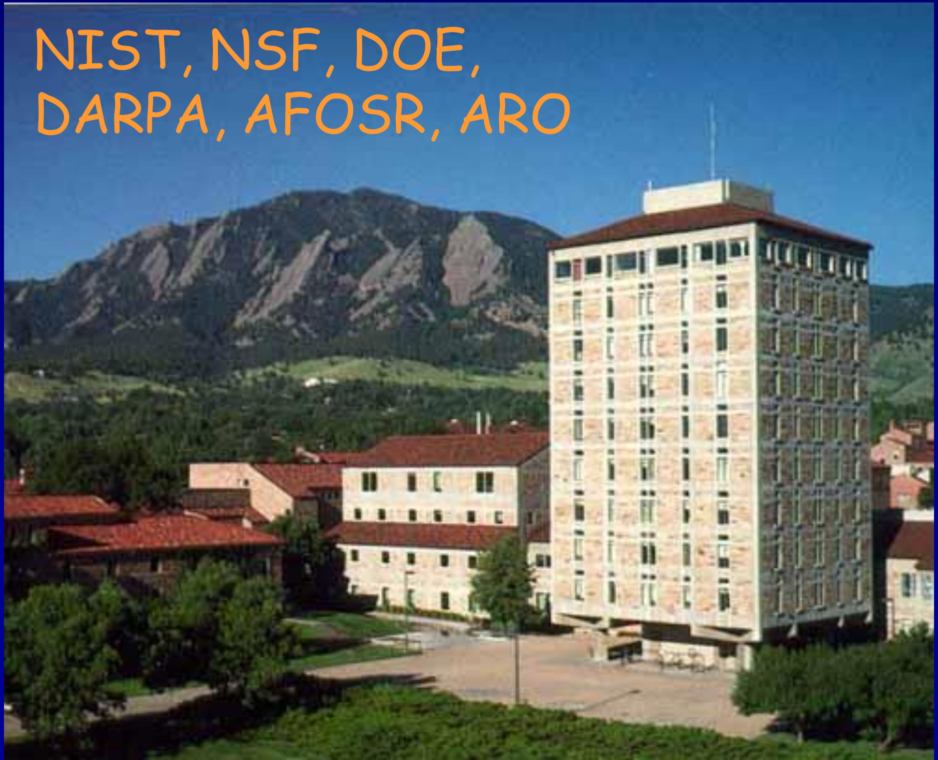
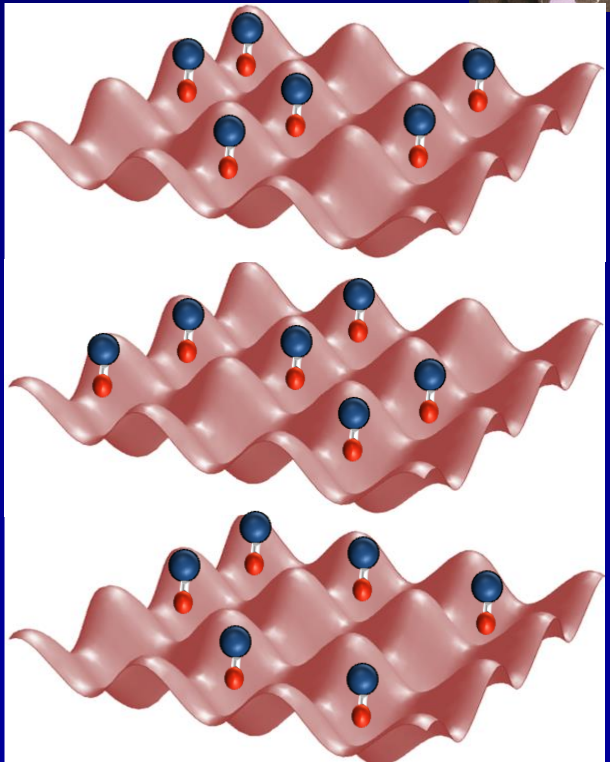
JILA, NIST & CU, Boulder

Fermi School Course 191
Quantum Matter at Ultralow Temperatures
July 9, 2014


NIST, NSF, DOE,
DARPA, AFOSR, ARO

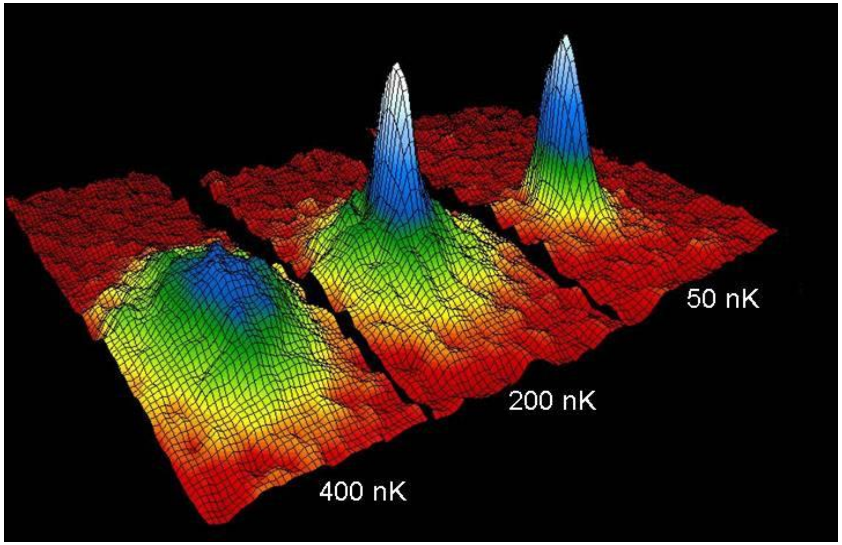


Ultracold quantum gases

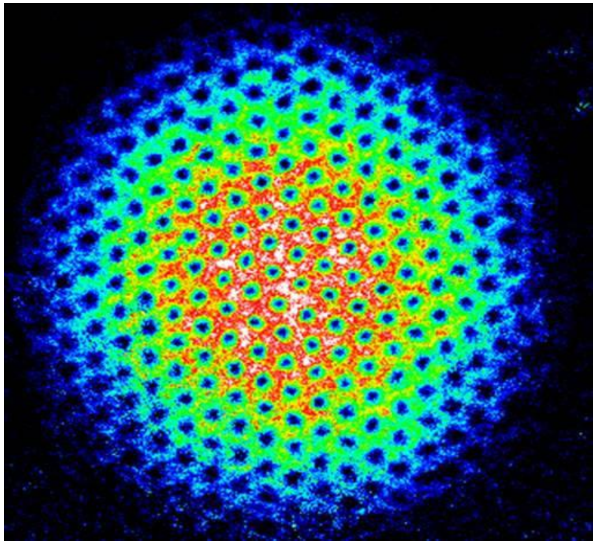
$T = 100$ nK

$N = 10^6$ atoms

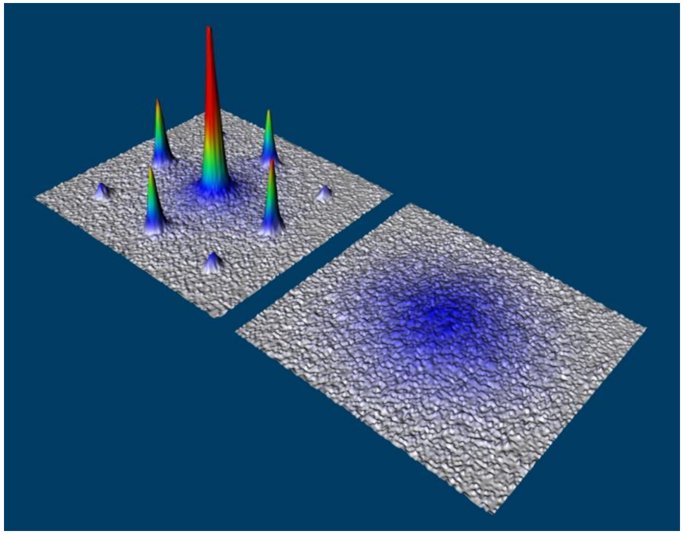
$n = 10^{13}$ cm^{-3}



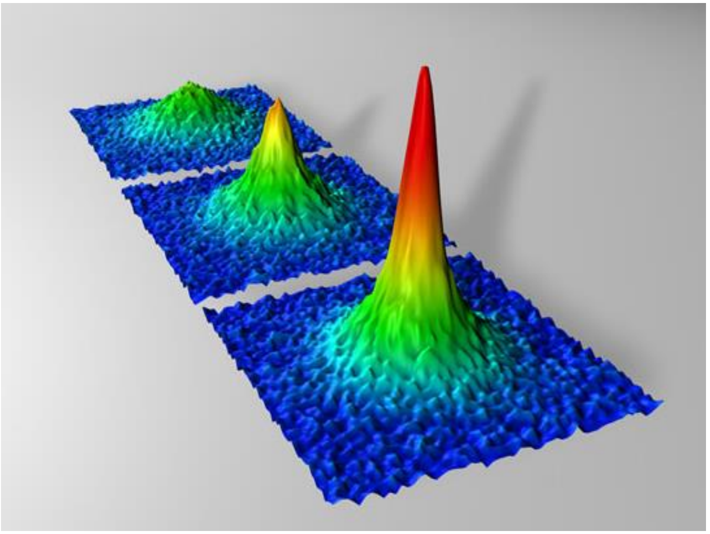
Bose-Einstein condensate



superfluidity

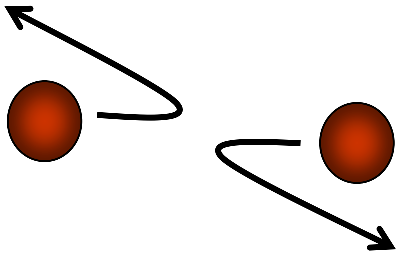


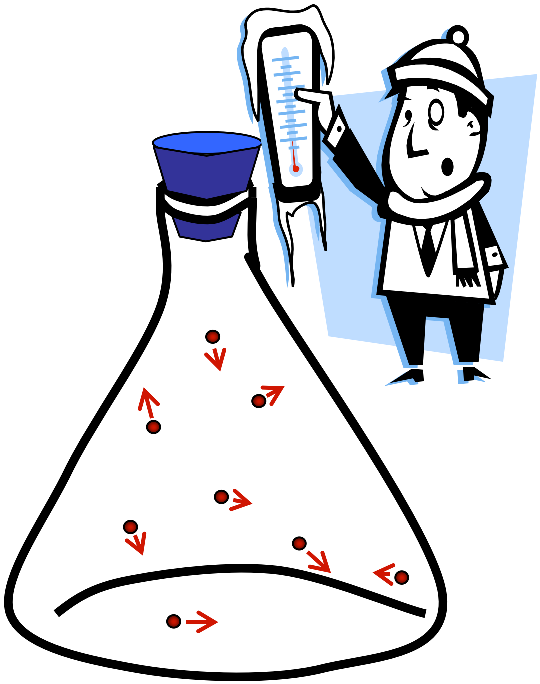
Superfluidity – MOTT insulator



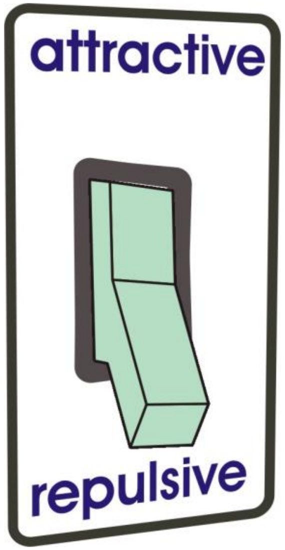
Fermi superfluidity

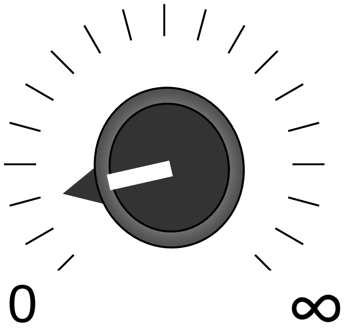
Atomic Interactions





We can understand &
control the interactions!

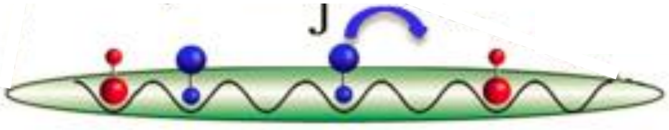


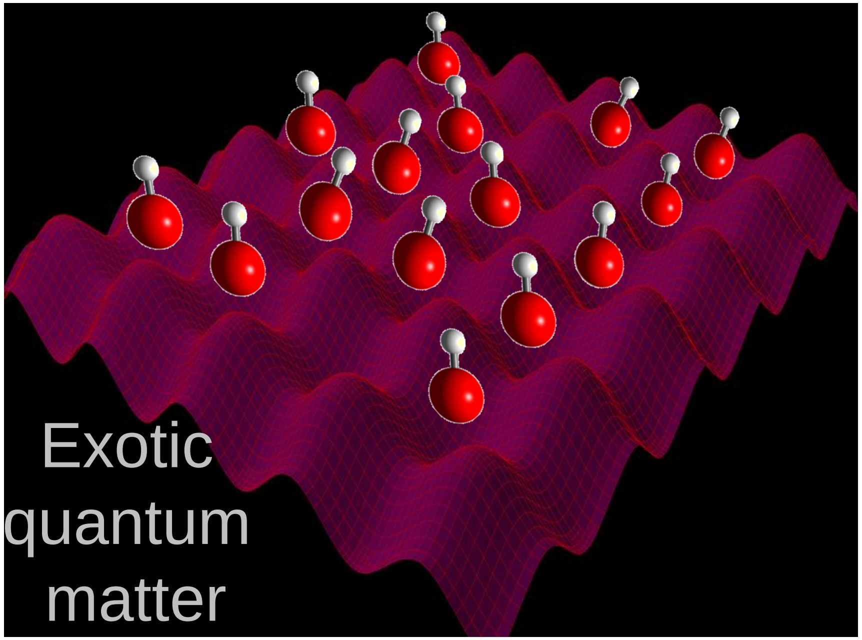


interaction strength

Quantum gas of polar molecules

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

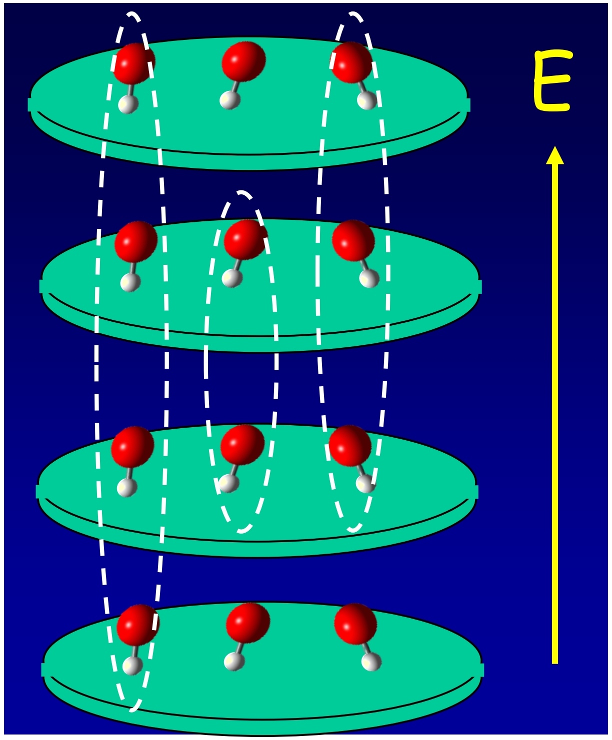




Quantum metrology,

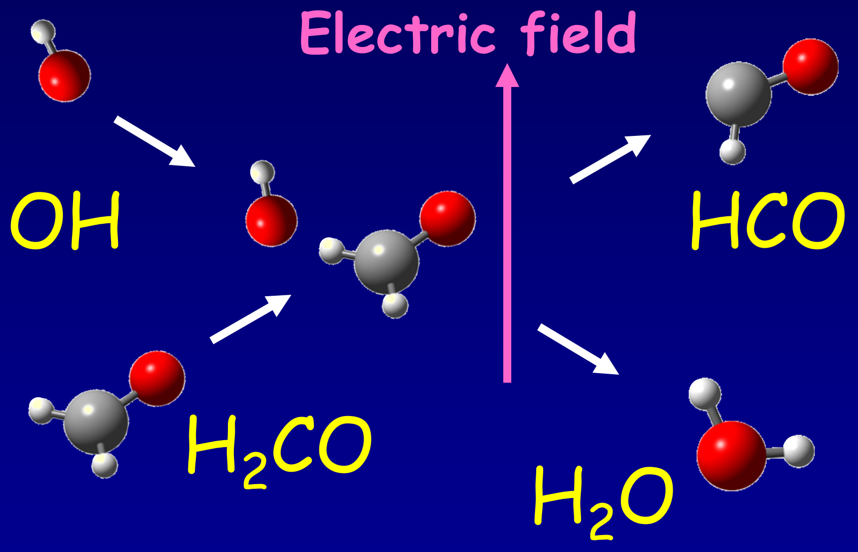
Correlated material,

Chemistry



Control reactions at ultralow energy

Hudson *et al.*, Phys. Rev. A **73**, 063404 (2006).



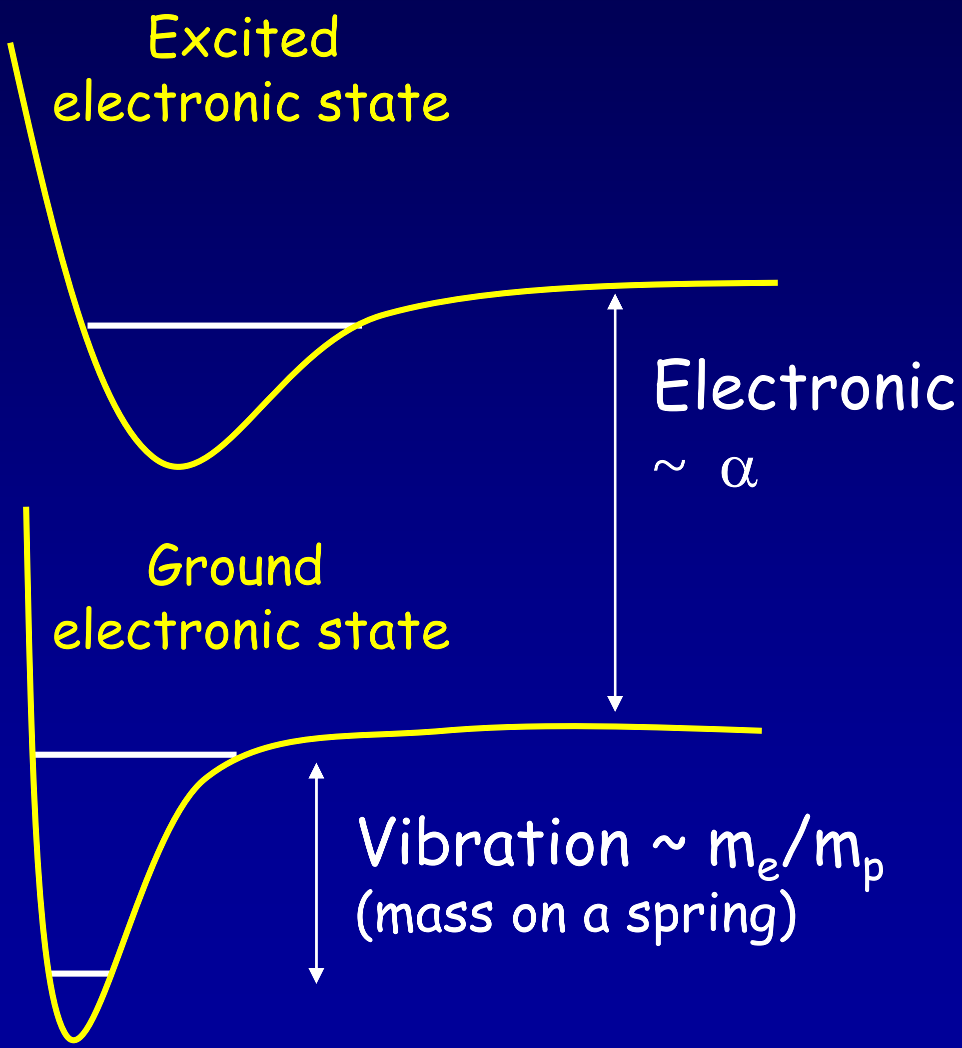
Controlled molecular collisions,
Ultracold chemical reactions

- Molecules in well-defined quantum states (internal & external)
- Long-range approaches precisely controlled

"Can a pico-eV knob control a 1 eV reaction ?" - J. L. Bohn

Ultracold molecules: Test fundamental principles

Hudson *et al.*, PRL 96, 143004 (2006).



Excited electronic state

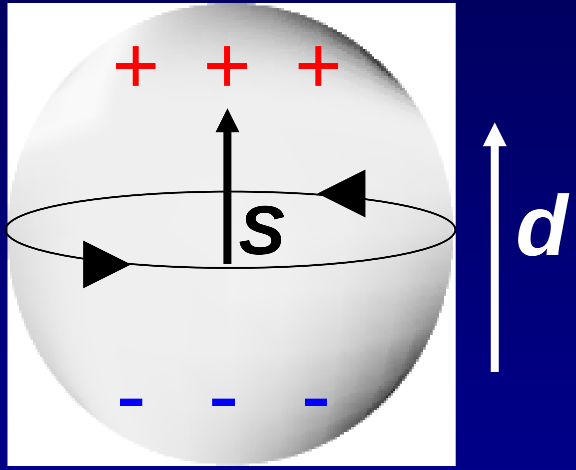
Electronic ~ α

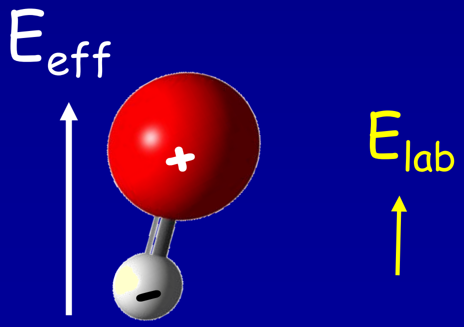
Ground electronic state

Vibration ~ m_e/m_p
(mass on a spring)

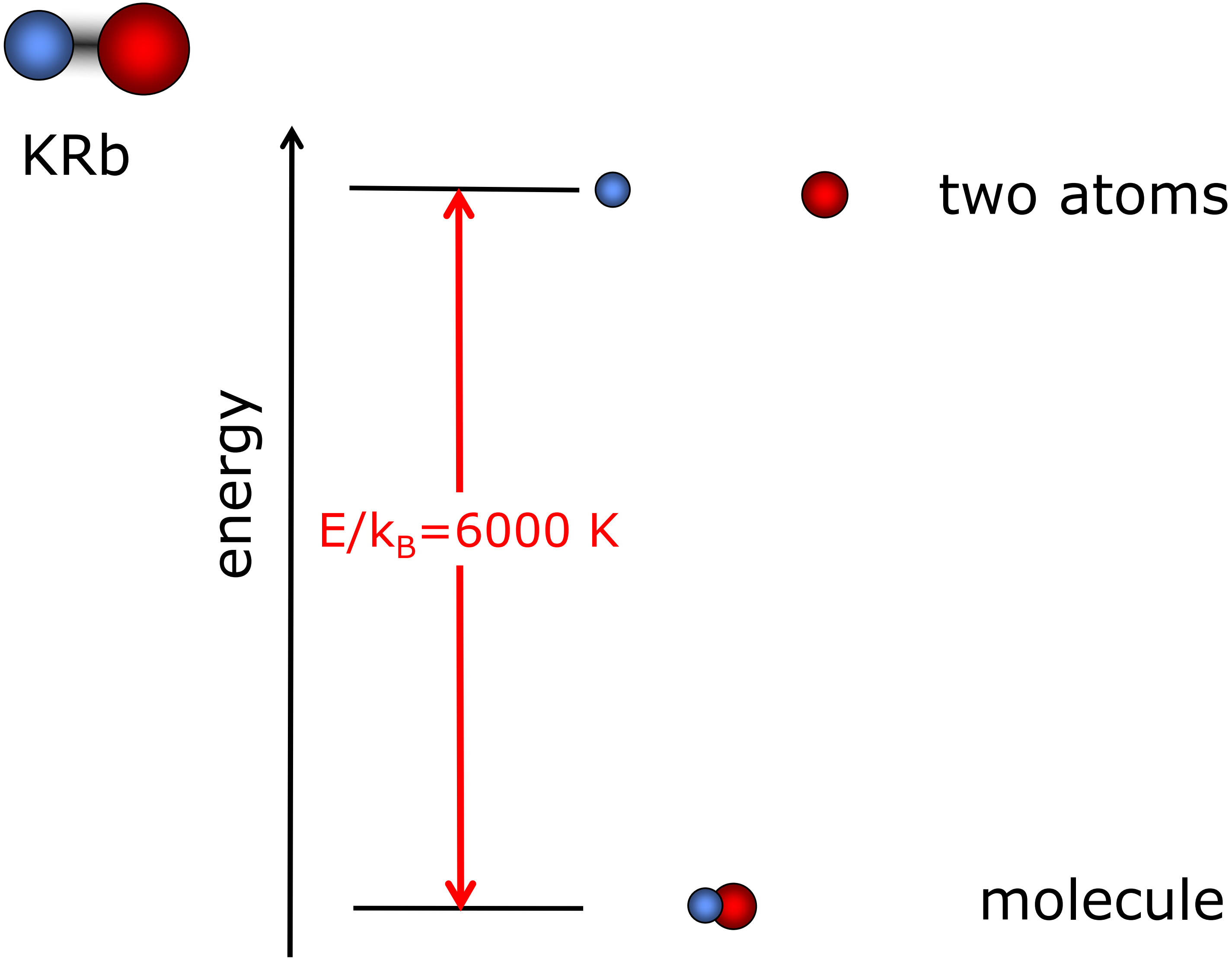
Hinds, Physica Scripta (1997).

