# MOTION trap: A hybrid neutral-ion environment

## for fundamental physics, quantum chemistry, and quantum information

Eric R. Hudson
Department of Physics and Astronomy

### A new direction:

- Introduction to the MOTION trap
- The Neutral-Ion Interaction
- Ultracold charged Molecules

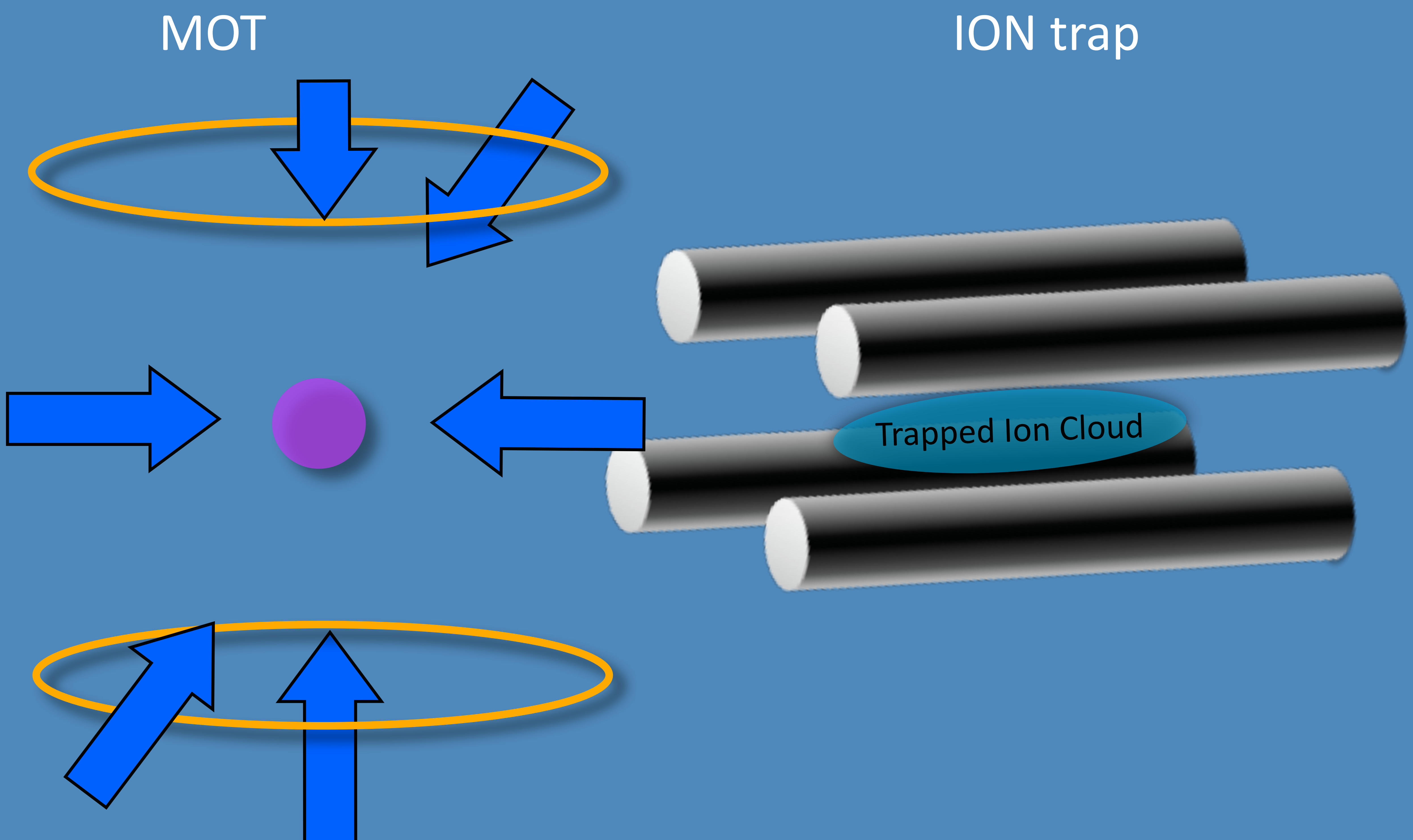
### The MOTION trap:

- Part 1: atom-ion quantum chemistry
- Part 2: Cold Molecular ions

This work was funded by the US ARO and NSF

- INTRODUCTION TO THE MOTION TRAP

MOTION TRAP:
COMBINATION OF MAGNETO-OPTICAL TRAP (MOT) AND ION TRAP



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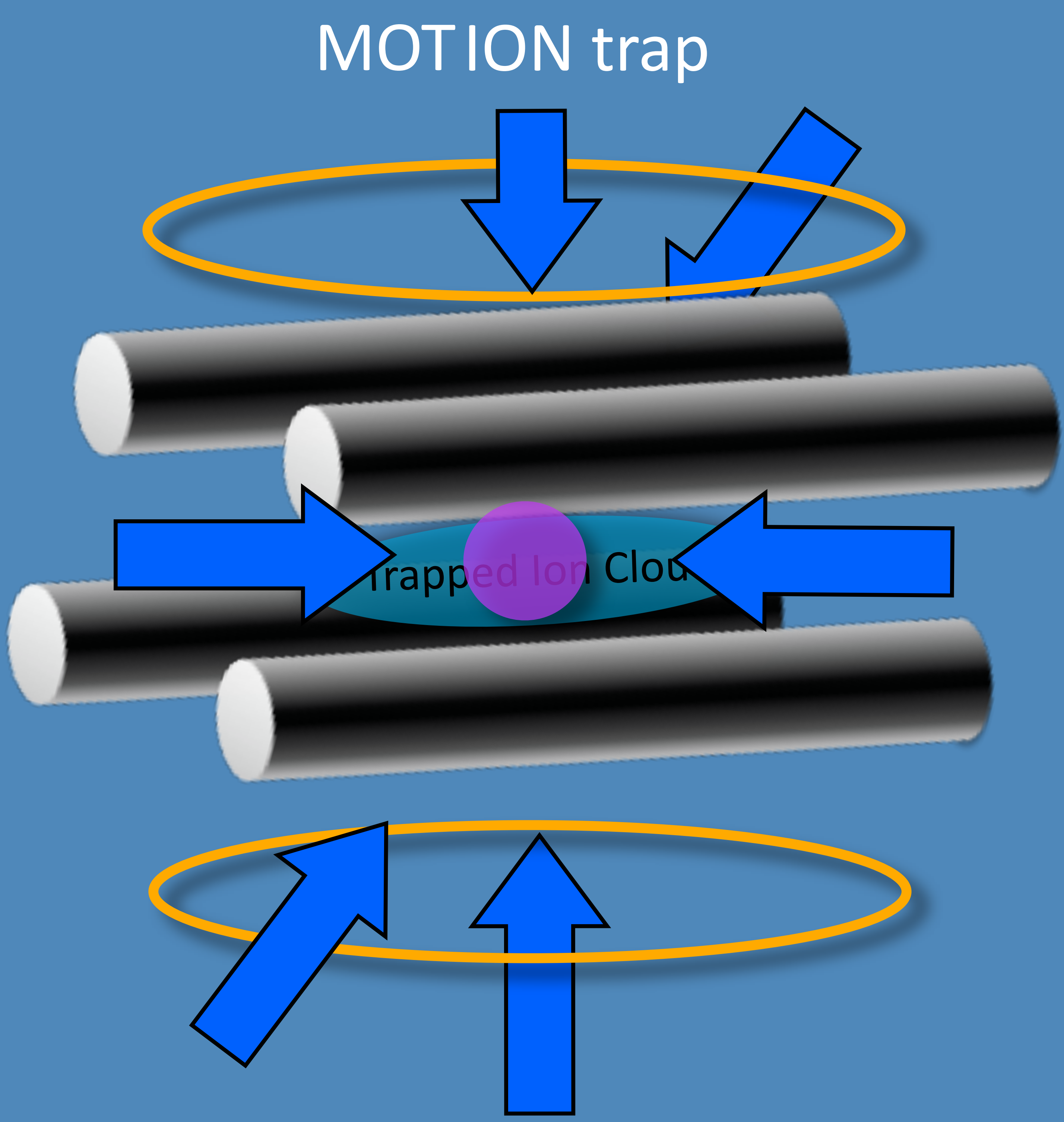

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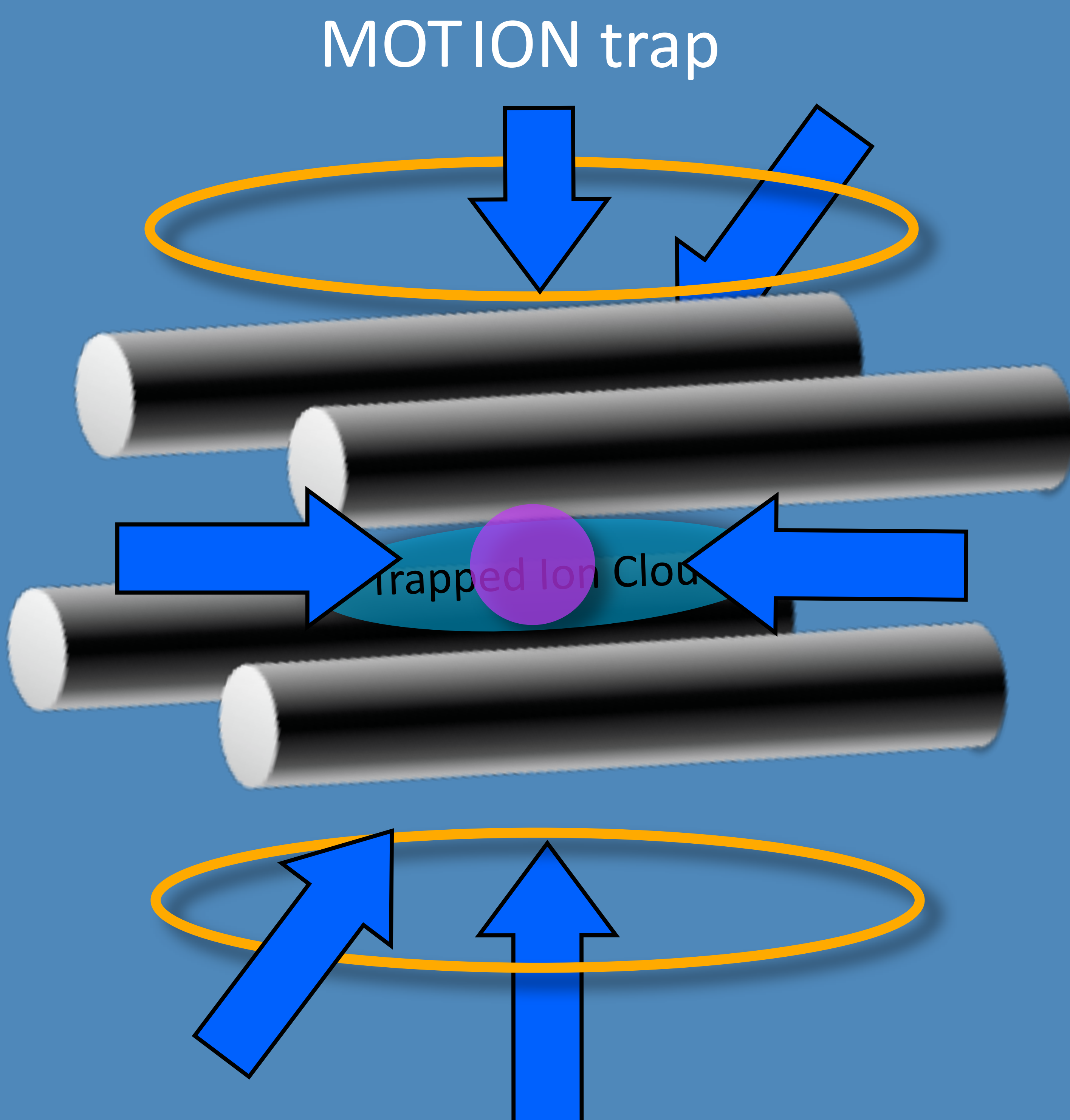


Brings ultracold neutral particles in contact with ultracold charged particles. Allows the study of:

- atom-ion interactions
- quantum chemistry
- quantum information
- cold molecular ions
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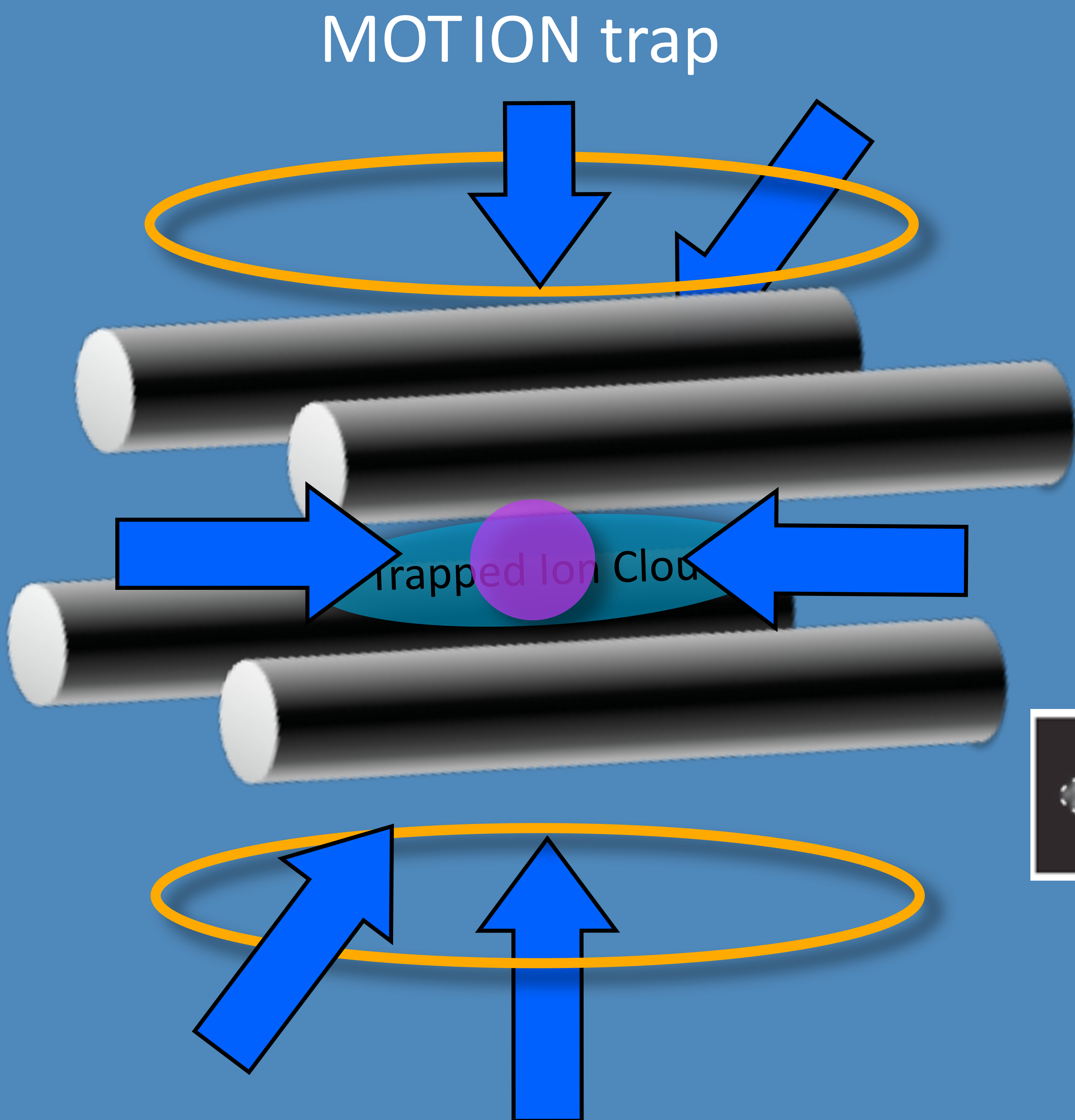


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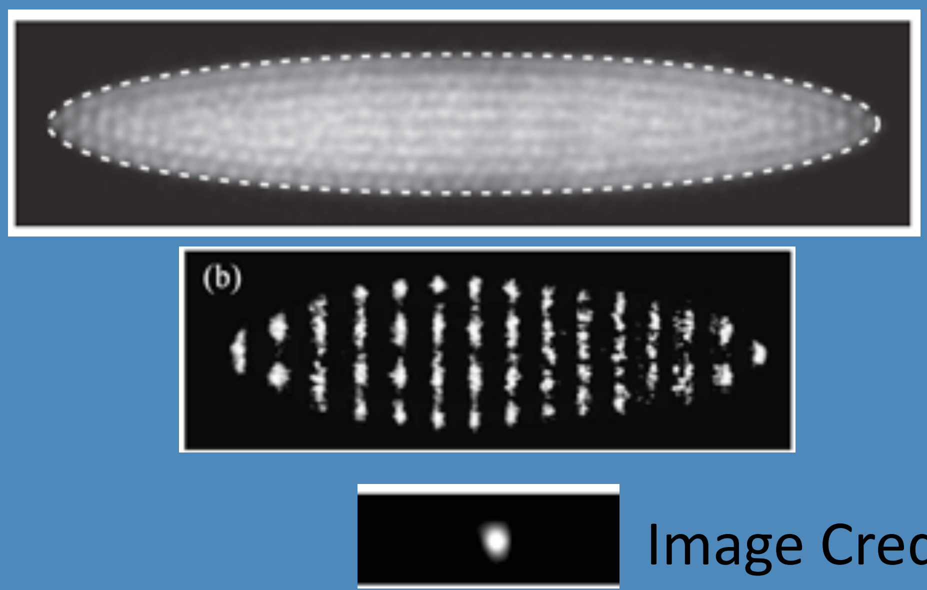
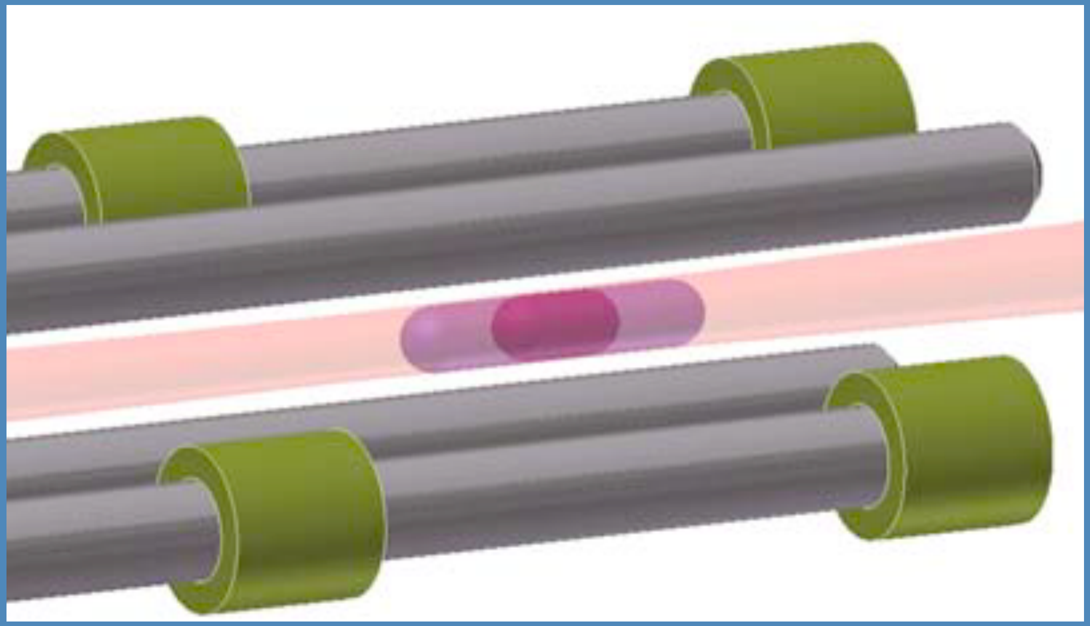



Image Credit: M. Drewsen

# Experimental Implementation of MOT-ION trap



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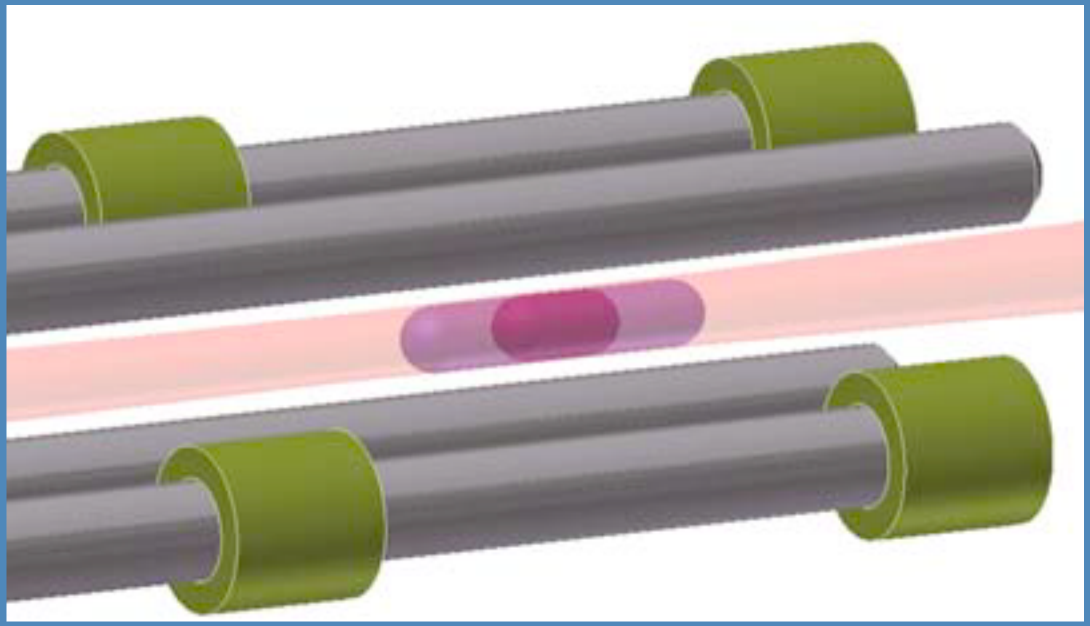

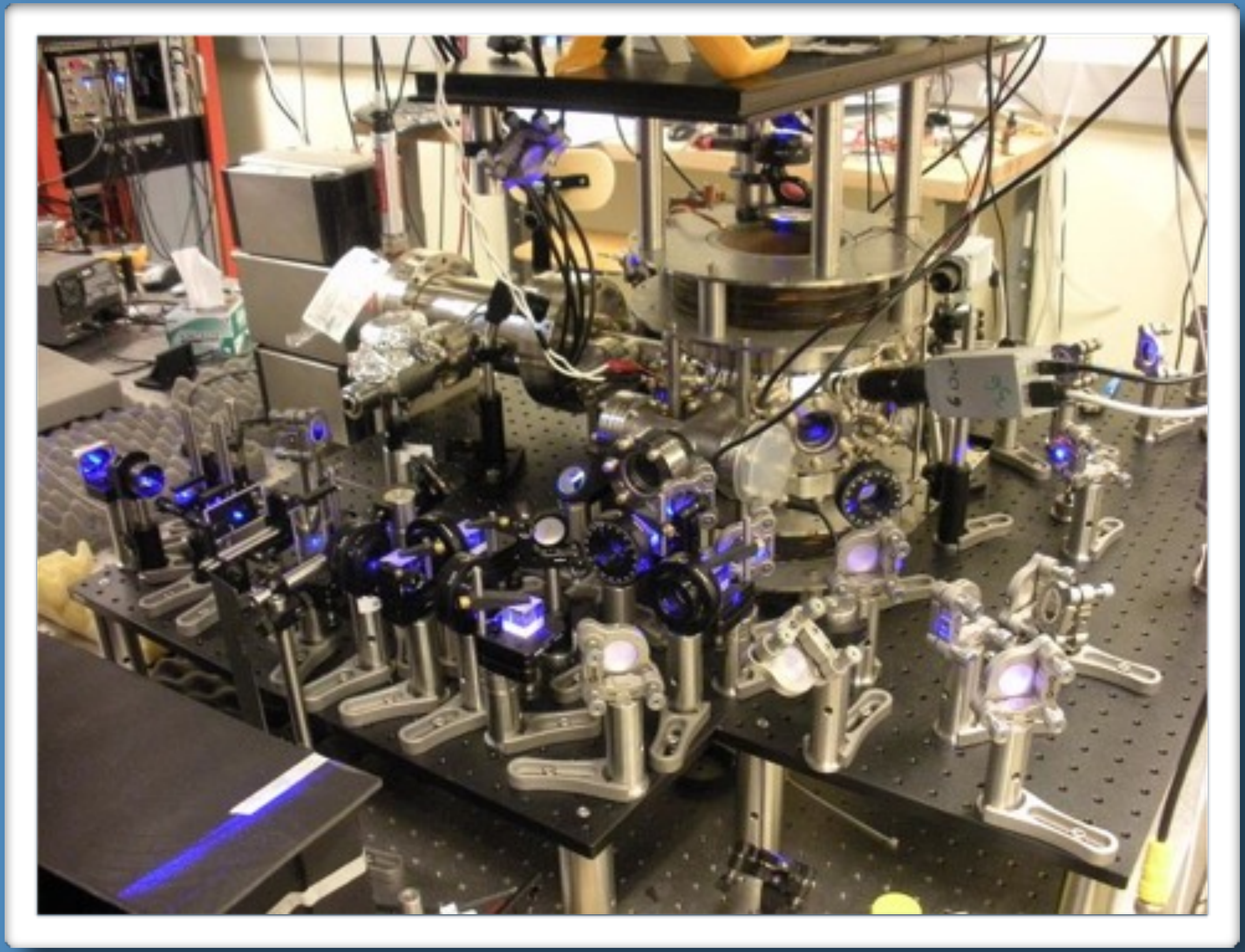

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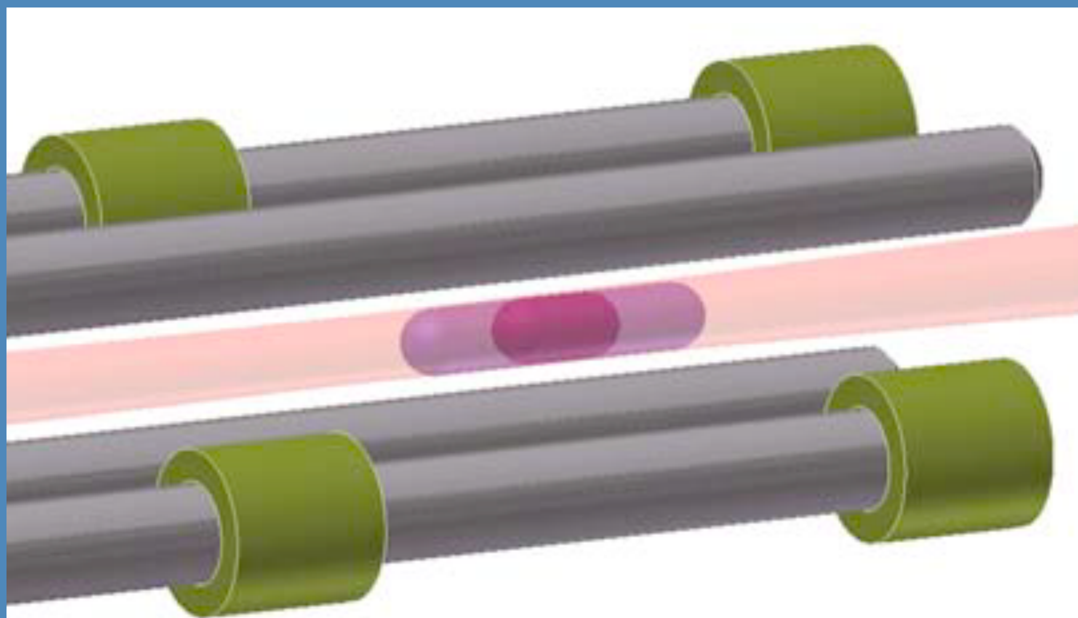

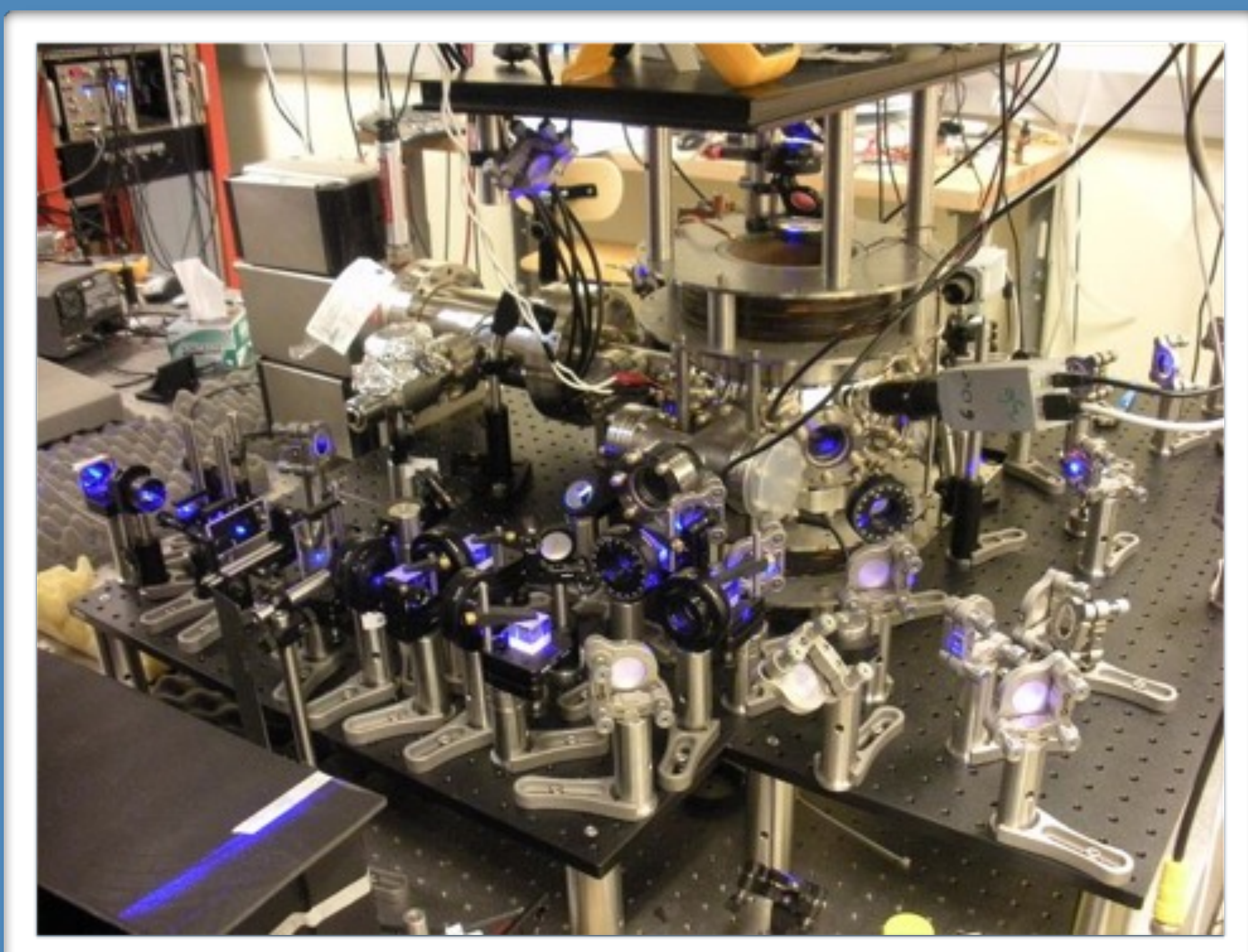

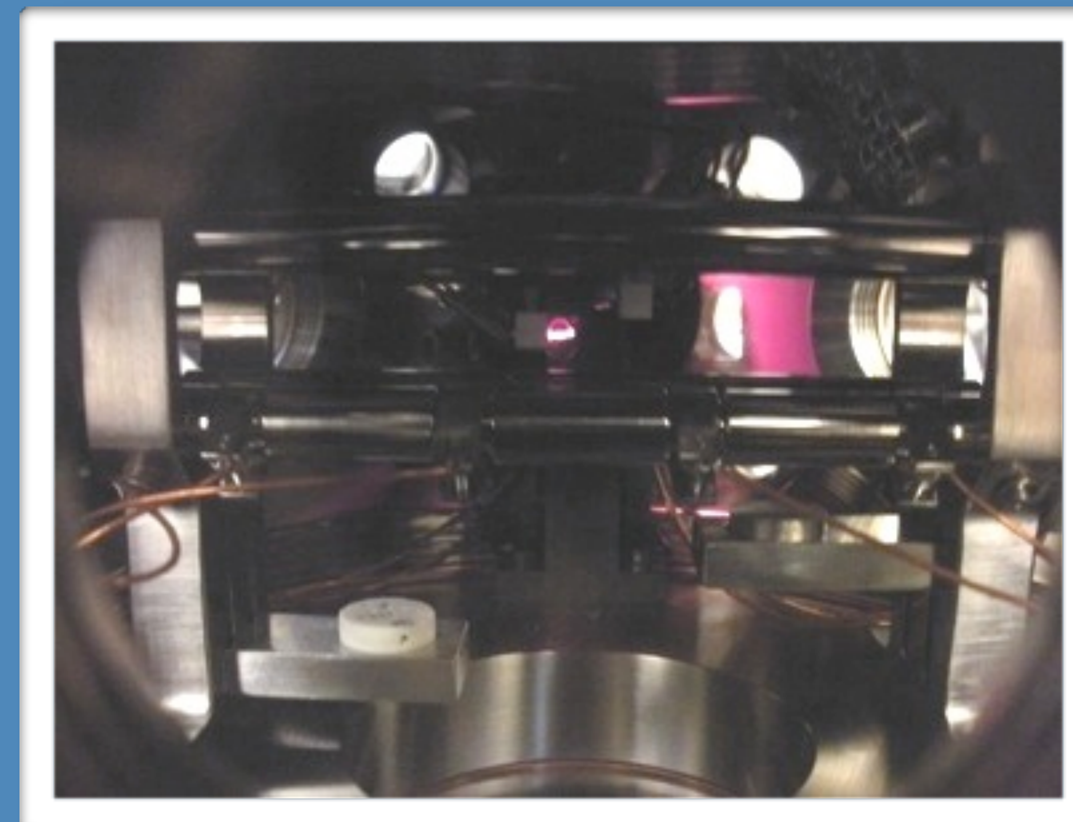

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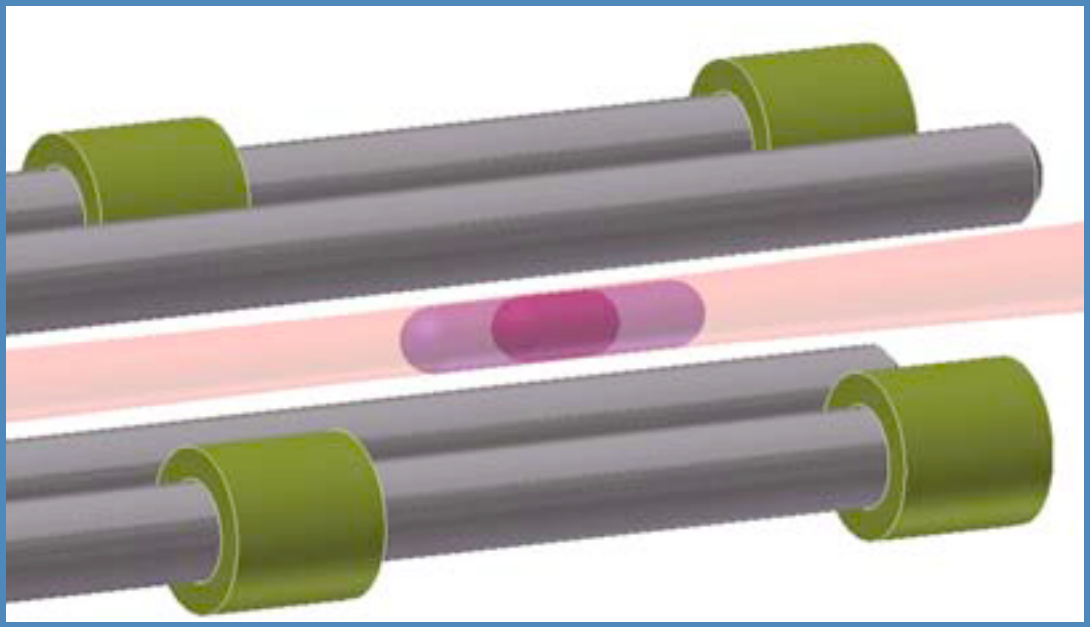
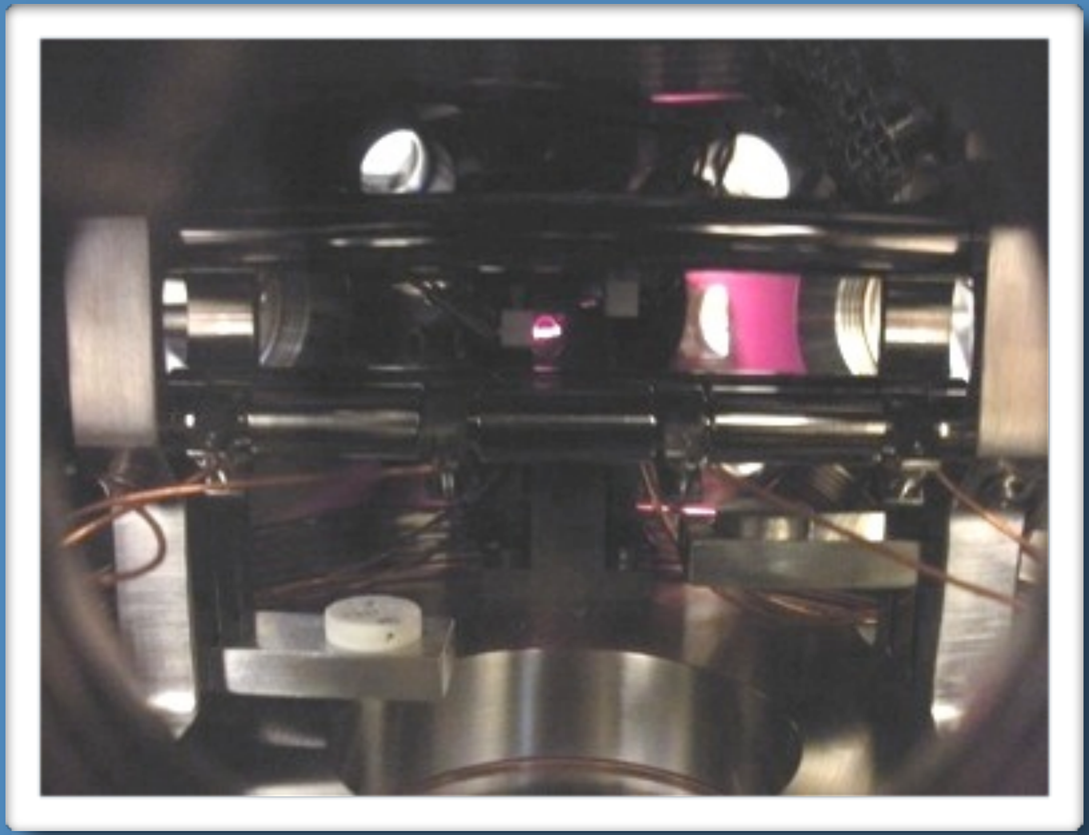
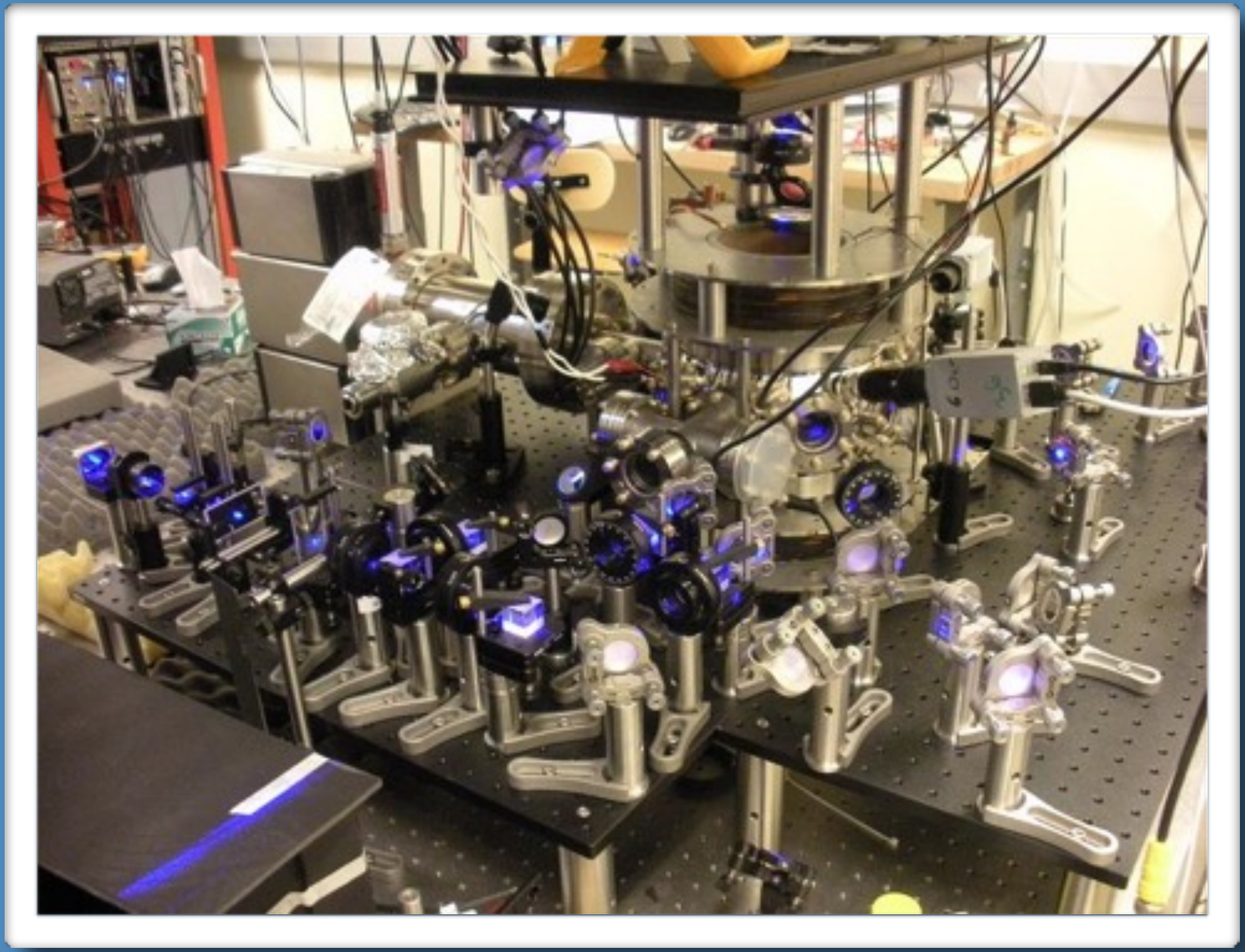
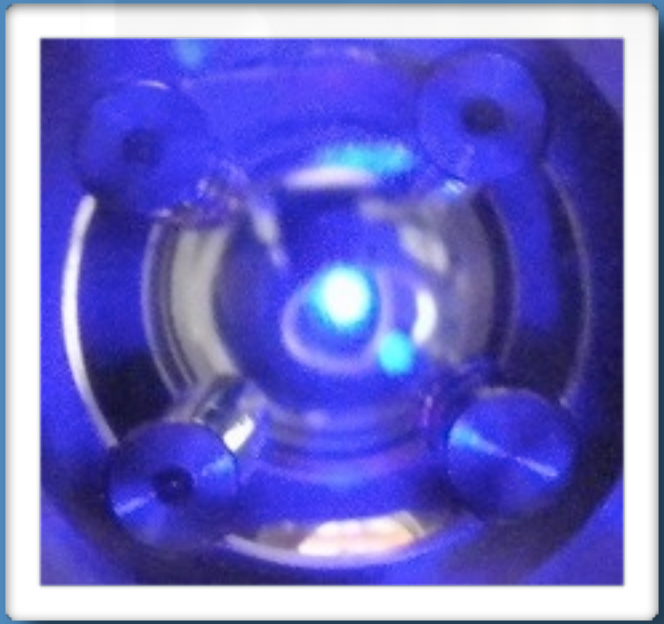








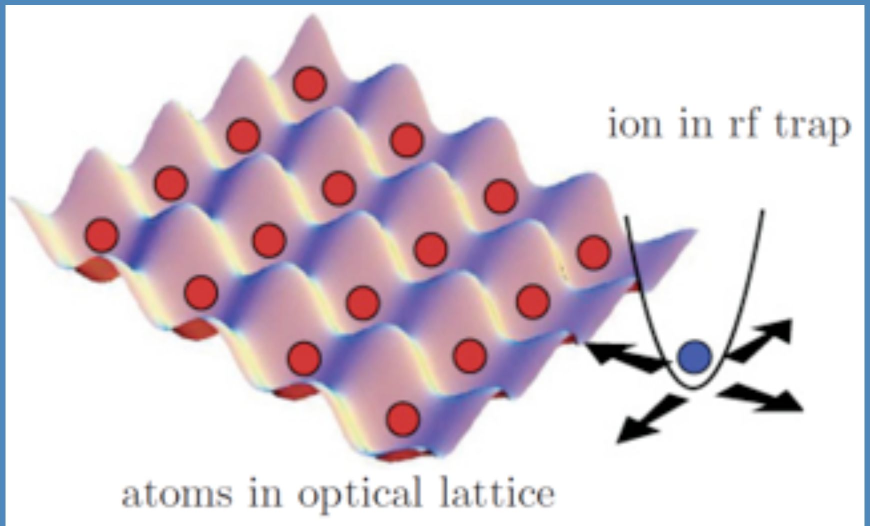
Ca MOT

low mass

reduced

reactivity

# • Using the Neutral - ion interactions




## Implementing Quantum gates

Ion entangles atoms

Idziaszek & Zoller

e.g. PRA 76 033409 (2007)

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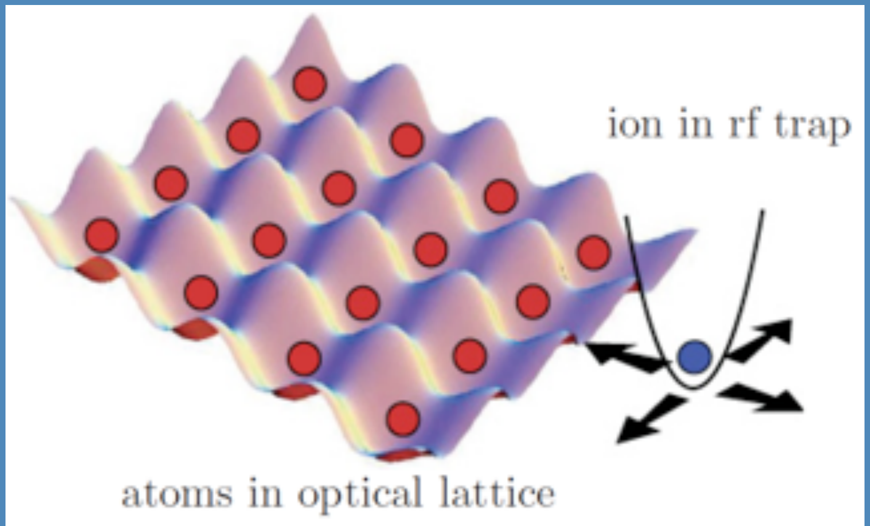


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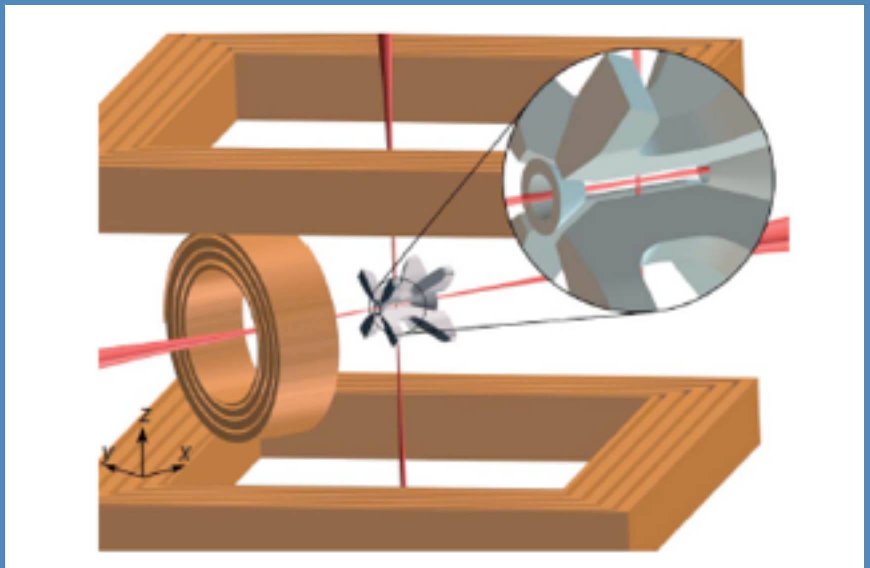

### Probing Quantum gases

Ion alters quantum gas, novel interactions, potential probe

Koehl & Denschlag

e.g. Nature 464 388 (2010).

