High-Resolution Quantum Control of Chemical Reactions

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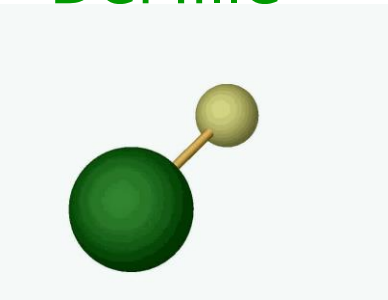
DeMille



Group

MURI Overview

--Team composition & organization

--Scientific background

--Themes, goals, and recent results

MURI Team & Budget

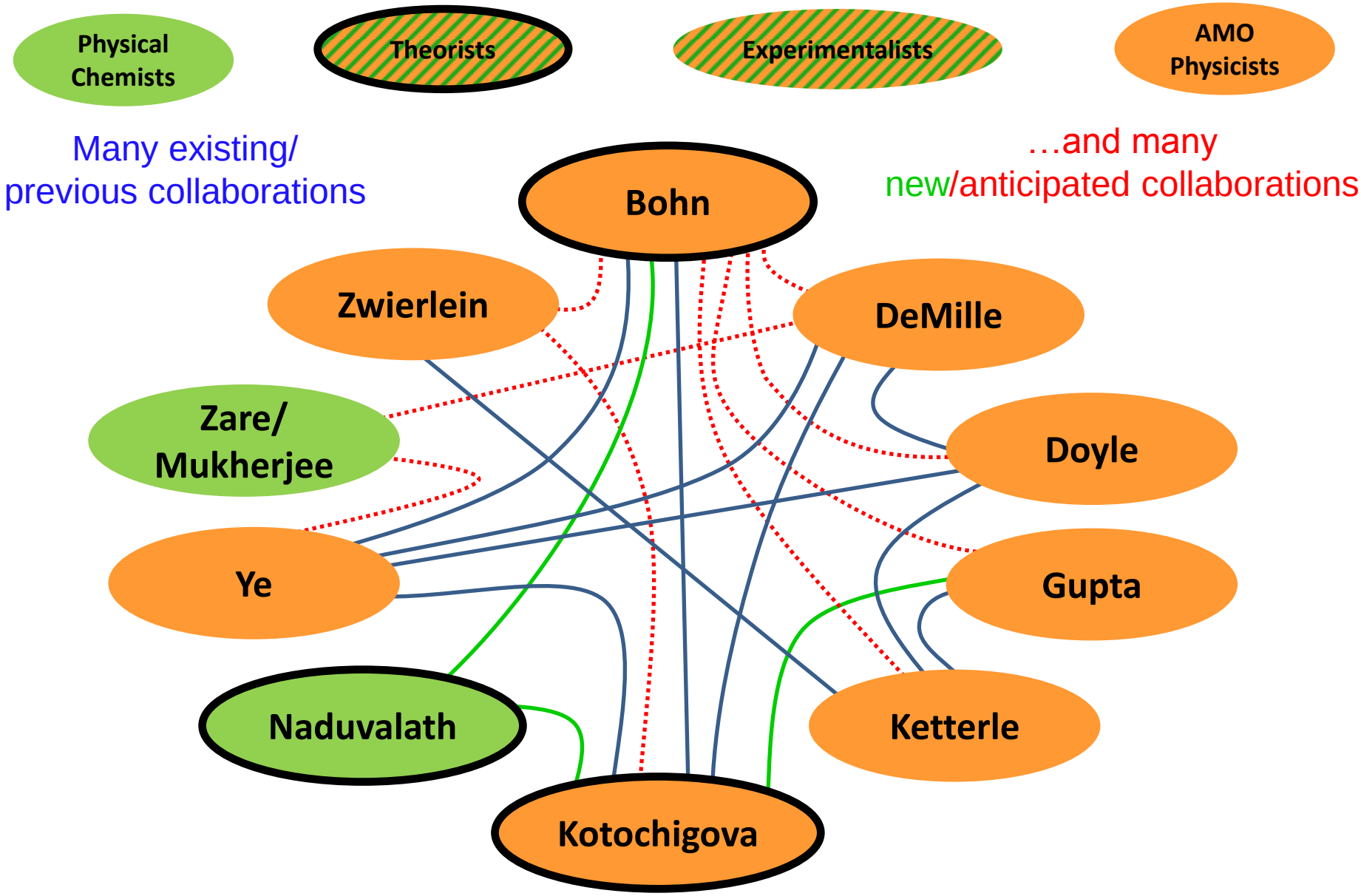
- Prof. John Bohn (U. Colorado/JILA)
- Prof. David DeMille (Yale)
- Prof. John Doyle (Harvard)
- Prof. Subhadeep Gupta (U. Washington)
- Prof. Svetlana Kotochigova (Temple)
- Prof. Wolfgang Ketterle*,§ (MIT)
- Prof. Balakrishnan Naduvalath (U. Nevada, Las Vegas)
- Prof. Jun Ye* (U. Colorado/JILA)
- Prof. Dick Zare* & Dr. Nandini Mukherjee (Stanford)
- Prof. Martin Zwierlein (MIT)

*Member, National Academy of Science §Nobel Laureate

PLUS: 19 Ph.D. students & 13 postdocs fully or partially supported

Budget: $1.25M/year

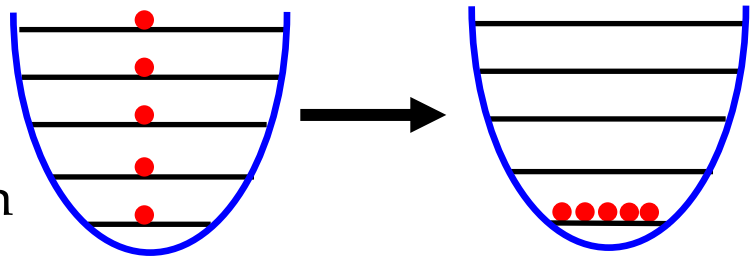
Interdisciplinary, multiply-connected team



