Quantum gases of polar molecules

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Leibniz Universität Hannover, Germany

Outline

- Why quantum gases of molecules?
- How to prepare?
- Collisions and collisional control
- Outlook

Our research

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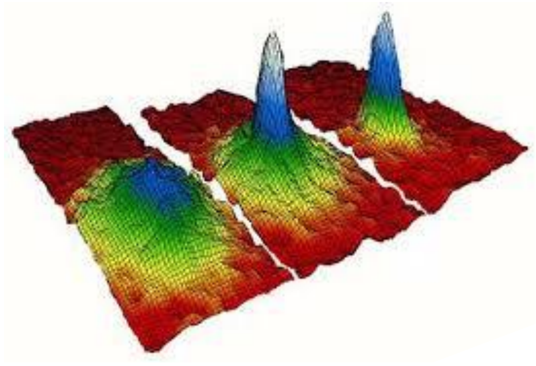
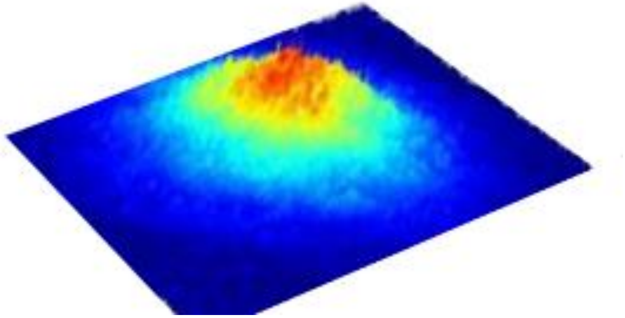
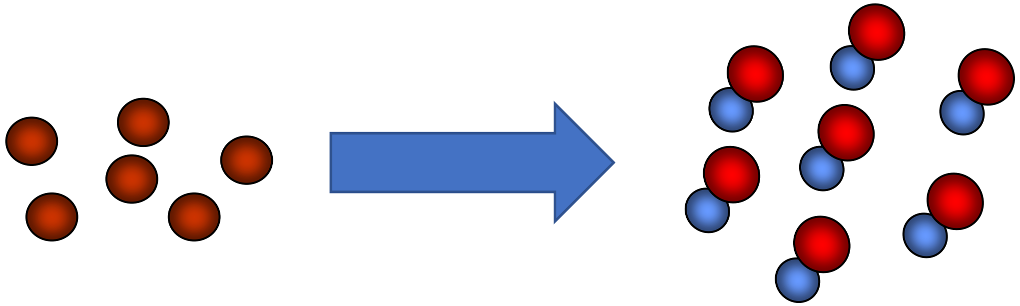
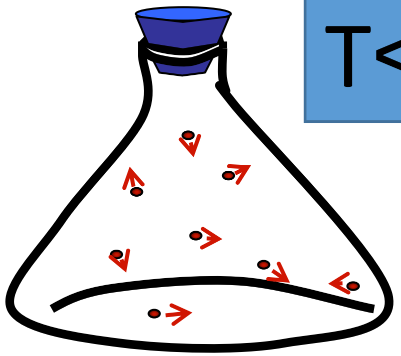


Quantum degenerate gases



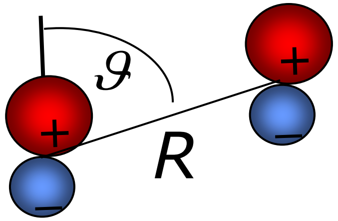




T<1 μK

Perspectives: Quantum physics

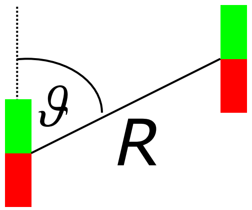
Polar molecules: Electric dipoles



d ~ Debye

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

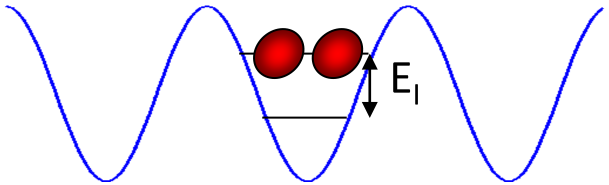
Atoms: Magnetic dipoles



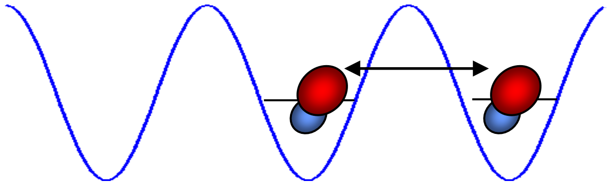
d ~ Bohr magneton

Quantum phases and dynamics of many-body systems with finite range interactions

Contact interactions

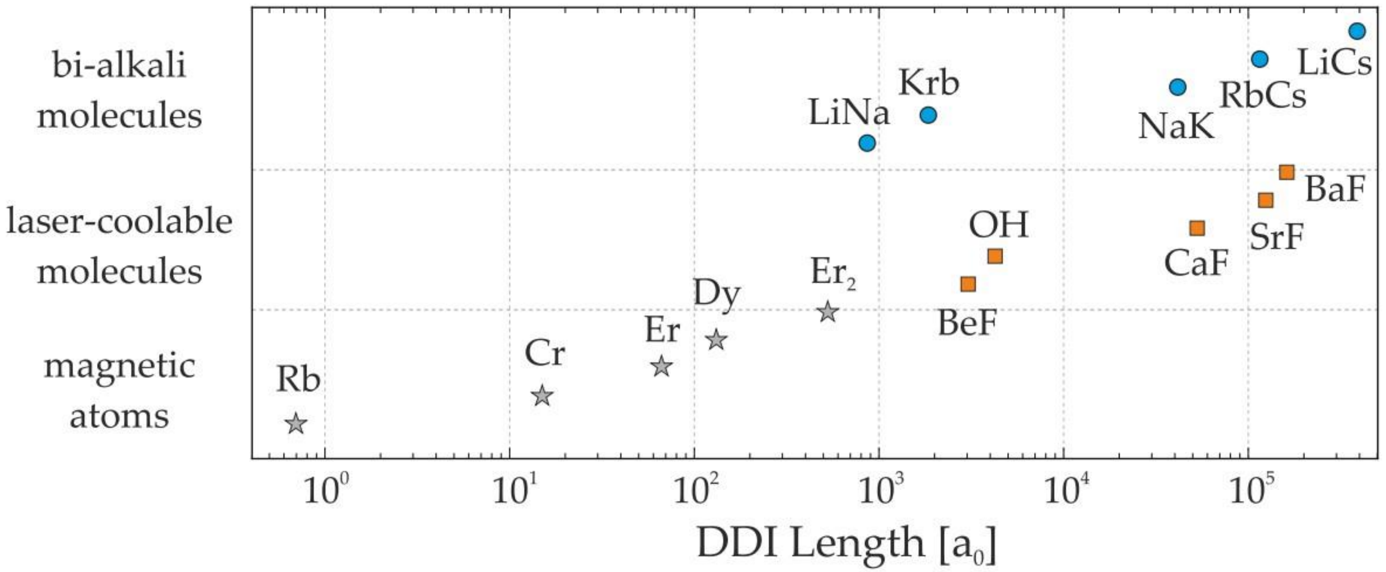


Dipole-dipole interactions



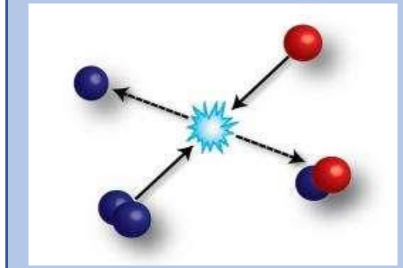
Why ultracold molecules?

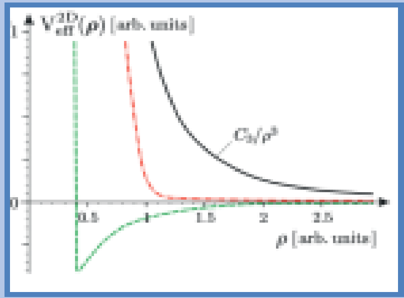
Dipole dipole interactions length selected atoms/molecules

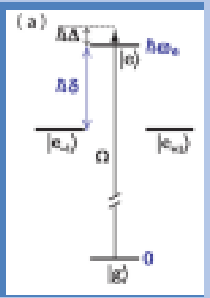


Perspectives: Collisions

Collisions and collisional control



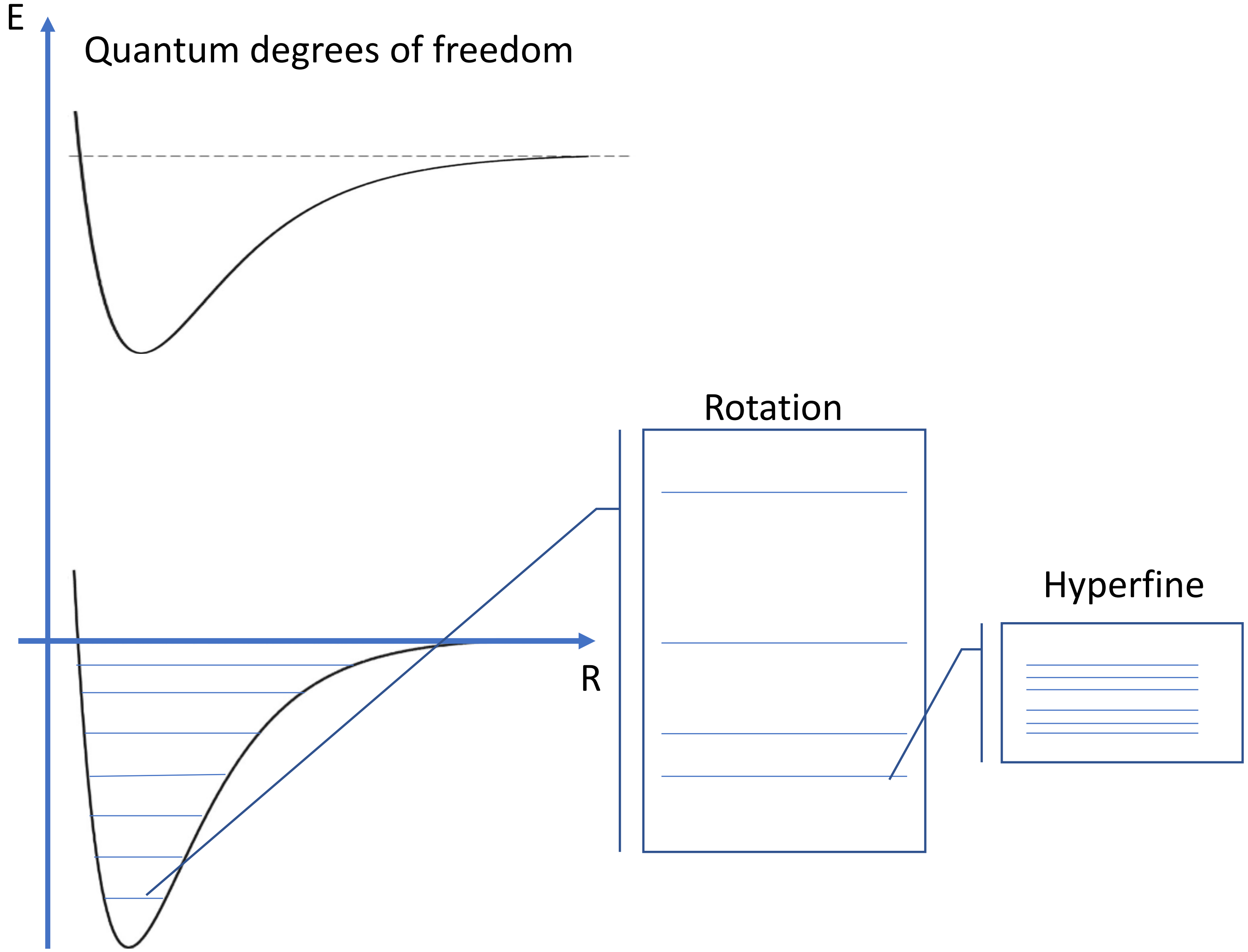




- Collisions and collisional control

- Quantum chemistry
- Complex formation and sticky collisions

Preparing ultracold molecules

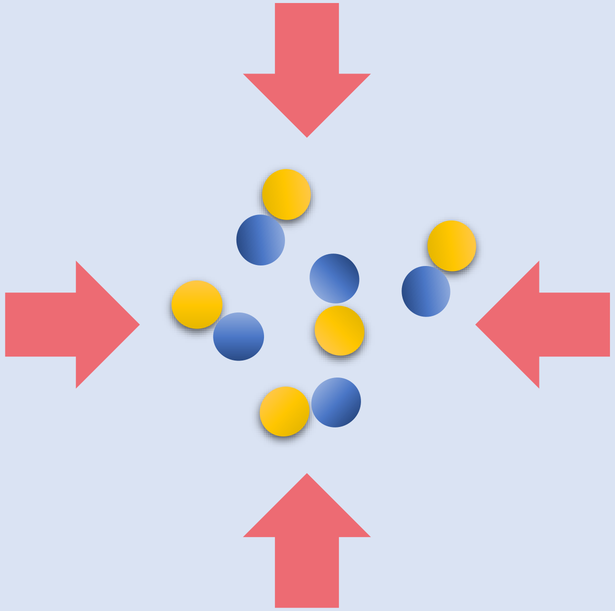


Complicates

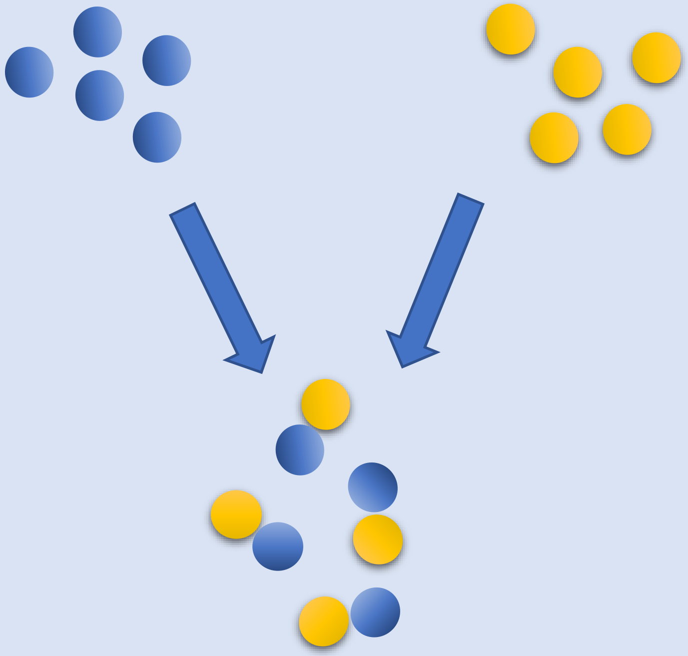
- Interaction with light
- Collisional processes