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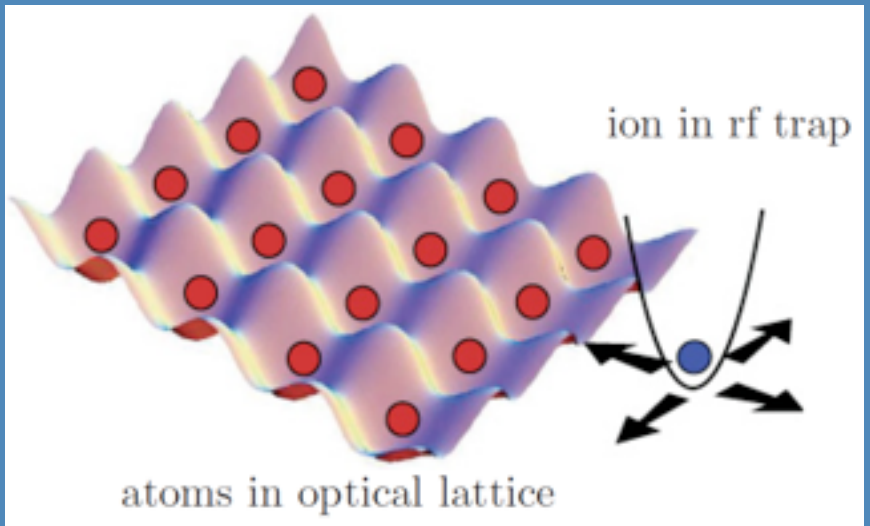


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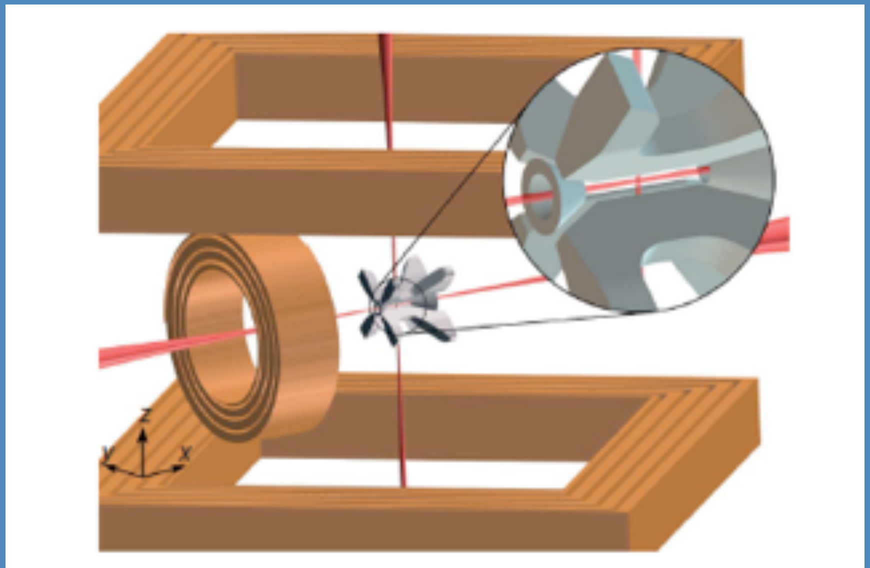

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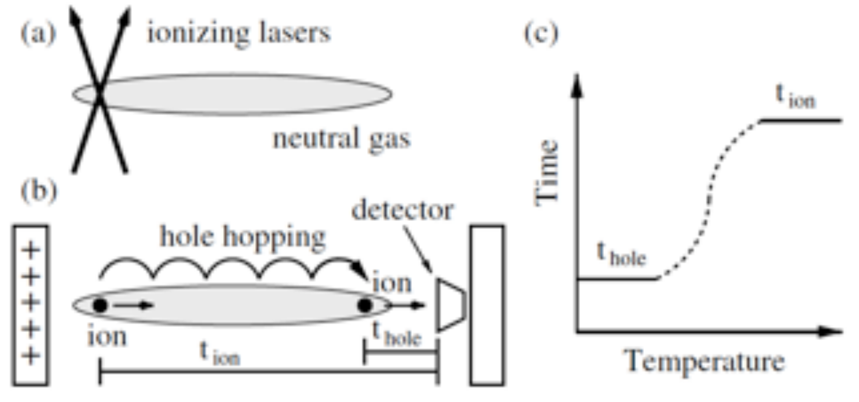


## Novel Charge transport

Quantum simulation of charge transport
Cote
e.g. PRL 85 5316 (2000).

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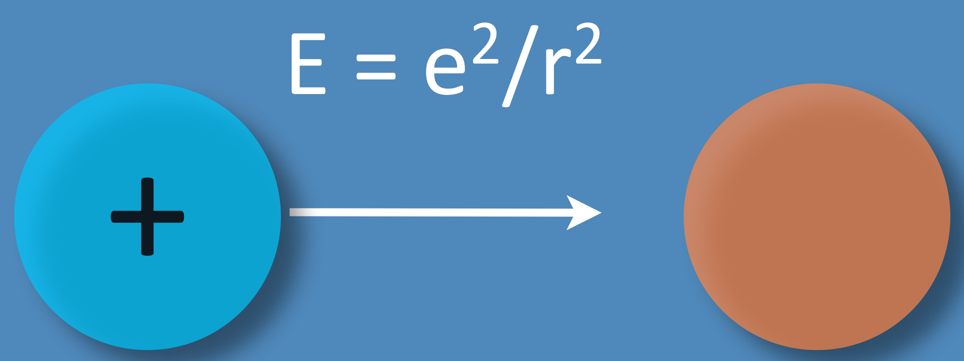

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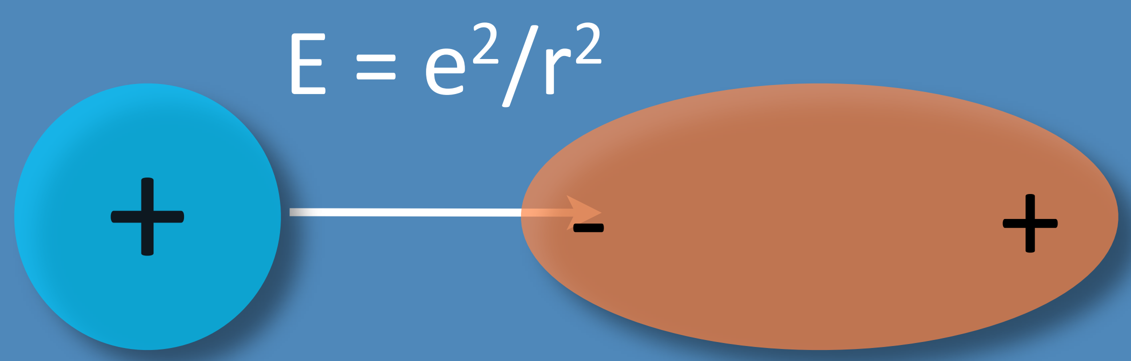
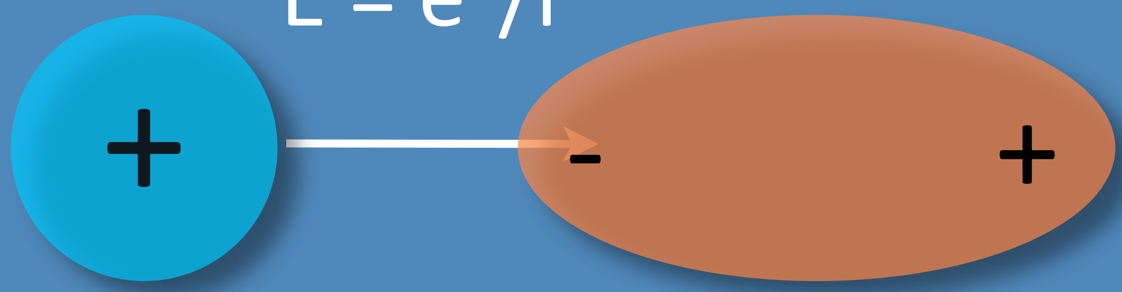


- THE NEUTRAL - ION INTERACTIONS

# ATOM AND ION INTERACT WITH $1/r^4$ SEPARATION DEPENDENCE

$E = e^2/r^2$

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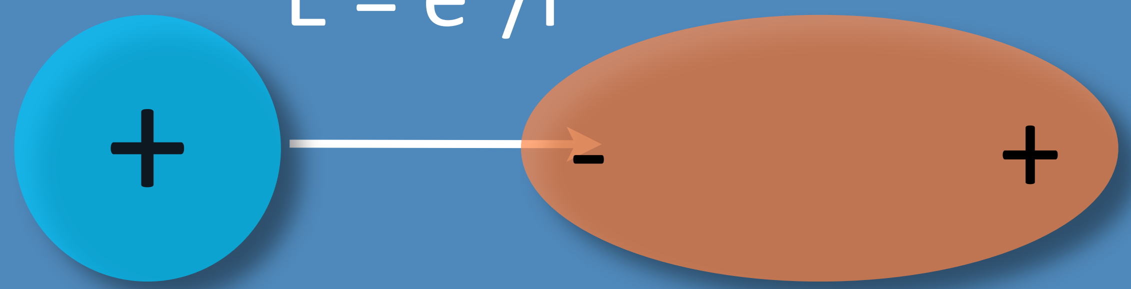

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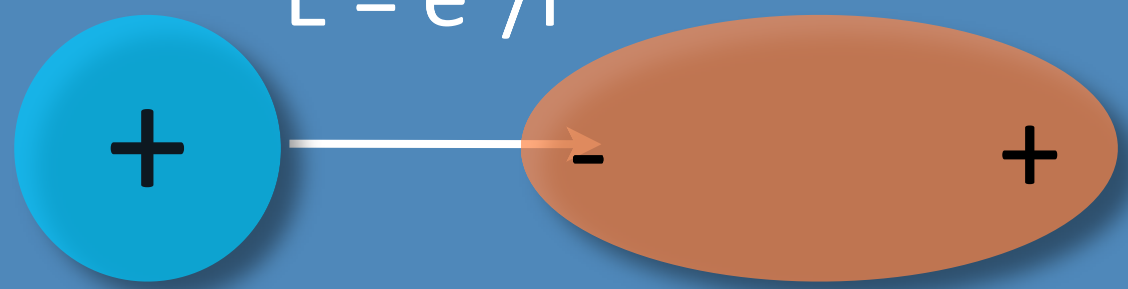


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Before applications & Technologies are possible there is still Lots to learn! Primarily:

- atom-ion (quantum) Chemistry
- ultracold atom-ion collisions

# • Charged Molecules Vs. Neutral Molecules

## They still rotate

Moment of inertia

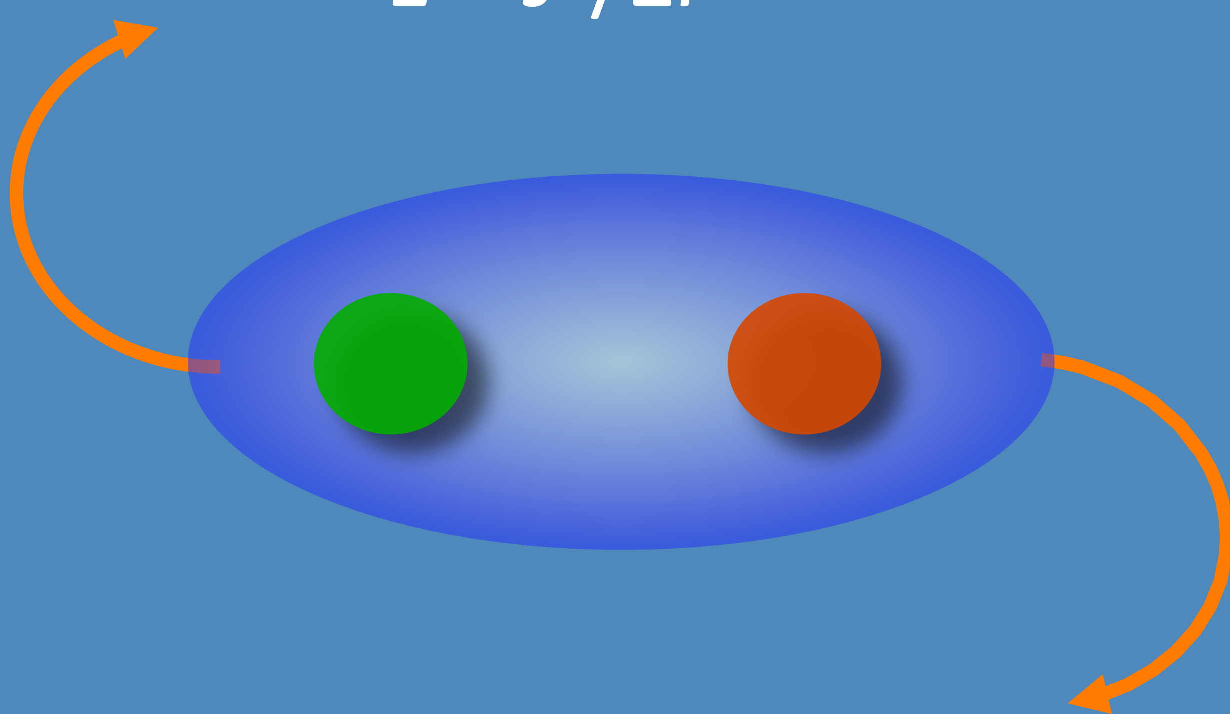
$I = MR^2$

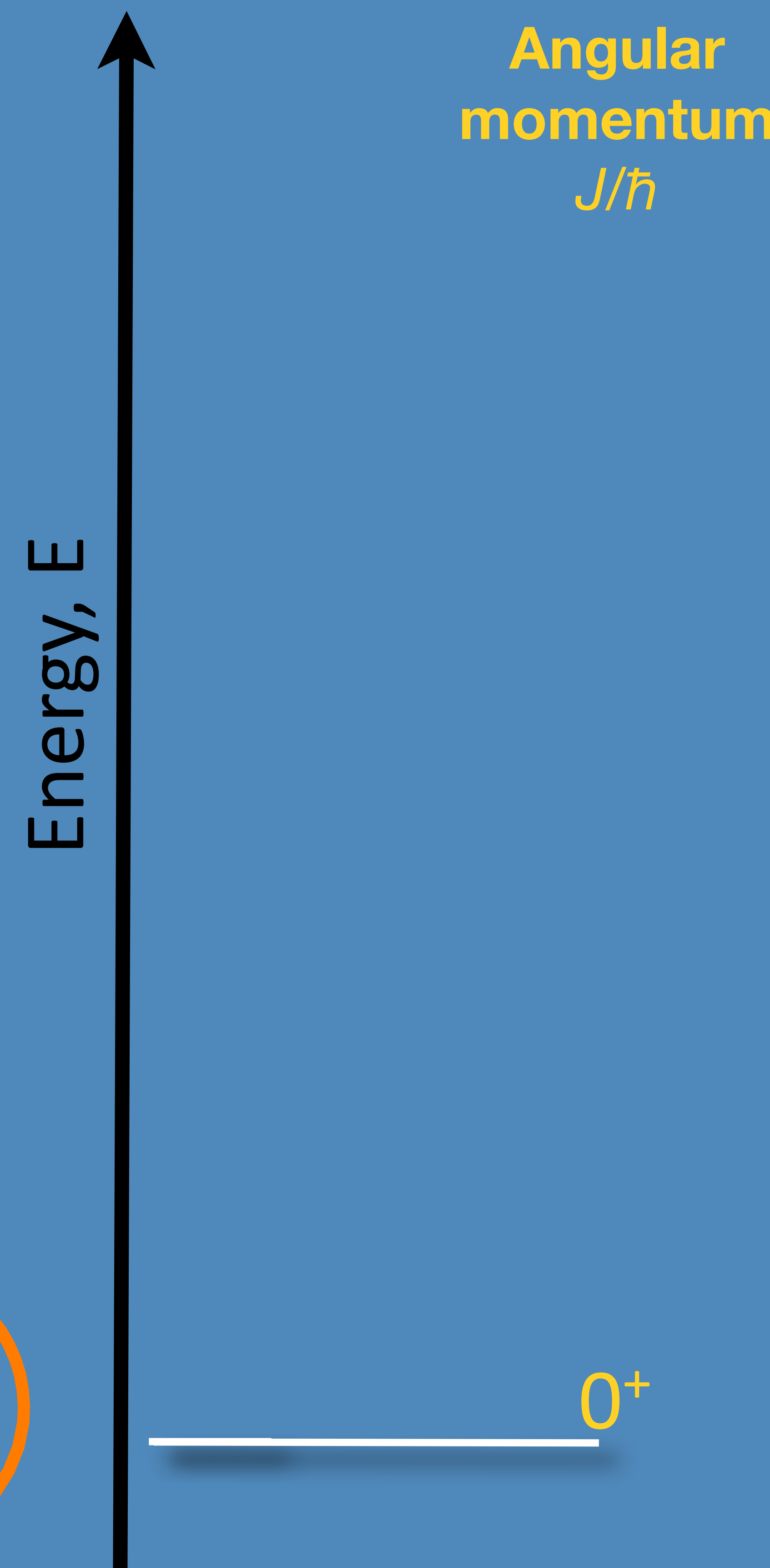
Angular momentum

$J = n\hbar$

Energy of rotation

$E = J^2/2I$





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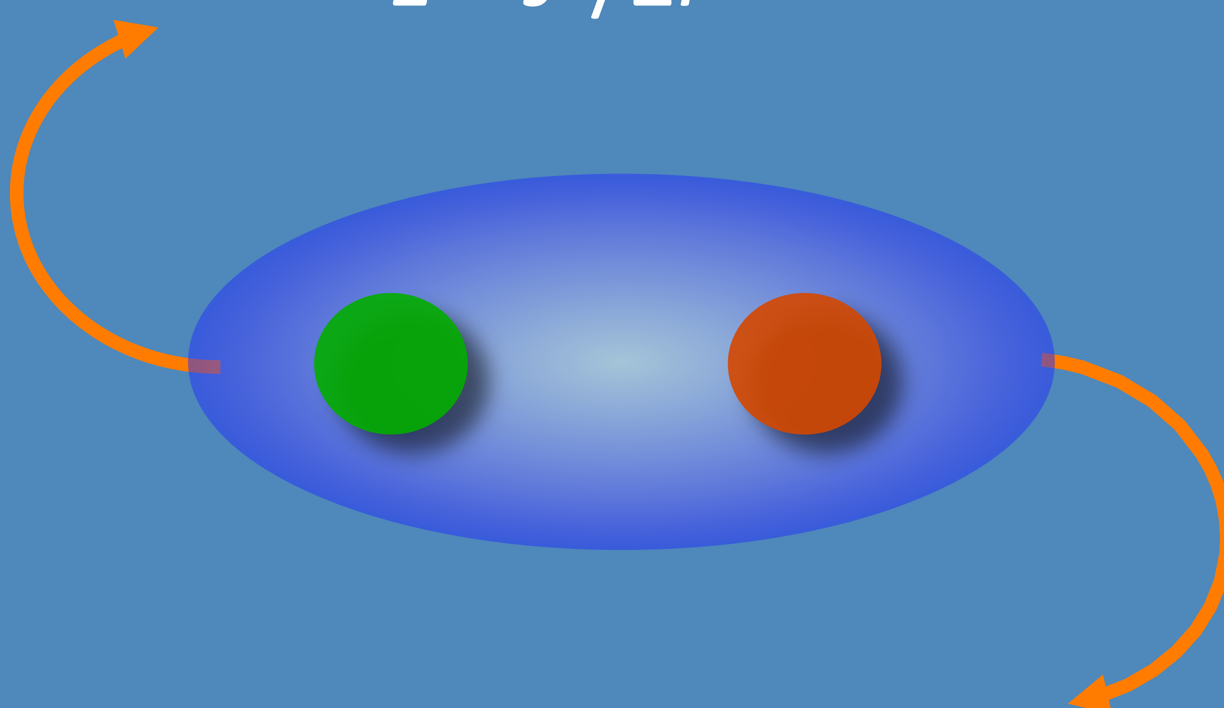

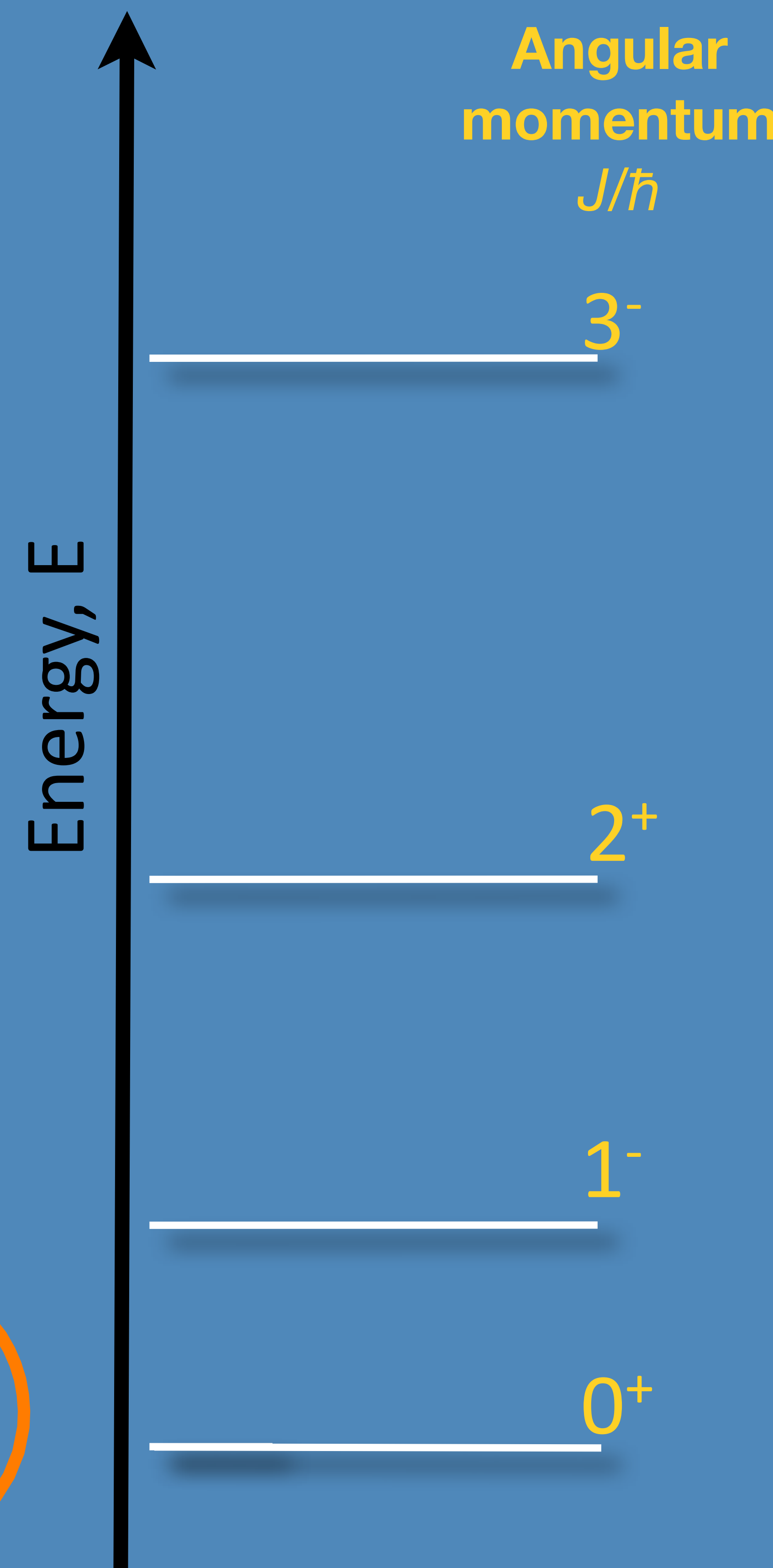

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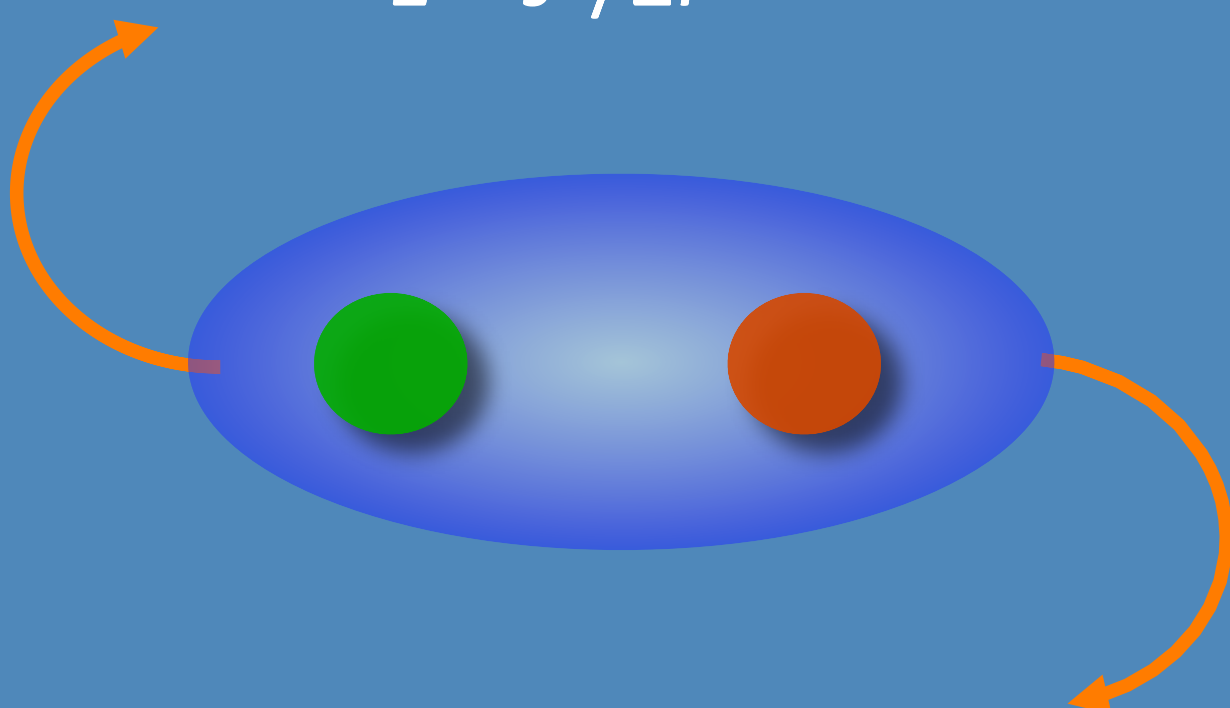

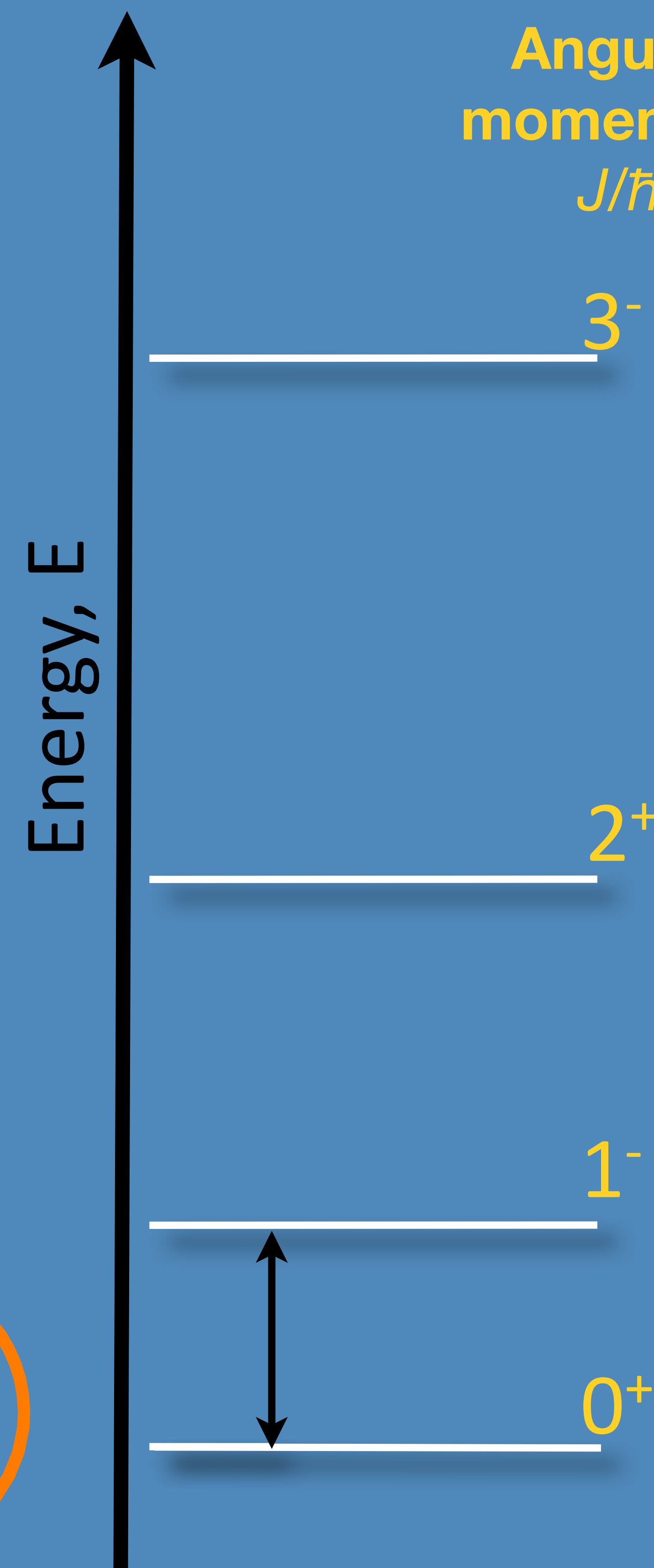


- microwave (GHz-THz)
- control with EM rad.
- Two-level system w/ long lifetime: $10\text{-}10^7$ s

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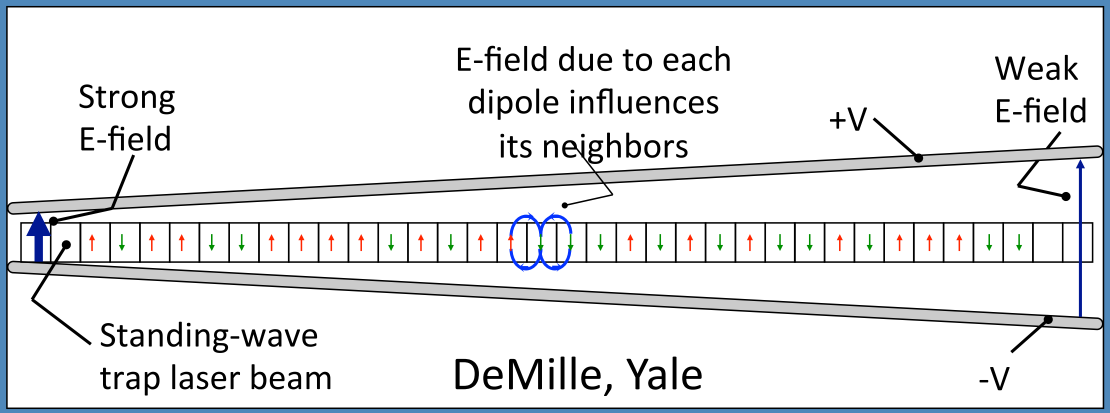
### Quantum computation with ultracold polar molecules



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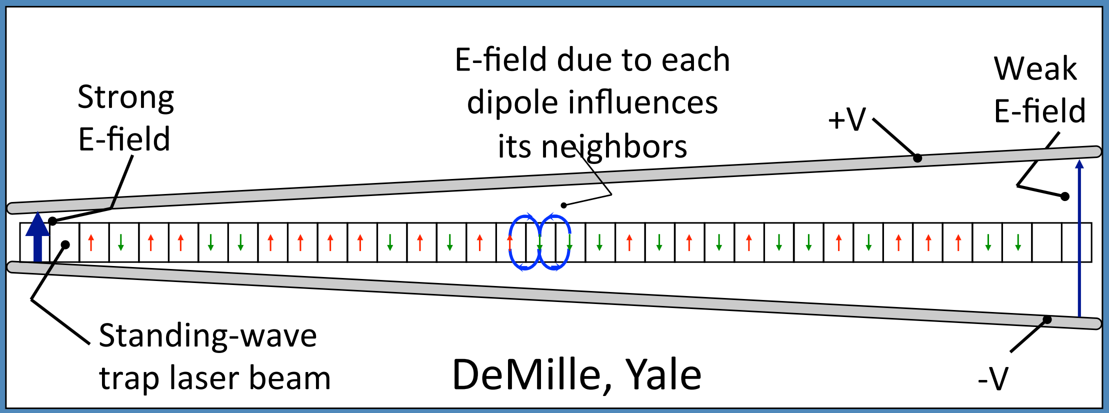
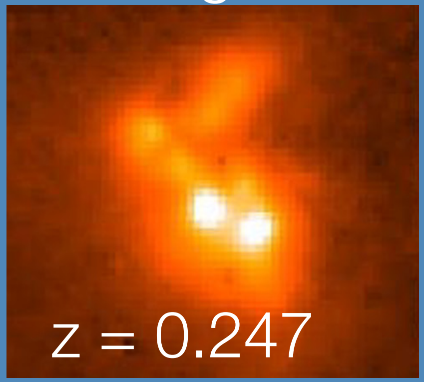
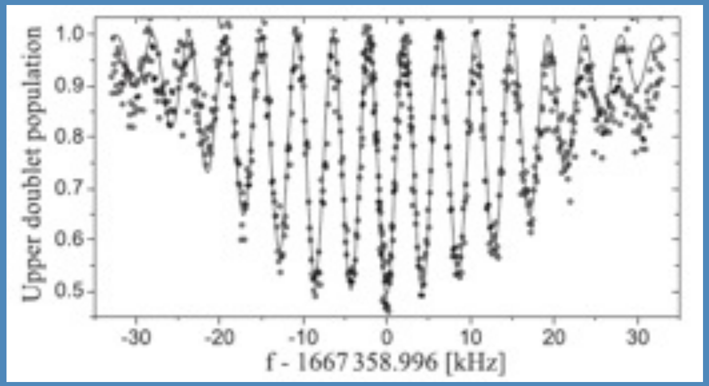



### Fundamental Physics Test

OH Megamasers




Kanekar et al., APJ 716 L23 (2010)

Phys. Rev. Lett. 96 143004 (2006)

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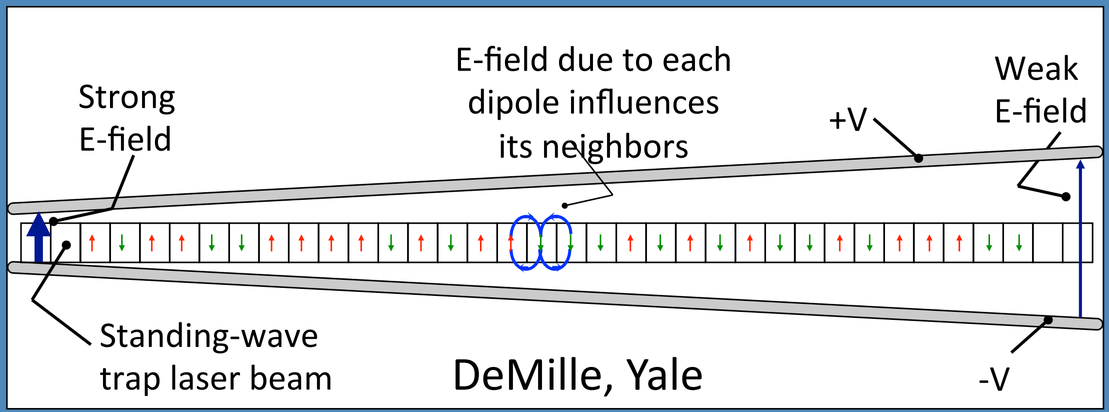


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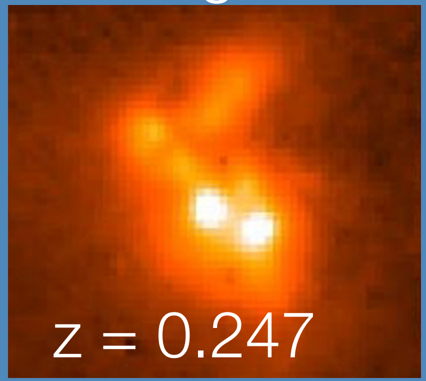


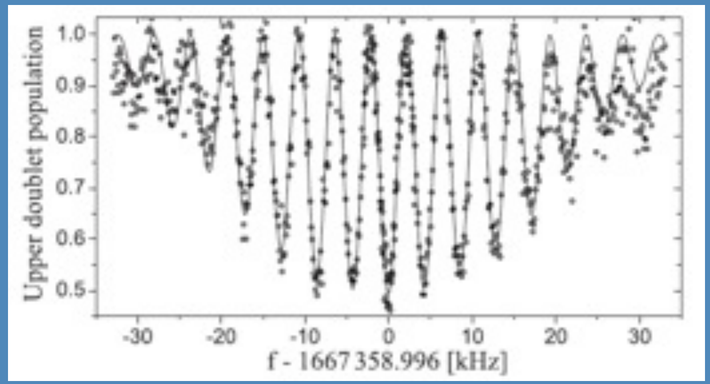


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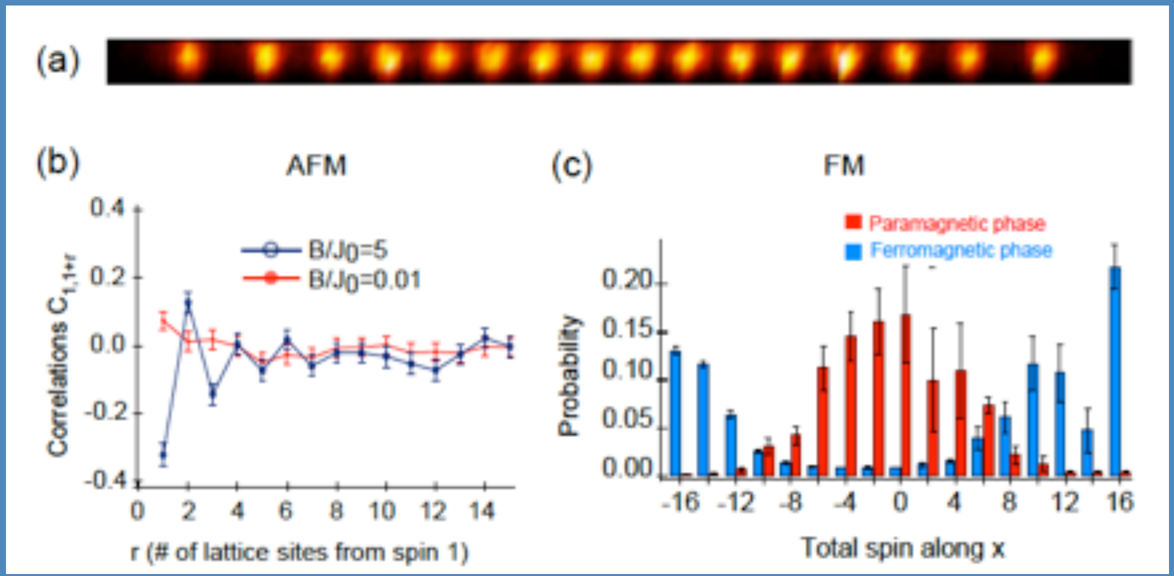
## Simulation of condensed matter systems (w/ internal DOF)




Monroe, JQI

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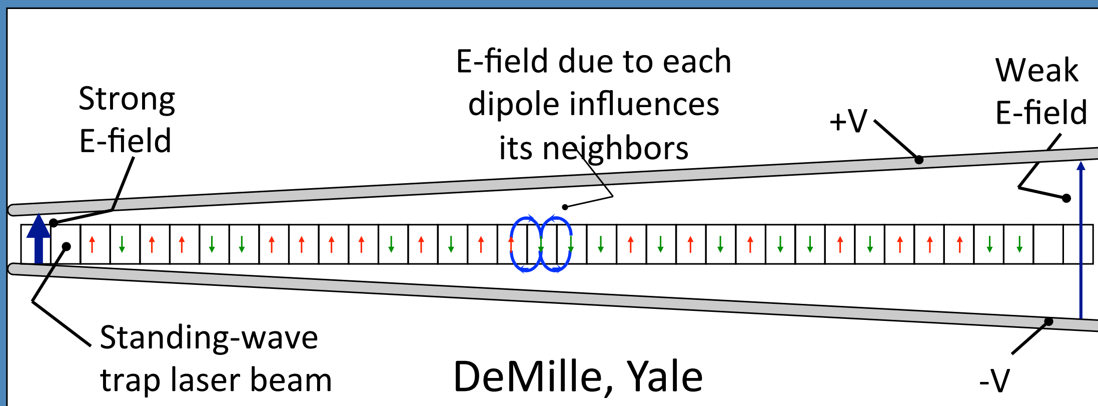


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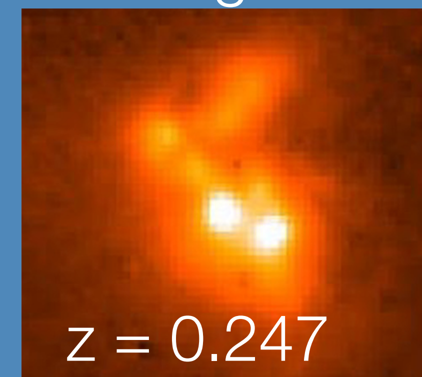


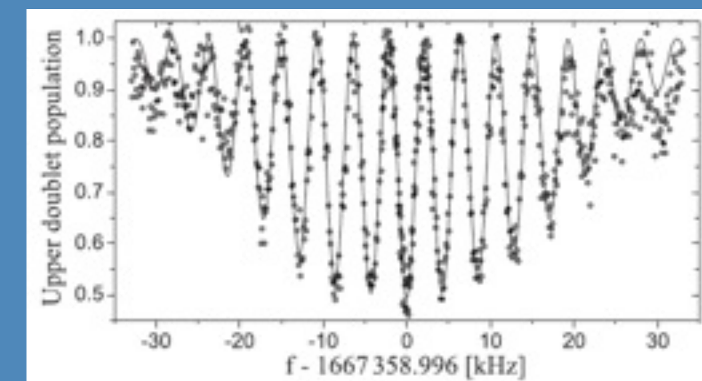


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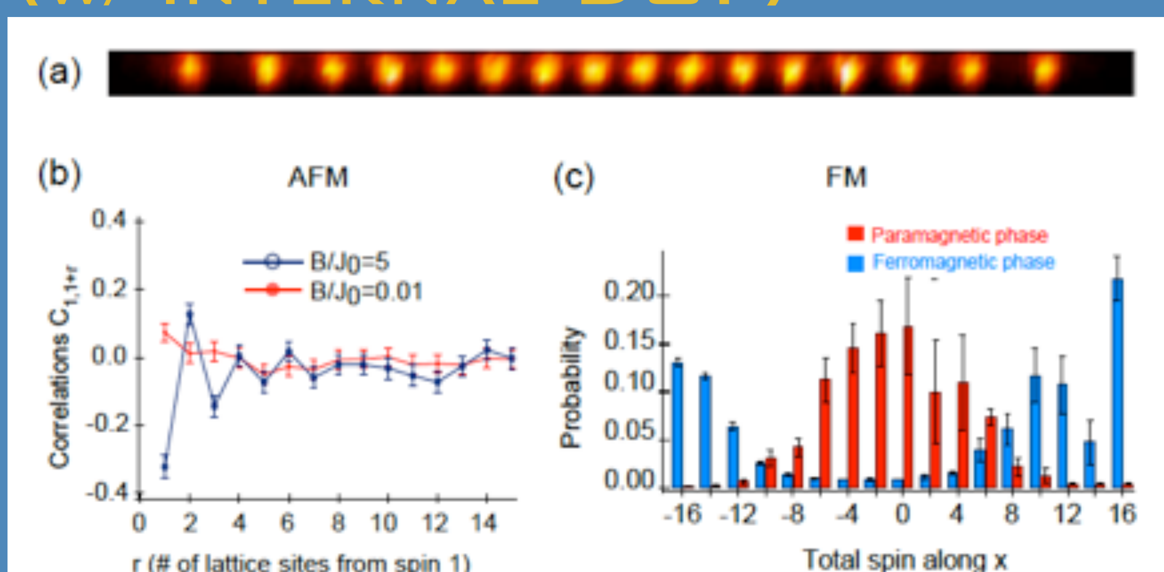


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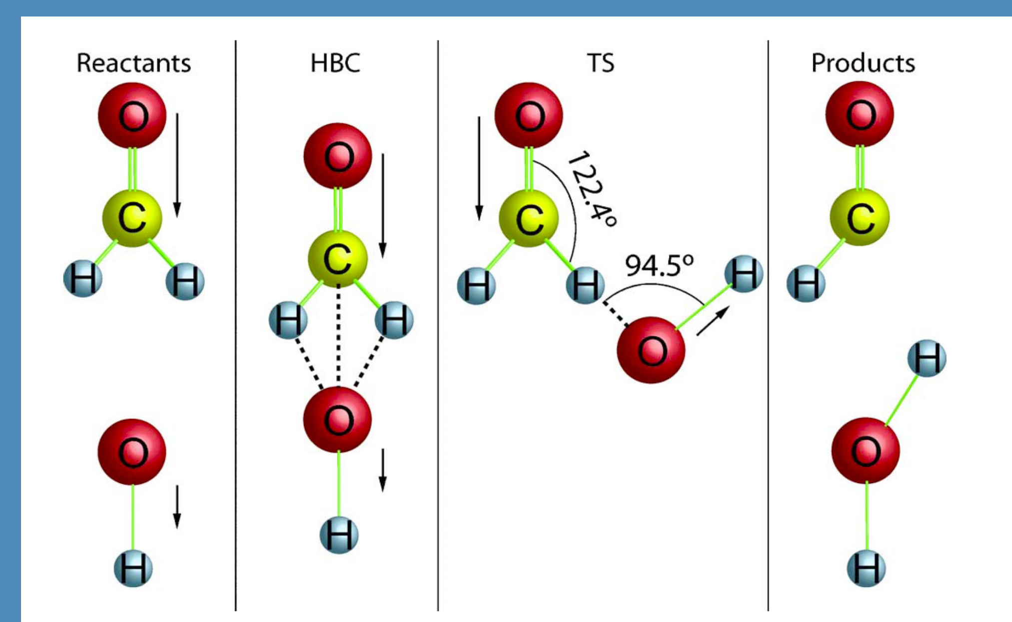


