Ultracold Molecules for physics and chemistry

molecules in tweezers

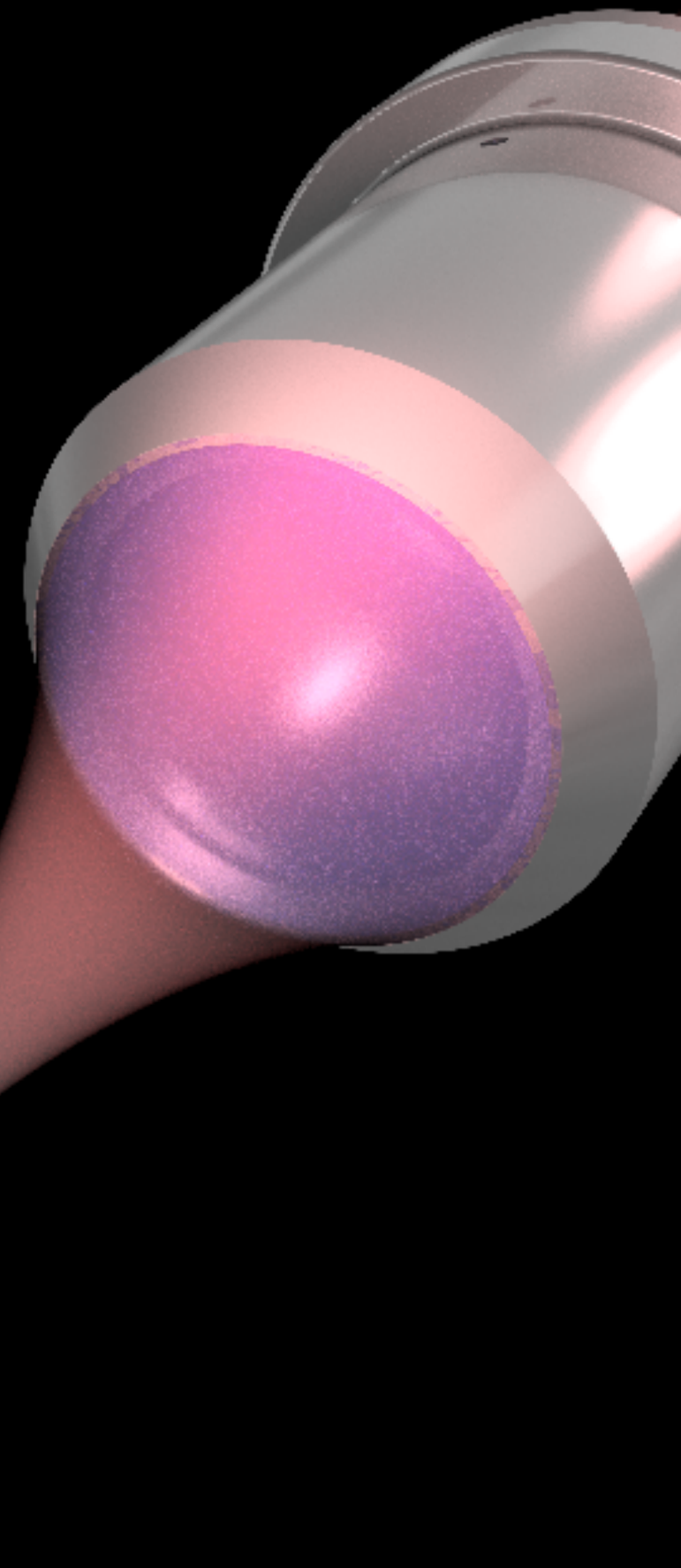
Kang-Kuen Ni

Department of Chemistry

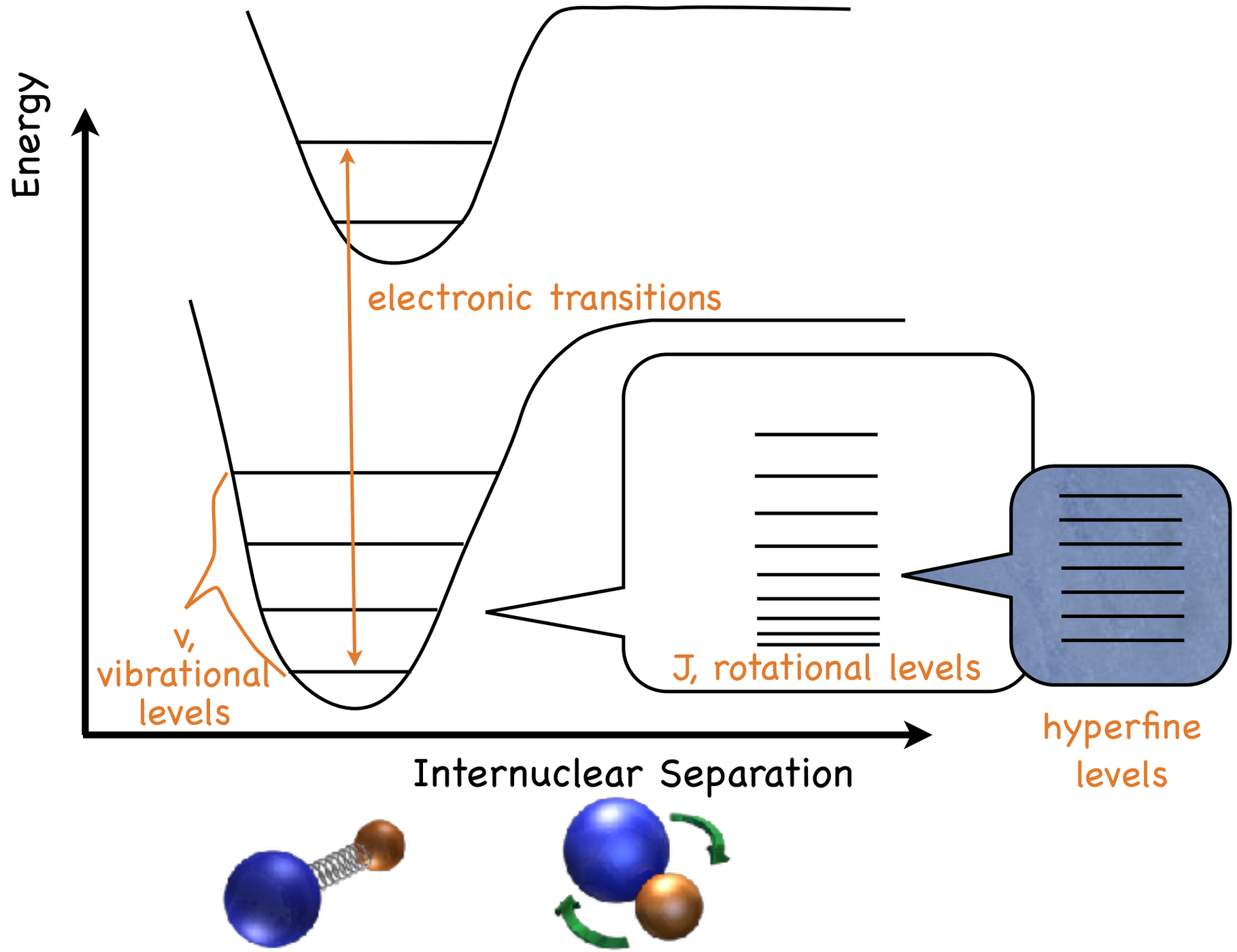
and Chemical Biology

Harvard University

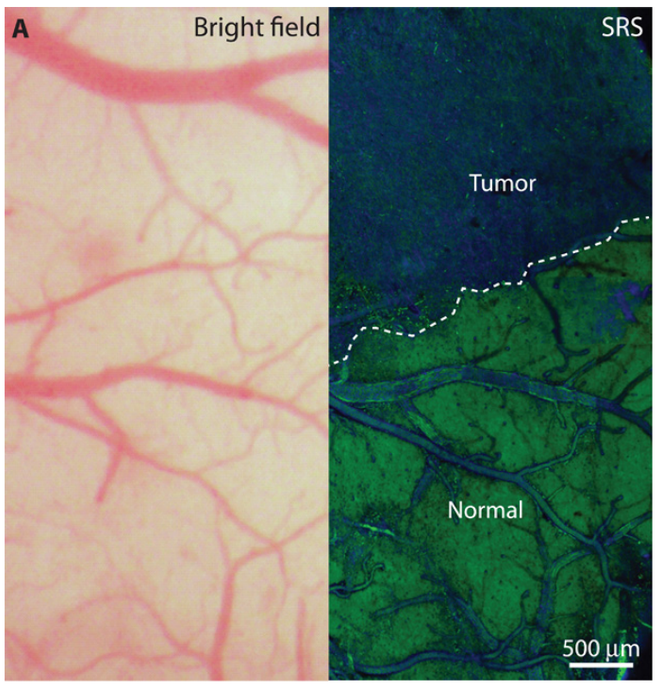
Boulder Summer School 2021



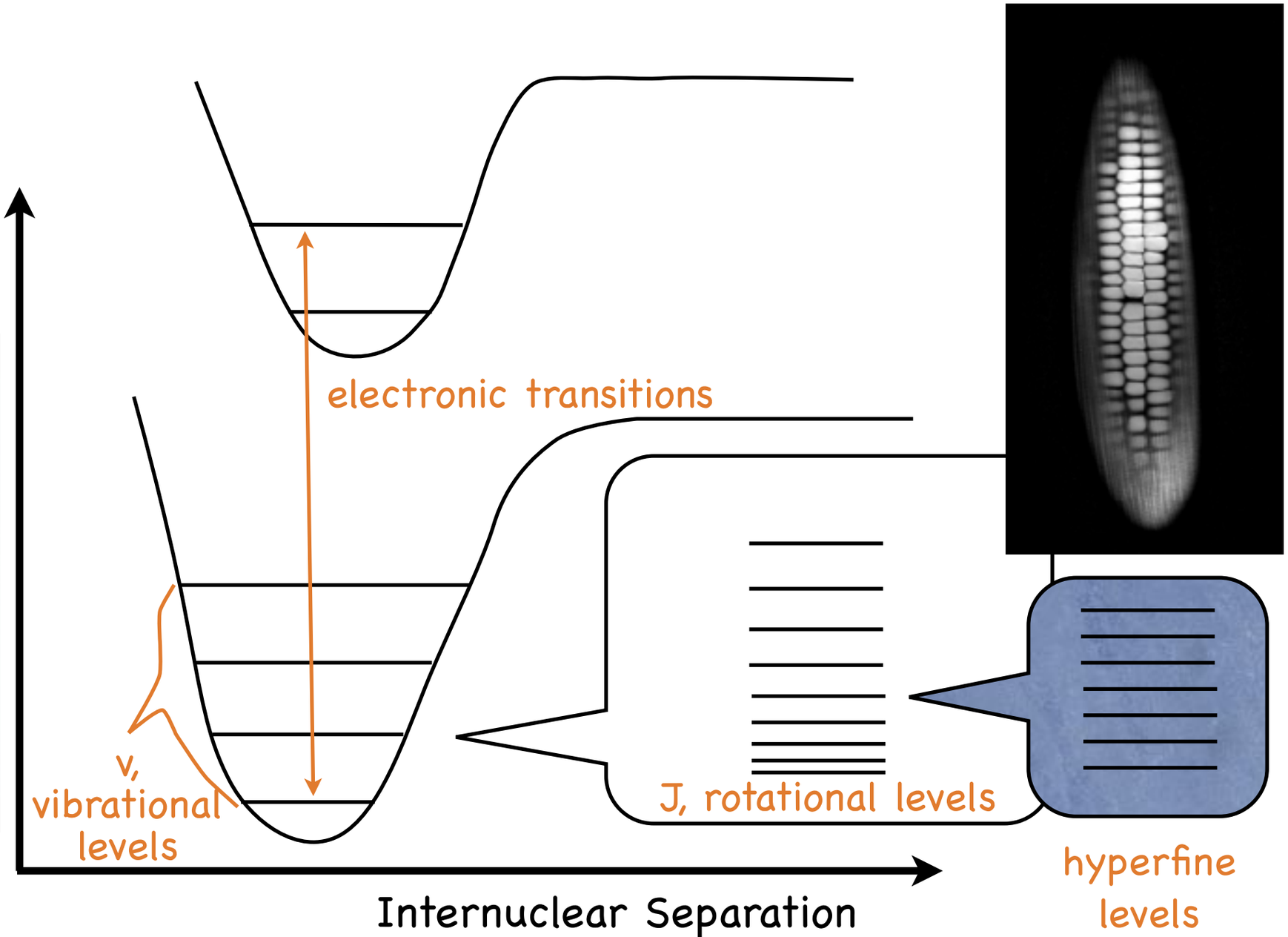
Molecular Quantum Degrees of Freedom

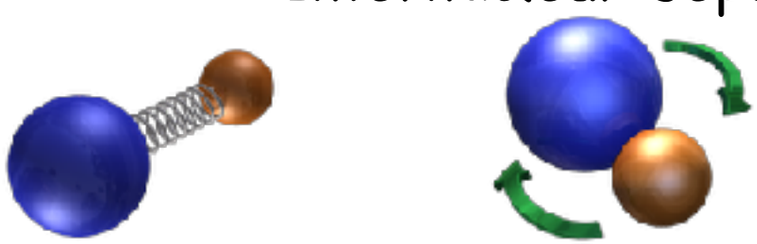


Molecular Quantum Degrees of Freedom



Sci. Transl. Med. 5, 201 (2013)