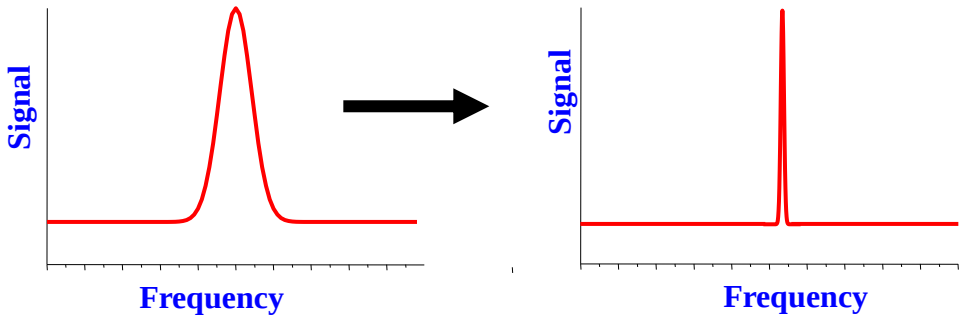
21 refereed publications in <2 years including 4 in Nature & 4 in Phys. Rev. Lett.

Colder = more control, better understanding

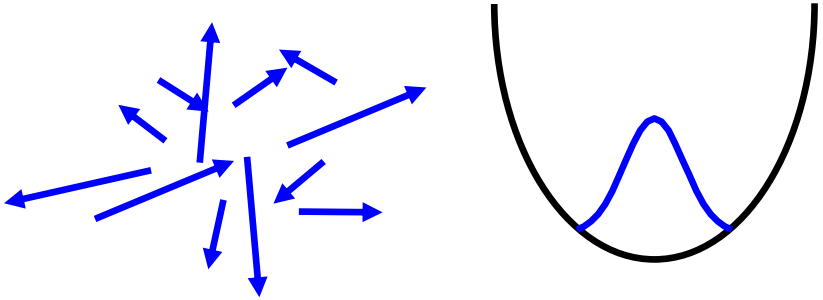
- **Collapse of internal state distribution:**
  --bigger signals
  --initialized for internal state manipulation



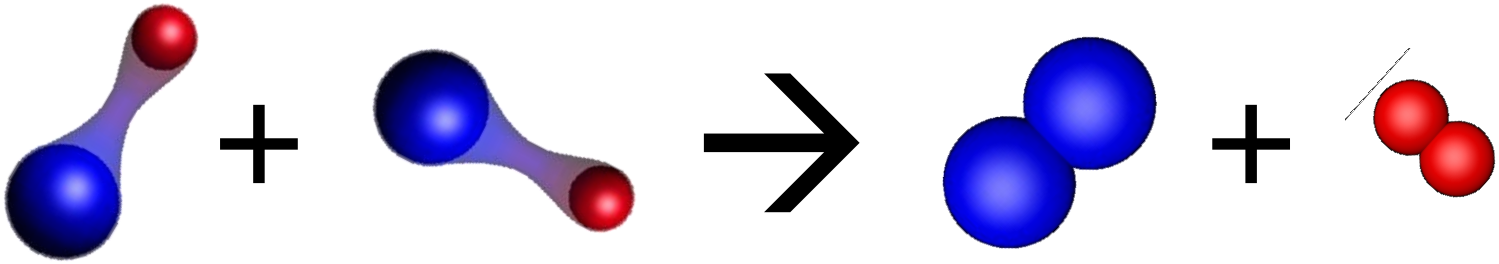
- **Collapse of velocity distribution:**
  --no Doppler
  --long observation times
  ⇒ narrower spectroscopic lines,
  better energy resolution
  --precise control of kinetic energy



- **Trapping:**
  --longer observation (measure small rates)
  --precise control over motional state
  --easier manipulation of internal states
  --enables further cooling
  --ultimately: single quantum internal AND motional state of each particle



An ultimate goal: Controlled quantum chemistry

e.g.: 

Electric Fields

Magnetic Fields

Quantum Statistics

Dimensionality

Electron/nuclear spin states

Rovibronic excitation

Kinetic Energy/ Temperature

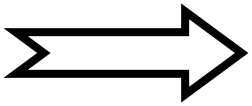
Rotational state orientation/alignment

→ Dramatic, controlled changes in reaction rates/branching ratios

→ Improved understanding of general reaction mechanisms

How do you make cold molecules?

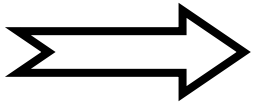
Chemistry
(make molecules)