**PRA 73 063404 (2006)**

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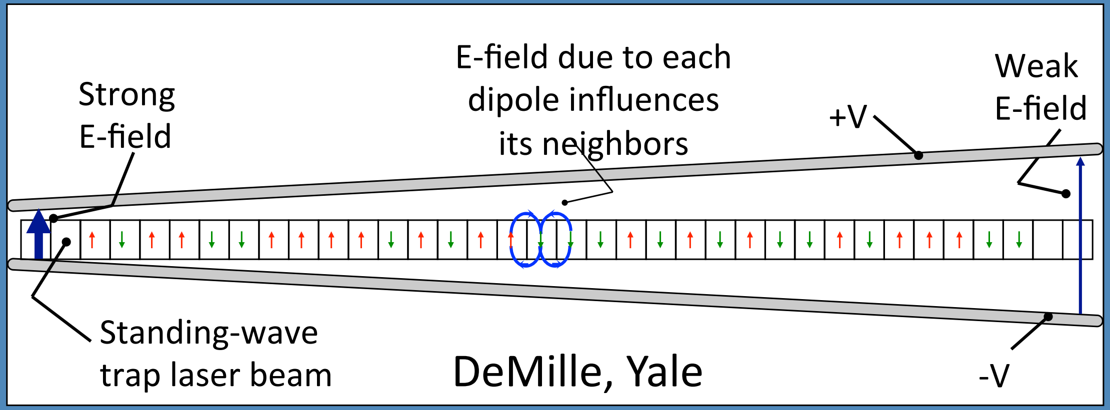


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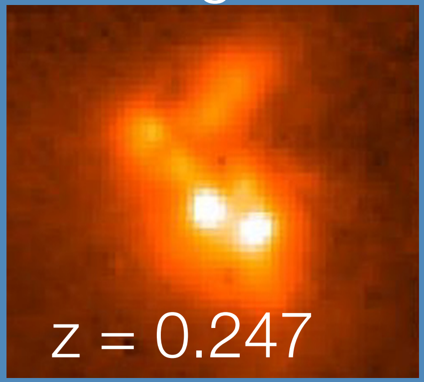


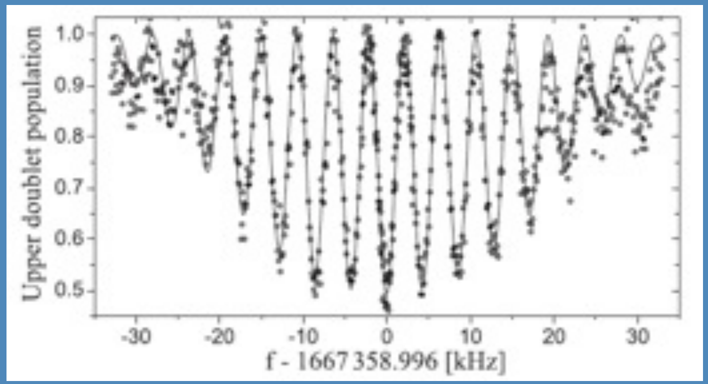


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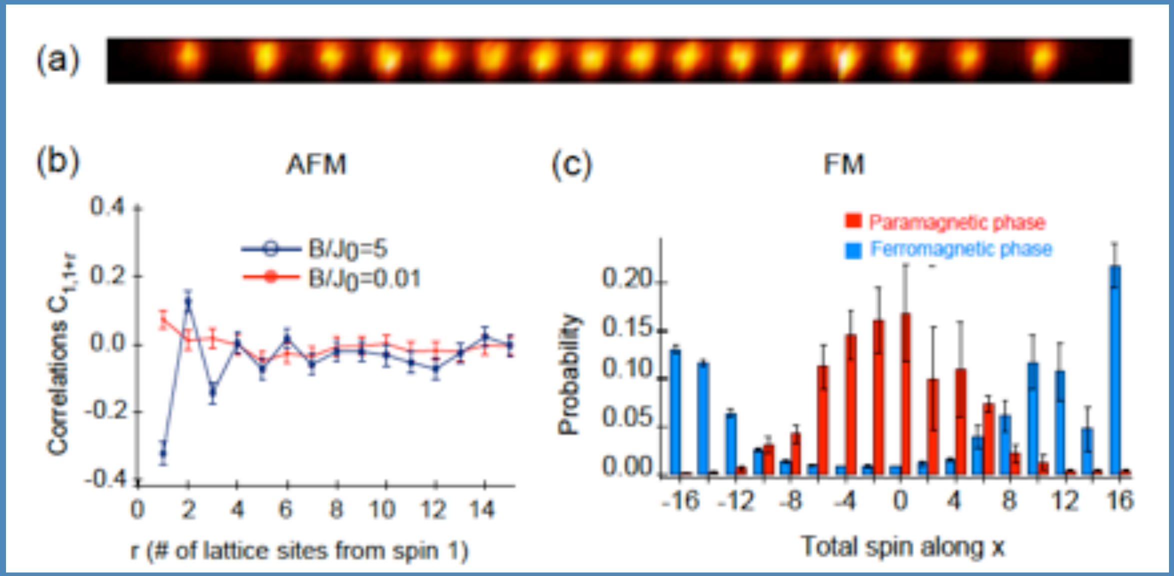


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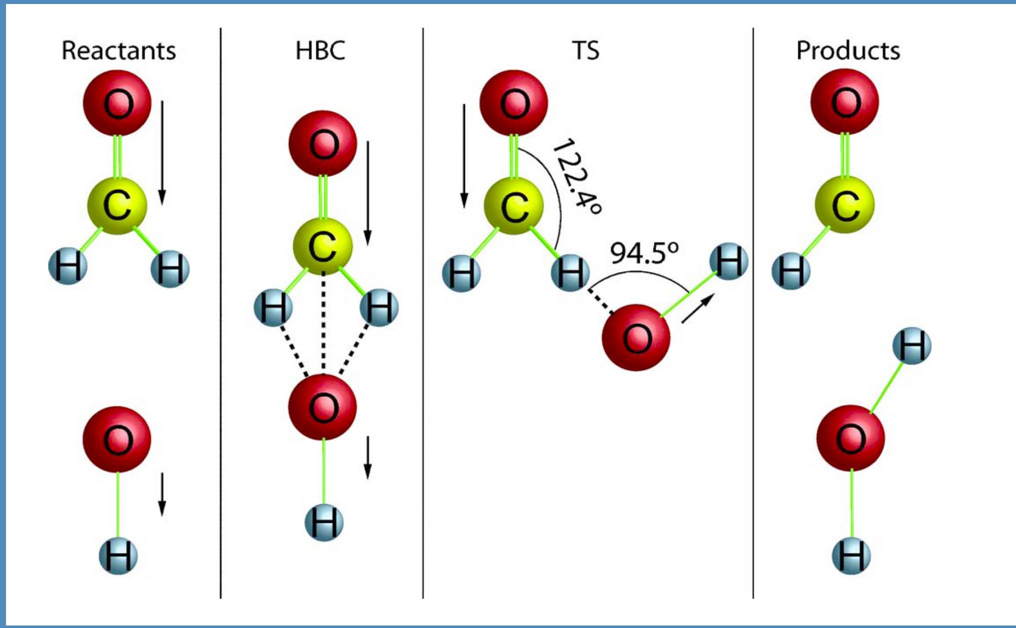
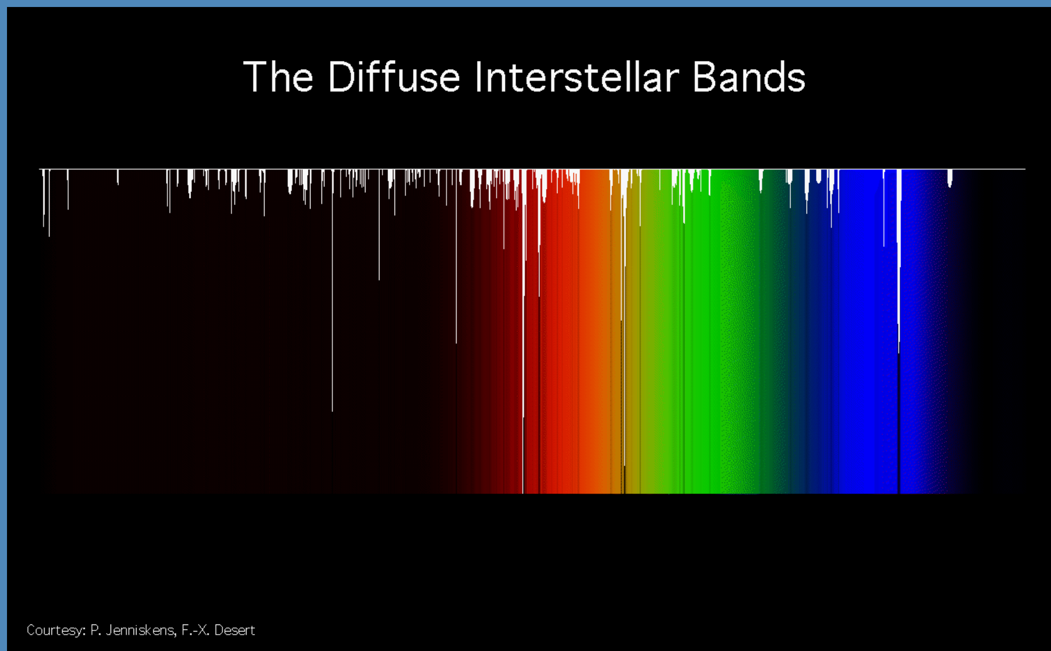



PRA 73 063404 (2006)

Dipolar physics seems harder, but perhaps recoverable.

# • Why use ultracold molecular ions?




Ion-atom chemistry not well-understood:

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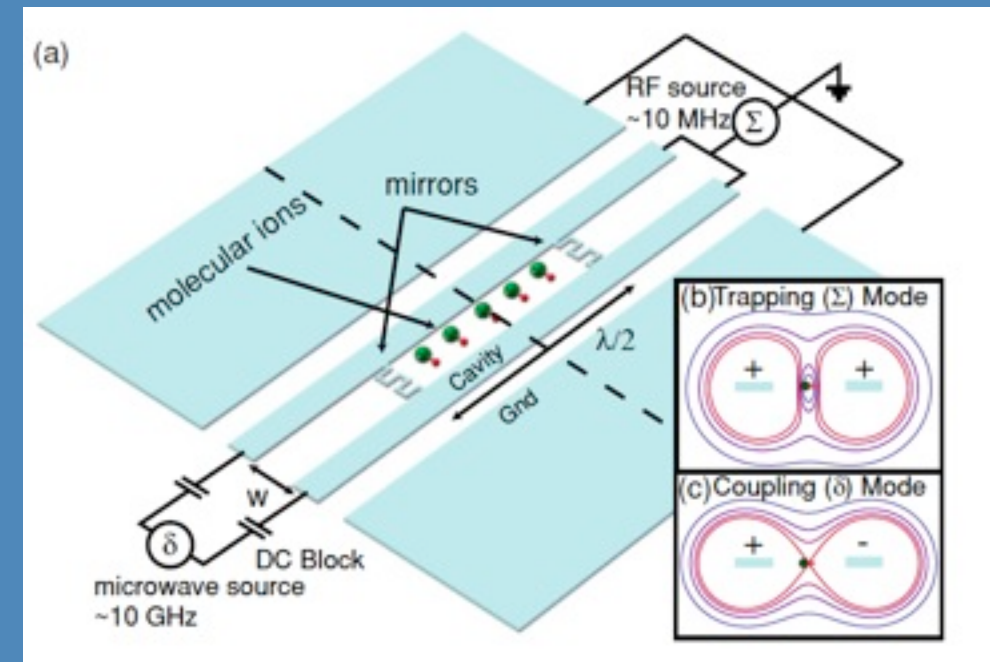


Cavity QED with molecular ions:

D.I. Schuster et al., PRA 83 012311 (2011).

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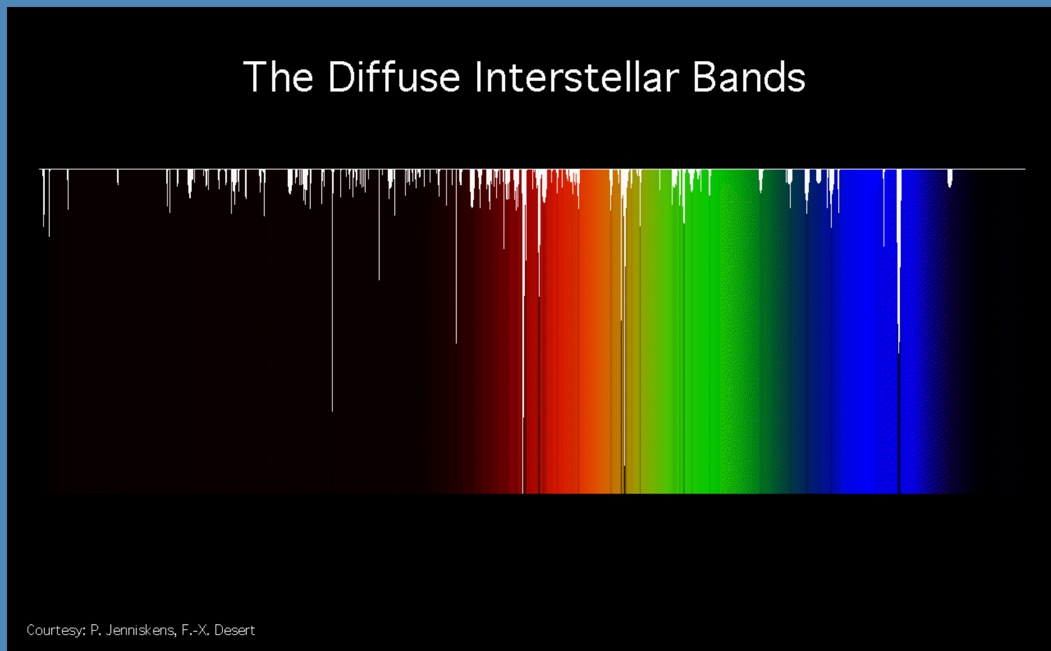


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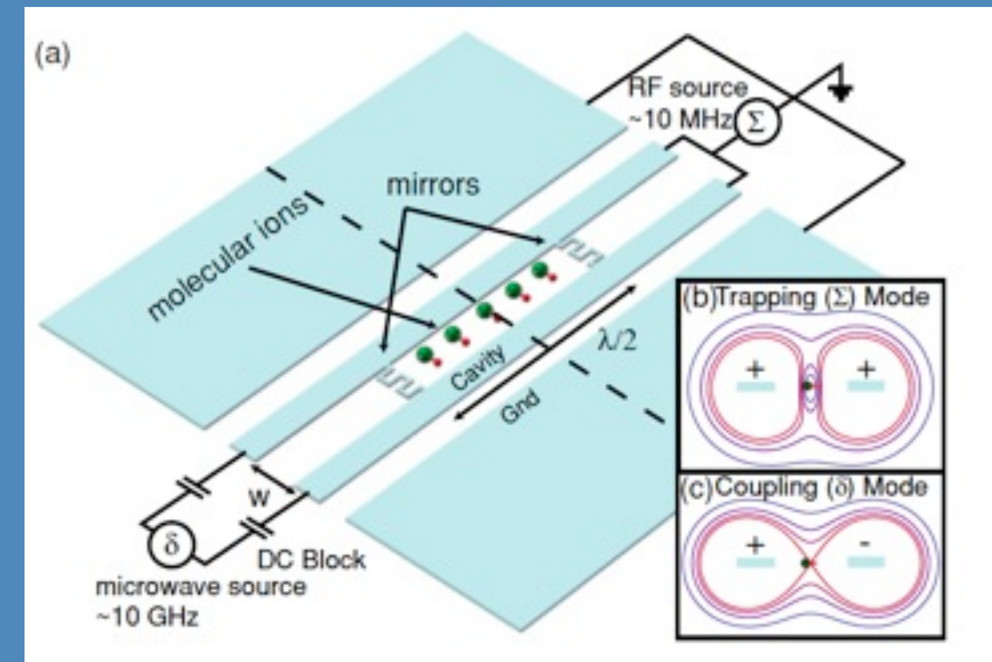


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## Taking a step back

Progress has been hampered because Neutral molecules are difficult to control at room temperature

- Laser and Static/Dynamic E-B fields produce potentials with characteristic depths of ~ 0.01 K

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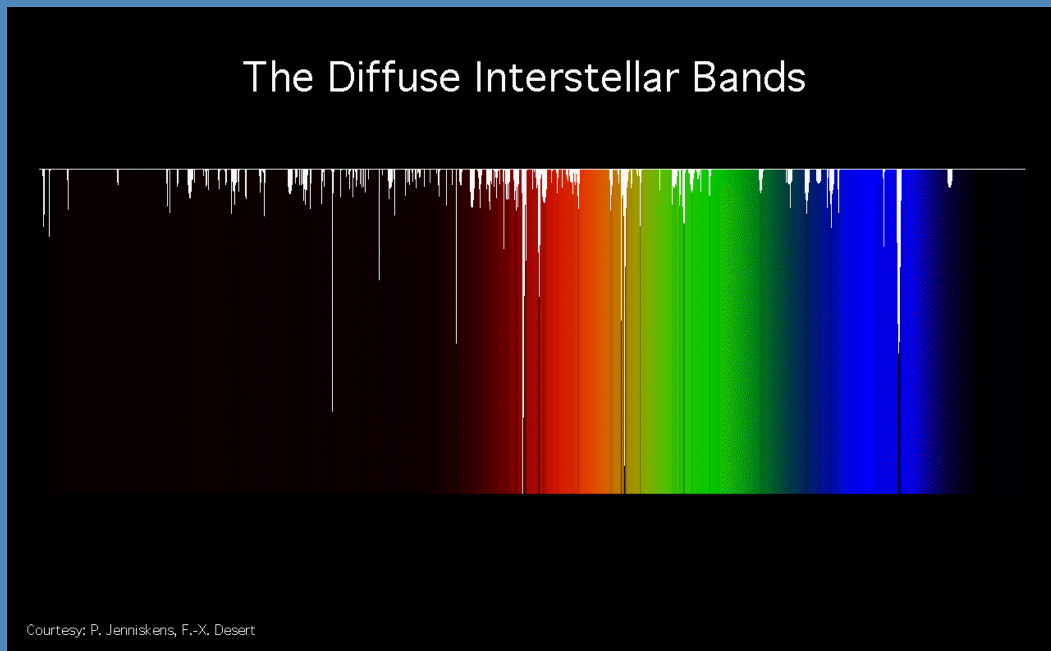


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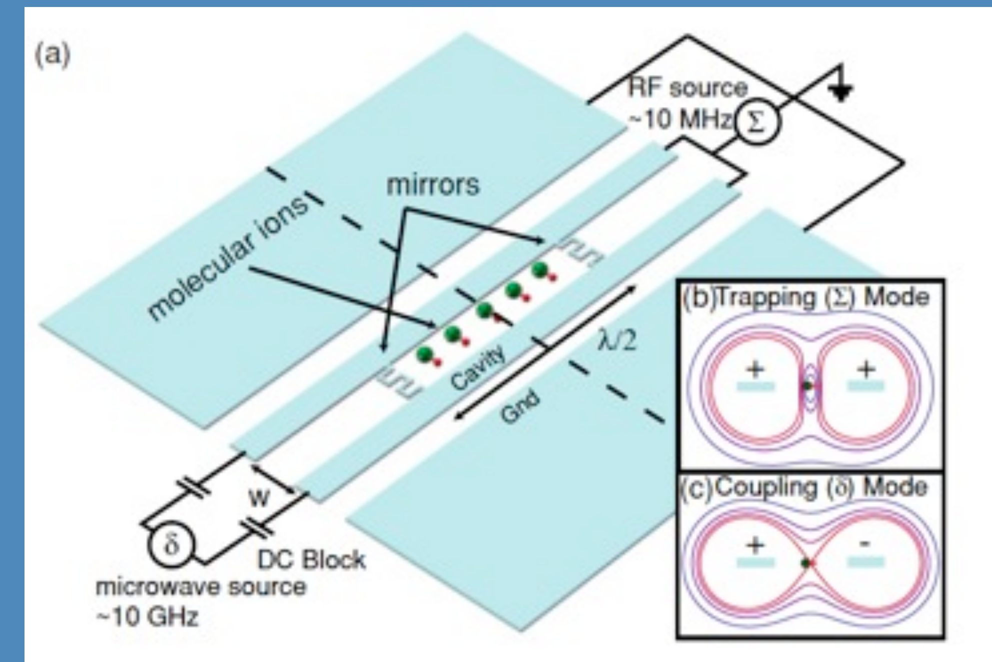


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- Laser and Static/Dynamic E-B fields produce potentials with characteristic depths of ~ 0.01 K

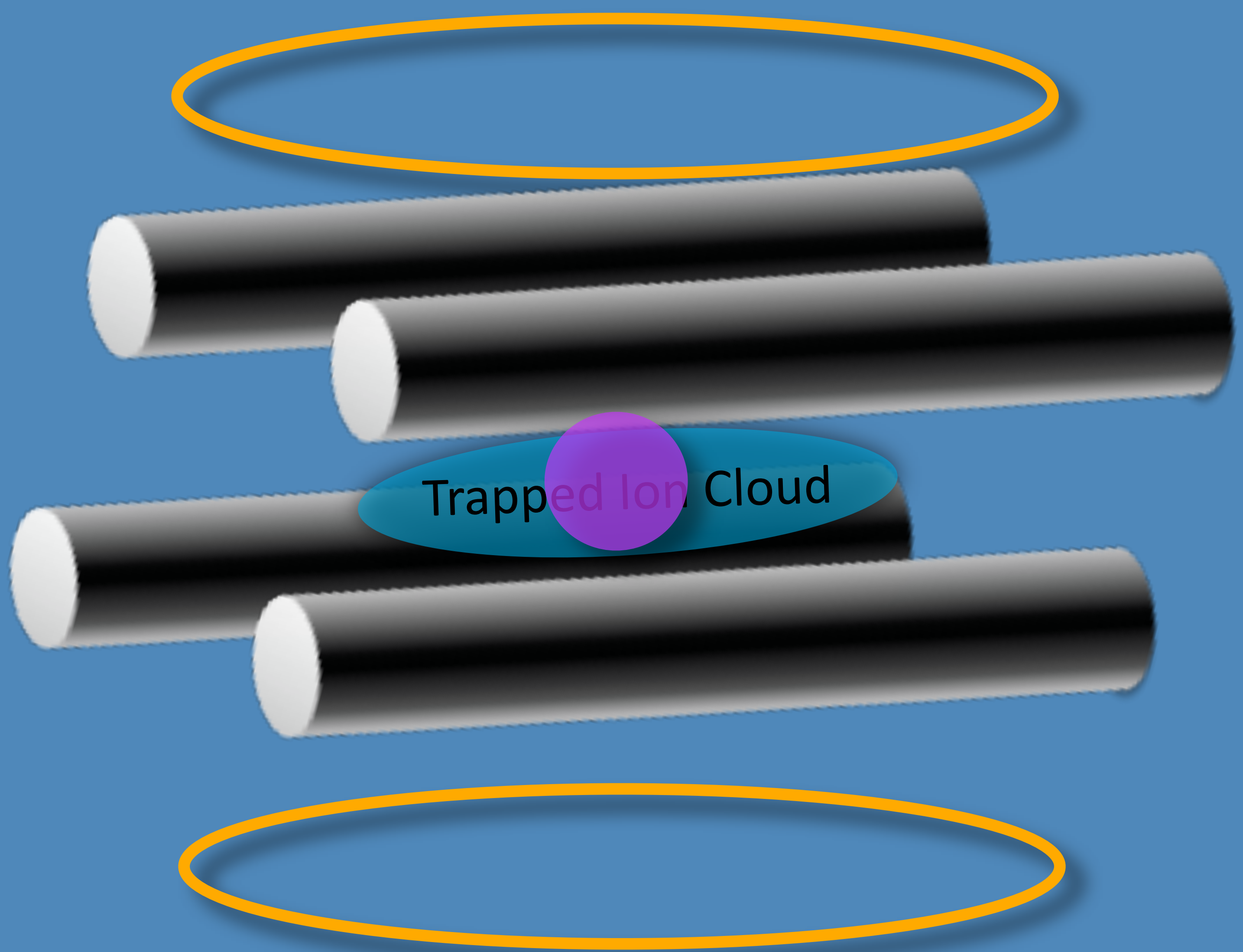
Charged molecules are much easier to control

- Static/Dynamic E-B fields produce potentials with characteristic depths > 10,000 K

# MOTION: Atom-ion quantum chemistry

## Our first surprise: Photo-Associative ionization

MOTION trap



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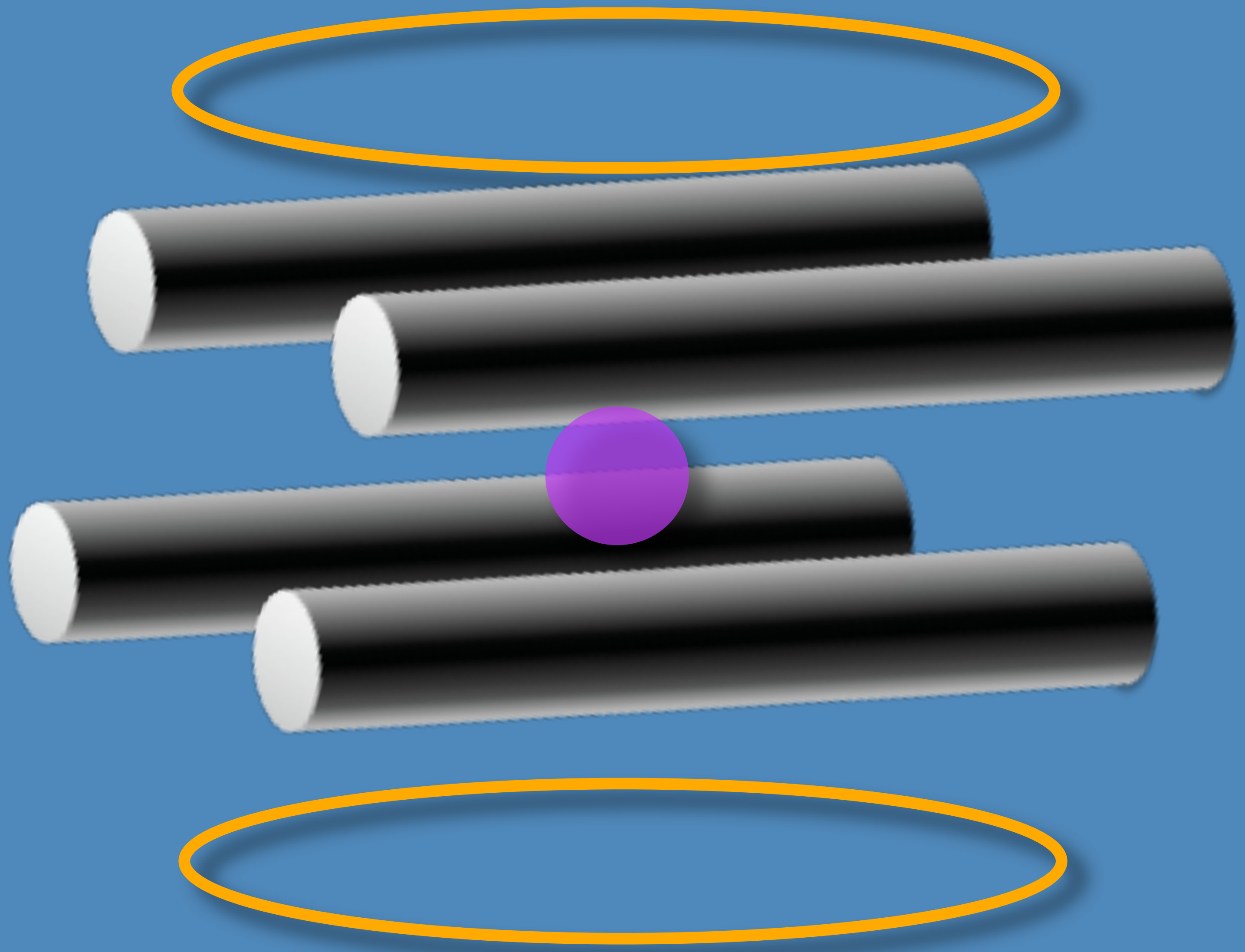
## Our first surprise: Photo-Associative ionization

MOTION trap



# MOTION: Atom-ion quantum chemistry

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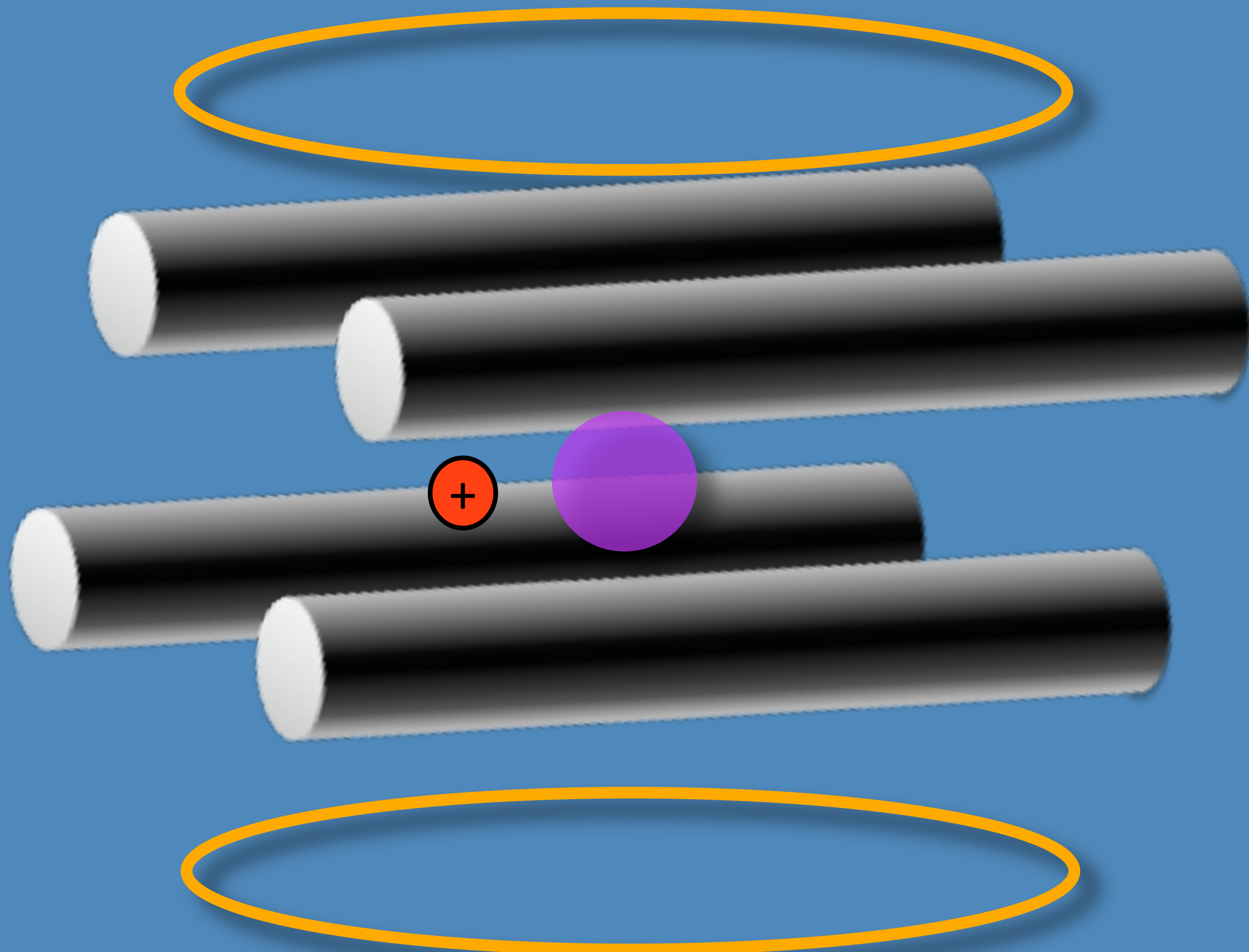

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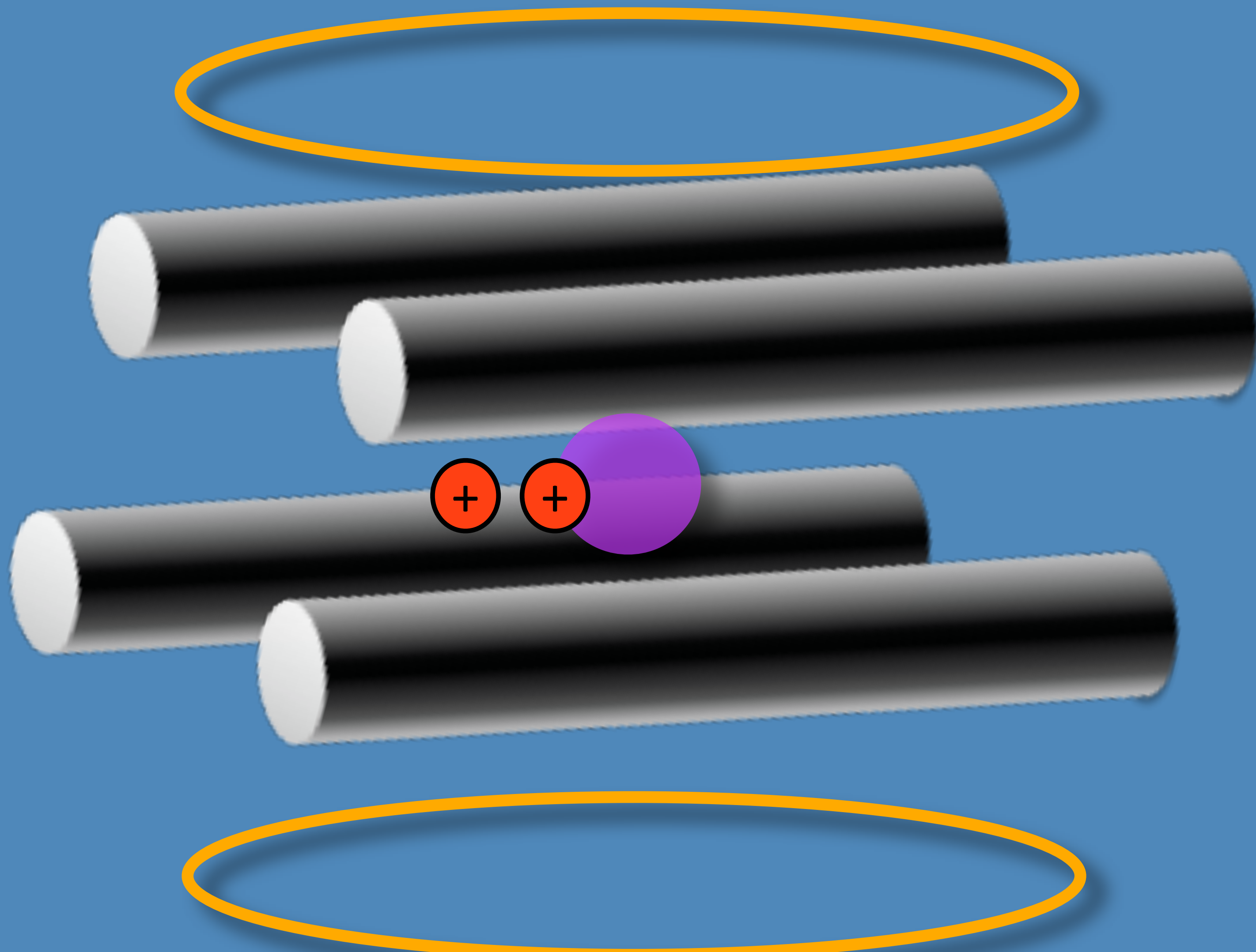

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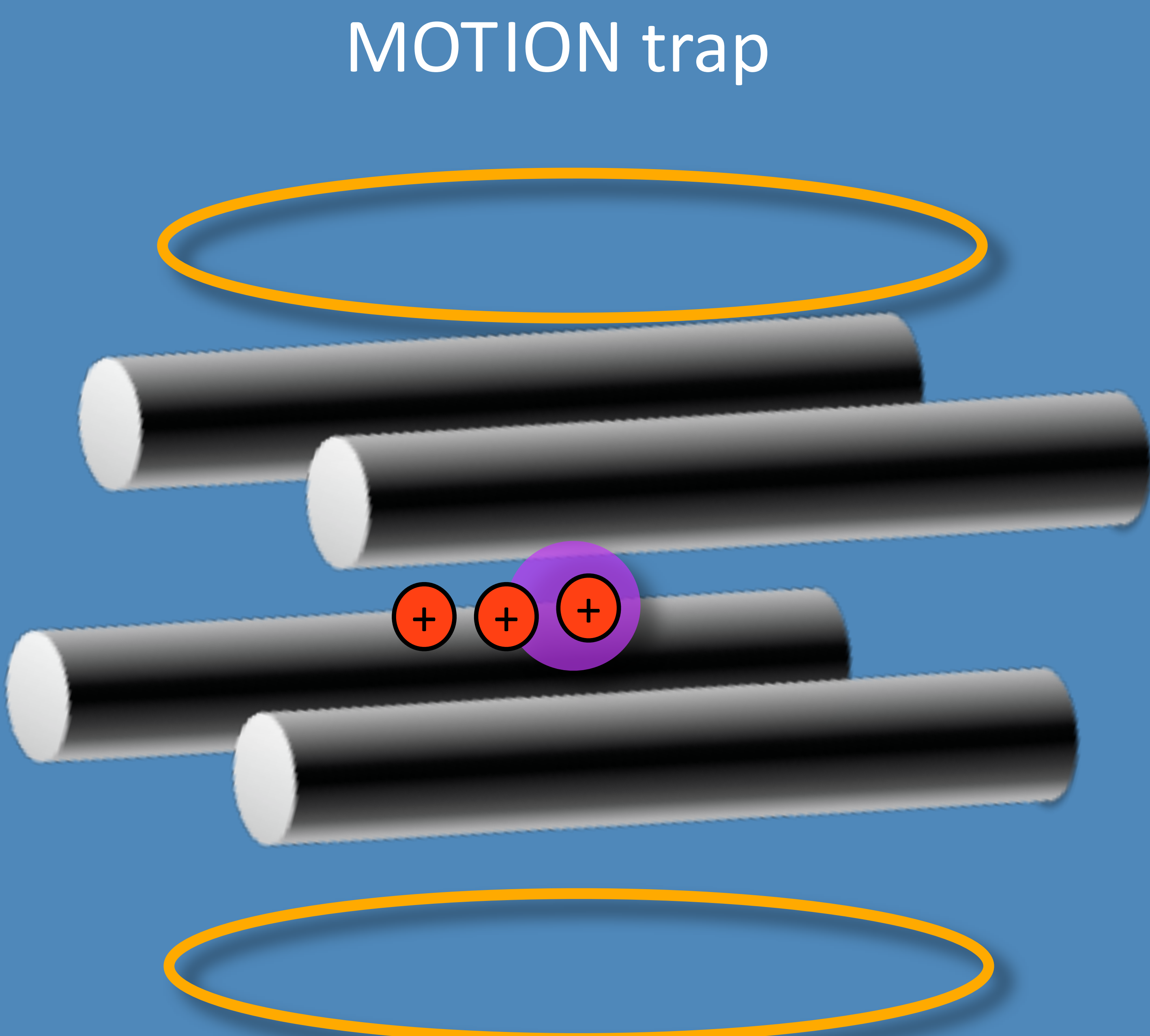

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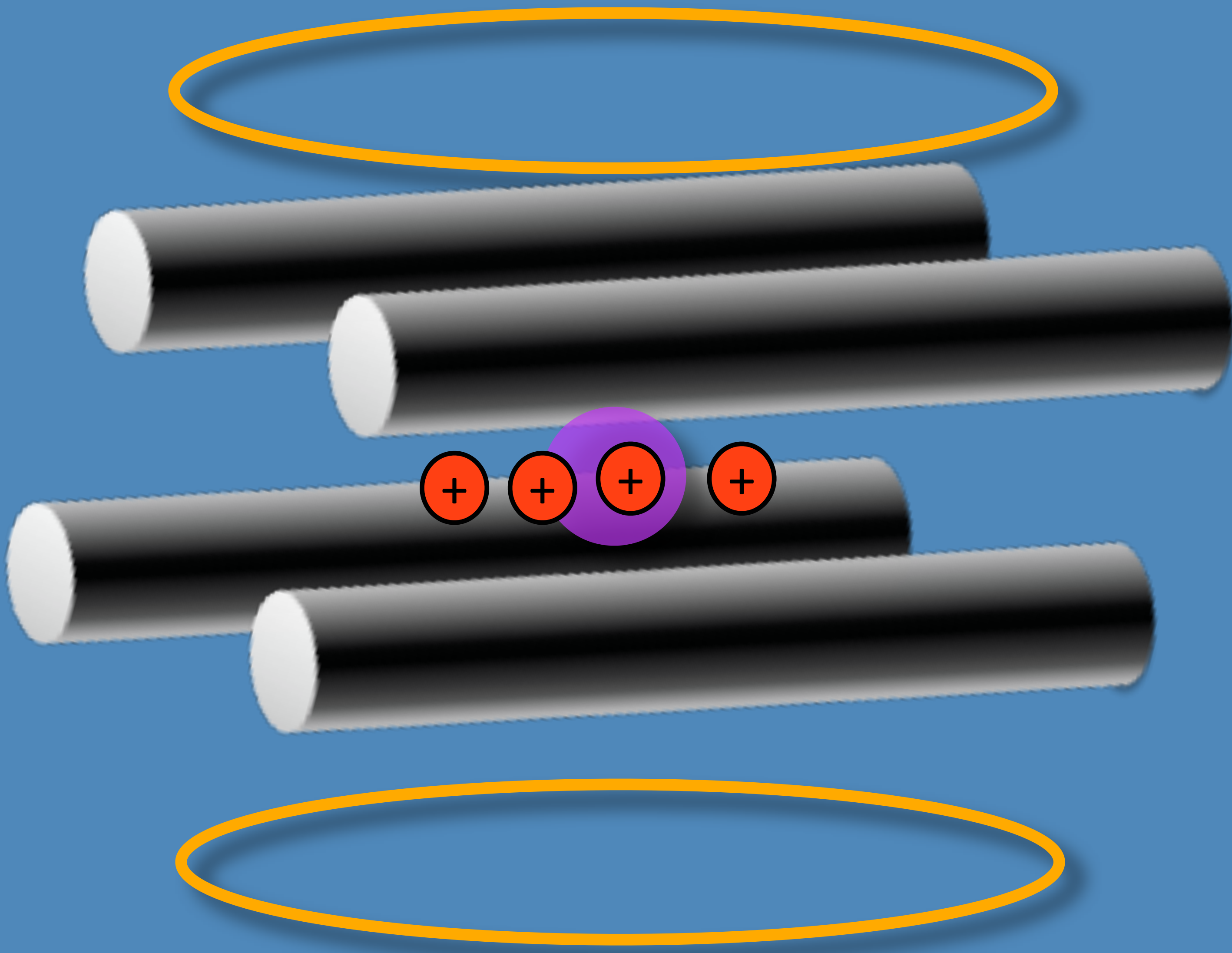

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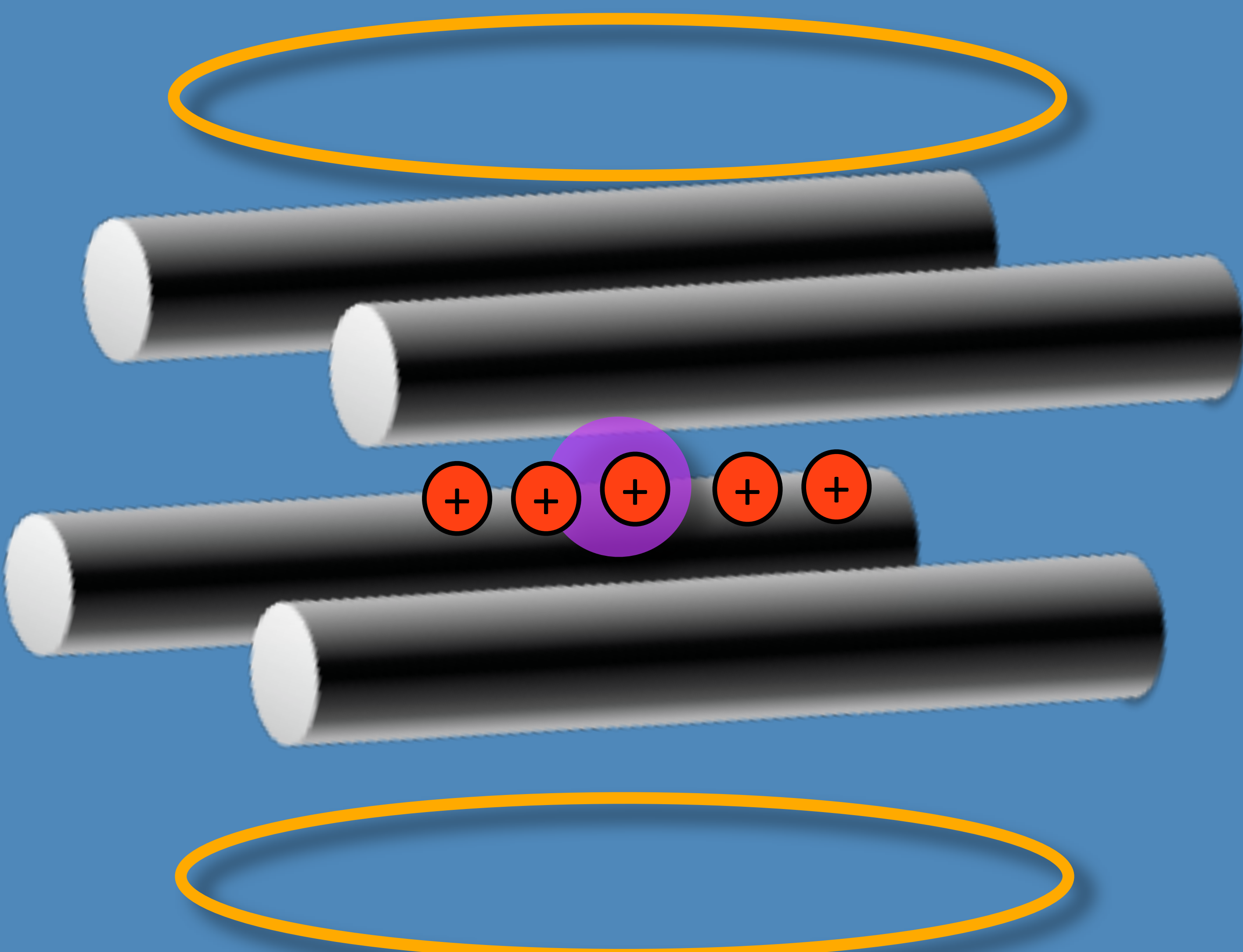
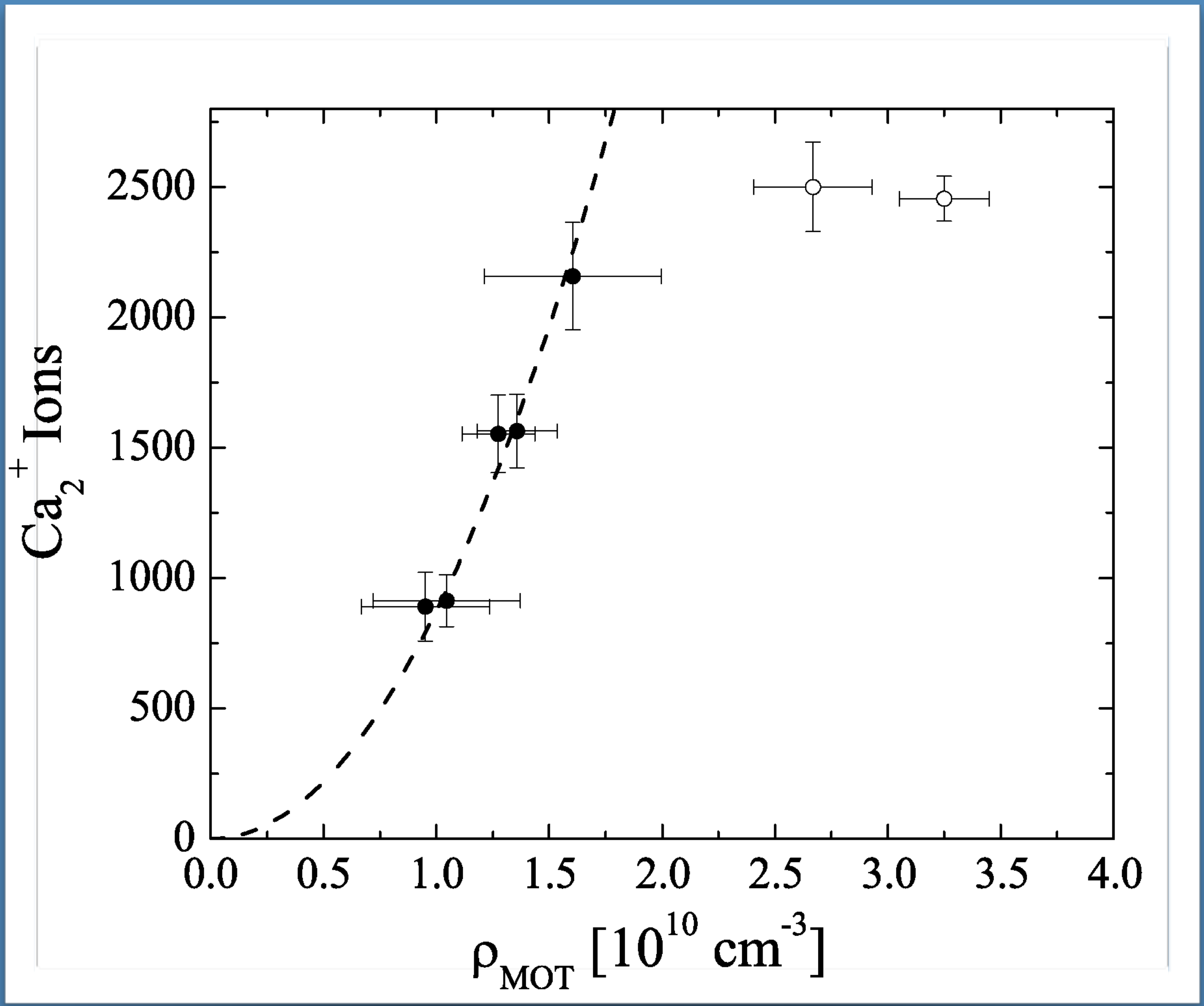