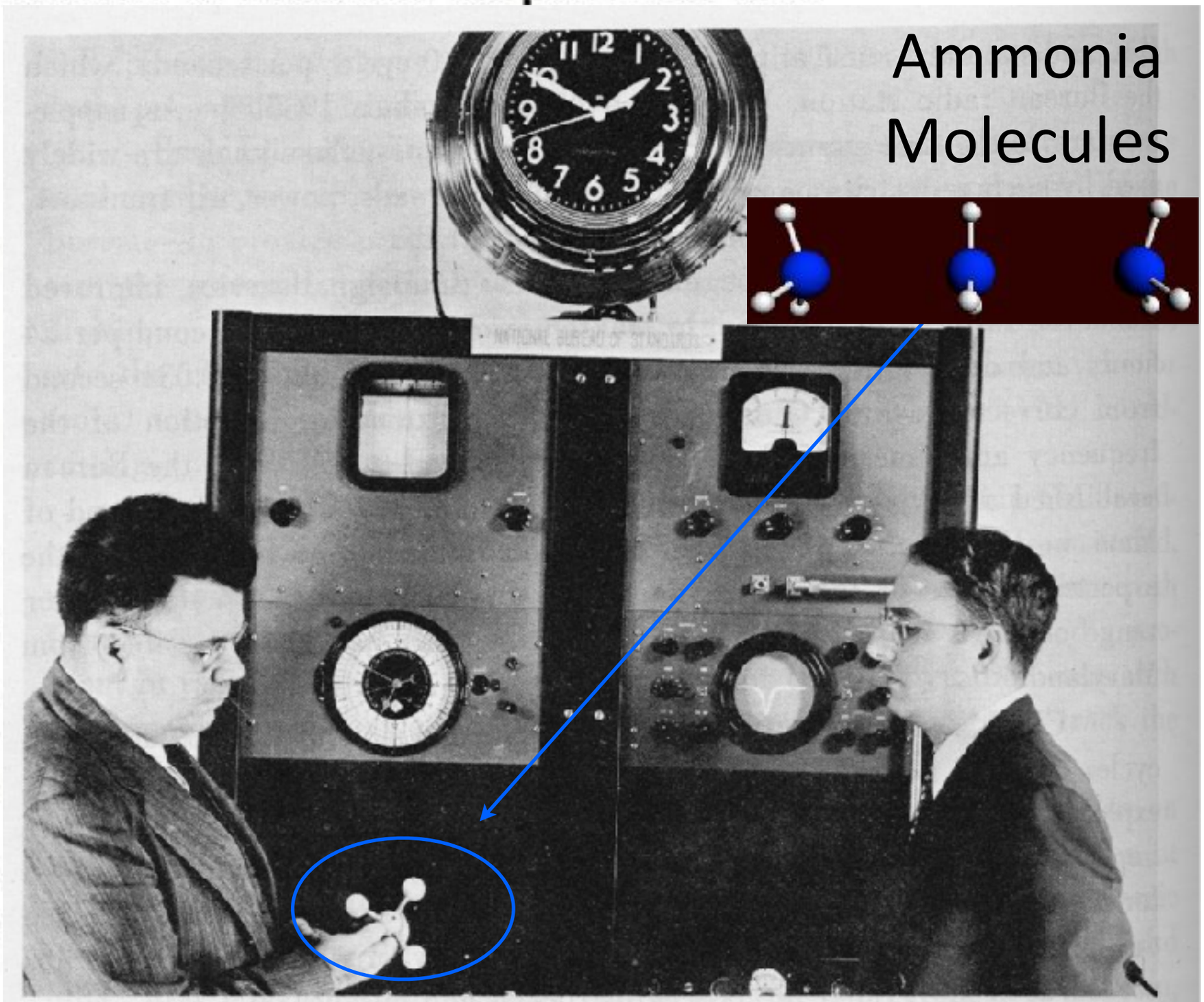
https://experiencetalk.files.wordpress.com/2014/04/corn-mri-scan.gif

First “Atomic” Clock, 1949

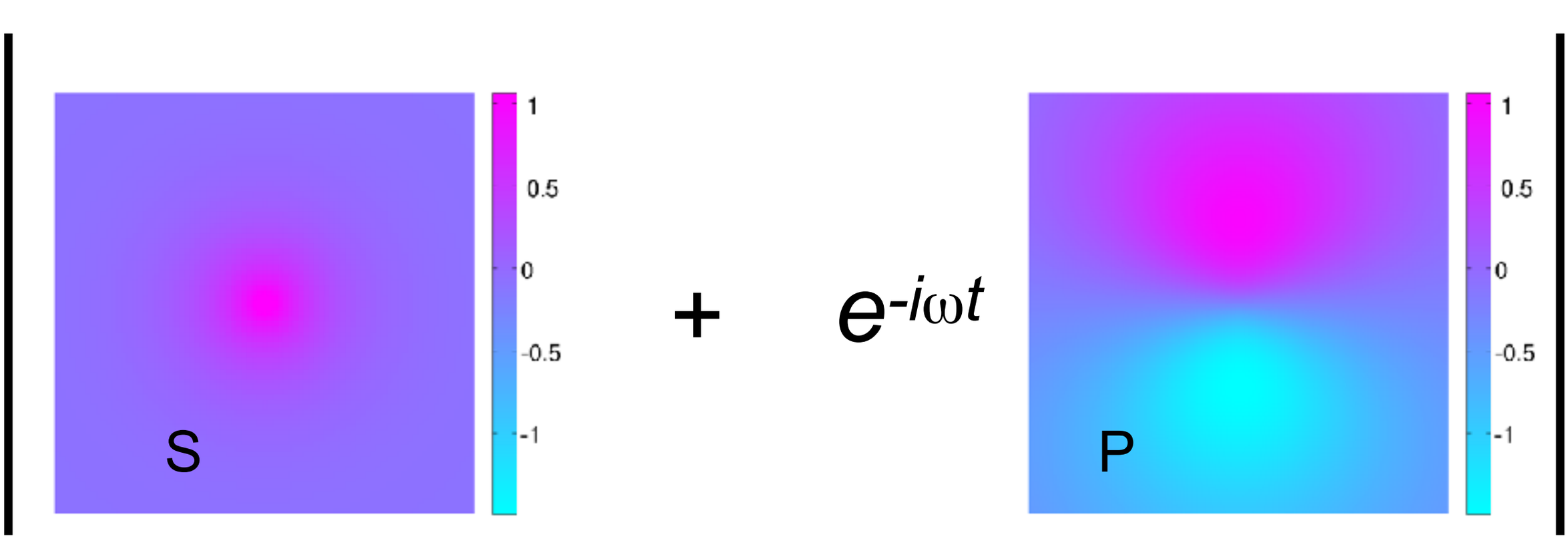
Clock Regulated by Vibrations of Molecules Keeps Perfect Time

So accurate that it will lose only one second in 300 years, an atomic clock developed at the Bureau of Standards is controlled by the constant, natural vibration of atoms in the ammonia molecule. The atomic timepiece is the first clock not dependent on astronomical observation.

Driven by a quartz-crystal oscillator, the clock is a complex electronic instrument. A low-frequency radio signal, generated by the crystal oscillator, is transformed into a very high frequency signal. This signal is then compared with the natural vibration frequency of the ammonia molecule, which oscillates or turns itself inside-out 24 billion times a second. When the two frequencies differ, an “error signal” adjusts the oscillator, bringing them into agreement. The natural frequency of the ammonia molecule being invariable, this continuous synchronization enables the clock to tick off seconds far more accurately than any other method.



“Ticking” of atomic clock



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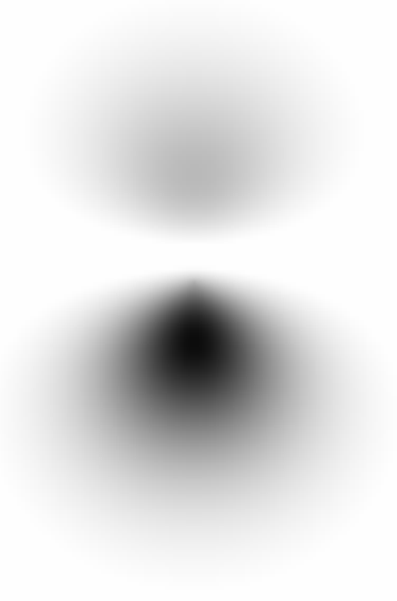


$Al^+/Hg^+/Sr/Yb$

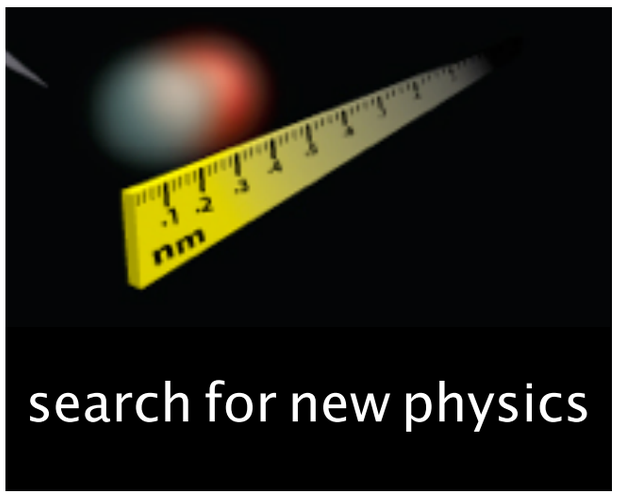
$\omega = \sim 2\pi \times 10^{15}$ Hz

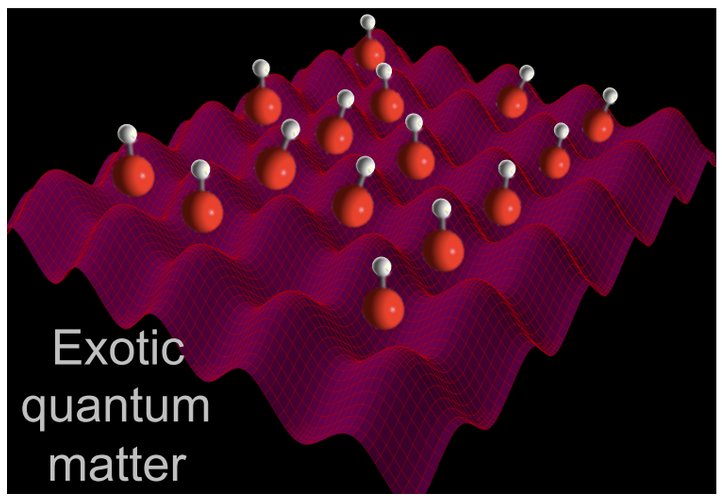
slide courtesy of T. Rosenband

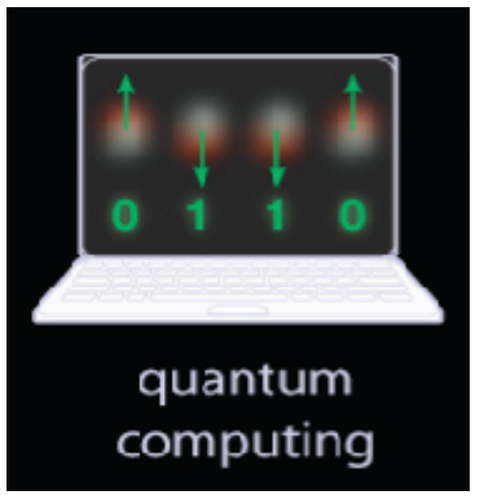
Quantum Control of Molecules

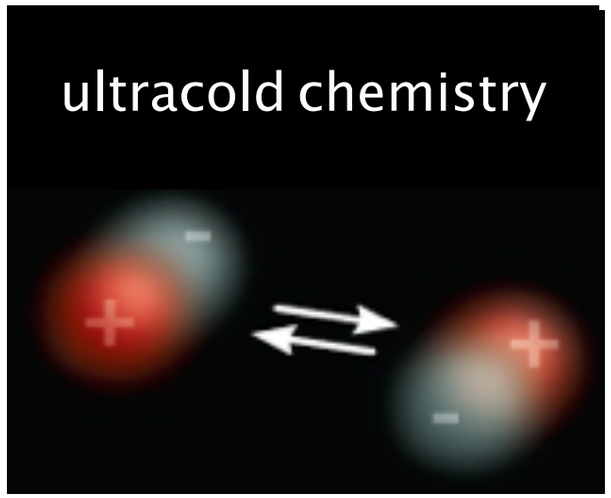
- cooling and trapping
- long interaction and probe time
- building complex system from the bottom up

Why Ultracold Molecules?





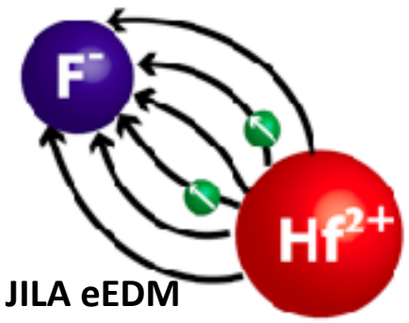




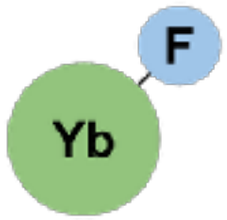
Fundamental Physics with Molecules

"Nature's high E-field laboratory"

- Searches for EDMs and other symmetry violations
- Bond distorts atomic orbitals (enhanced sensitivities)
- Molecular symmetries cancel systematics (co-magnetometer)

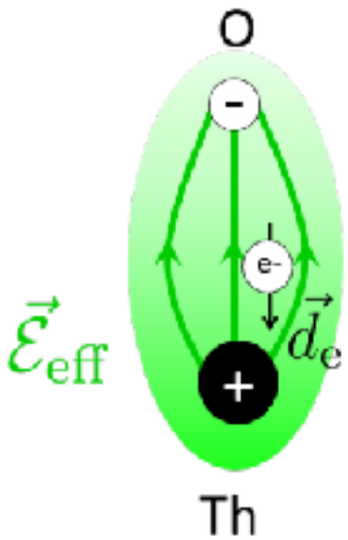


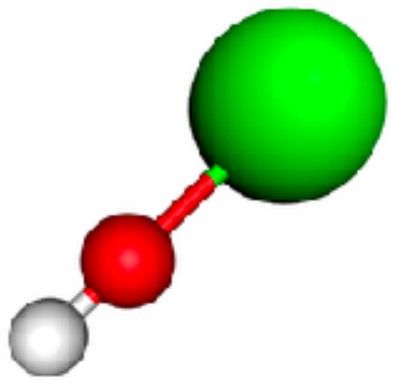
JILA eEDM molecular ions



Laser-cooled YbF (Imperial College)

ACME eEDM

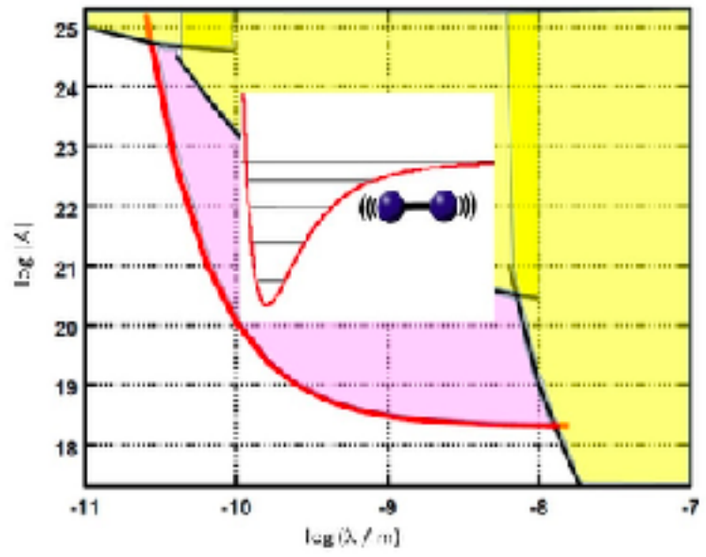




Future: laser-cooled YbOH (Hutzler, Doyle)

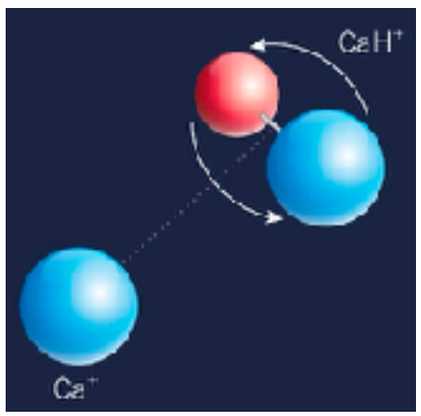
Bonded nuclei as "test masses"