Electron Electric Dipole Moment ?





Ultracold molecules: The challenge



KRb

energy

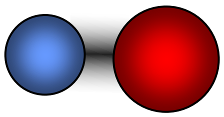
$E/k_B=6000$ K

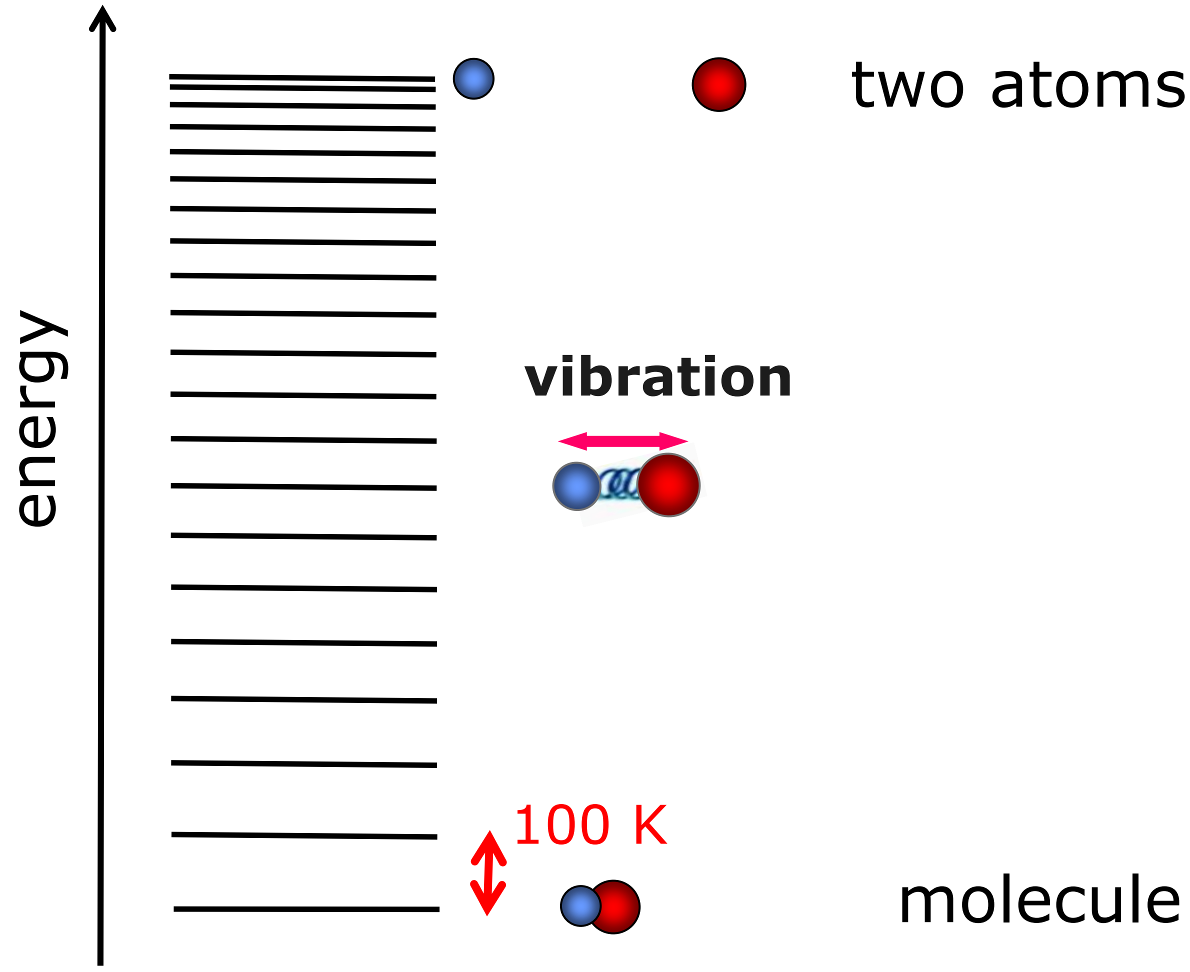
two atoms

molecule

Ultracold molecules: The challenge

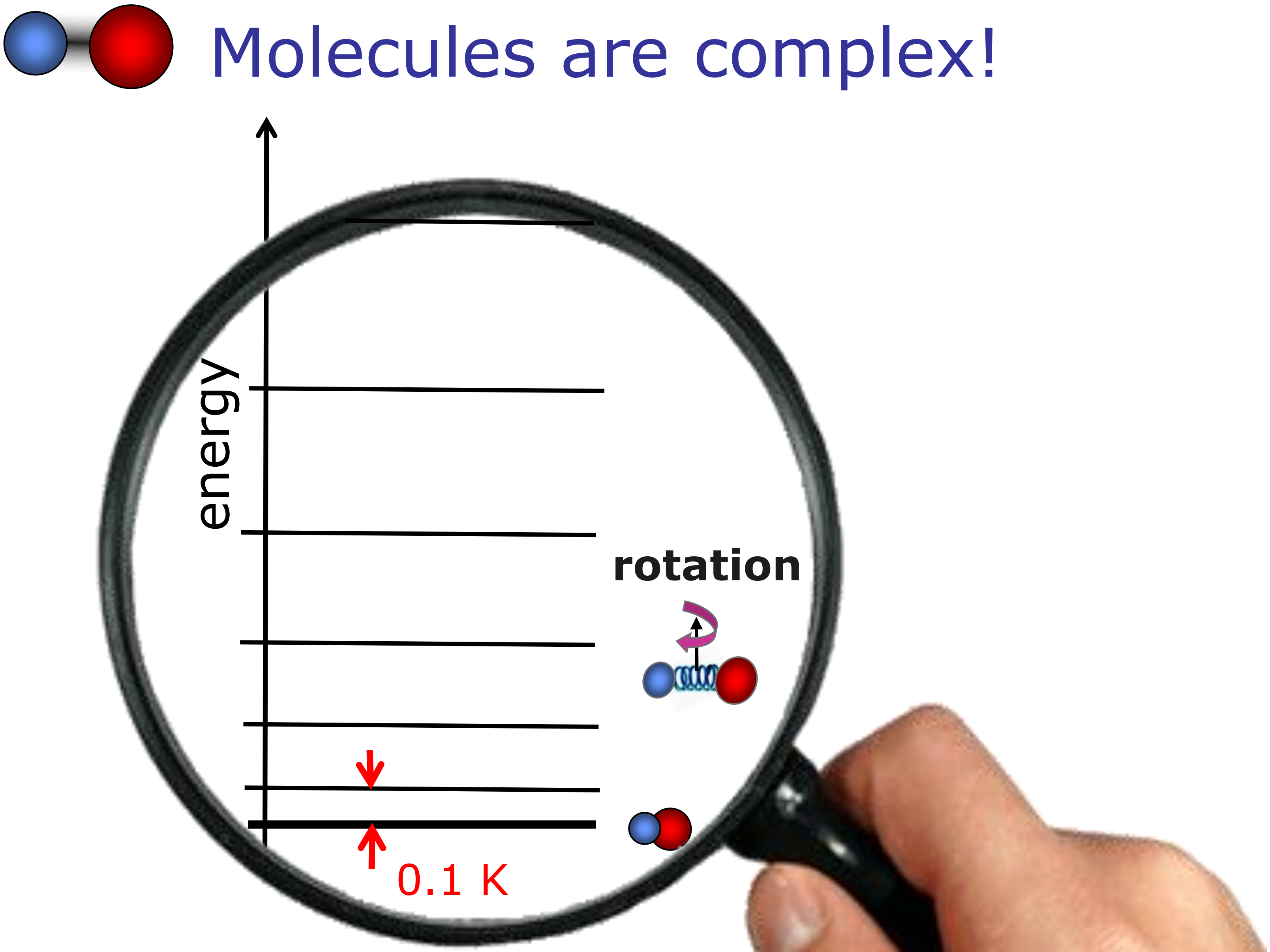
Molecules are complex!





Ultracold molecules: The challenge

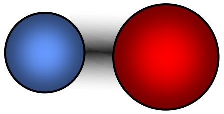
Molecules are complex!



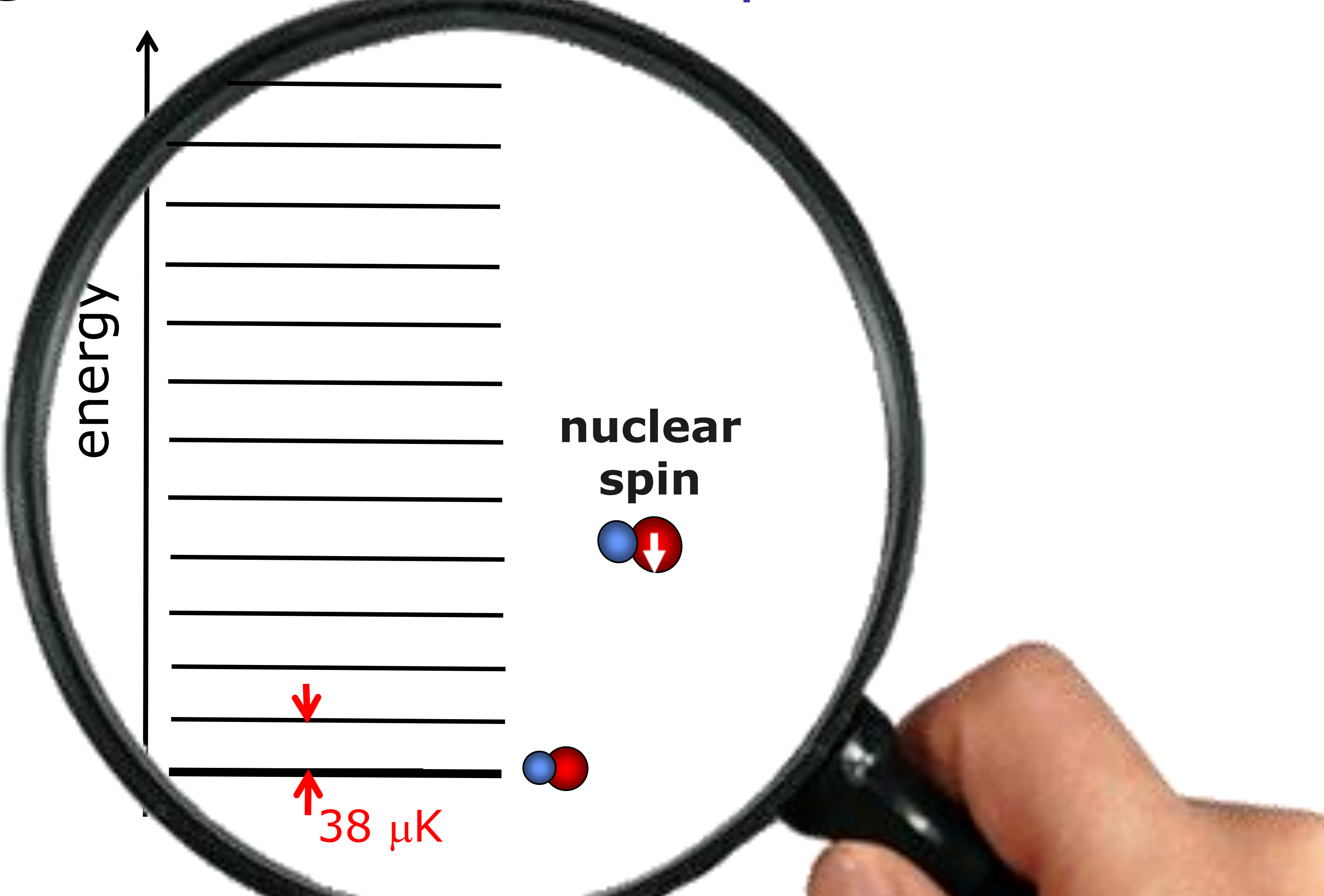
Ultracold molecules: The challenge

Molecules are complex!

Ultracold molecules: The challenge



Molecules are complex!

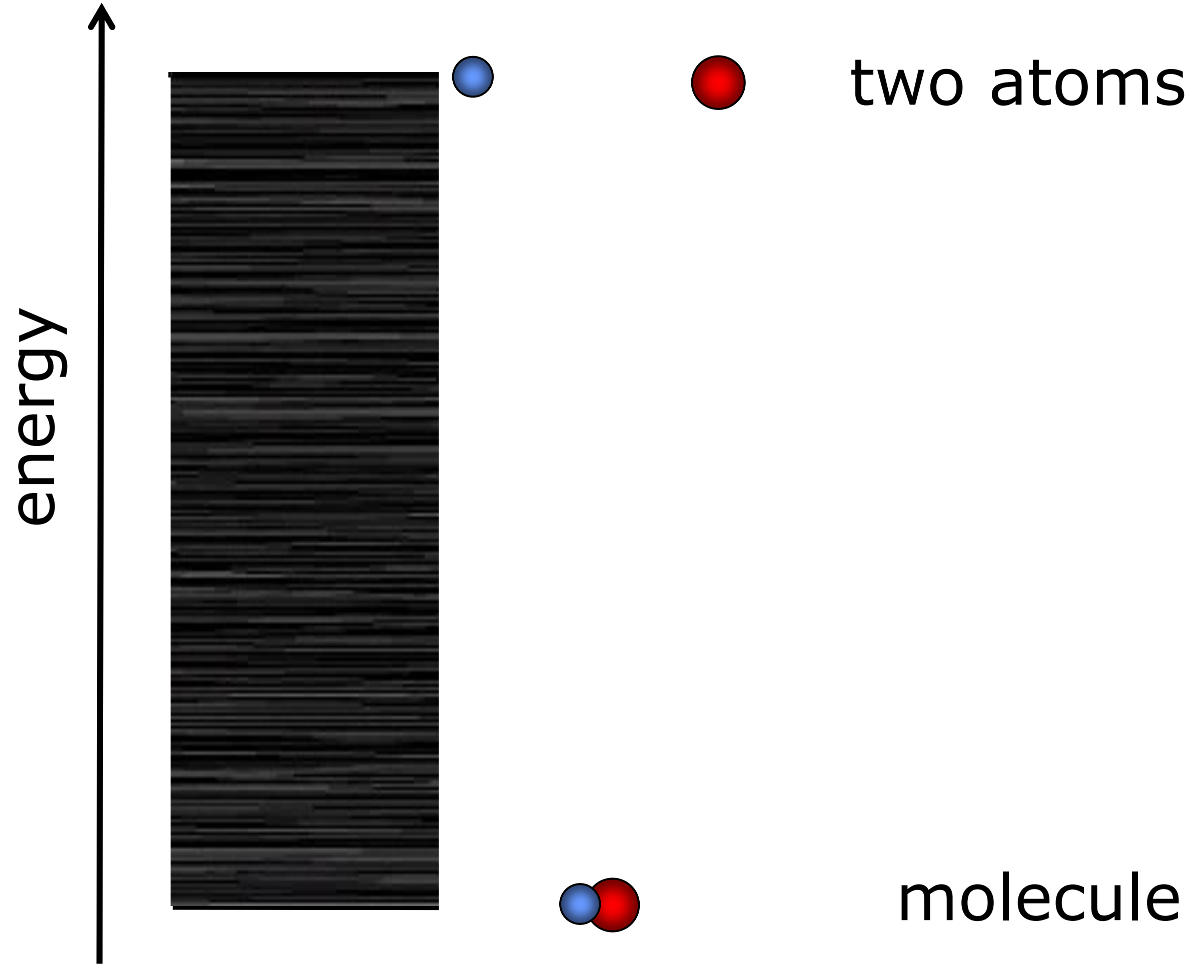


nuclear spin

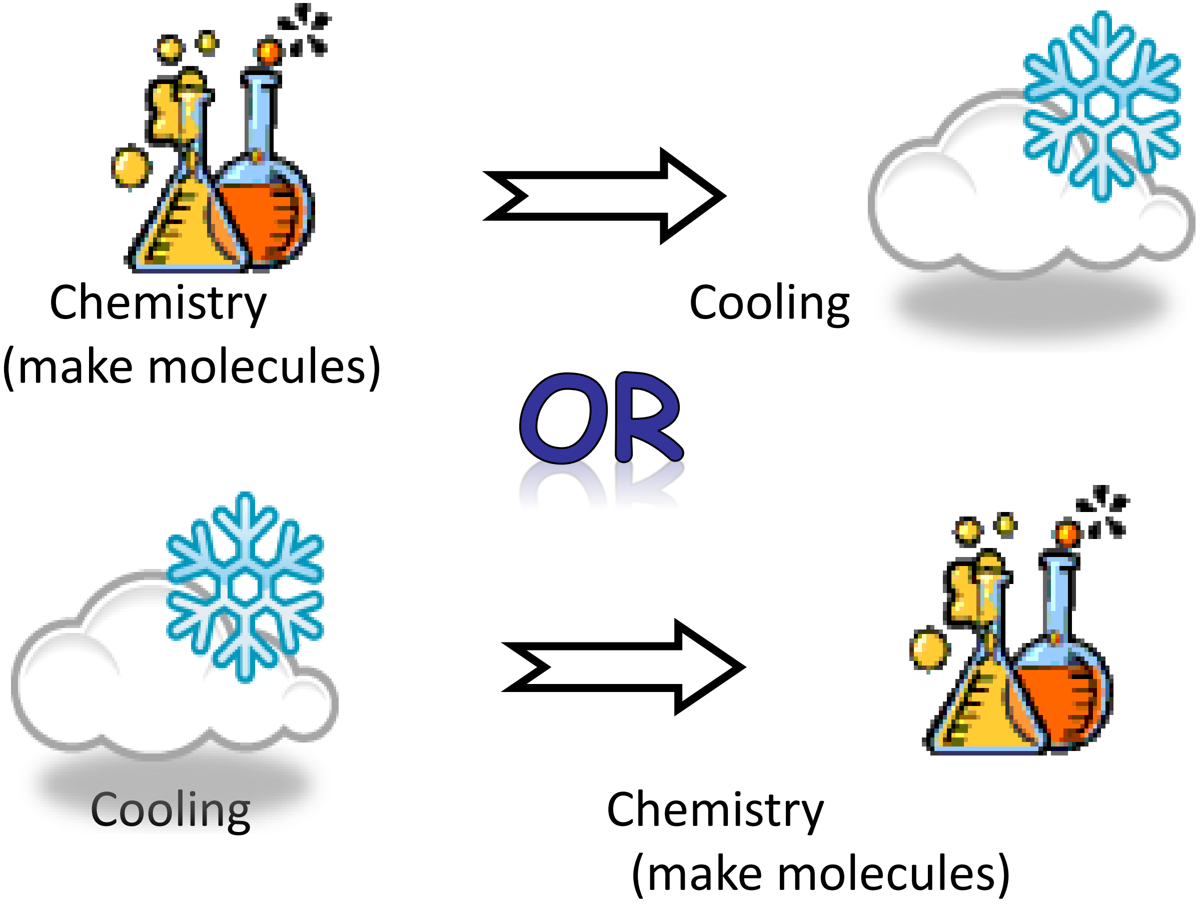
38 μK

Ultracold molecules: The challenge

Molecules are complex!



How do we cool molecules



Chemistry
(make molecules)

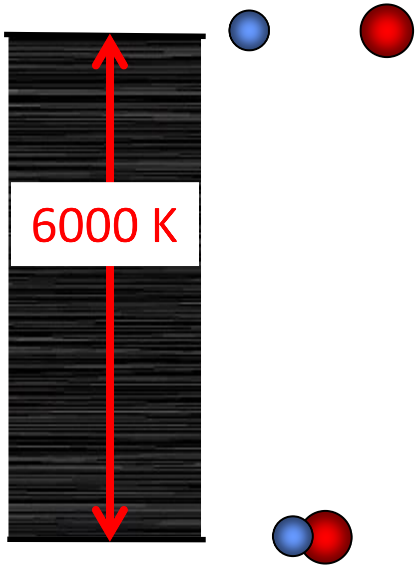
Cooling

OR

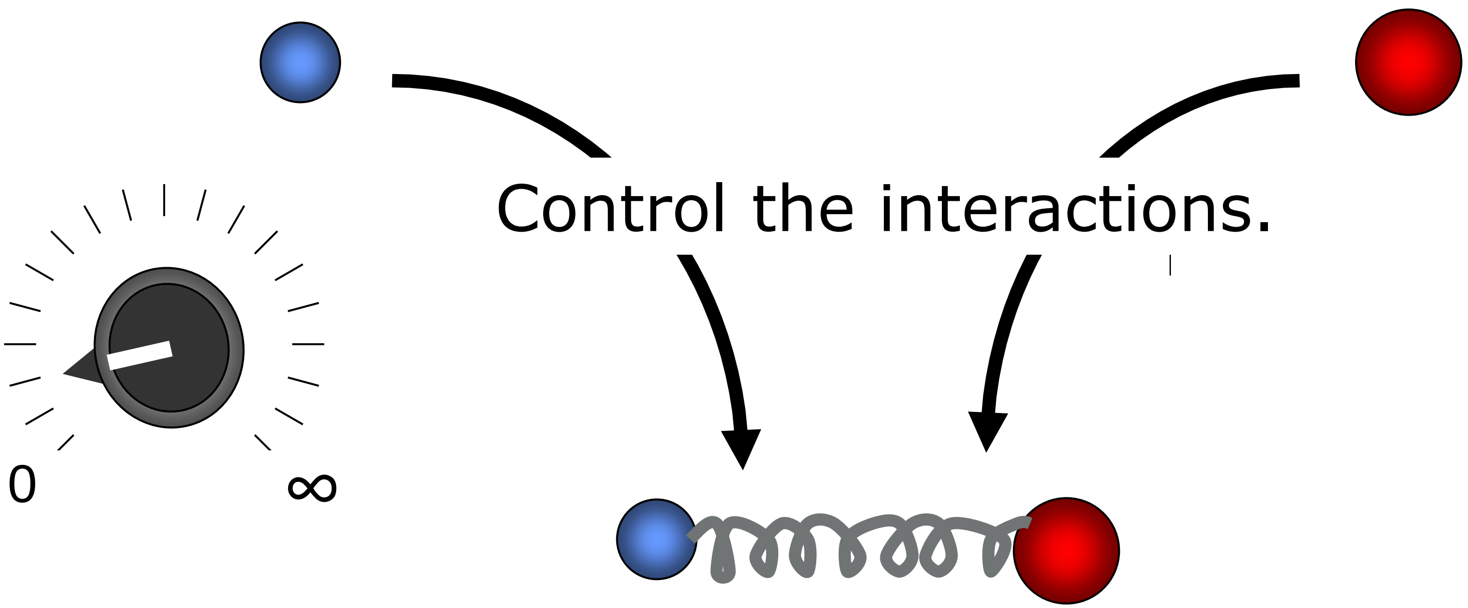
Cooling

Chemistry
(make molecules)

Buffer gas cooling
Stark deceleration
Laser cooling
Evaporative cooling



Make Feshbach molecules



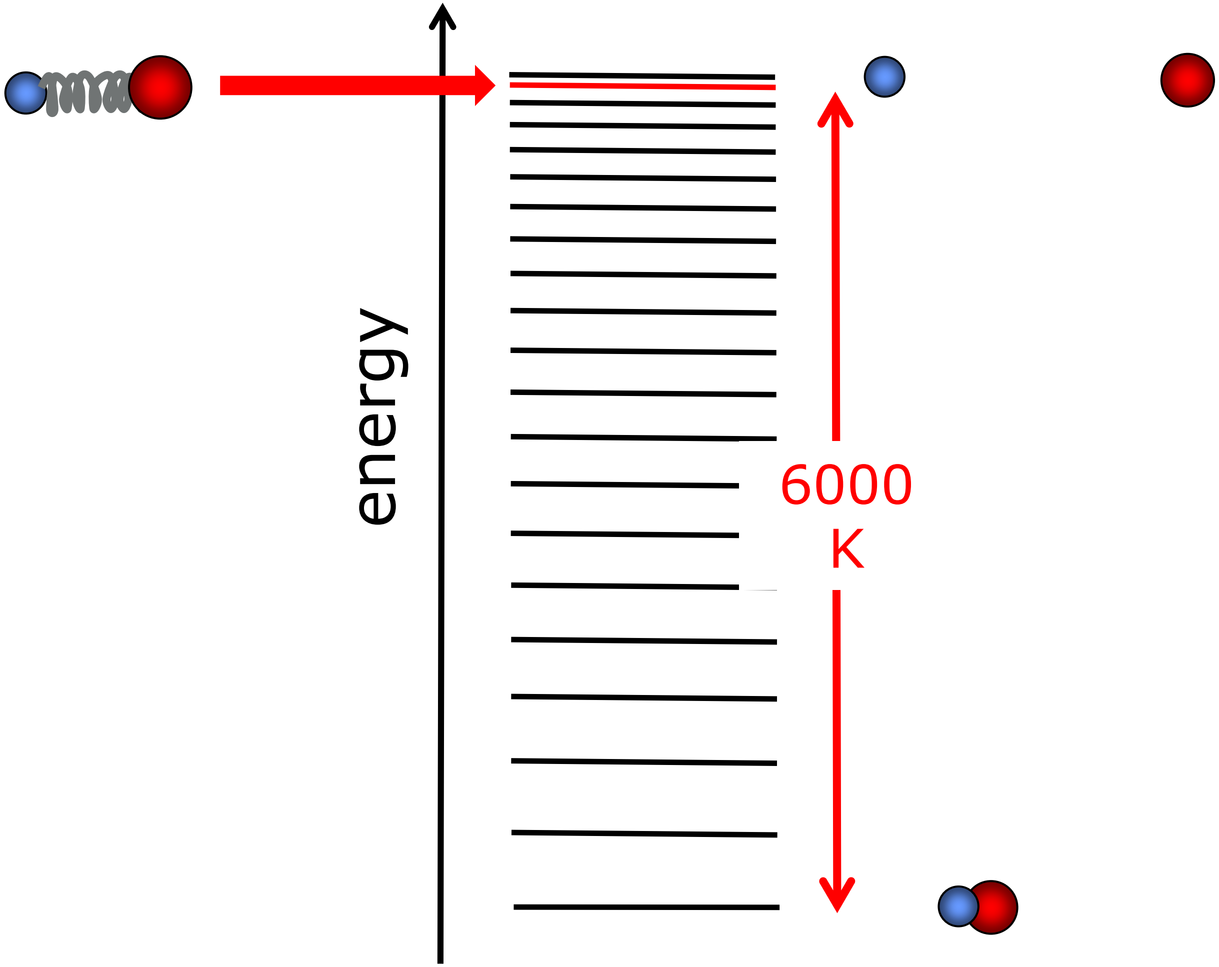
^{40}K Start with ultracold atoms. ^{87}Rb

Control the interactions.