# Some surprising chemistry

## Our first surprise: Photo-Associative ionization

When operating ONLY the MOT we observe an anomalous, large ion signal:

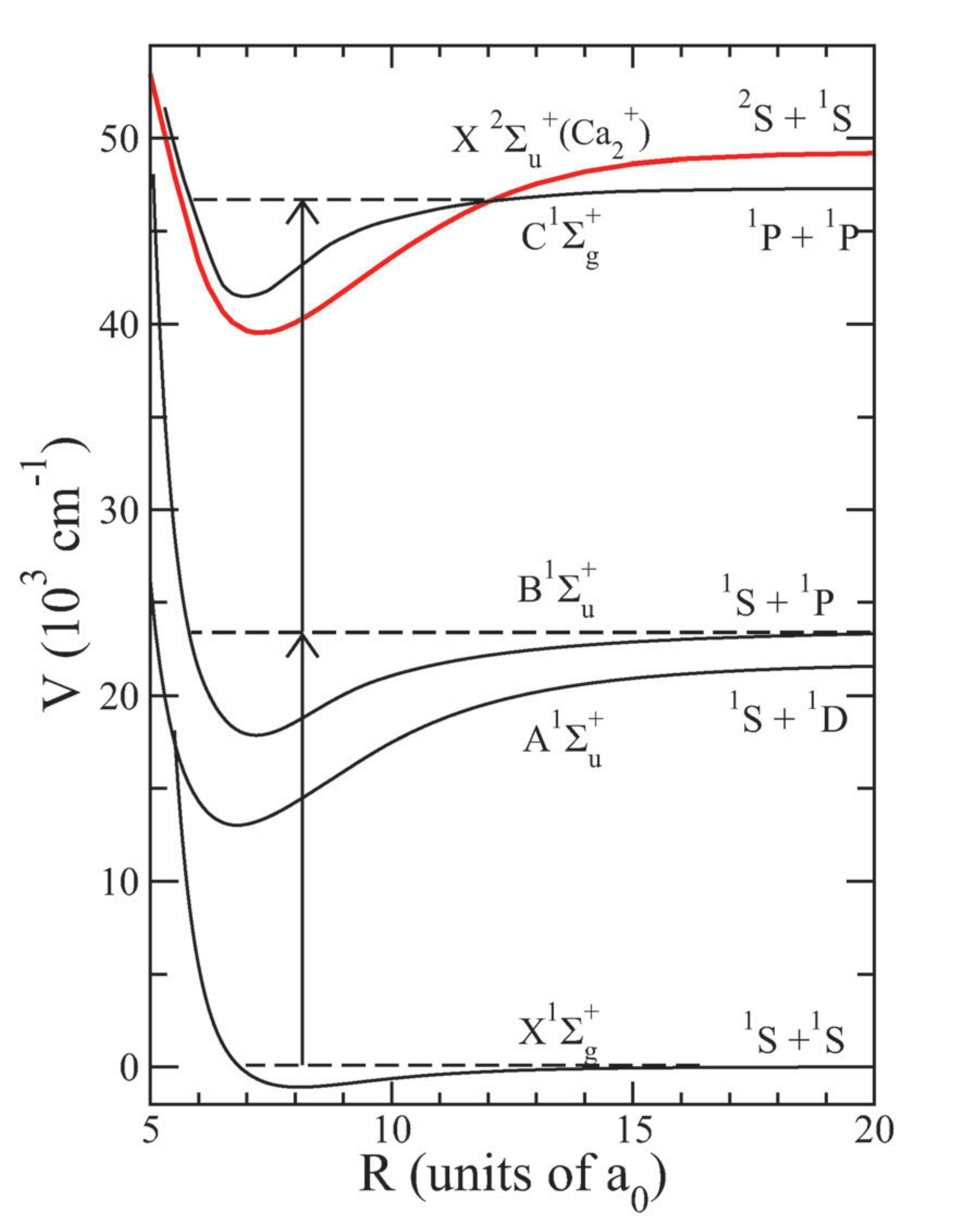



- Secular frequency scans indicate mass = 80 amu
- Production increases as the square of the density of the MOT indicating a two-body process
- Production also appears quadratic in the intensity of the MOT beams indicating a two-photon process

$$\mathrm{Ca} + \mathrm{Ca} + 2\gamma \xrightarrow{??} \mathrm{Ca_2}^{+} + e^{-}$$

# Some surprising chemistry

## Our first surprise: Photo-Associative ionization




$$Ca + Ca^+ \rightarrow Ca_2^+ + \gamma \quad (1)$$

$$Ca(P) + Ca(S) + \gamma \rightarrow Ca_2^+ + e^- \quad (2)$$

$$Ca(P) + Ca(P) \rightarrow Ca_2^+ + e^- \quad (3)$$

$$Ca(D) + Ca(S) + \gamma \rightarrow Ca_2^+ + e^- \quad (4)$$

$$Ca(S) + Ca(S) + \gamma \rightarrow Ca_2^* + \gamma \rightarrow Ca_2^+ + e^- \quad (5)$$

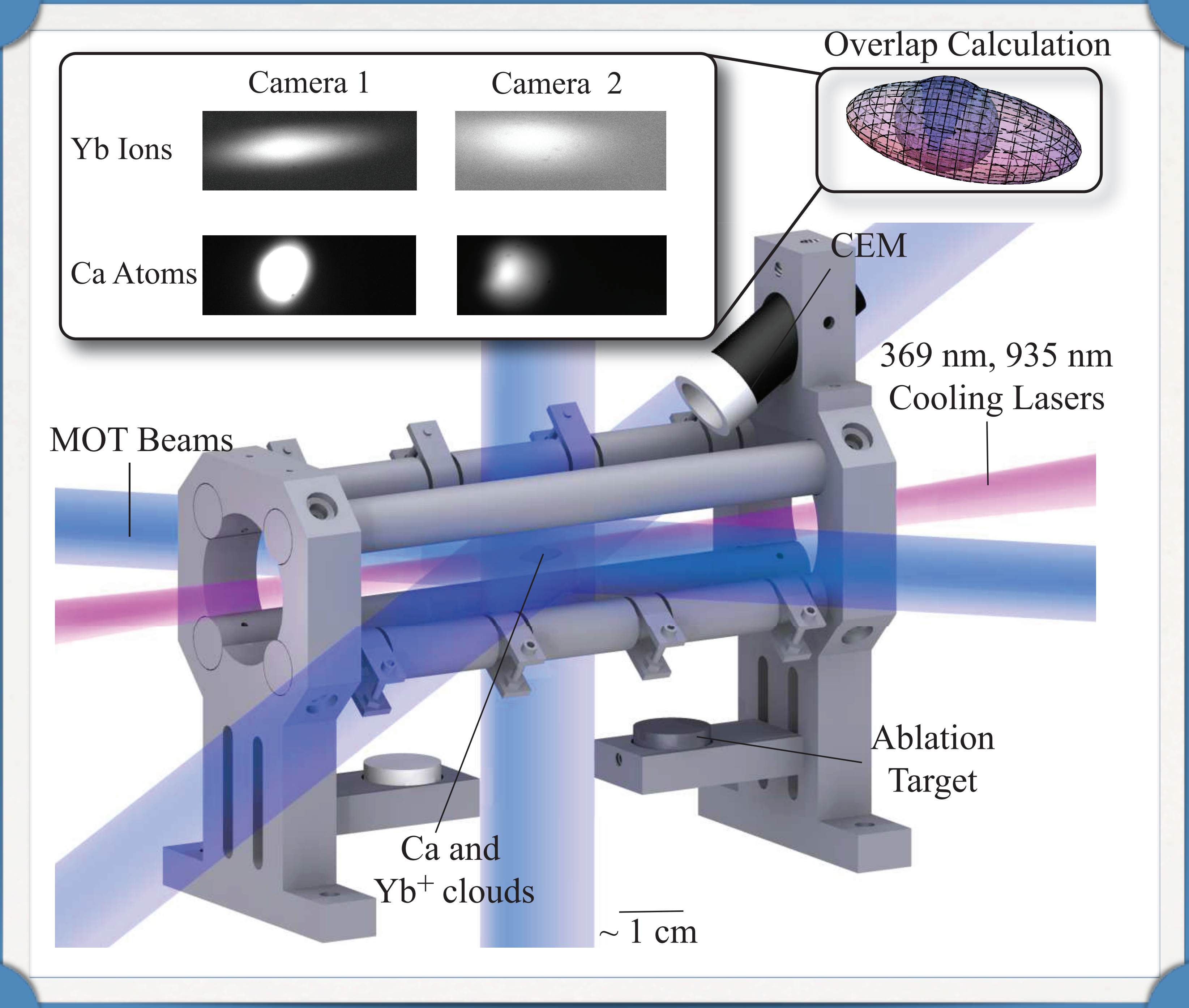
$$\beta \geq 2 \pm 1 \times 10^{-15} cm^3 s^{-1}$$

Similar results observed in Na -- Lett & Gould Julienne

In collaboration with Prof. Svetlana Kotochigova of Temple University

S.T. Sullivan et al., Phys. Chem. Chem. Phys., 2011, DOI: 10.1039/C1CP21205B

# • Our first implementation of MOTION trap



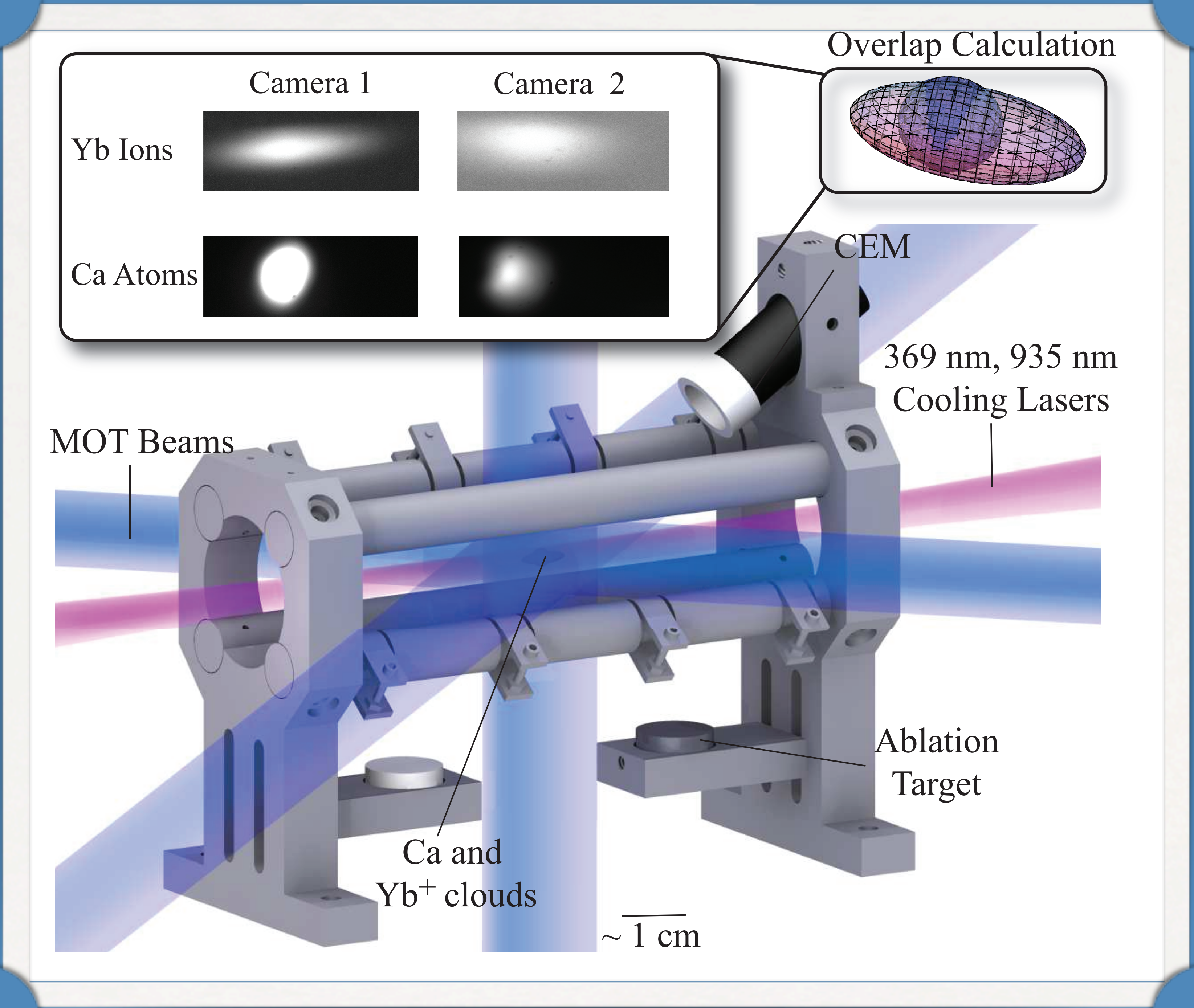

Imaging laser cooled $Yb^{+}$ ions

+

Imaging Ca MOT

Precise Overlap and characterization

# • Our first implementation of MOTION trap

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↓

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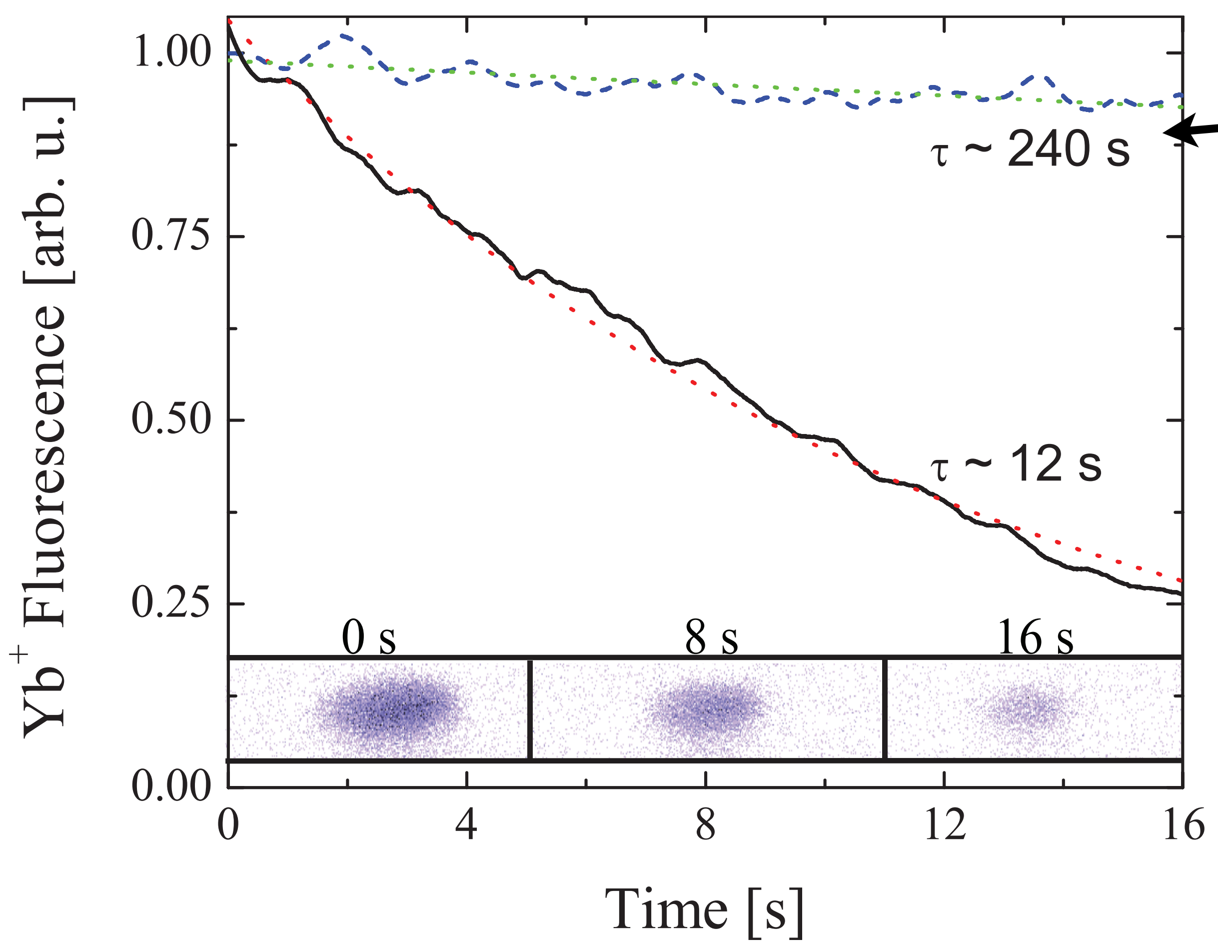
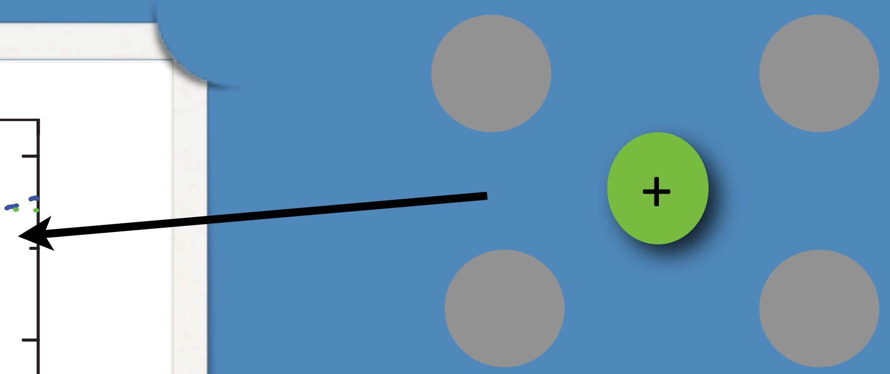
CHEMISTRY

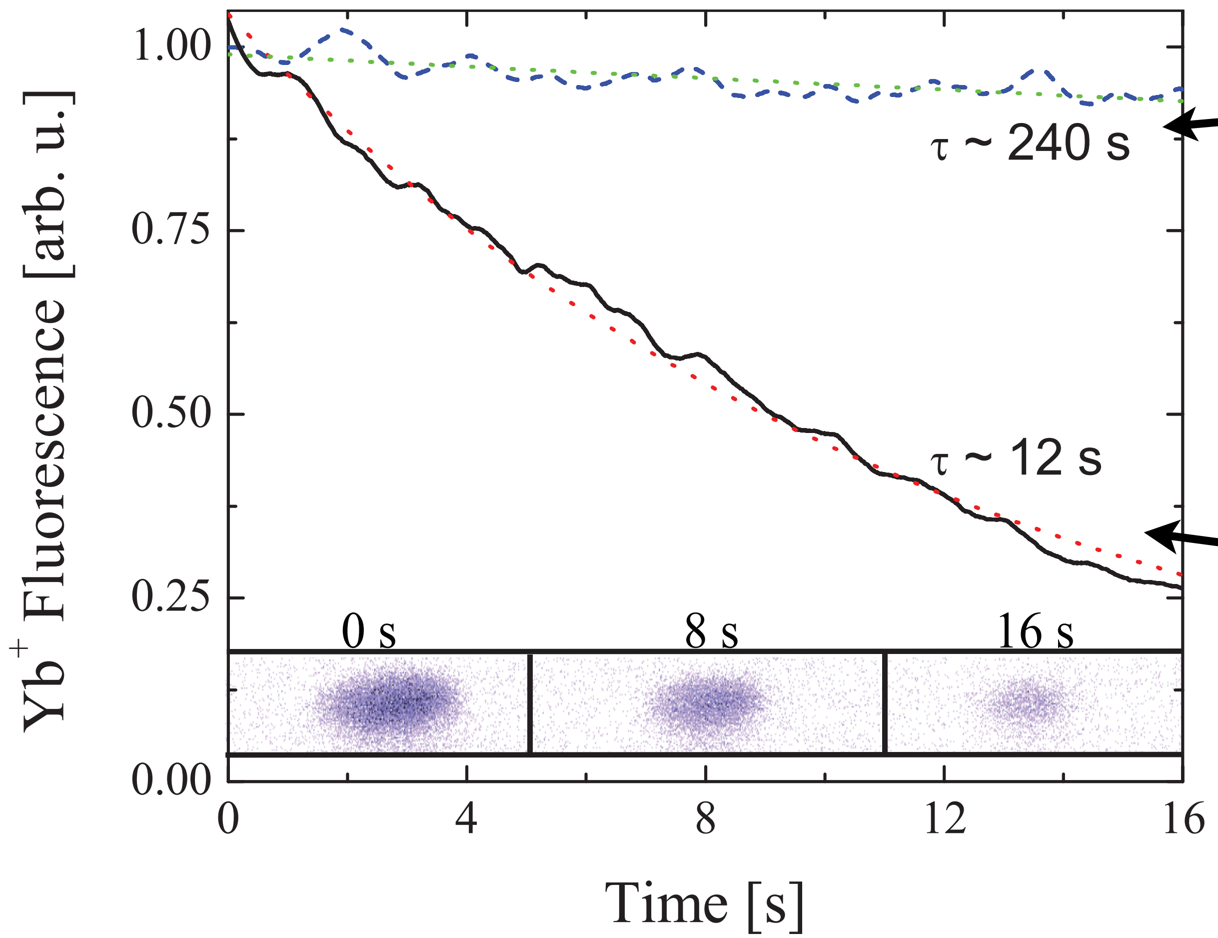
# • Some surprising chemistry

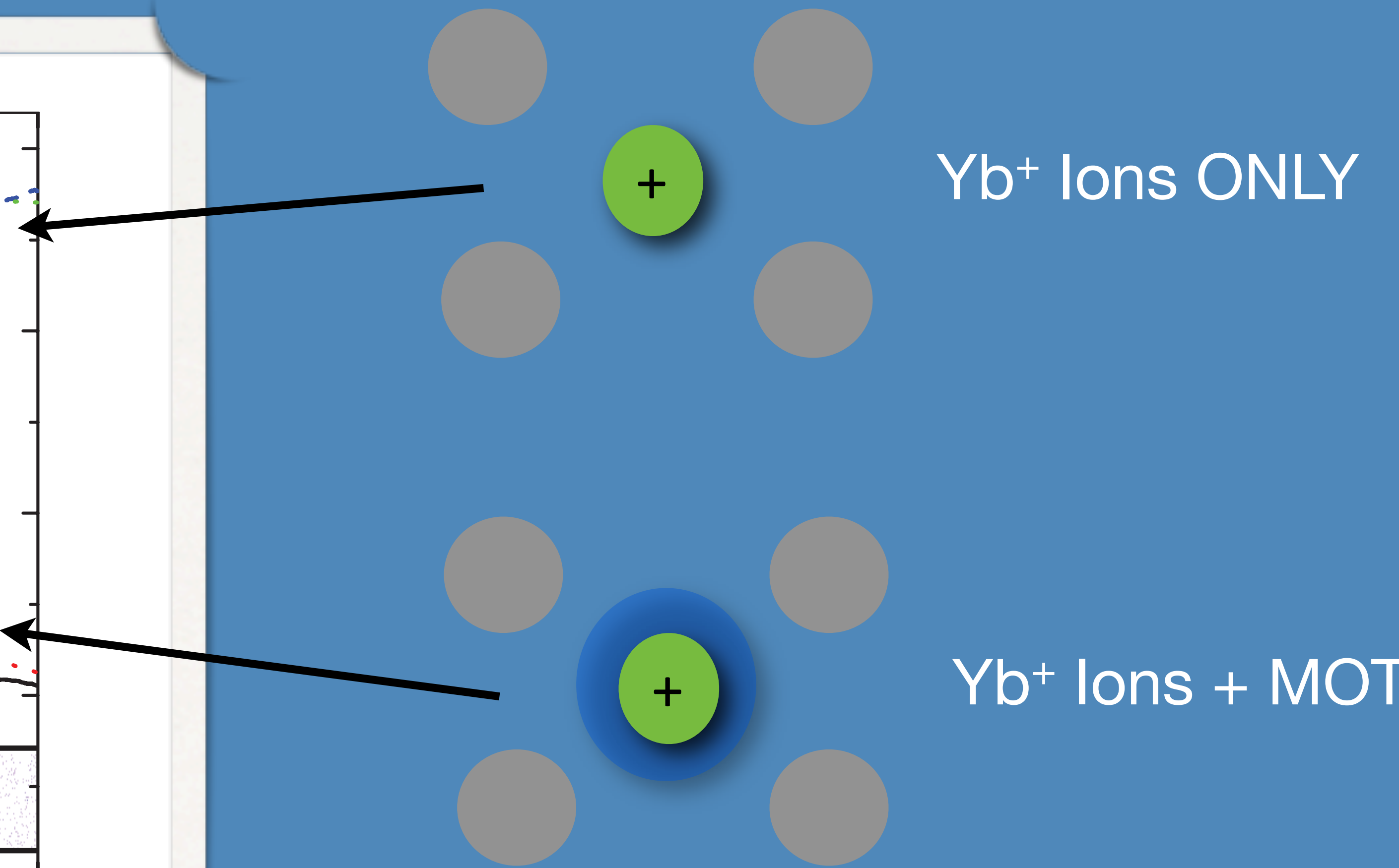



Yb⁺ Ions ONLY

# • Some surprising chemistry



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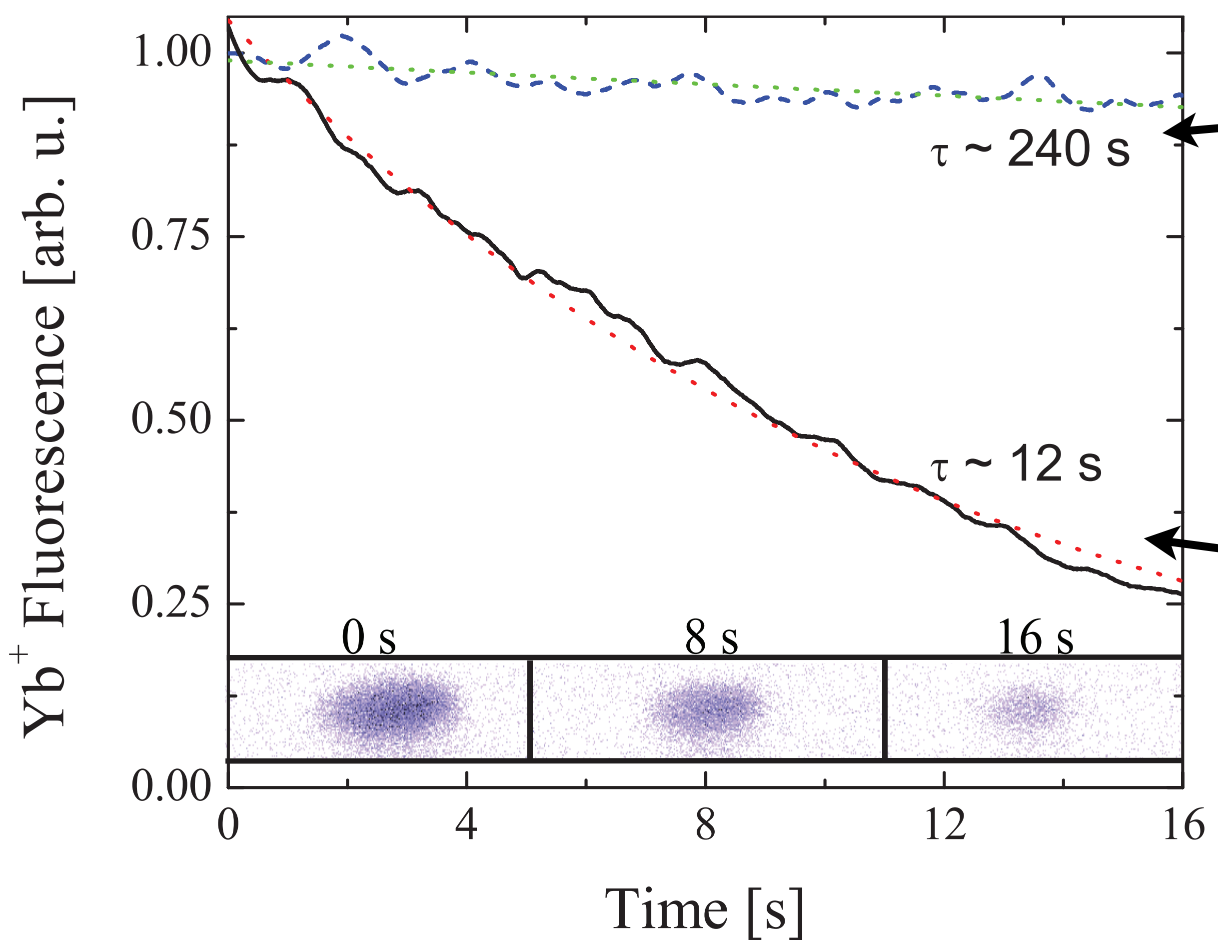


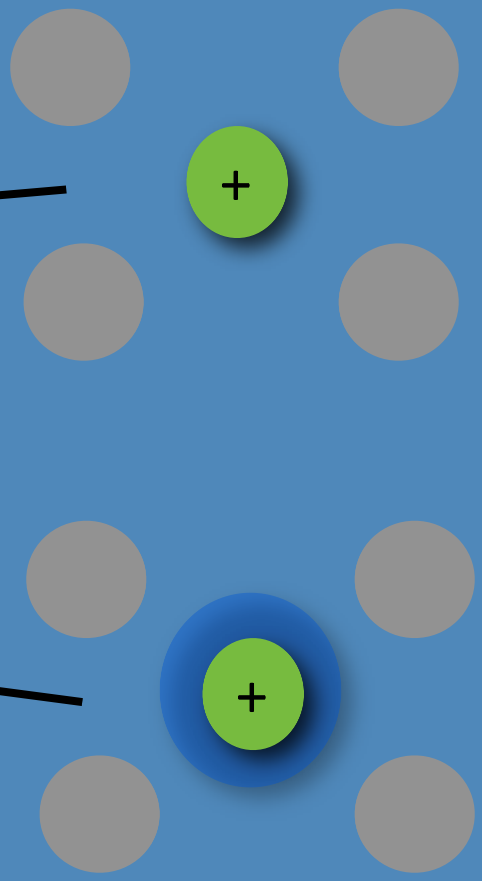


IP(Ca) = 6.1 eV
IP(Yb) = 6.3 eV

→ Charge-exchange chemical rxn?

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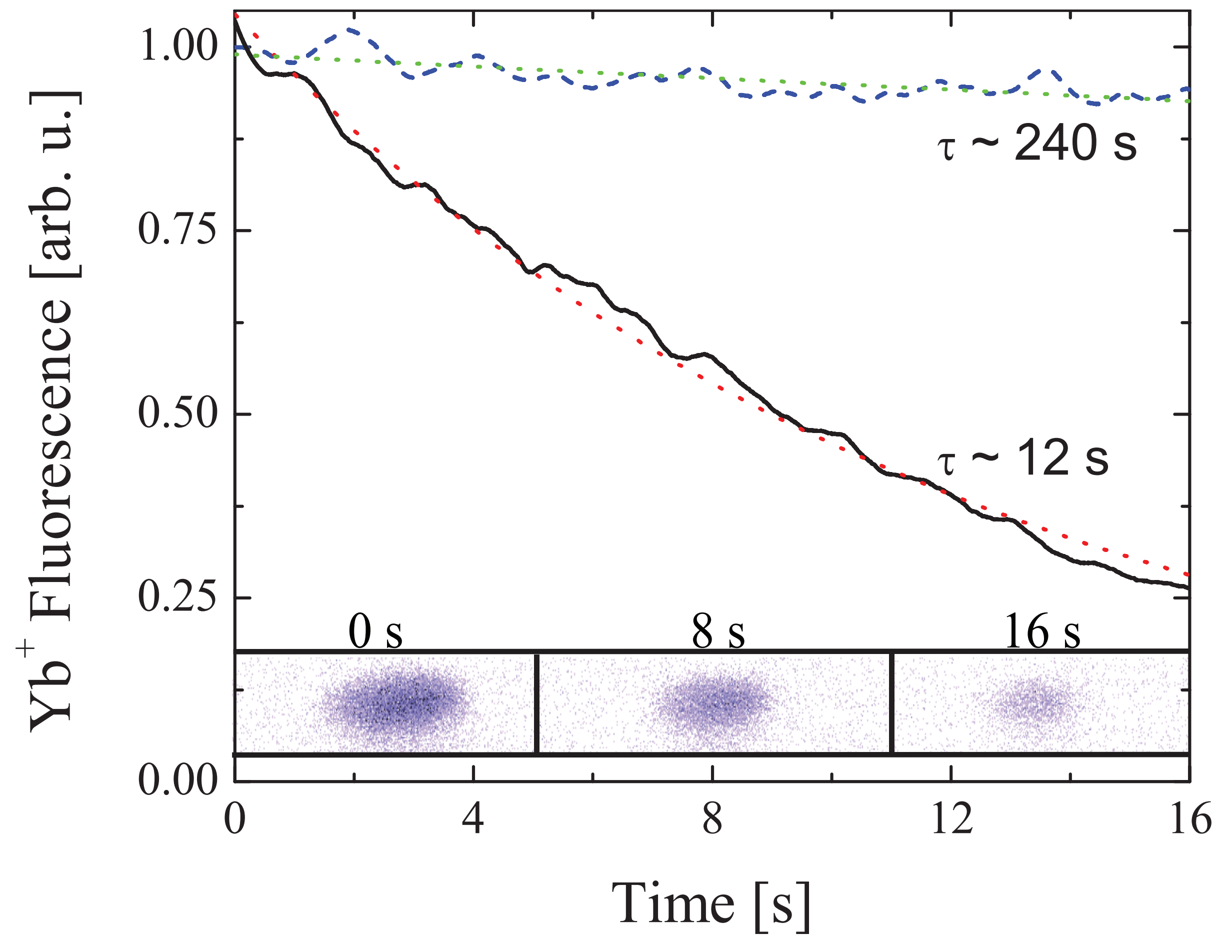


$$1/\tau = \rho \langle \sigma v \rangle = \rho K$$

$$K \approx \frac{1}{12\ \mathrm{s} \cdot 5 \times 10^9\ \mathrm{cm}^{-3}} = 10^{-10} \mathrm{cm}^3 \mathrm{s}^{-1}$$

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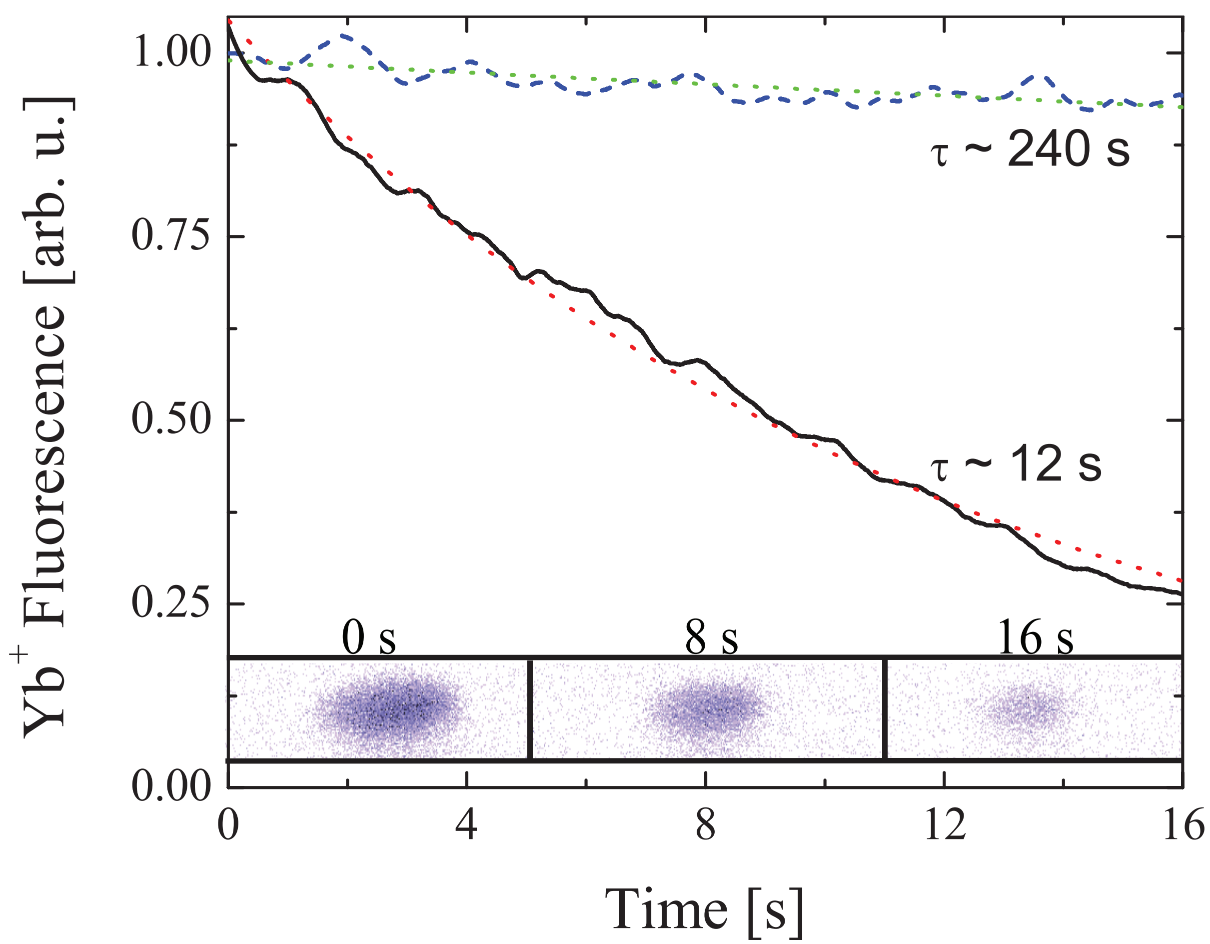


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**$Yb^+$ + Rb ~ 3.5 x $10^{-14}$ $cm^3s^{-1}$**
[C. Zipkes et al., Phys. Rev. Lett. 105, 133201 (2010).]

**$Ba^+$ + Rb ~ Similar rate**
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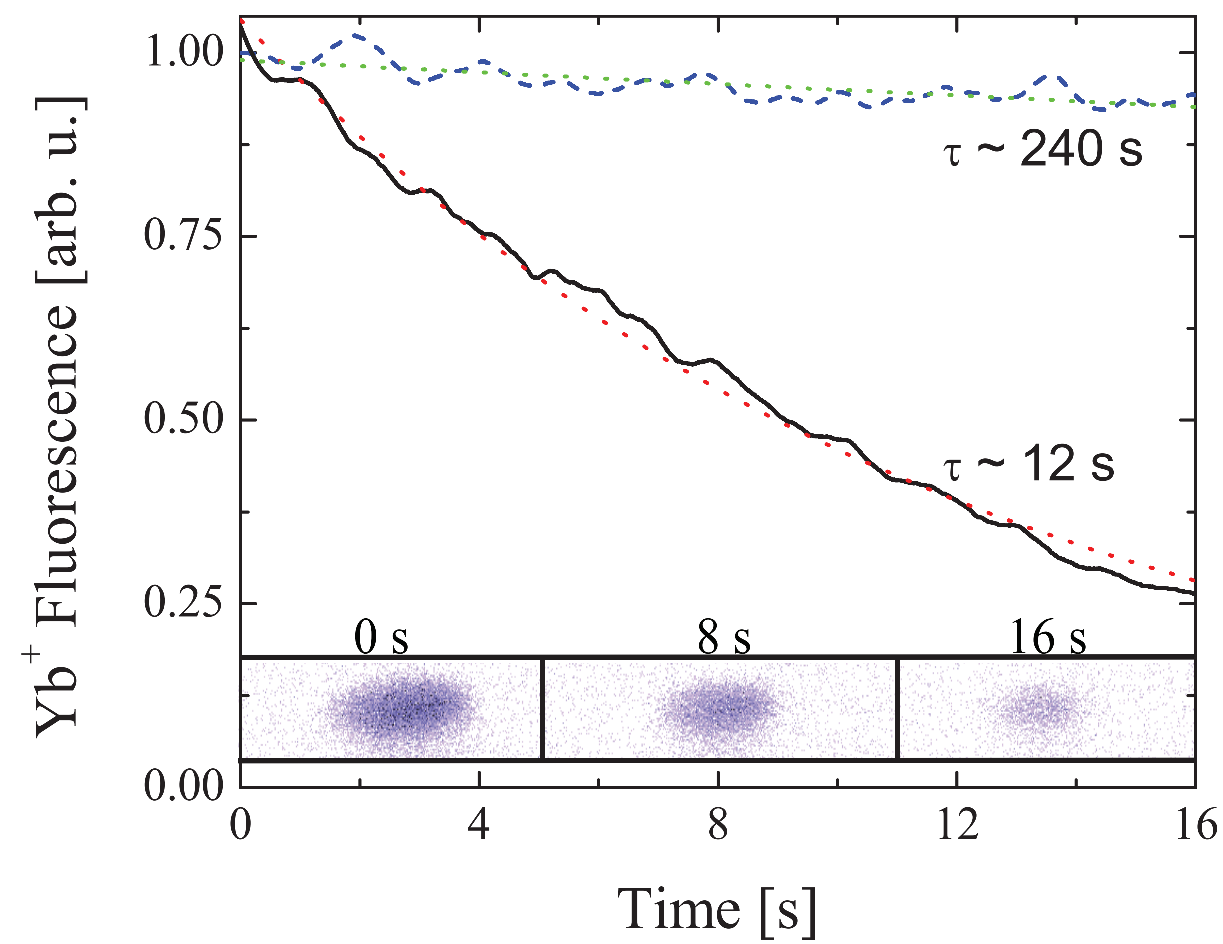


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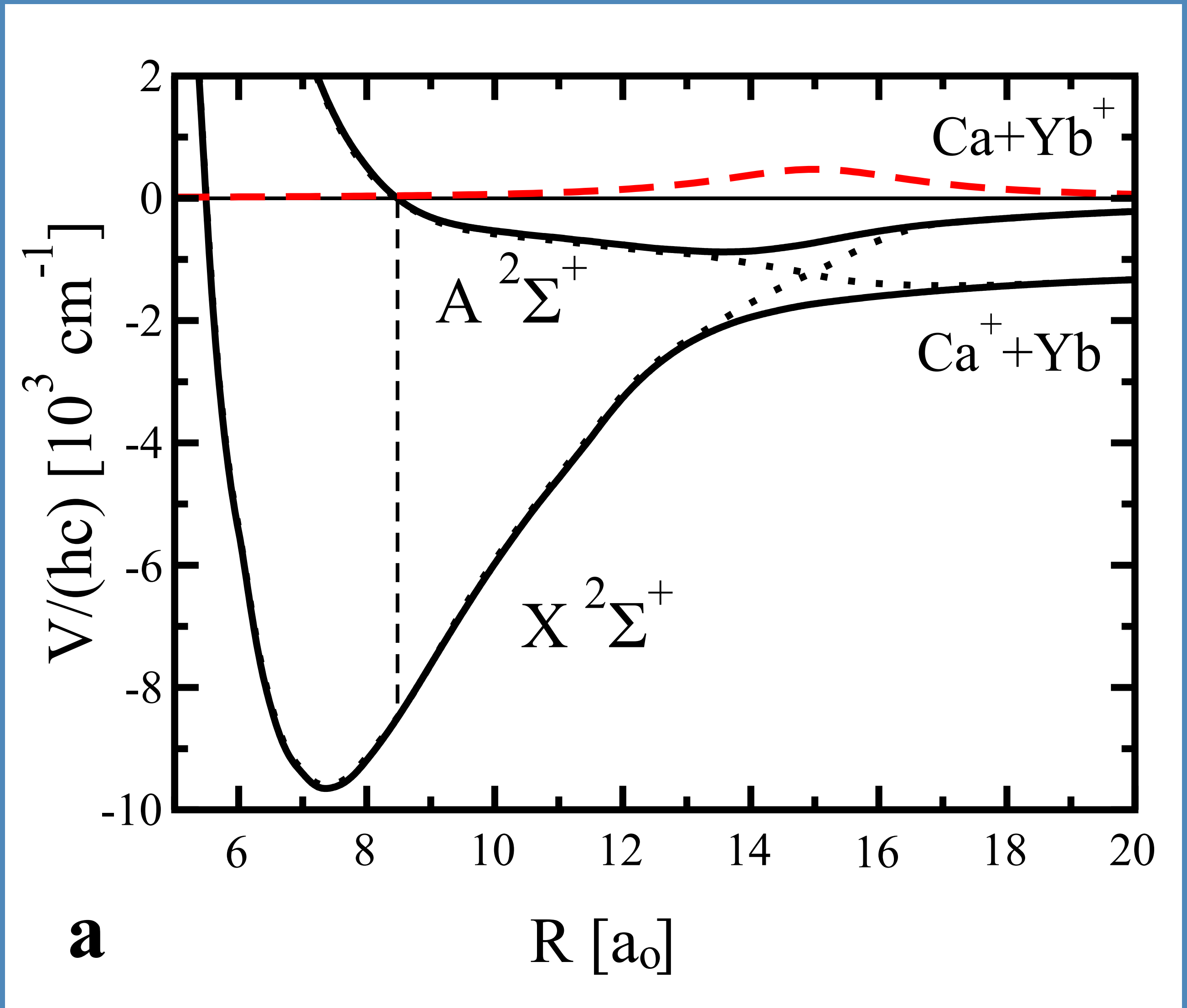
In these cases, radiative charge transfer is the most likely mode of transfer, and radiative charge transfer rates are all set to $1 \times 10^{-14}$ $cm^3$ $s^{-1}$ (Butler, Guberman & Dalgarno 1977). This should be correct to within an order of magnitude.