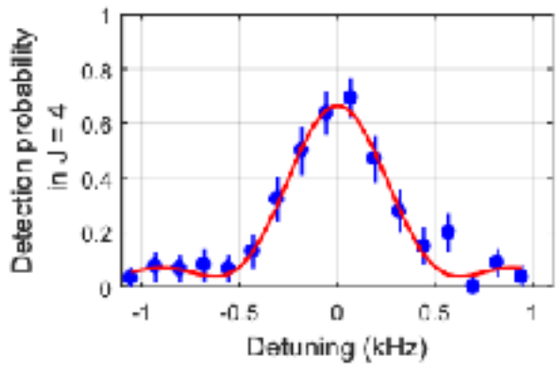
- Testing gravity at nanometer scales
- Ultracold Sr_2 – T. Zelevinsky, Columbia U



Testing precision theory

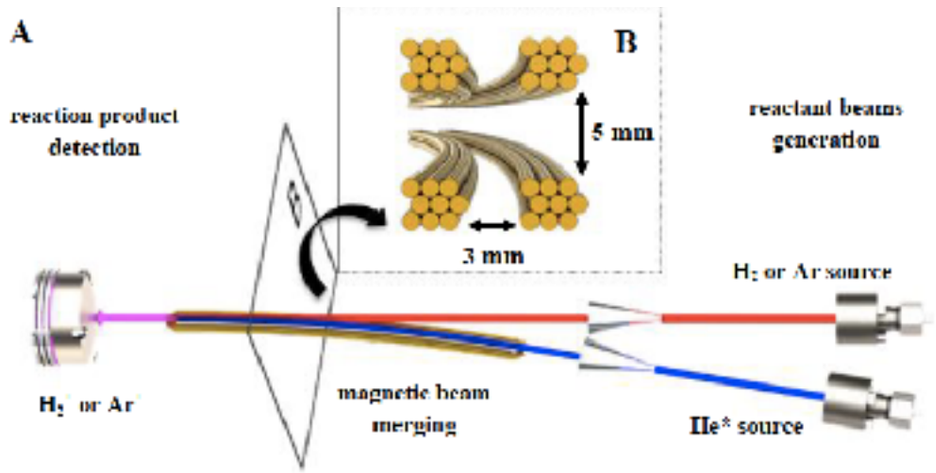
- High precision spectroscopy benchmarks theory
- CaH^+, NIST ion storage group
- HD^+, Vrije U. Amsterdam. Theory: C. Pachucki & co.



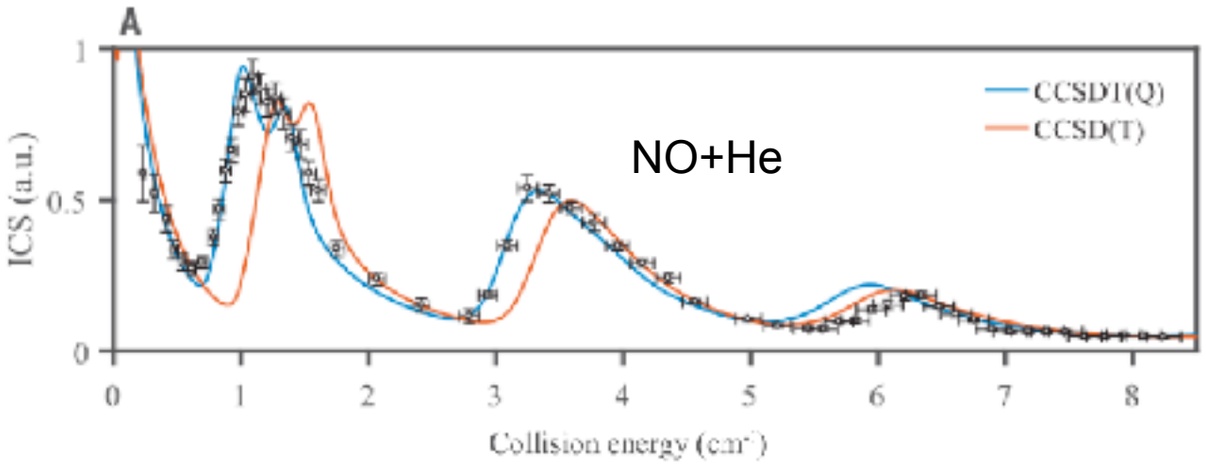


Fundamental Chemistry with Cold Molecules

probing potential energy surfaces beyond "gold-standard" quantum chemistry calculation



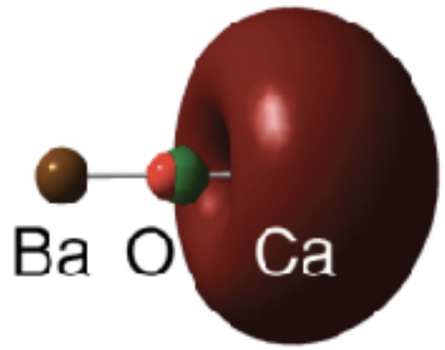
Narevicius (Wiezmann)
Nature Phys. 13, 35 (2017)



van de Meerakker (Radboud)
Science 368, 626 (2020)

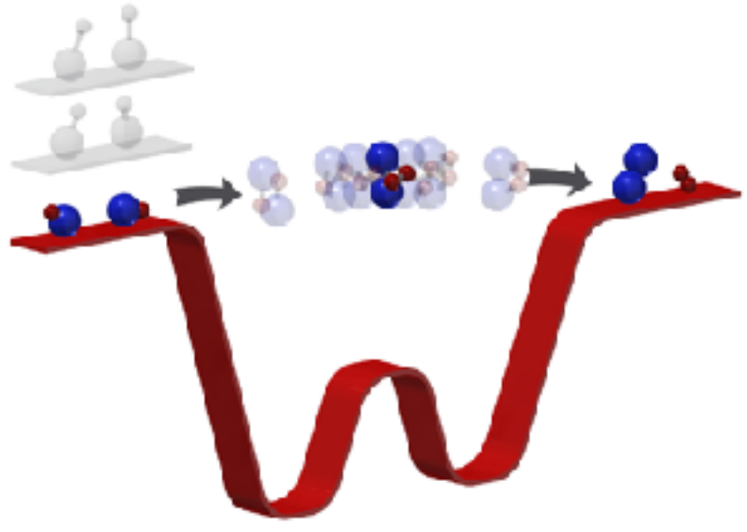
Other work: Ye, Denschlag, Zelevinsky, Willitsch, Zare, …

synthesizing new chemical species



Hudson (UCLA)
Science **357**, 1370 (2017)

Direct detection of reaction intermediate

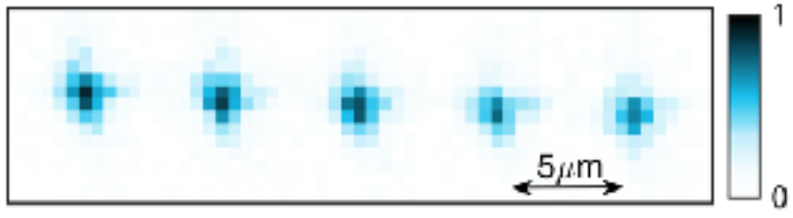


Hu, Liu, et al., Science **366**, 1111 (2019)

Outline (Lecture 1)

- Quantum computing/simulations with molecules (theoretical ideas)
- Preparing single ultracold polar molecules
  - Assembling molecules from atoms

Laser-cooled molecules

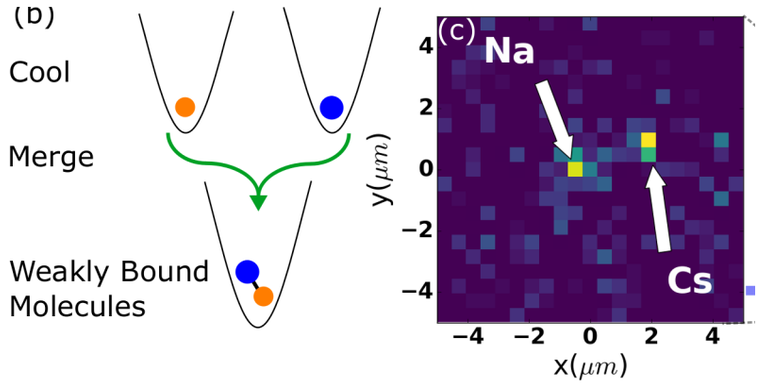


CUA Doyle/Ketterle/Ni Collaboration

Science 365, 1156 (2019)

Yale, Imperial, JILA…

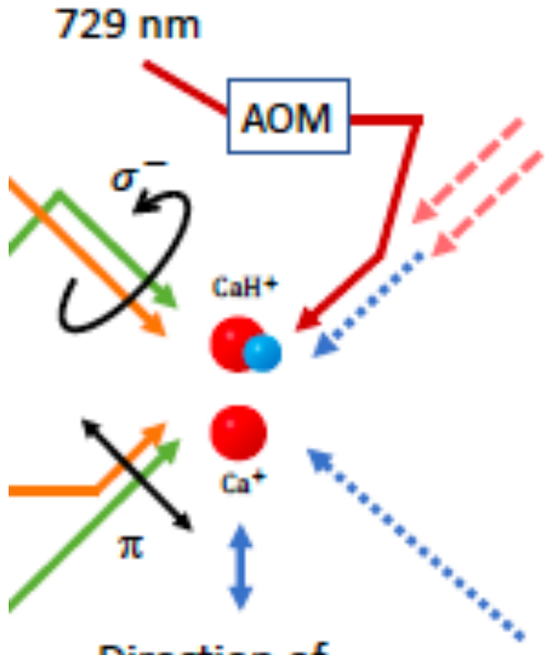
Assembly molecules from atoms



Science 360, 900 (2018)
Cairncross*, Zhang* et al., PRL **126**, 123402 (2021)

Wuhan, Durham, JILA, MIT, Innsbruck
Hong Kong, MPQ, Hannover,…

Molecular ions



NIST, Nature 581, 273 (2020)

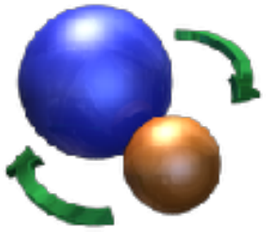
PTB, Basel, UCLA, UCSB,…

Outline (Lecture 2)

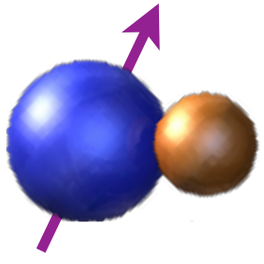
- Probing sub-microkelvin chemistry

(Designer) Ultracold Molecules for quantum simulation and computation

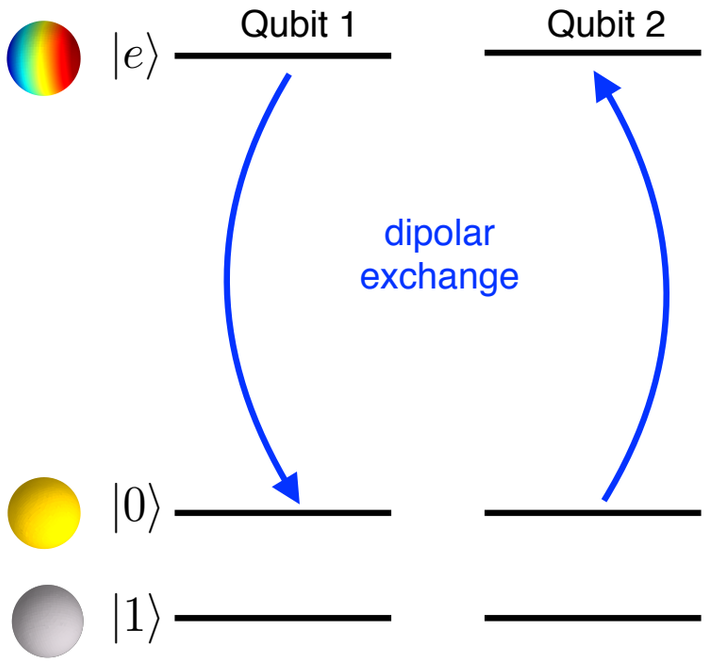
* larger variety of internal states for storage and interaction
* intrinsic molecular couplings between internal degrees of freedom



rotation

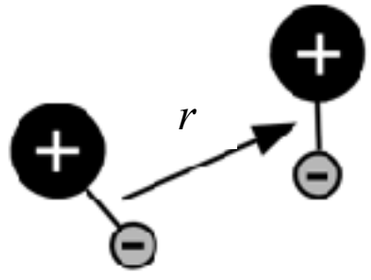


hyperfine



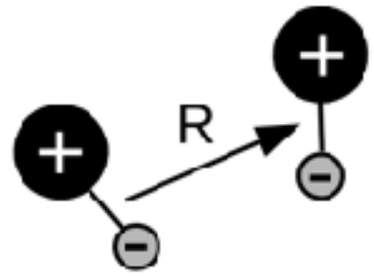
Dipole-dipole interaction

$$\hat{H}_{DD} = \frac{1}{4\pi\epsilon_0 r^3}\left[\hat{d}_1 \cdot \hat{d}_2 - 3(\hat{d}_1 \cdot \hat{e}_r)(\hat{d}_2 \cdot \hat{e}_r)\right]$$



DeMille, PRL **88**, 067901 (2002), Yelin, others…
Ni group, Chemical Science **9**, 6830-6838 (2018)
Jin and Ye (JILA) Nature **501**, 521 (2013)
Zwierlein (MIT) Science **357**, 372 (2017)