Two approaches to ultracold molecules

Direct laser cooling of molecules

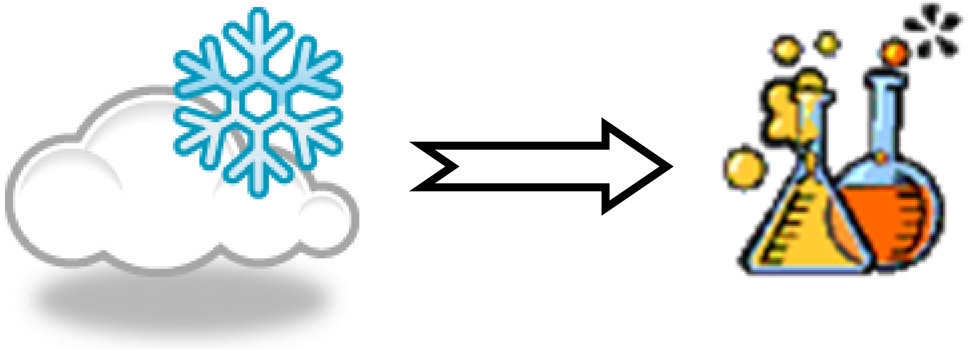


Association of ultracold atoms to bialkali molecules



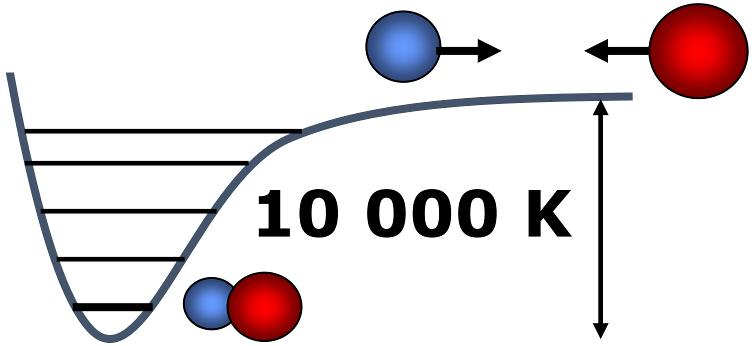
Association of atoms to molecules

Controlled chemistry at ultracold temperatures



Advantage: Start ultracold

Challenge: Staying ultracold

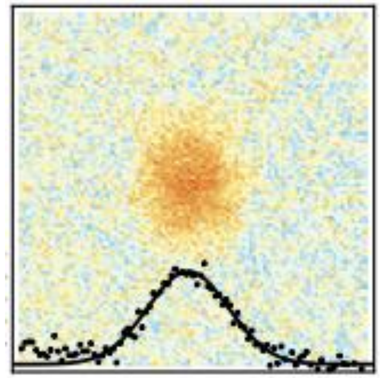


Ultracold dense gas of bosonic $^{23}Na^{39}K$

Hannover

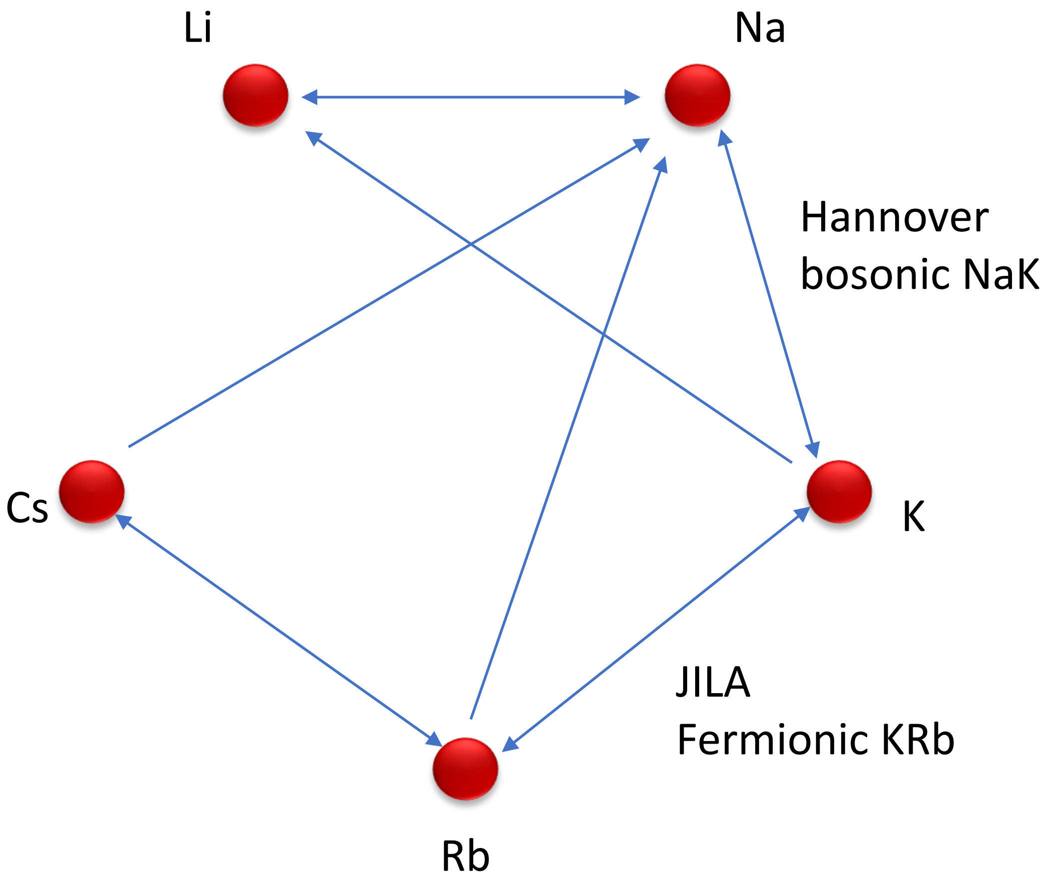
- 15 000 molecules in rovibrational ground state
- 300 nK in cODT at 1064 nm
- ρ~0.6

Quantum gas of bosonic polar $^{23}Na^{39}K$ molecules



Phys. Rev. Lett. 125, 083401 (2020)

New J. Phys. 21, 123034 (2019)

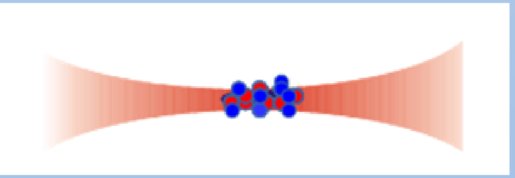


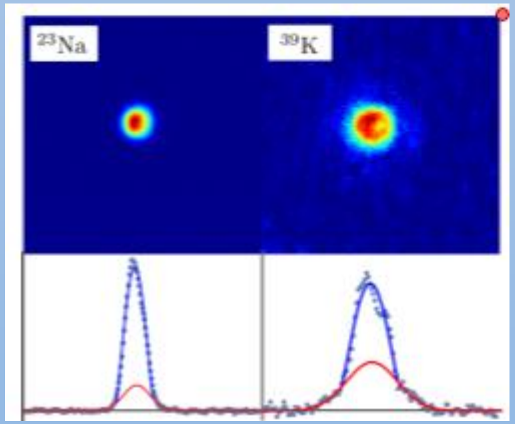
Preparation of ultracold bialkali molecules

Preparation follows the three-step process from the pioneering $^{40}K^{87}Rb$ JILA experiment.

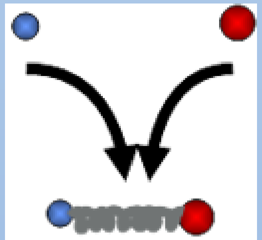
Ni, Ospelkaus et al. Science 322, 231 (2008), Ospelkaus, Ni et al. Faraday Discussions 142, 351 (2009).

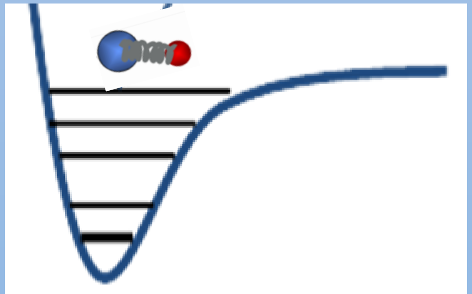
Dual species quantum gas mixture



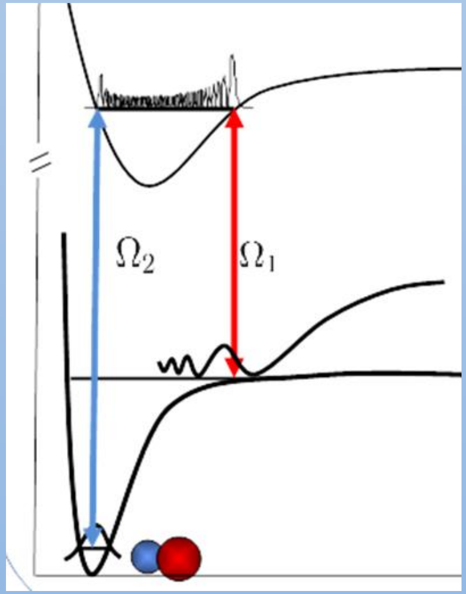


Feshbach molecule creation

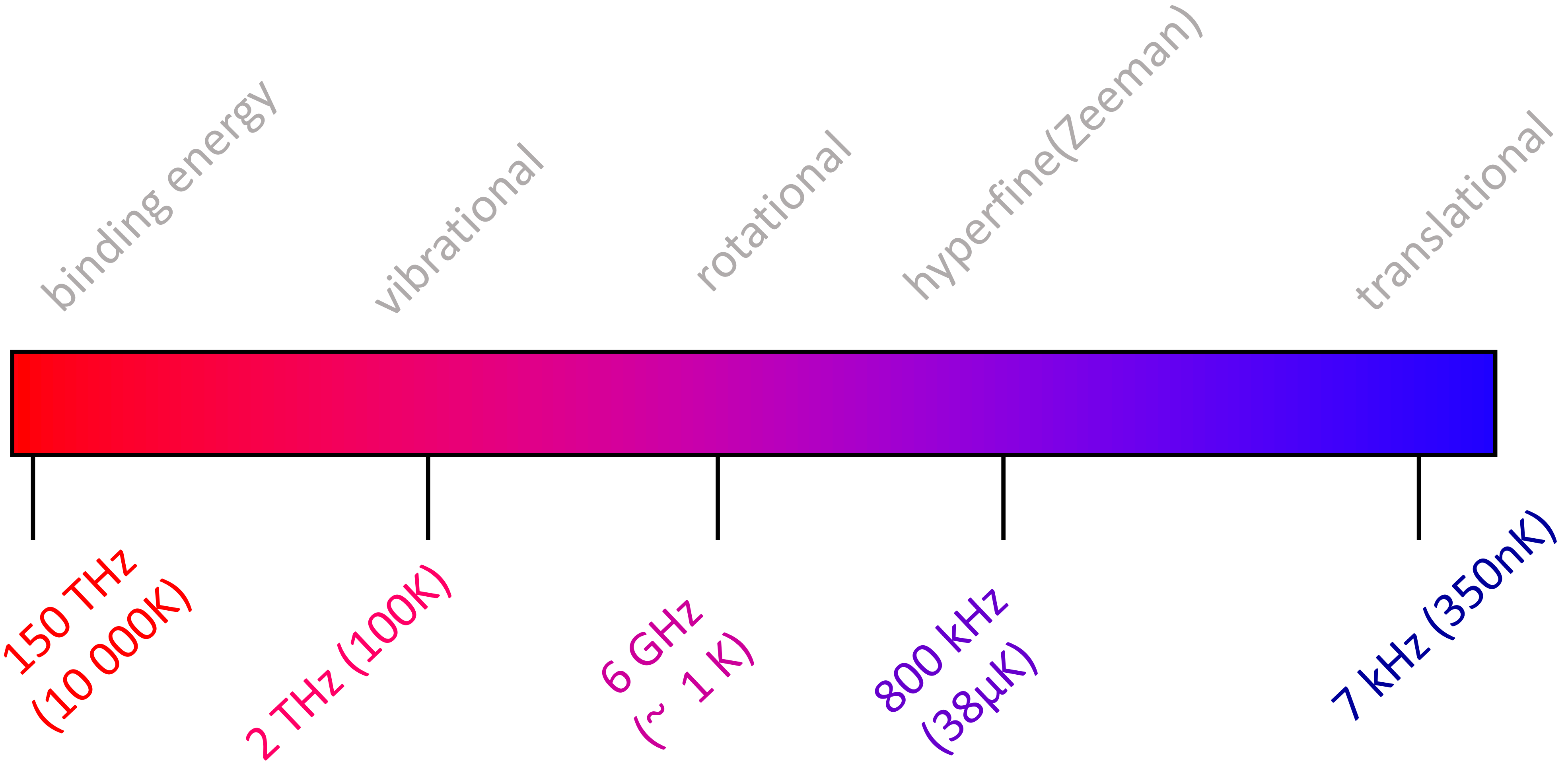




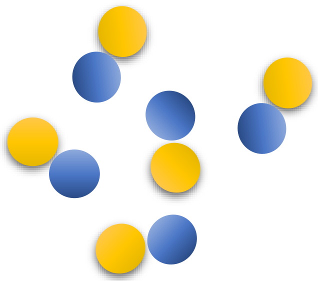
Two-photon transfer (STIRAP) to rovibrational ground state (v=0, N=0)



Energy scale



Molecule-molecule collisions

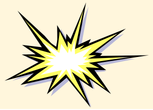


Collisions of ultracold molecules?

Chemically reactive

	Na	K	Rb	Cs
Li				
Na		NR	NR	NR
K				NR
Rb				NR

Chemically reactive molecules:
Exoergic chemical reaction

XY+ XY → X_2+Y_2 + 

Non-chemically reactive molecules:
Endoergic exchange reaction
Forbidden at ultracold temperature

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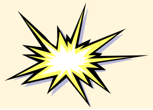
$XY + XY \rightarrow X_2 + Y_2 +$ 

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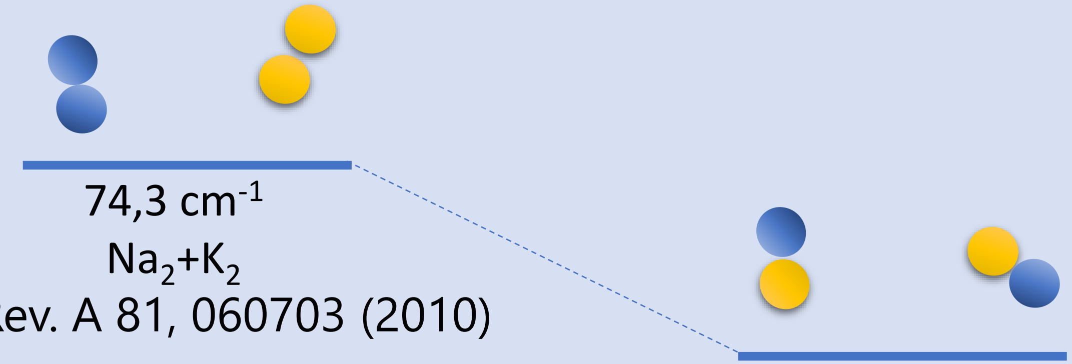
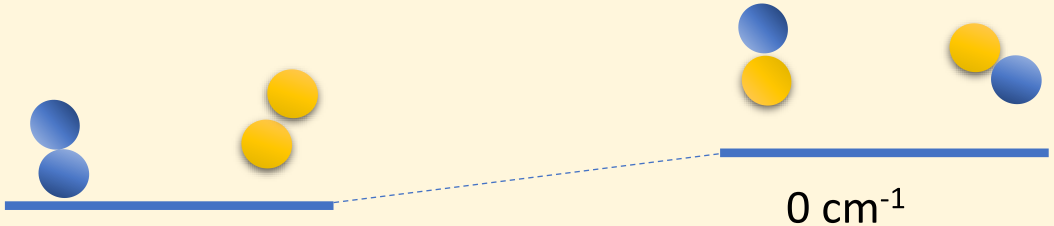


Phys. Rev. A 81, 060703 (2010)



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Collisions of ultracold molecules?