[J.B. Kingdon, Mon. Not. R. Astron. Soc., 274 425 (1995).]

# • Some surprising chemistry

## What is a charge-exchange chemical rxn?




### Three types of rxns:

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$Ca + Yb^{+} \rightarrow Ca^{+} + Yb$

- Landau-Zener type transition

In collaboration with Prof. Svetlana Kotochigova of Temple University

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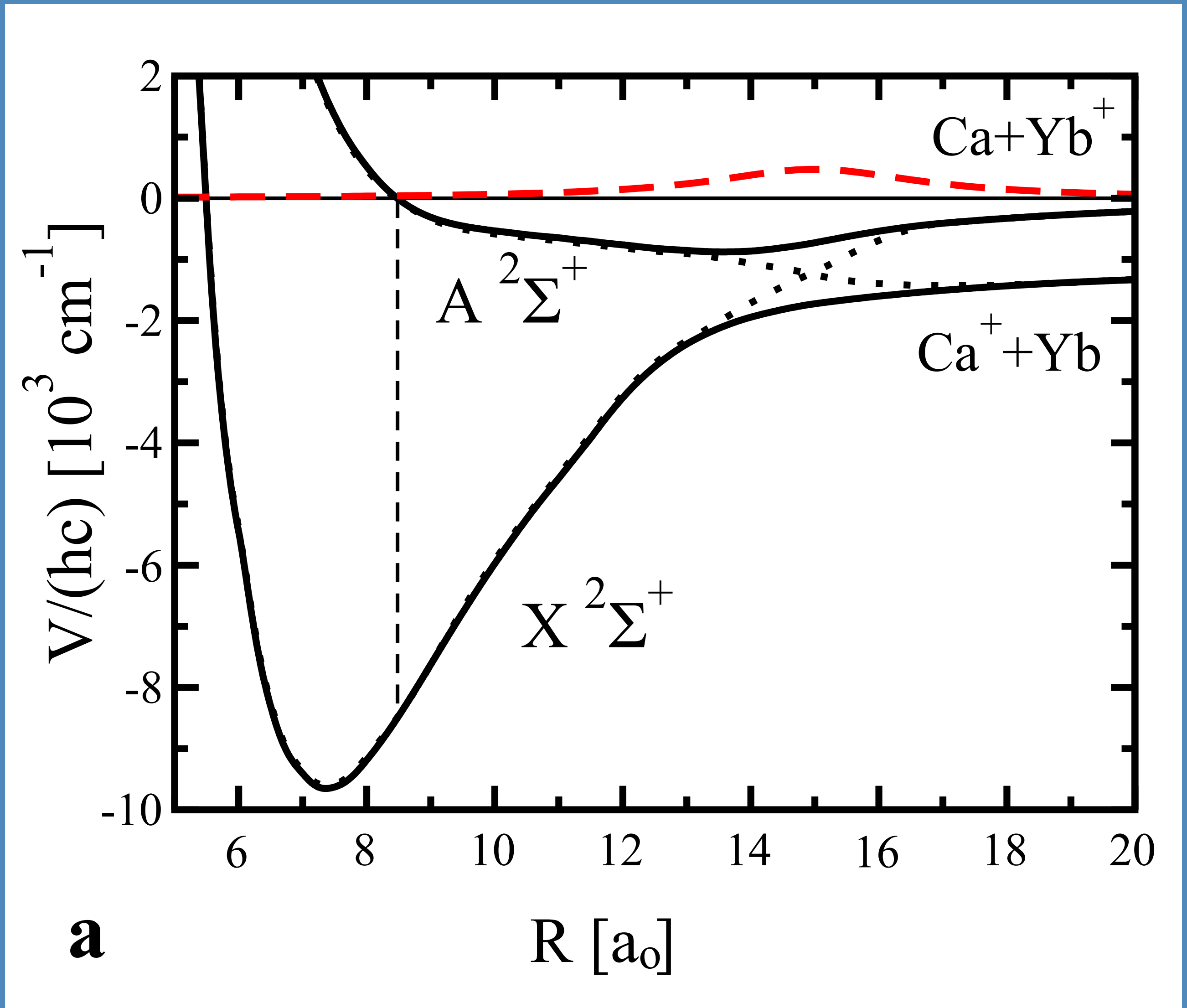


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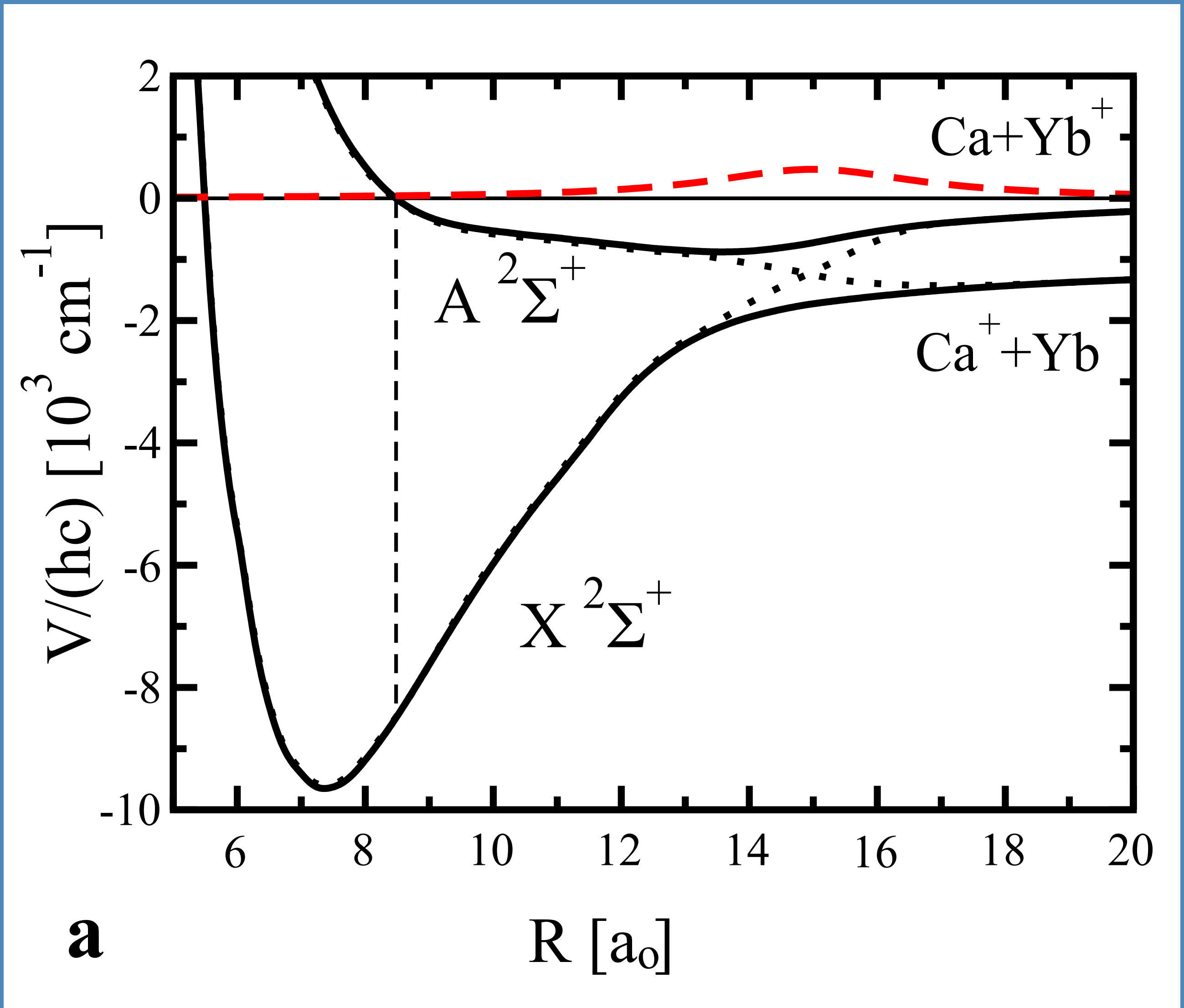


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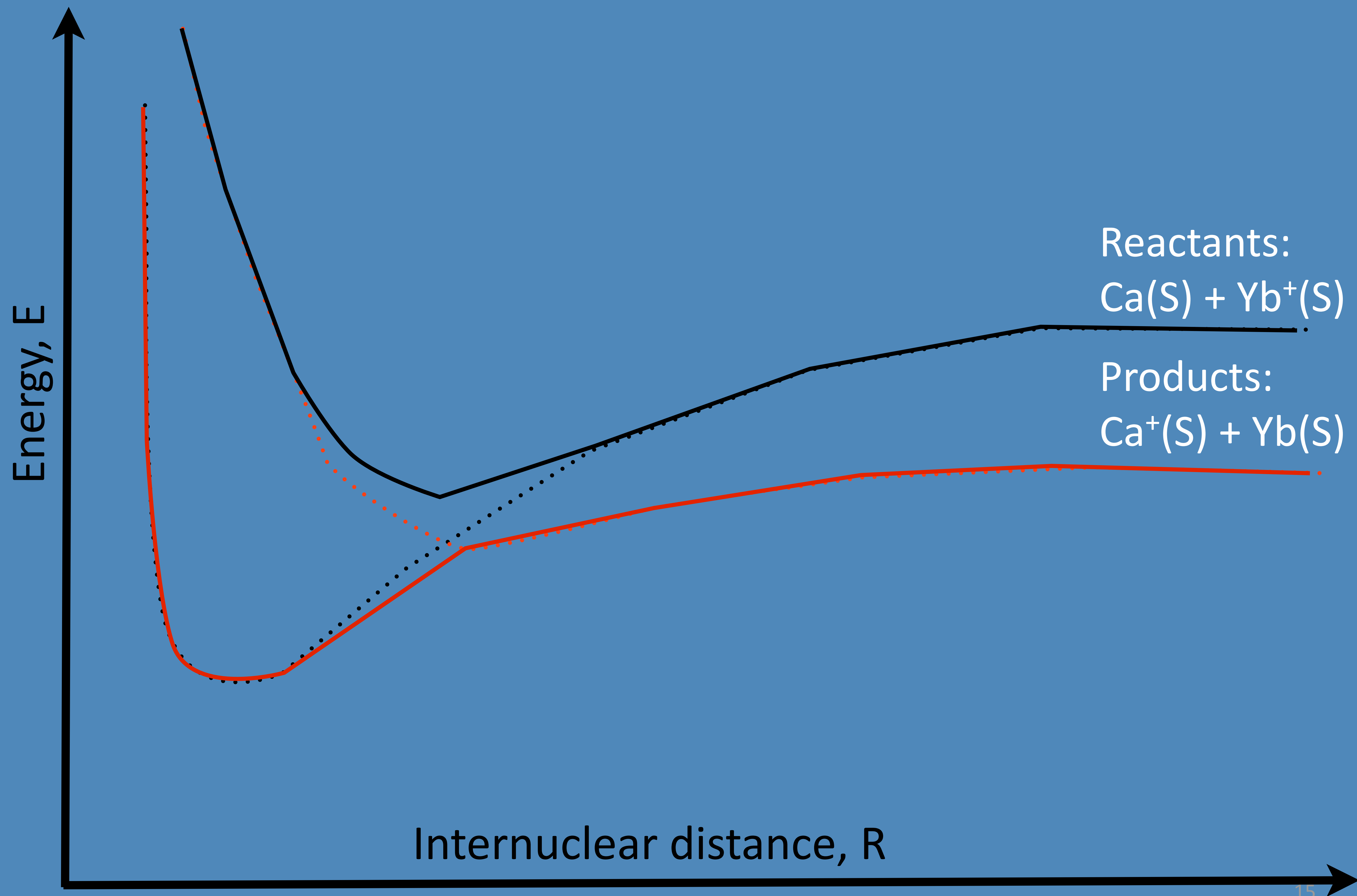

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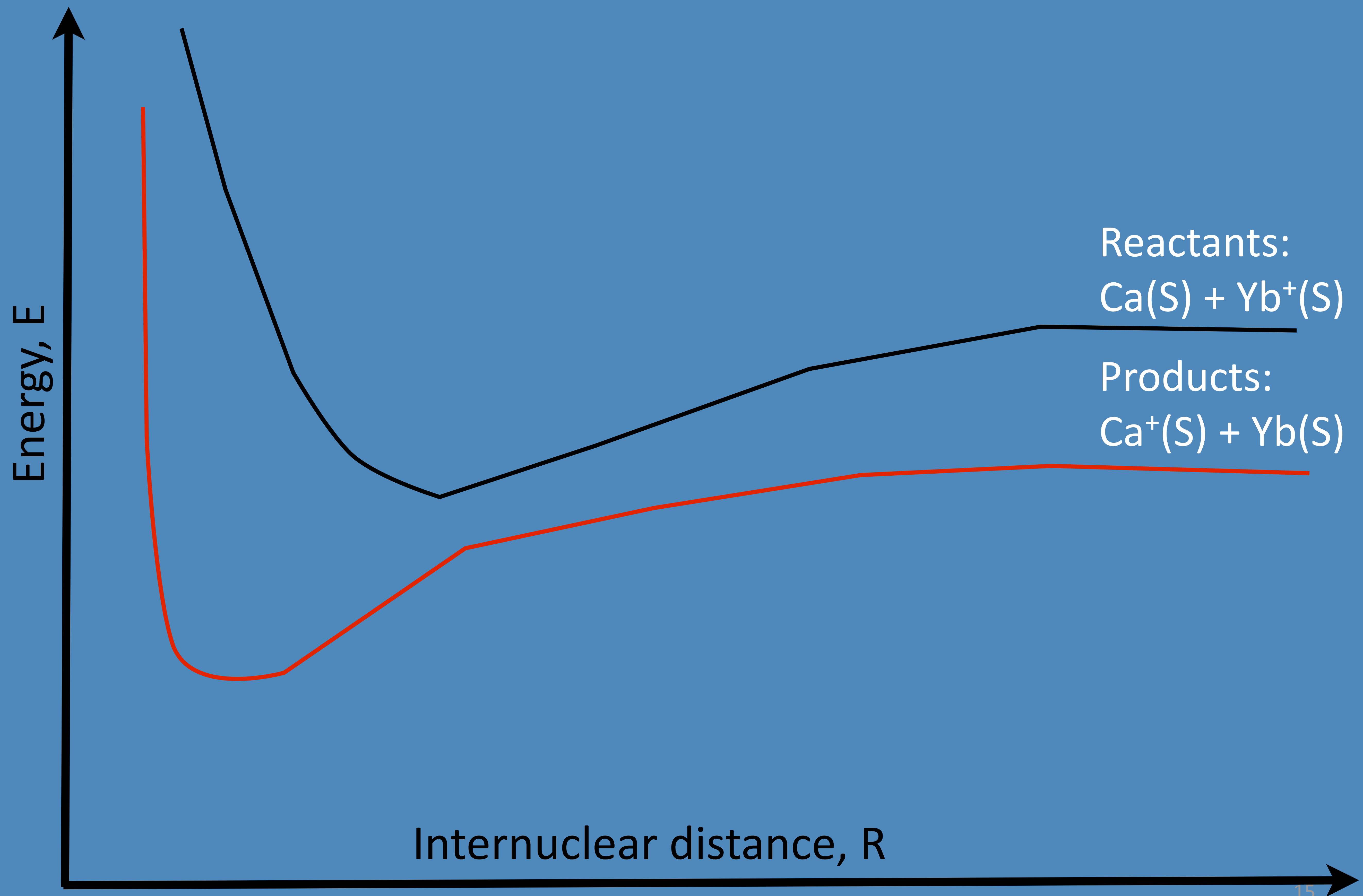

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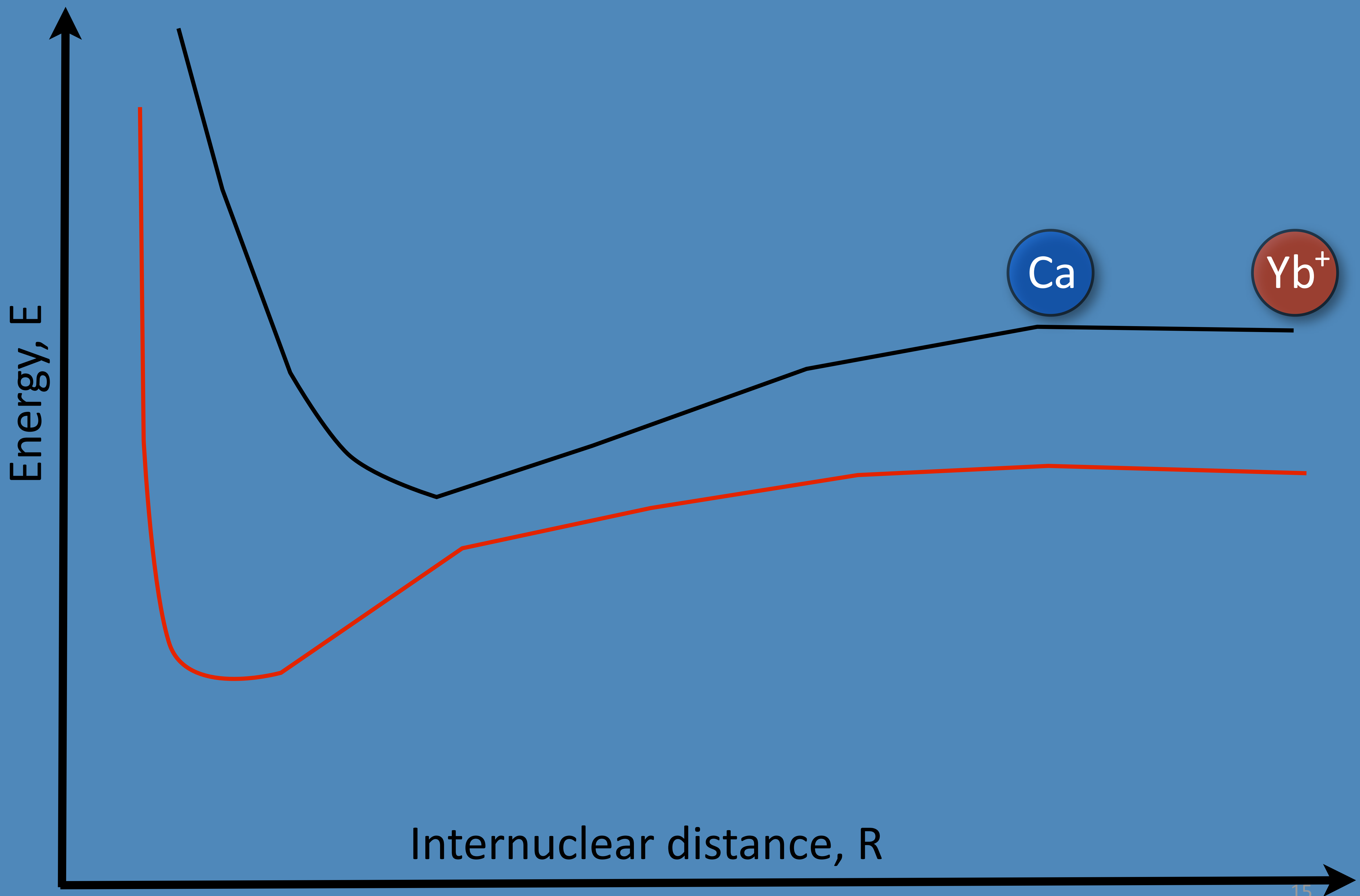

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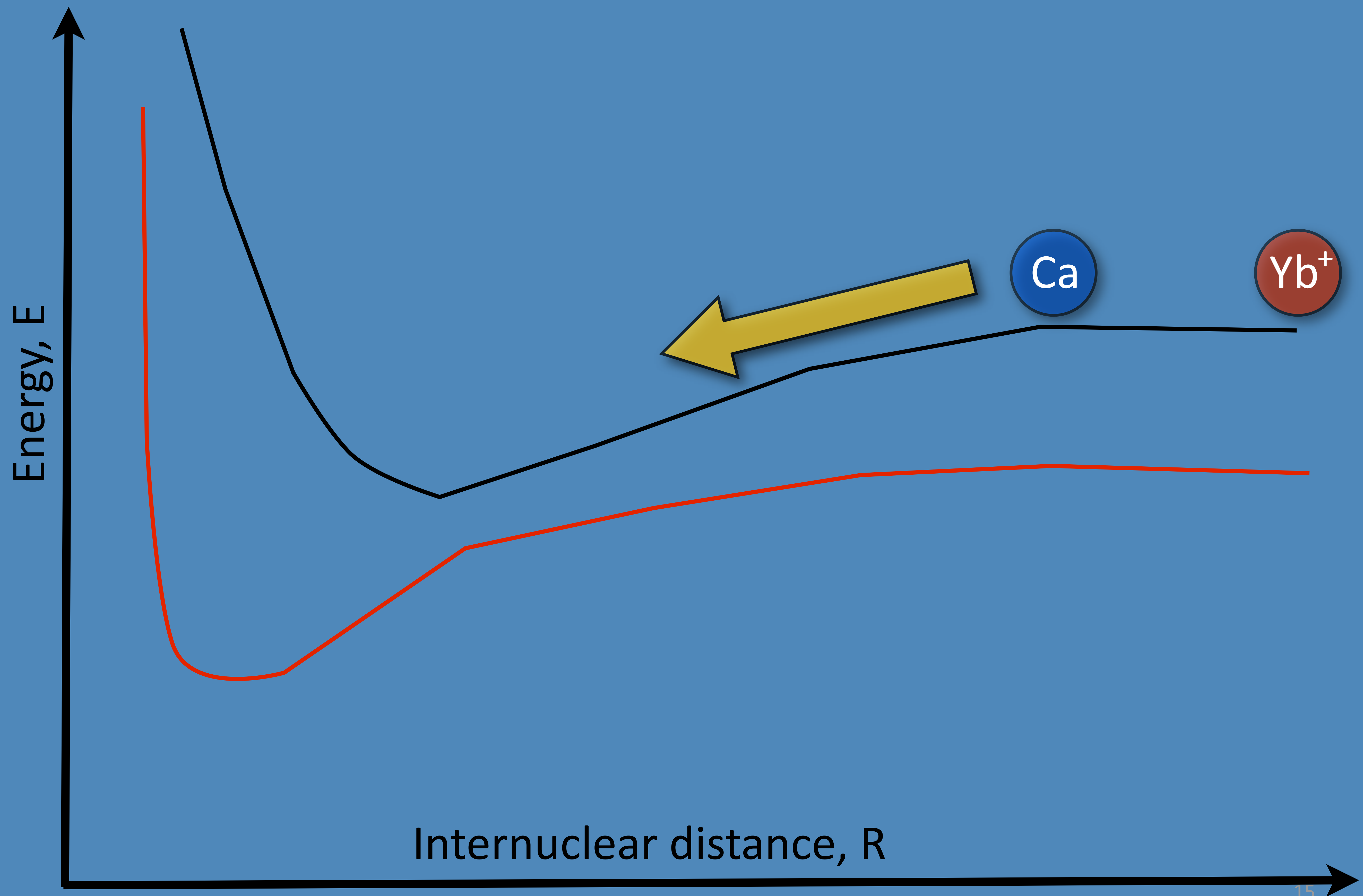

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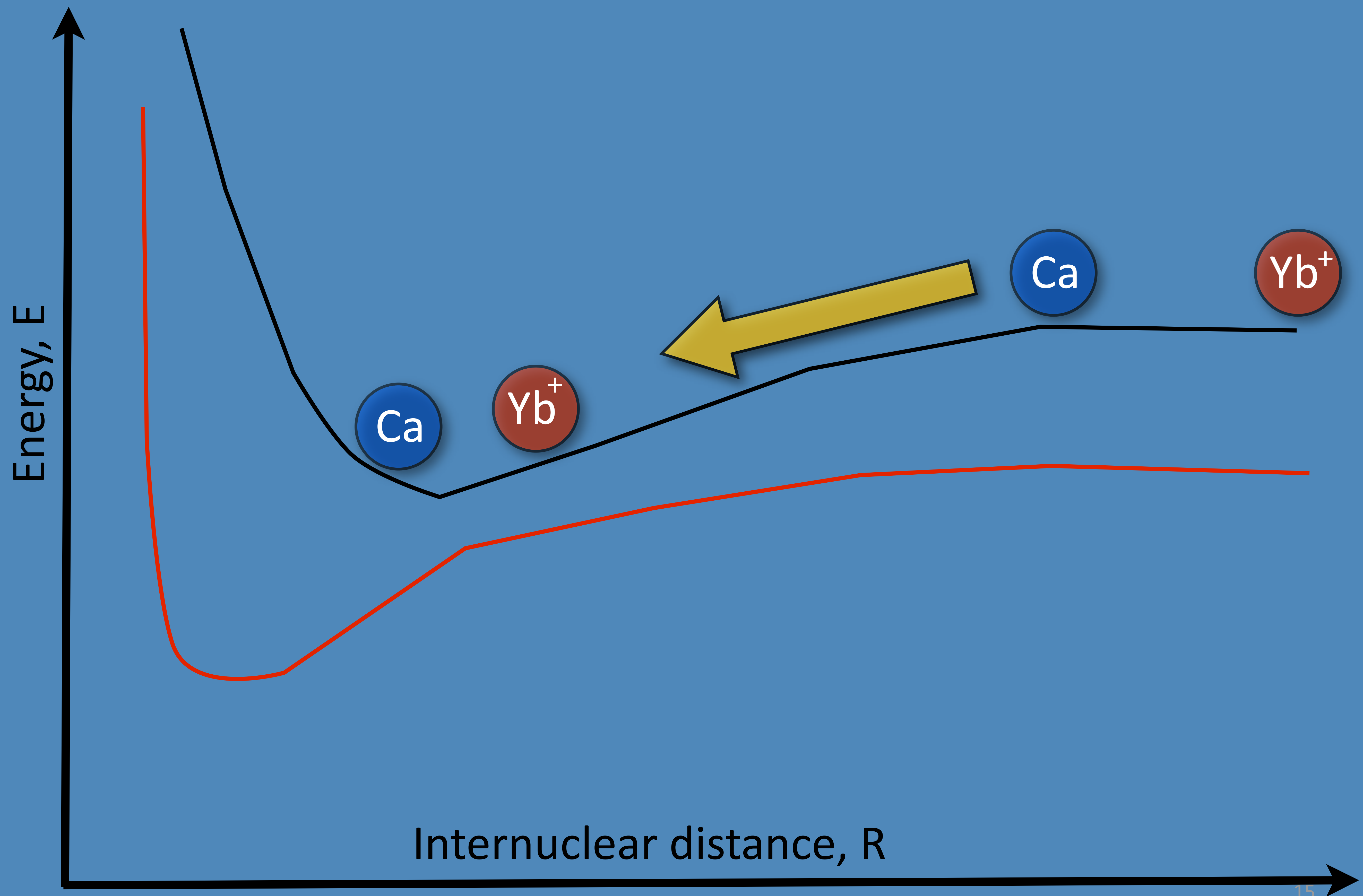

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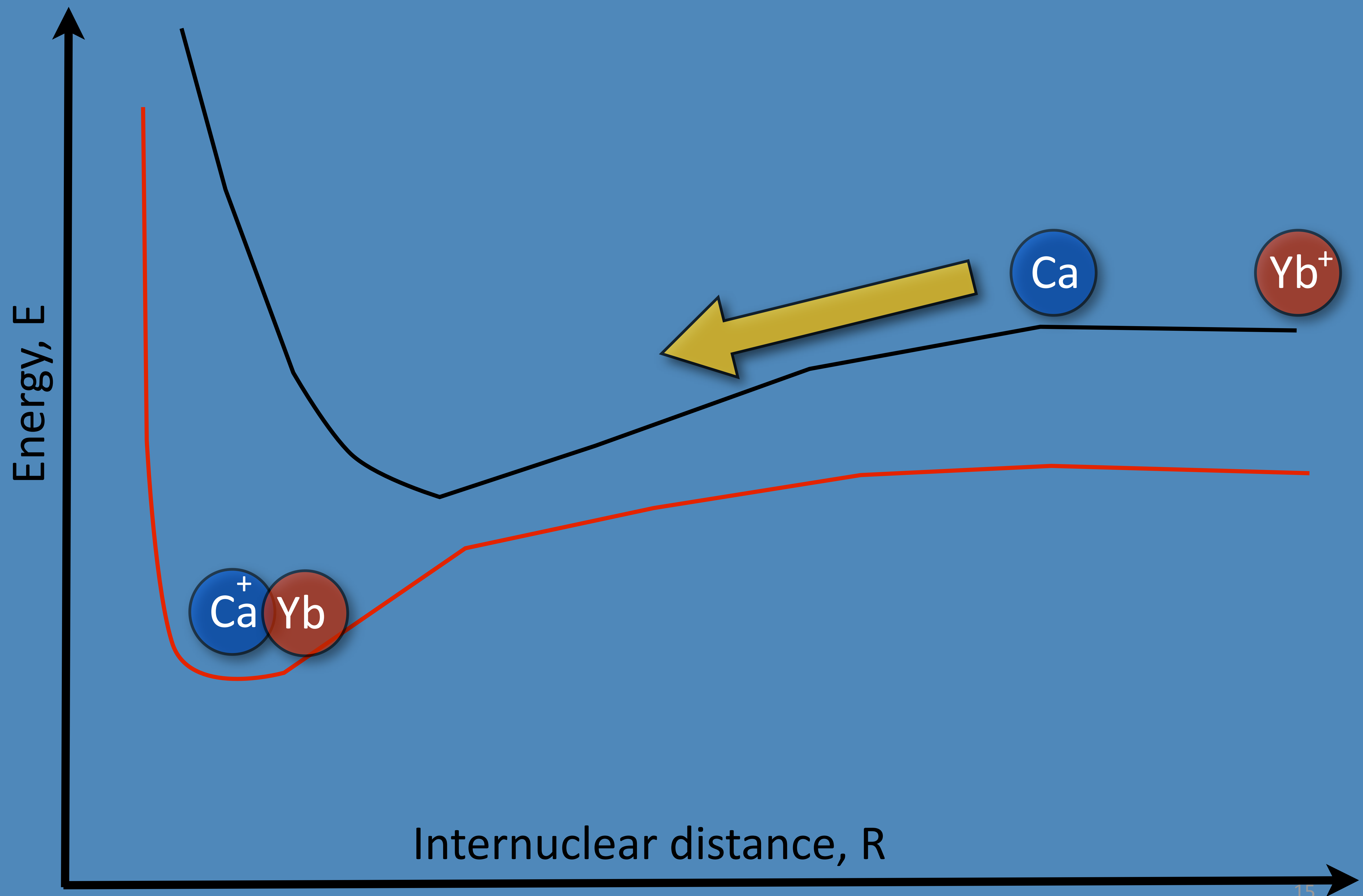

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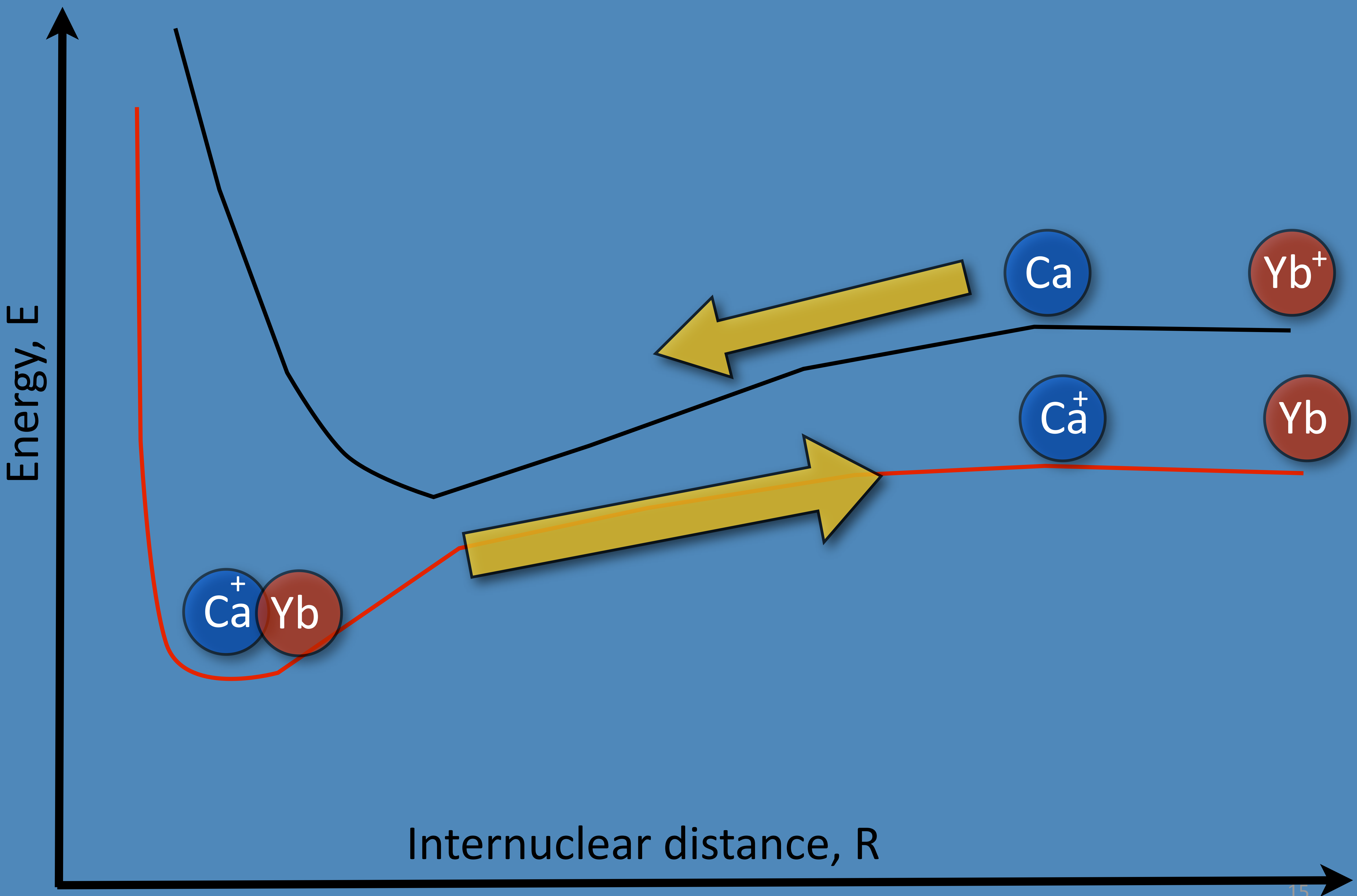


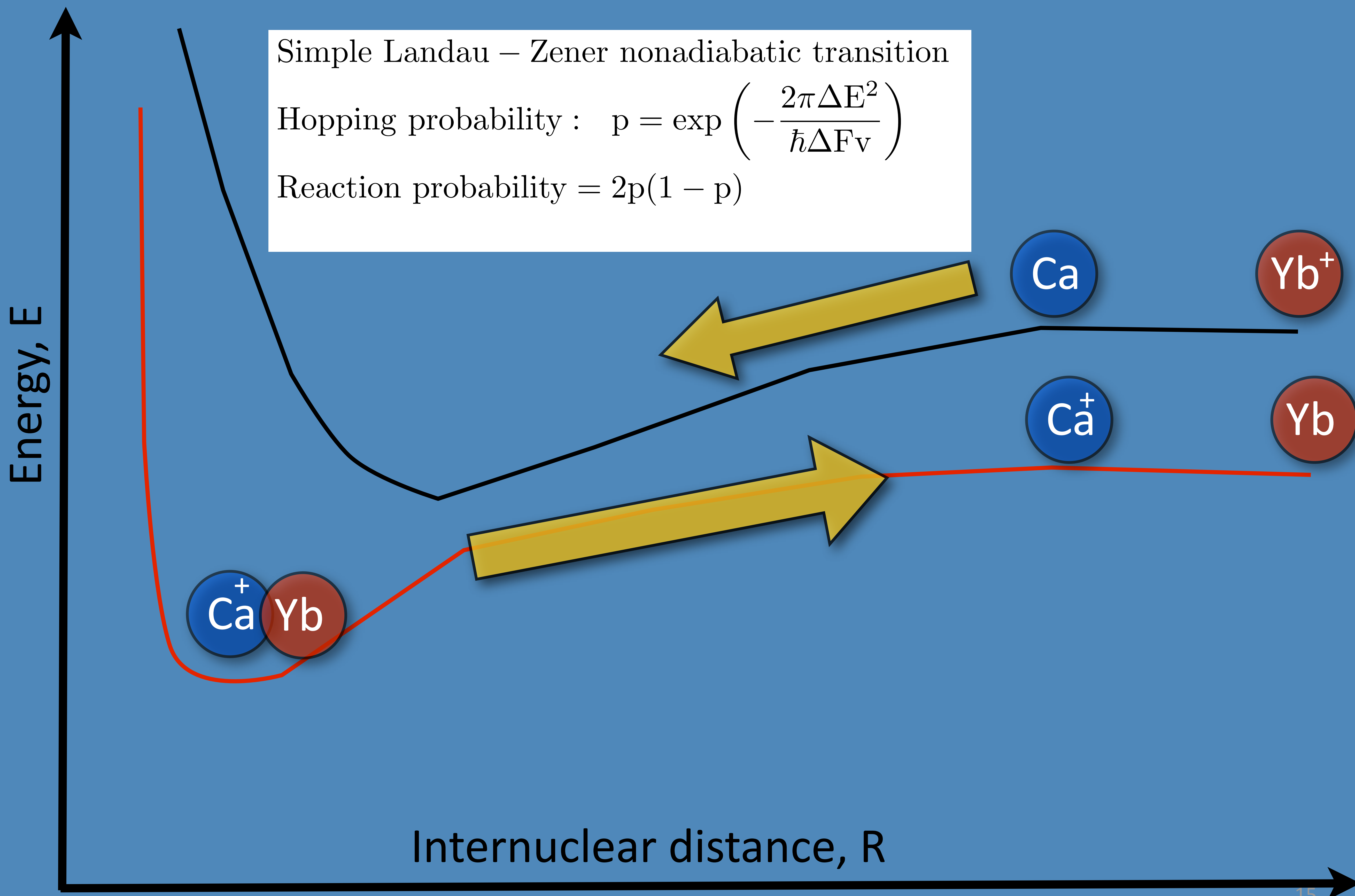
# Some surprising chemistry

## What is a charge-exchange chemical rxn?

Simple Landau – Zener nonadiabatic transition

Hopping probability : $p = \exp\left(-\frac{2\pi \Delta E^2}{\hbar \Delta F v}\right)$

Reaction probability $= 2p(1-p)$



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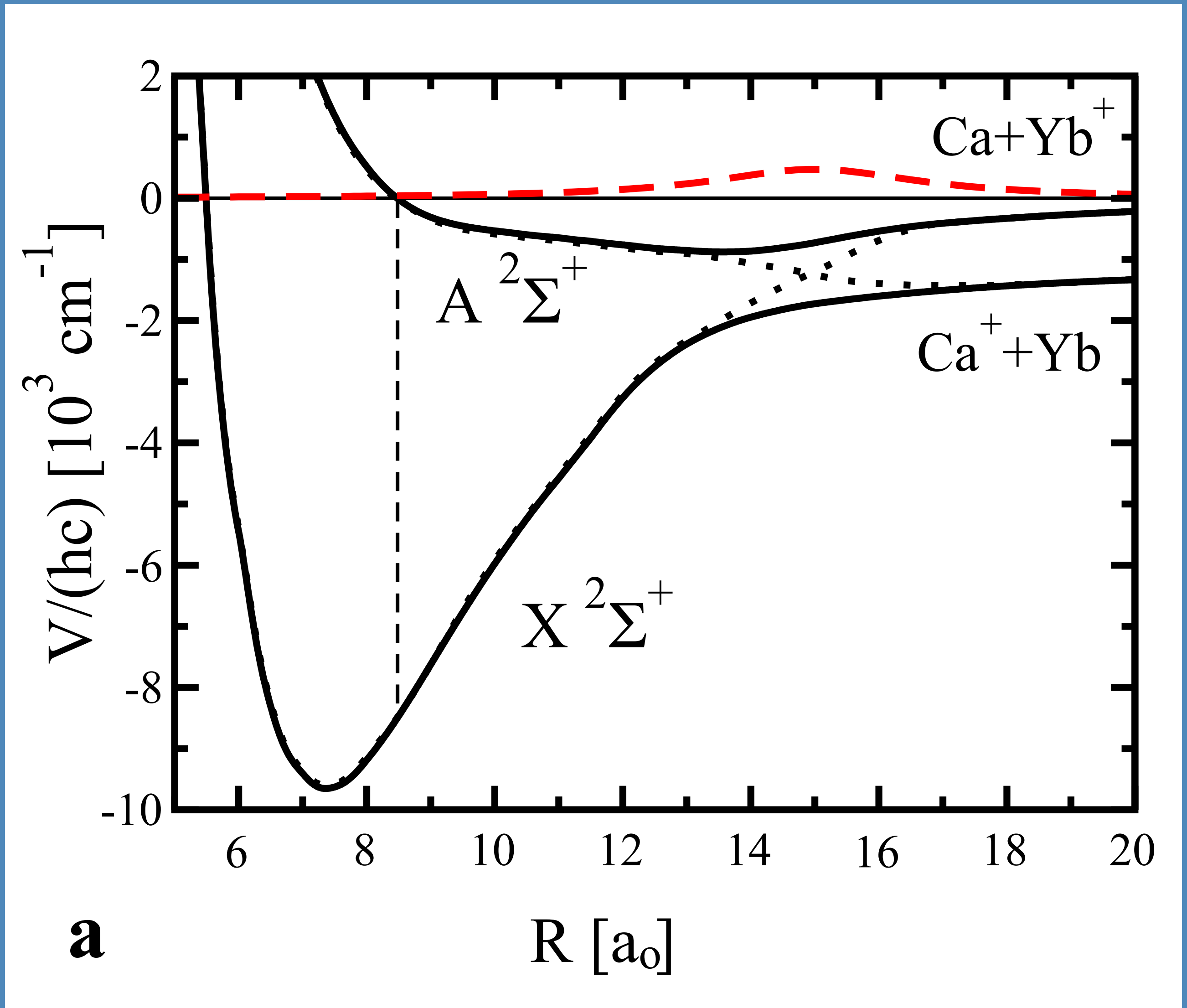


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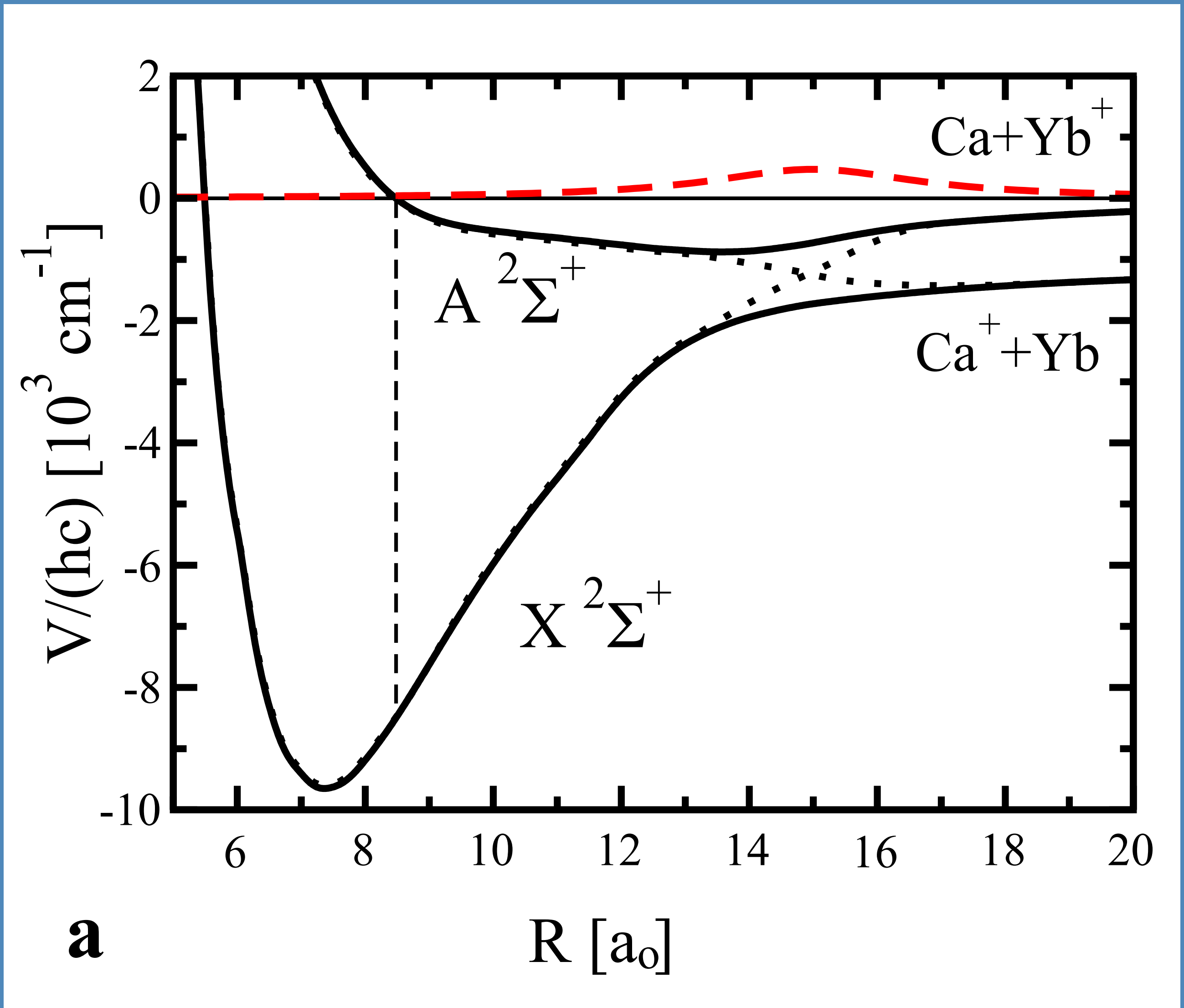


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# Some surprising chemistry

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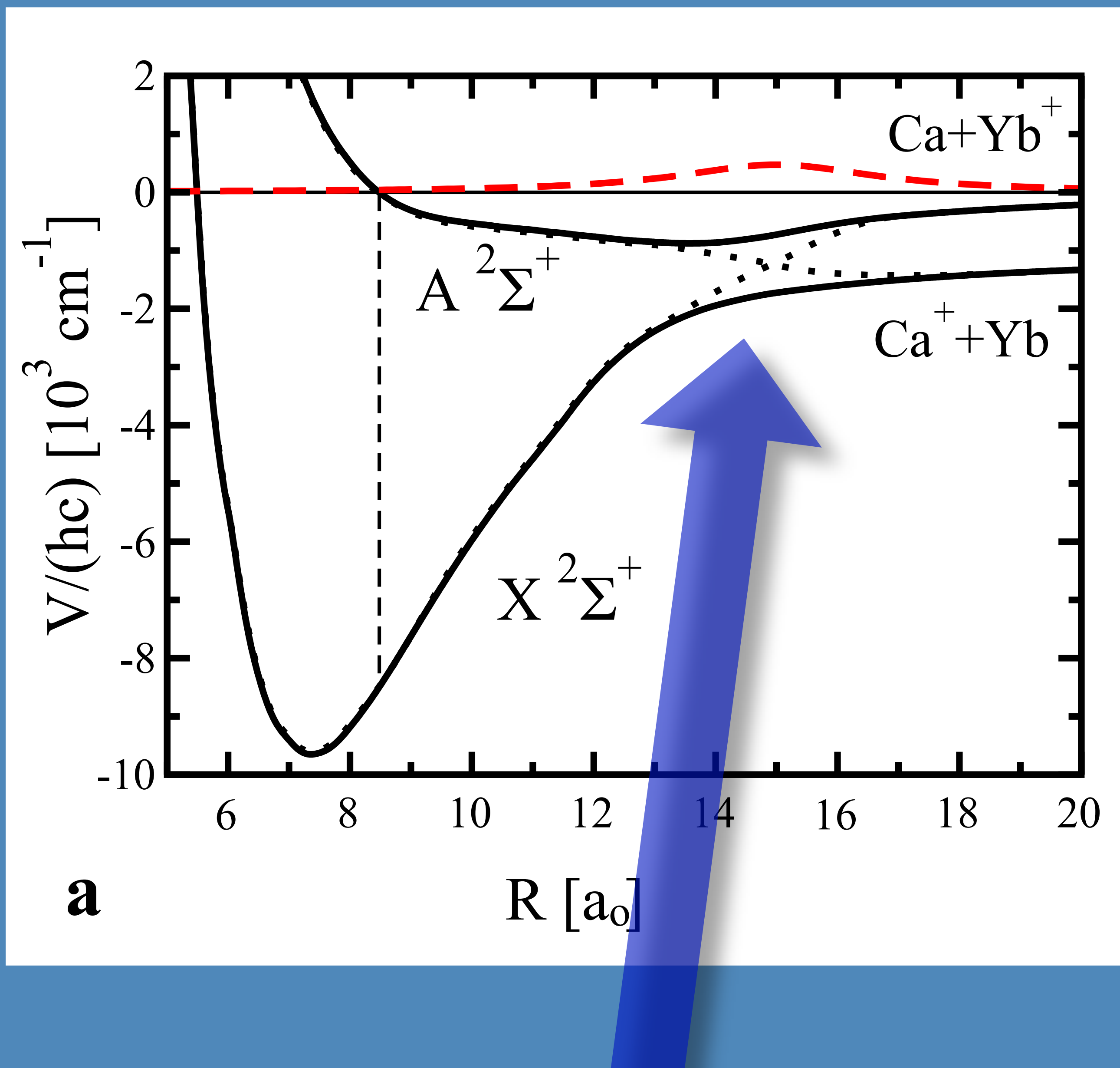


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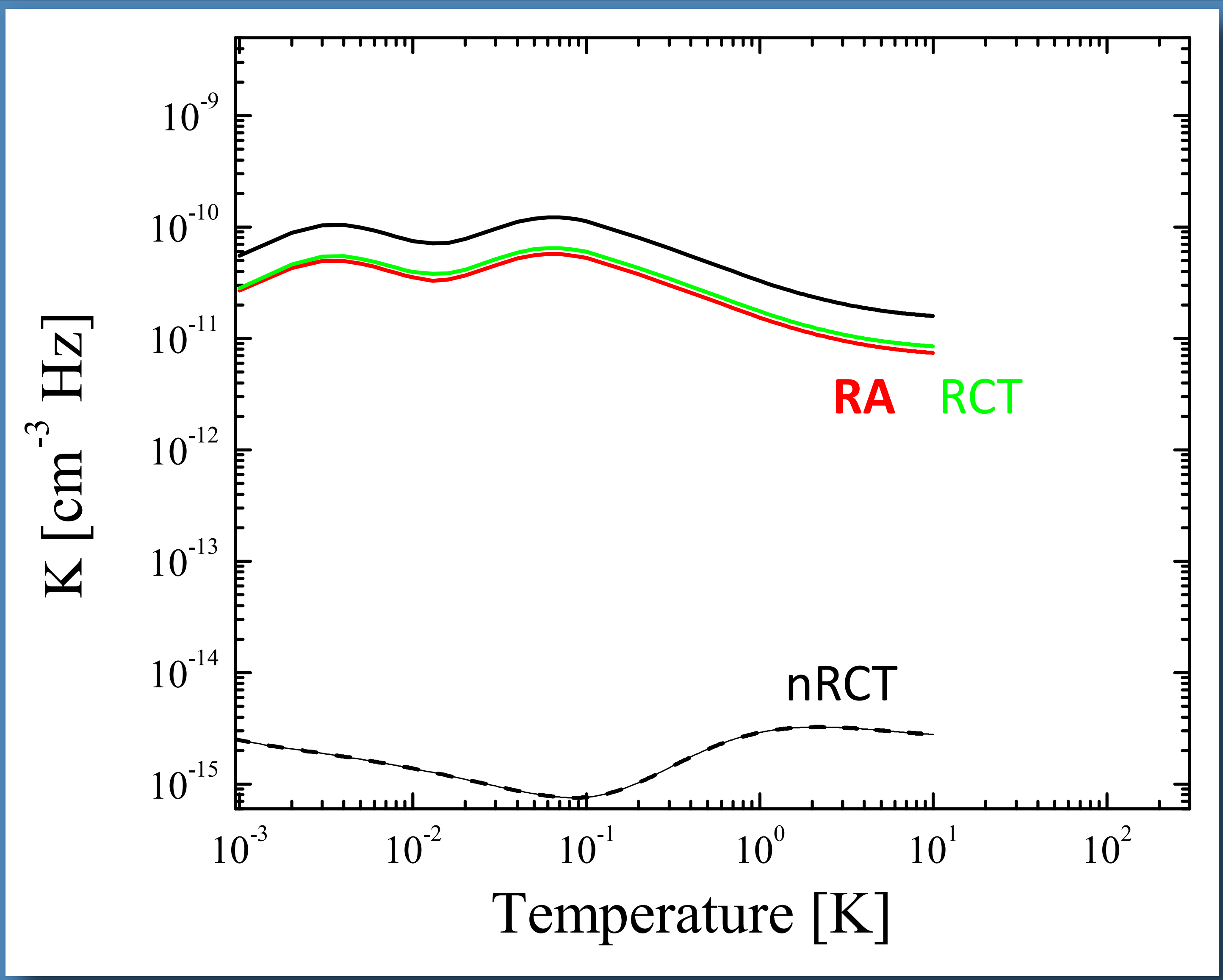
- Free-to-bound molecular E1 trans.