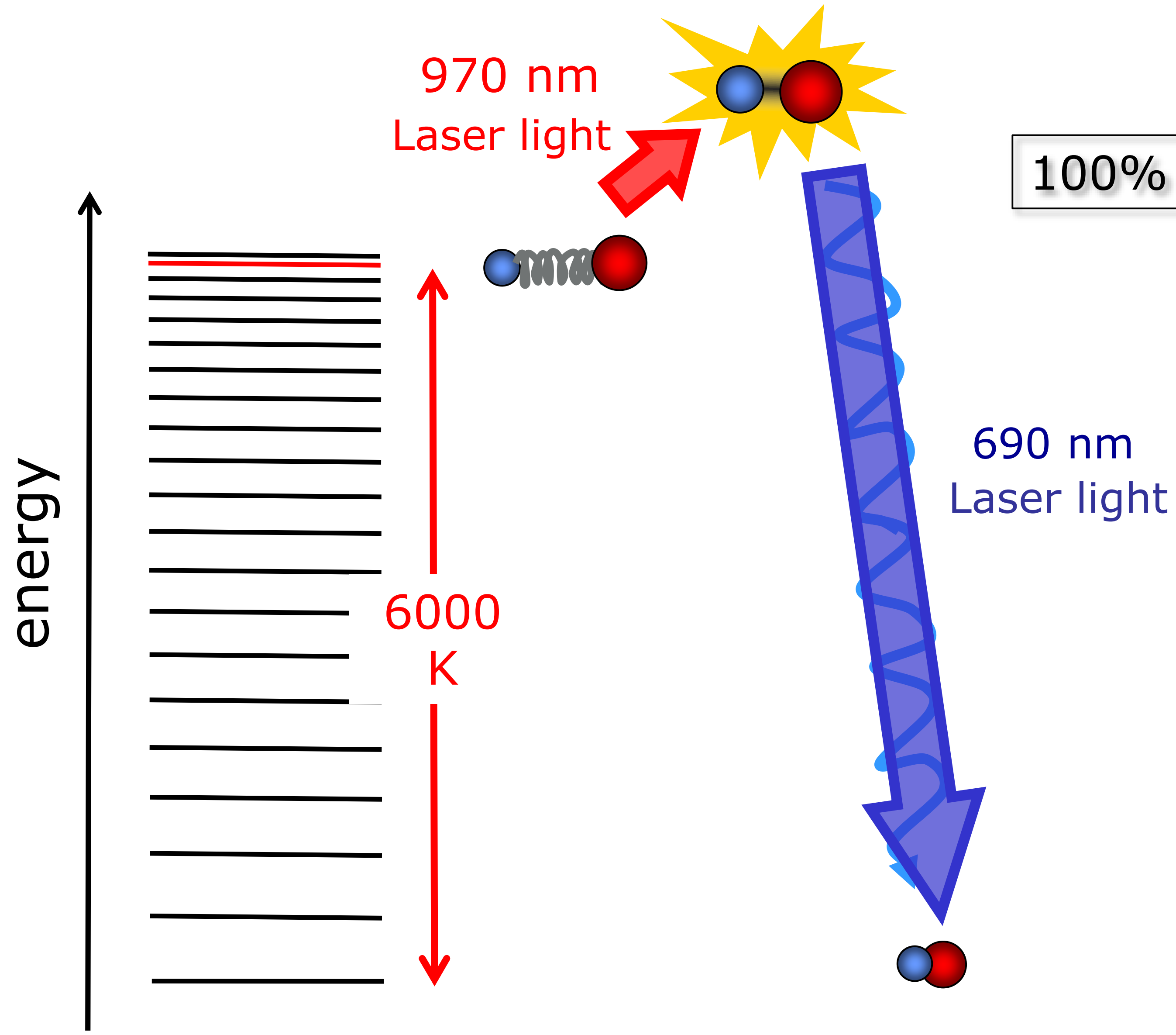
Make large, floppy molecules

Weakly bound molecules

- no dipole moment
- inelastic collisions



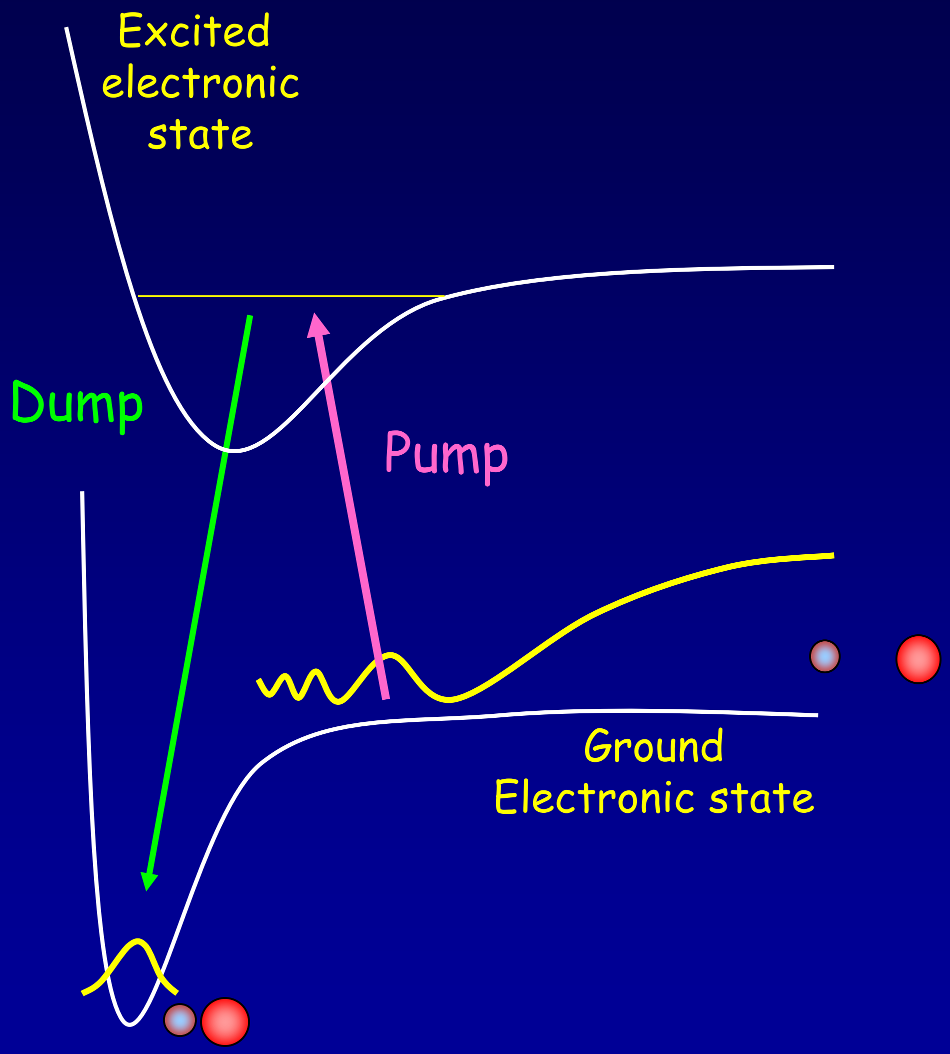
Transfer the molecules to the ground state



100% efficient ?

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

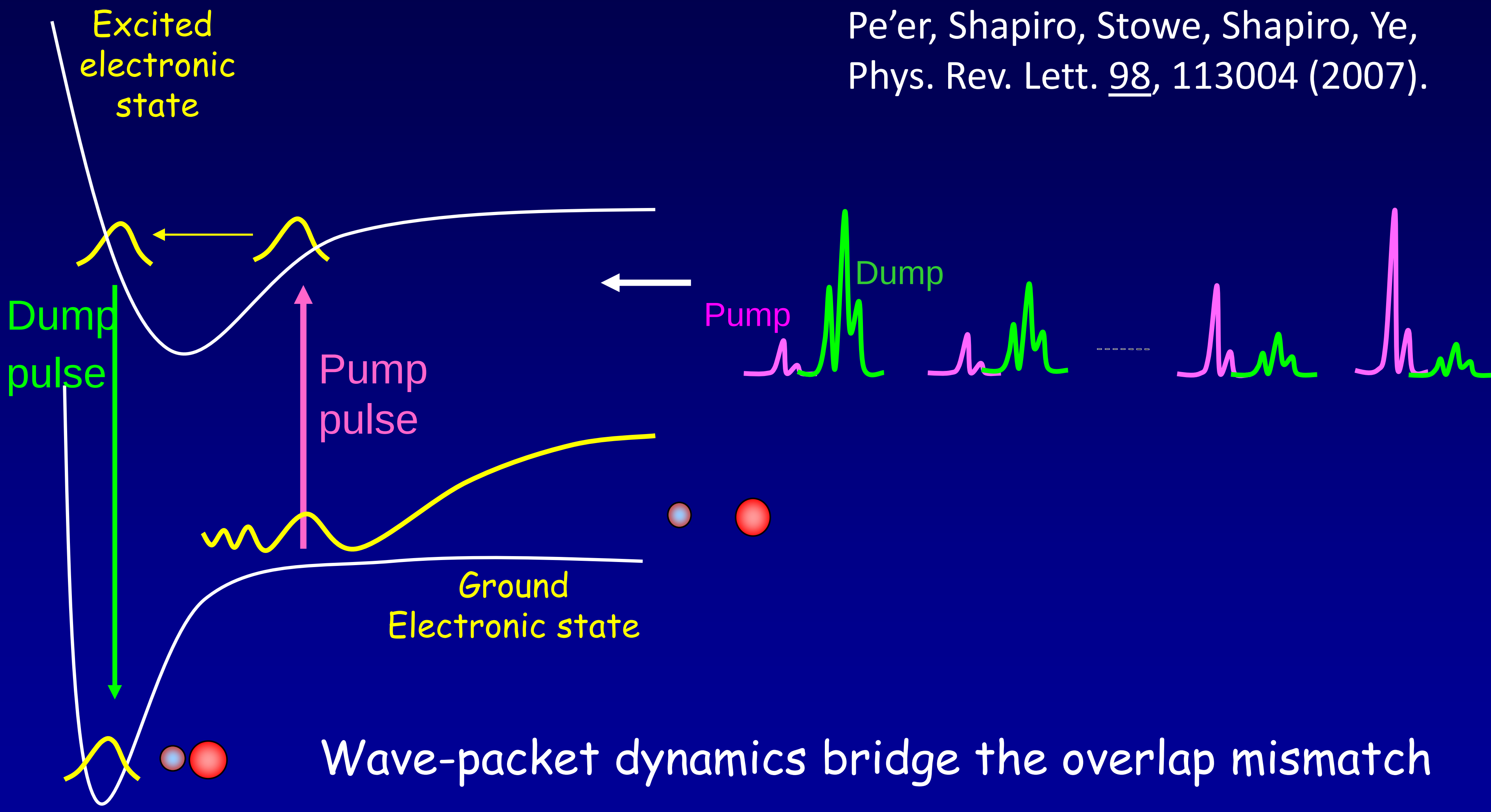
Going to really deep ground potential



The problem: ~ zero wavefunction overlap (pre 2008)

Ultracold meets ultrafast

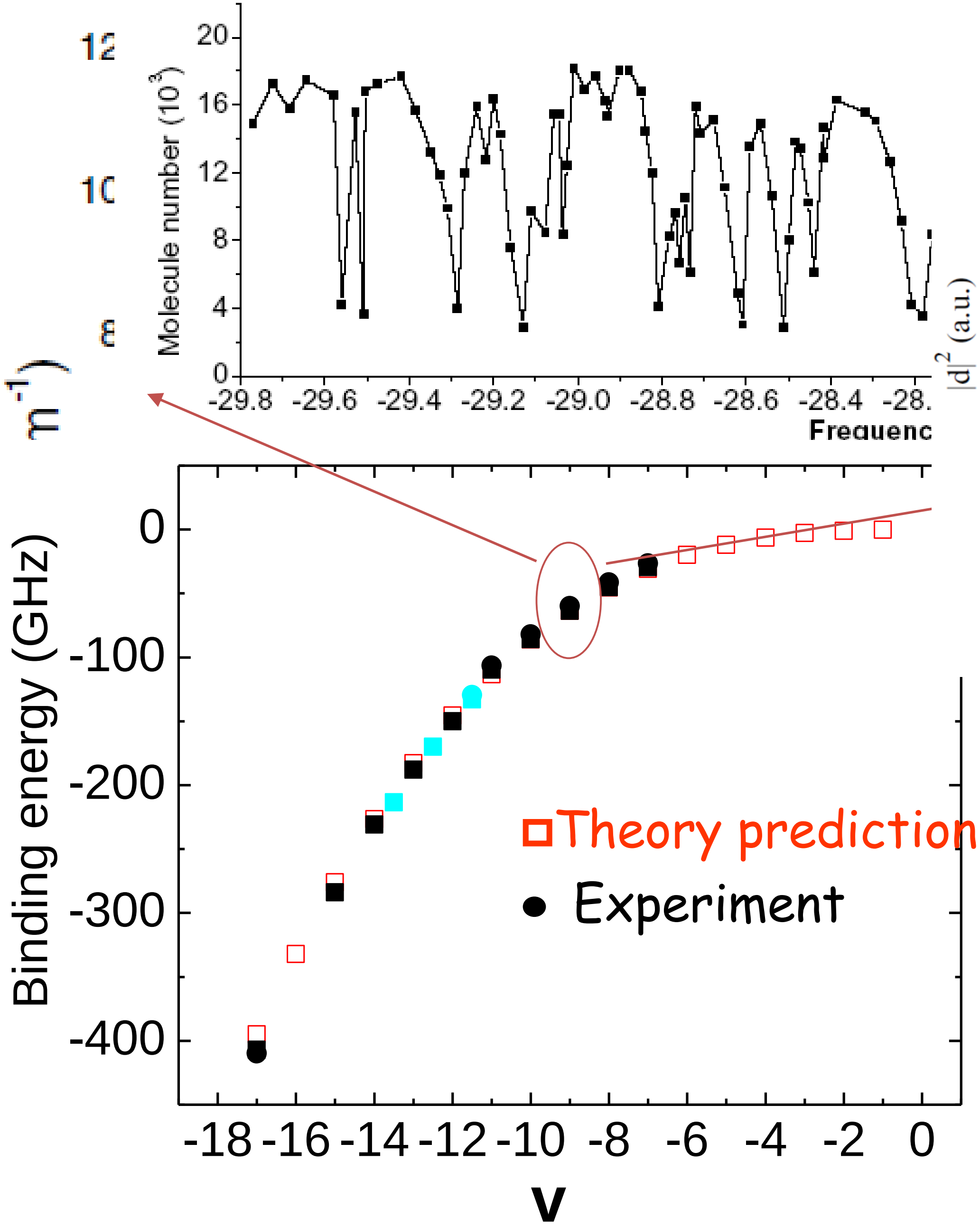
Pe’er, Shapiro, Stowe, Shapiro, Ye,
Phys. Rev. Lett. 98, 113004 (2007).

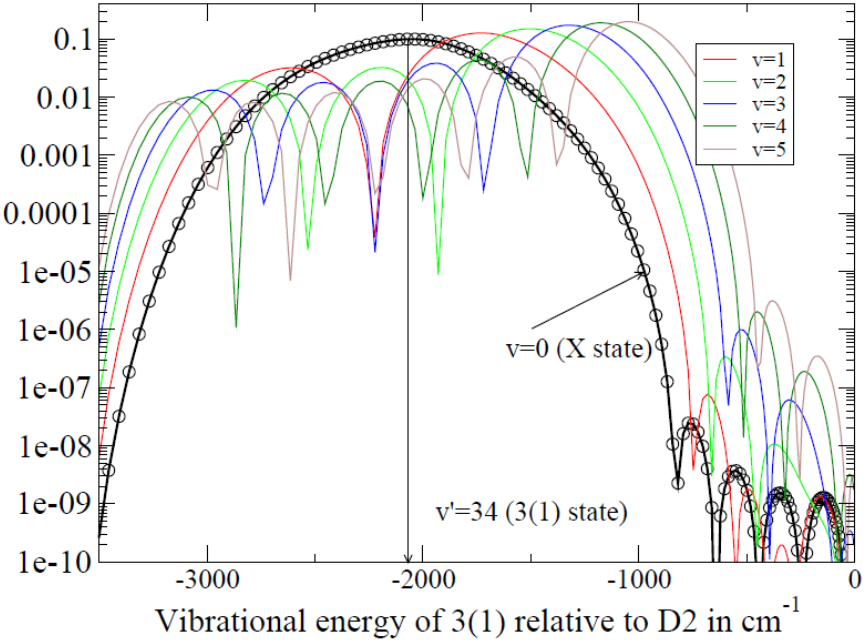


Wave-packet dynamics bridge the overlap mismatch

Coherent accumulations resolve single quantum state

Hiking through the forest of potentials





became clear

Good Franck-Condon for both up and down transitions.

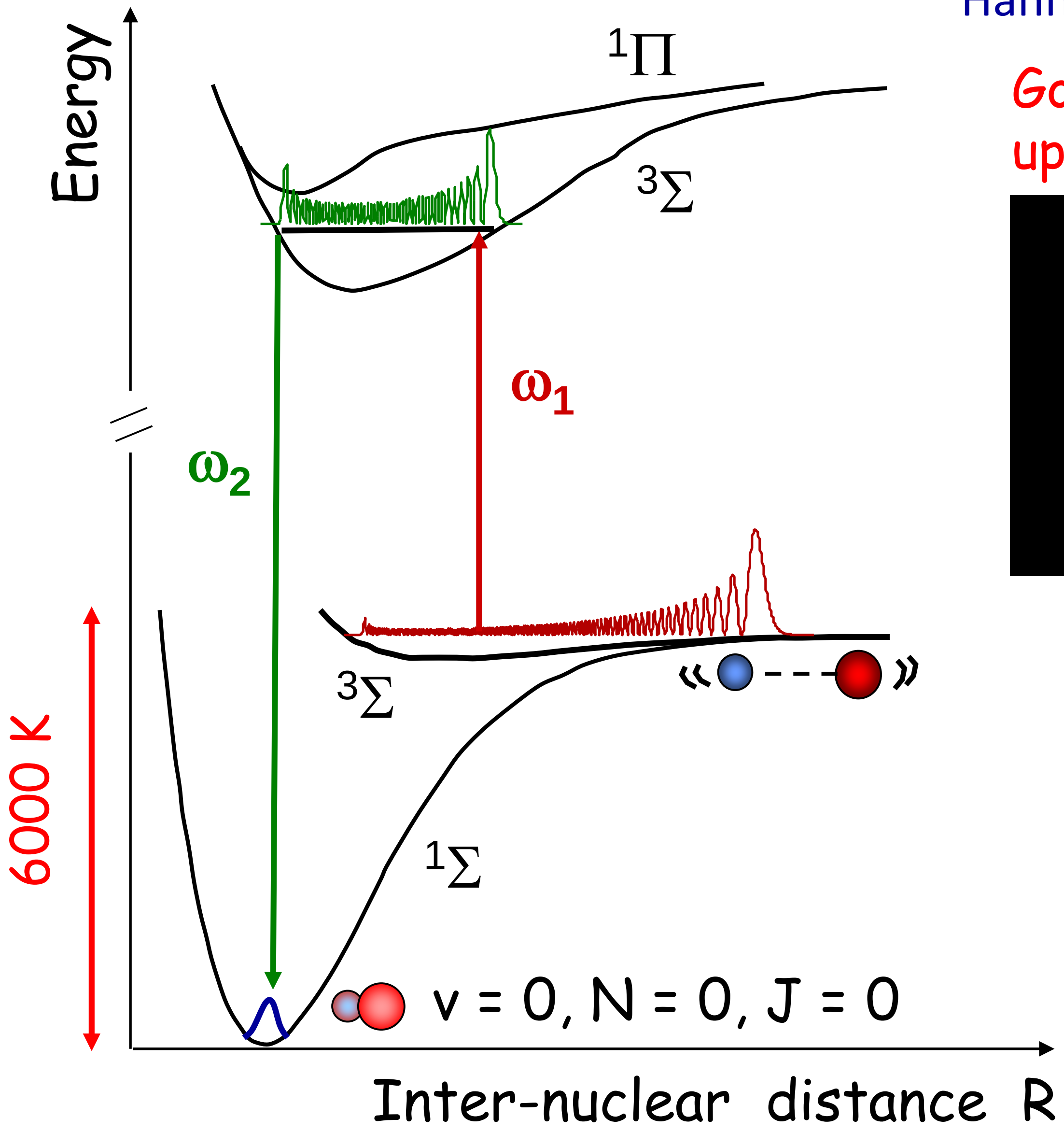
Julienne, Kotochigova

Tiemann, Bohn, Hutson, …

Coherent two-photon transfer

Ospelkaus *et al.*, Nature Phys. **4**, 622 (2008); Ni *et al.*, Science **322**, 231 (2008).