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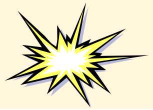
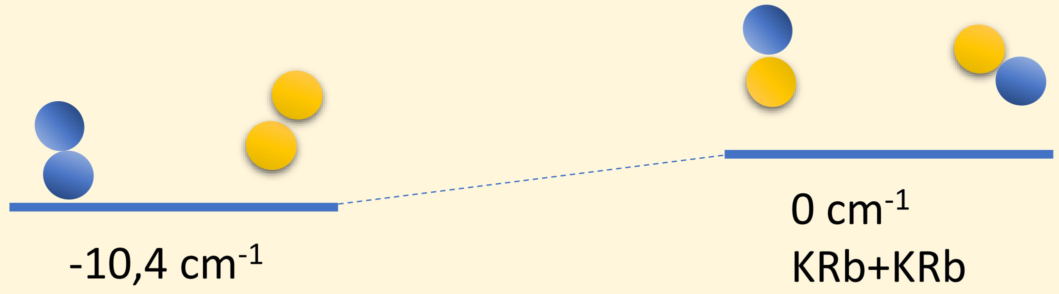
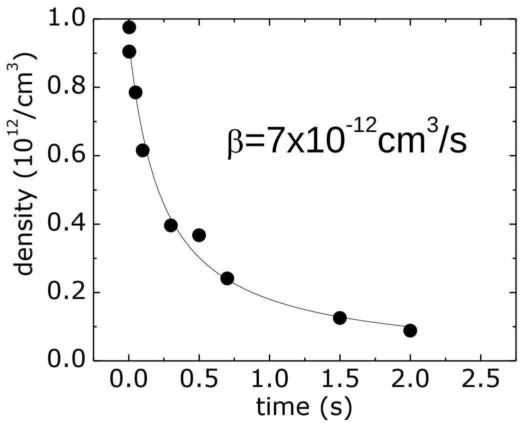
XY+ XY → X_2+Y_2 + 



Phys. Rev. A 81, 060703 (2010)

- 2010: Fast two-body loss at universal limit due to exoergic exchange reaction in KRb
- 2019: Direct observation using VMI



Two-body loss

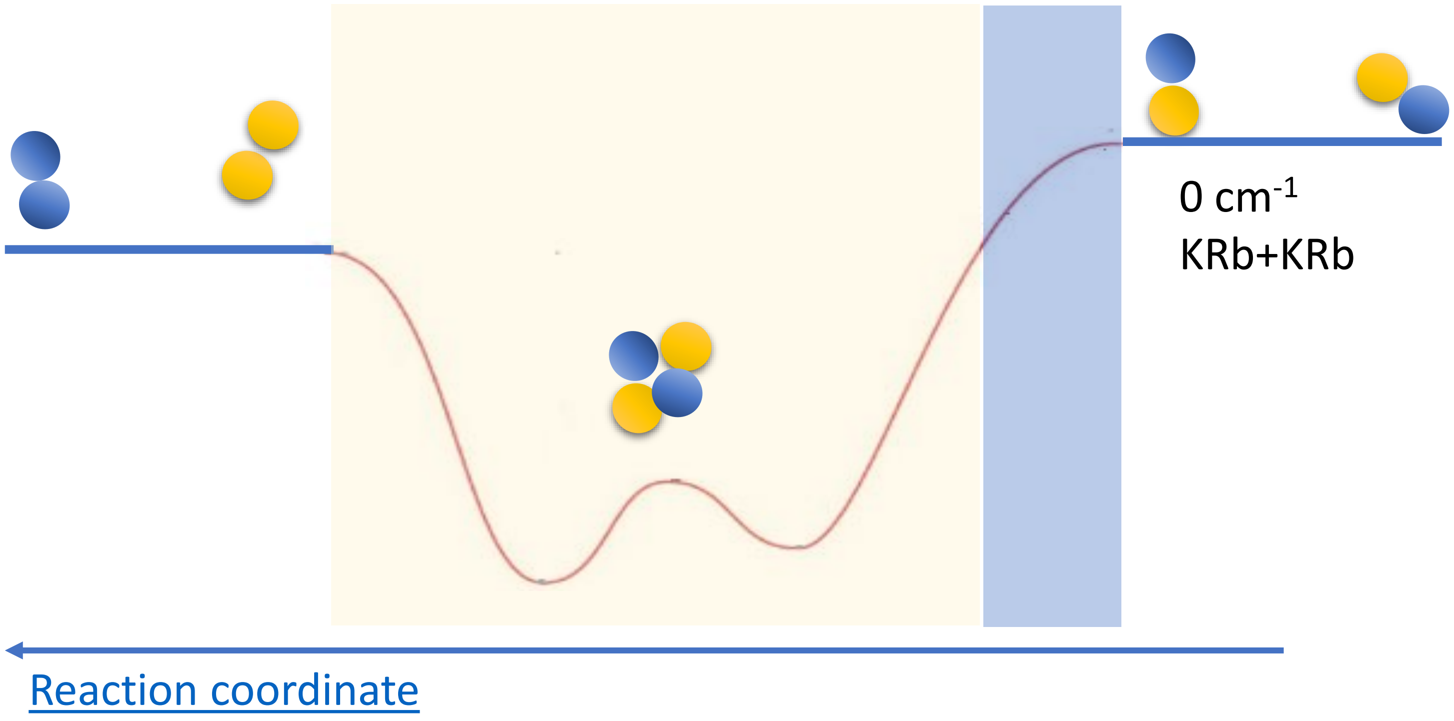
$$\dot{n}(t) = -\beta n(t)^2$$

$$n(t) = \frac{n_0}{1+n_0\beta t}$$

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

Chemical reactions rate – simple model

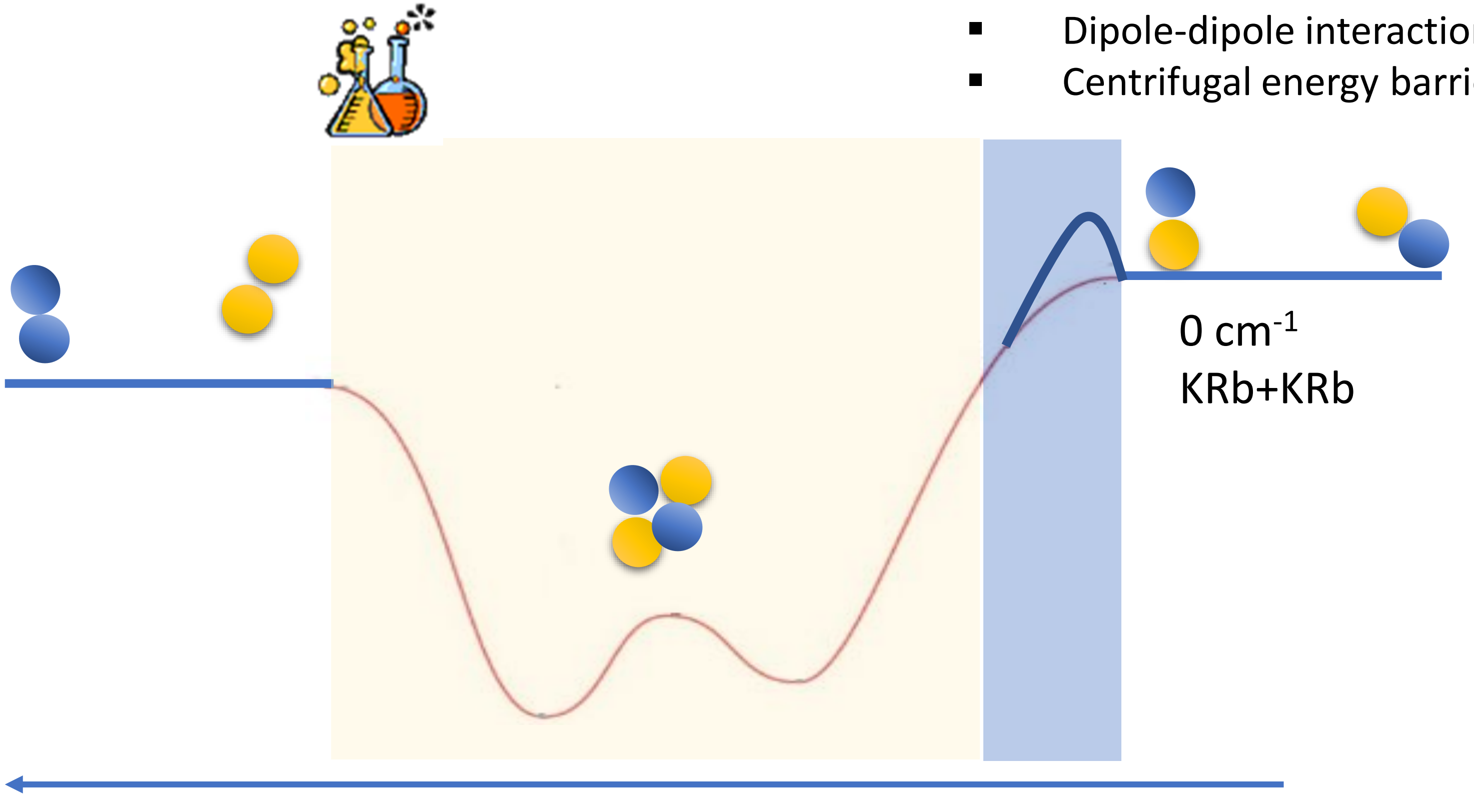
Long-range: e.g.
- van der Waals interaction
- Dipole-dipole interactions
- Centrifugal energy barriers



Chemical reactions rate – simple model

Short-range
→ Unity probability for loss

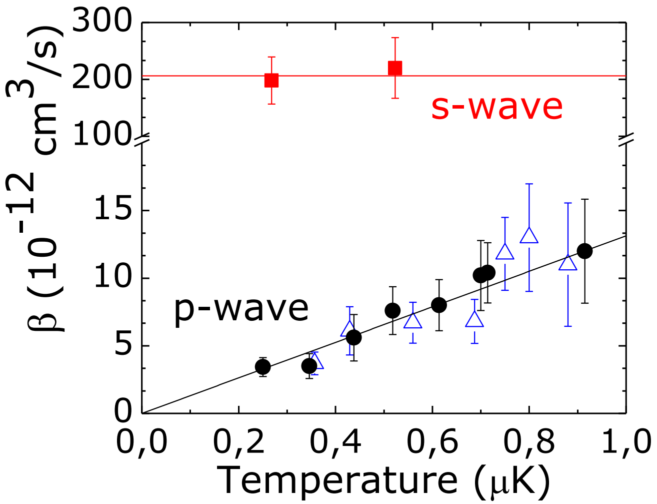
Long-range: e.g.
- van der Waals interaction
- Dipole-dipole interactions
- Centrifugal energy barriers



Long-range for $^{40}K^{87}Rb$ molecules?
- Single quantum state fermionic molecules
- Ultracold

→ p-wave scattering
→ centrifugal energy barrier ~16µK

$$\sim l(l+1)/r^2$$



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

Collisions of ultracold molecules?

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Non-chemically reactive molecules:
Endoergic exchange reaction
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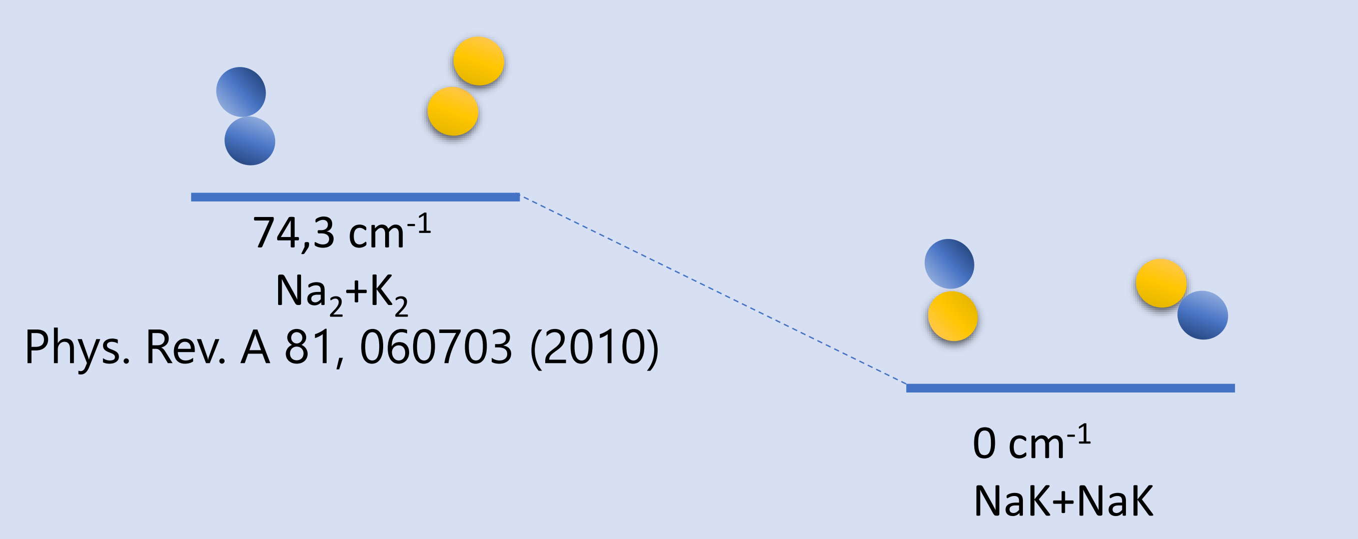
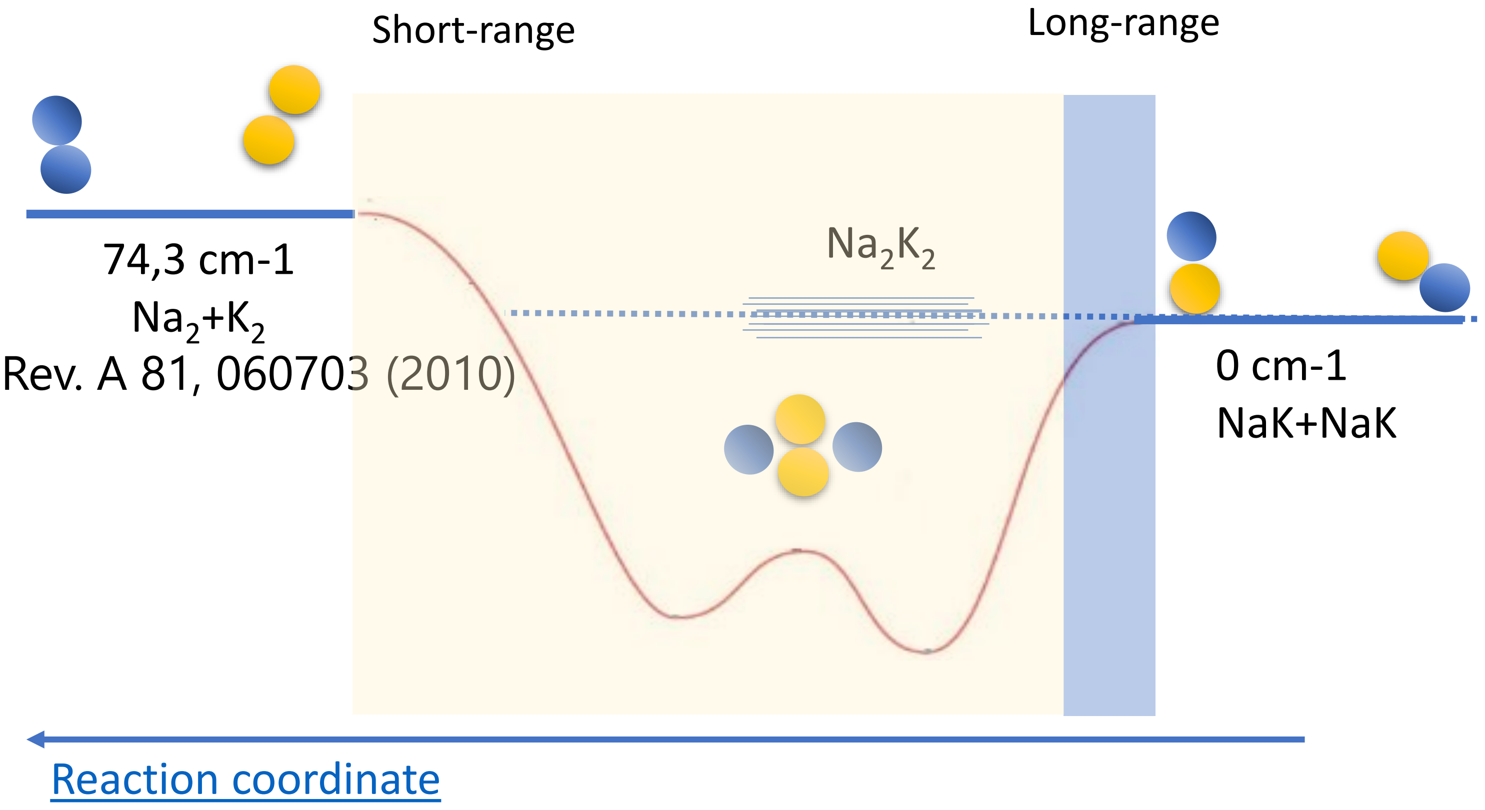
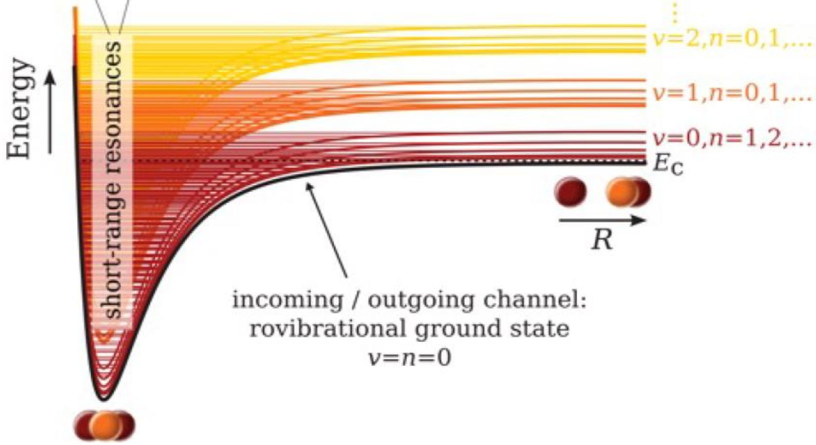