Avoided crossing gives rise to a large (several Debye) transition moment
+ Ca and Yb have similar dispersion coefficient

Lots of shape resonances , Large radiative rates

In collaboration with **Prof. Svetlana Kotochigova** of Temple University

# Some surprising chemistry




- Good Agreement with total rate
- Remaining mystery: No molecular ion formation observed
- Generic theory result, but most ultracold experiments have never observed the formation of a single molecular ion...

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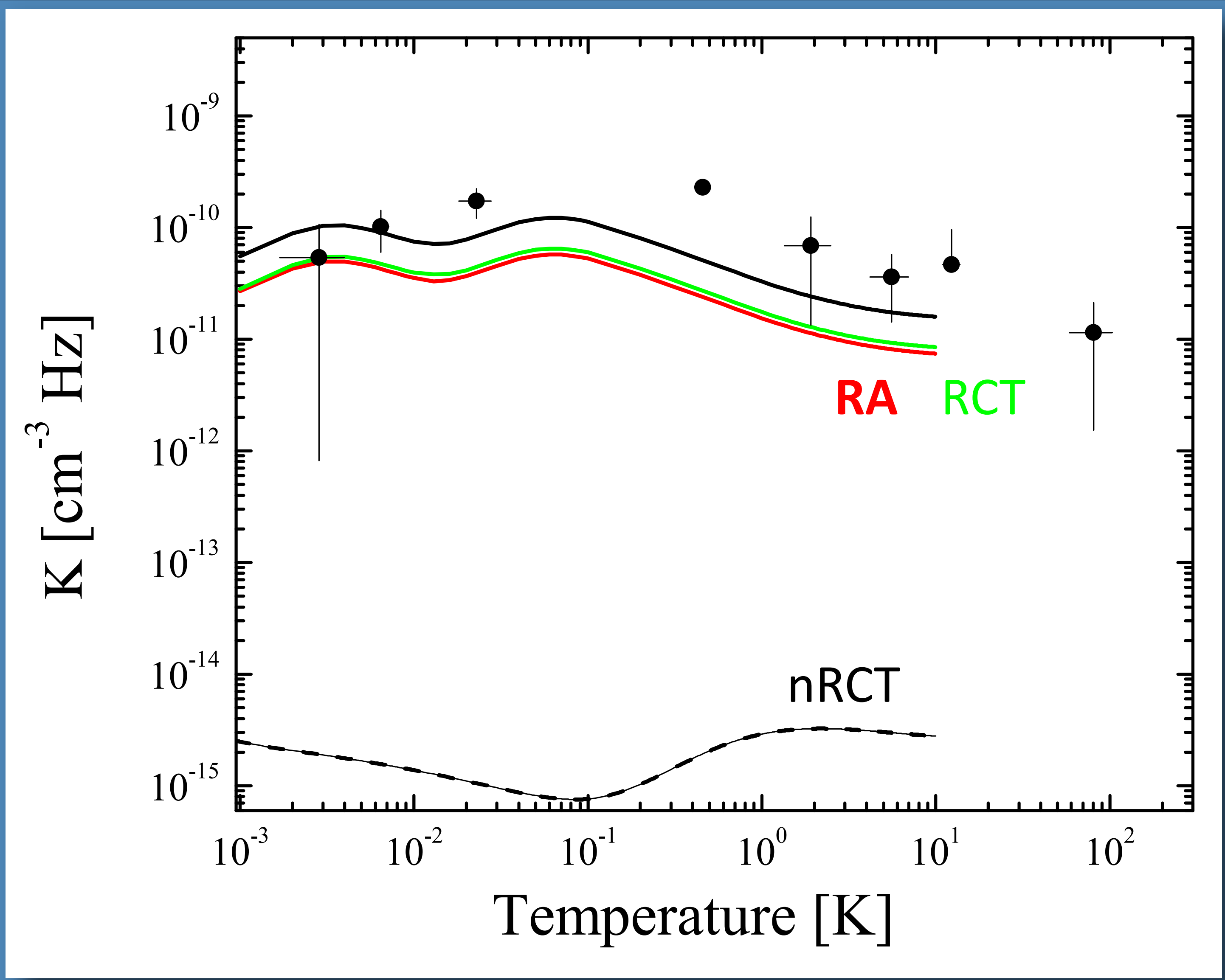


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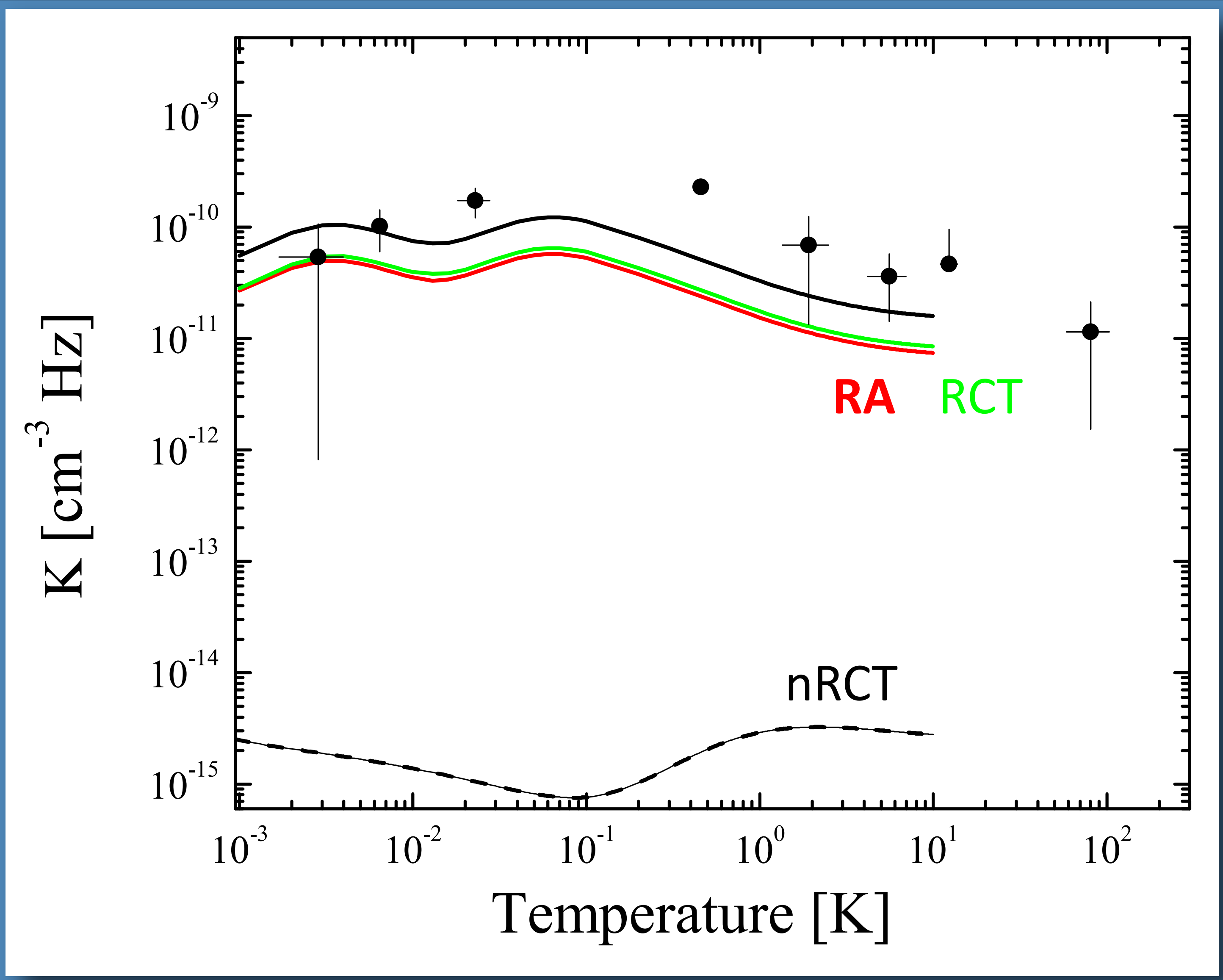


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Need for more work

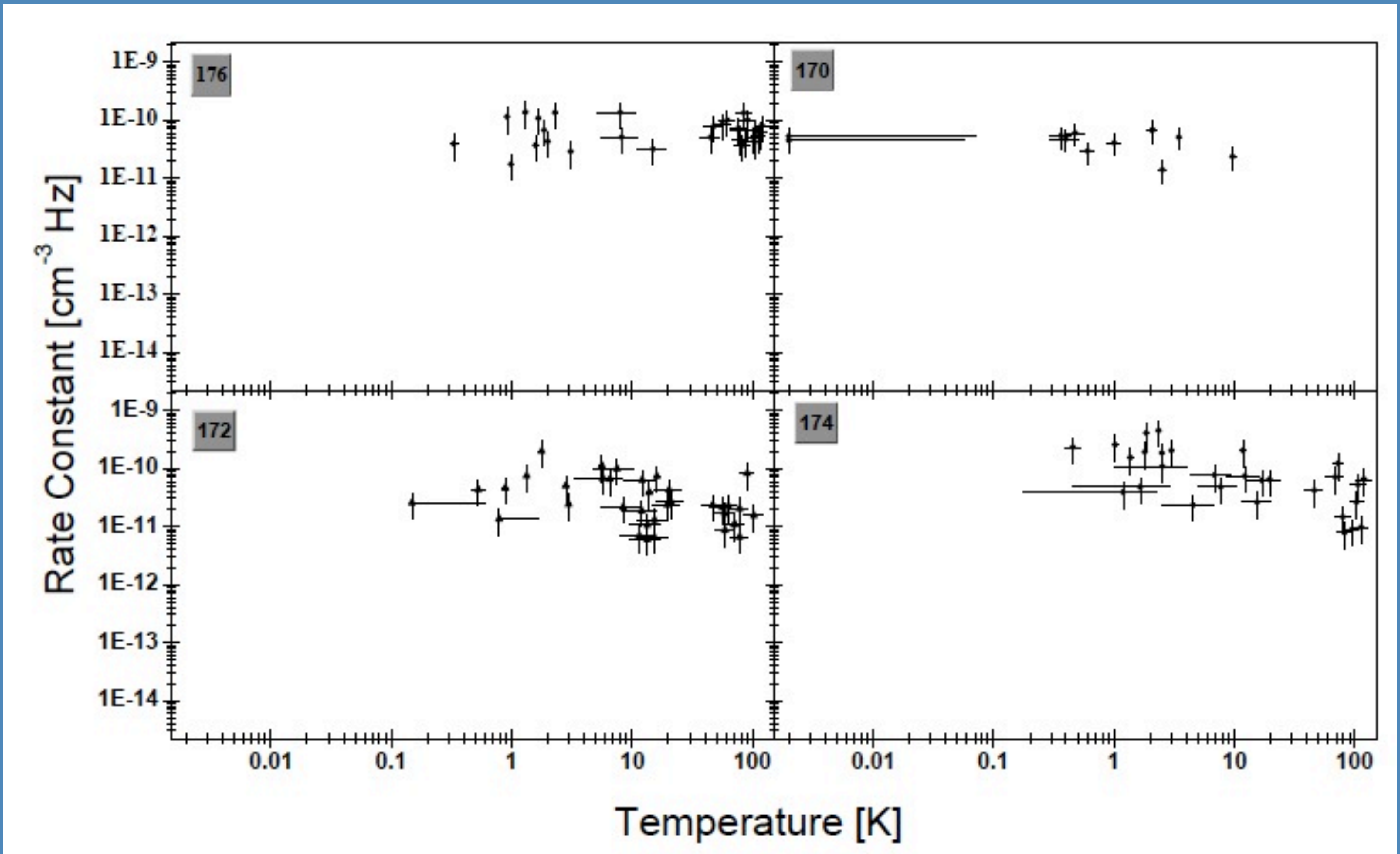
→ Impact Physics, chemistry, astrophysics

More details: W.G. Rellergert et al., Phys. Rev. Lett. 107, 243201 (2011)
in collaboration with Svetlana Kotochigova

# Some surprising chemistry

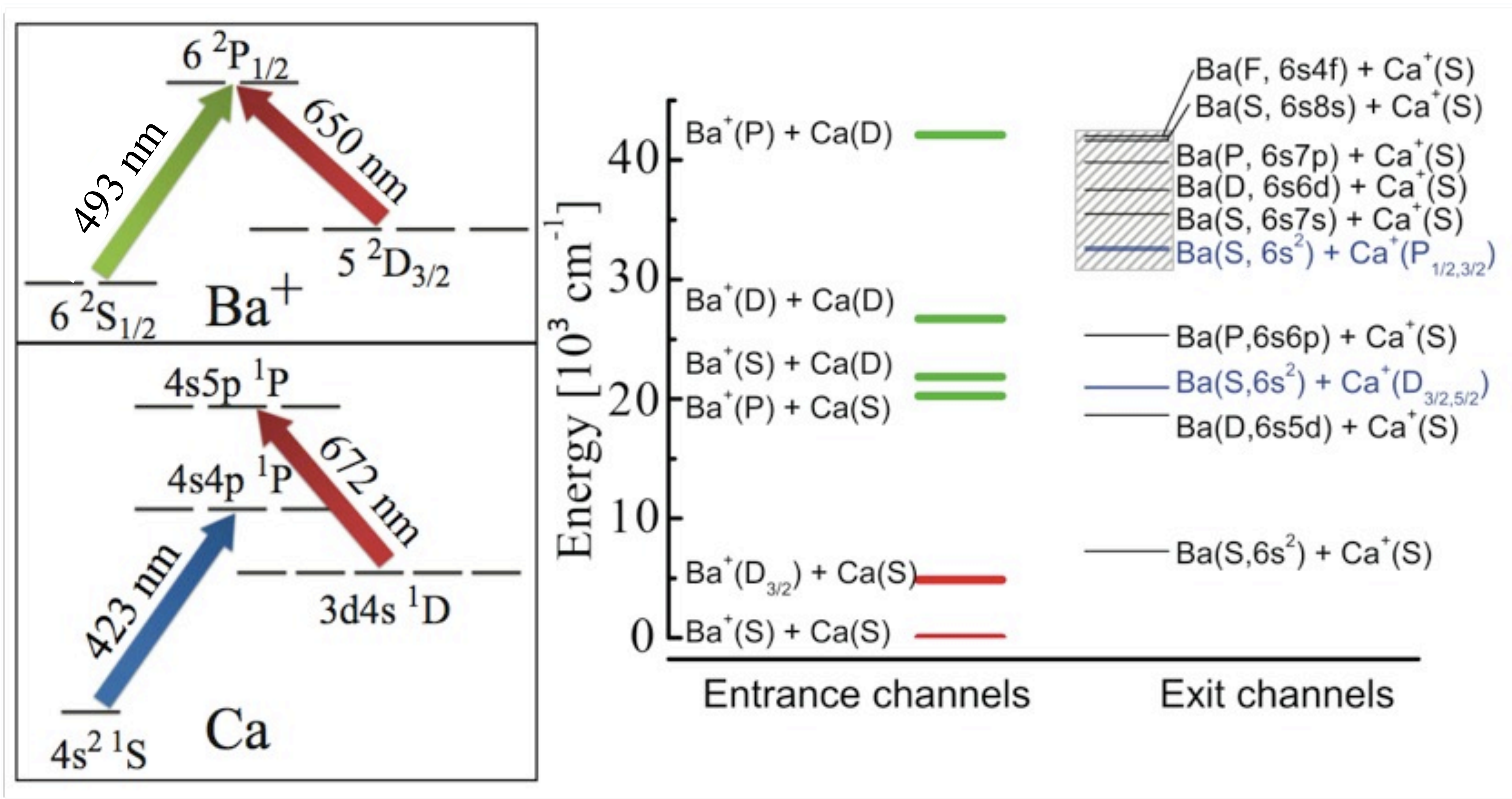
## Resolving single isotope chemistry

Reactants: $^{40}Ca + {}^{n}Yb^{+}$



# • Some surprising chemistry

## Solution → The Ca + $Ba^+$ System



# • Some surprising chemistry

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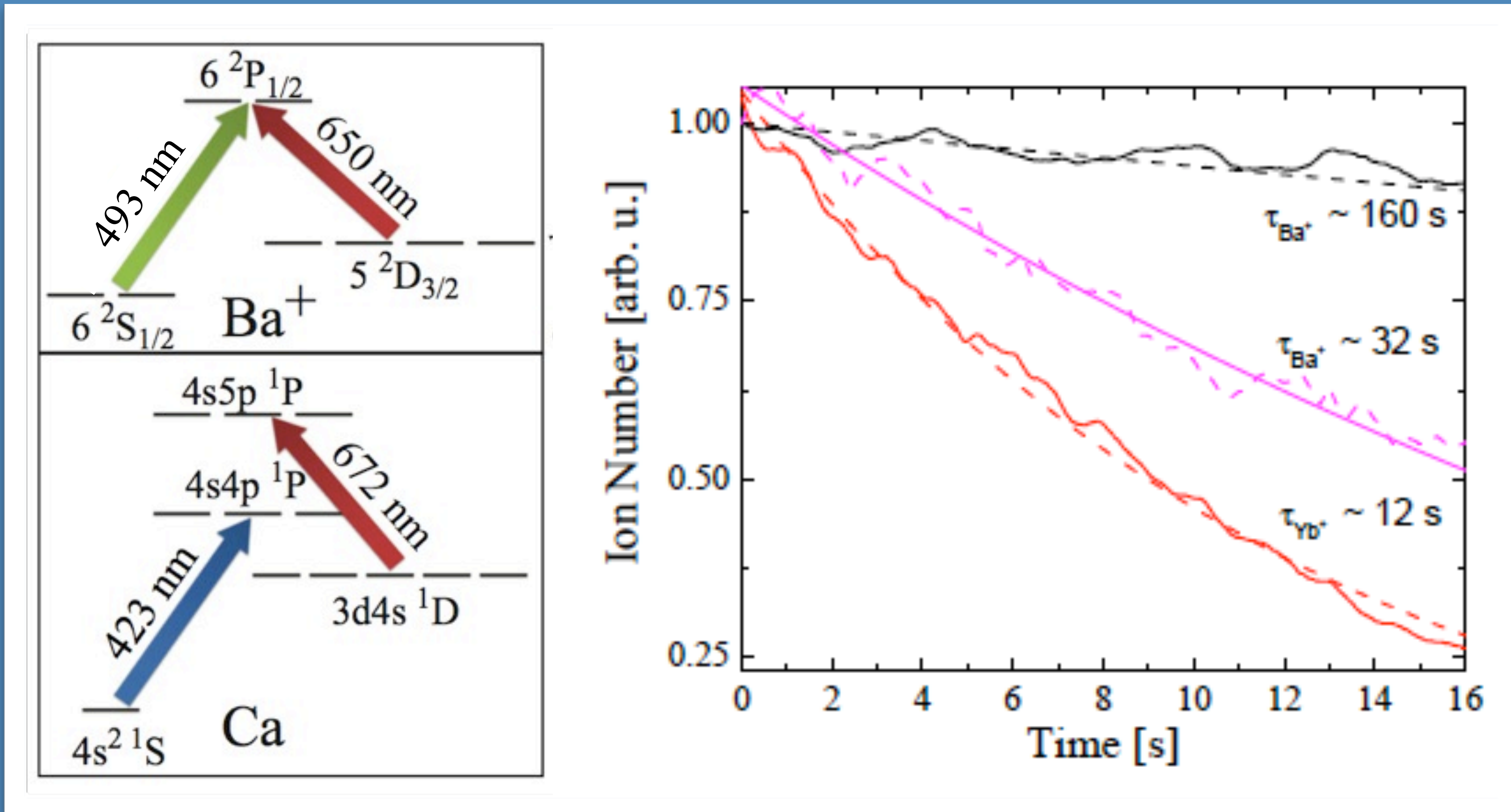

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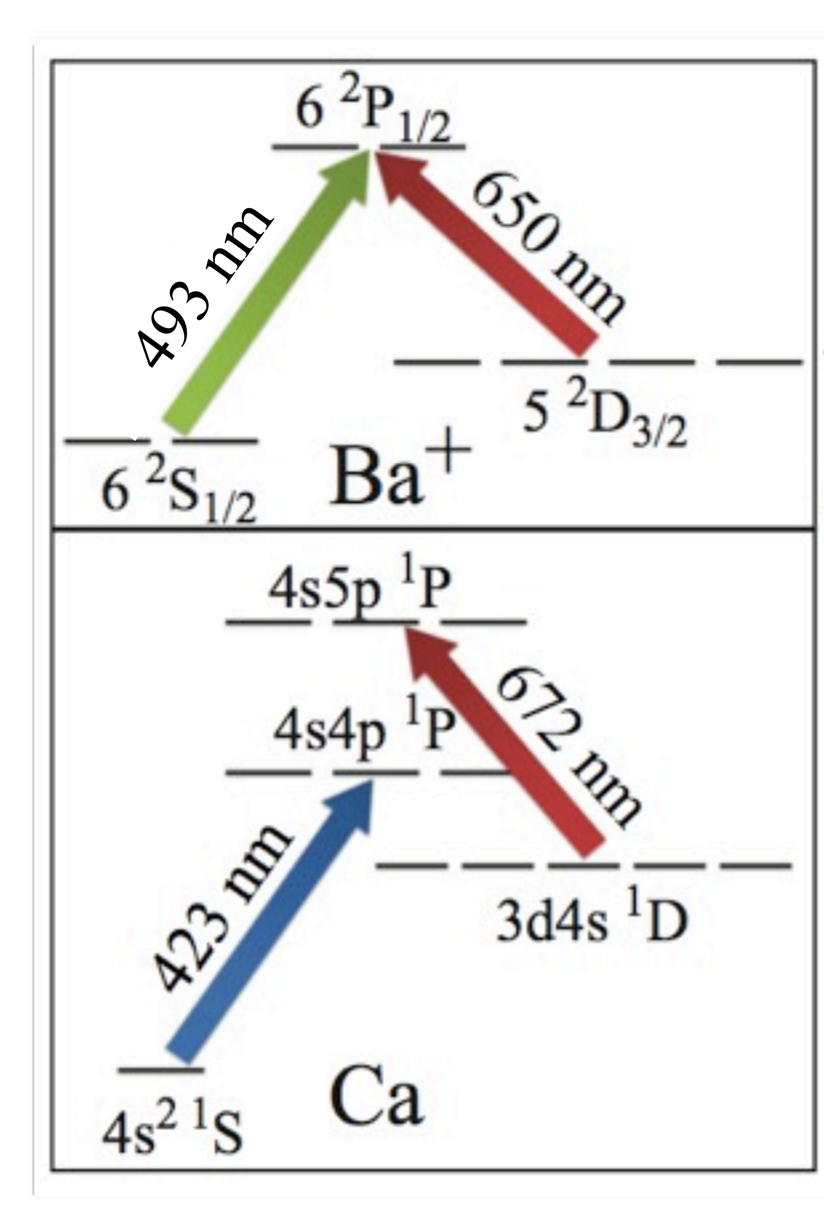


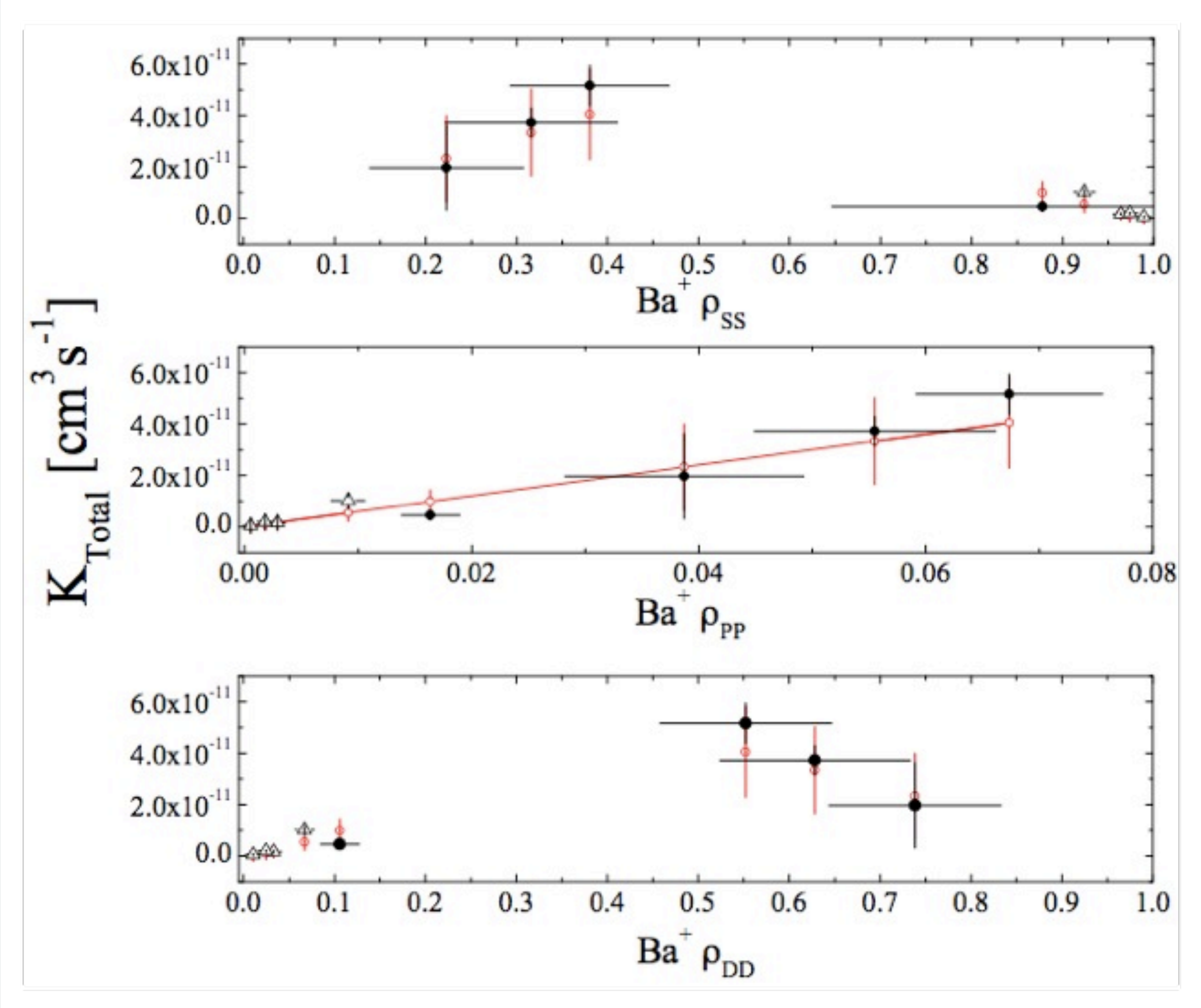

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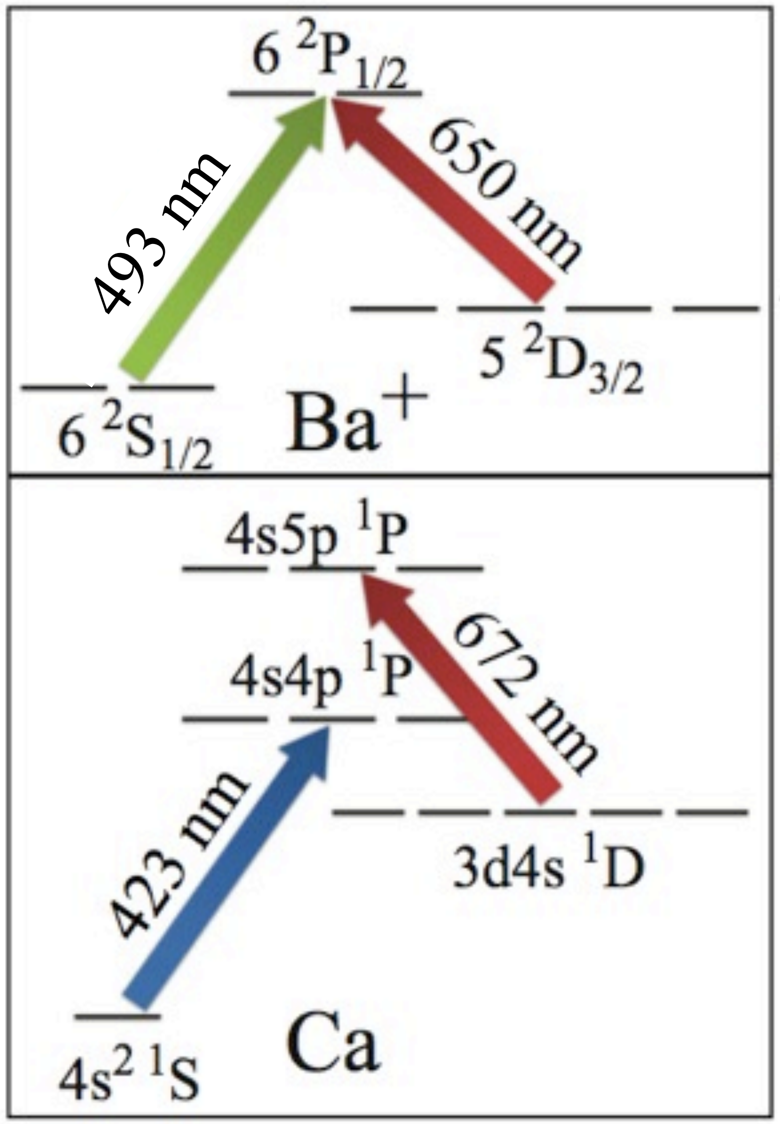


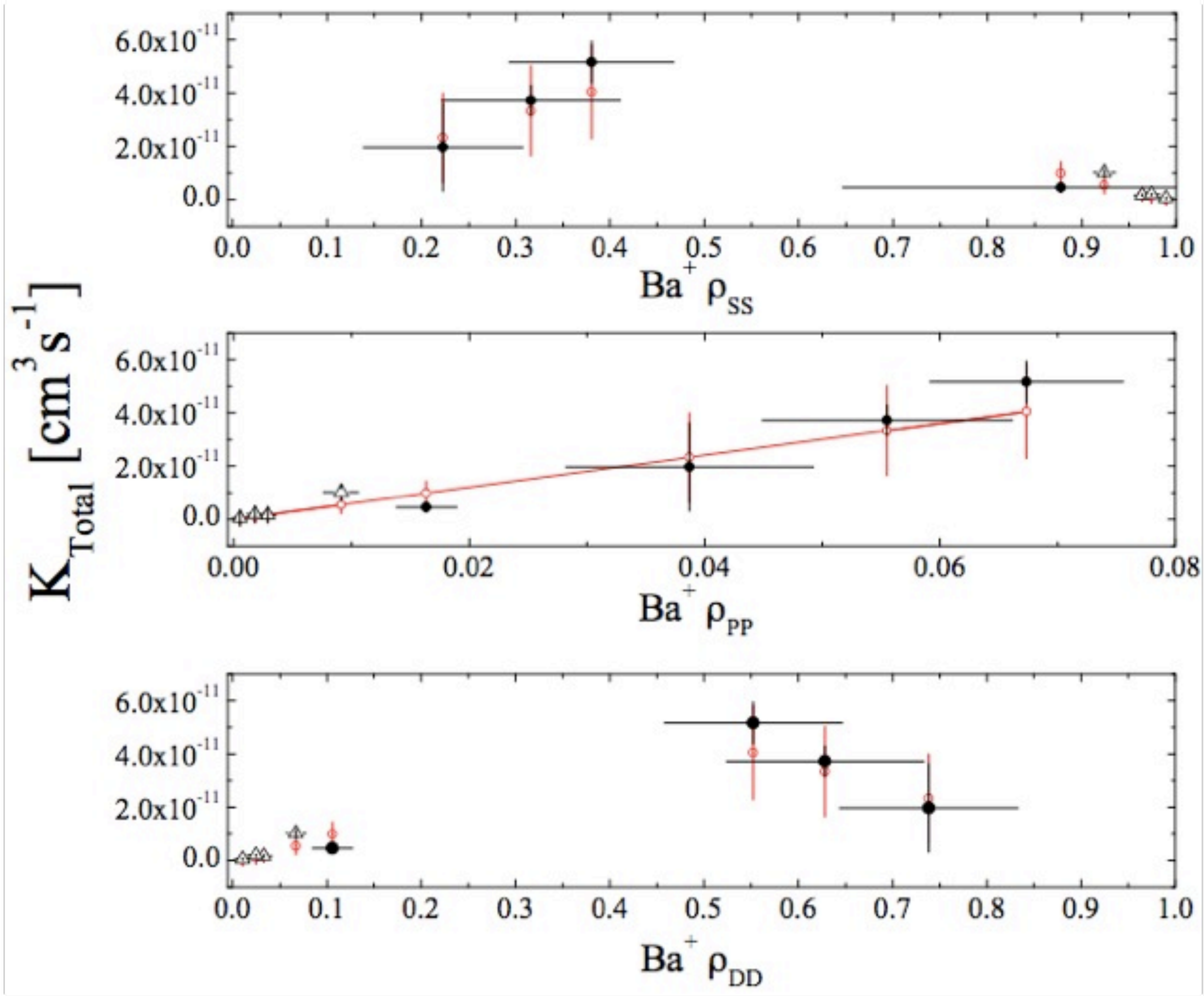


$$K = \rho_{pp}^{Ba}\rho_{ss}^{*Ca}K_{ps} + \rho_{ss}^{Ba}\rho_{dd}^{Ca}K_{sd} + \rho_{dd}^{Ba}\rho_{dd}^{Ca}K_{dd} + \rho_{pp}^{Ba}\rho_{dd}^{Ca}K_{pd}$$

# Some surprising chemistry

## CaBa$^+$ channel dependent rate constants

$$K = \rho_{pp}^{Ba}\rho_{ss}^{*Ca}K_{ps} + \rho_{ss}^{Ba}\rho_{dd}^{Ca}K_{sd} + \rho_{dd}^{Ba}\rho_{dd}^{Ca}K_{dd} + \rho_{pp}^{Ba}\rho_{dd}^{Ca}K_{pd}$$

TABLE I: Measured reaction rate constants.

| Entrance Channel | $K_{ab}$ [$cm^3s^{-1}$] |
|---|---|
| $Ba^+(6^2P_{1/2})$ + $Ca(4^1S)$ | $4.3(1.1) \times 10^{-10}$ |
| $Ba^+(6^2S_{1/2})$ + $Ca(3^1D)$ | $\leq 1.5 \times 10^{-12}$ |
| $Ba^+(5^2D_{3/2})$ + $Ca(3^1D)$ | $\leq 8 \times 10^{-12}$ |
| $Ba^+(6^2P_{1/2})$ + $Ca(3^1D)$ | $\leq 1 \times 10^{-9}$ |

# • Some surprising chemistry

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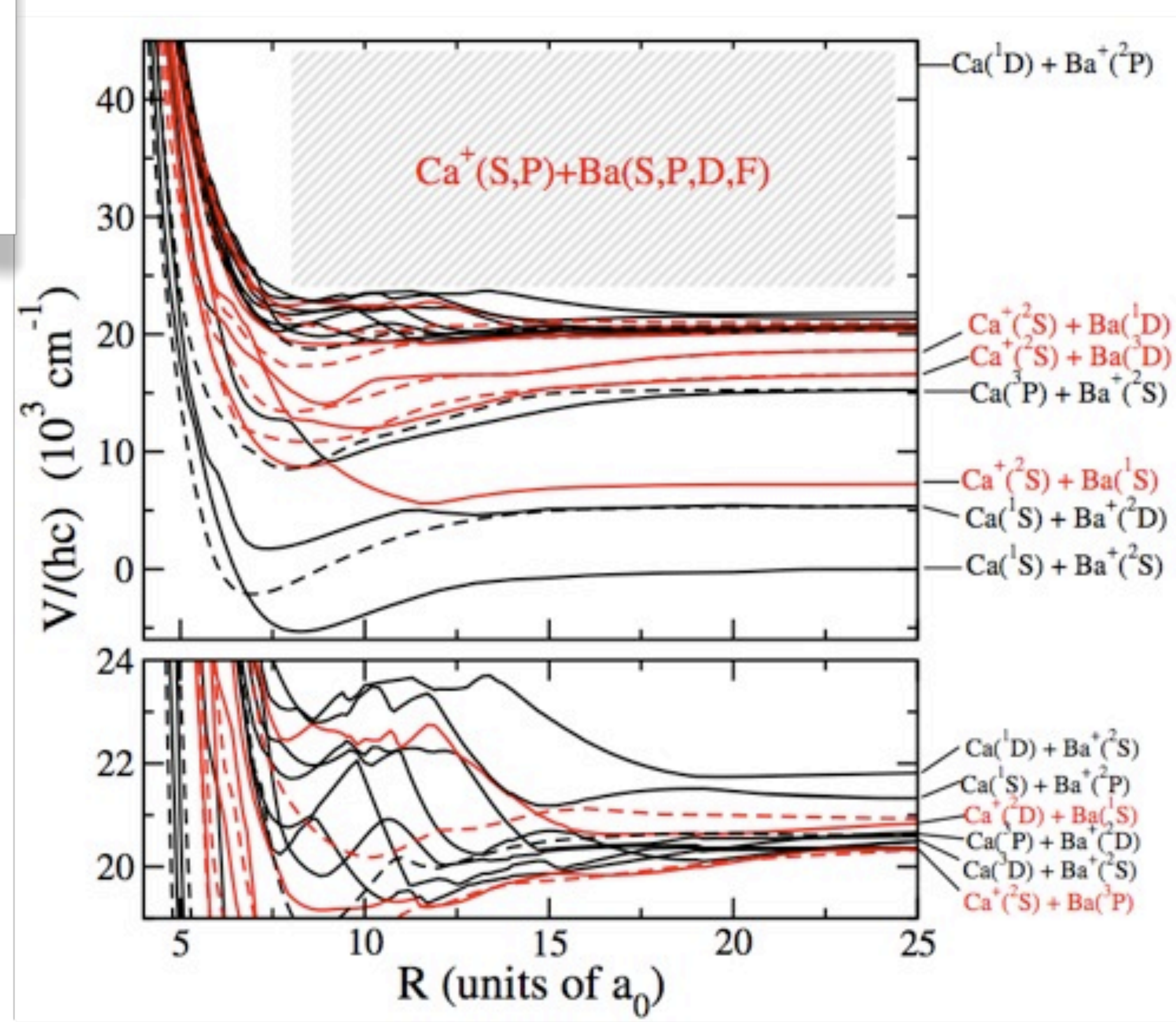

# • Some surprising chemistry

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- Short-lived atom excitation is unimportant
- Avoided Crossings (nRCT)
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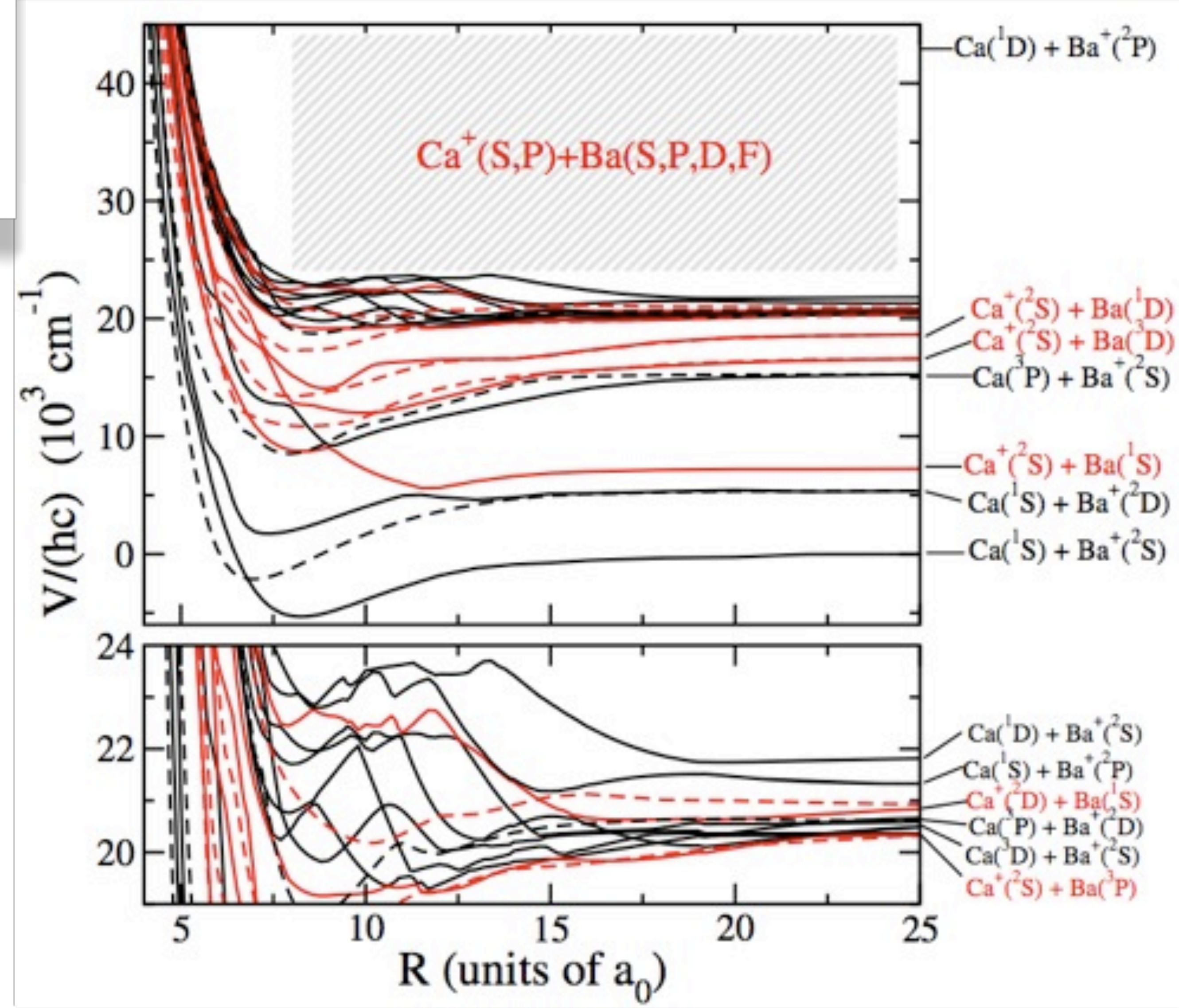

# • Some surprising chemistry

## $CaBa^+$ channel dependent rate constants

$$K = \rho_{pp}^{Ba}\rho_{ss}^{*Ca}K_{ps} + \rho_{ss}^{Ba}\rho_{dd}^{Ca}K_{sd} + \rho_{dd}^{Ba}\rho_{dd}^{Ca}K_{dd} + \rho_{pp}^{Ba}\rho_{dd}^{Ca}K_{pd}$$

TABLE I: Measured reaction rate constants.