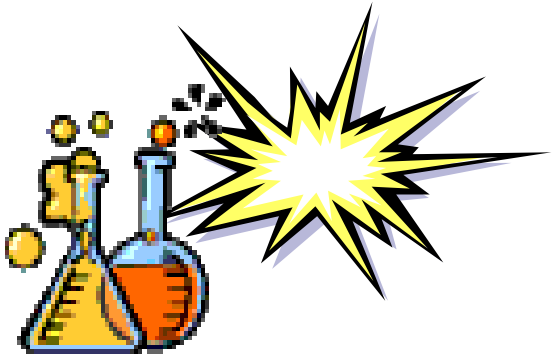


Cooling

OR

Cooling

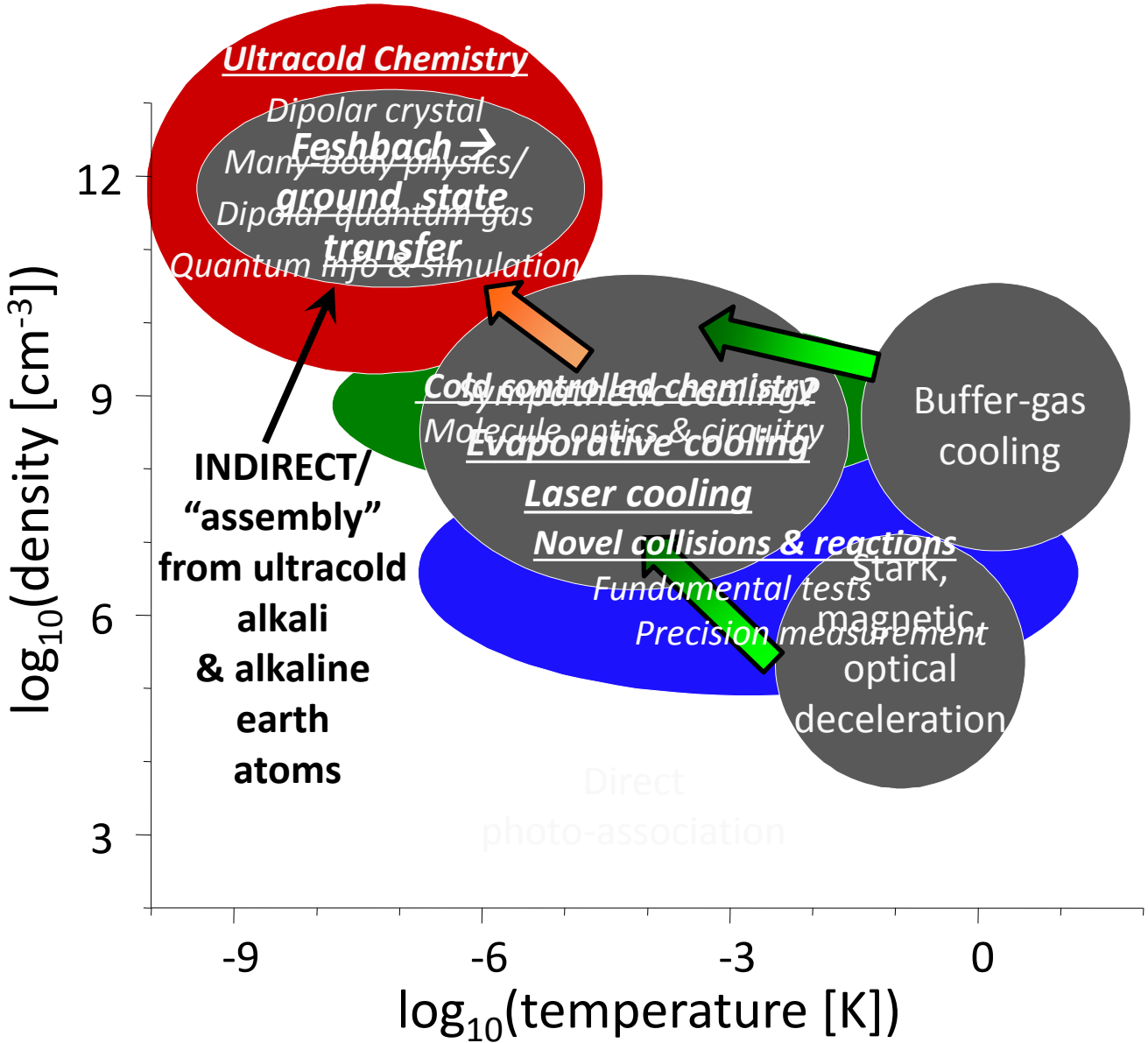




Chemistry
(make molecules)

Diverse physics goals w/cold & ultracold molecules:

- *Different conditions & types of molecules desired*
- *Different techniques address different needs*



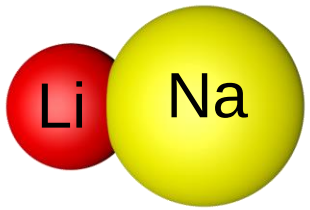
DIRECT

Cooling of wide variety of chemically diverse species

New molecular species for quantum-degenerate gases

Starting point for field: ultracold KRb (Ye/JILA)
NOW: *many interesting new features to explore*