Non-chemically reactive molecules - simplified



Phys. Rev. A 81, 060703 (2010)

Mayle et al., Phys. Rev. A **87**, 012709

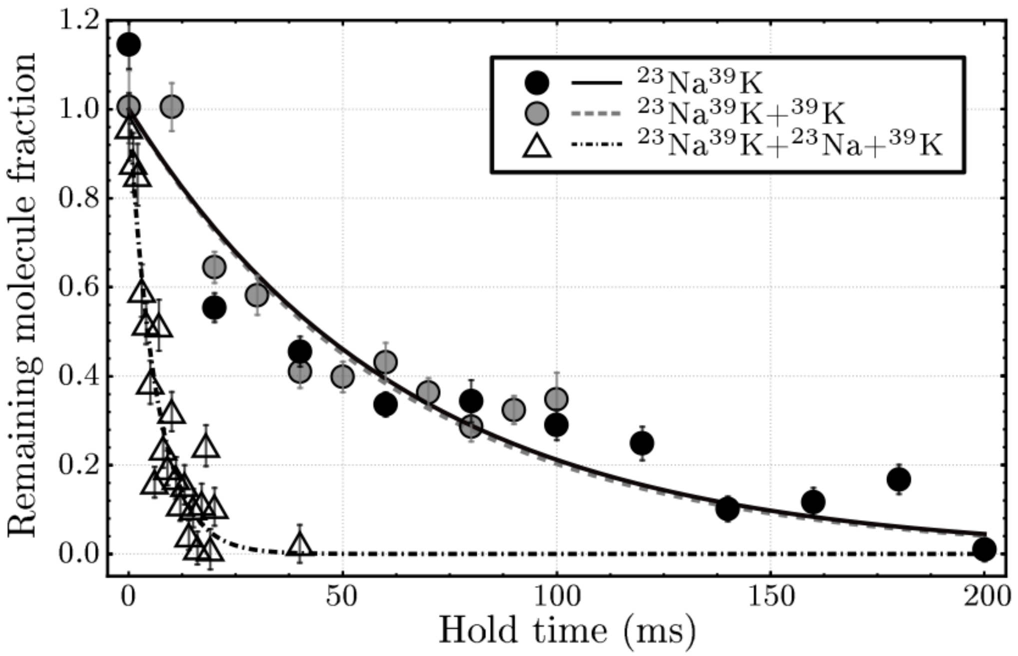


- Large density of states→ergodic exploration with sticking time
- Long-lived tetramer complexes
- RRKM theory: $\tau = \frac{2\pi\hbar\rho}{N}$

	^{23}Na	^{39}K	^{87}Rb	^{133}Cs
^{7}Li	0,25µs	0,67µs	1,17µs	3,3µs
^{23}Na		6µs	12,9µs	40µs
^{39}K			23µs	72µs
^{87}Rb				253µs

Adapted from Phys. Rev. A 100, 032708 (2019)

Molecule-molecule collisions of NaK molecules



Two-body decay

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

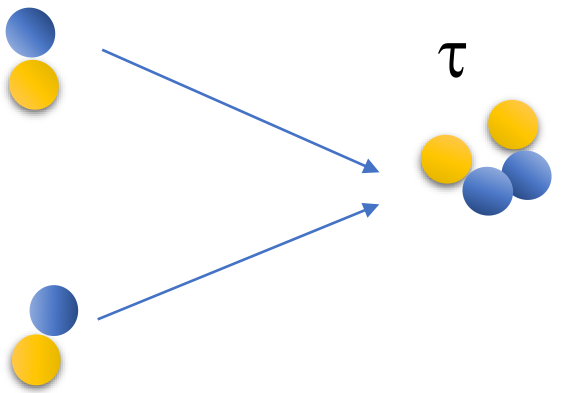
β from fit of two-body decay including the effect from antievaporation

$\beta_{exp} = 4.5(1.2) \times 10^{-10}\ \mathrm{cm}^3\ \mathrm{s}^{-1}$

→ Close to the universal limit ($\sim 7 \times 10^{-10}\ \mathrm{cm}^3\ \mathrm{s}^{-1}$)

Similar observations
Takekoshi et al., Phys. Rev. Lett., **113**, 205301 (2014);
Park et al., Phys. Rev. Lett., **114**, 205302 (2015);
Guo et al., Phys. Rev. Lett., **116**, 205303 (2016);
Gregory et al., Nat Commun, **10**, 3104 (2019);

Measure complex lifetime



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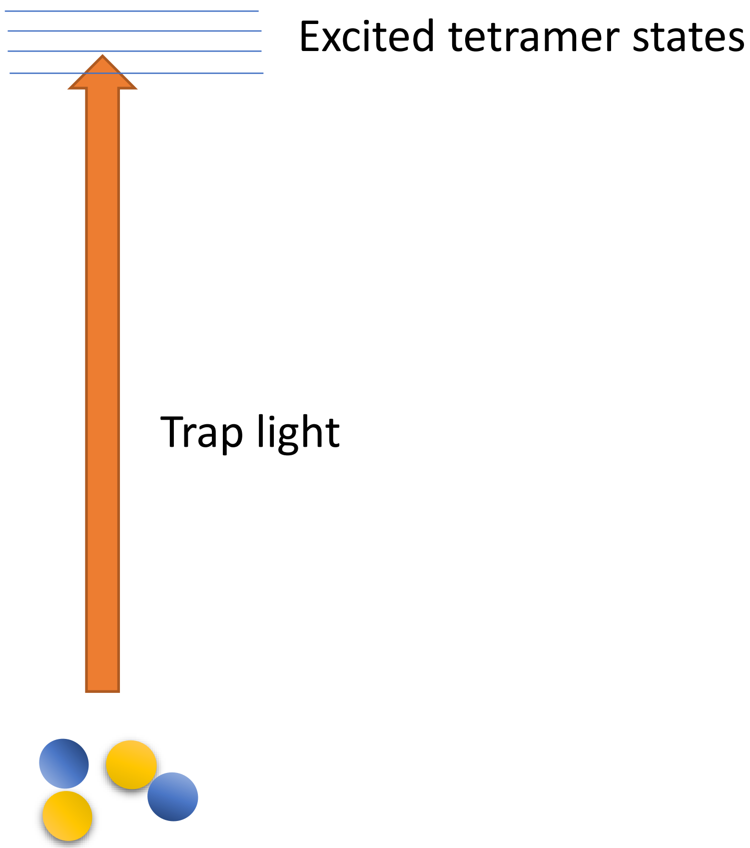
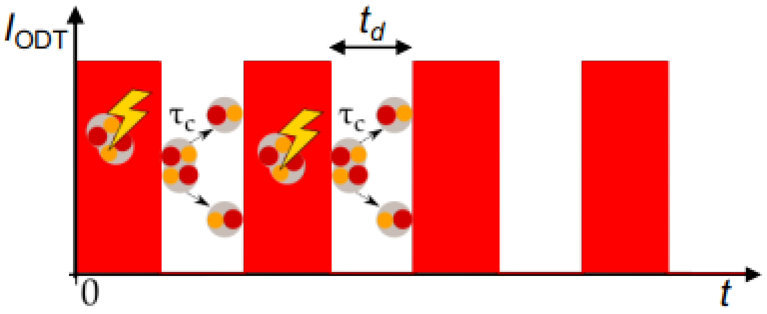


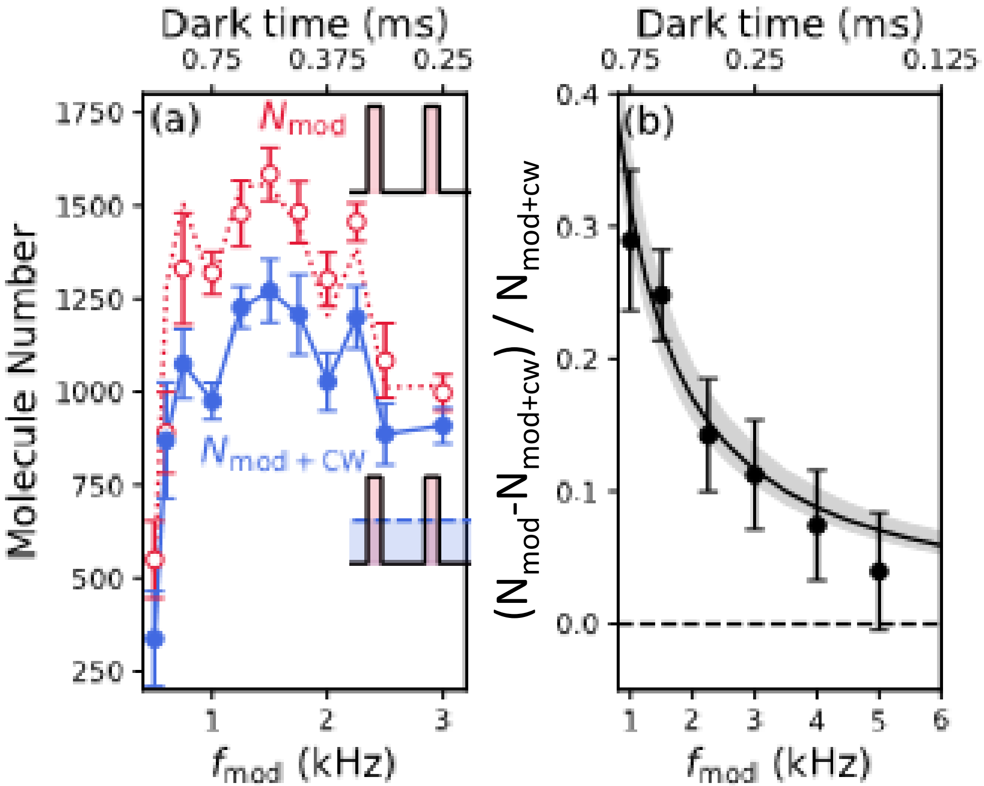
A. Christianen et al.
Phys. Rev. Lett. 123, 123402 (2019)

Collisions of nonreactive molecules -RbCs

First evidence for photoinduced loss by Durham group with RbCs

Chopped trap





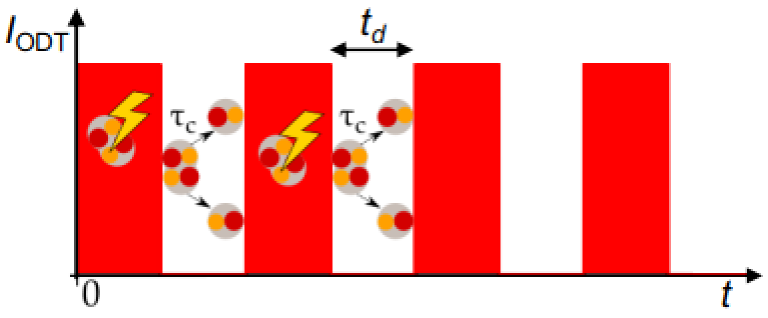
Gregory et al., Phys. Rev. Lett. 124, 163402 (2020)

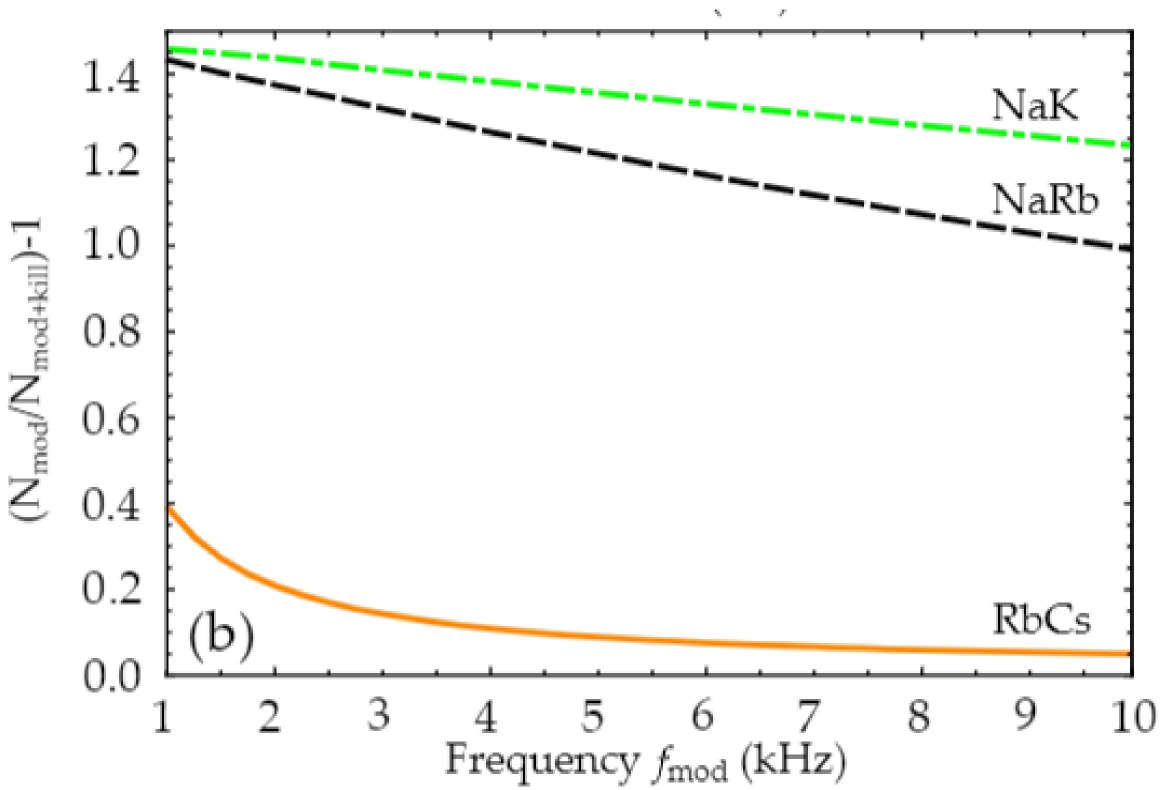
- Measured complex lifetime ~500µs within a factor of two of predicted lifetime!