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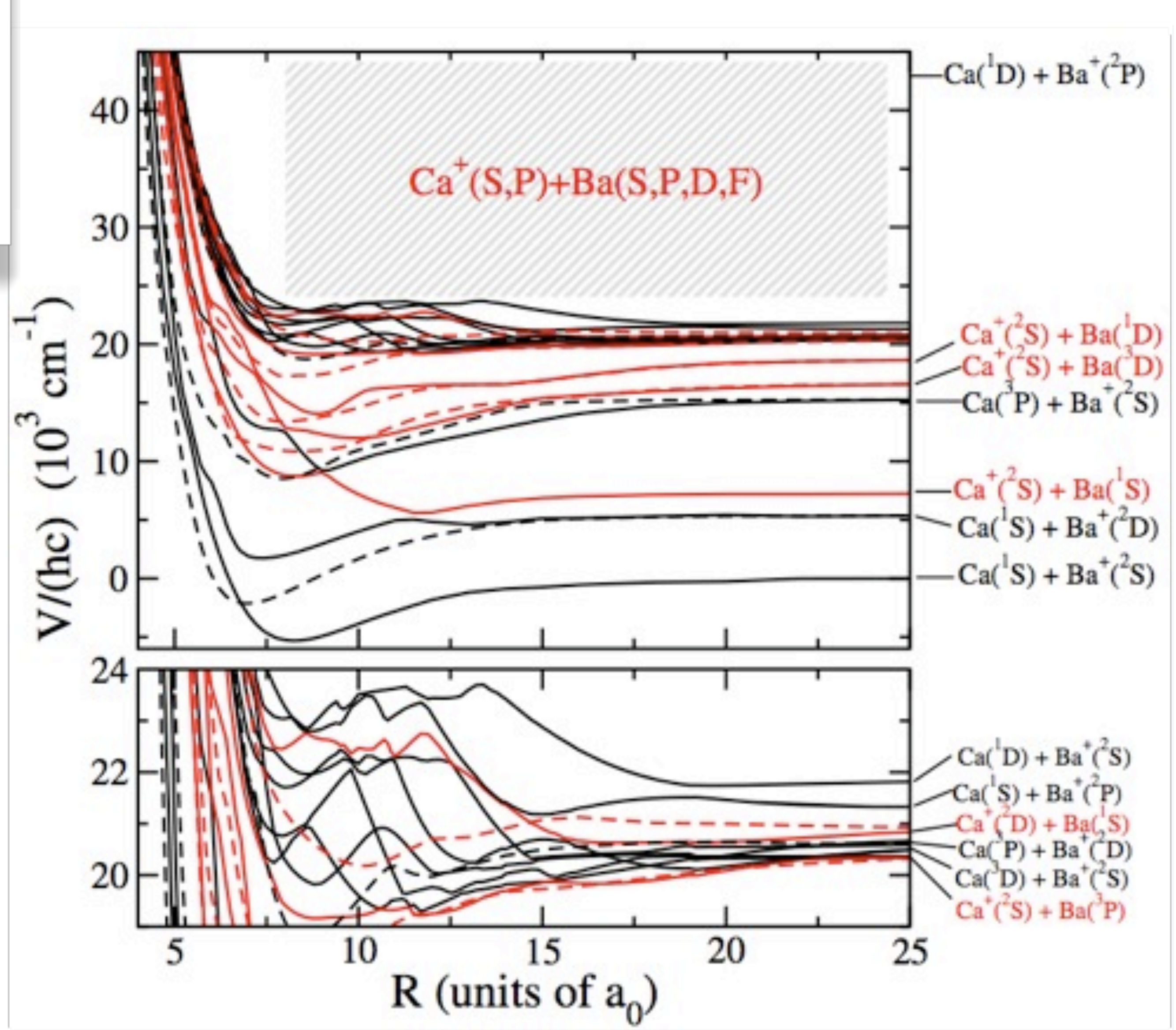
Sullivan *et al.*, Phys. Rev. Lett. **109,** 223002 (2012)
in collaboration with Svetlana Kotochigova



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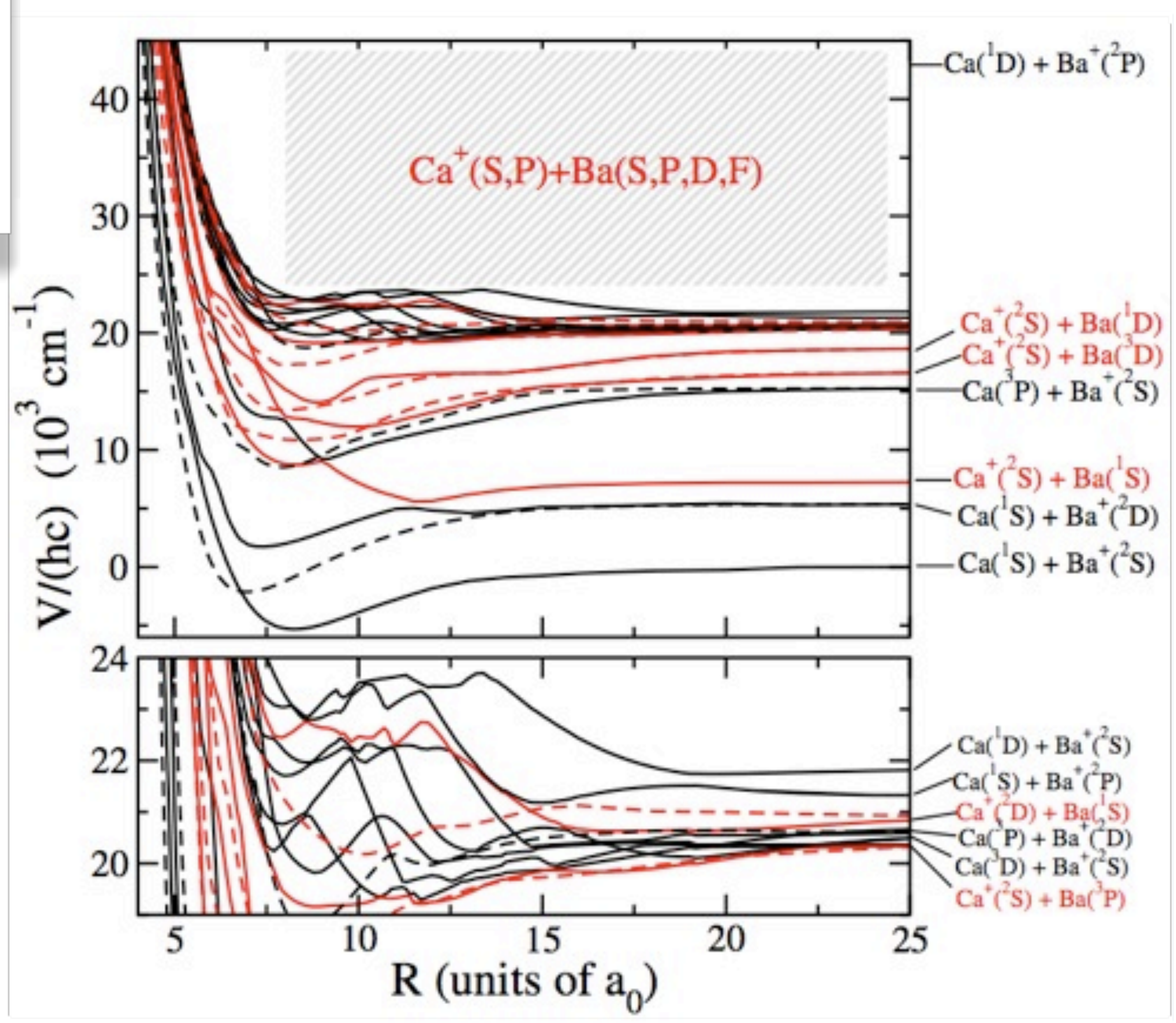


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<u>$Ca^+$ + Rb</u>

Hall *et al.*, Phys. Rev. Lett. **107**, 243202 (2011)

# Making ultracold molecular ions

## cooling methods

Traditional sympathetic cooling

- Atomic ions → Long range collisions

  (no internal state cooling)

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Traditional sympathetic cooling

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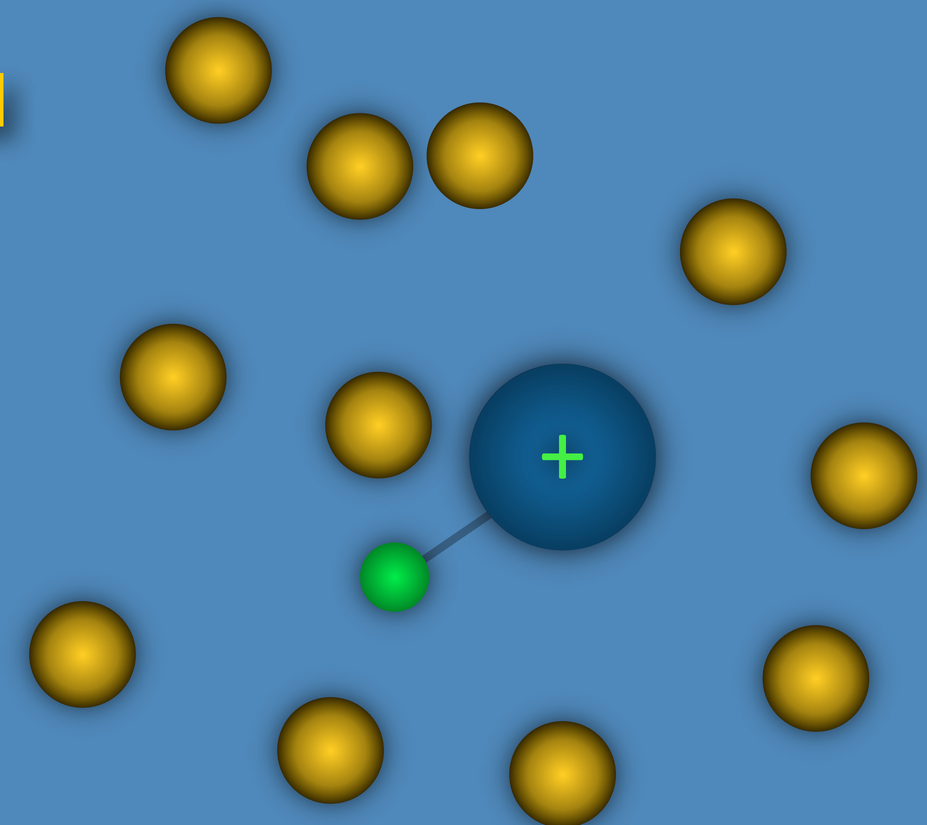

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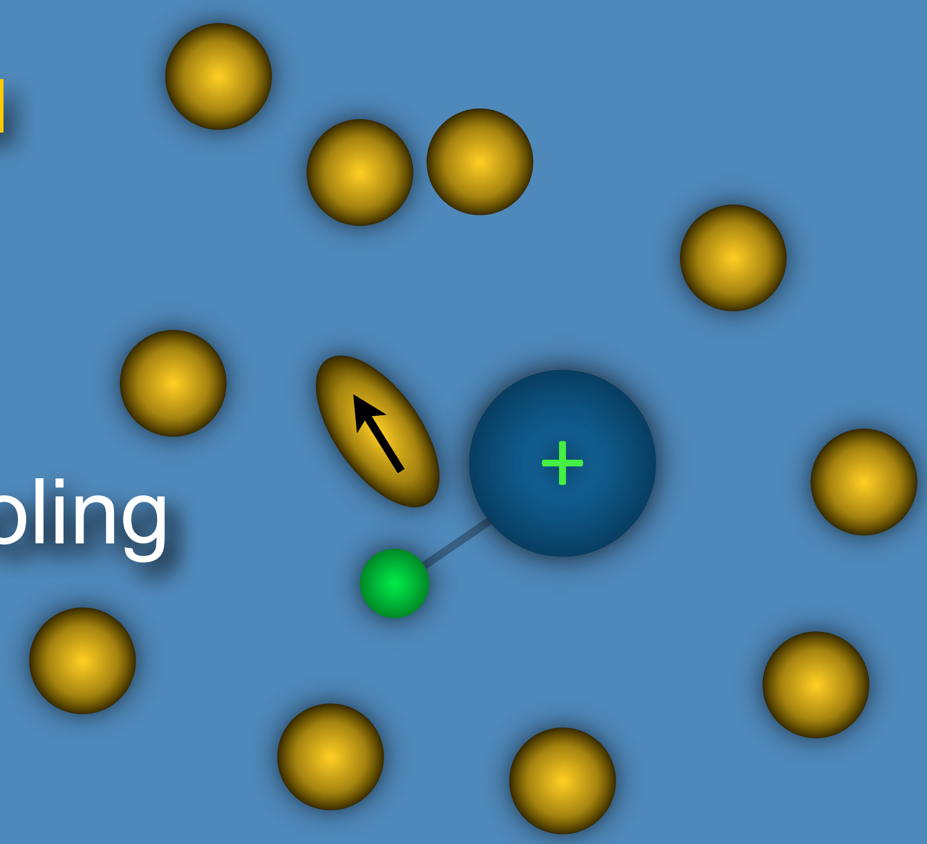

### Ultra-cold neutral atom sympathetic cooling

- Laser cooled atoms have high polarizability

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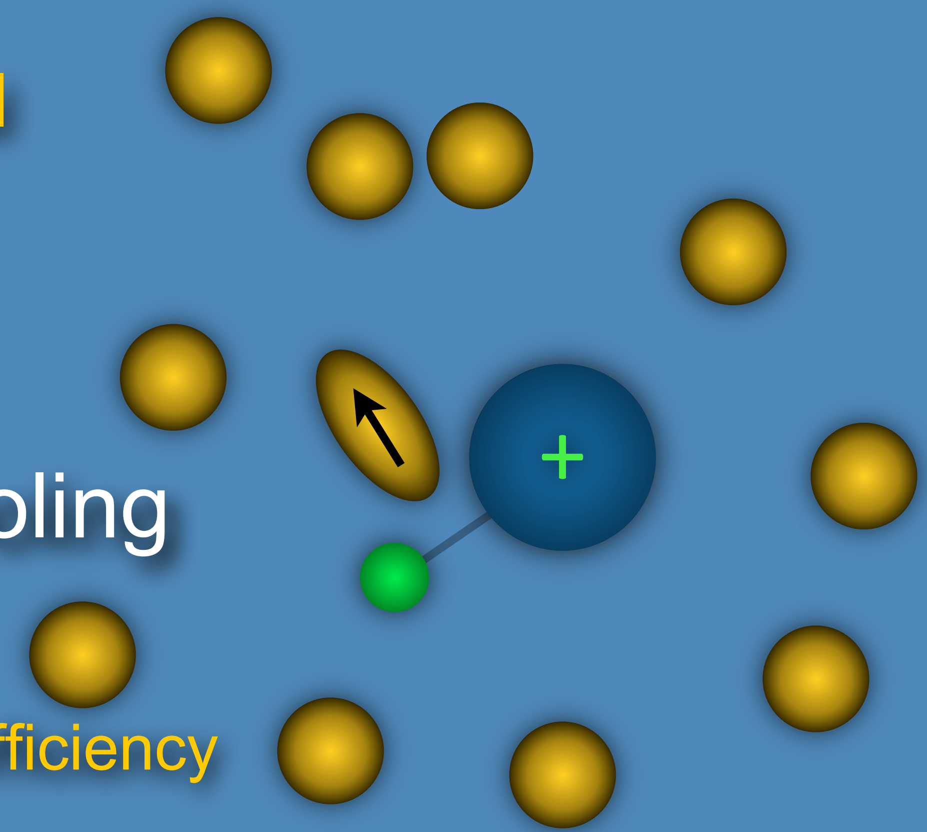

### Ultra-cold neutral atom sympathetic cooling

- Laser cooled atoms have high polarizability
  - $|\alpha_{Ca}/\alpha_{He}|^2 \approx 10^4$ → much higher relaxation efficiency
  - Simultaneous translation cooling

# • Making ultracold molecular ions

## Ingredients

## Ionization Potentials

| Atom | IP [eV] | λ [nm] | Molecule | IP [eV] | Molecule | IP [eV] | Molecule | IP [eV] | Molecule | IP [eV] | Molecule | IP [eV] |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Be | 9.3 | 235 | $Rb_2$ | 3.5 | $Li_2$ | 5.1 | $YH_2$ | 6.2 | TiS | 7.1 | NaI | 7.64 |
| Mg | 7.6 | 285 | $Cs_2$ | 3.6 | SrF | 5.3 | HoF | 6.2 | GeCl | 7.2 | RbBr | 7.7 |
| Yb | 6.3 | 399 | $K_2$ | 4.1 | SrCl | 5.5 | $Ti_2$ | 6.3 | CsI | 7.25 | $Pd_2$ | 7.7 |
| Ca | 6.1 | 423 | KNa | 4.4 | SrBr | 5.5 | US | 6.3 | SiF | 7.26 | $C_6H_7N$ | 7.7 |
| Sr | 5.7 | 461 | KLi | 4.6 | SrI | 5.5 | ErF | 6.3 | GeBr | 7.3 | (Aniline) | |
| Li | 5.4 | 671 | BaI | 4.7 | CaBr | 5.6 | TiO | 6.4 | SnF | 7.4 | LiD | 7.7 |
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| K | 4.3 | 771 | SrF | 4.9 | CaF | 5.8 | CaO | 6.5 | $Mn_2$ | 7.4 | PbBr | 7.8 |
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| | | | BaF | 4.9 | CaCl | 6 | TiO | 6.8 | MgCl | 7.5 | MgF | 7.8 |
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| | | | $Na_2$ | 4.9 | TaO | 6 | UN | 7 | PbCl | 7.5 | (Trimethylamine ) | |
| | | | LaO | 4.9 | TiH | 6 | InS | 7 | $N(CH_2CH_3)_3$ | 7.5 | LiH | 7.85 |
| | | | BaCl | 5.0 | SrO | 6.1 | $ZrH_2$ | 7 | (Triethylamine) | | $Cu_2$ | 7.89 |
| | | | LiNa | 5.0 | ZrO | 6.1 | InSe | 7.1 | HfO | 7.5 | PtSi | 7.9 |
| | | | VO | 5.0 | CaI | 6.1 | TiS | 7.1 | GeF | 7.5 | $Ge_2$ | 7.9 |
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| | | | LiNa | 5.0 | ZrO | 6.1 | InSe | 7.1 | HfO | 7.5 | PtSi | 7.9 |
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| | | | PrO | 4.9 | Dy[illegible] | [illegible] | [illegible] | [illegible] | [illegible] | [illegible] | [illegible] | 7.82 |
| | | | $Na_2$ | 4.9 | Ta[illegible] | [illegible] | [illegible] | [illegible] | [illegible] | [illegible] | [illegible] | |
| | | | LaO | 4.9 | Ti[illegible] | [illegible] | [illegible] | [illegible] | [illegible] | [illegible] | [illegible] | 7.85 |
| | | | BaCl | 5.0 | Sr[illegible] | [illegible] | [illegible] | [illegible] | [illegible] | [illegible] | [illegible] | 7.89 |
| | | | LiNa | 5.0 | Zr[illegible] | [illegible] | [illegible] | [illegible] | [illegible] | [illegible] | [illegible] | 7.9 |
| | | | VO | 5.0 | Ca[illegible] | [illegible] | [illegible] | [illegible] | [illegible] | [illegible] | [illegible] | 7.9 |
| | | | NdO | 5.0 | UO[illegible] | [illegible] | [illegible] | [illegible] | [illegible] | [illegible] | [illegible] | 7.9 |

- $BaCl^+$ produced via laser ablation of $BaCl_2$
- Simple electronic structure
- Appears to have UV transitions (rather than VUV)
- Has rotational transitions in the  1-10 GHz

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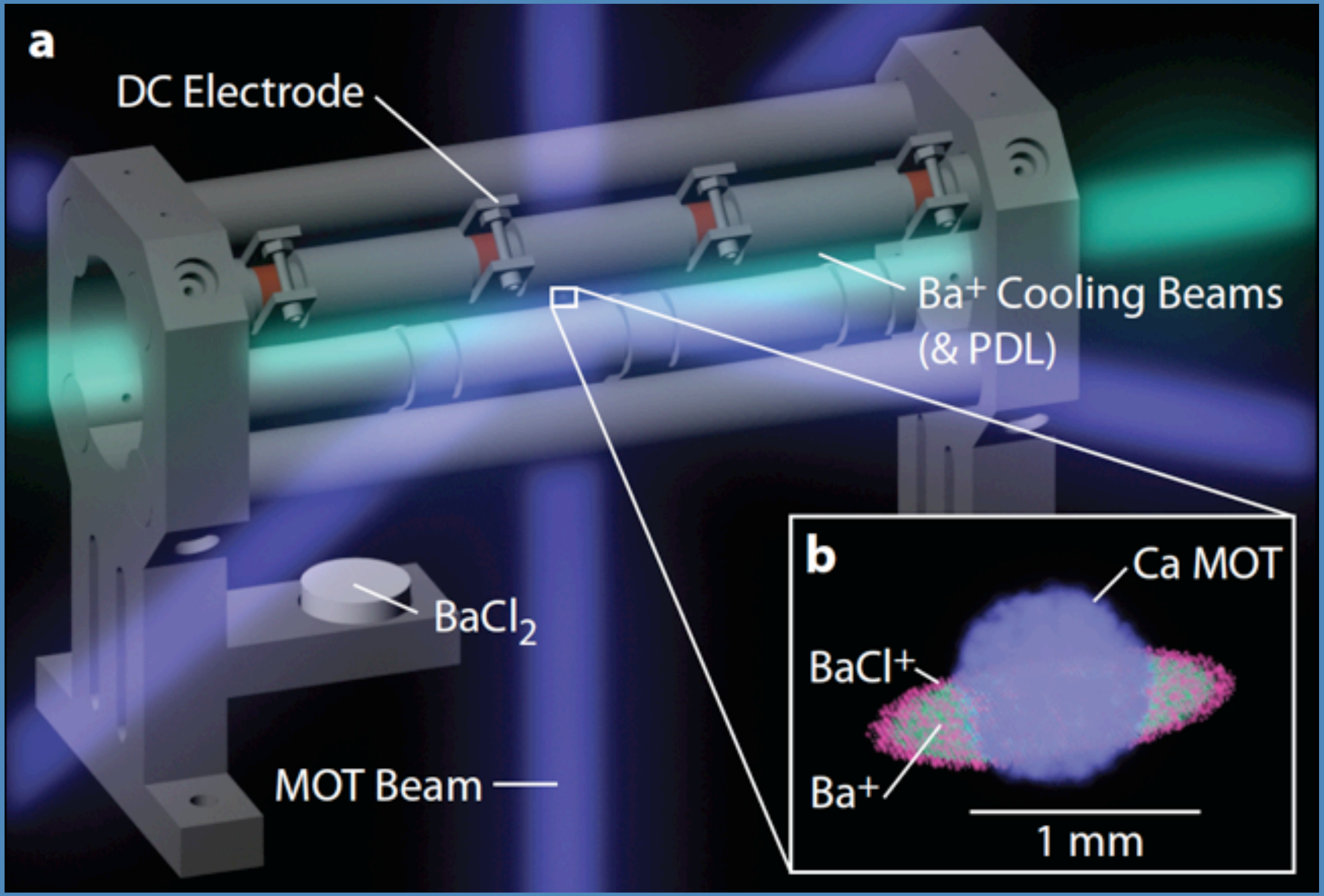
# Experimental arrangement




$Ba^+$ ions are used for imaging and also cool the $BaCl^+$ motion

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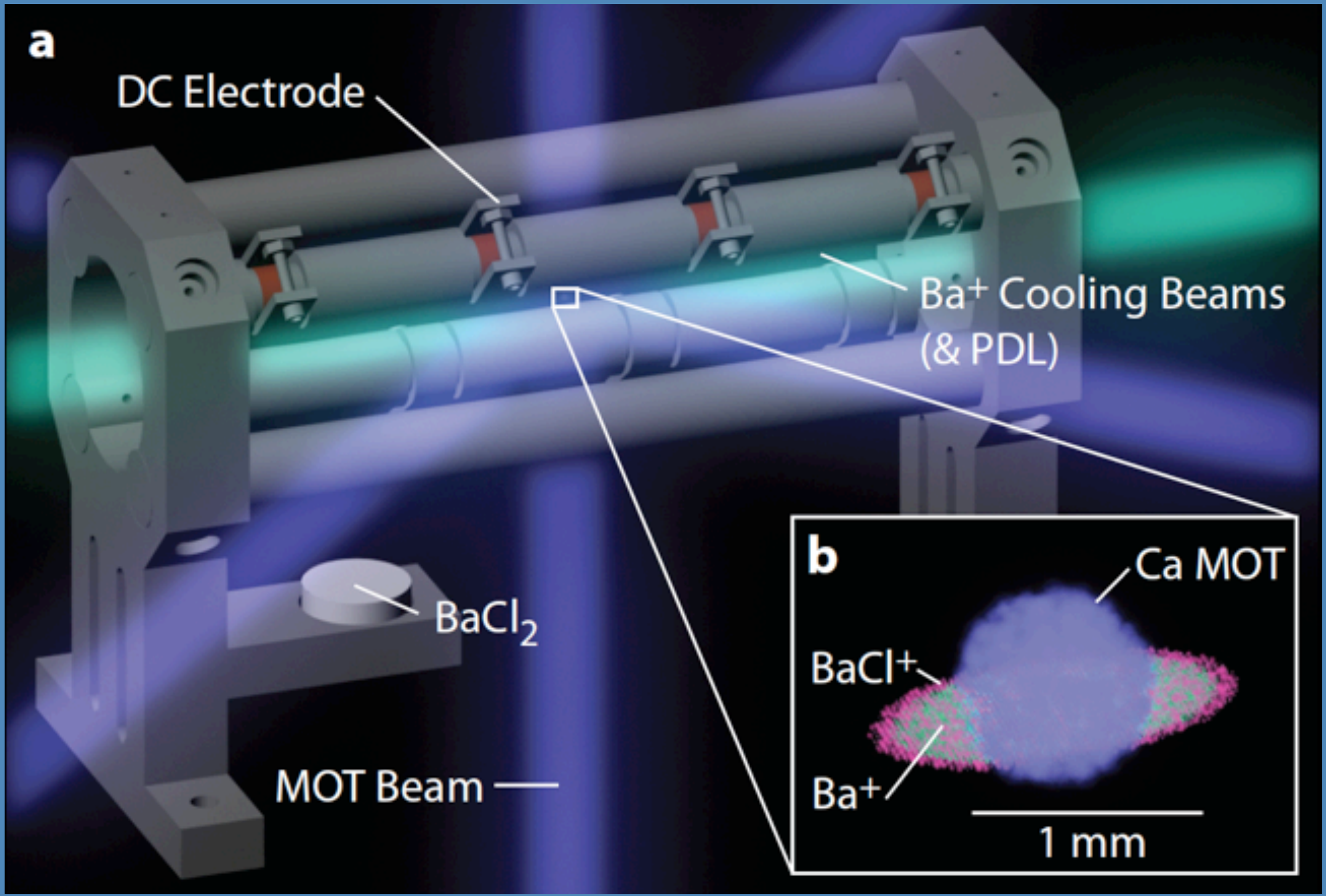


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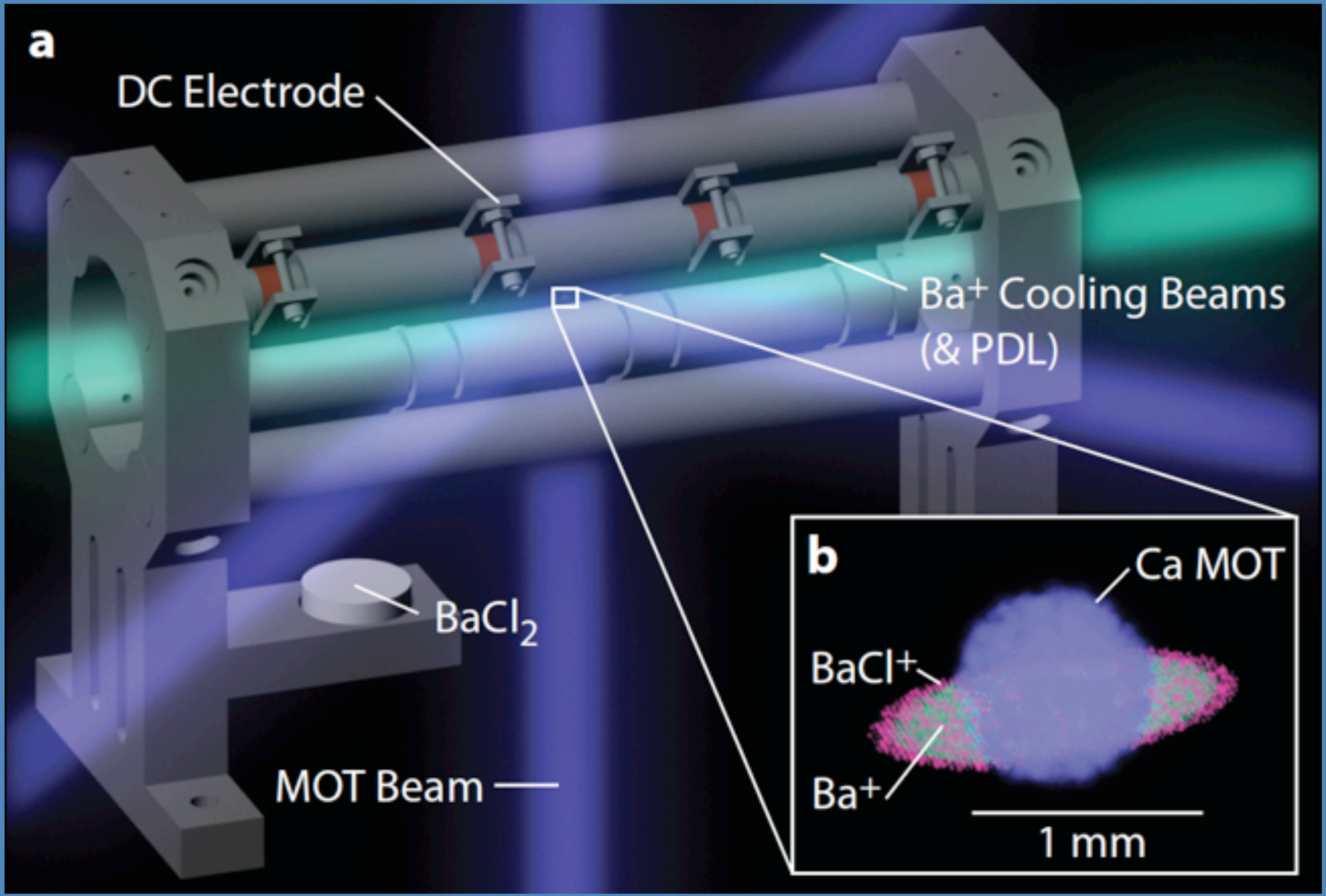


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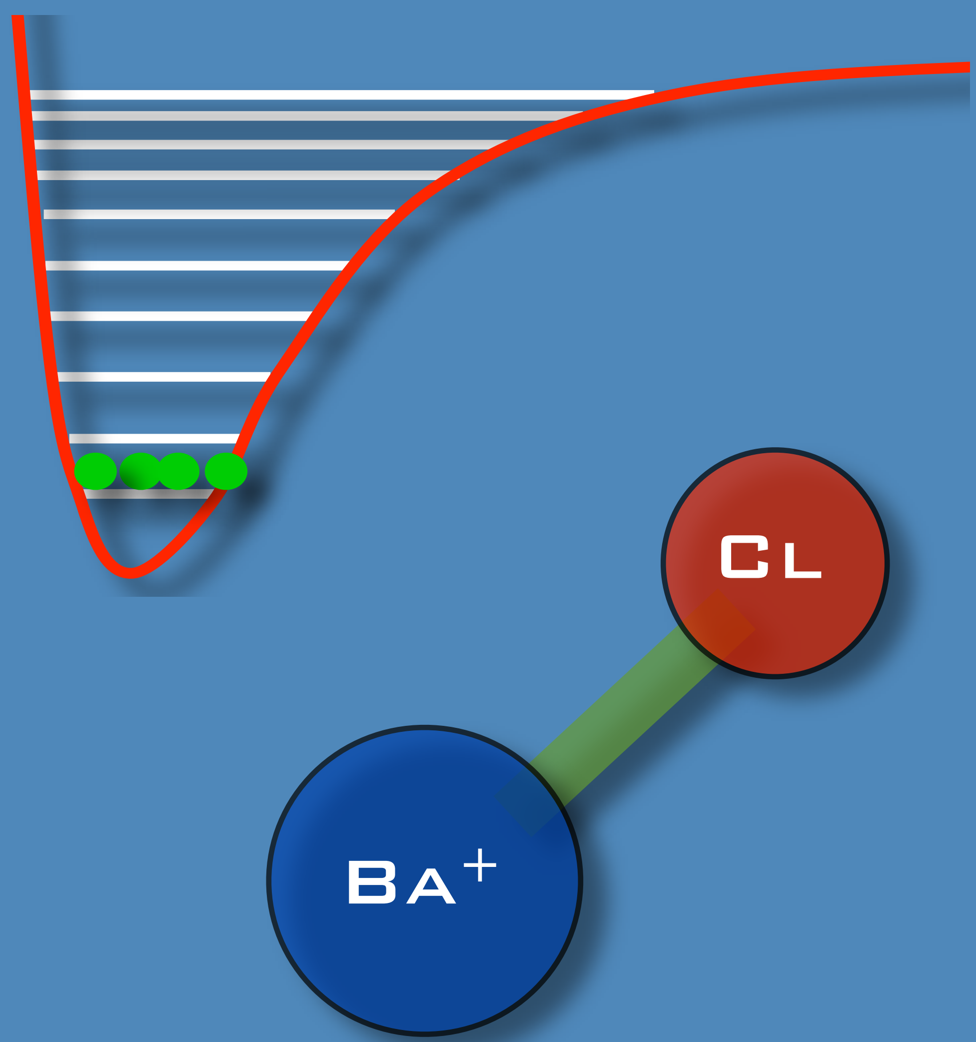
Does it work?

✓ Translationally cold

? Internal states

# • cold Molecular ions

## 1st step: Spectroscopy




25

# cold Molecular ions

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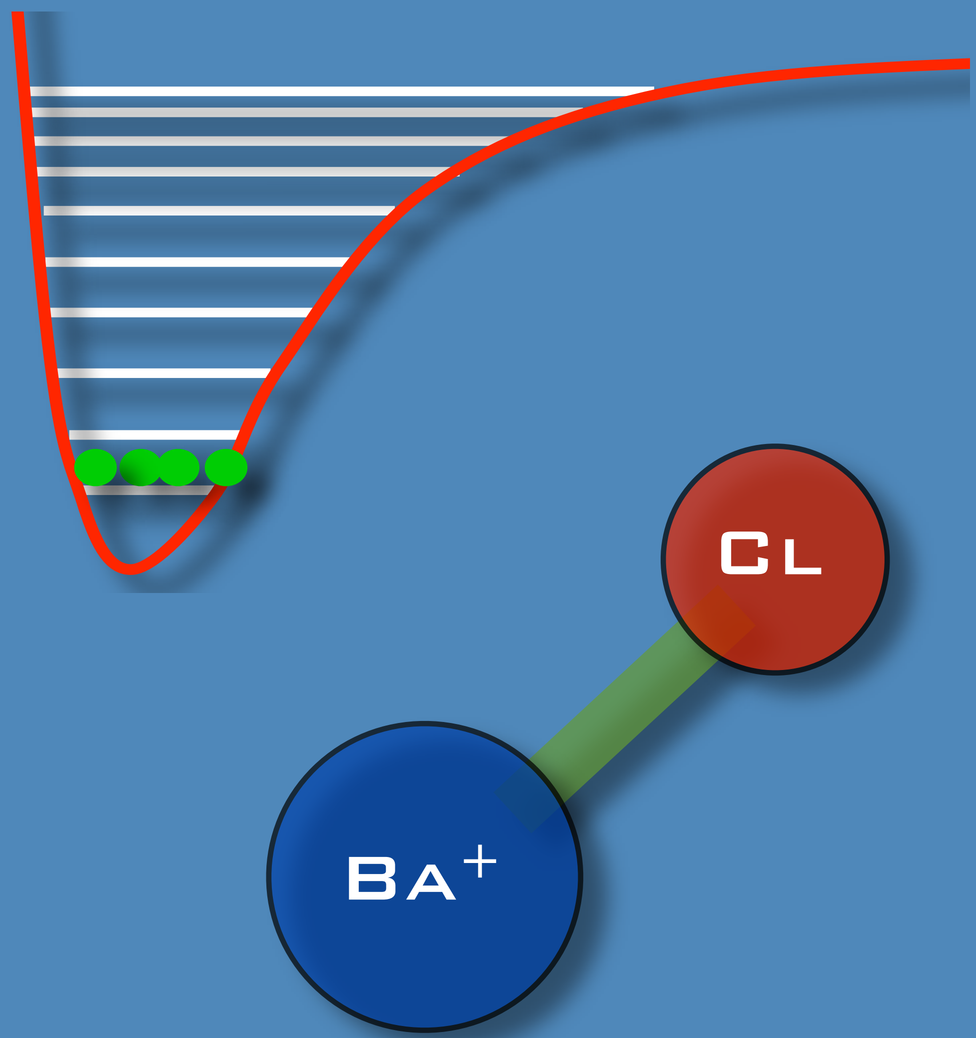


What we knew:

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25

# • Cold Molecular Ions

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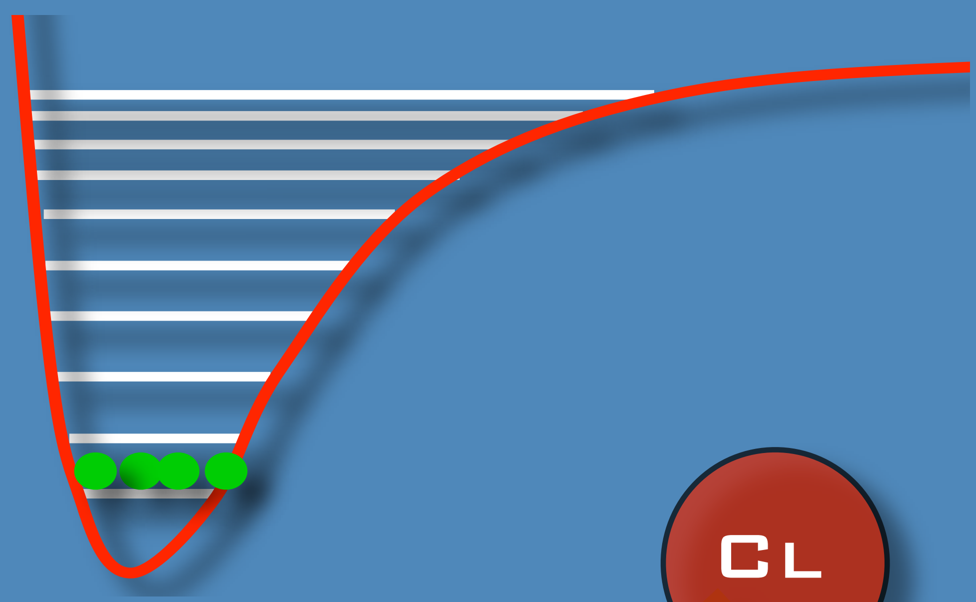

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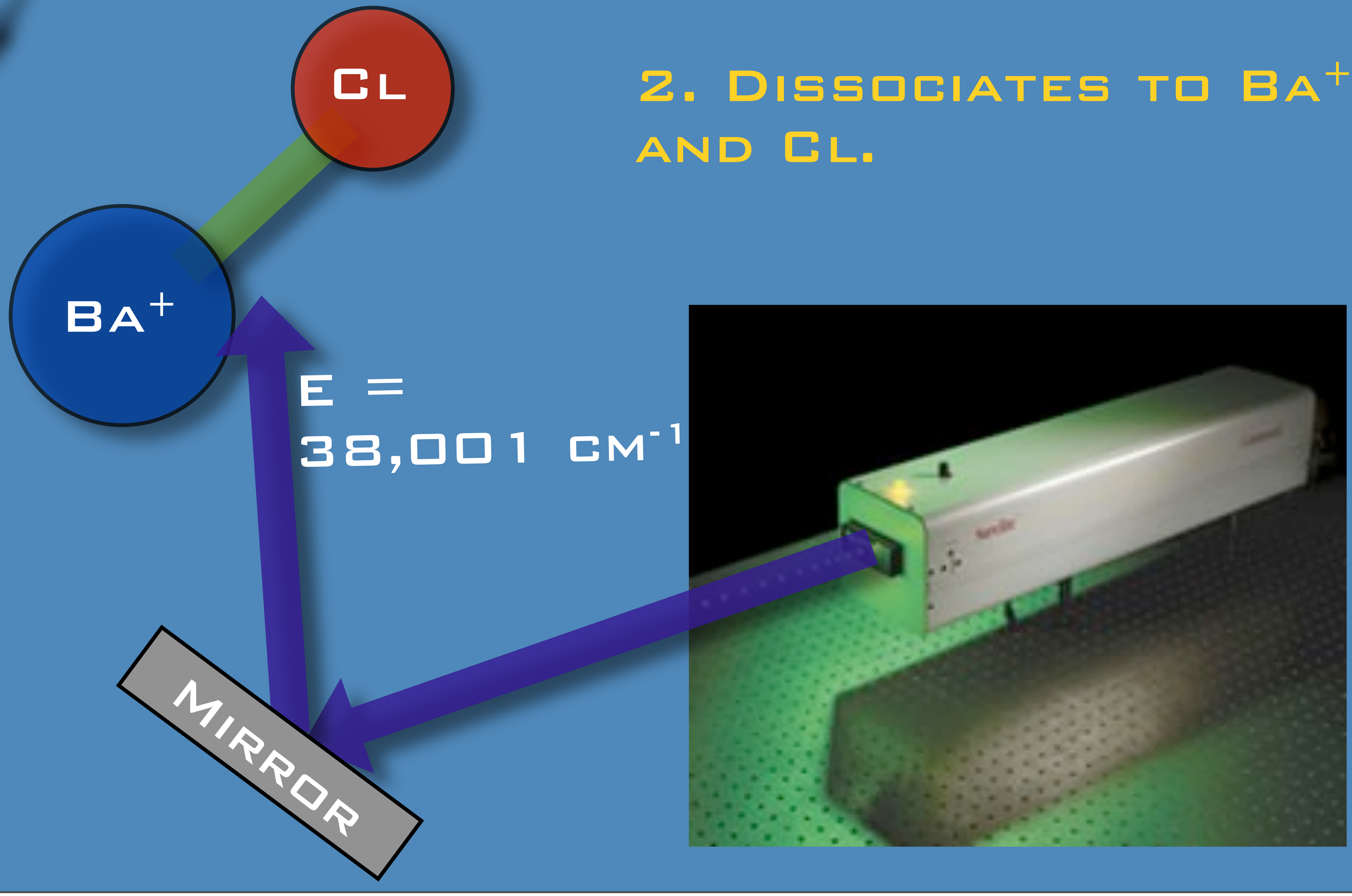
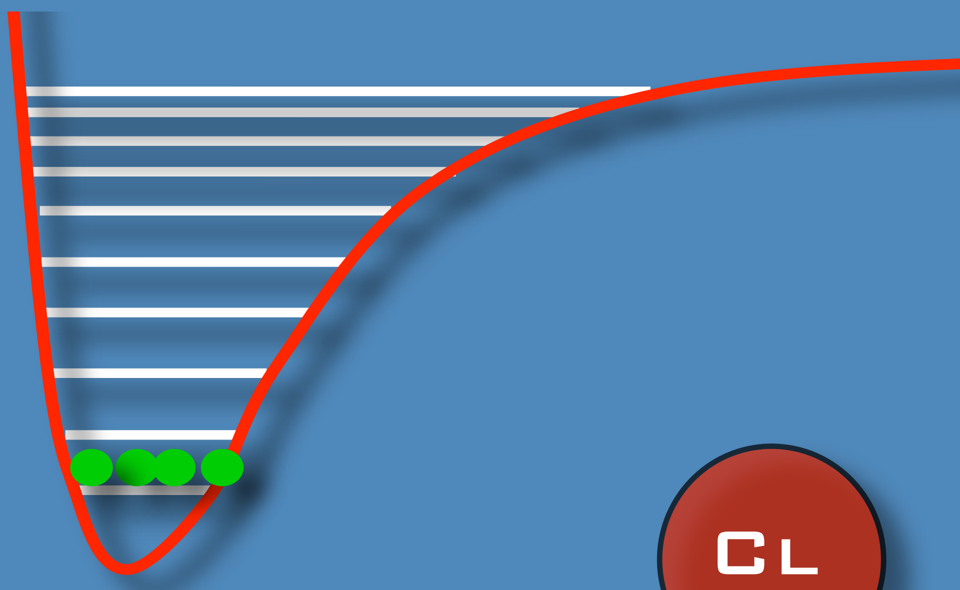