LiNa



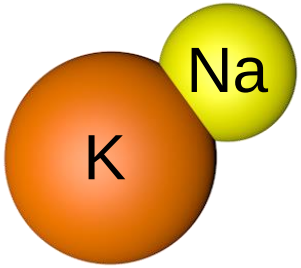
Ground state
long-lived

Large rotational energy

Moderate dipole moment

Ketterle

NaK



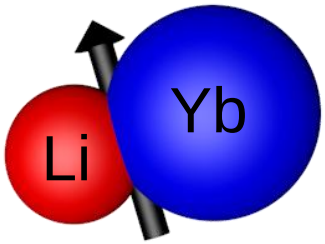
Ground state
chemically stable

Wide Feshbach
resonances

Large dipole moment

Zwierlein

LiYb



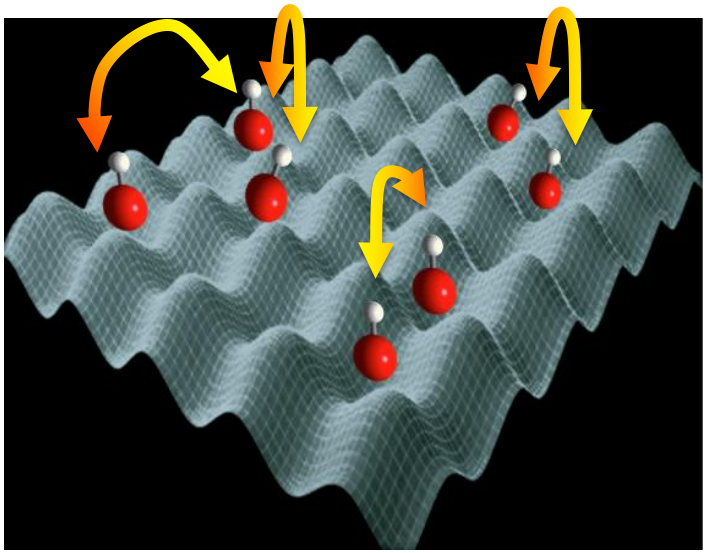
Ground state has
magnetic moment

Long-lived
excited states

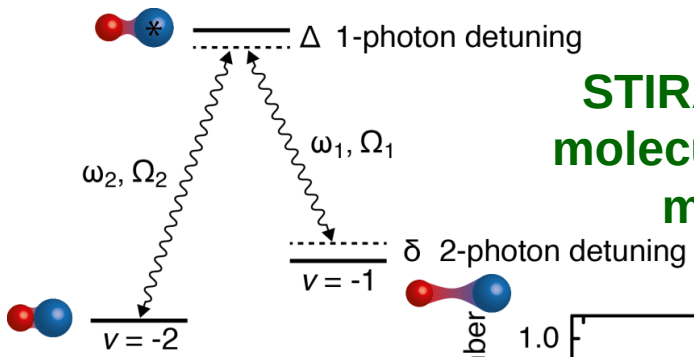
Small dipole moment

Gupta

Molecular gases near quantum degeneracy in the MURI

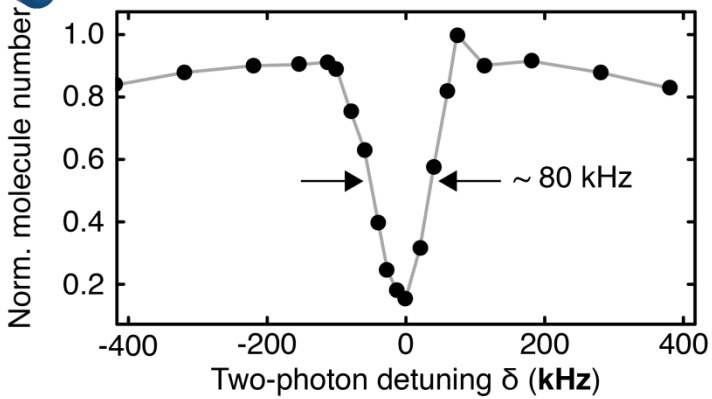


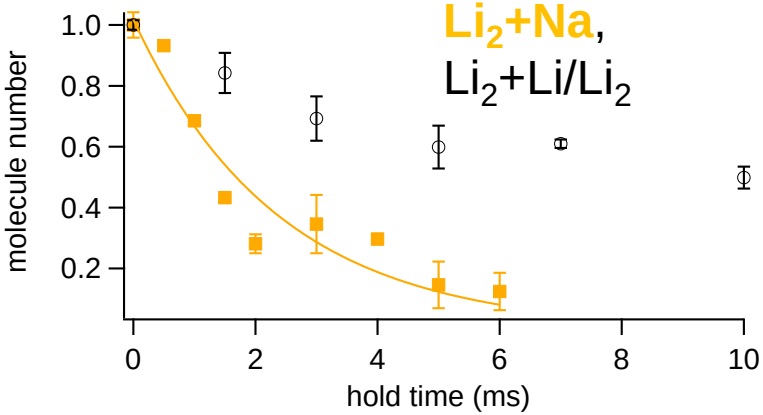
Quantum-Zeno-suppressed chemical reactions with KRb



STIRAP from Feshbach molecule to deeply bound molecule in NaK

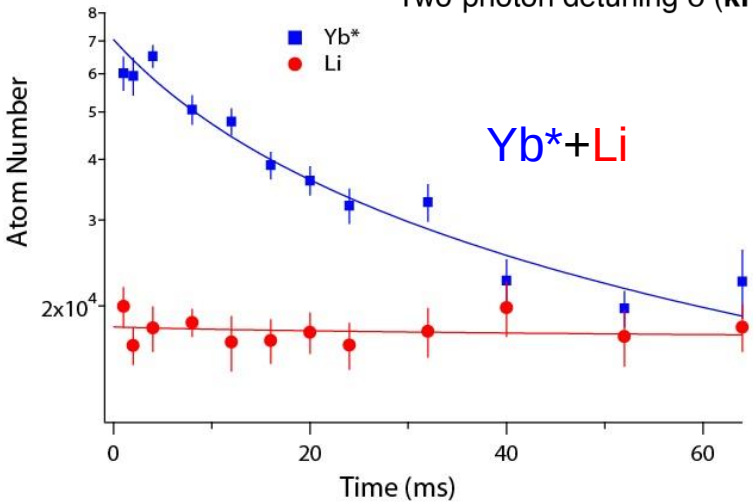
Towards NaK





Deviation from universal behavior in $^6Li_2$ collisions.

Towards NaLi

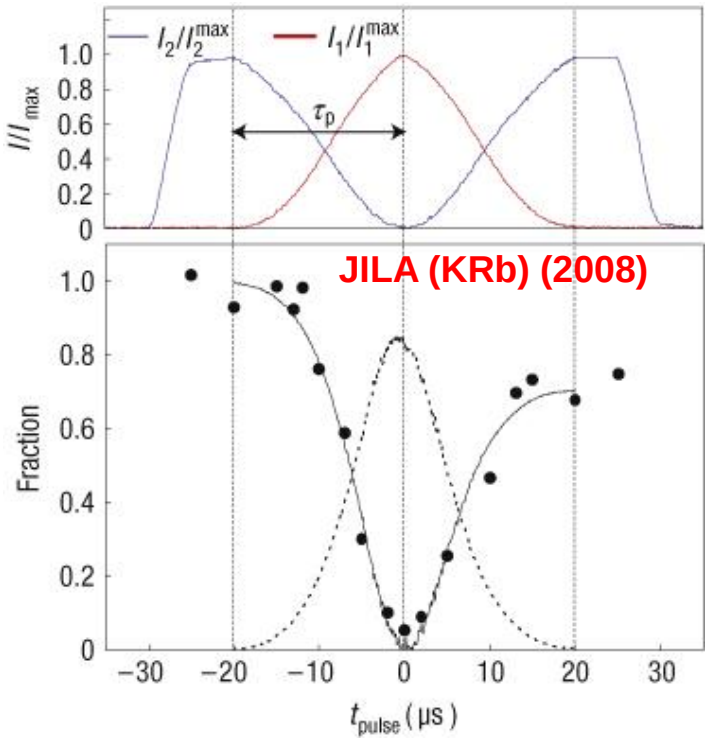


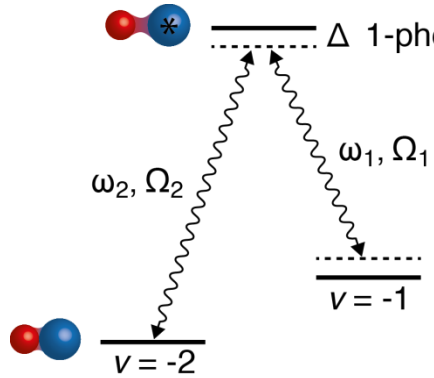
Combining metastable Yb and ground Li.