Collisions of nonreactive molecules - NaK

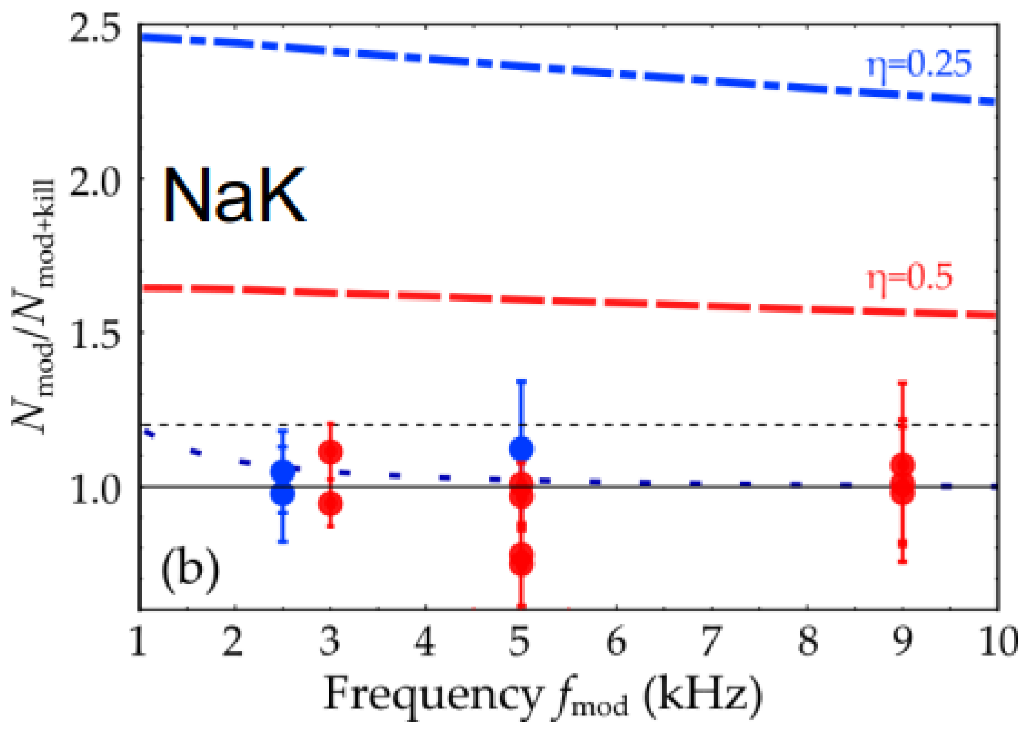
→ Short expected complex lifetime of ~6µs; $t_d \gg \tau$
→ Molecule number should double in chopped trap!

Chopped trap

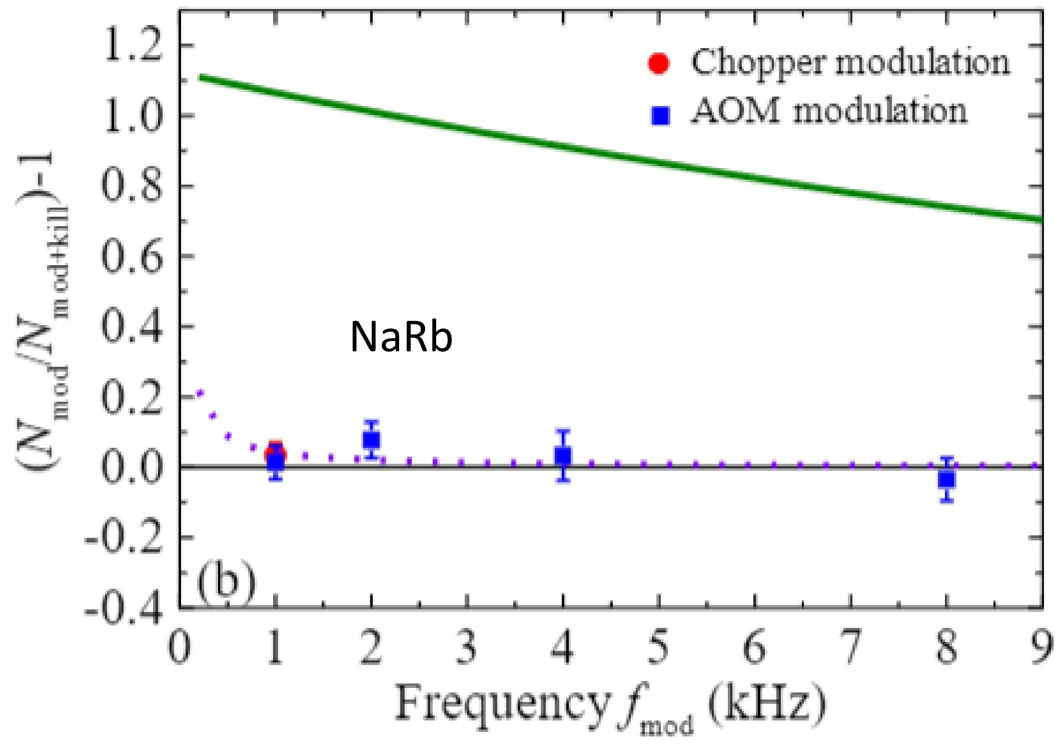




Collisions of ultracold NaK molecules



τ>1 ms



τ>4 ms

P. Gersema et al. , Phys. Rev. Lett. 127, 163401 (2021)

Related results

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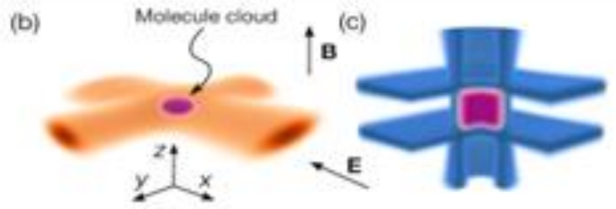
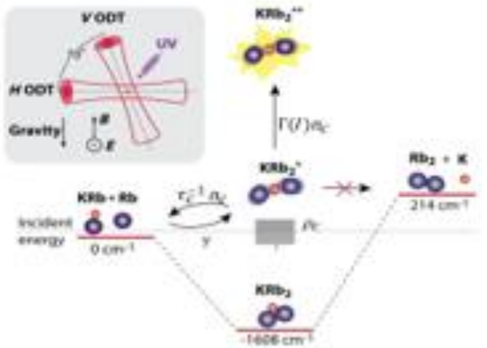
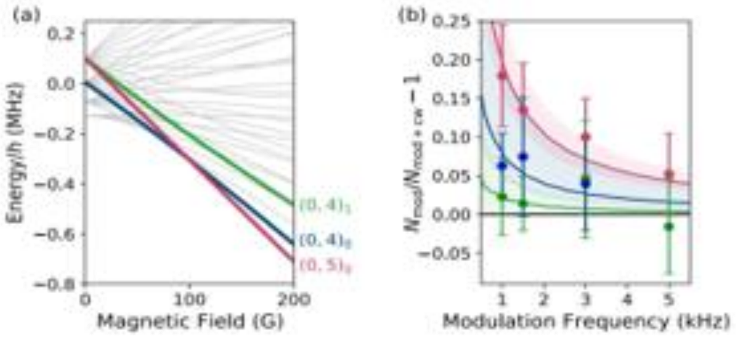
- fermionic NaK (Munich): no enhancement at low trap intensities
  Bause et al., Phys. Rev. Res., **3**, 033013 (2021)
- trimer complex lifetime of $^{40}K^{87}Rb_2^*$ much longer than expected
  Nichols et al., Phys. Rev. X 12, 011049 (2022)
- RbCs: $\tau^c \approx 0.5$ ms $\approx \tau^{cth}$ for spin-stretched state but disagreement for other states
  Gregory et al., Phys. Rev. Lett. **124**, 163402 (2020)
  Gregory et al., New J. Phys. 23, 125004 (2021)
- Anomalous lifetimes of ultracold complexes with few open channels
  Croft et al., Phys. Rev. A 107, 023304 (2023)
- Severely underestimated DOS? Nuclear spin changes?
  Jachymski et al. Phys. Rev. A106, L041301 (2022)
- Roaming pathways in real-time collisional simulations
  Kłos et al., Sci Rep 11, 10598 (2021)







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Interpretation

Subject of ongoing debate:

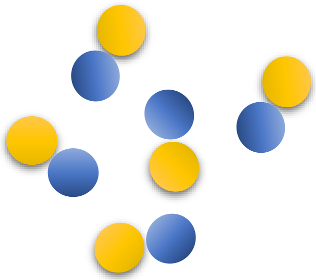
- Parameters relevant for lifetime estimate using RRKM theory (DOS, outgoing channels, ...)?
- Relevant couplings?
- Applicability of RRKM theory?

Christianen et al., Phys. Rev. A **100**, 032708 (2019)

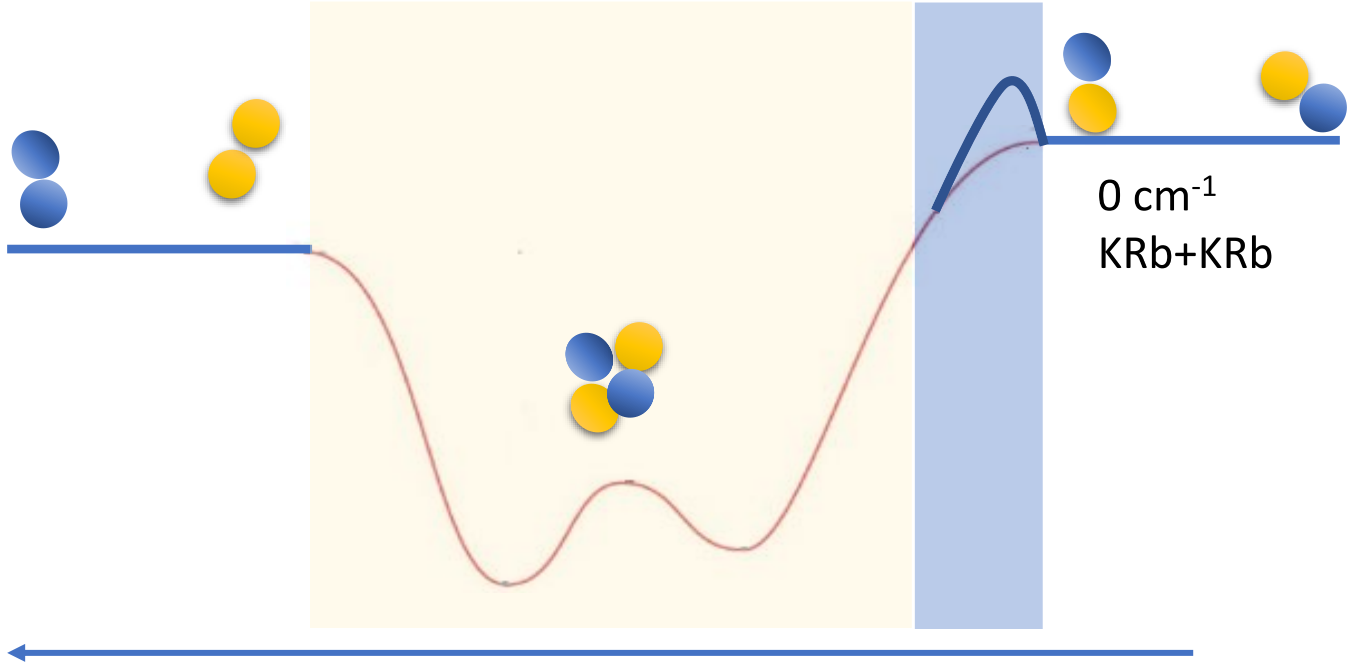
Jachymski et al. Phys. Rev. A106, L041301 (2022)

Croft et al., Phys. Rev. A 107, 023304 (2023)

Controlling collisions



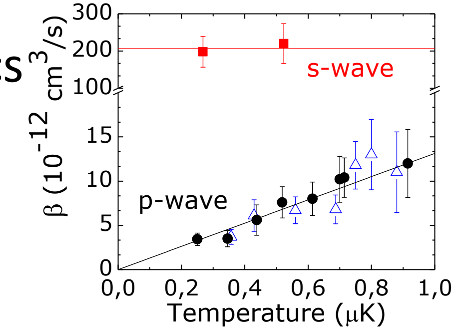
Manipulating the long-range



Reaction coordinate

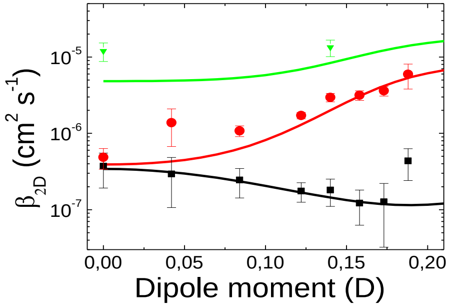
- Quantum statistics

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

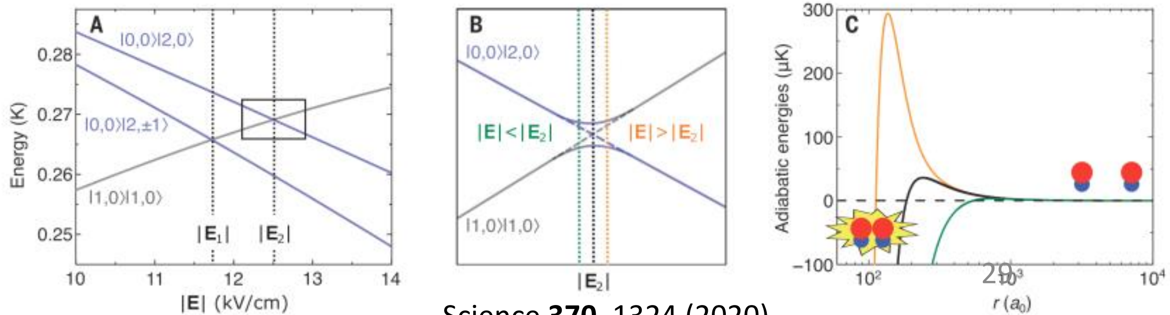


- Dipolar interactions + confinement in restricted geometries

  K. K. Ni, S. Ospelkaus et al.,
  Nature 464(7293) (2010)
  Nature Physics 7, 502 (2011)



- Avoided crossings in static electric fields



Science **370**, 1324 (2020)