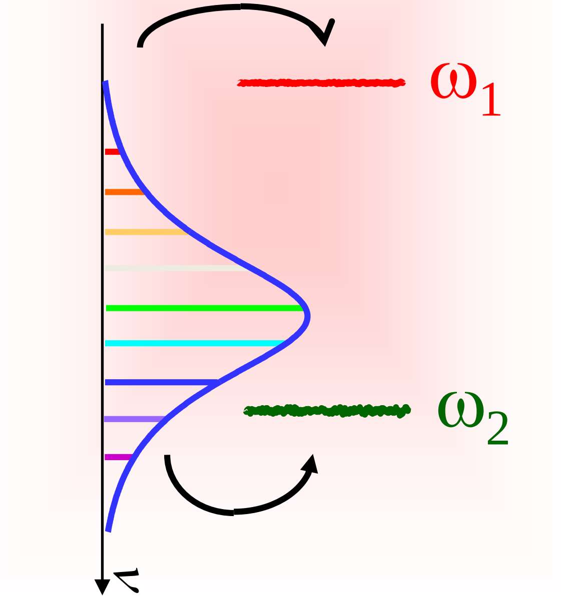
Hanns-Christoph Nägerl, Innsbruck, Cs_2



Good Franck-Condon for both up and down transitions.

Population transfer

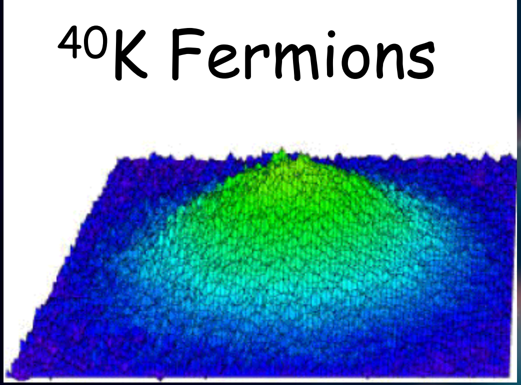
- 92% efficiency
- Fully coherent (no heating)

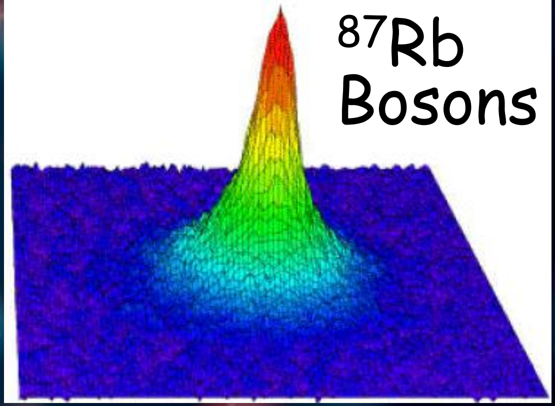


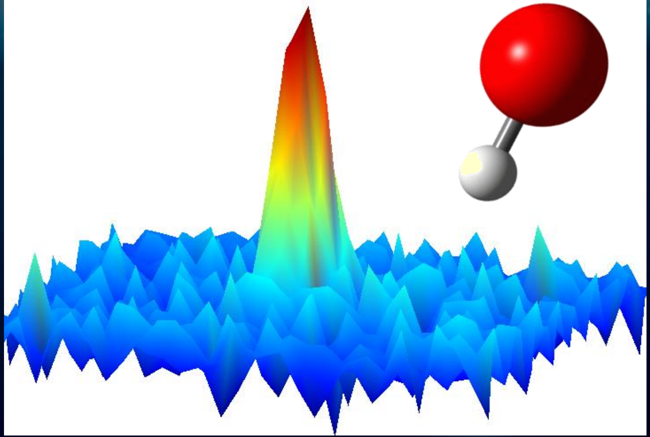
Quantum gas of polar molecules

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

- Temperature ~ 120 nK
- $T/T_F = 1.0$
- Density ~$10^{12}/cm^3$



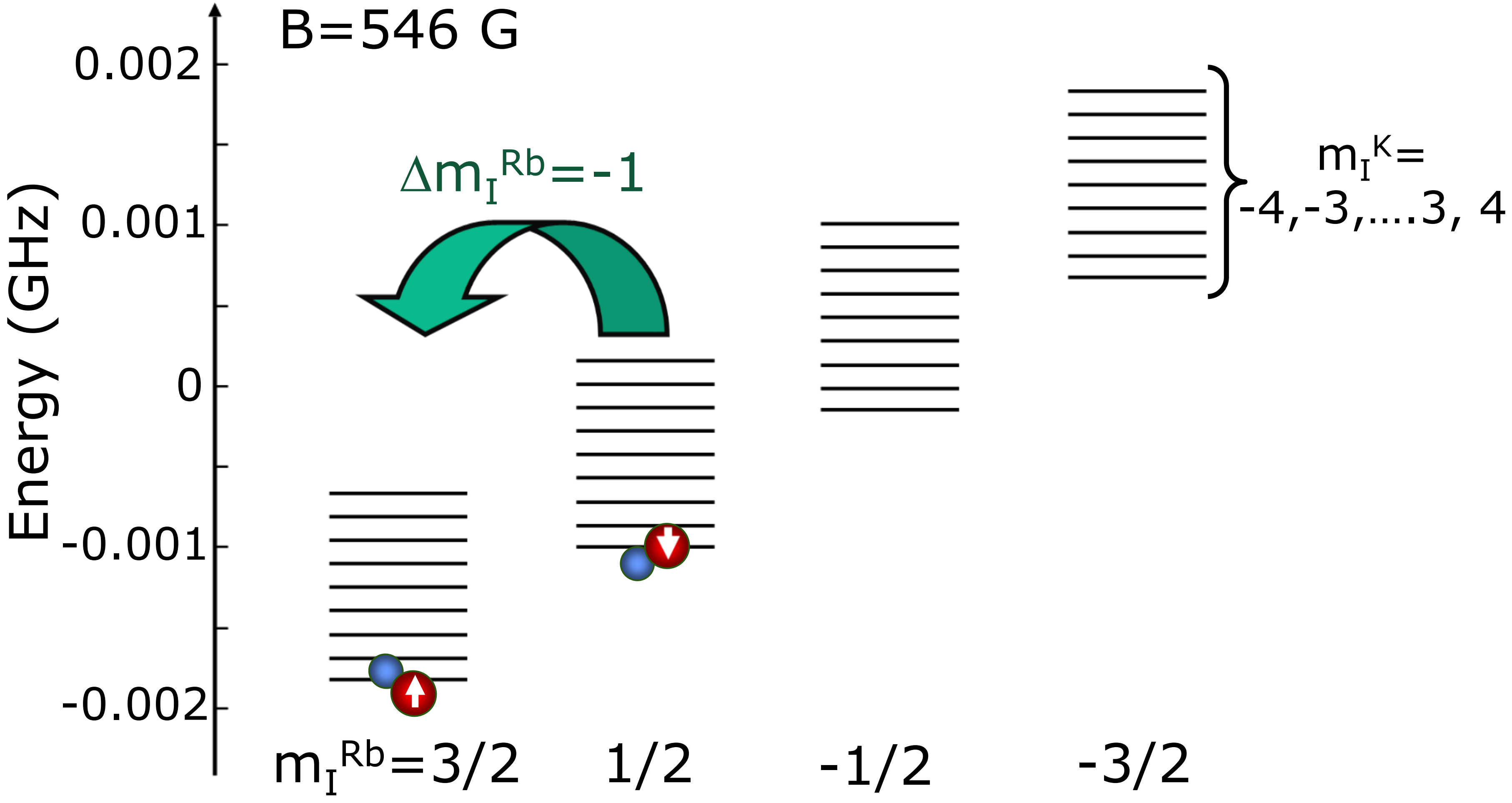




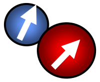
KRb molecules (Dipole ~0.5 Debye)

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

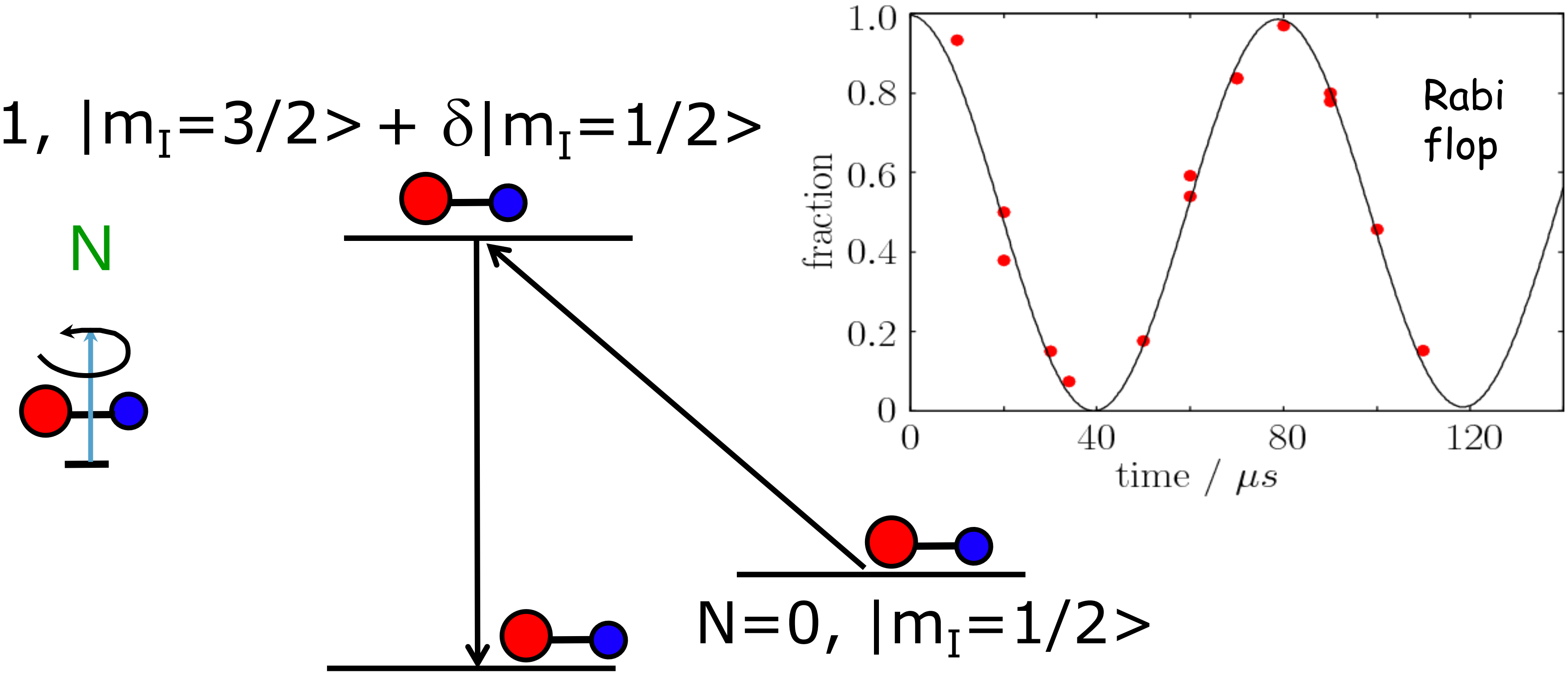
$I^{Rb}=3/2$ $I^{K}=4$ 36 states



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



$I^{Rb}=3/2$ $I^{K}=4$ **36 states**



N=1, $|m_I=3/2\rangle + \delta|m_I=1/2\rangle$

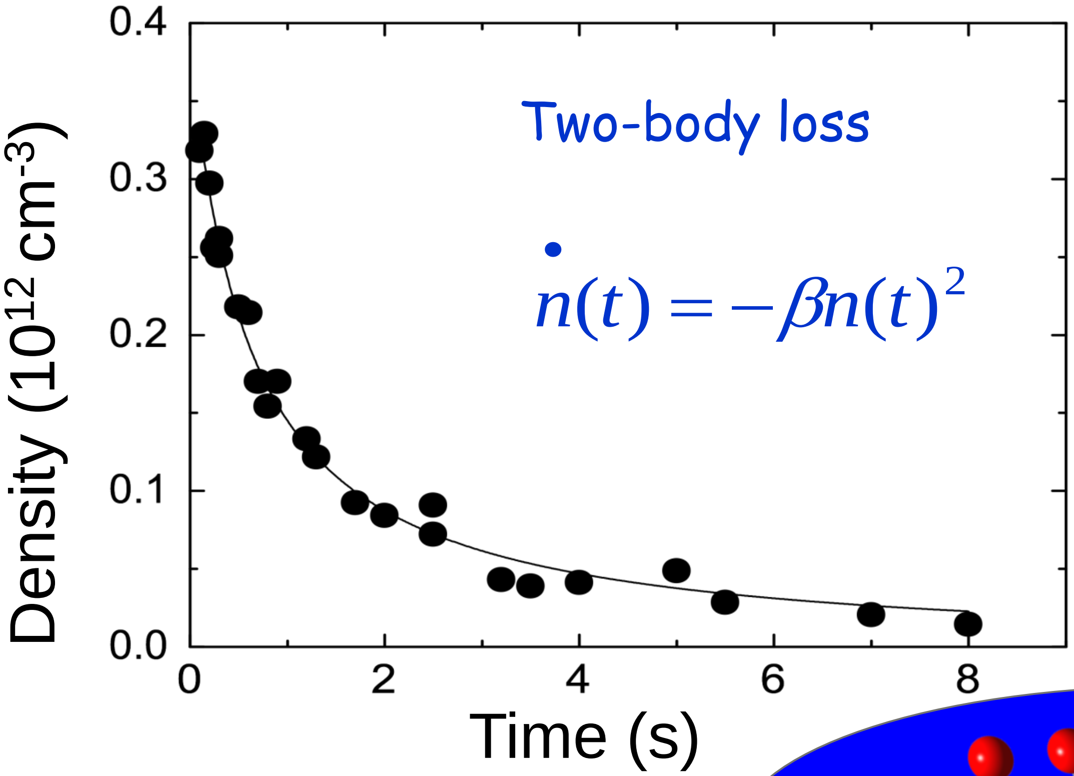
N

N=0, $|m_I=1/2\rangle$

N=0, $|m_I=3/2\rangle$

S. Ospelkaus *et al.*, Phys. Rev. Lett. **104**, 030402 (2010).

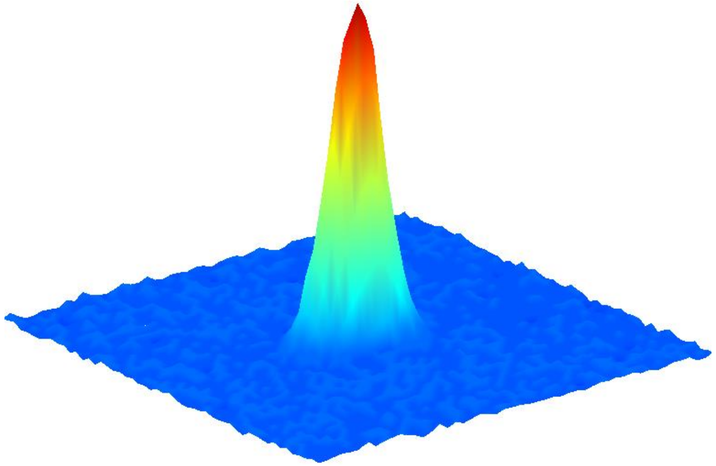
Trapped molecules in the lowest energy state (electronic, vibrational, rotational, hyperfine)

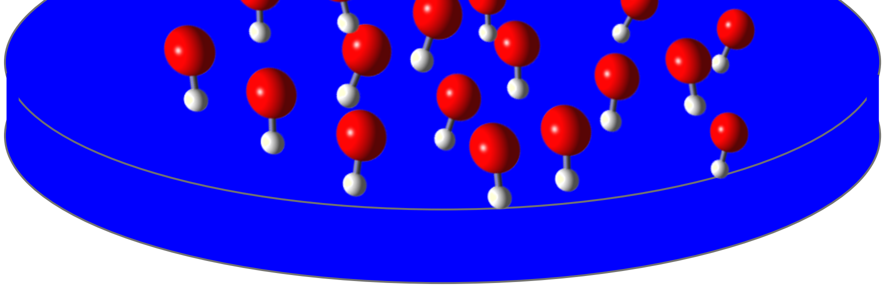


Two-body loss

$$\dot{n}(t) = -\beta n(t)^2$$

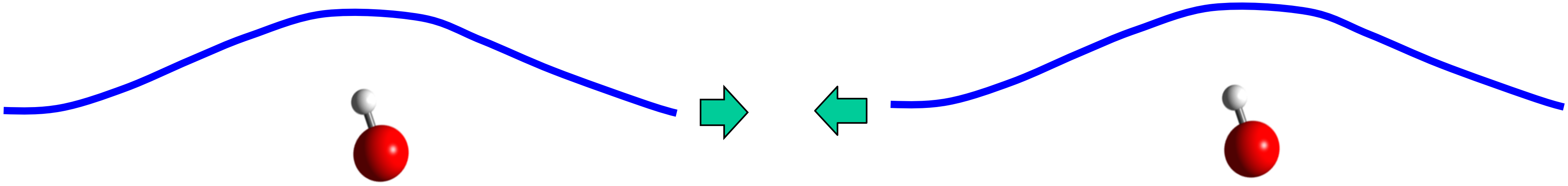
T = 200 nK
> 1 s lifetime



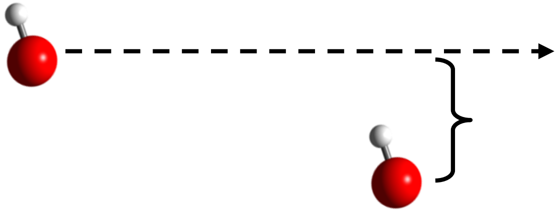


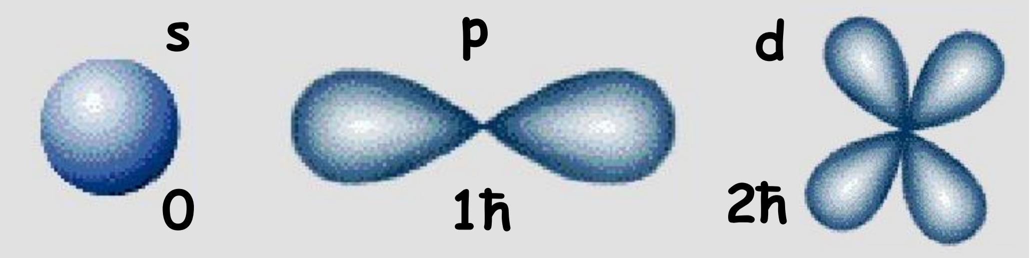
Cold collisions between identical Fermions

(1) Particles behave like waves

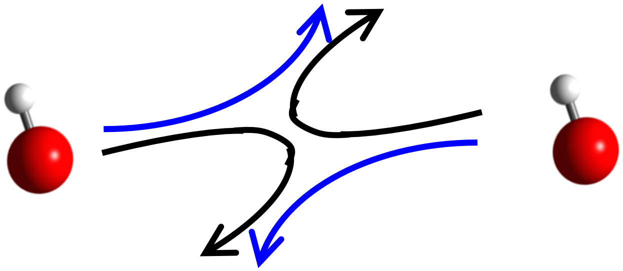


(2) Angular momentum is quantized

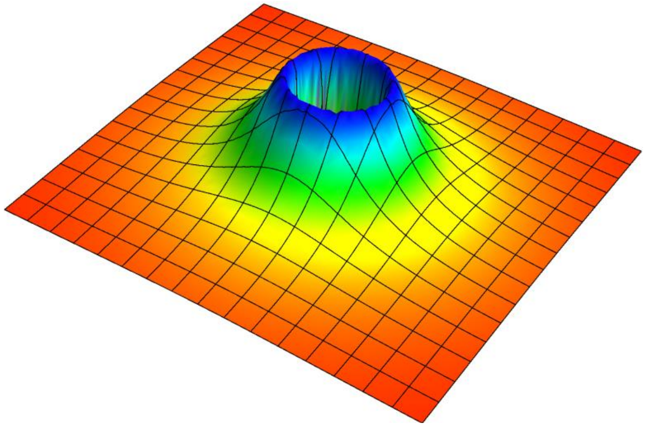




(3) Quantum statistics matter

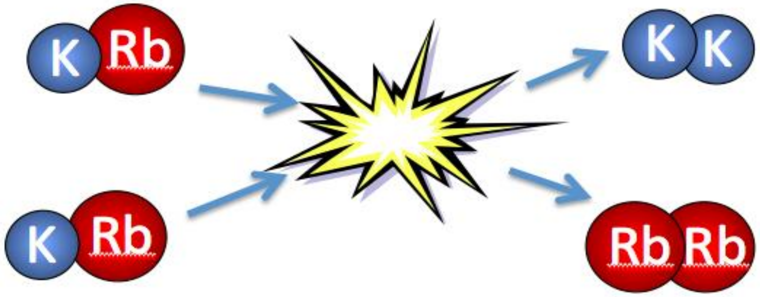


$$|\psi_0\rangle|\psi_1\rangle - |\psi_1\rangle|\psi_0\rangle$$



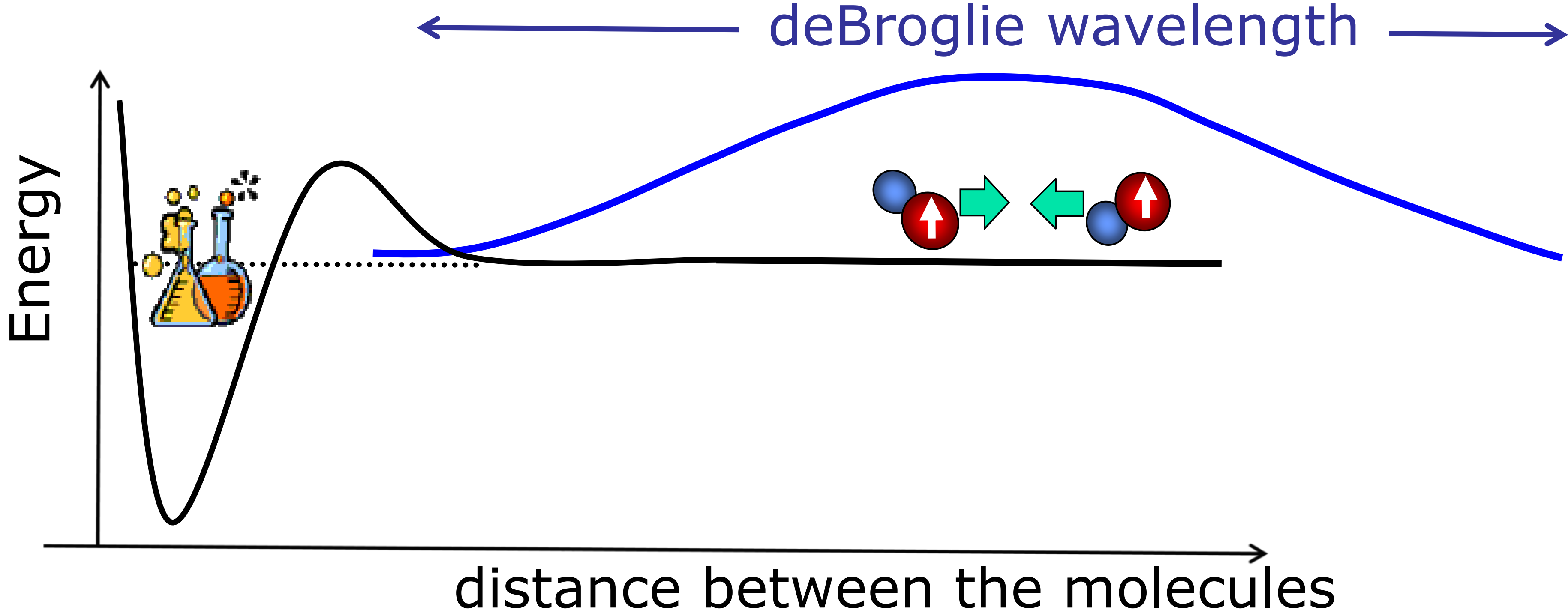
Fermions ⇨ $L = 1$, *p*-wave collisions

Ultracold quantum chemistry

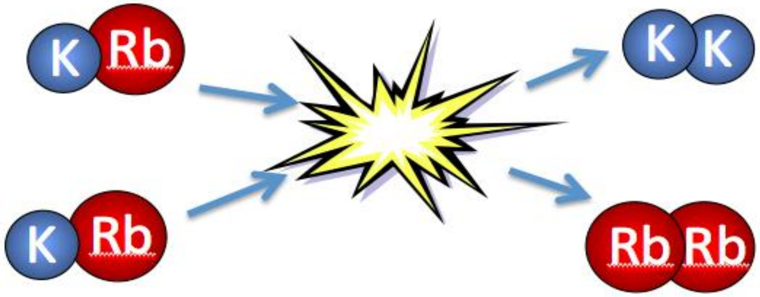


Ospelkaus *et al.*,
Science **327**, 853 (2010).