(Designer) Ultracold Molecules for quantum simulation and computation



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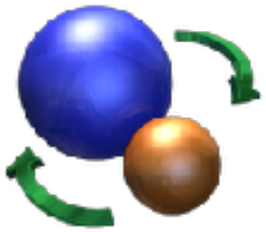
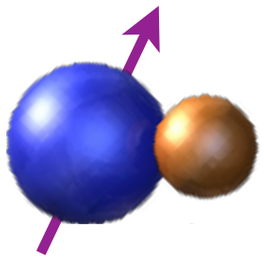
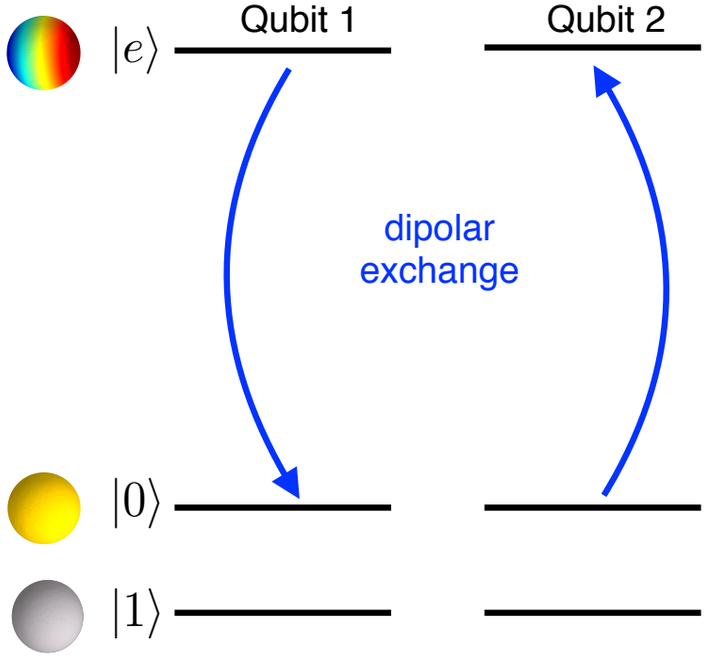


rotation



hyperfine



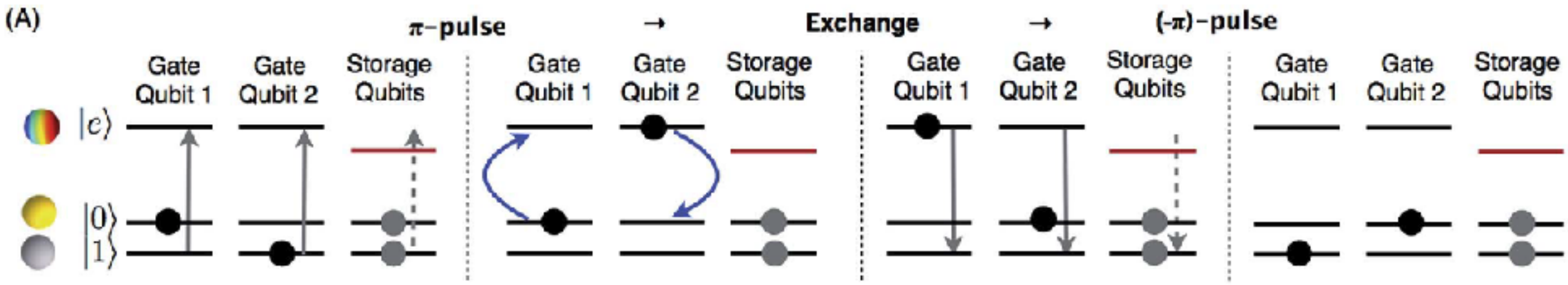
$$\widehat{H}_{DD} = \begin{array}{c} \begin{matrix} 00 & 0e & e0 & ee \end{matrix} \\ \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & \hbar\Omega & 0 \\ 0 & \hbar\Omega & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix} \end{array} \begin{matrix} 00 \\ 0e \\ e0 \\ ee \end{matrix}$$

1. Prepare a 50/50 superposition of |0> and |e> [a global single-particle pi/2-pulse]
2. Wait for t=pi/(2 Omega)
3. Apply another global single-particle pi/2-pulse
4. Realize a Bell state (|00>+|ee>)/2

$$U = e^{i\widehat{H}_{DD}t/\hbar} = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & cos\Omega t & isin\Omega t & 0 \\ 0 & isin\Omega t & cos\Omega t & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix}$$

Two-Qubit (iSWAP) Gates (scheme)

* Avoid DC electric-field or dressed field

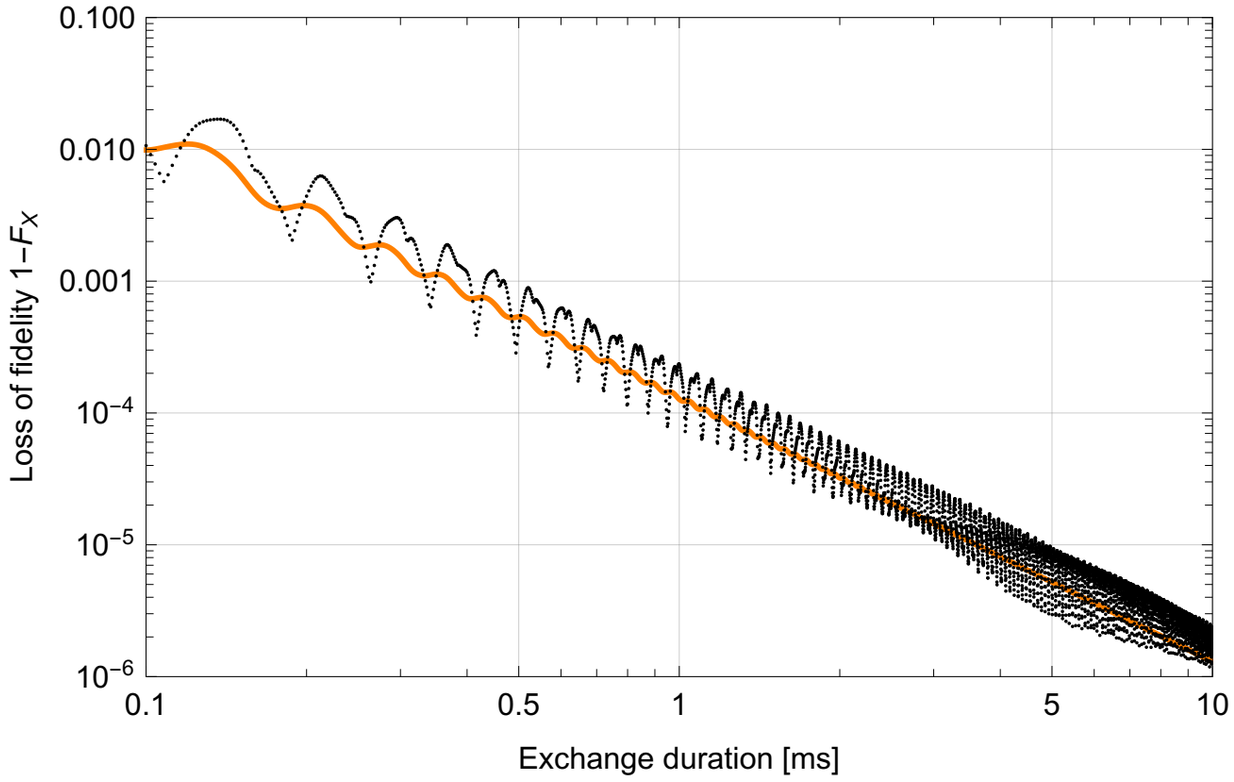


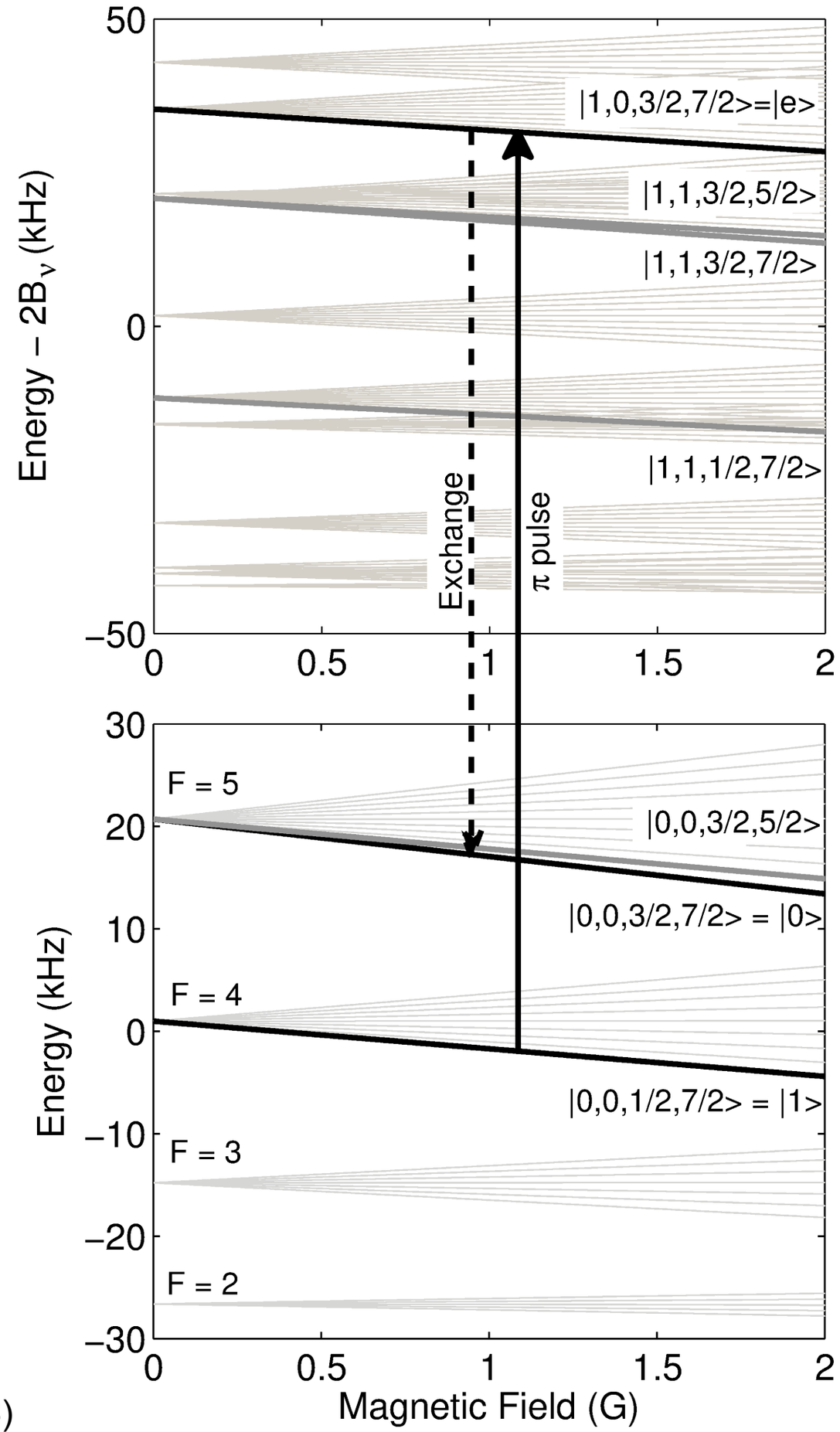
All states are field insensitive & long-lived
Exchange occurs at zero external E-fields

(Ni group) Chemical Science **9**, 6830-6838 (2018)
Hudson & Campbell, PRA 98, 040302 (2018)
motivated by Theoretical work: Demler, Lukin, Rey, Gorshkov; Exp work: Jin/Ye, Zwierlein

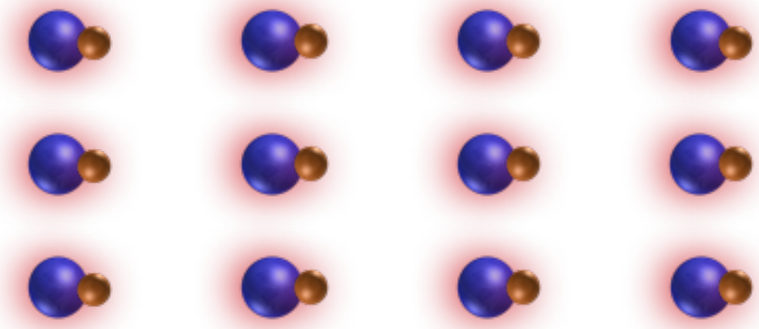
The full picture

- molecule of choice: NaCs
- exchange time of 50 μs, for F>99.6% @ B-field = 35G (limited by off-resonance excitations)
- exchange time of ~3ms (gate time~10ms), F>99.99% @ B-field = 1 G





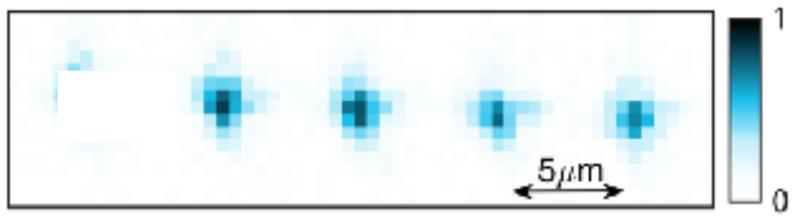
Ni, Rosenband, Grimes, Chemical Science **9**, 6830-6838 (2018)



building single molecules

in the rovibrational ground state
and the motional ground state

Laser-cooled molecules

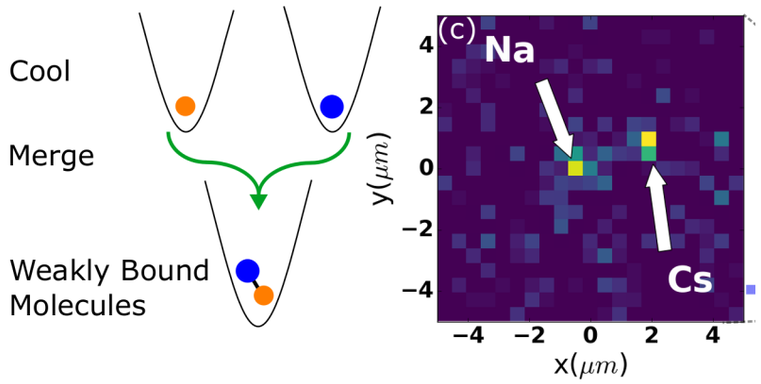


CUA Doyle/Ketterle/Ni Collaboration

Anderegg et al., Science **365**, 1156 (2019)

Other work: Molecular ions

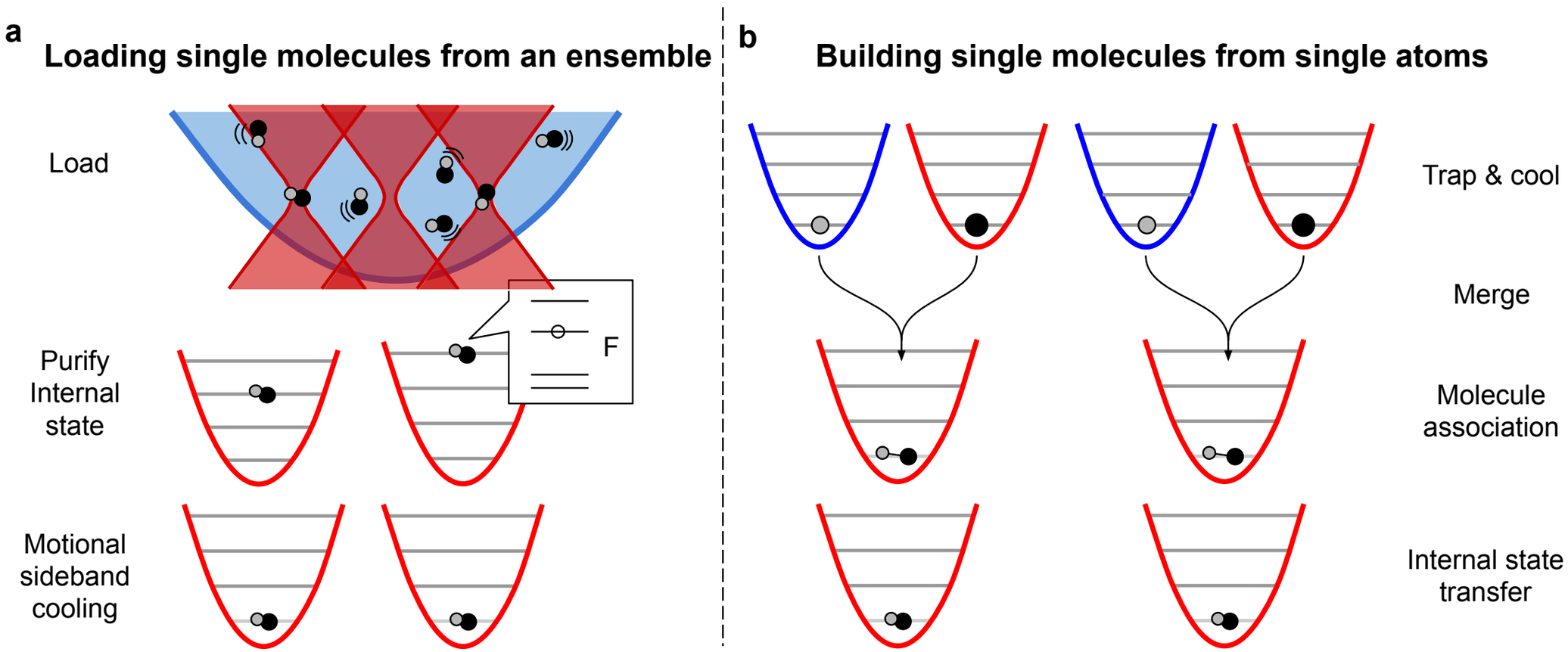
Assembly molecules from atoms



(Ni Group) Science **360**, 900 (2018)