Towards LiYb

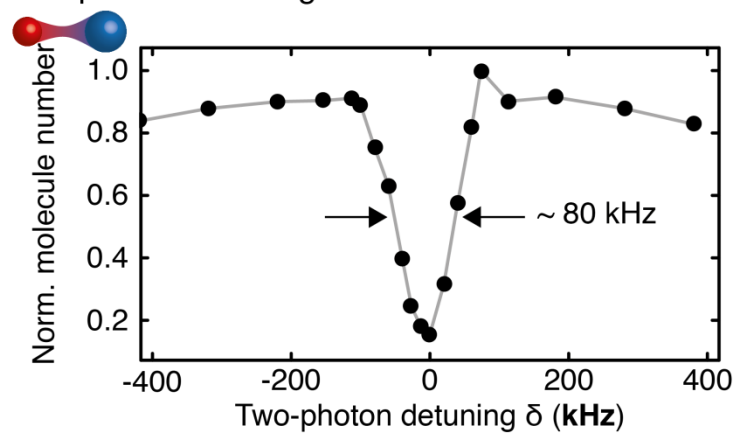
Internal state dependence of reactive collisions

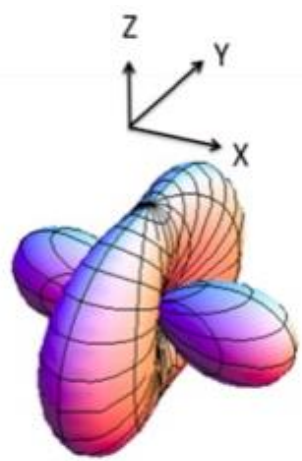
STIRAP: general-purpose tool for preparing arbitrary states





STIRAP from Feshbach molecule to deeply bound molecule in NaK



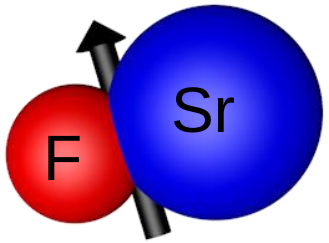


Aligned H2 molecular state Produced via Stark-chirped STIRAP

- State-to-state reactive collision cross-sections w/*aligned* H_2 (Zare/Mukherjee)
- Preparation of targeted vibrational wavepackets for control of collisions (Ye)
- Vibrational "activation" of NaK reactions, producing bound NaK (Zwierlein)
- Singlet vs. triplet electronic states for reactions (Ketterle/Zwierlein)
- Etc.

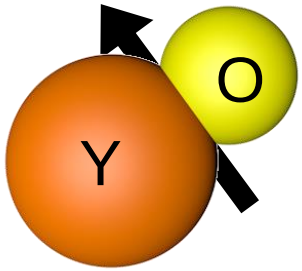
Direct laser/evaporative/sympathetic cooling

SrF



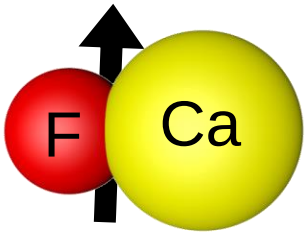
DeMille

YO



Ye

CaF



Doyle

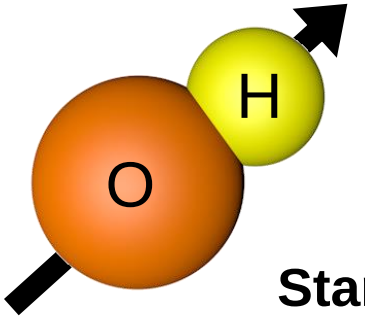
Similar structures: unpaired electron, large dipole *d* >3 Debye, etc.

Starting point: laser cooling demonstrated in SrF

Cooling in YO, CaF; Magneto-Optical Trap for SrF now demonstrated

then collisions, reactions, evaporation, E&B-field control, etc.