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

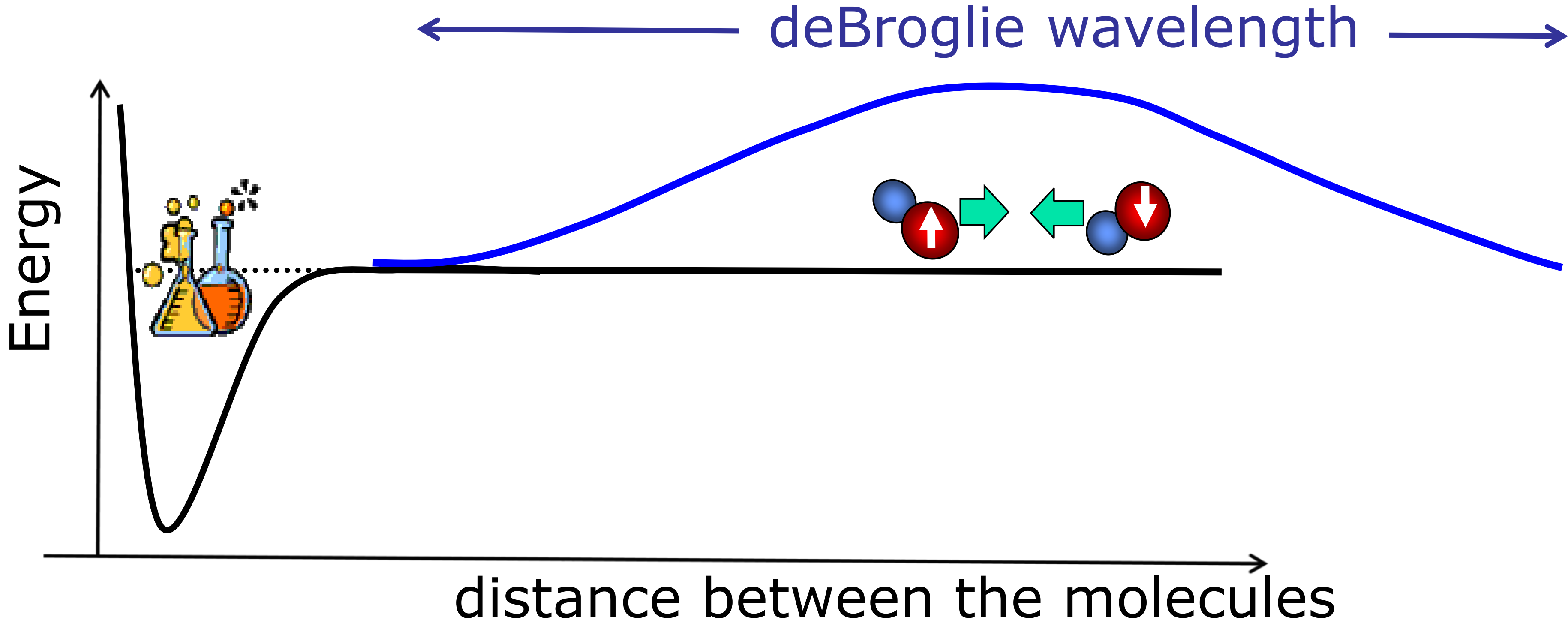


Ultracold quantum chemistry

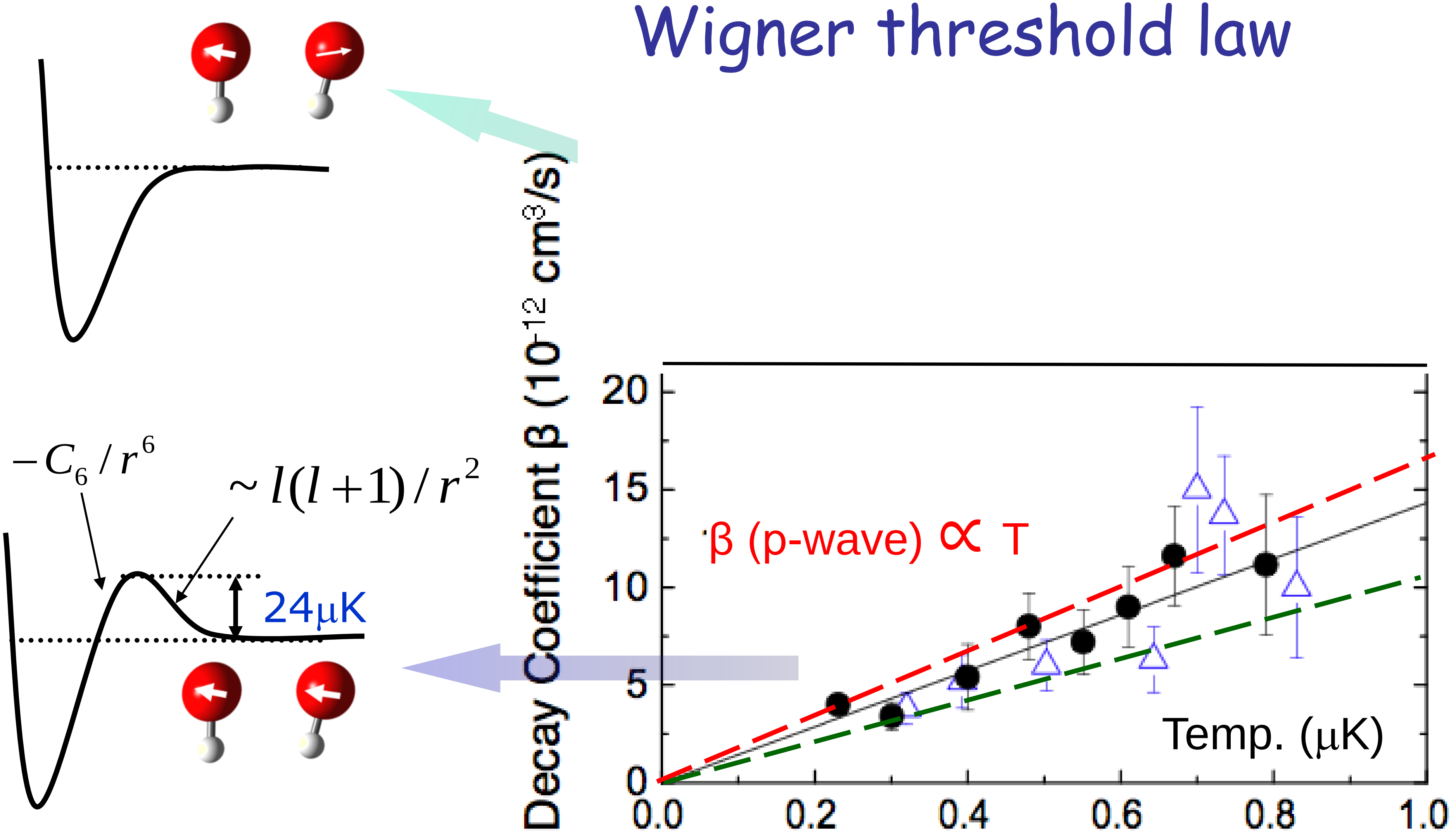


Ospelkaus *et al.*,
Science **327**, 853 (2010).

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



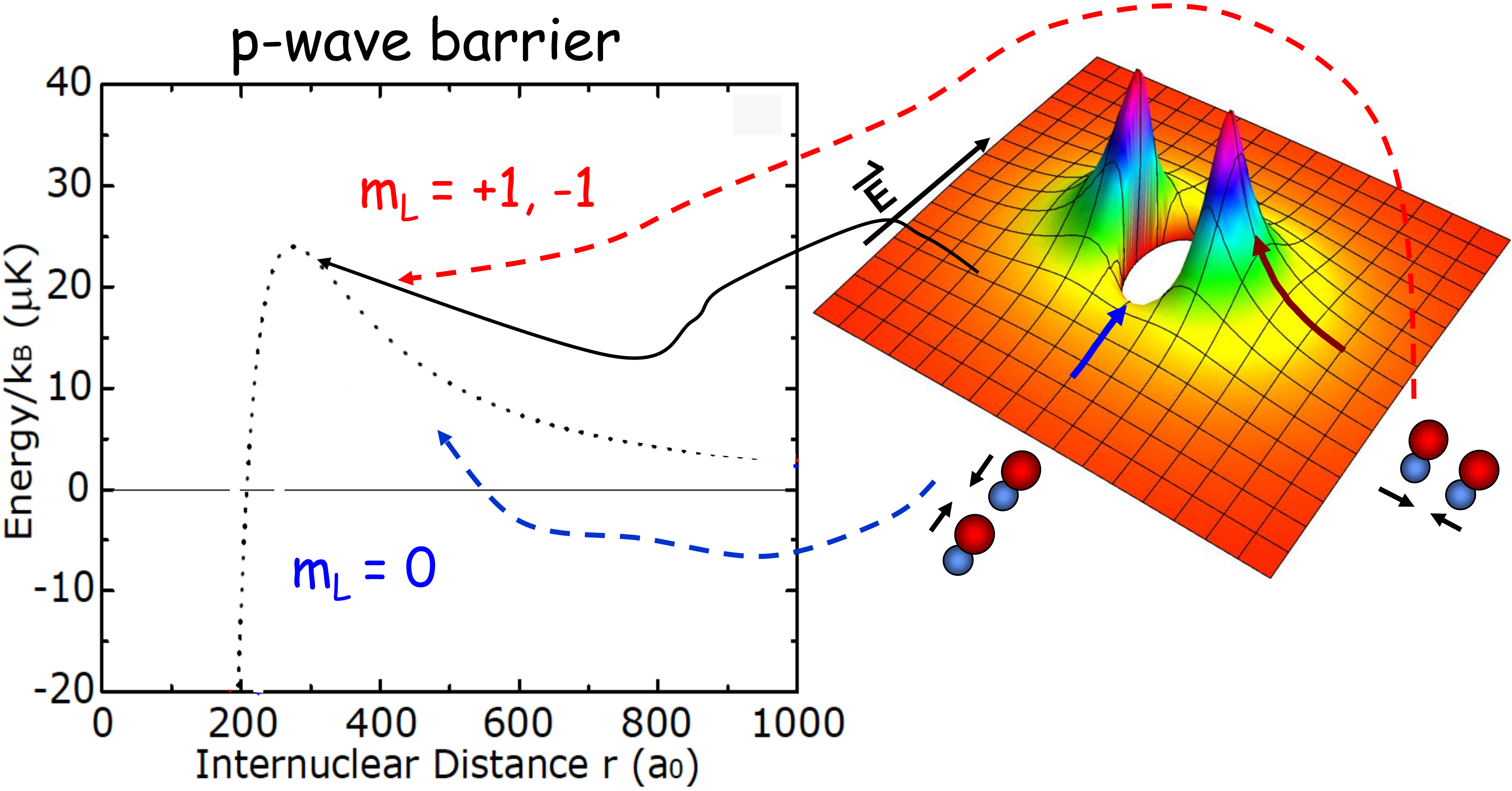
Bimolecular reactions under Wigner threshold law



β (p)= 1.2(3)x10^{-5}cm^3/(s K) T

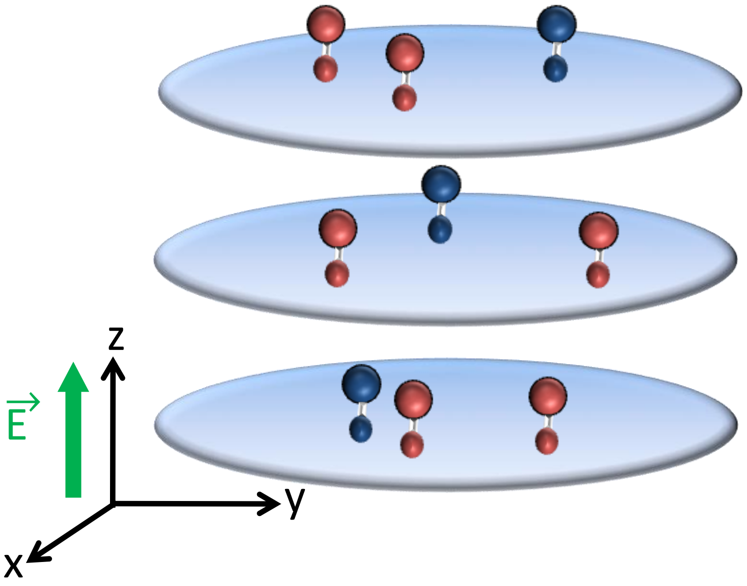
Anisotropic dipolar collisions

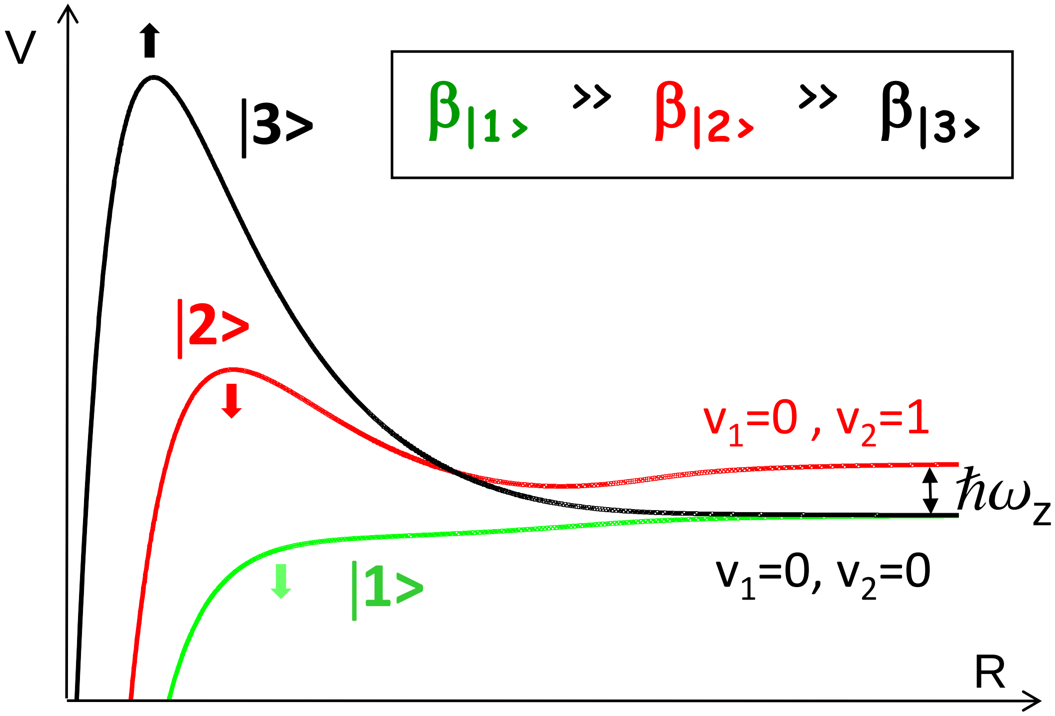
K.-K. Ni *et al.*, Nature **464**, 1324 (2010).



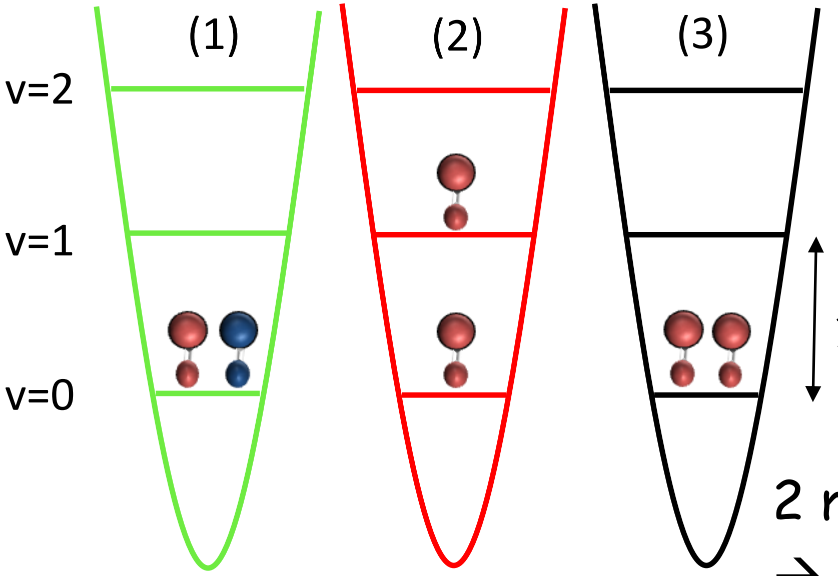
Collisions under a single partial wave ($L = 1\hbar$).

2D quantum gas - suppress losses





$\beta_{|1>} \gg \beta_{|2>} \gg \beta_{|3>}$

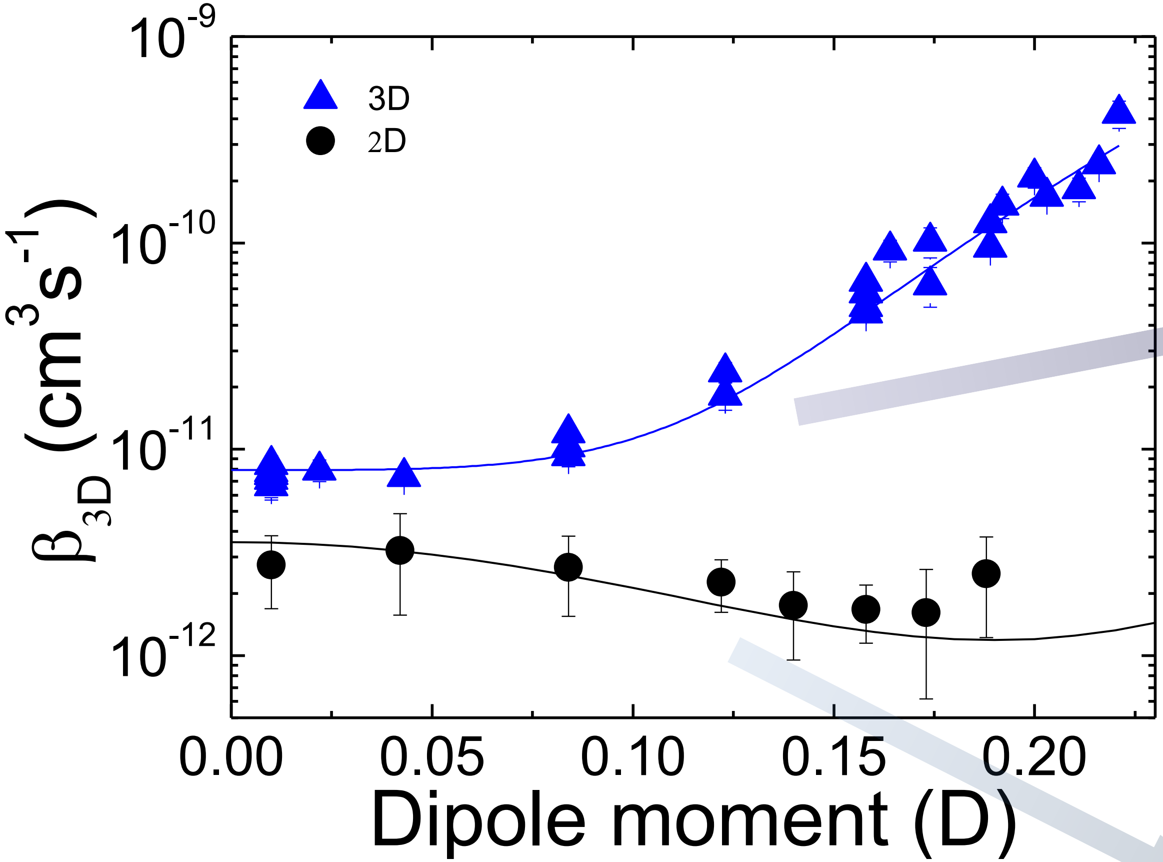


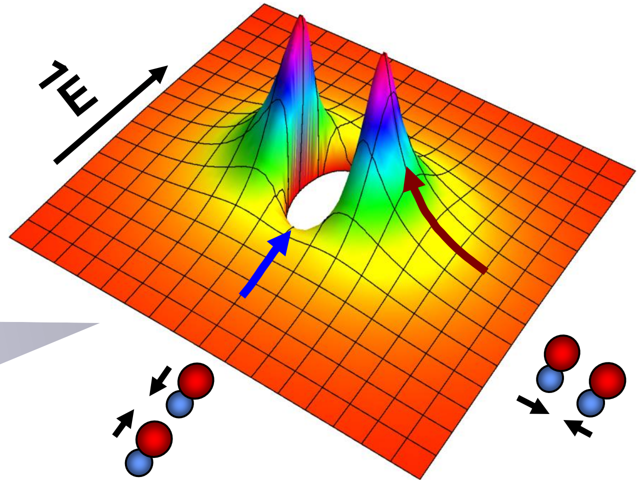
Quantized stereo-dynamics of chemical reactions

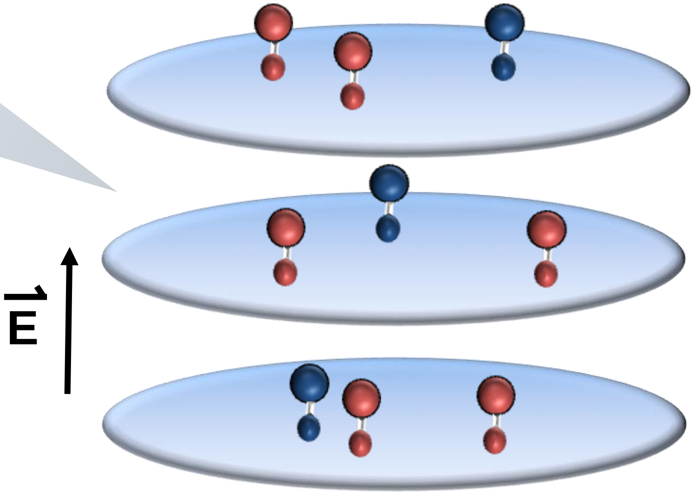
2 molecules in the same internal & |v> states