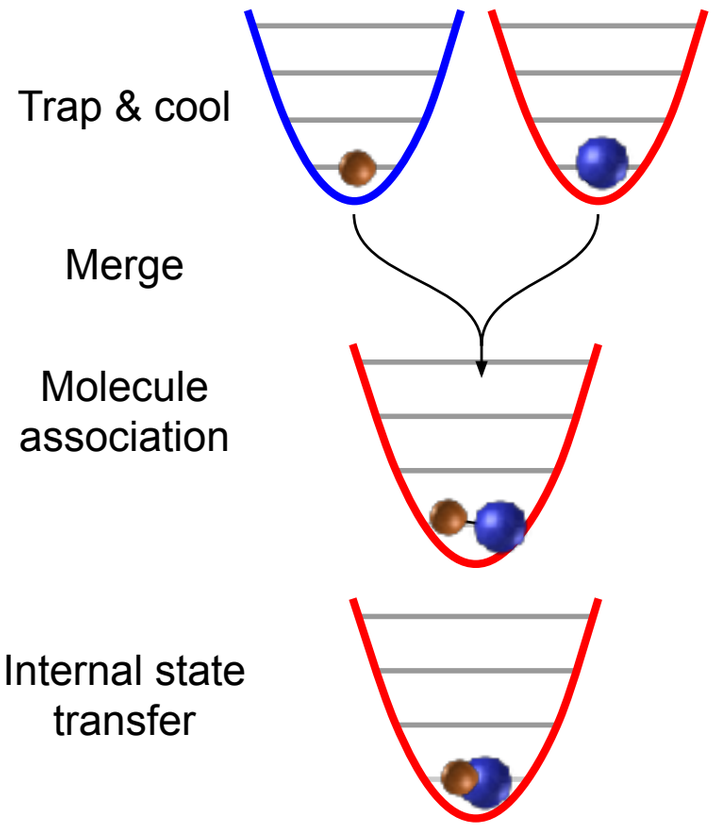
related work: Wuhan group

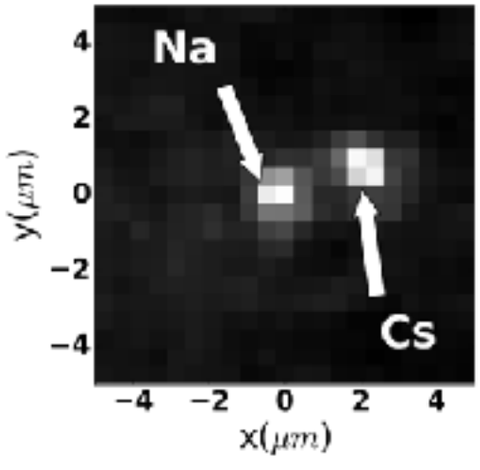
Two approaches



Building Single Molecules

Na+Cs → NaCs





New J. Phys. **19**, 023007 (2017)

Science **360**, 900 (2018)

Phys. Rev. A **97**, 063423 (2018)

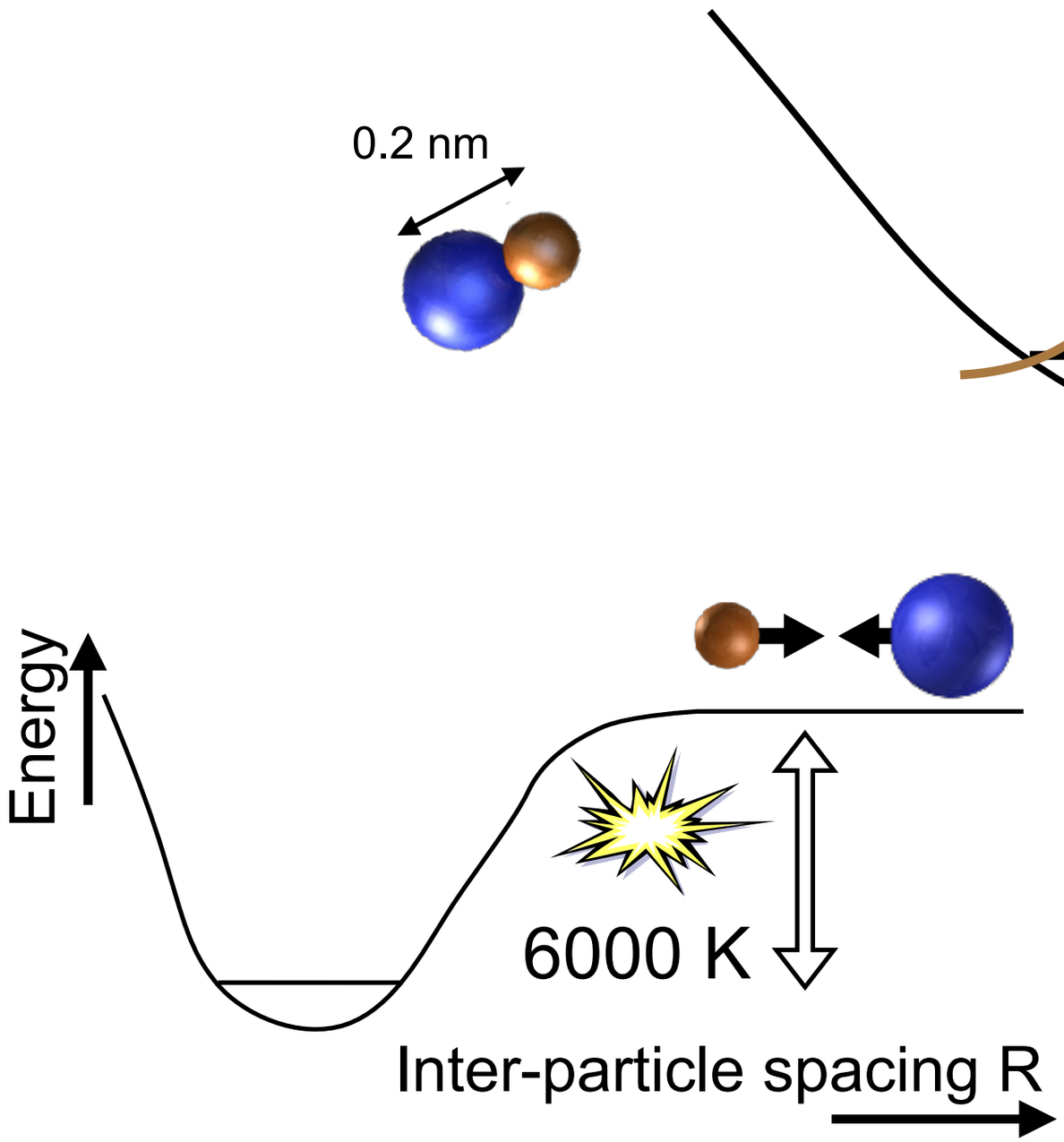
Phys. Rev. X **9**, 021039 (2019)

Phys. Rev. Lett. **124**, 253401 (2020)

Phys. Rev. Lett. **126**, 123402 (2021)

Associate Molecules from Ultracold Atoms

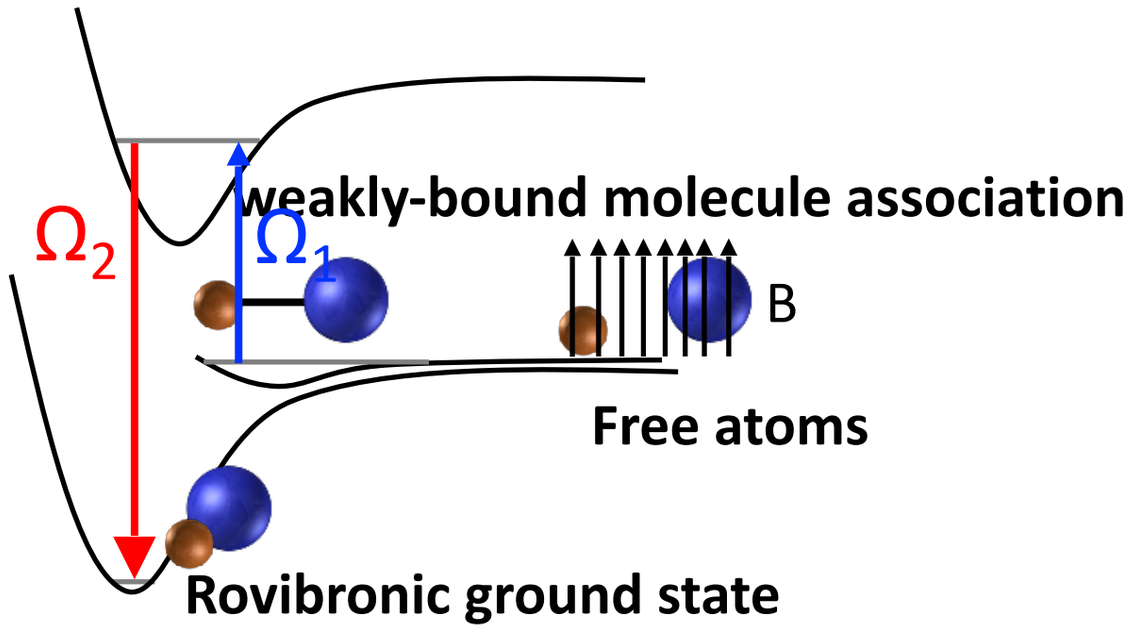
Main challenge: efficient molecule creation while staying cold



Associate Molecules from Ultracold Atoms

Two-step solution:

Step 1: bind atoms into weakly bound molecules through a Fano-Feshbach resonance, “Feshbach molecules” (this prepares them in a single quantum state)

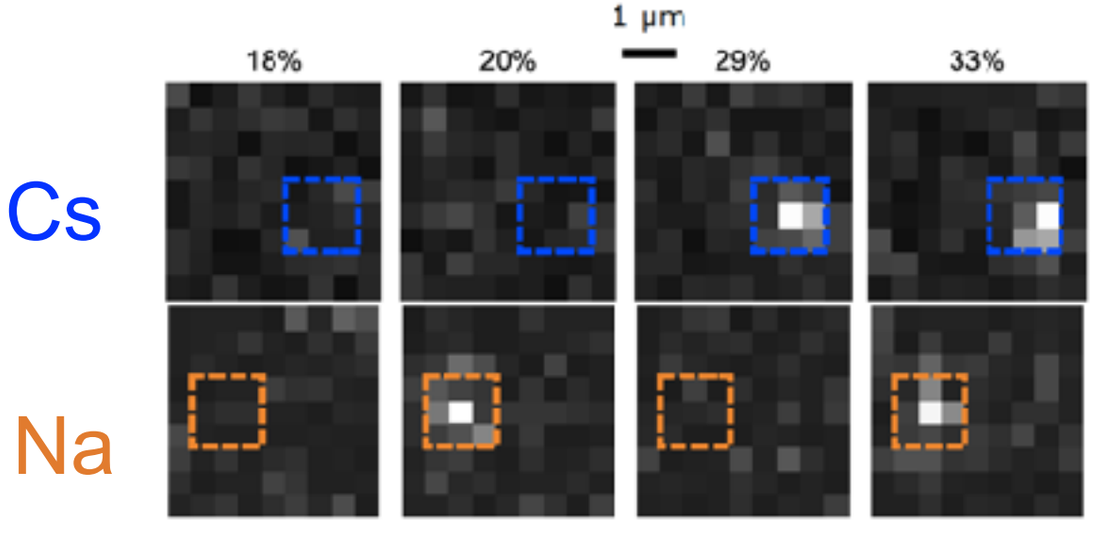


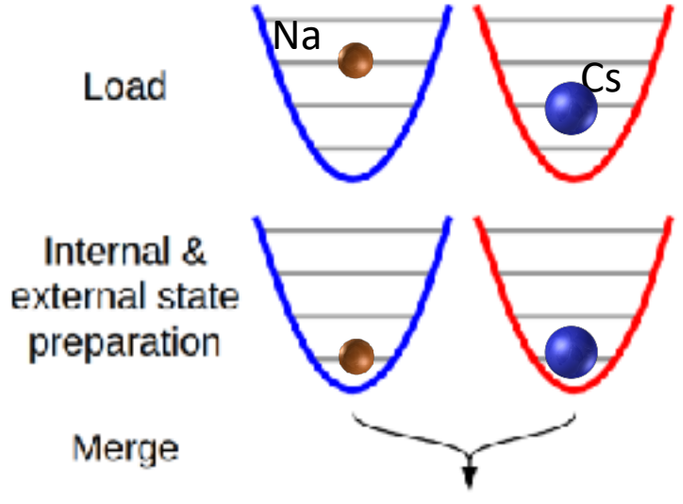
Step 2: two-photon coherent transfer to ro-vibrational ground state.
Stimulated photon takes away excess binding energy

from exactly two atoms,
we can make a
molecule with ~30%
efficiency

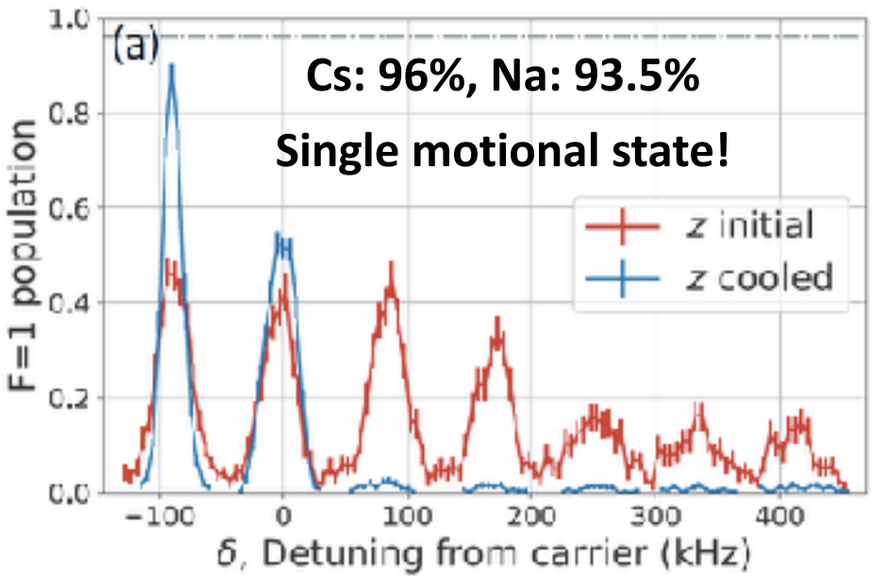
Innsbruck and JILA (2008)
Durham, MIT, Hong Kong,
MPQ, USTC, Harvard, Hannover