Controlling collisions – blue shielding

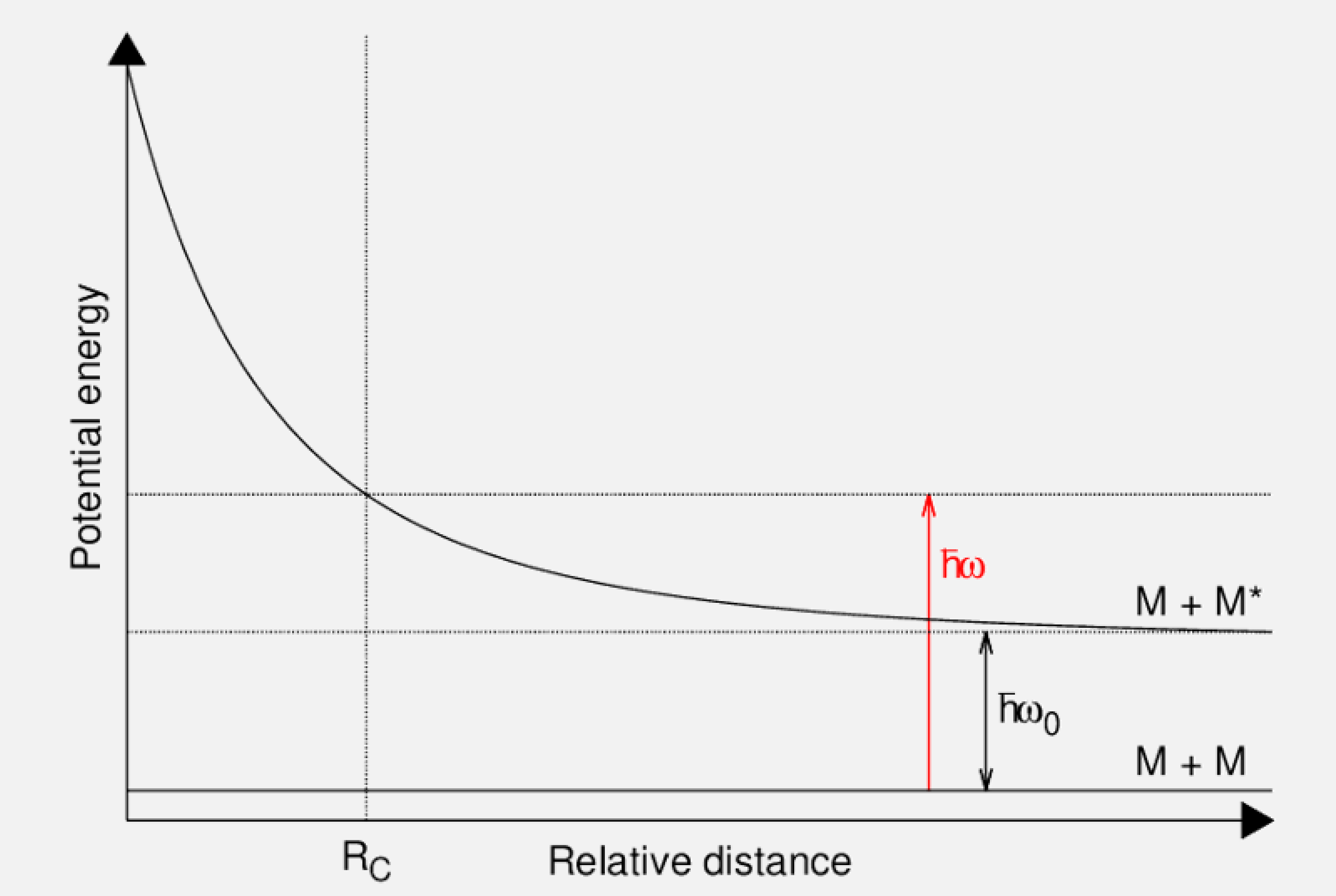


Figure courtesy:
Maxence Lepers

Controlling collisions – blue shielding

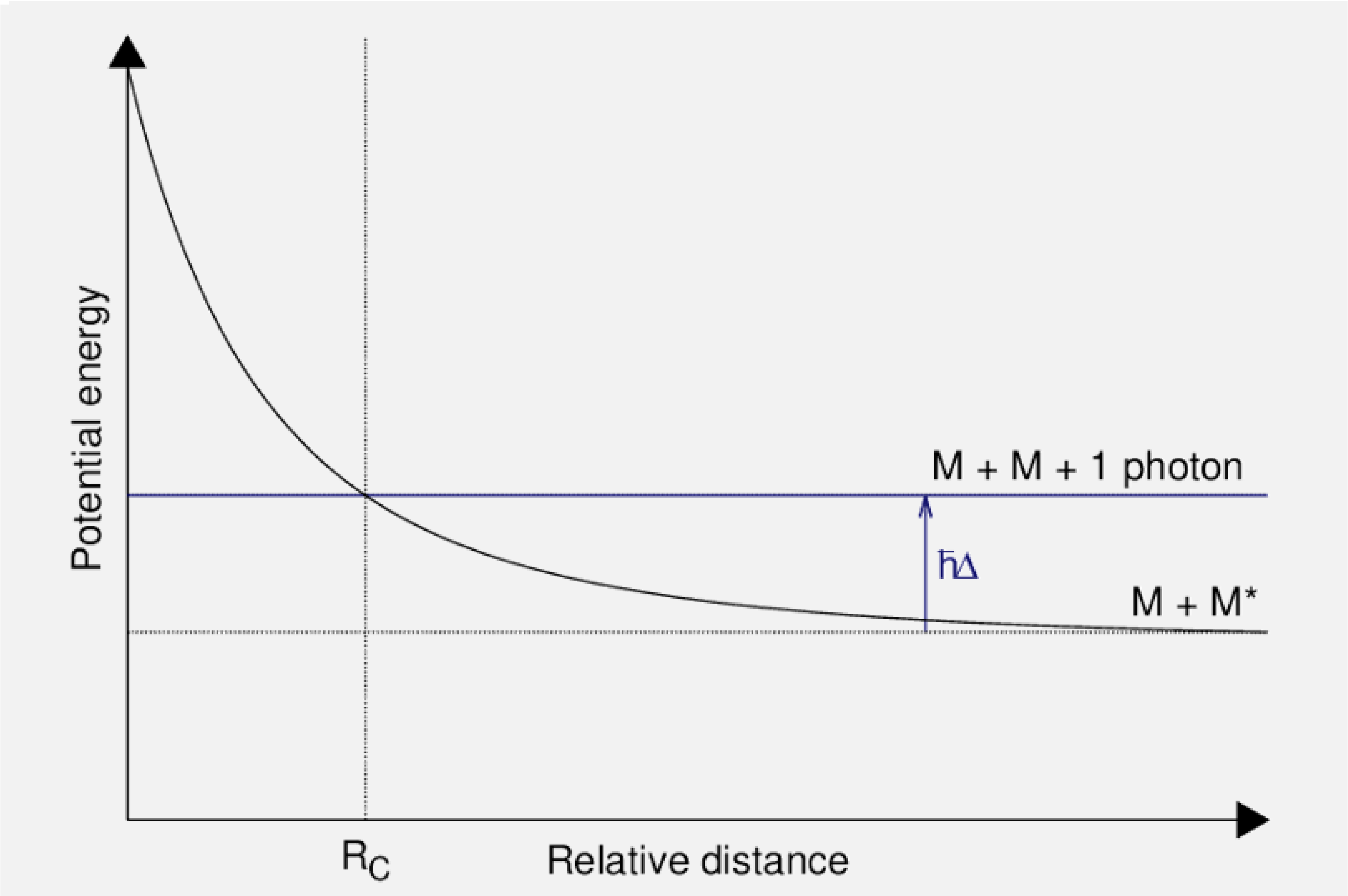


Figure courtesy:
Maxence Lepers

Controlling collisions – blue shielding

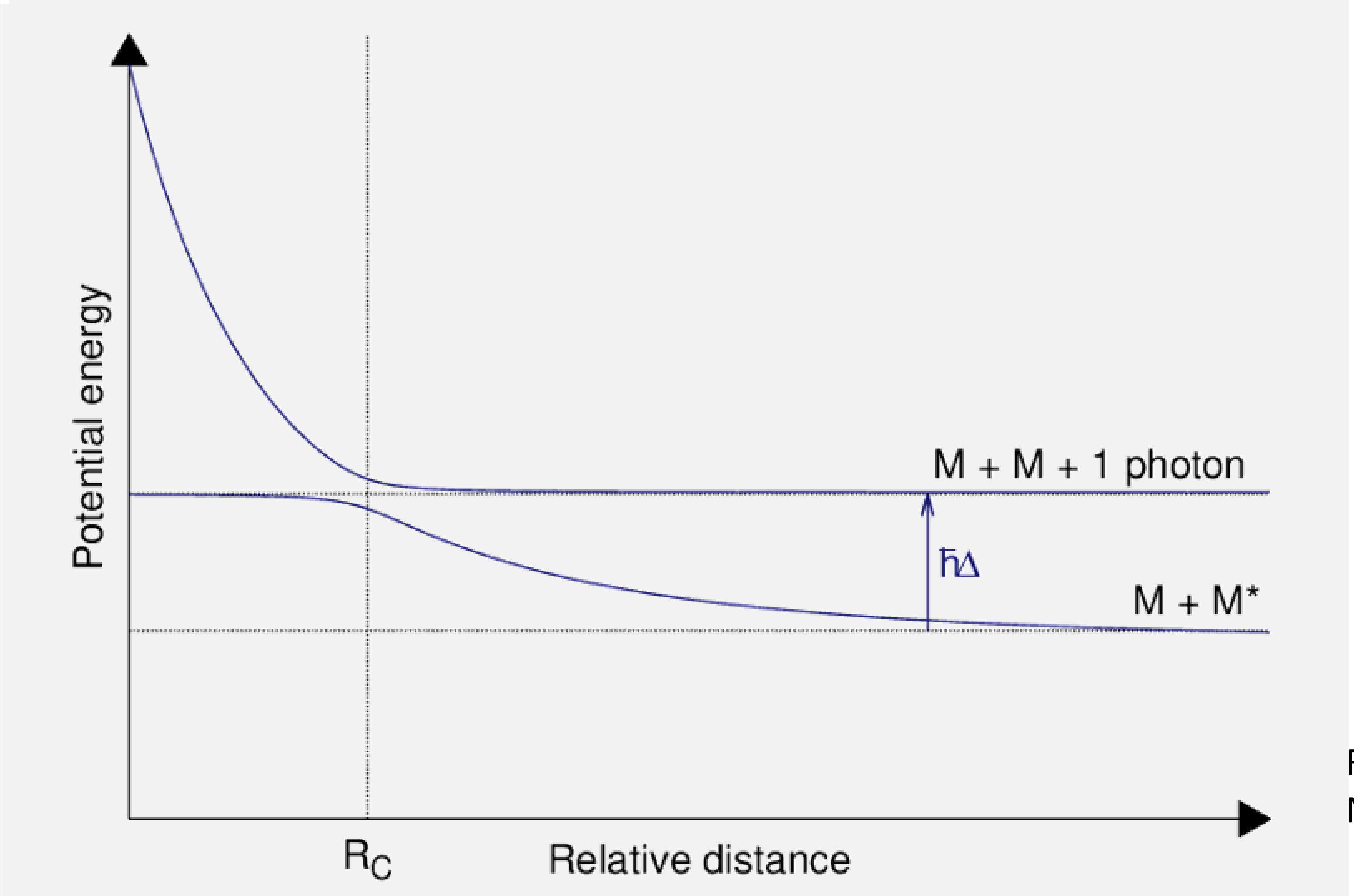


Figure courtesy:
Maxence Lepers

MW blue shielding of collisions

Molecules: Blue shielding of collisions using microwave coupling within the rotational structure
Experiment with CaF molecules

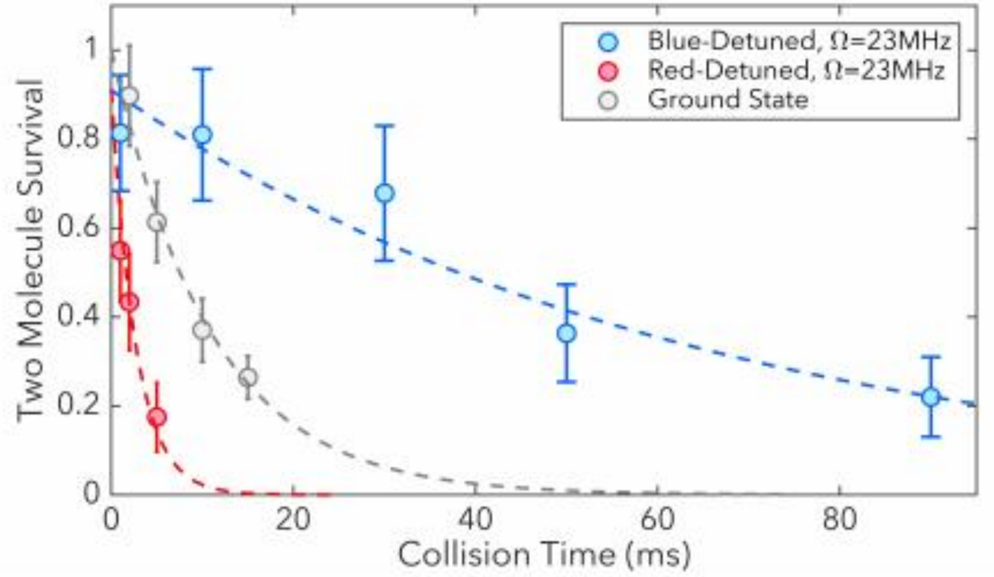


Figure 2: **Microwave shielding of CaF collisions** The grey trace (10.8 ms) shows the bare two body loss of unshielded ground state collisions. The blue trace (64 ms) shows the shielded loss rate at a Rabi frequency of 23 MHz, and magnetic field of 27 G while blue detuned. The red trace (2.7 ms) shows the loss rate while red detuned with a Rabi frequency of 20 MHz, and magnetic field of 27 G.

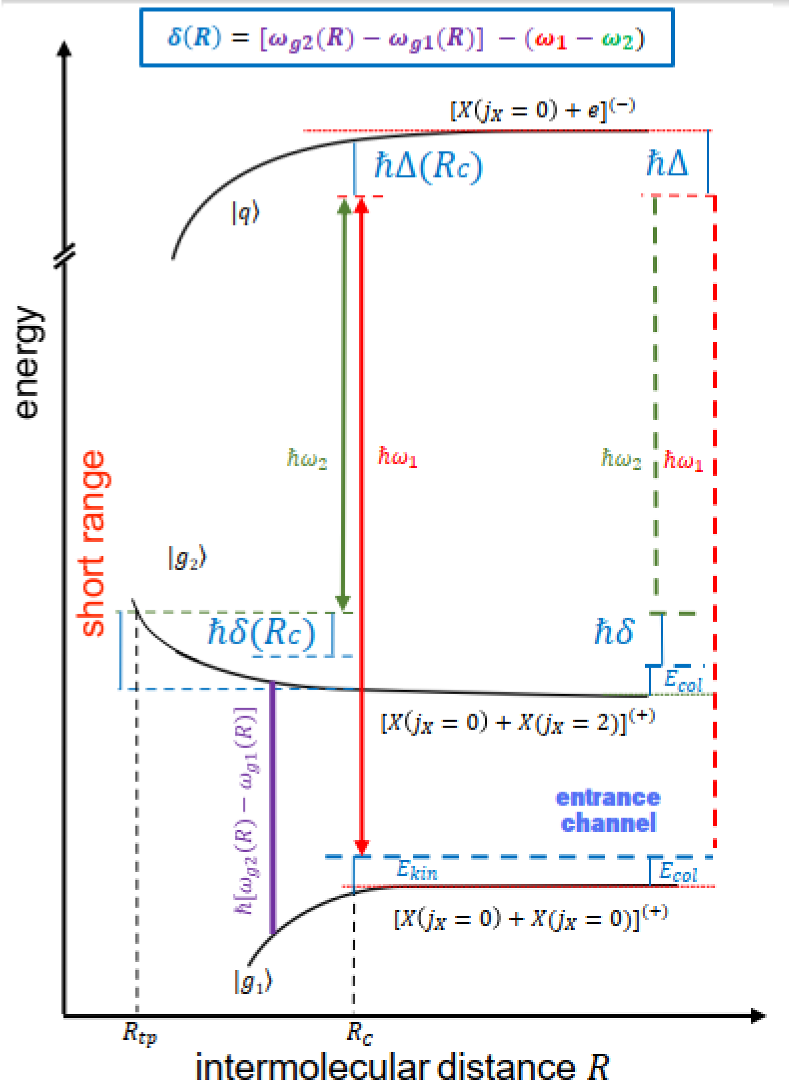
Anderegg et al. , Science 373, 779 (2022)

MPQ with fermionic NaK:
Schindewolf et al., Nature **607**, 677 (2022)