→ $|L = 1, m_L = \pm 1>$

2D quantum gas - suppress losses





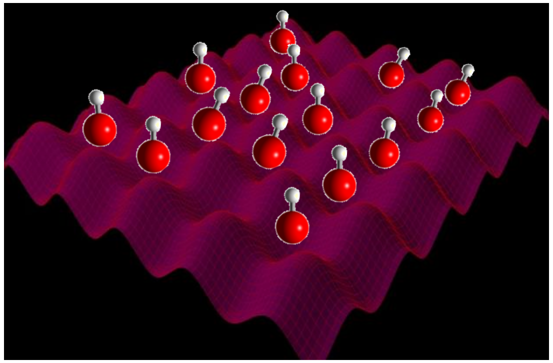


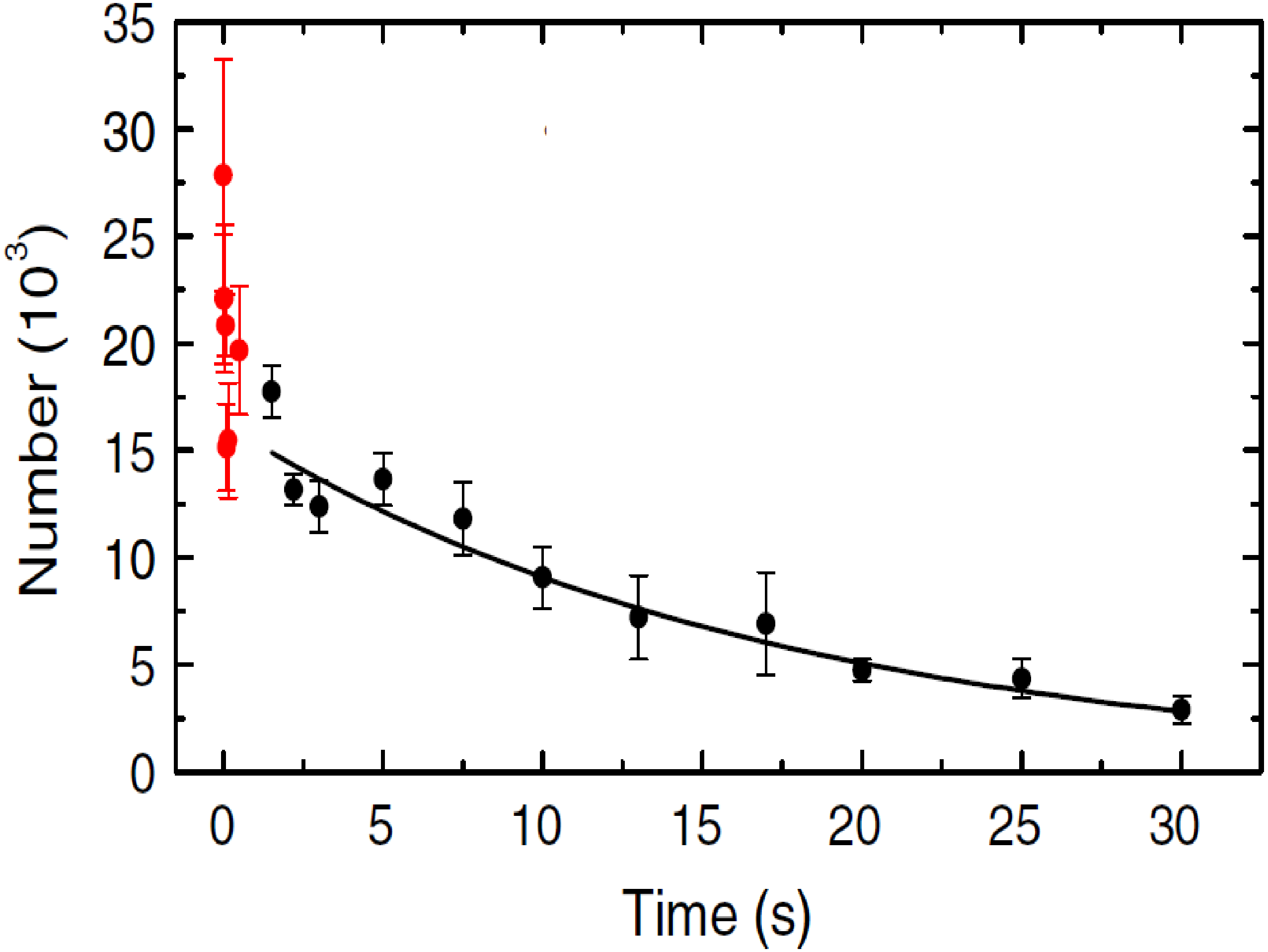
Theory: Büchler, Zoller, Bohn, Julienne

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

3D Optical Lattice – ruins all that beautiful chemistry

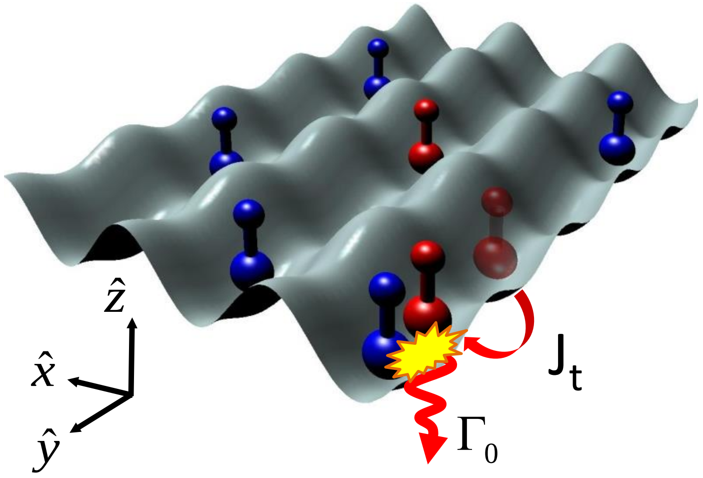
Lifetime ~ 20 s





A. Chotia *et al.*, "Long-Lived Dipolar Molecules and Feshbach Molecules in a 3D Optical Lattice", Phys. Rev. Lett. **108**, 080405 (2012).

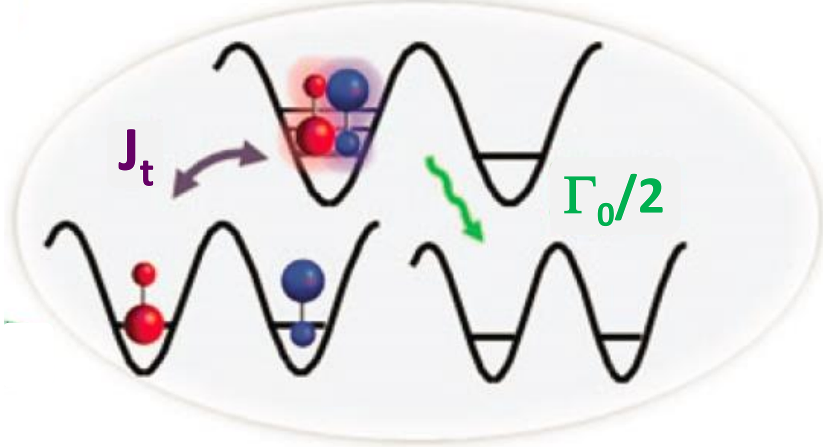
Characterize filling factor in 3D lattice



Continuous Quantum Zeno suppression

- *Create an incoherent spin mixture.*
- *Lower lattice along **y** to allow tunneling.*
- *Measure the loss rate.*

$$\frac{dN_\downarrow}{dt} = -\frac{\kappa N_\downarrow^2}{N_{\downarrow,0}} \qquad \kappa = 8\Gamma_{\mathrm{eff}} n_0$$



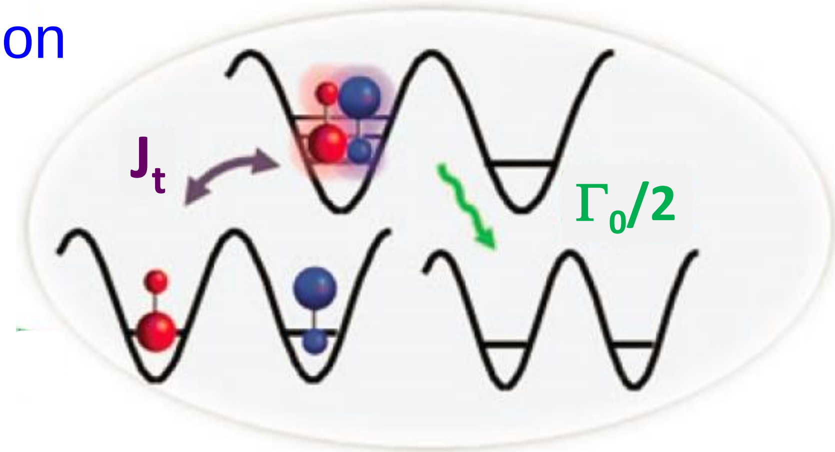
$$\Gamma_0 \gg J_\mathrm{t} \quad \boxed{\Rightarrow \Gamma_{\mathrm{eff}} = \frac{2J_\mathrm{t}^2}{\Gamma_0}}$$

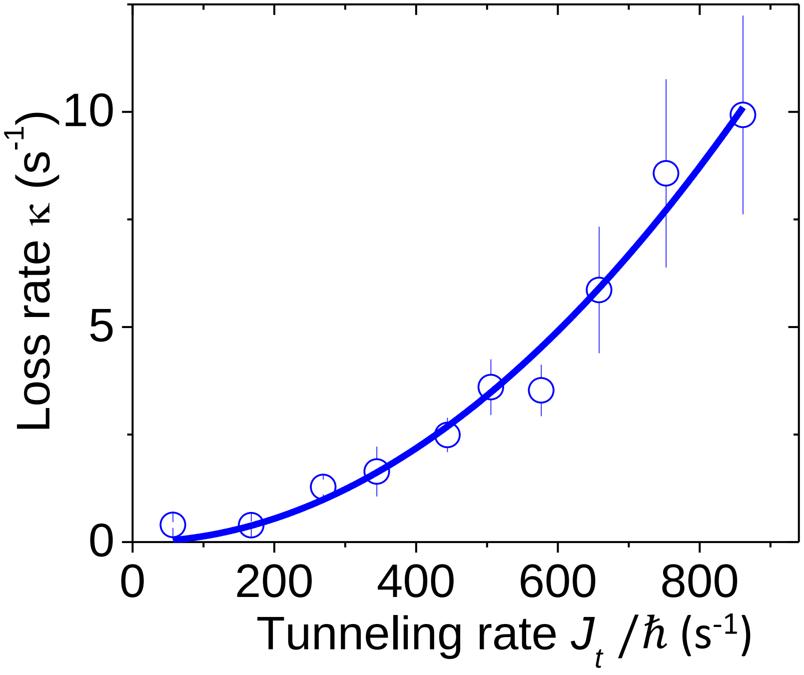
Syassen *et al.*, Science 320, 1329 (2008).

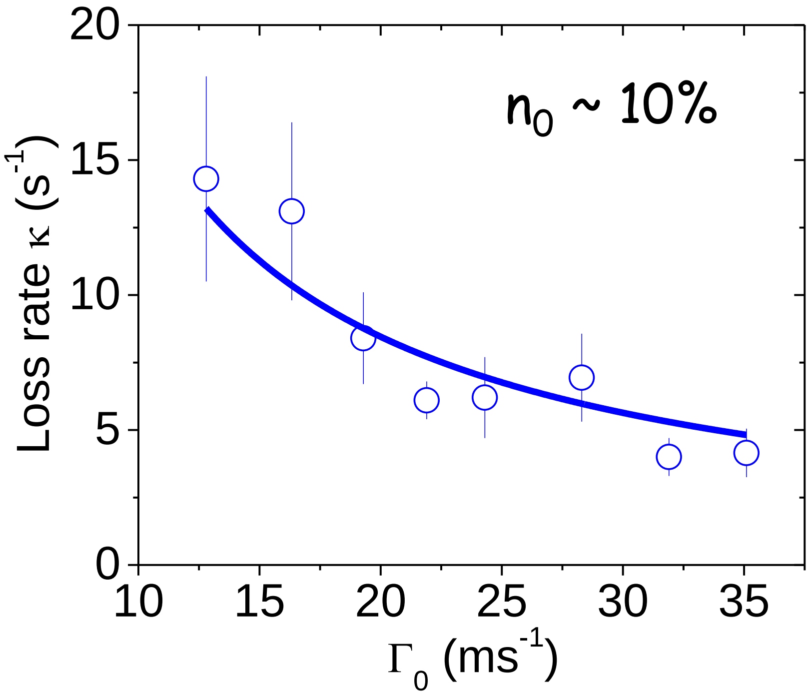
Characterize filling factor in 3D lattice

Continuous Quantum Zeno suppression

Loss ~ J_t^2/Γ_0

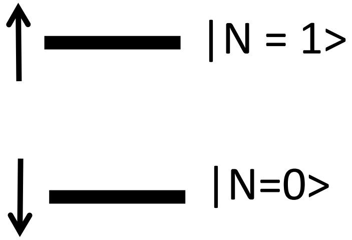




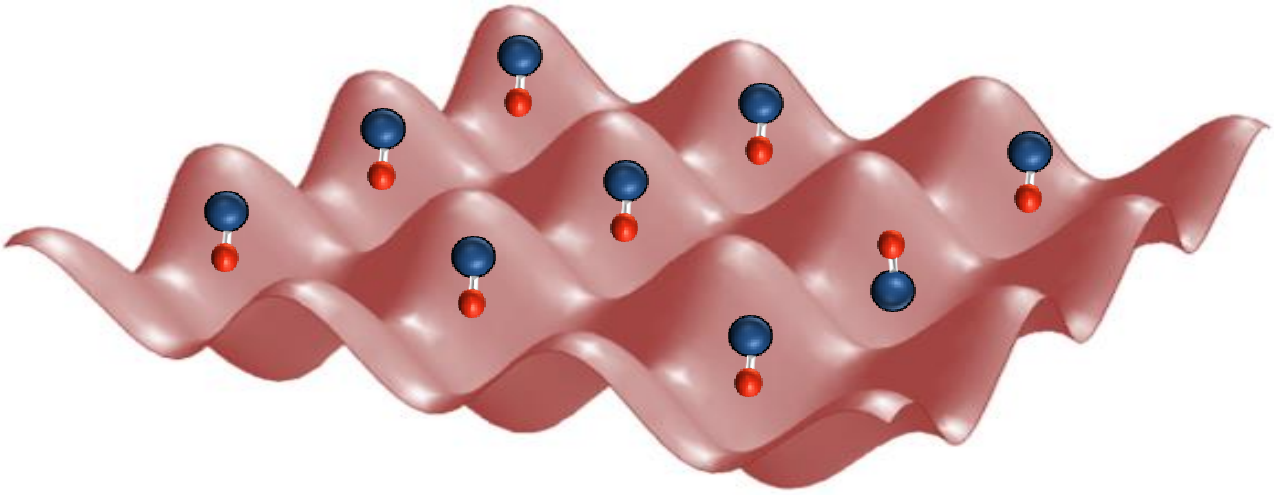


Dipolar exchange interaction in a 3D lattice

Barnett, Petrov, Lukin, Demler, Phys. Rev. Lett. **96**, 190401 (2006).
Micheli, Brennen, & Zoller, Nature Phys. **2**, 341 (2006).
Gorshkov *et al*., Phys. Rev. Lett. **107**, 115301 (2011).



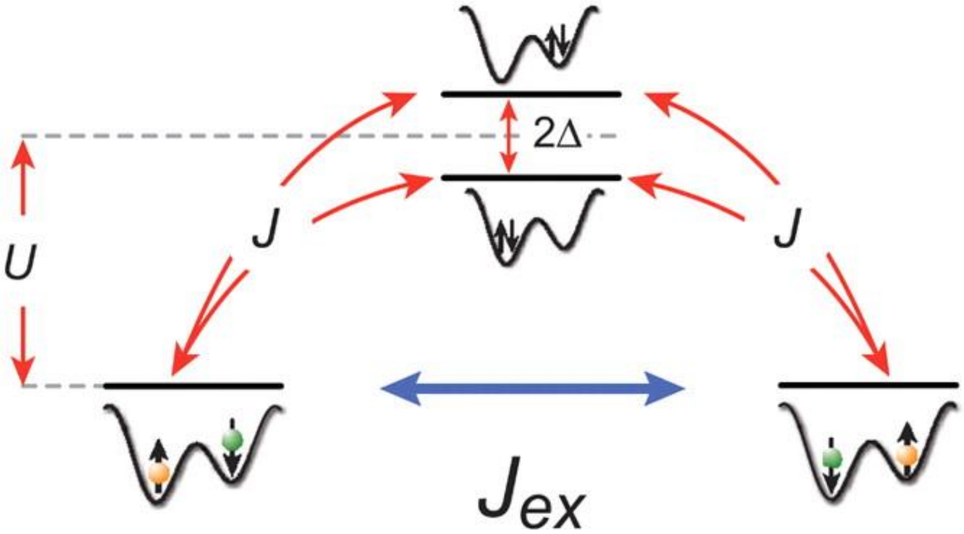
Molecules (material) are physically pinned down,
but spins (excitations) can be exchanged and mobile !



Many-body quantum localization – an old/new frontier
(David Huse, Gora Shlyapnikov, Misha Lukin, Eugene Demler …)

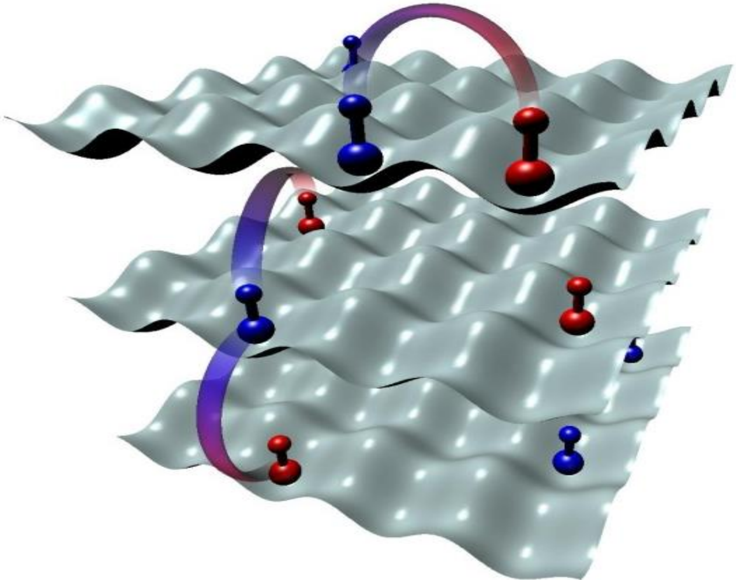
Spin exchange in ultracold systems

Super-exchange for atoms:
2nd order process via tunneling.



S. Trotsky *et al.*, Science 319, 5861 (2008)

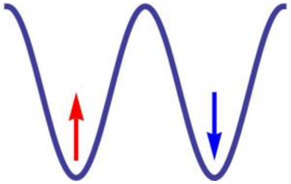
Spin exchange for molecules:
long-range dipolar interactions
-- motion & spin decoupled.



Interaction strength in quantum magnetism

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

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



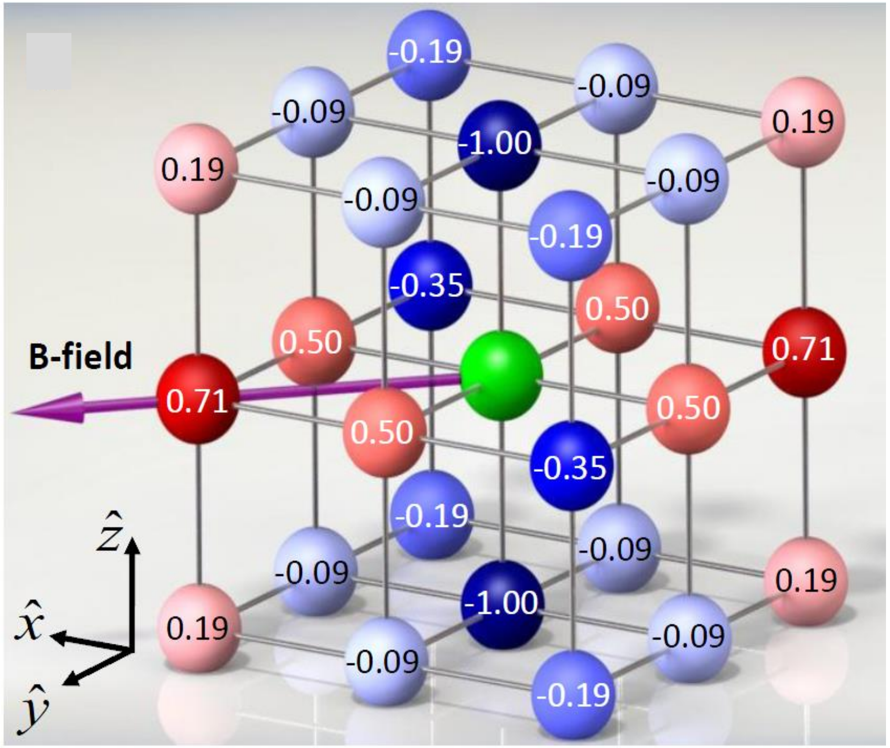
$$= S_i^x S_j^x + S_i^y S_j^y$$

Flip-flop term

$$\frac{1 - 3\cos^2\theta}{|\mathbf{r_i} - \mathbf{r_j}|^3}$$

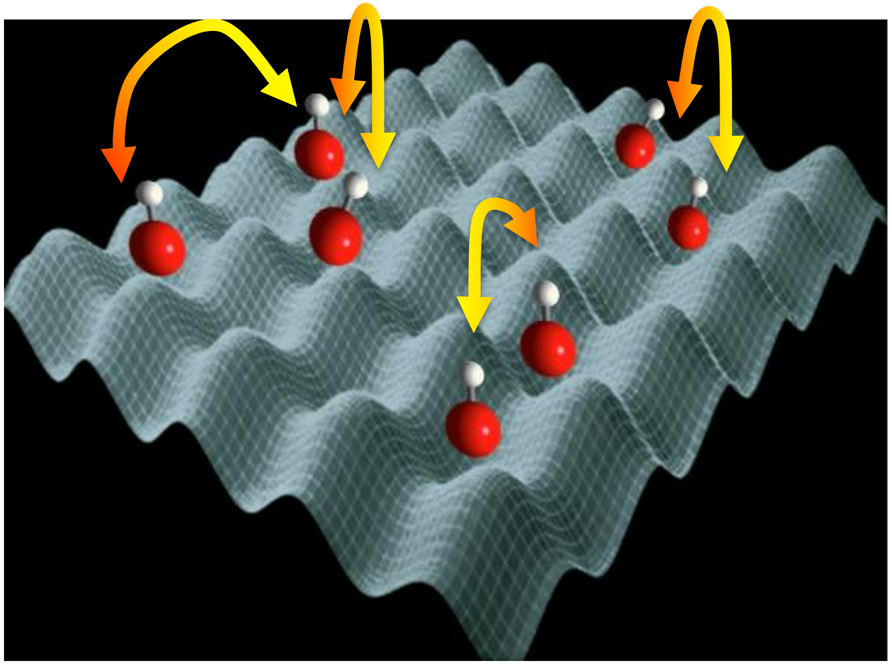
$$\frac{J_\perp}{h} = \frac{d_{\uparrow\downarrow}^2}{4\pi\epsilon_0 h a_{\mathrm{lat}}^3}$$

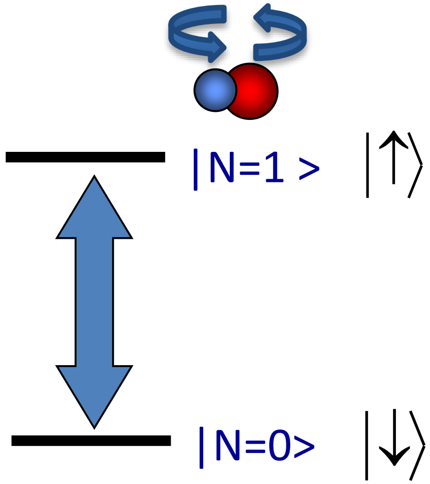
The oscillation frequency for a pair of molecules is $J_\perp/2h$.