Atom state preparation

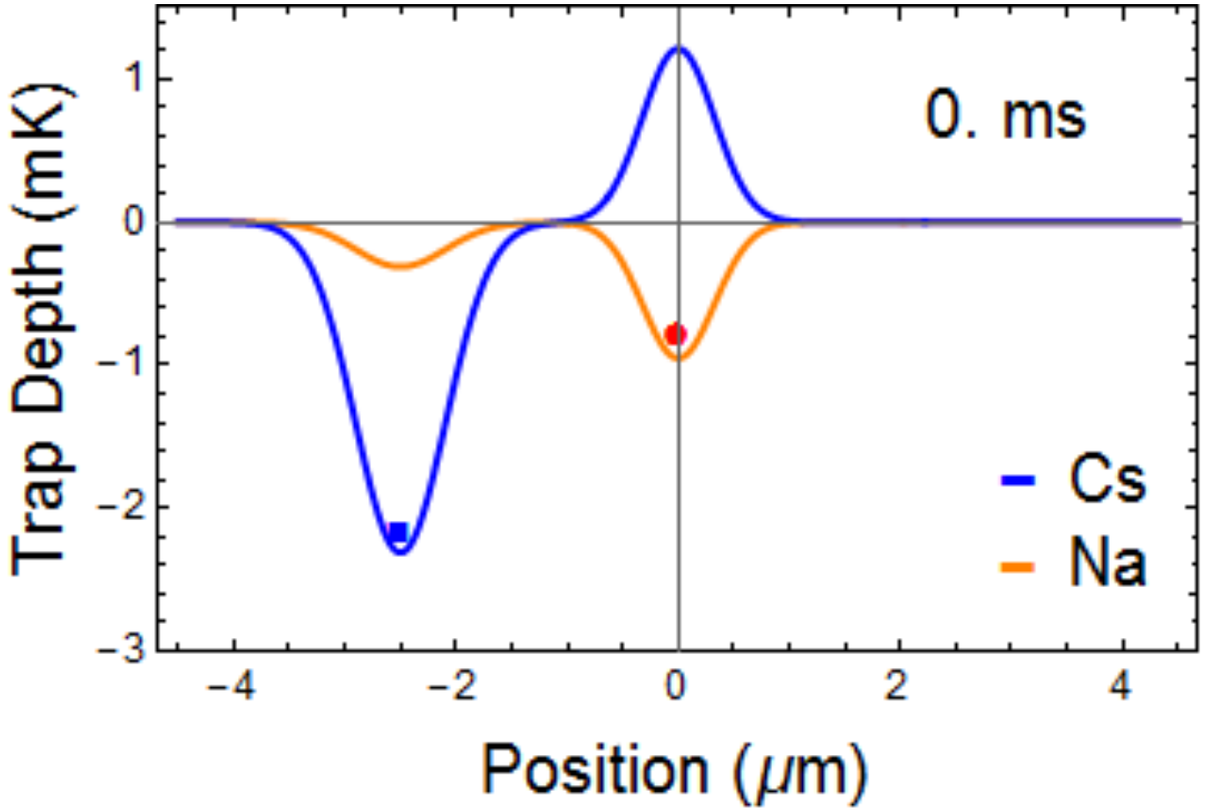




Raman sideband cooling



PRA 97, 063423 (2018)



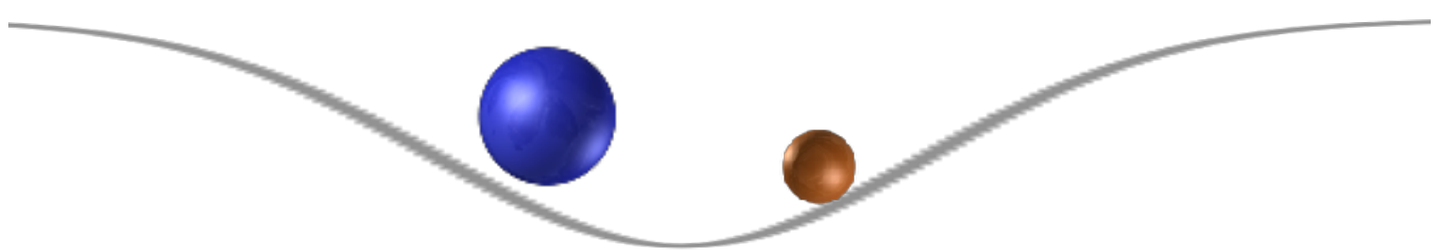
PRX 9, 021039 (2019)

Collisions of two atoms

- no reactions

(because needs to simultaneous fulfill conservations of energy and momentum)

- To make a molecule, additional stimulated fields (light or magnetic) will need to be added



$$H = \underbrace{\sum_{i=x,y,z} \left(\frac{m_1\omega_{1,i}^2 x_{1,i}^2}{2} + \frac{p_{1,i}^2}{2m_1}\right)}_{\text{Na}} + \underbrace{\sum_{i=x,y,z} \left(\frac{m_2\omega_{2,i}^2 x_{2,i}^2}{2} + \frac{p_{2,i}^2}{2m_2}\right)}_{\text{Cs}} + \underbrace{V_{int}(\vec{r}_1 - \vec{r}_2)}_{\text{Interaction}}$$

To center of mass
and relative coordinates

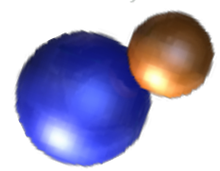
$$M = m_1 + m_2 \qquad \mu = \frac{m_1 m_2}{m_1 + m_2}$$

$$\Omega_i^2 = \frac{m_1\omega_{1,i}^2 + m_2\omega_{2,i}^2}{m_1 + m_2} \qquad \omega_{R,i}^2 = \frac{m_2\omega_{1,i}^2 + m_1\omega_{2,i}^2}{m_1 + m_2}$$

$$X_i = \frac{m_1 x_{1,i} + m_2 x_{2,i}}{m_1 + m_2} \qquad x_{R,i} = x_{1,i} - x_{2,i}$$

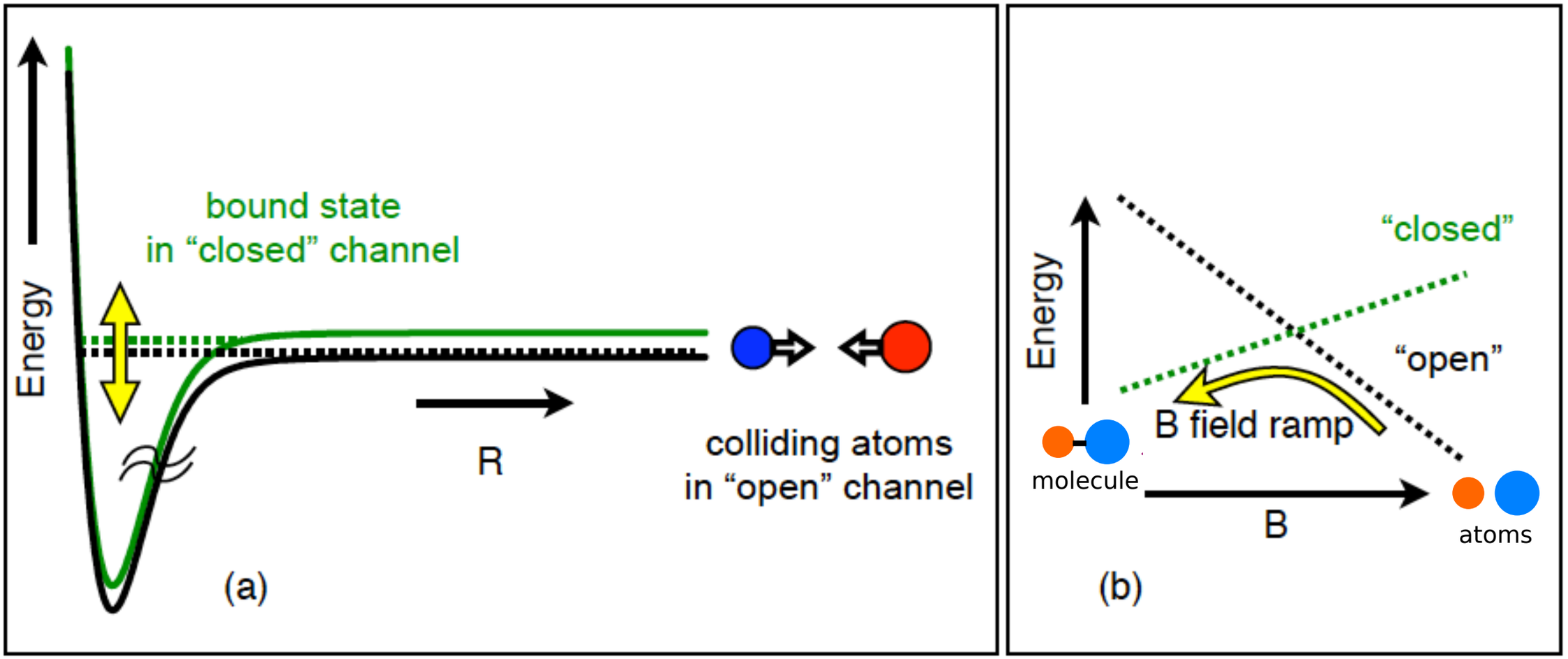
$$P_i = p_{1,i} + p_{2,i} \qquad p_{R,i} = \frac{m_2 p_{1,i} - m_1 p_{2,i}}{m_1 + m_2}$$

Turning a two-body problem to an one-body problem



$$H = \overbrace{\sum_{i=x,y,z} \left(\frac{M\Omega_i^2 X_i^2}{2} + \frac{P_i^2}{2M}\right)}^{\text{Center of mass}} + \overbrace{\sum_{i=x,y,z} \left(\frac{\mu\omega_{R,i}^2 x_{R,i}^2}{2} + \frac{p_{R,i}^2}{2\mu}\right) + V_{int}(\vec{r}_R)}^{\text{Relative}} + \overbrace{\sum_{i=x,y,z} \mu(\omega_{1,i}^2 - \omega_{2,i}^2) X_i x_{R,i}}^{\text{Mixing}}$$