OH



Ye

Lambda-doublet substructure

Orbital + spin angular momentum pinned to axis

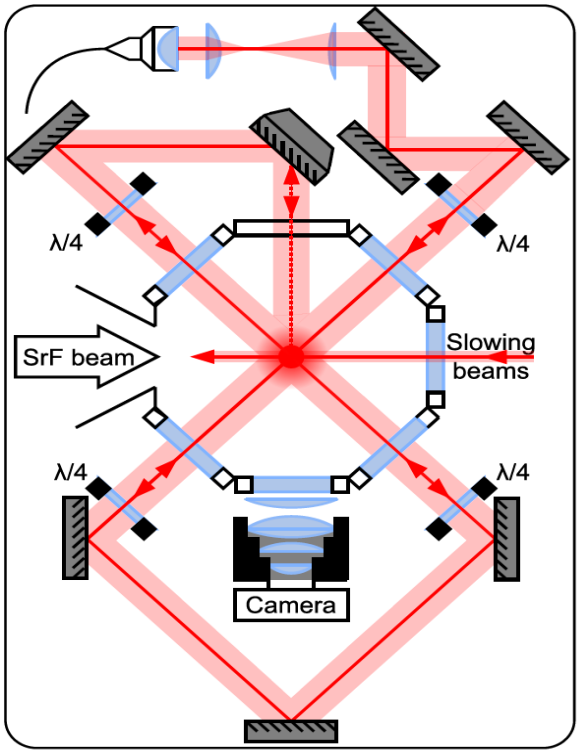
d ~ 1 Debye dipole moment

Large rotational splittings

Starting point: evaporative cooling of trapped OH demonstrated

***E*-field modification of collisions now demonstrated**

Magneto-Optical Trapping of Diatomic Molecules



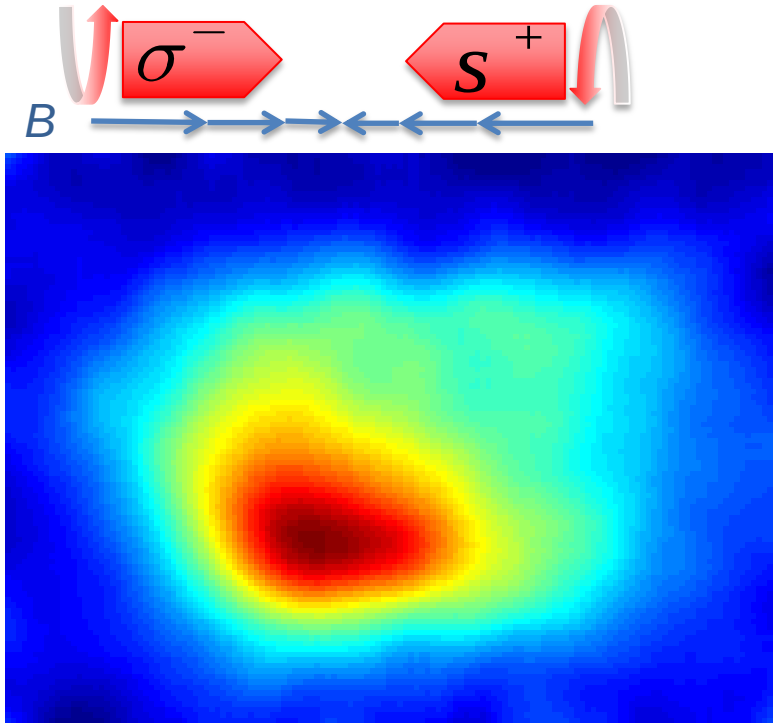
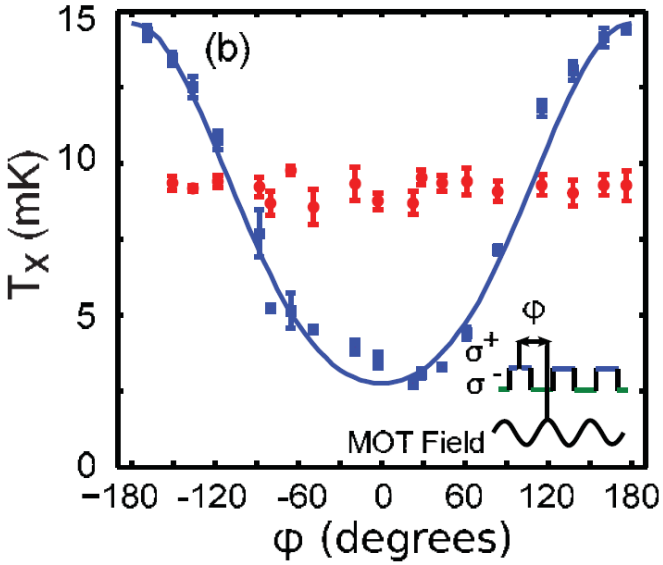
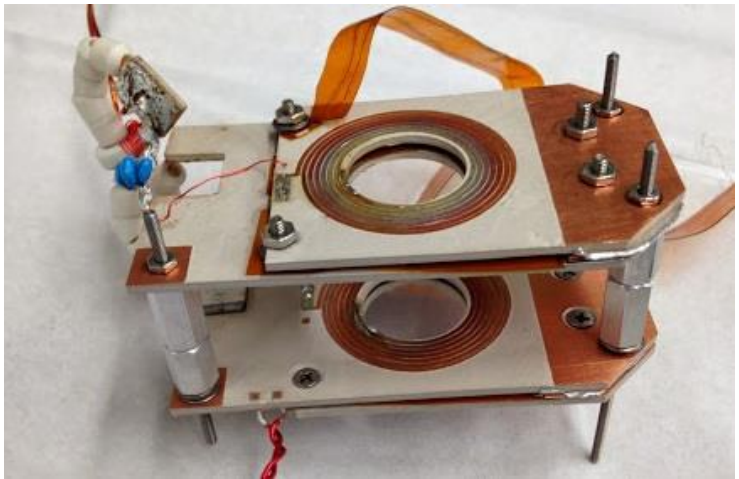


Image of fluorescence from trapped, levitated cloud of SrF molecules at T ~ 2.5 mK