A Dipolar Spin-Lattice Model

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

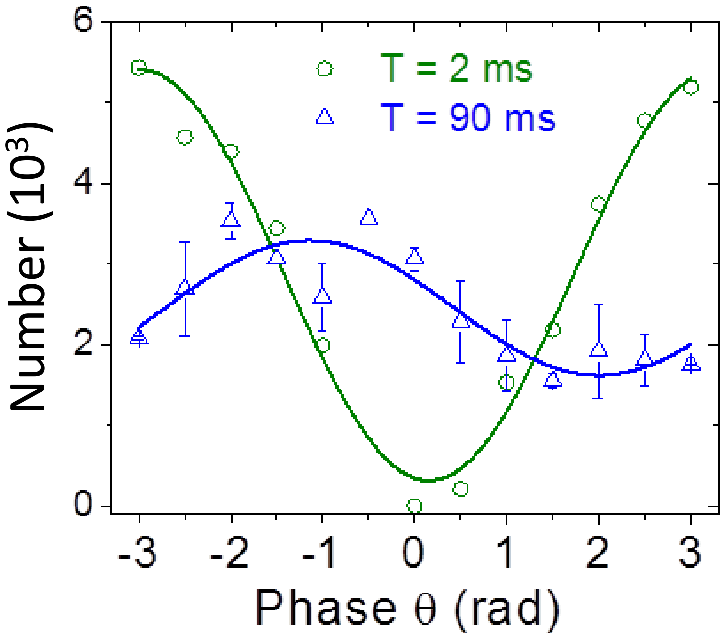




$|N=1>$ $|\uparrow\rangle$

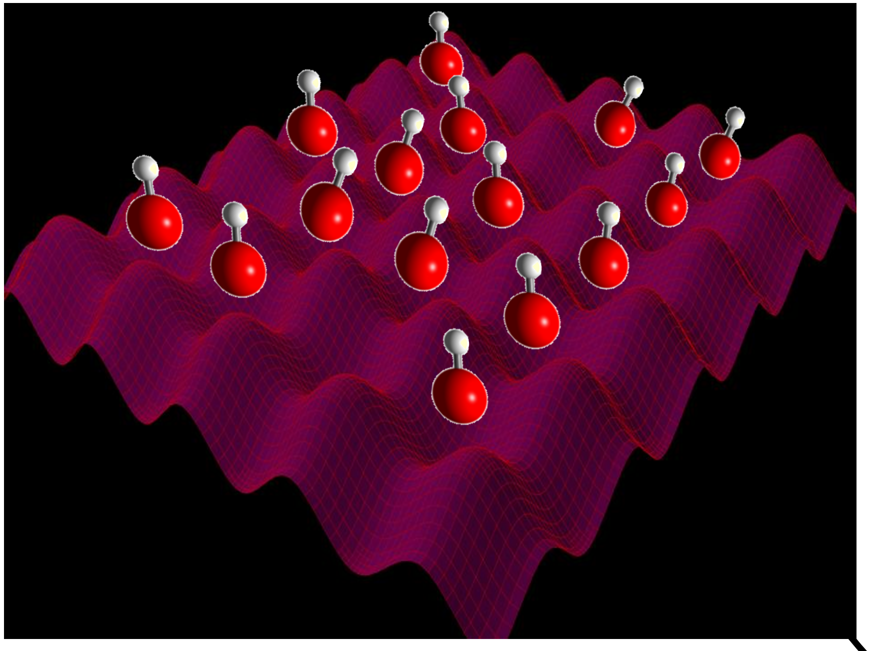
$|N=0>$ $|\downarrow\rangle$

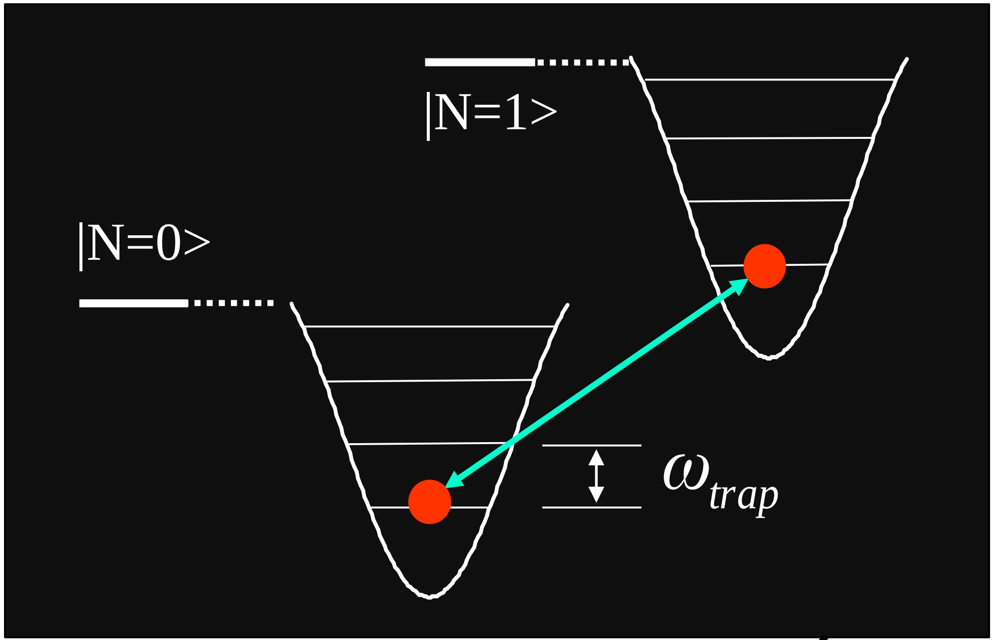
- Start with N=0. $|\downarrow\rangle$
- Drive a coherent spin superposition. $\frac{1}{\sqrt{2}}\left(|\uparrow\rangle+|\downarrow\rangle\right)$
- Probe spin coherence at T. (Ramsey spectroscopy)

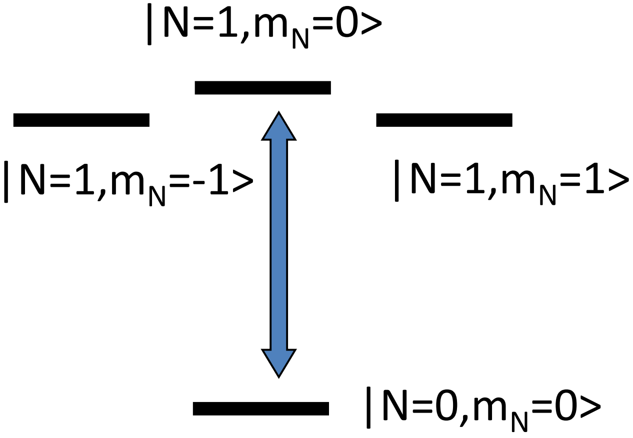


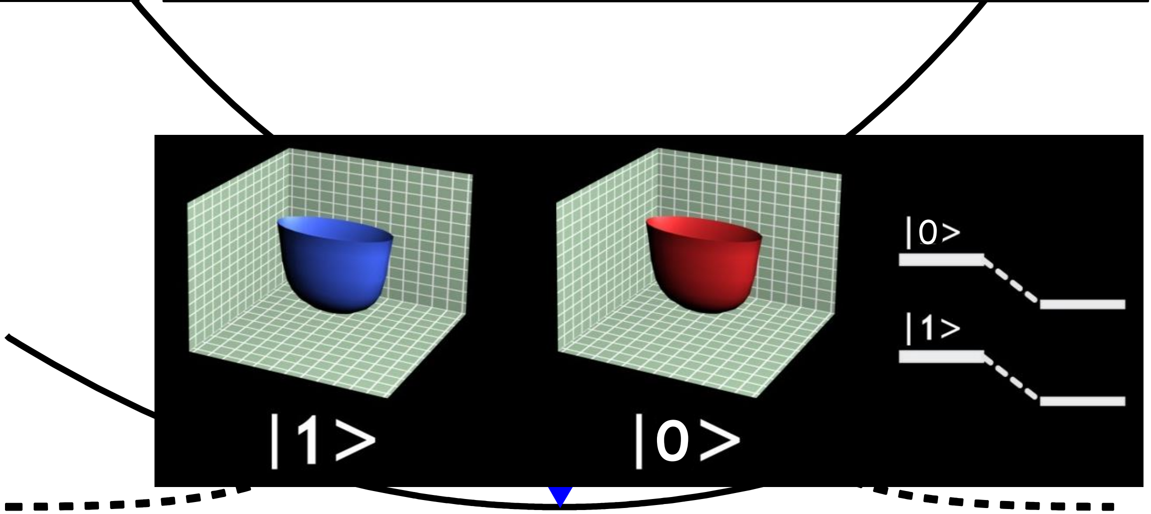
State-insensitive optical lattice

Nyenhuis *et al.*, PRL **109**, 230403 (2012).

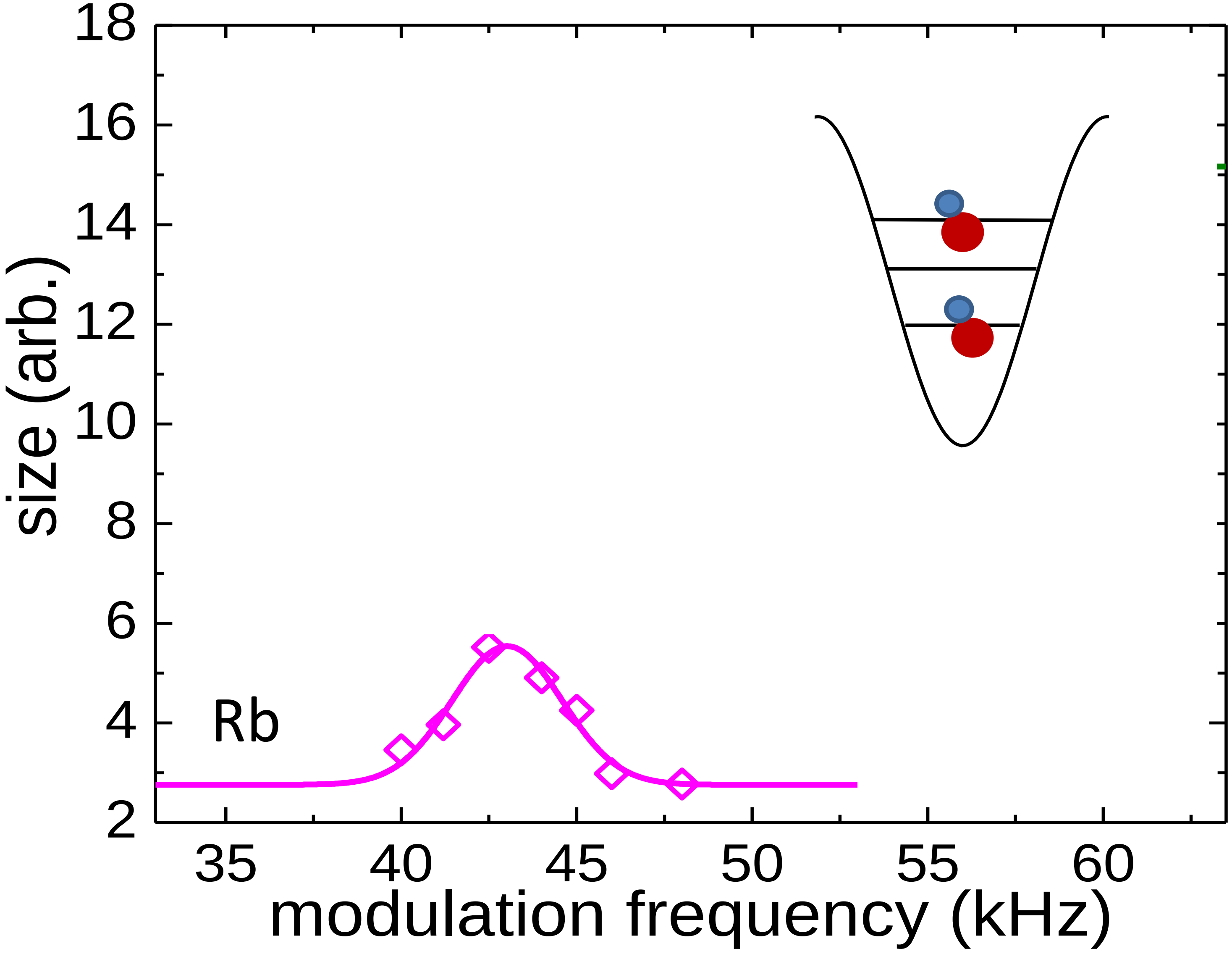




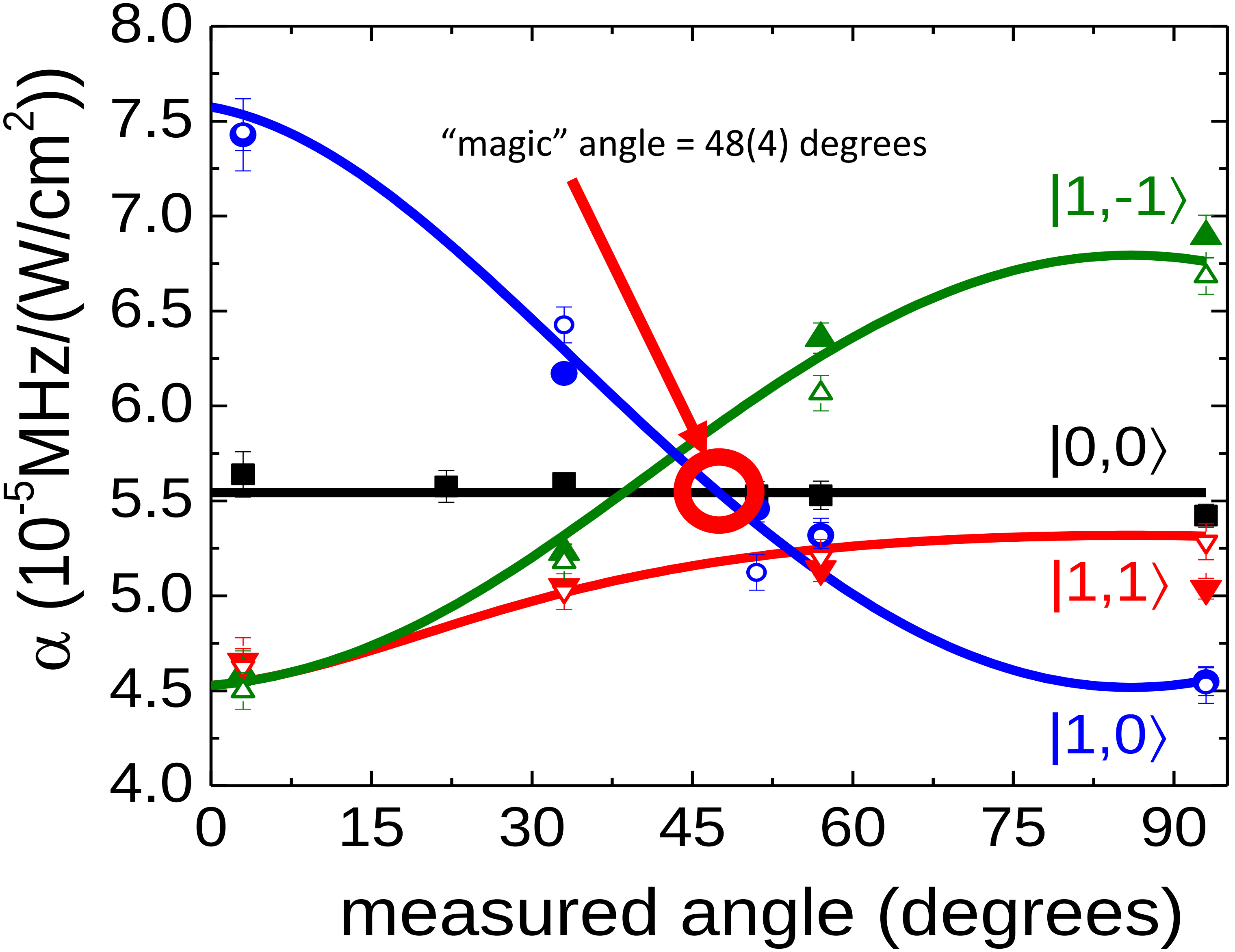




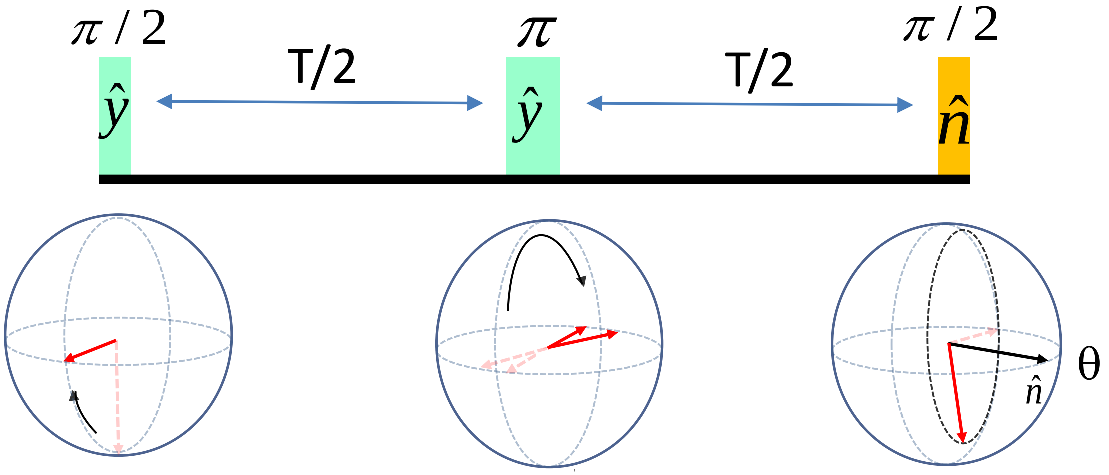
Measure trap strength with parametric heating



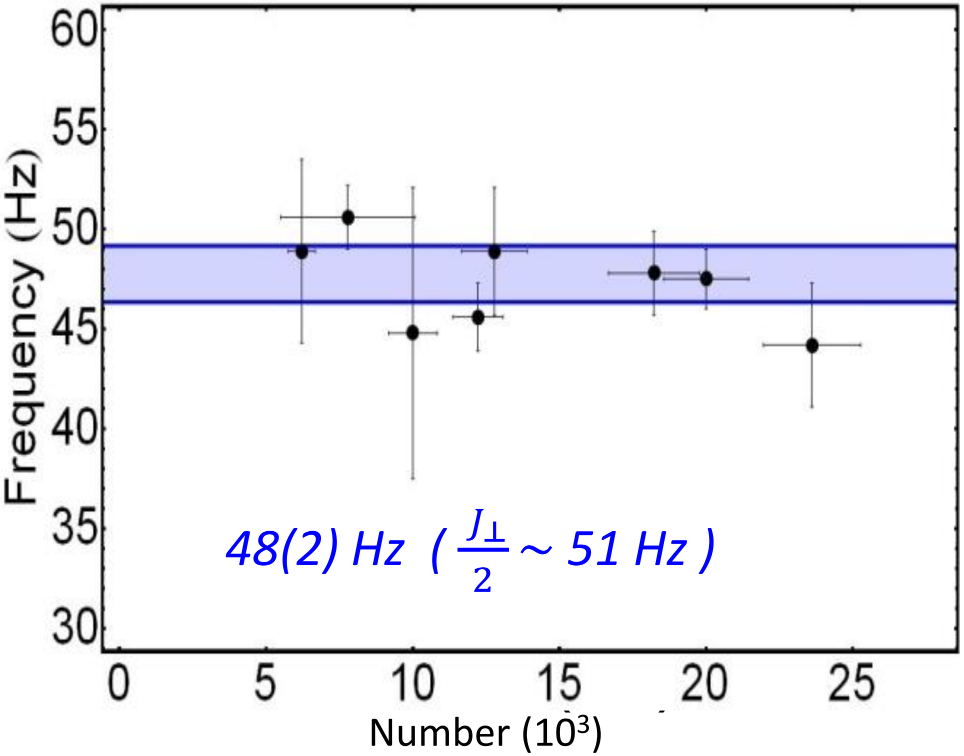
Angle-dependent polarizability



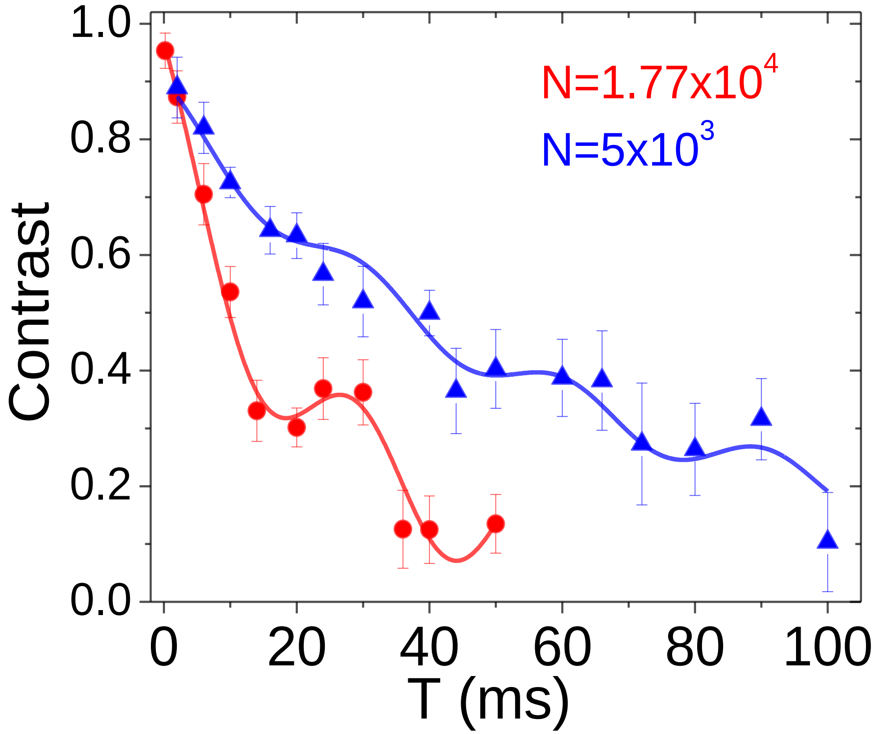
Ramsey & Spin-echo Spectroscopy



Scan θ for Ramsey fringe.

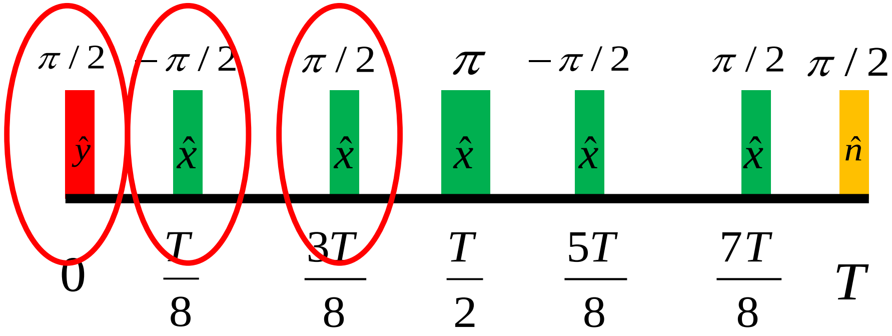


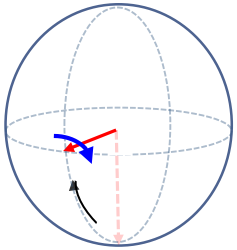
48(2) Hz ($\frac{J_\perp}{2} \sim$ 51 Hz)



Disentangling pulses

A multi-pulse sequence to suppress two particle interactions
(inspired by Misha Lukin, Hans-Peter Büchler)



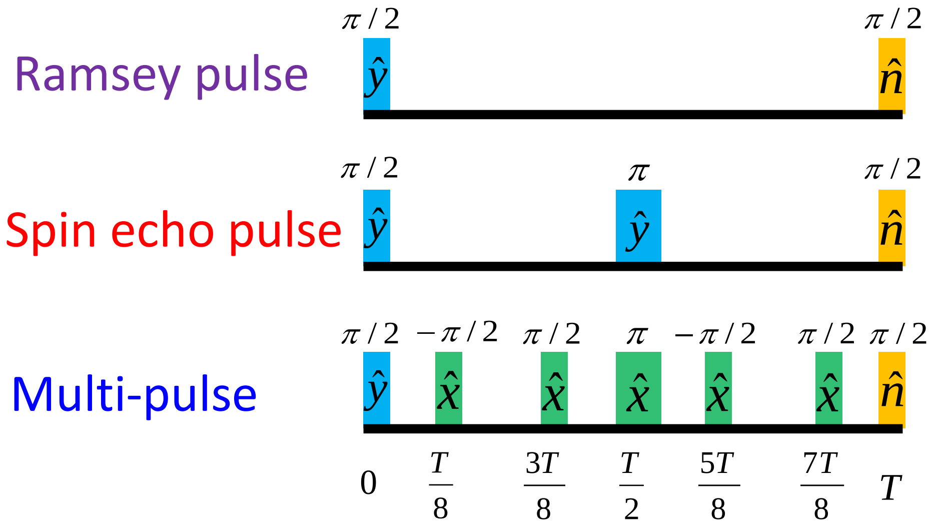


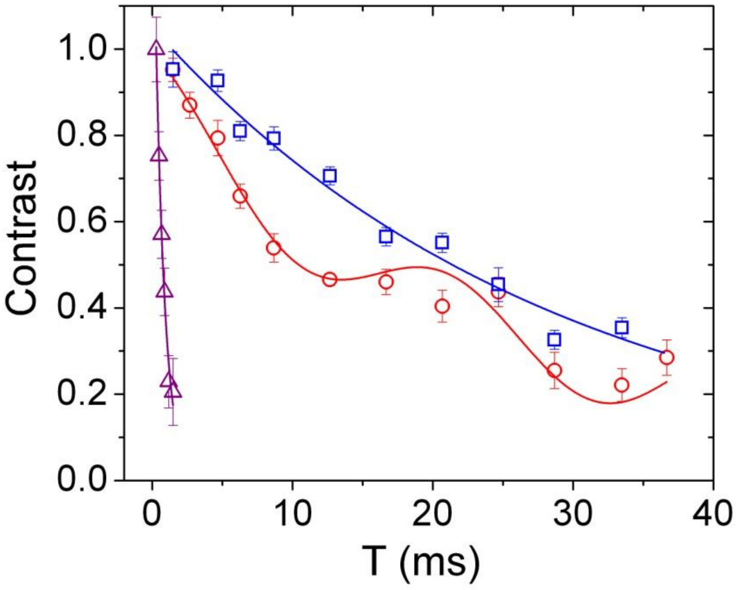
$$\frac{1}{\sqrt{2}}(|\downarrow\rangle+|\uparrow\rangle)\otimes\frac{1}{\sqrt{2}}(|\downarrow\rangle+|\uparrow\rangle)=\frac{1}{2}(|\downarrow\downarrow\rangle+|\uparrow\uparrow\rangle+|\downarrow\uparrow\rangle+|\uparrow\downarrow\rangle)$$

$$e^{-i(J/\hbar)(T/16)} \qquad e^{-i(J/\hbar)(T/16)}$$

$$\left(\frac{\pi}{2}\right)_x \qquad |\downarrow\uparrow\rangle+|\uparrow\downarrow\rangle \rightleftarrows |\downarrow\downarrow\rangle+|\uparrow\uparrow\rangle$$