Ramp Through a Fano-Feshbach Resonance

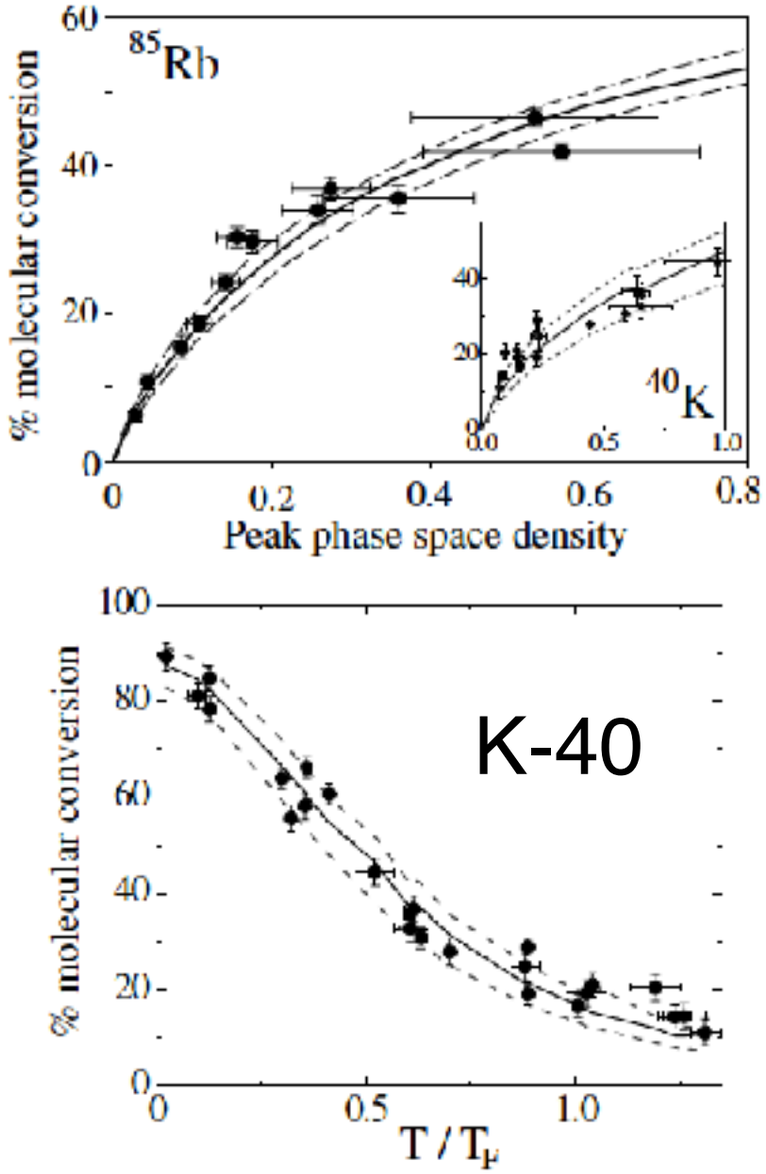


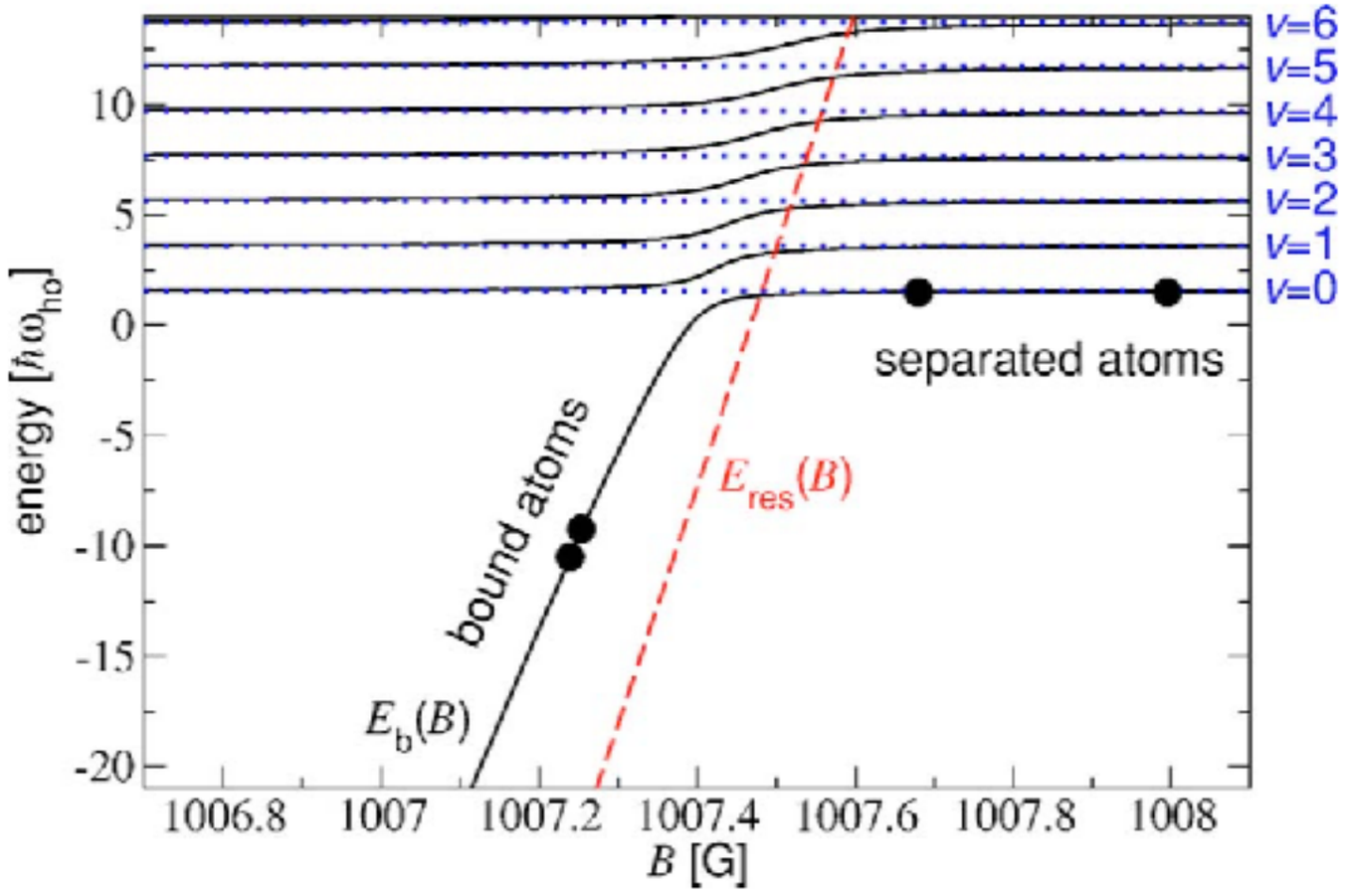
Feshbach Molecule Creation Efficiency

Most important parameters: PSD

For heteronuclear species, competing factors including cloud overlap and 3-body loss

Efficiency ~ 15%



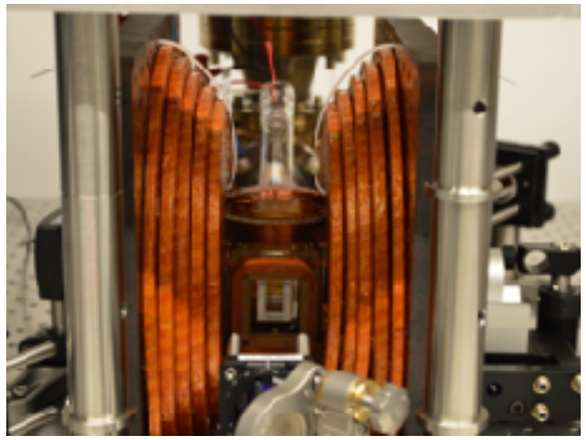


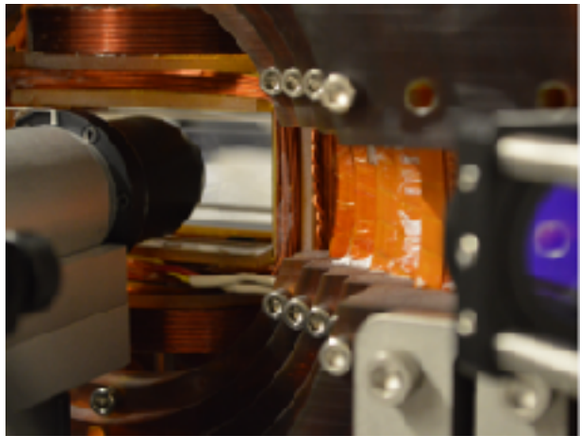
Hodby..Regal..Greiner..Jin..Cornell…Wieman, PRL 94, 120402 (2005)

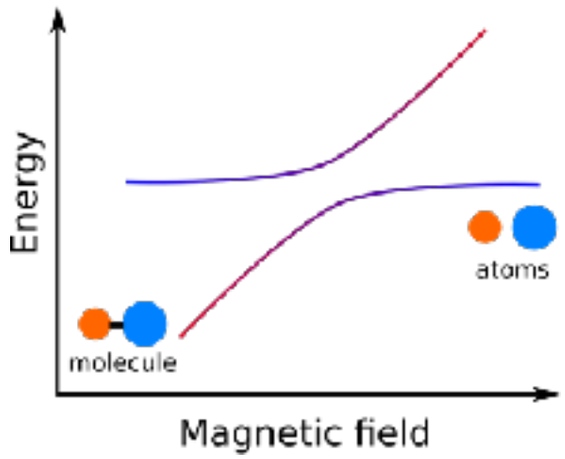
Köhler, Góral, and Julienne. Rev. Mod. Phys. vol 78, 1311 (2008)

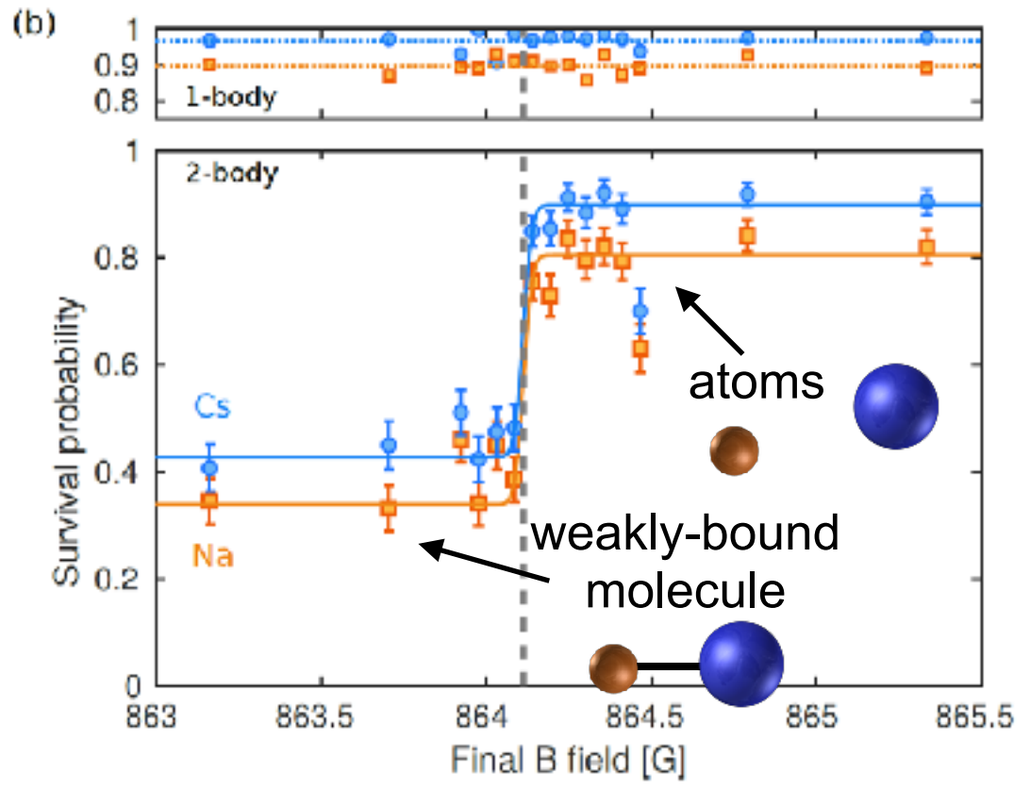
Weakly-bound molecule formation

* Absolute ground state atoms → External + rotational ground state molecules







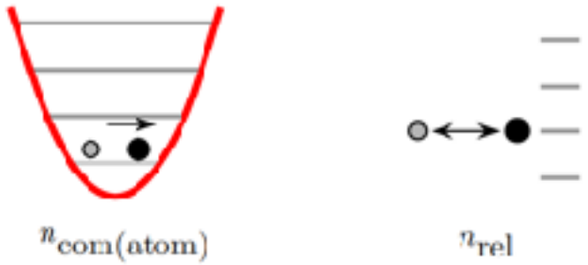


PRL **124**, 253401 (2020)

Mapping total control of atomic states to single molecules

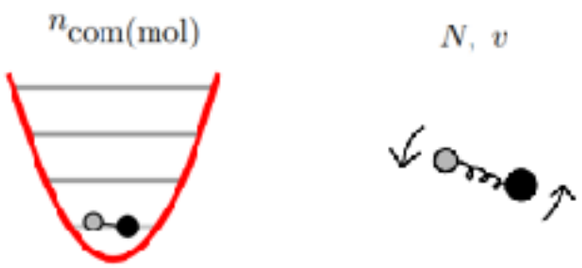
COM + Relative

Atom pair motional states



Magnetoassociation

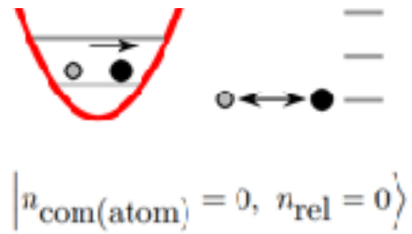
Molecular states



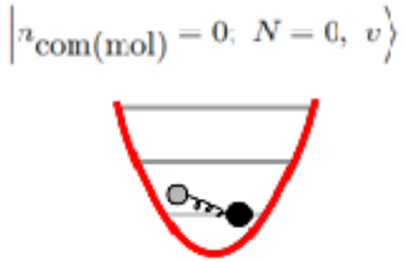
External

Internal - rotational, vibrational

S-wave Scattering resonance



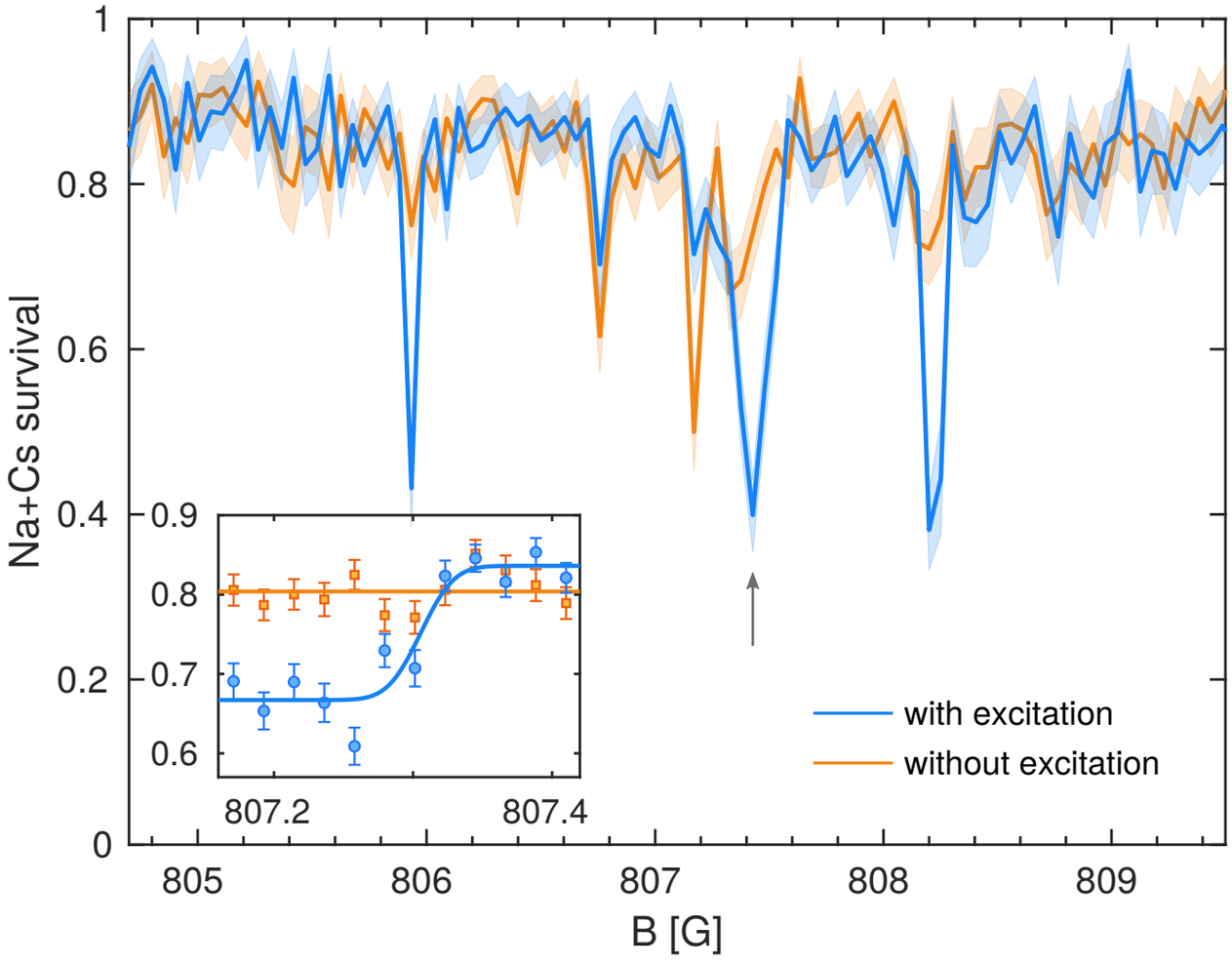
absolute ground state atoms



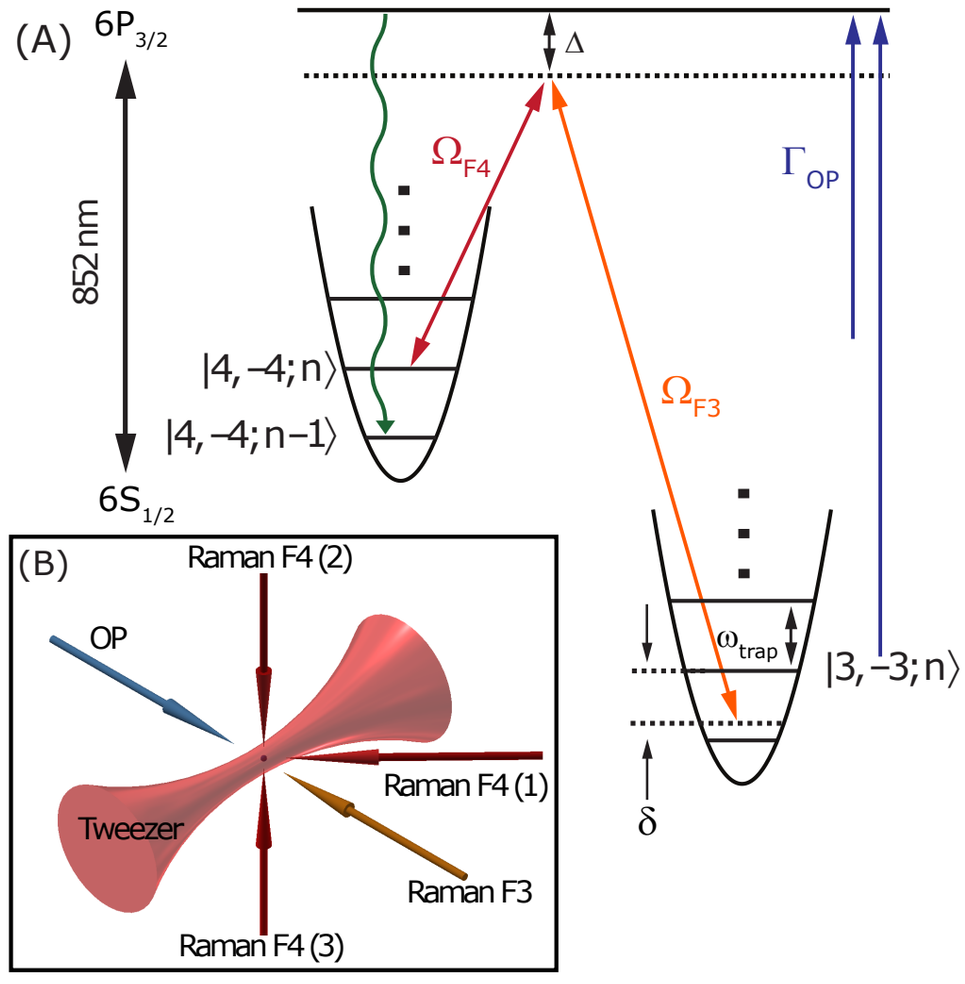
external + rotational ground state molecules