Increased trapping/ cooling forces using RF-modulated MOT

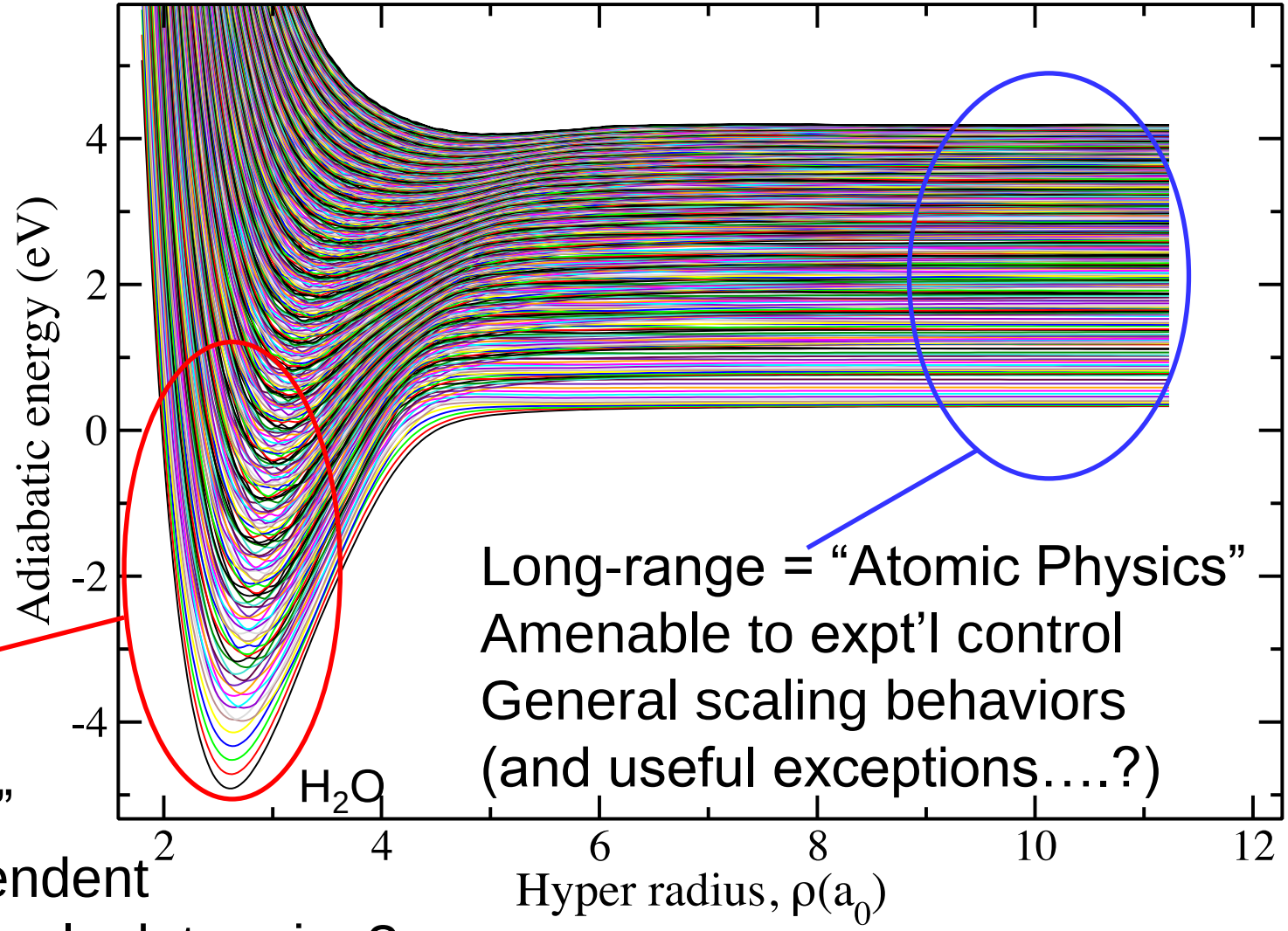


RF MOT coil



The general landscape of molecular collisions

Ex: O + H_2

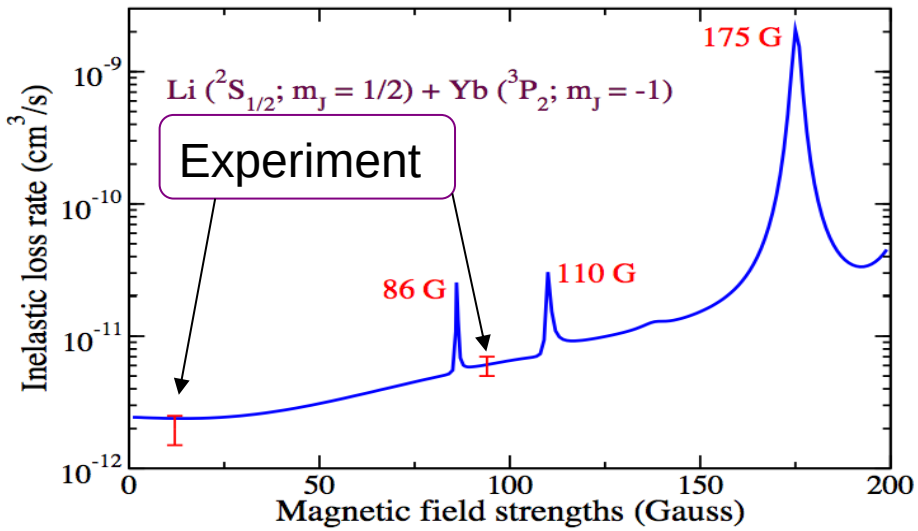


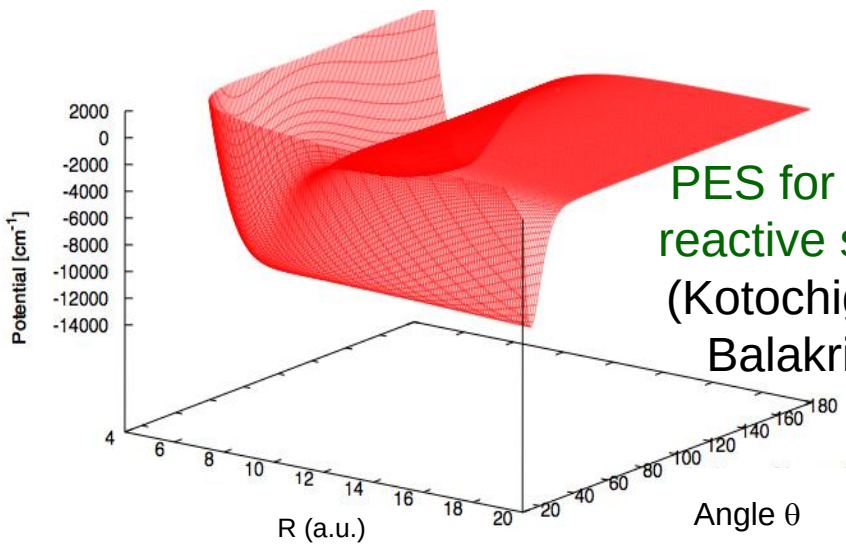
Long-range = "Atomic Physics"
Amenable to expt'l control
General scaling behaviors
(and useful exceptions….?)

Short-range
= "Chemistry"
species-dependent
Can we precisely determine?