HongKong with bosonic NaRb
Lin et al., arXiv:2304.08312

Cornell university with bosonic NaCs
Bigali et al., arxiv: 2303.16845

Optical shielding of collisions



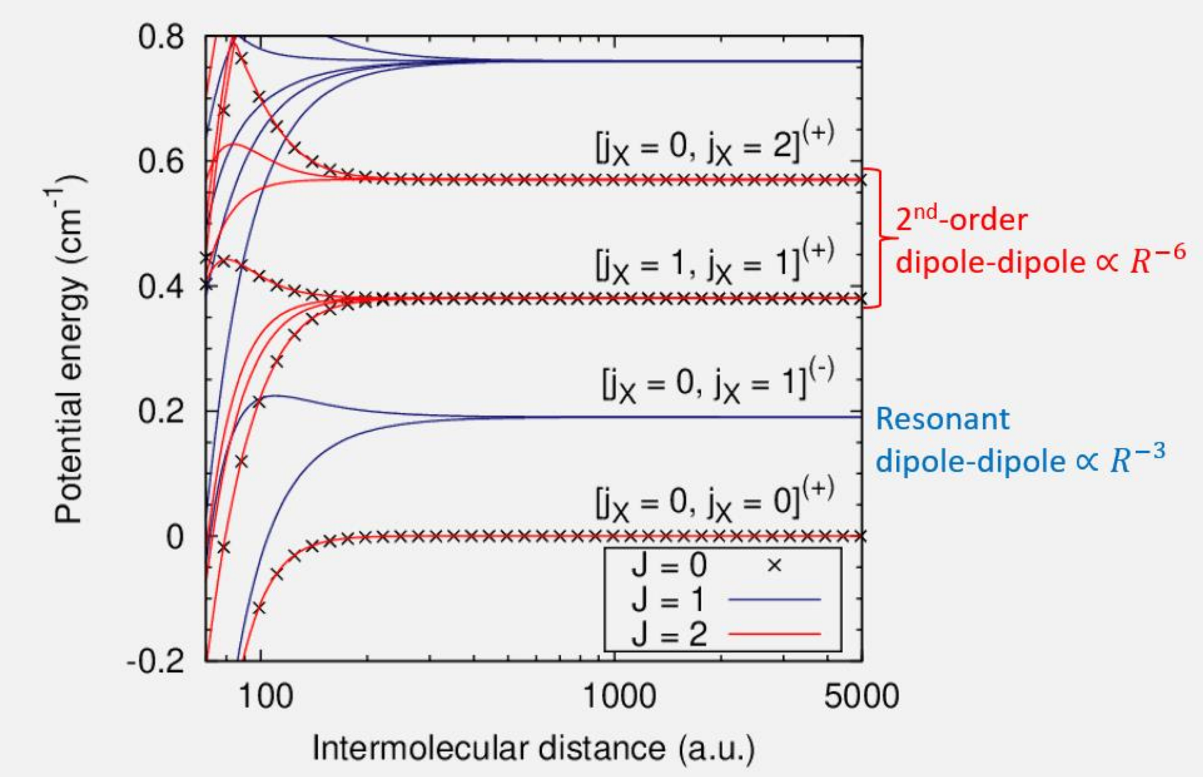
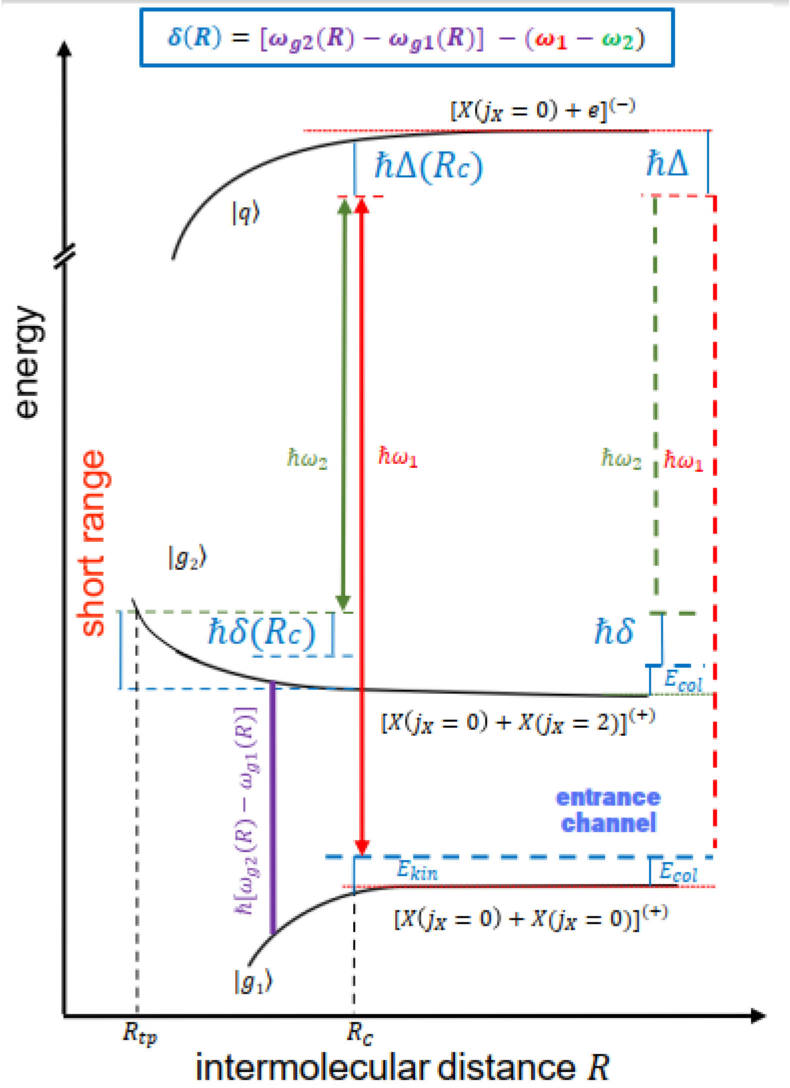


Figure courtesy: Maxence Lepers

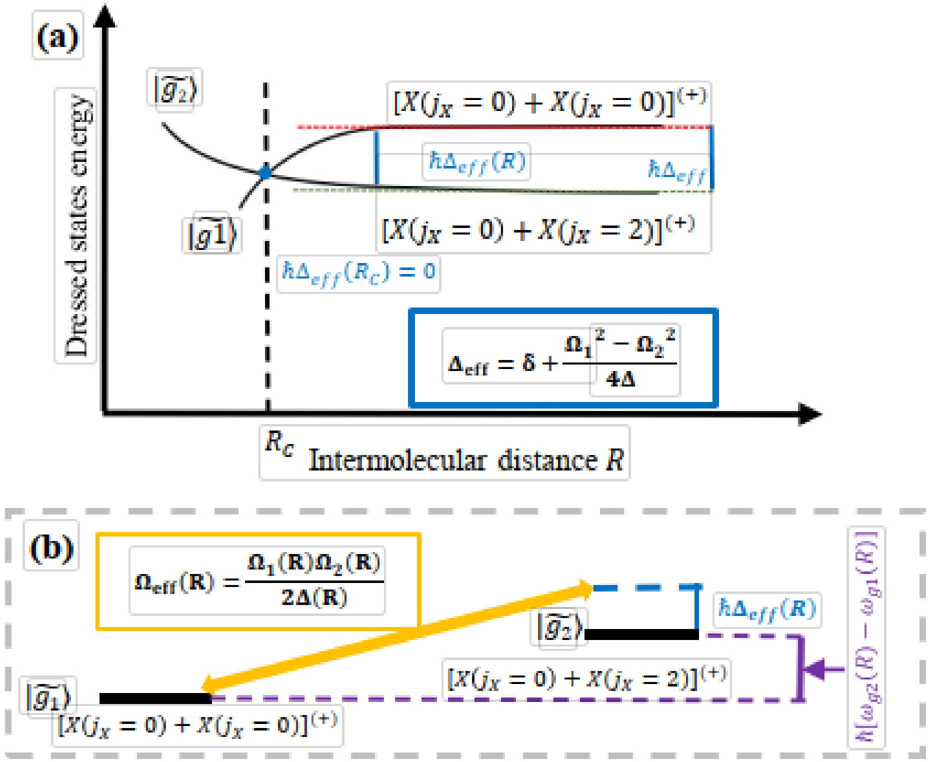
Hannover + Orsay proposal:
arXiv:2211.08950, accepted in PRR

Optical shielding of collisions



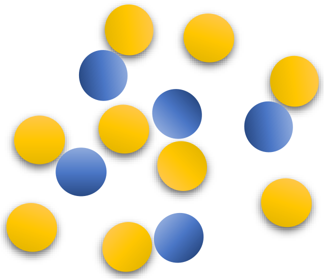
C. Karam et al. arXiv:2211.08950 (2022)

Effective 2-level system



- adiabatic elimination ⇒ effective 2-level coupling
- difference: need to couple states with same parity

Atom-molecule collisions



Atom-molecule collisions for the NaK molecule

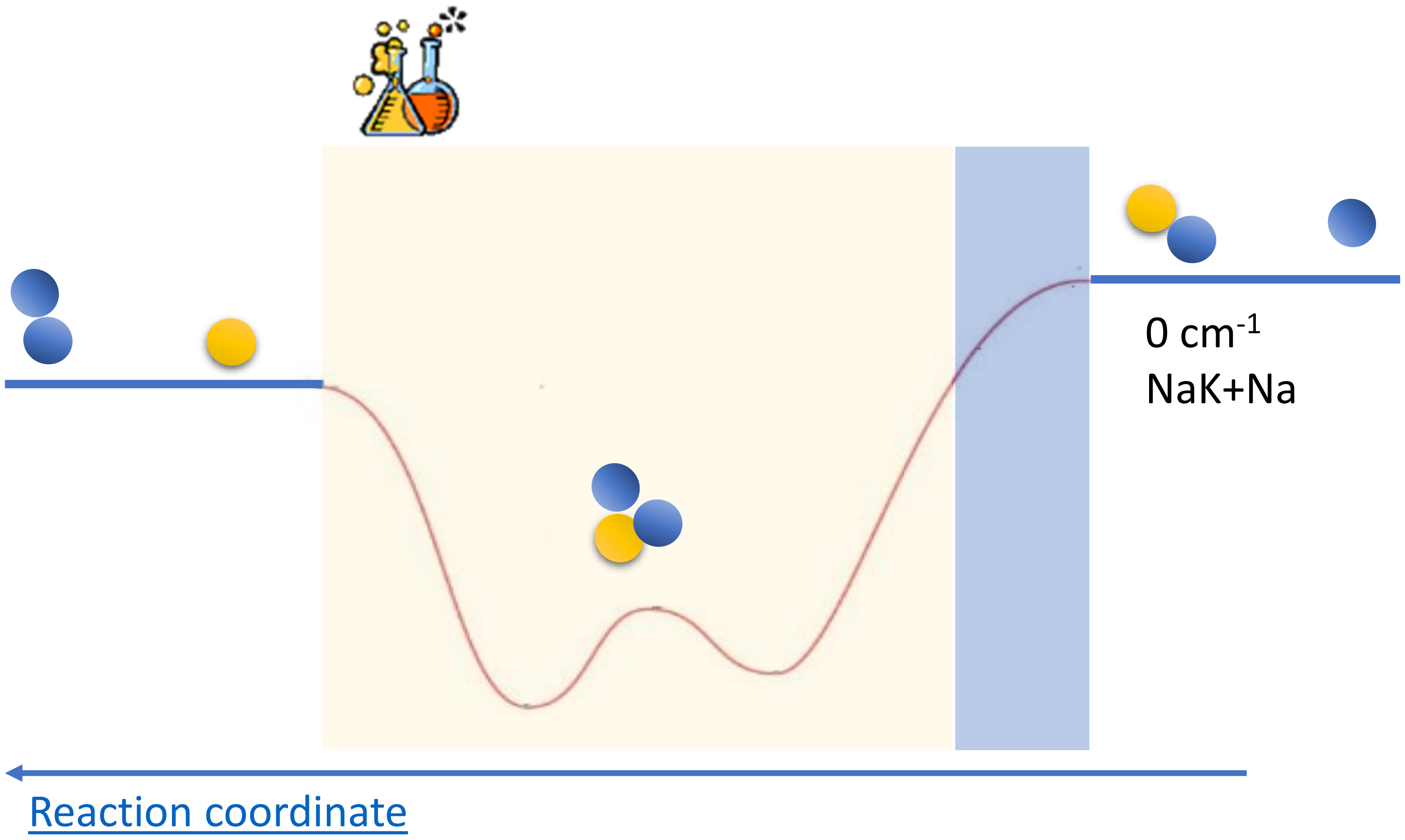
Leibniz Universität Hannover

NaK+Na

Exoergic chemical reaction

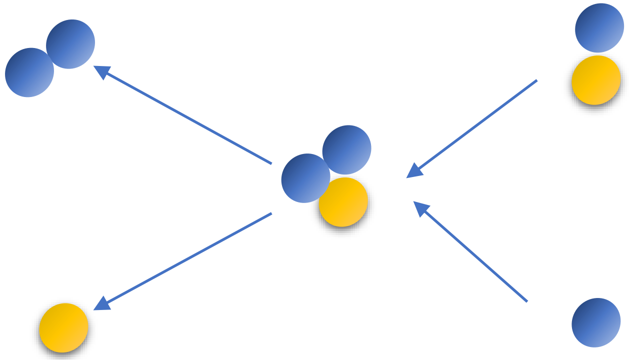
NaK+Na→ Na_2+K+

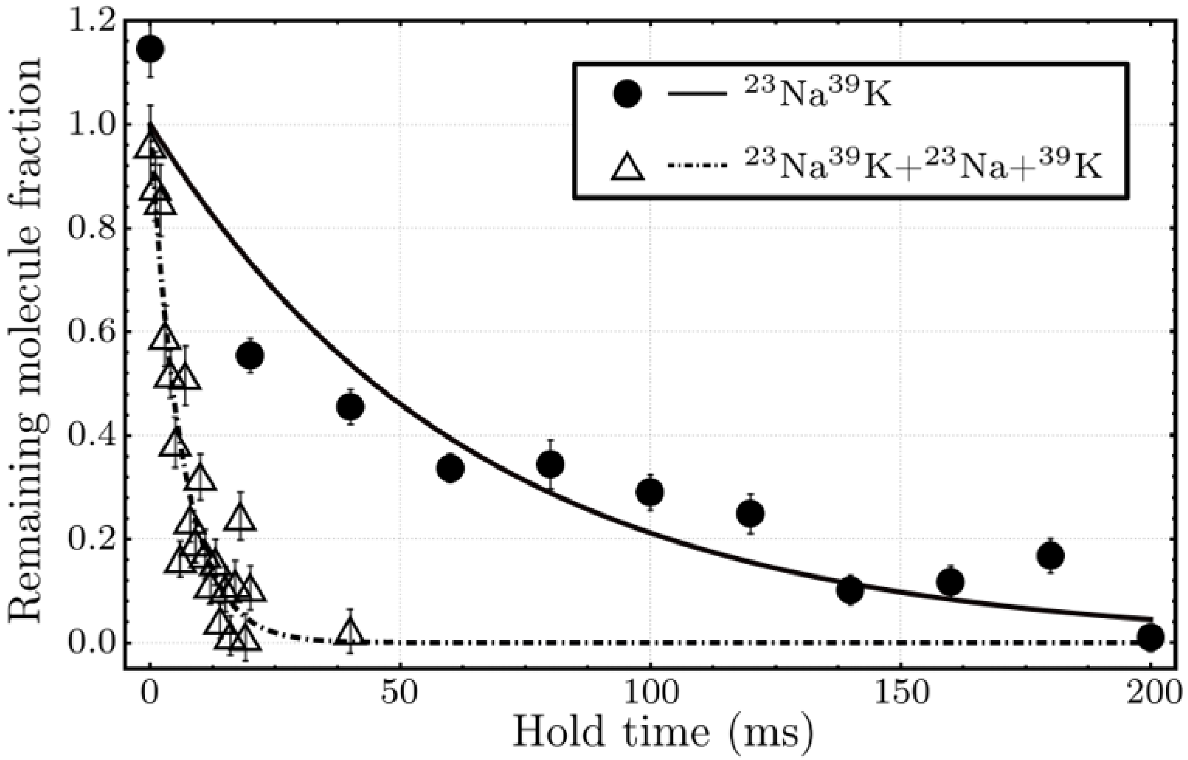
NaK+K



Atom-molecule collisions for the NaK molecule

NaK+Na





Exoergic chemical reaction

NaK+Na→ Na_2+K+

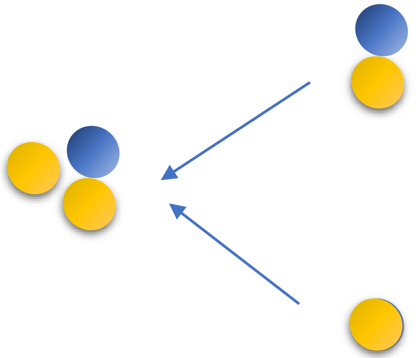
NaK+Na: β_{exp}= 1.25(14) × 10^{-10} cm^3 s^{-1}