Mapping an Atom motional excitation to a Molecular Internal state excitation

P-wave molecules



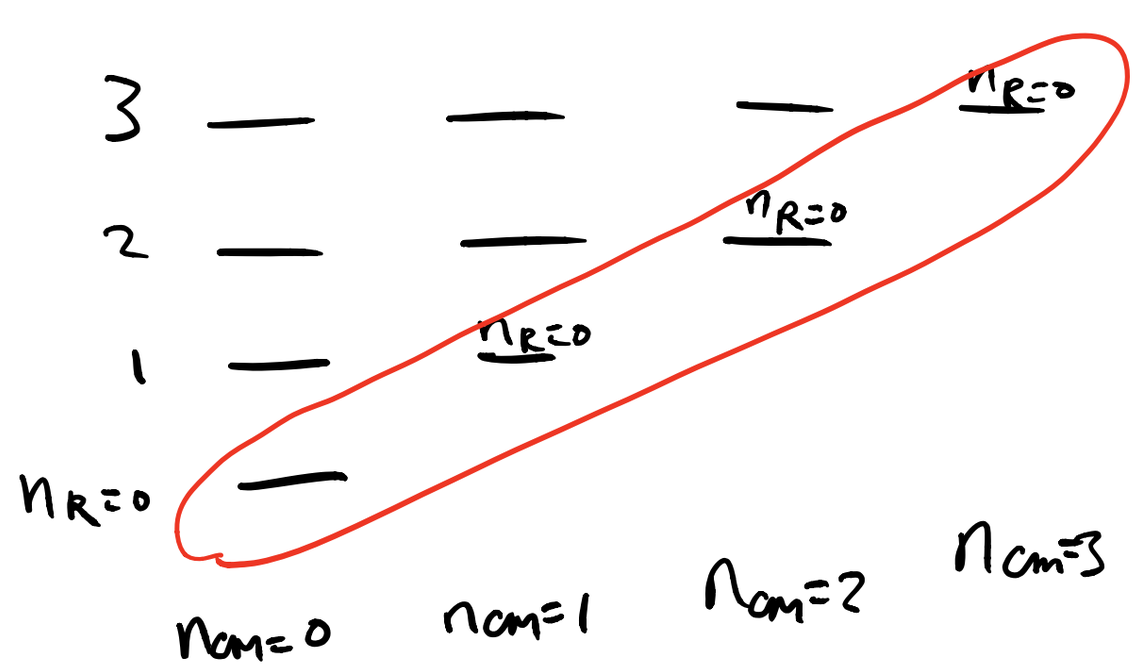
$\Delta n = 1$



Molecular Internal and External state control

- Molecule conversion efficiency
  - Determined by relative motional ground state population of atom pair
  - ~45-50% (of initial loaded atoms)
  - Not fundamentally limited
- Molecular state purity
  - Internal state pure
  - External state determined by COM motional state population of atom pair
    - ~77% in ground state

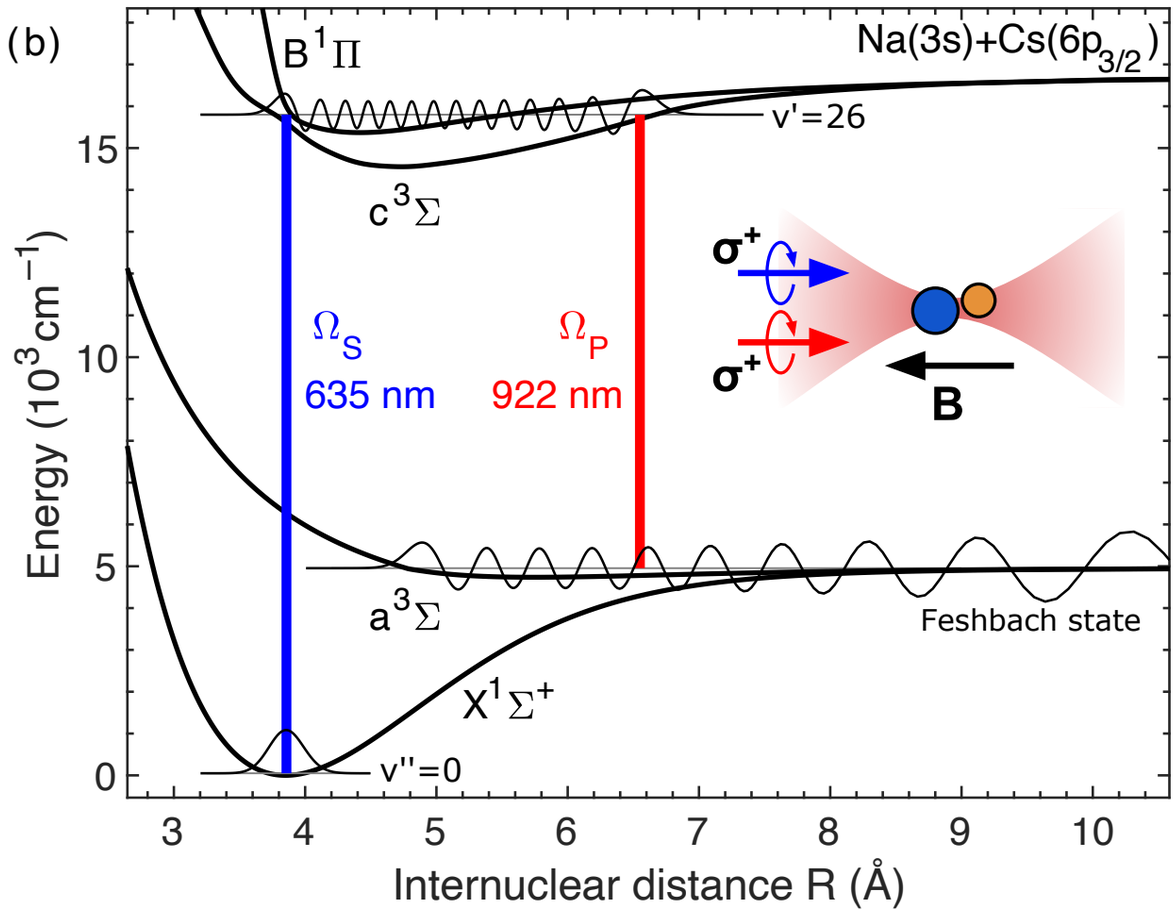
$$P(n_{\text{mol-com}}=0)=\frac{P(n_{\text{rel}}=0,n_{\text{com}}=0)}{P(n_{\text{rel}}=0)}$$
$$=1-\frac{m_{\text{Na}}}{M}\frac{\bar{n}_{\text{Na}}}{\bar{n}_{\text{Na}}+1}-\frac{m_{\text{Cs}}}{M}\frac{\bar{n}_{\text{Cs}}}{\bar{n}_{\text{Cs}}+1}.$$



Zhang et al., PRL **124**, 253401 (2020)

single NaCs molecules in the internal and motional *ground* state

Lifetime in excess of 3s

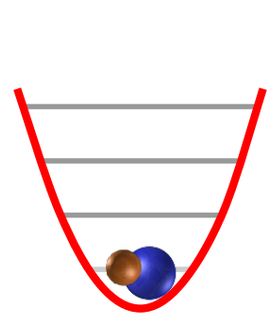


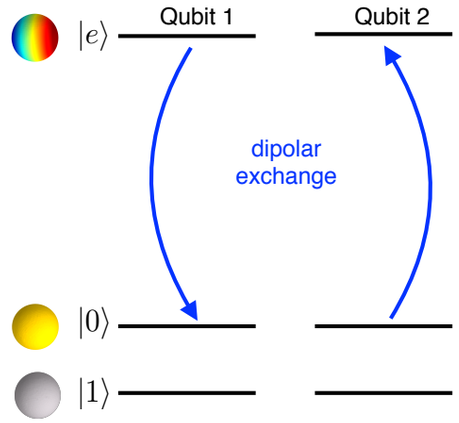
W. B. Cairncross*, J. T. Zhang*, L. R. B. Picard, Y. Yu, K. Wang, K.-K. Ni

Phys. Rev. Lett. 126, 123402 (2021)

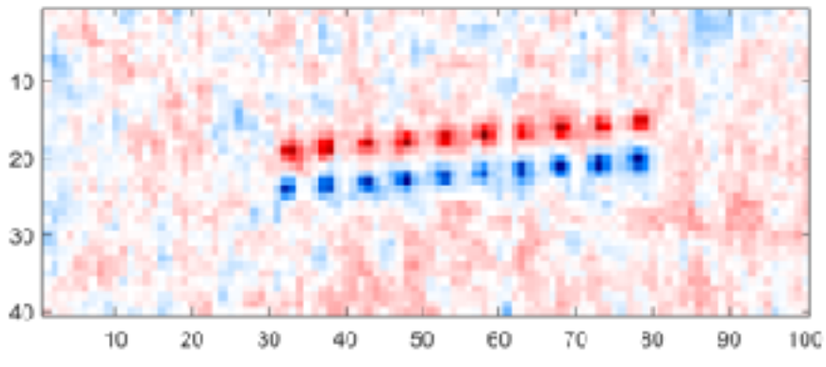
One molecular qubit is now fully controlled!

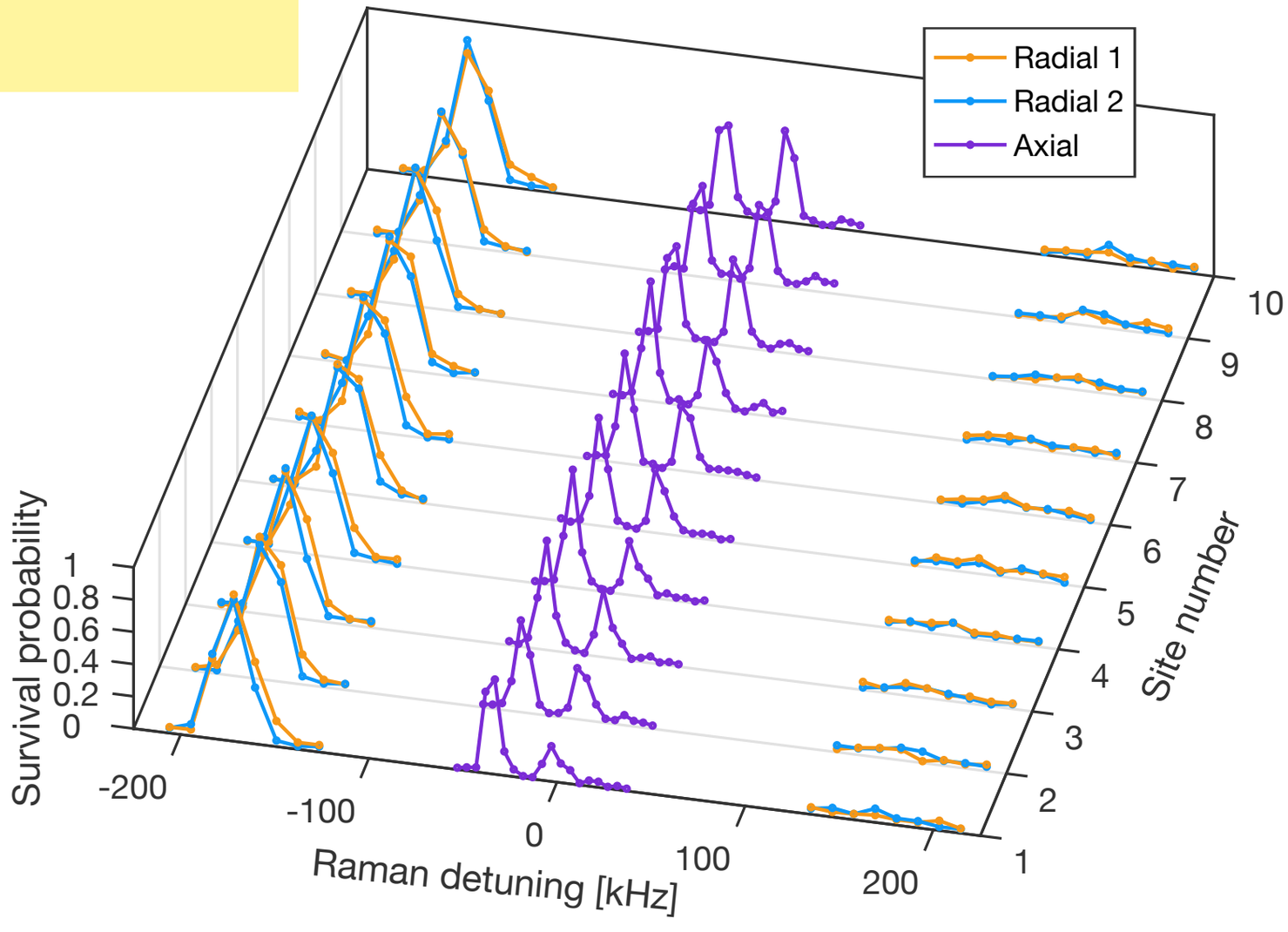
Next: Scaling up and entangle them





10 Na and 10 Cs atoms side-by-side





* 3D ground-state cooling in 10 traps, nbar=0.06(22), 0.07(2), 0.05(2)