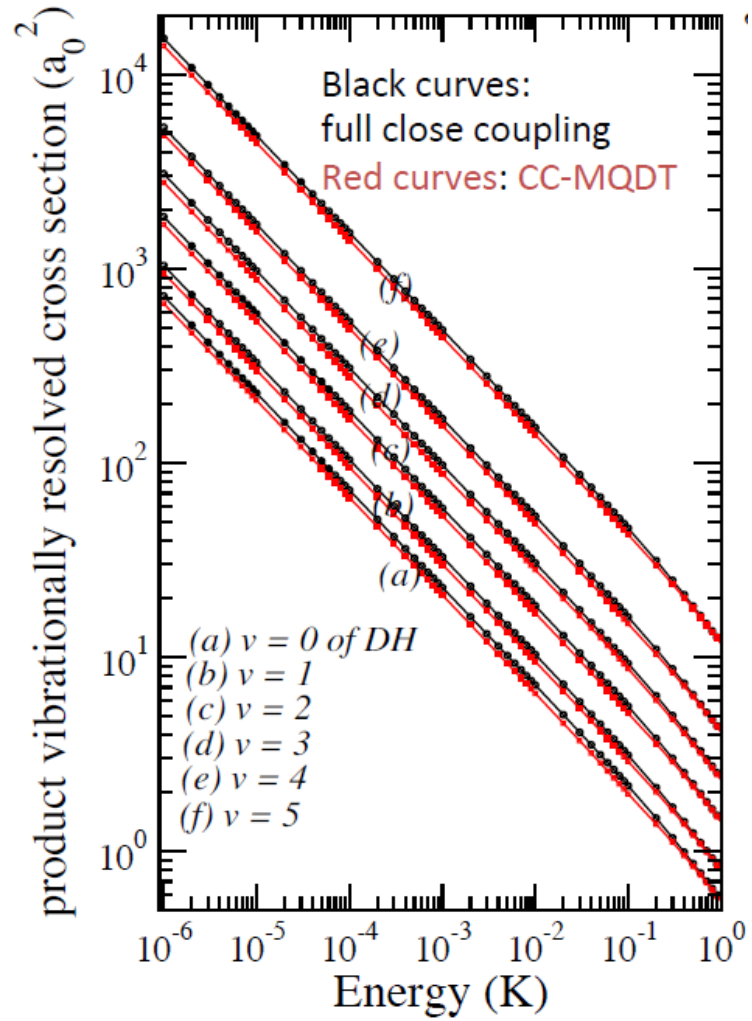
Some recent work from our MURI theorists

Fano-Feshbach resonances in metastable LiYb*
(Kotochigova and Gupta);



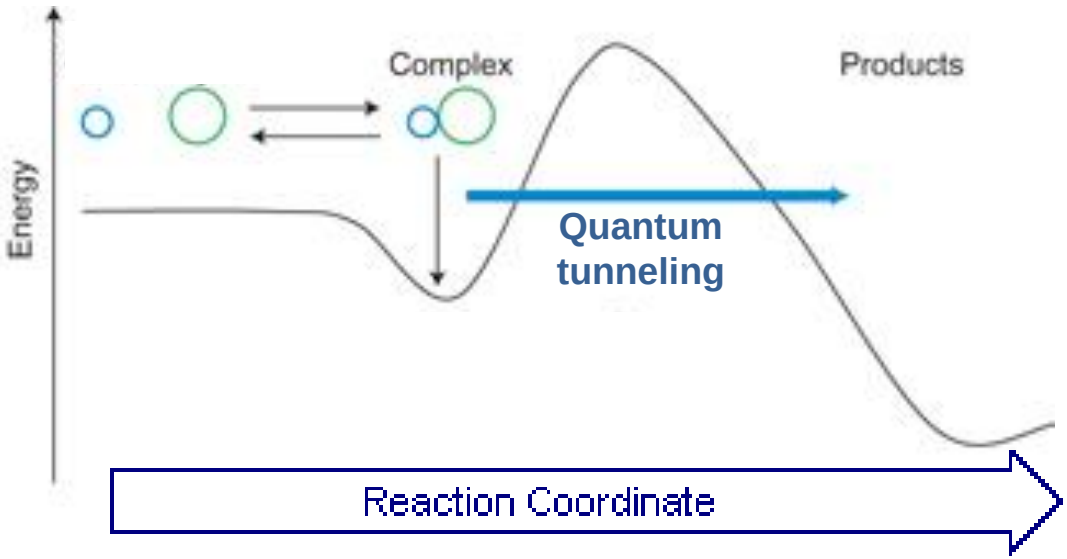


PES for Li + LiYb
reactive scattering
(Kotochigova and
Balakrishnan)

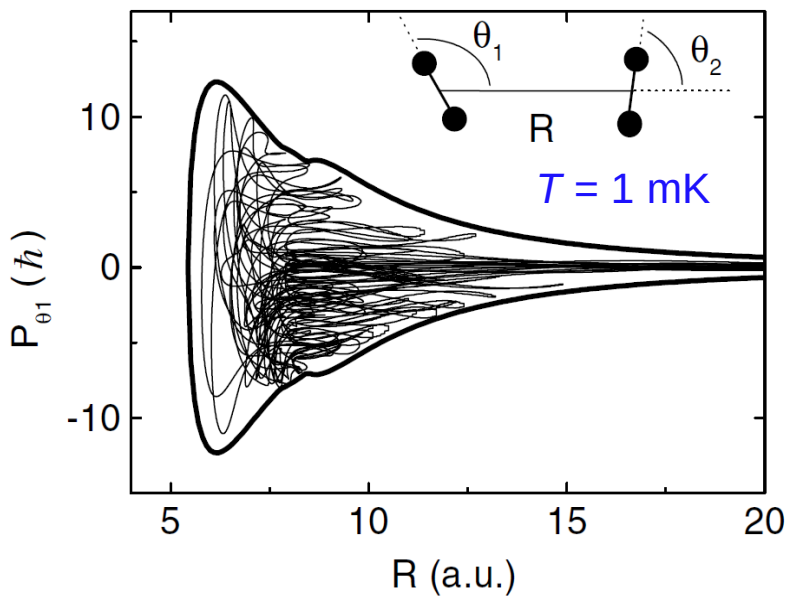


molecule-molecule
inelastic rate for D + H_2:
hybrid calculation method
dramatically simplifies computations
(Bohn and Balakrishnan)

Novel long-lived collisional resonances: gateway to chemical reactions at low temps?



Q: Is tunneling rate ever large enough to observe barrier reactions in ultracold gases?



Bohn:
Cold collisions *GENERICALLY* exhibit long-lived collisional resonances
⇒ many "attempts" to tunnel…?

Possible implications for chemical reactions in interstellar space/high atmosphere…?