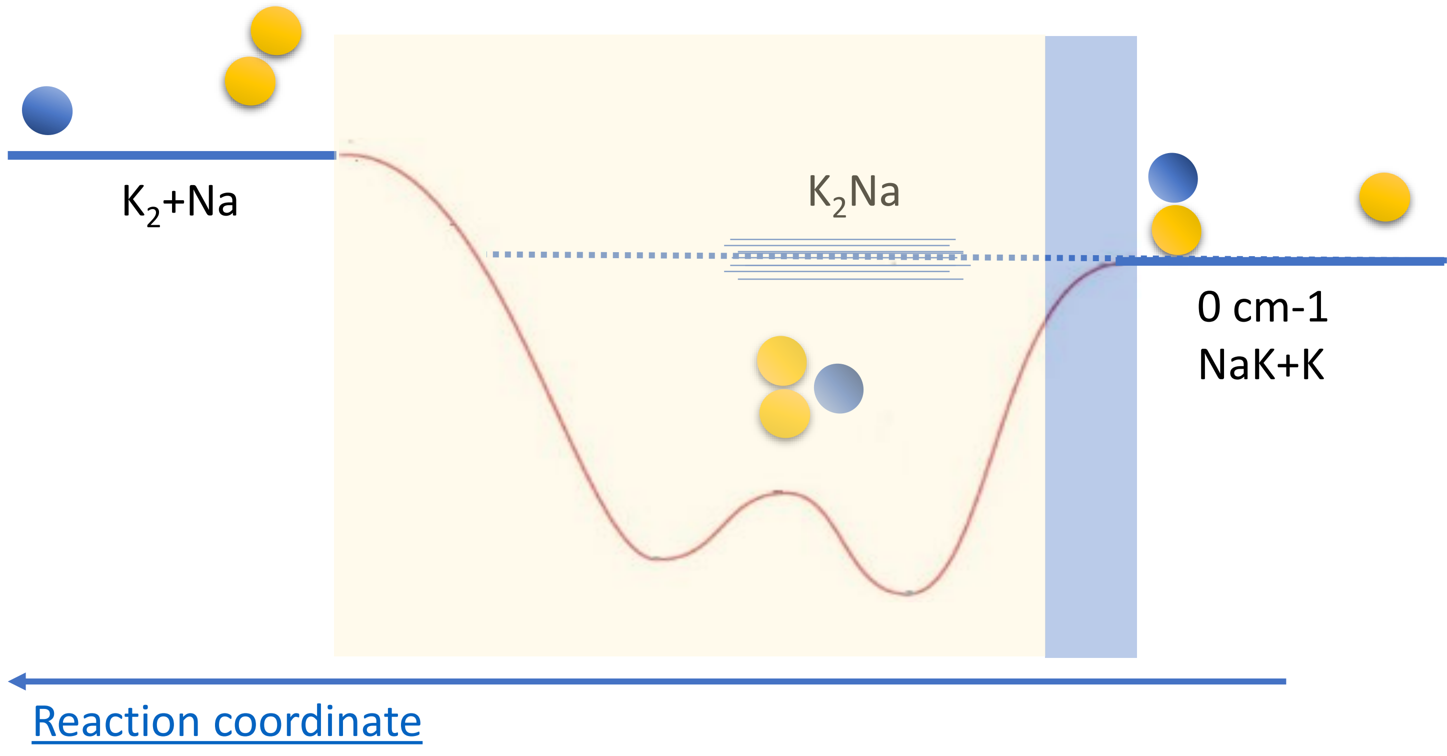
→ consistent the universal limit

(~1.3 × 10^{-10} cm^3 s^{-1})

Atom-molecule collisions for the NaK molecule

NaK+K

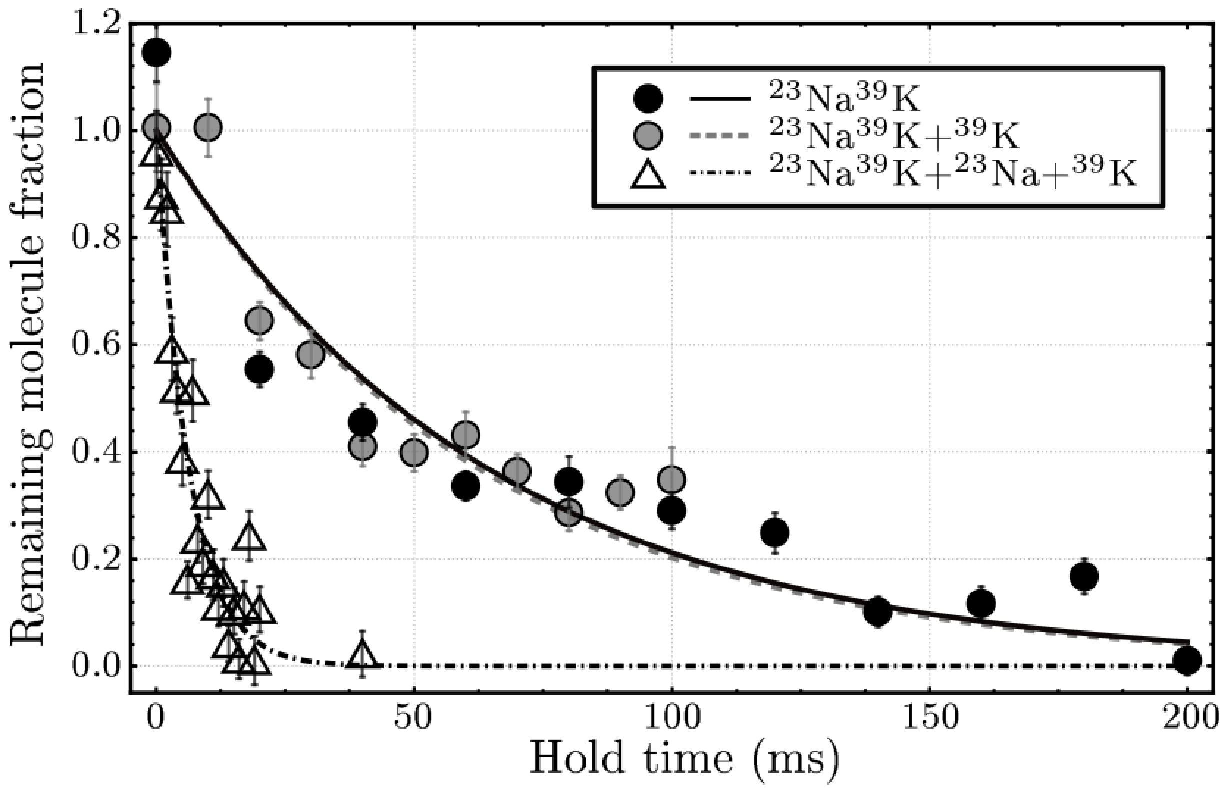




Endoergic chemical reaction
NaK+K→ K_2+Na
forbidden at ultracold temperatures

ρ~2x10^{-5}/μK (arxiv: 200805439)

Collisions of ultracold NaK molecules

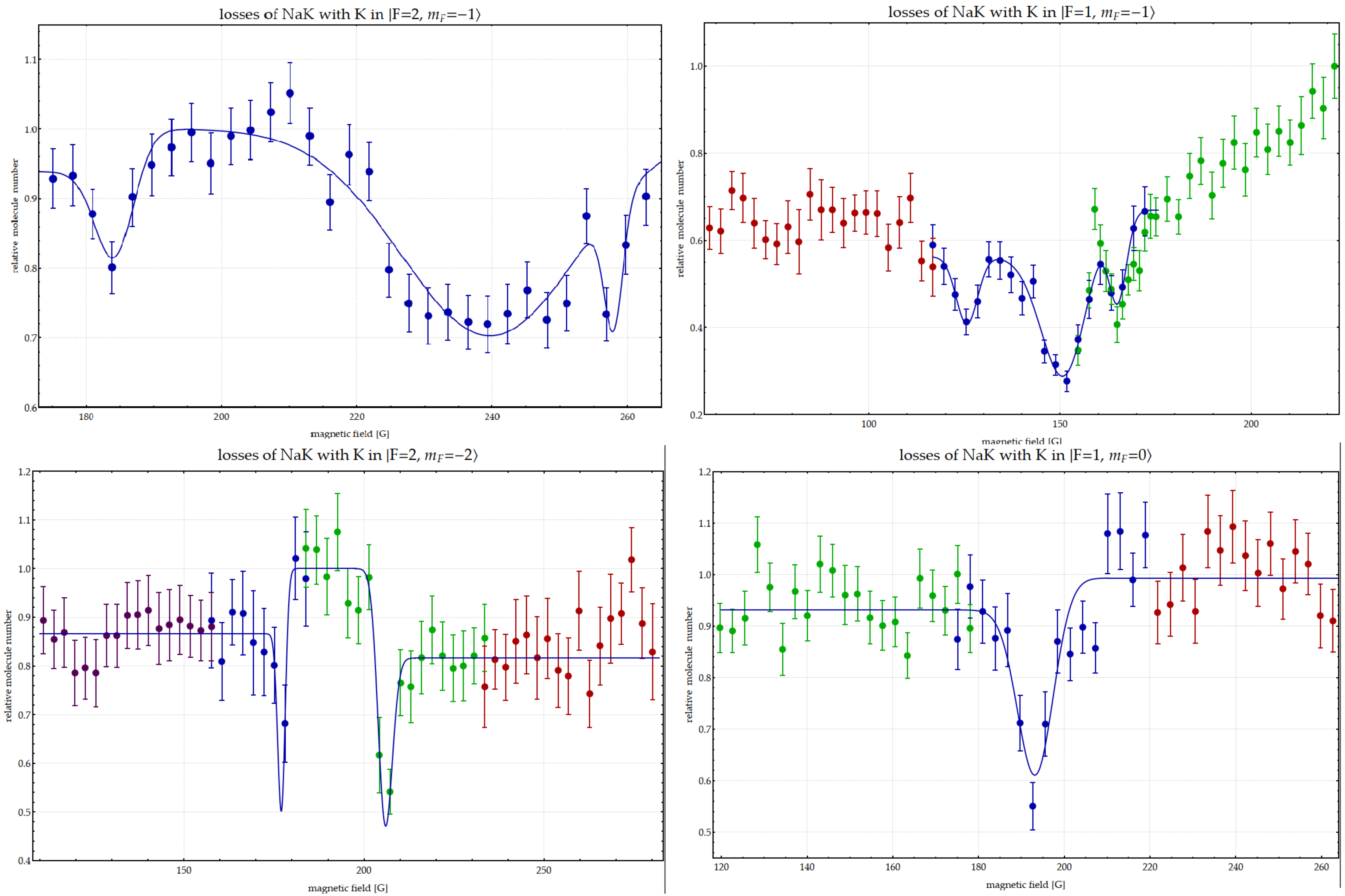


- Surprising: NaK + K in |1,-1> state
- Very low loss coefficient
- $\beta < 10^{-14} cm^3/s$
- Much lower than universal loss of $\sim 10^{-10} cm^3/s$

Atom-molecule Feshbach resonances

Leibniz Universität Hannover

Preliminary!!



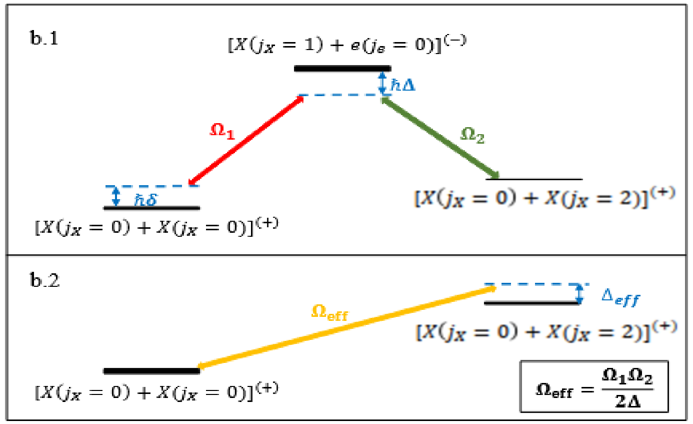
Theory of atom-molecule Feshbach resonances: M. Frye, J. Hutson arxiv:221208030

- J.W. Pan: Fermionic NaK
- W. Ketterle: Triplet LiK

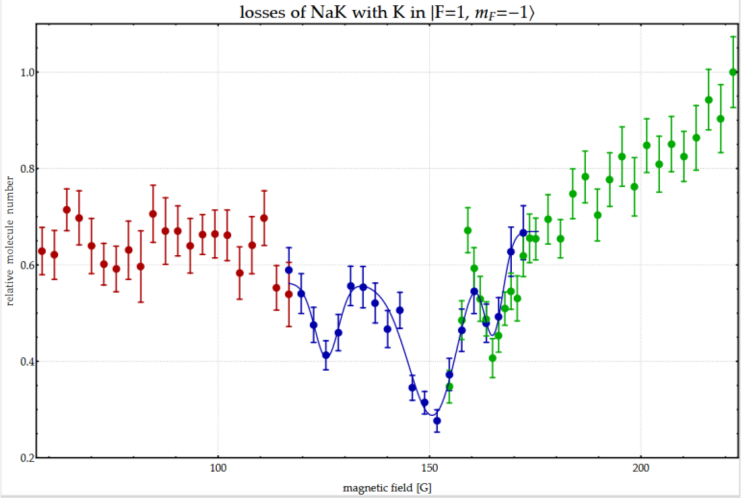
- Tuning of elastic atom-molecule interactions?

Outlook

Collisional control: Two-photon shielding
Long-lived molecular BEC?



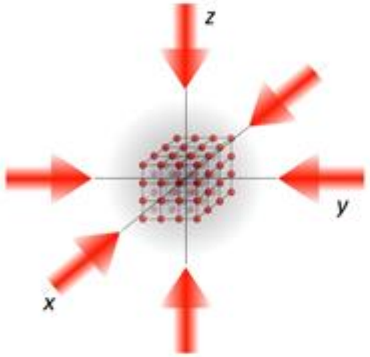
Tunable strongly interacting
atom-molecule mixtures?

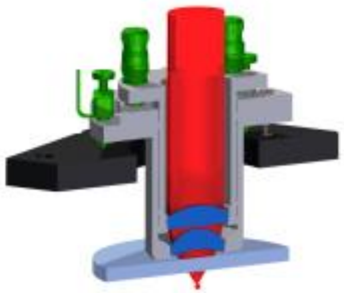


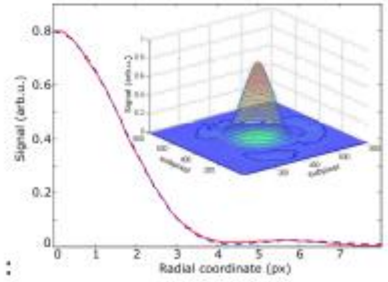
Trimer and tetramer states

Understanding collisions?
Ultracold polyatomic molecules?

Dipolar many-body physics in lattices







Thank you!

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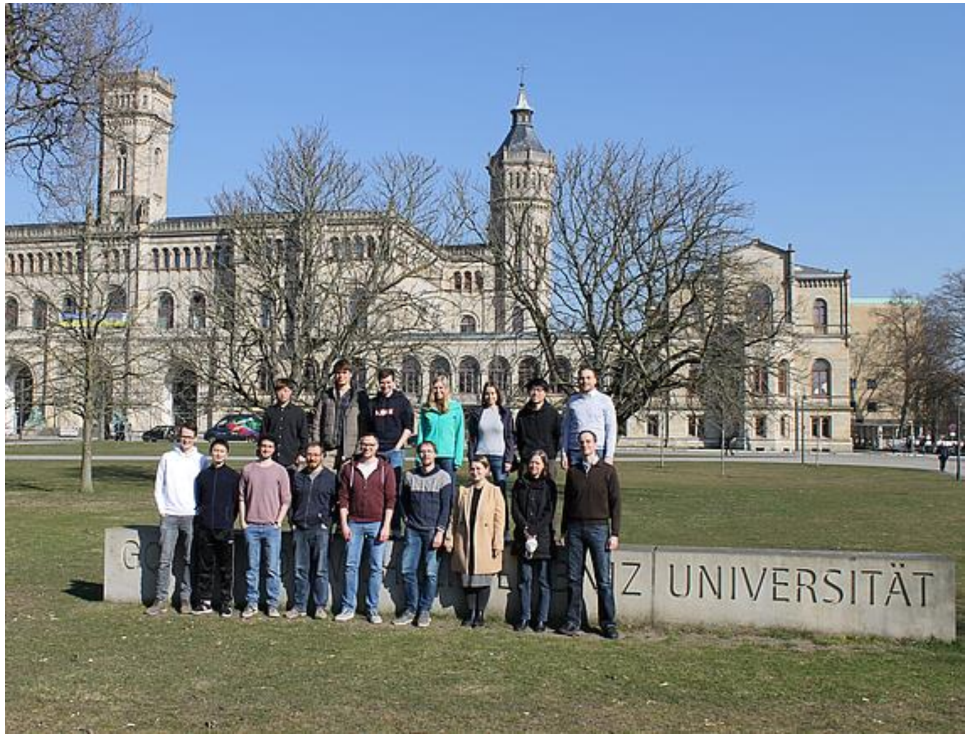
E. Tiemann

The experimental team at LUH



Funding

DFG Deutsche Forschungsgemeinschaft
German Research Foundation

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