Suppressing the oscillations

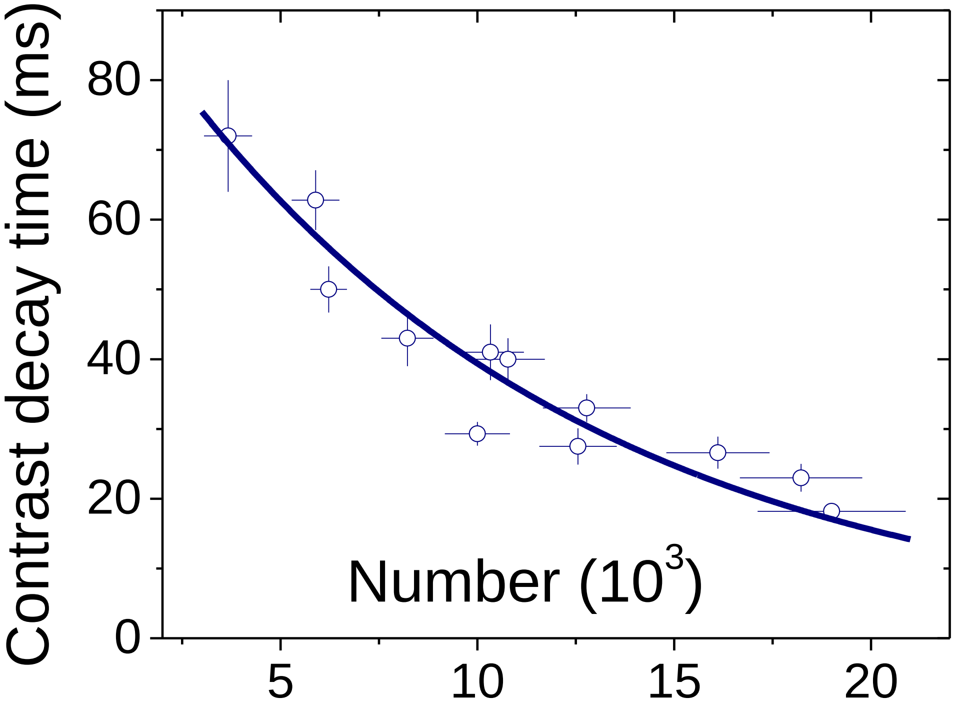


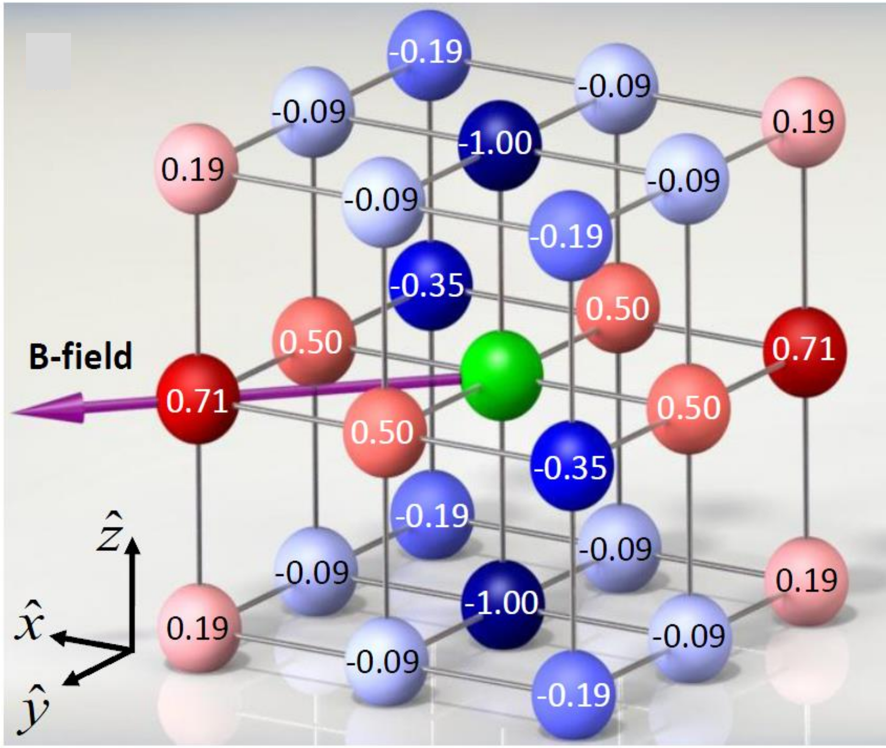


- *The spin-flip terms create entanglement*

- *The $(\pi/2)_x$ pulse swaps the eigenstates of the Hamiltonian, allowing the spins to rephase.*

Sum of pair-wise interactions or many-body interactions

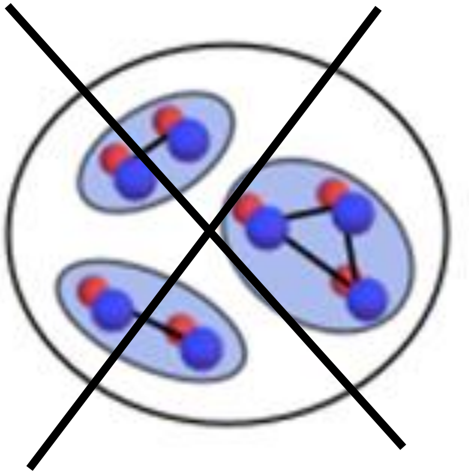




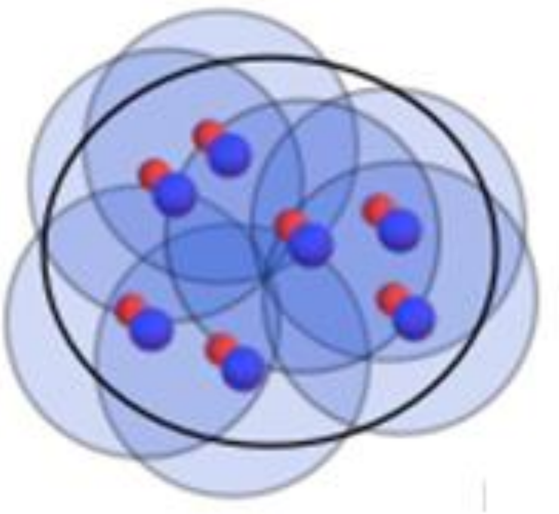
Simulation with
“Moving Average Cluster Expansion“

Hazzard *et al.*, arXiv:1402.2354

Standard

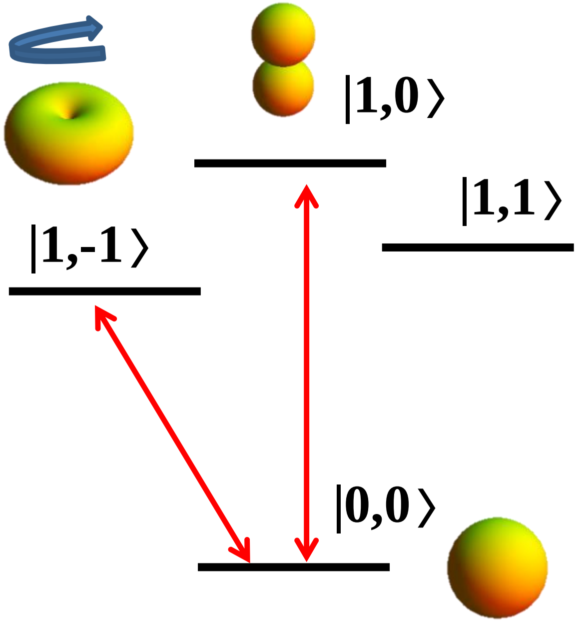


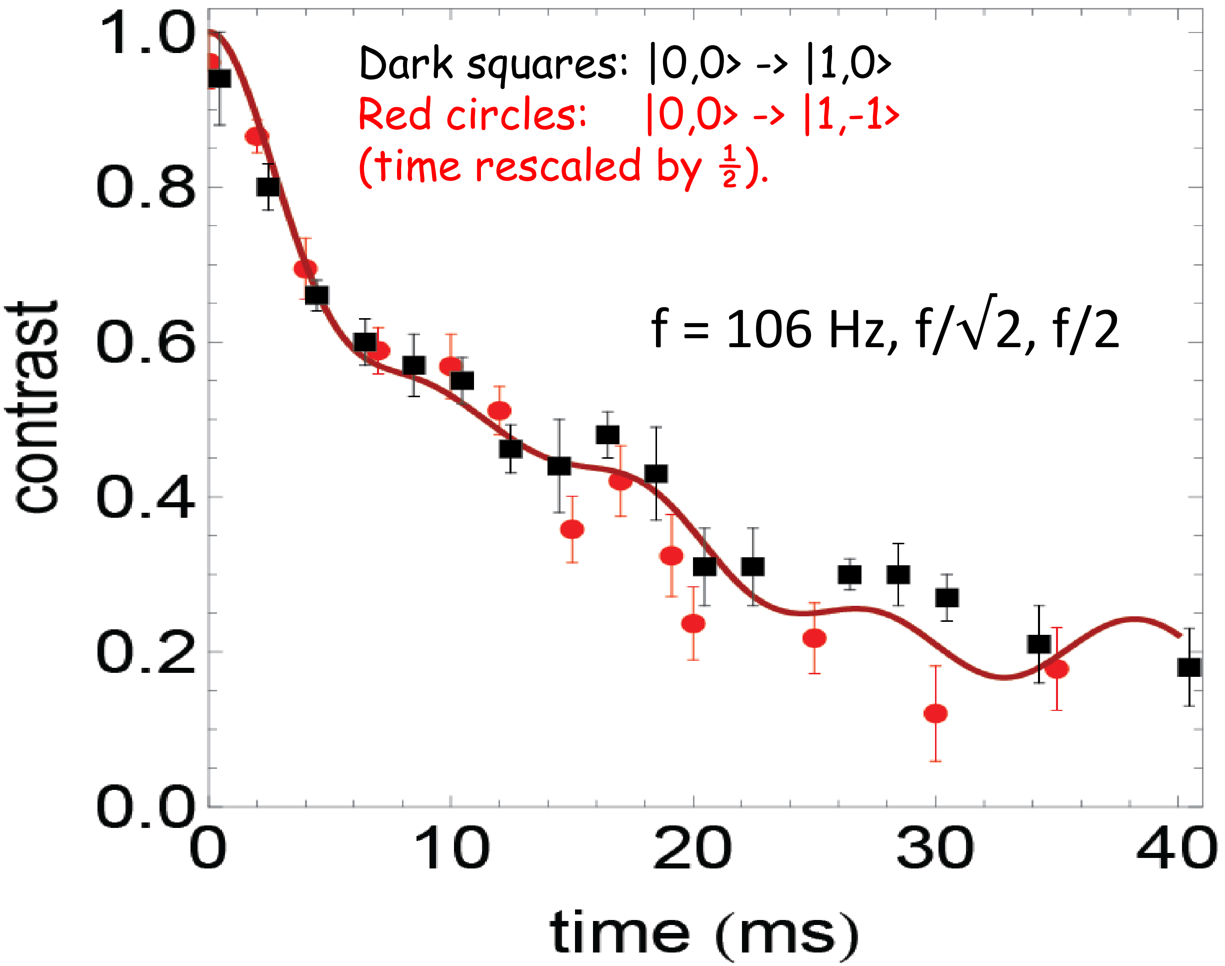
MACE



- **For each spin select an optimal cluster size**
- **Solve the dynamics for that cluster:** $\langle S_i^x \rangle$
- **Total dynamics: Sum over clusters** $\sum_{i=1}^{N} \langle S_i^x \rangle$

Control dipolar interaction





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

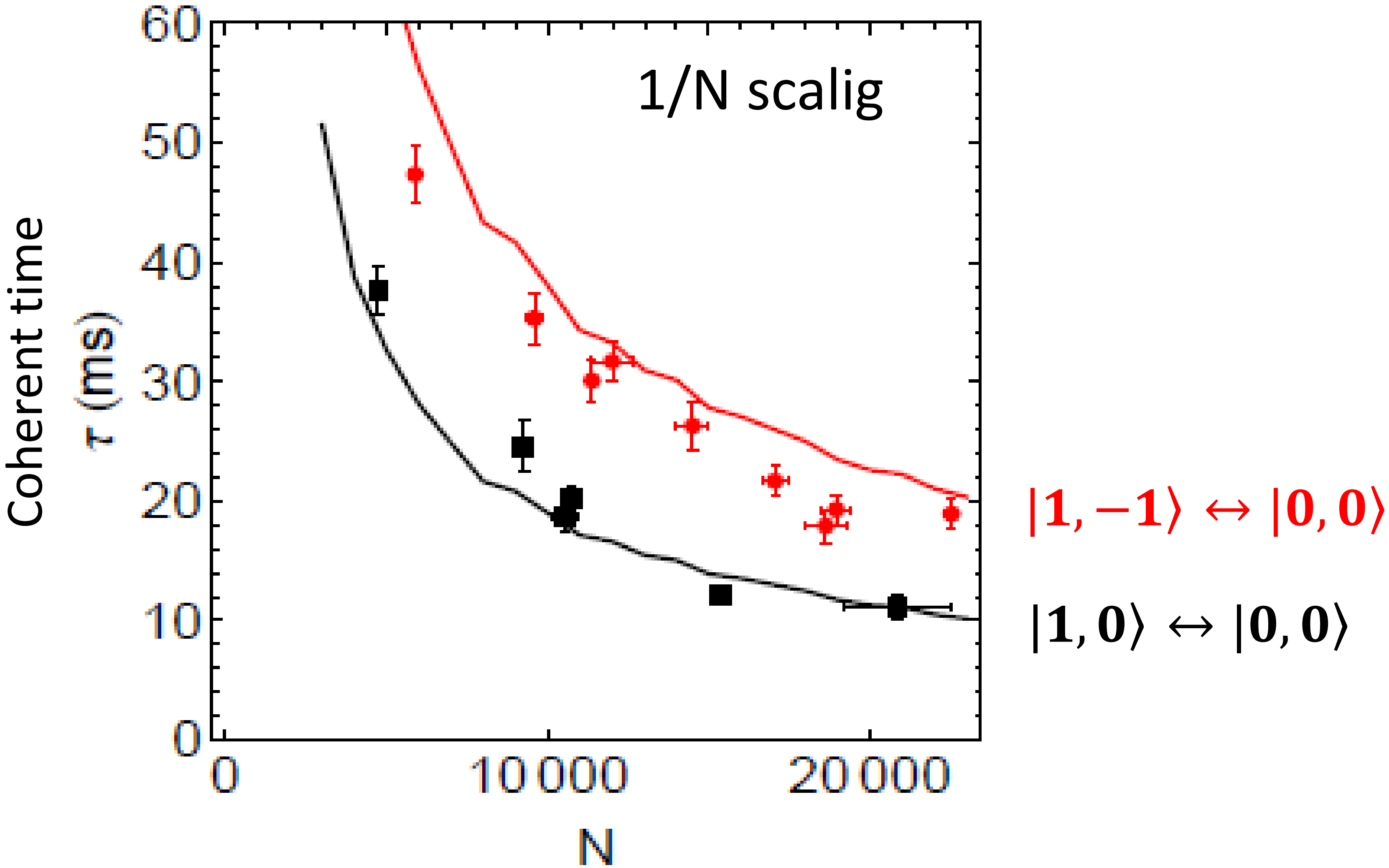
for |0,0> to |1,0>

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

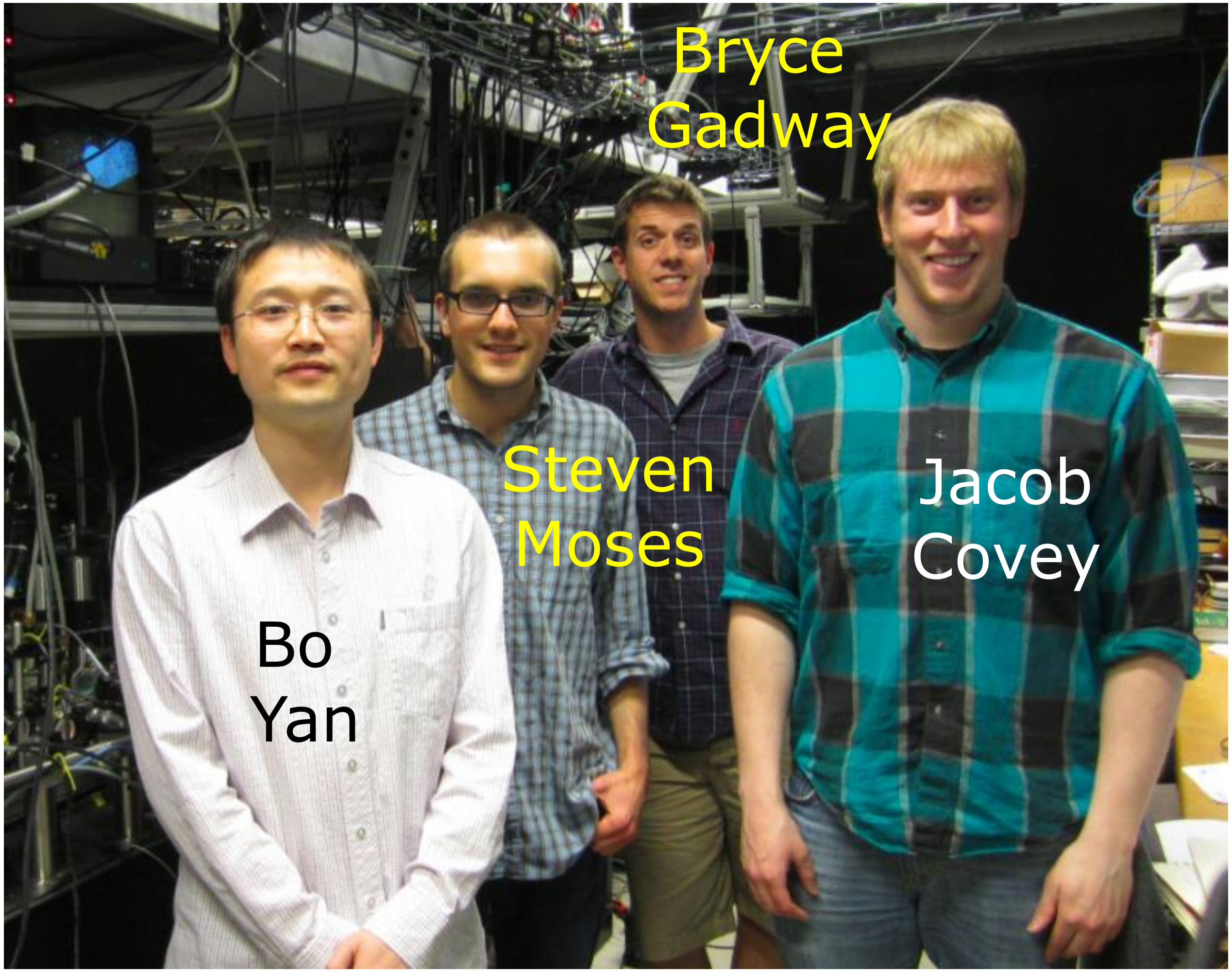
One fitting parameter (density) reproduces the experiment

Coherence time scales as 1/N

Experiment / theory benchmarking each other



Special Thanks (KRb team):



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Kang-Kuen Ni
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Theory collaborations:
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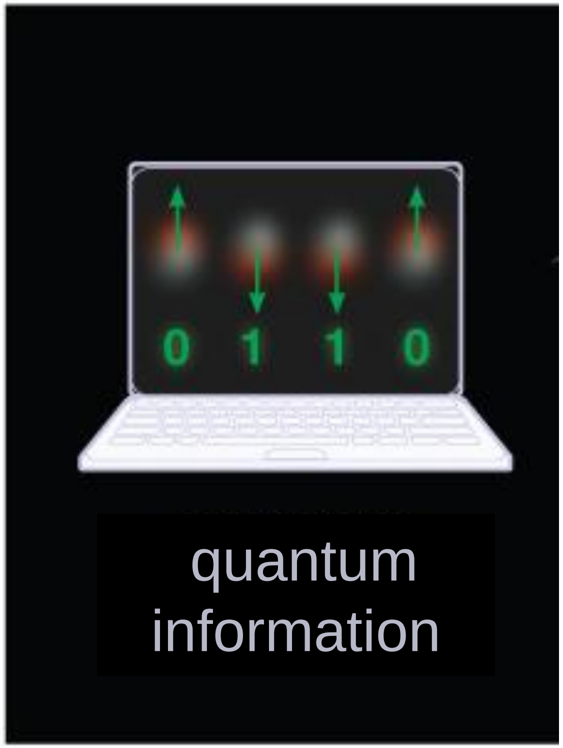
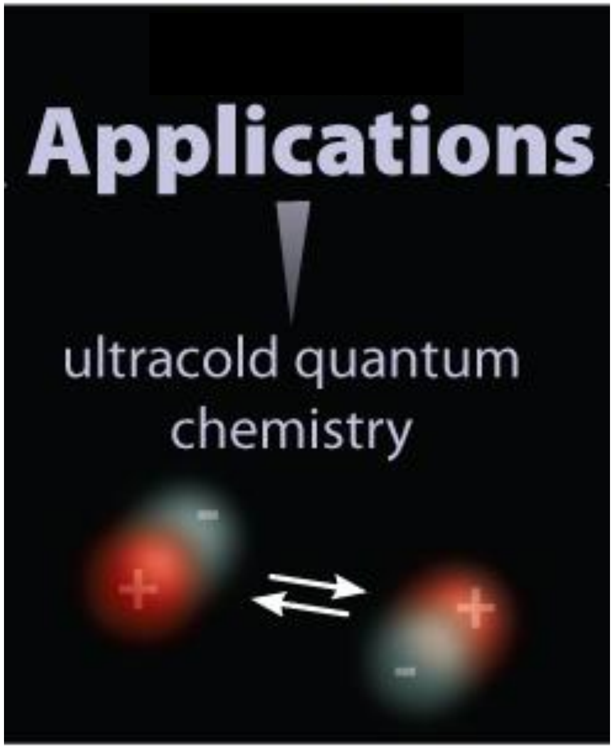
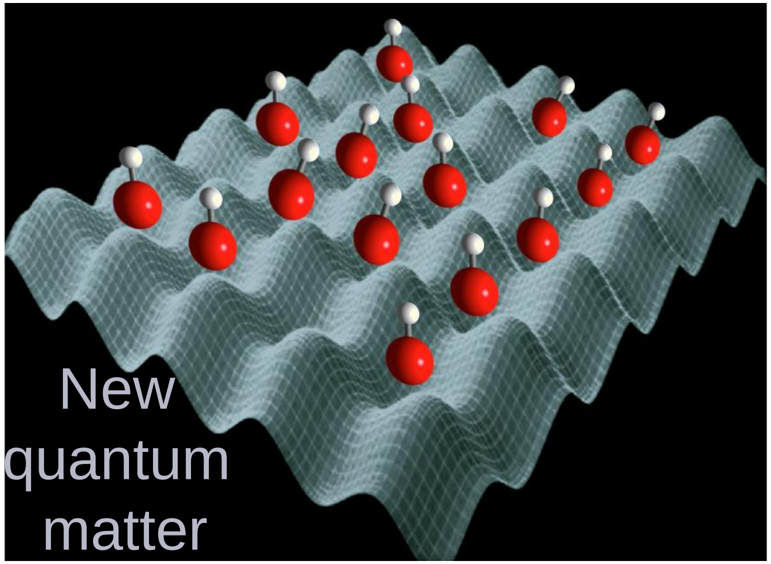
Science with cold molecules



quantum information



precision measurement & fundamental test



New quantum matter