# • cold Molecular ions

## 1st step: Spectroscopy



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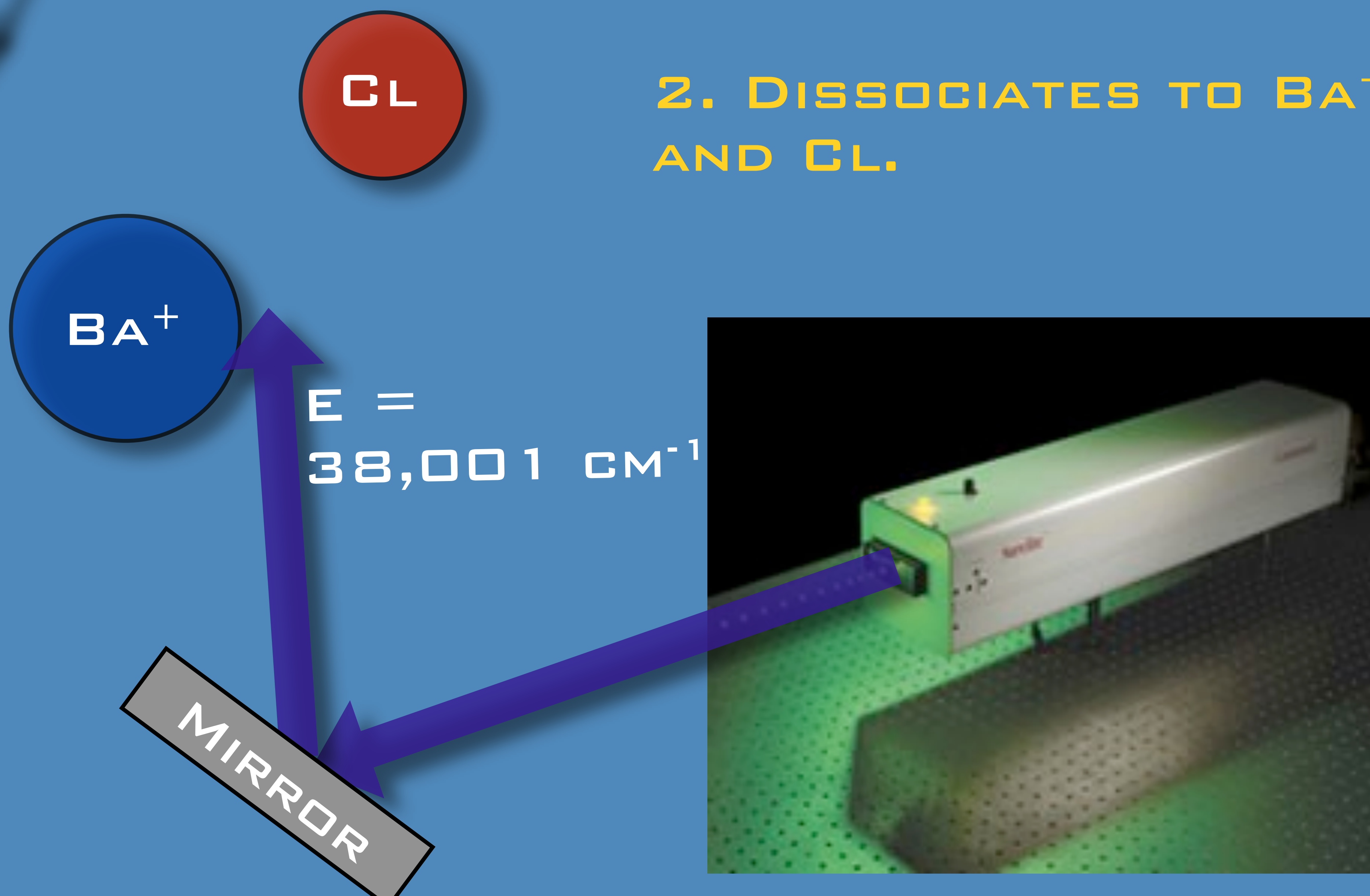




25

# cold Molecular ions

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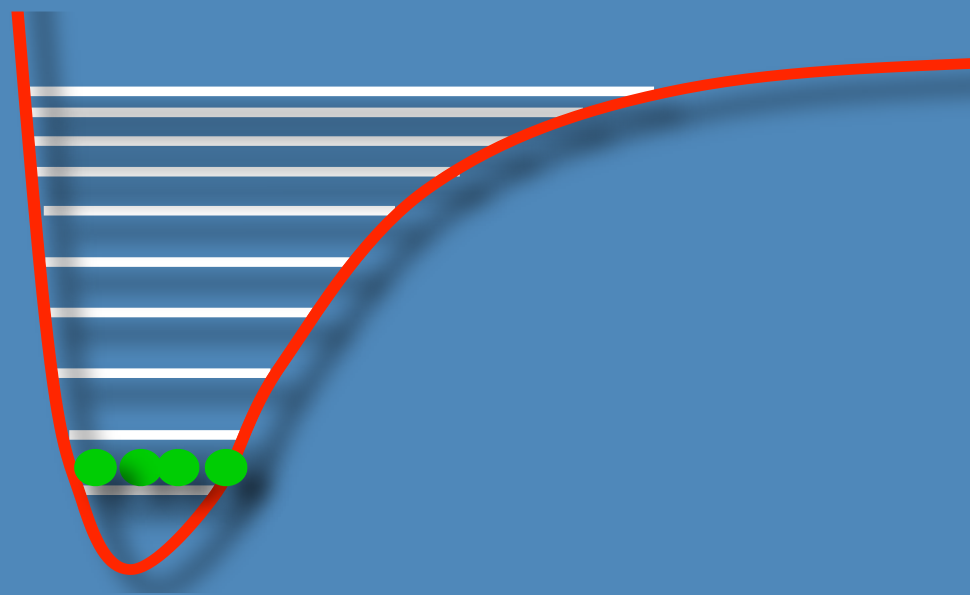

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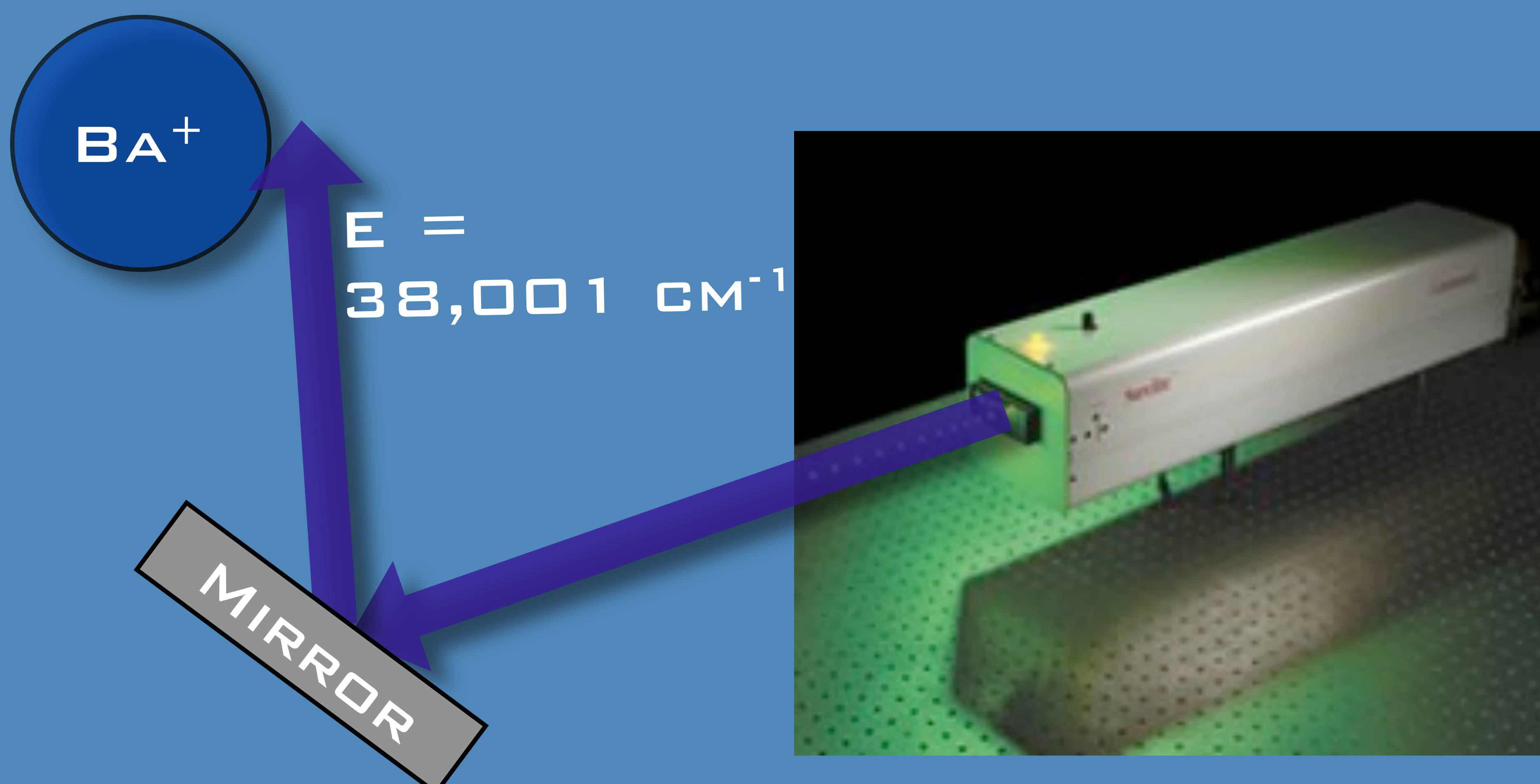
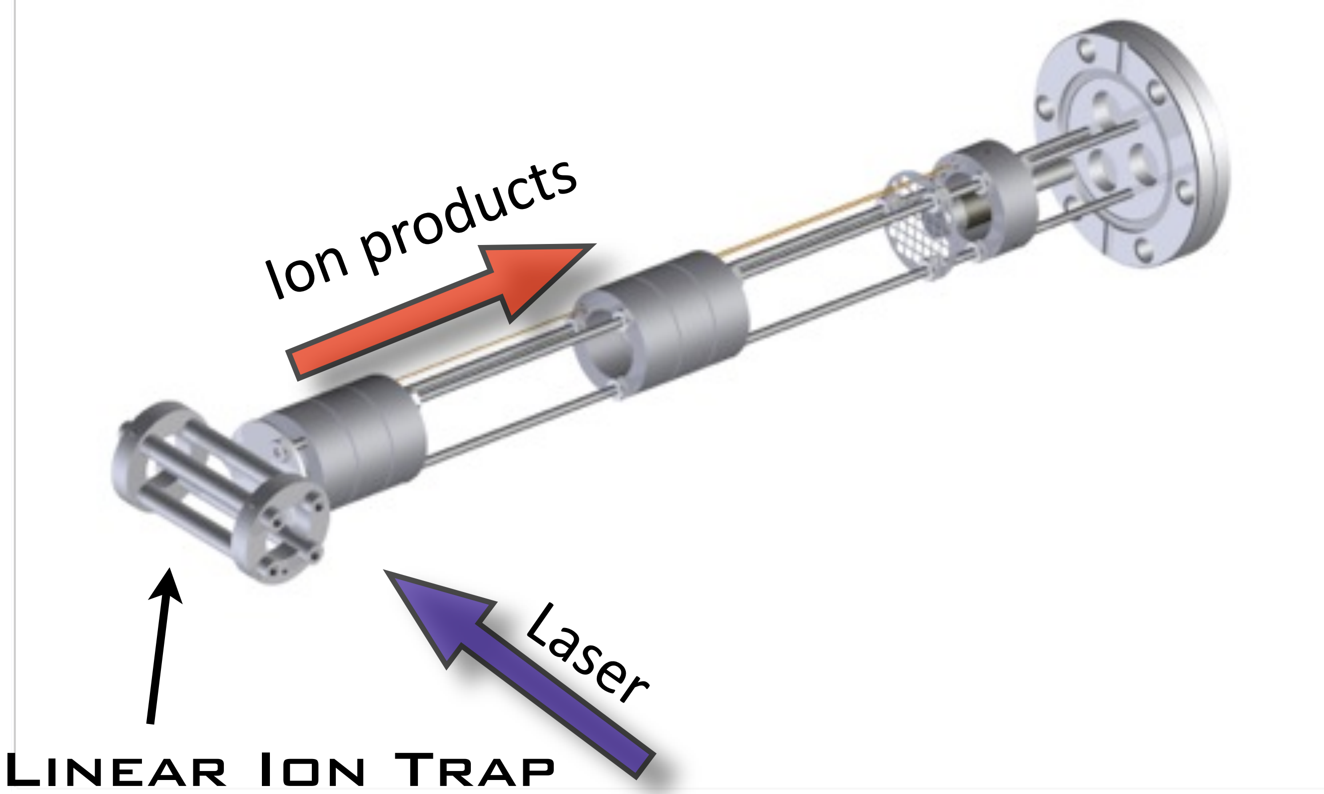


# • Cold Molecular ions

## 1st step: Spectroscopy

### Radial Ejection: TOF-MS




**S. J. Schowalter et al., Rev. Sci. Instrum., 83 043103 (2012)**

# • Cold Molecular ions

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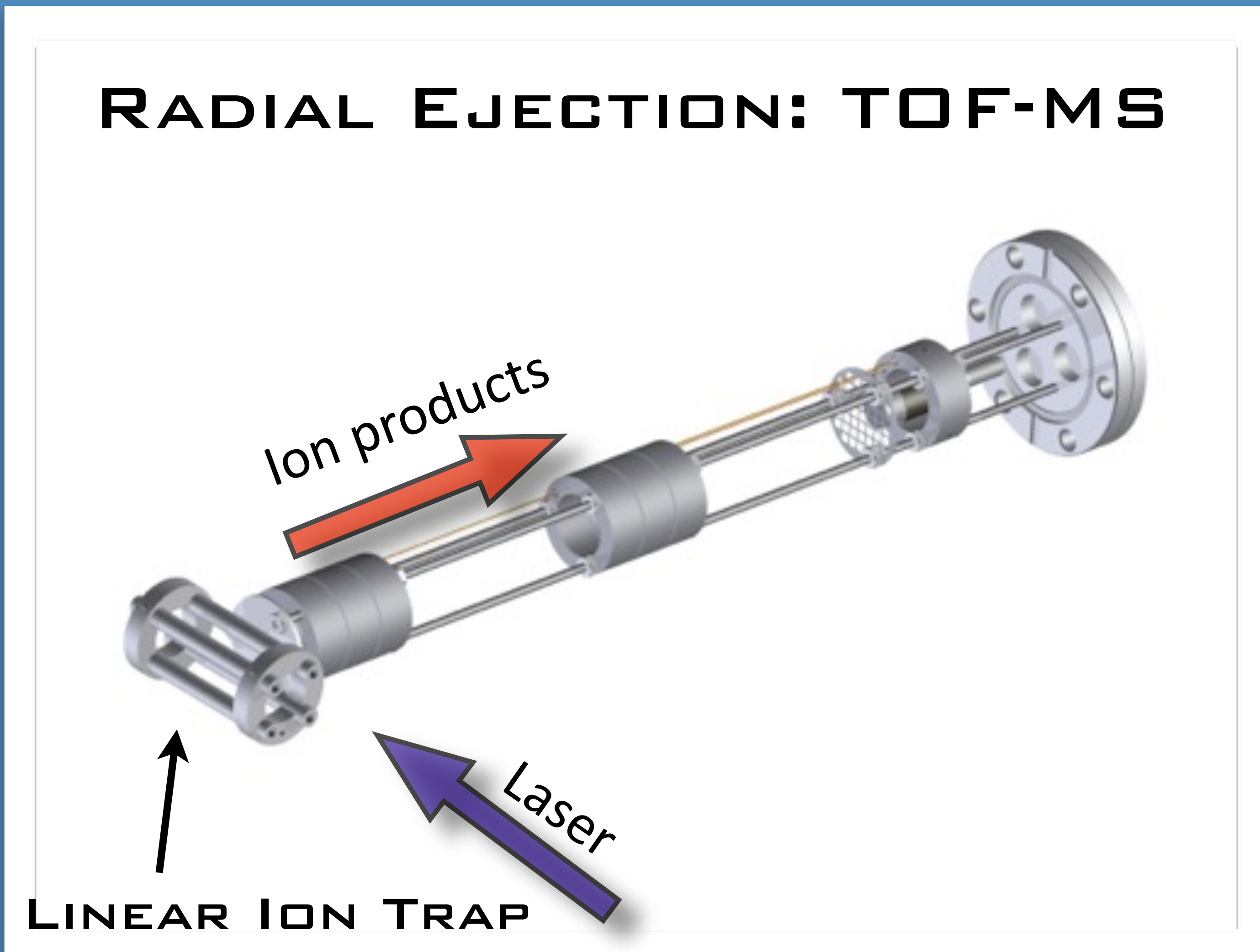


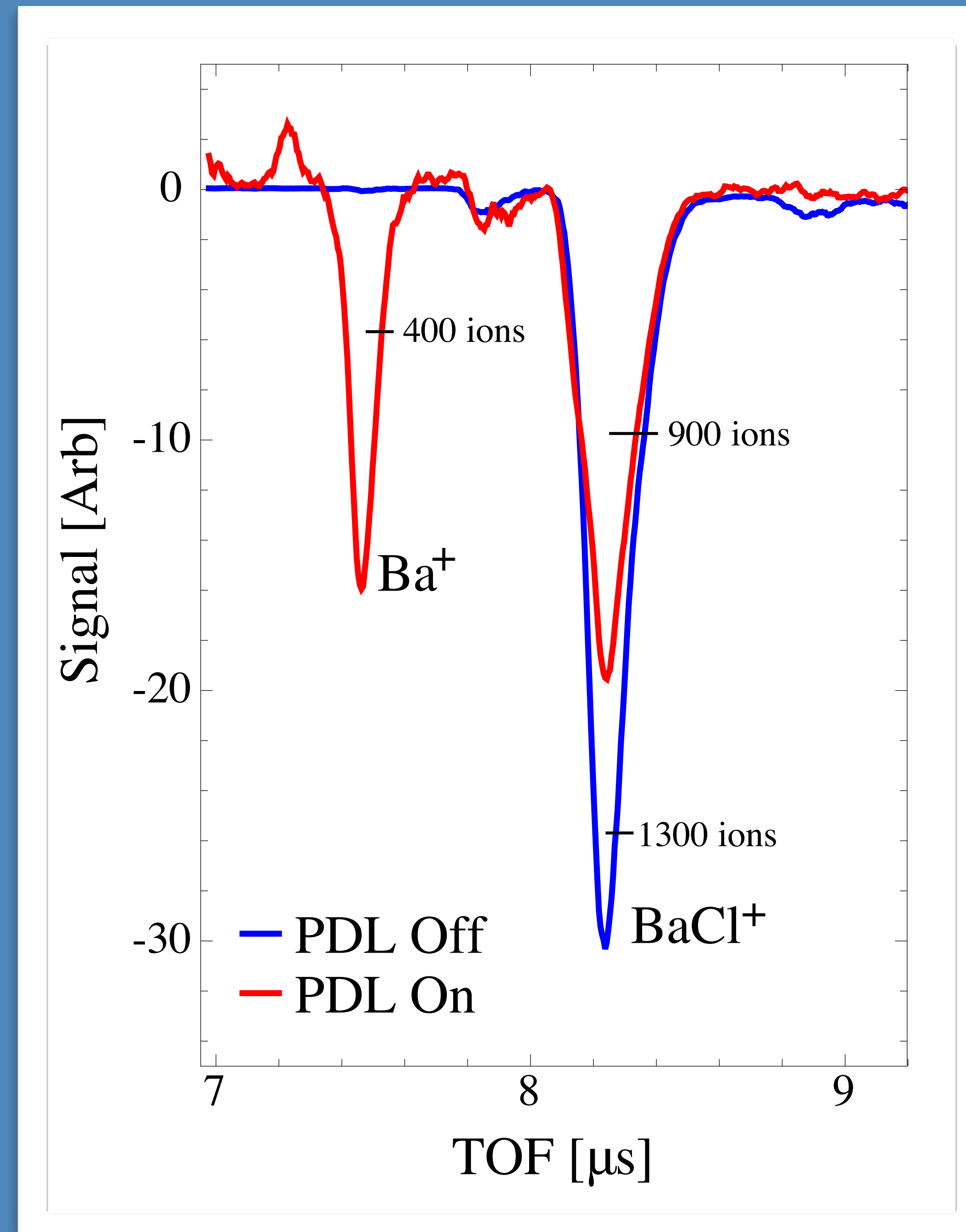


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# • Cold Molecular ions

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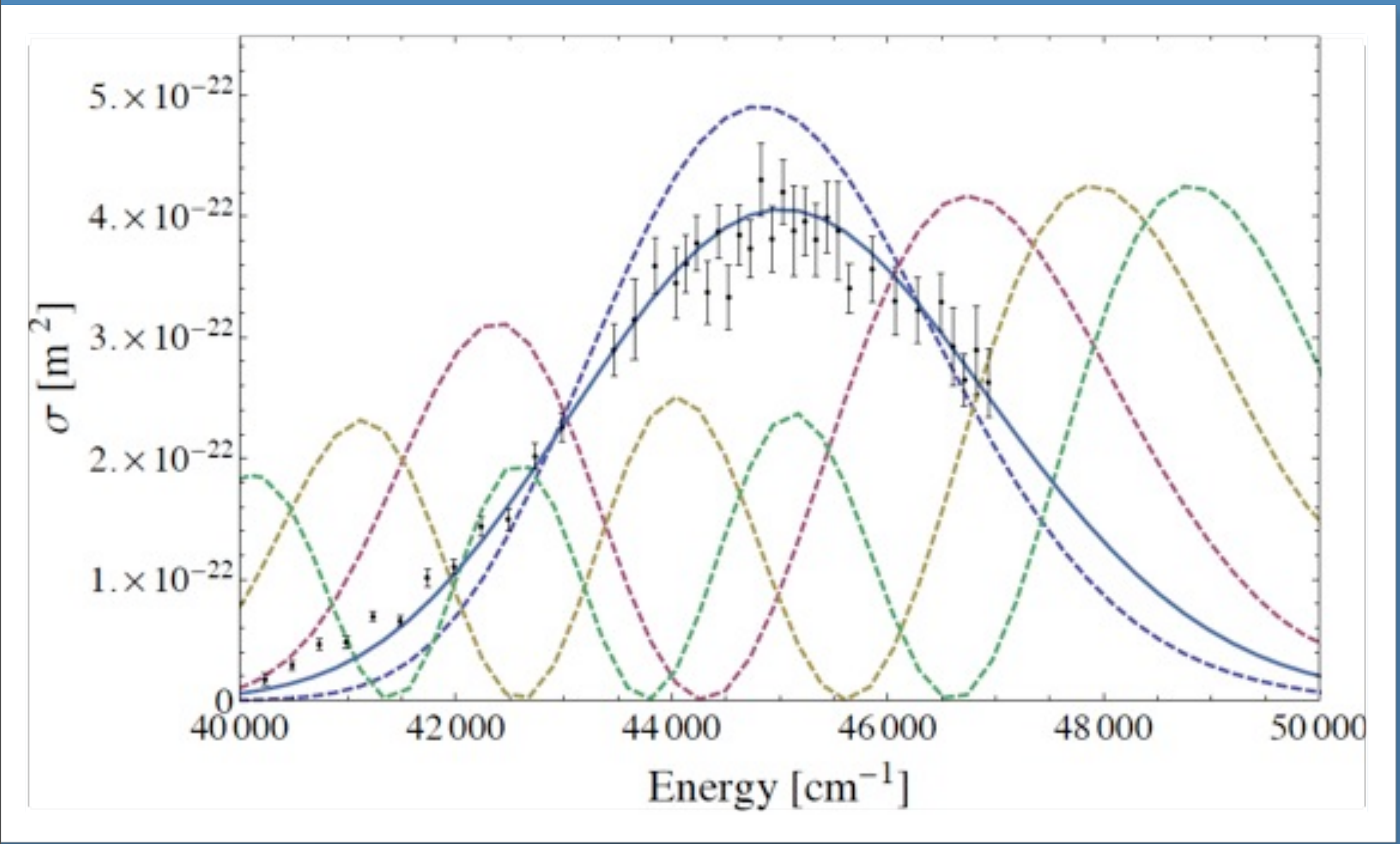
### Photodissociation Cross-section

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# Cold Molecular ions

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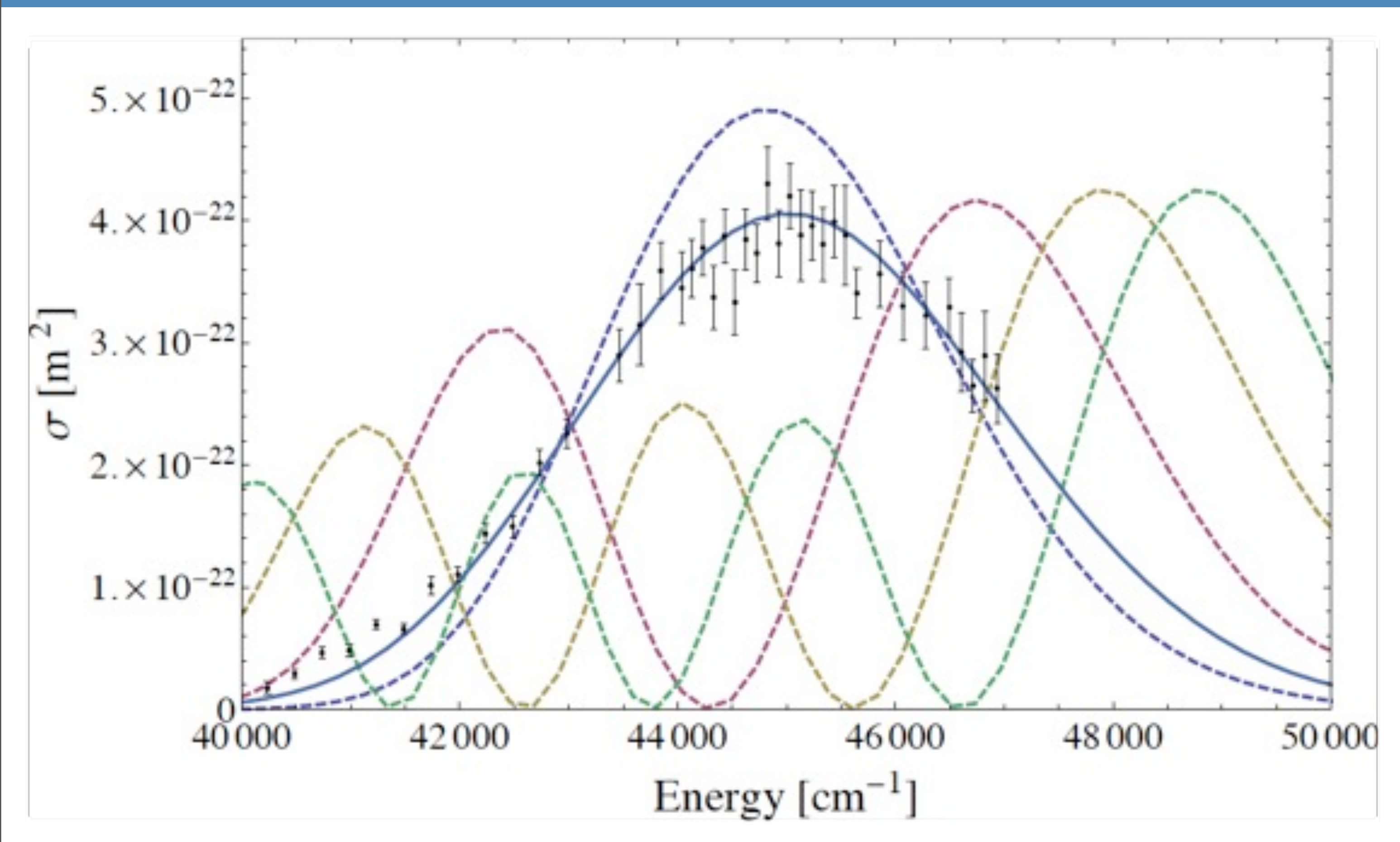


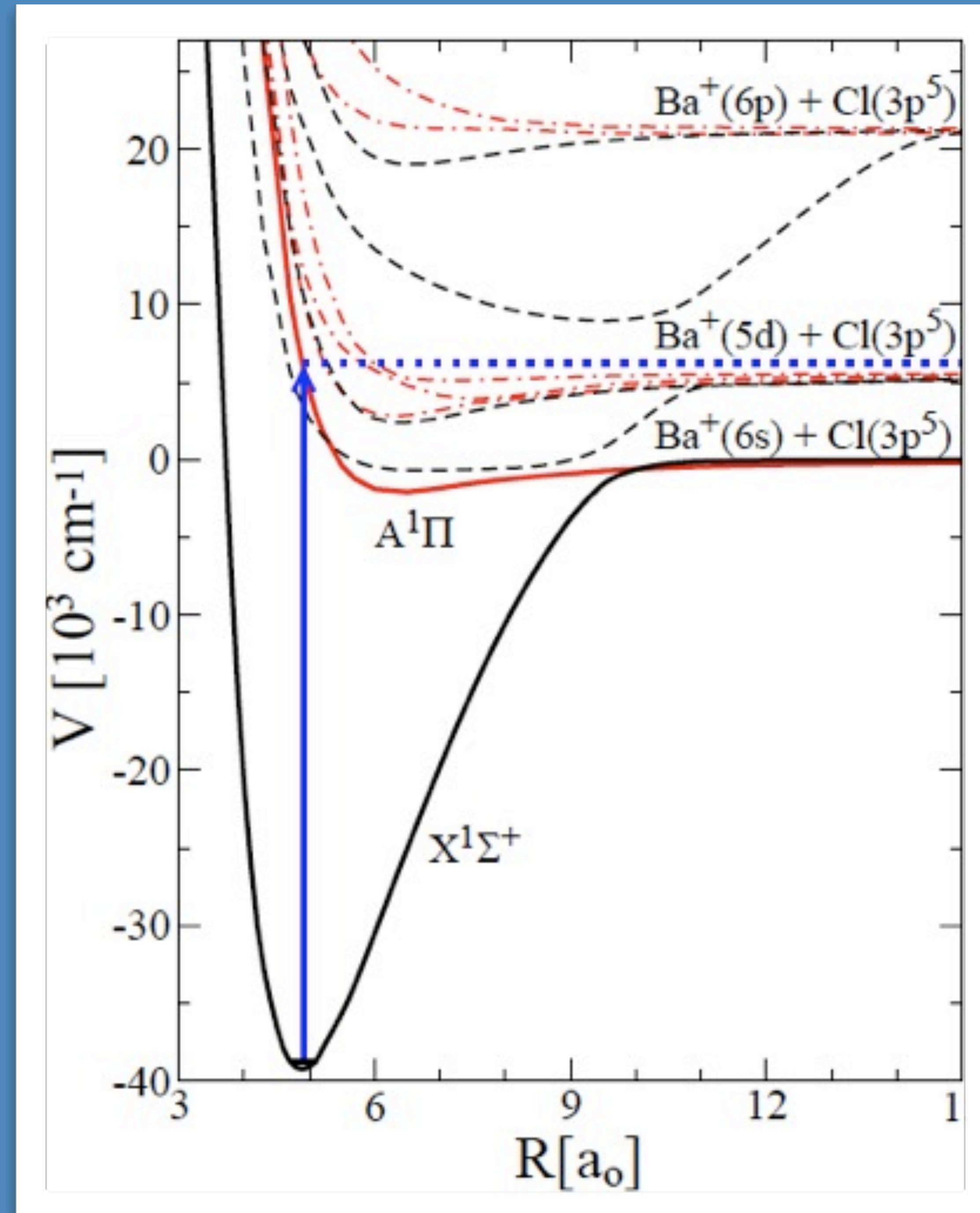


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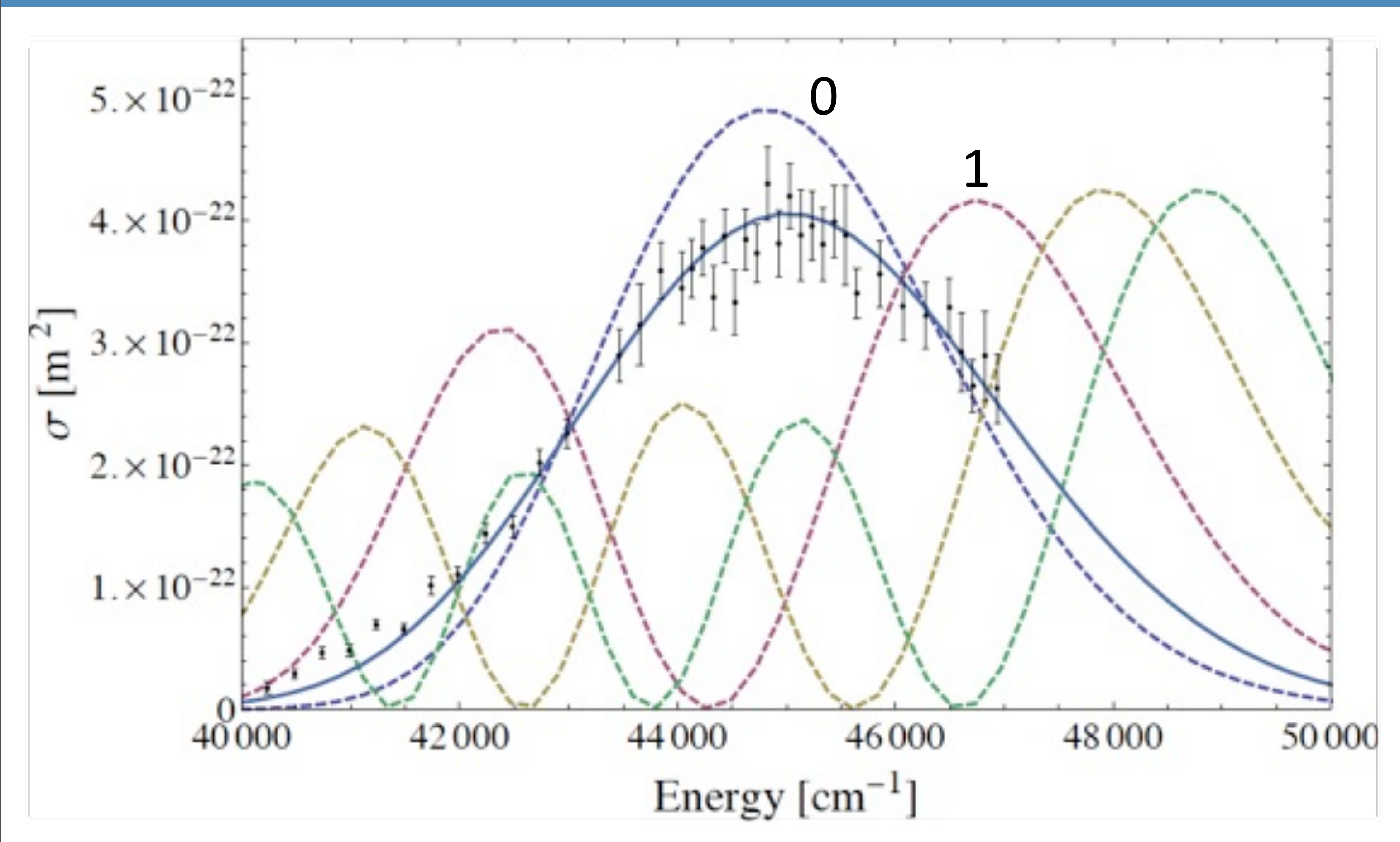


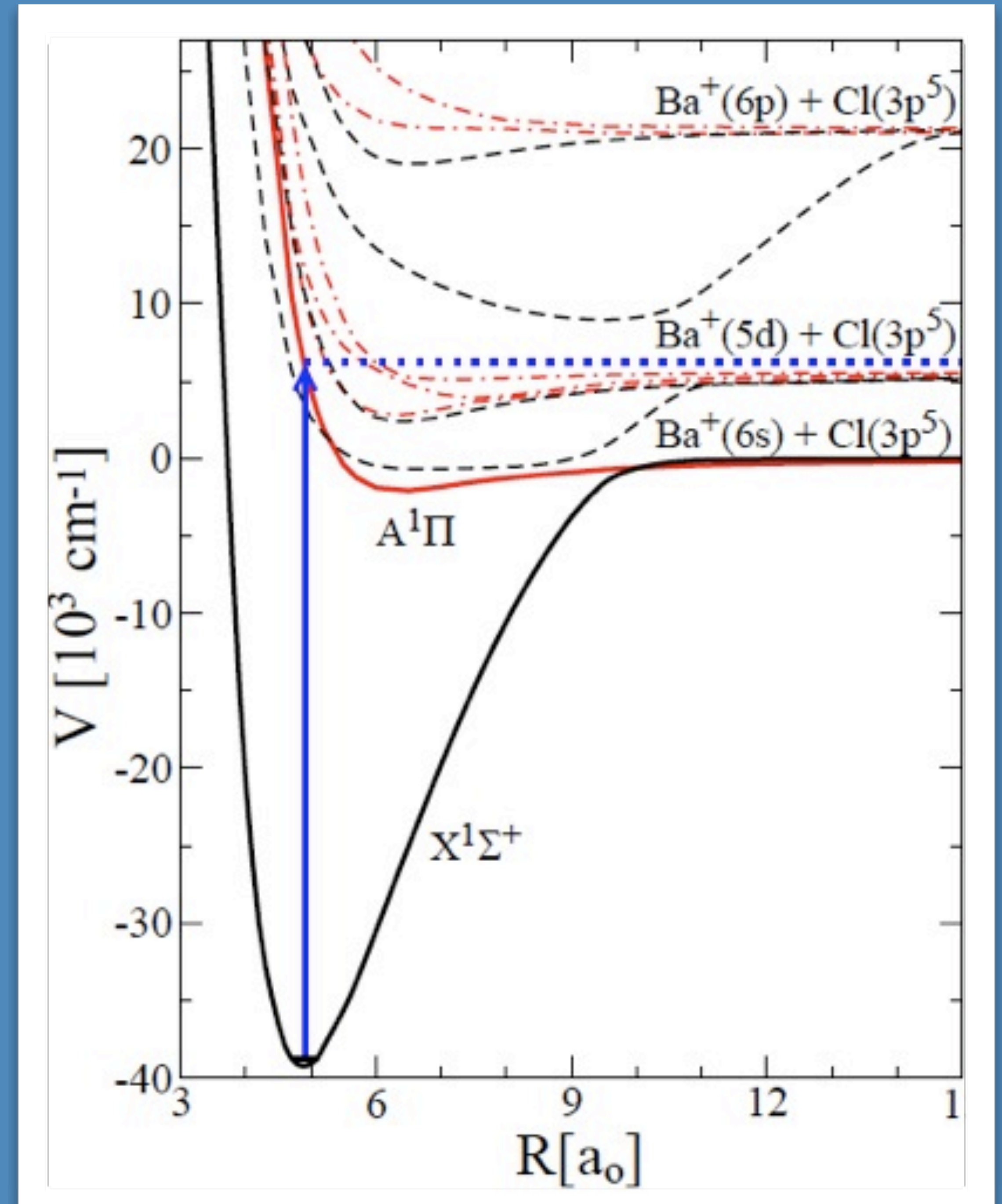


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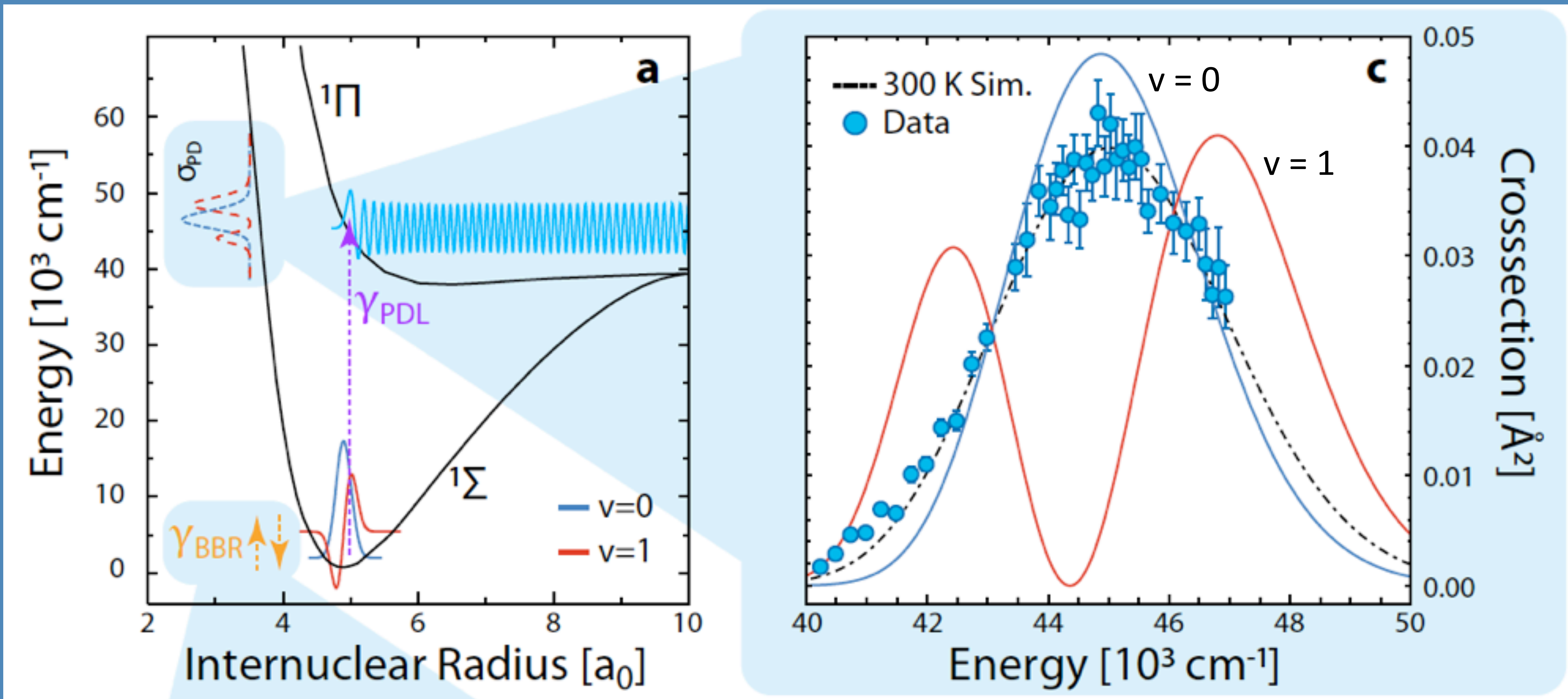
- Cold Molecular ions: Photodissociative thermometry



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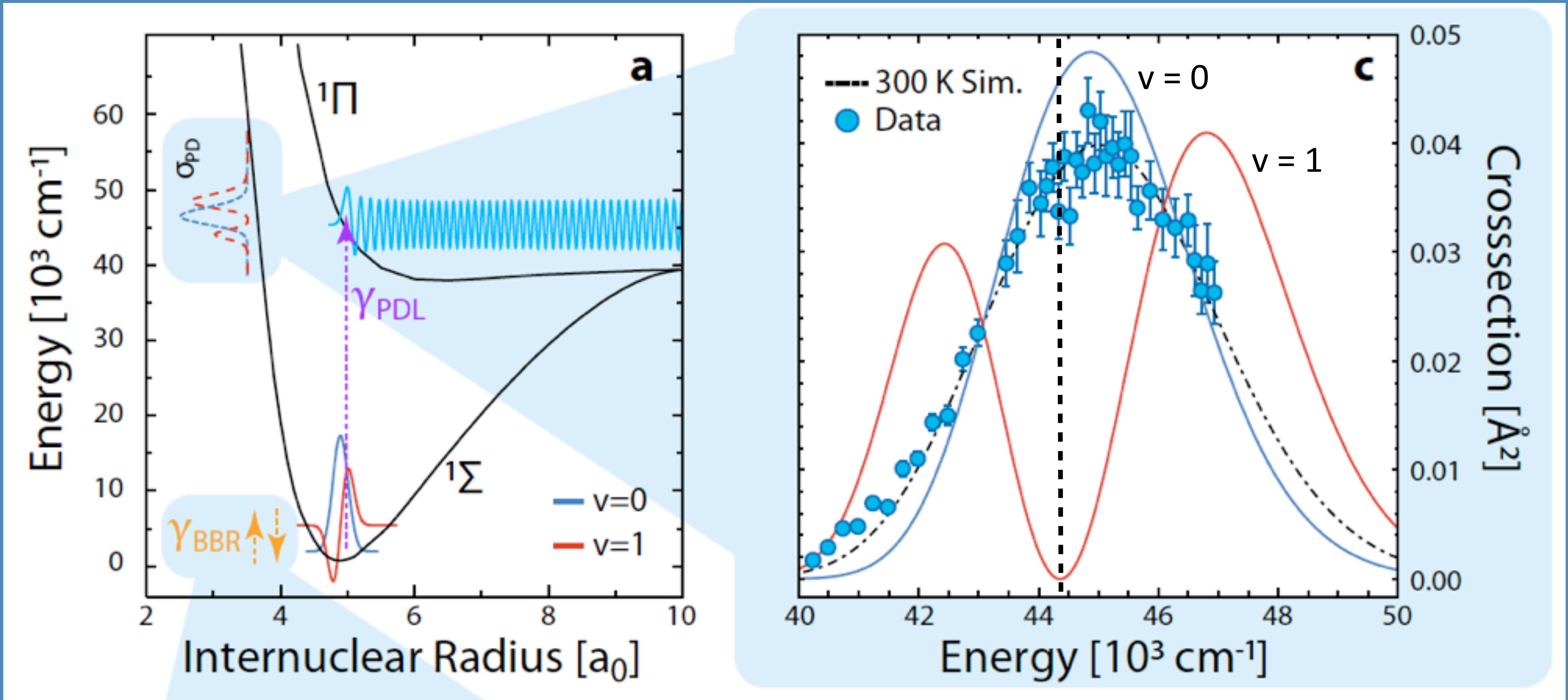

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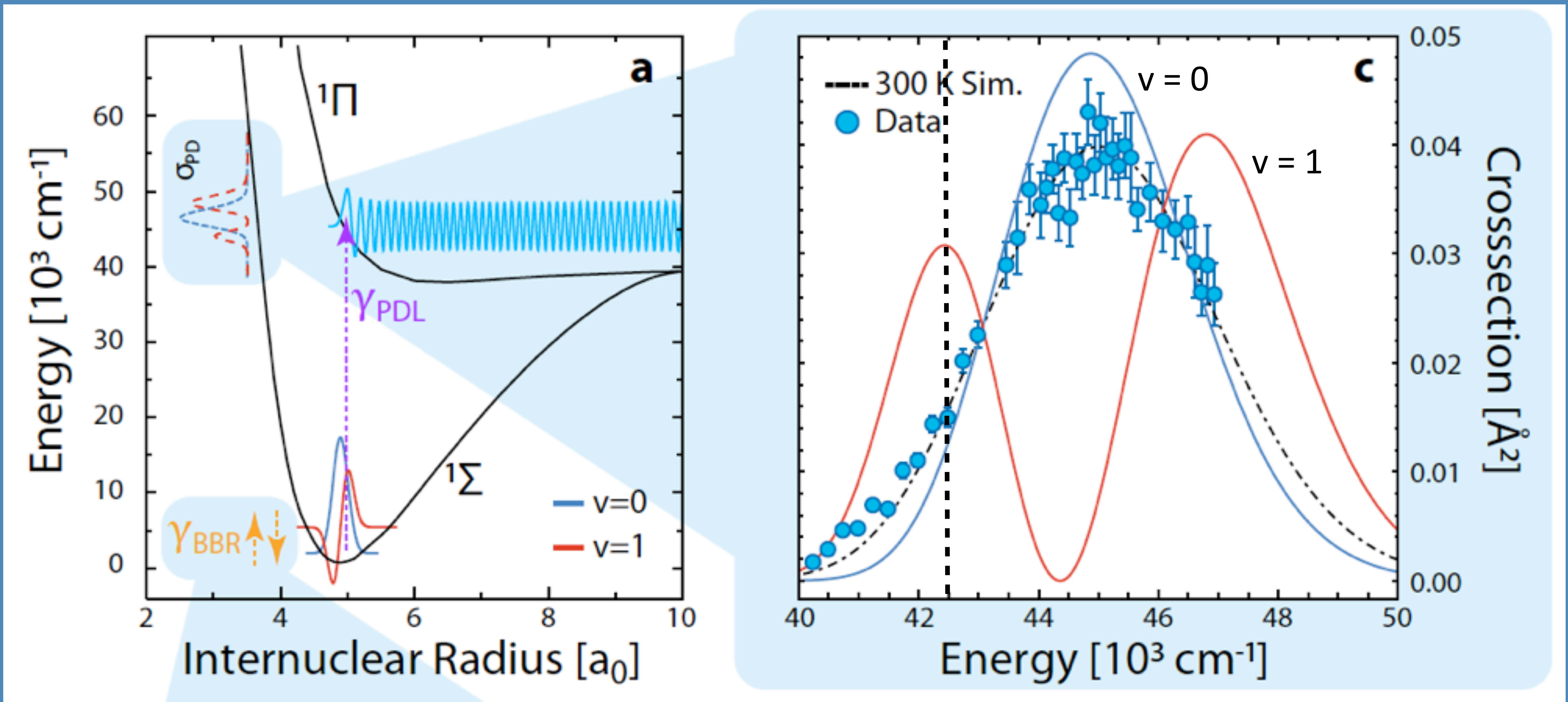
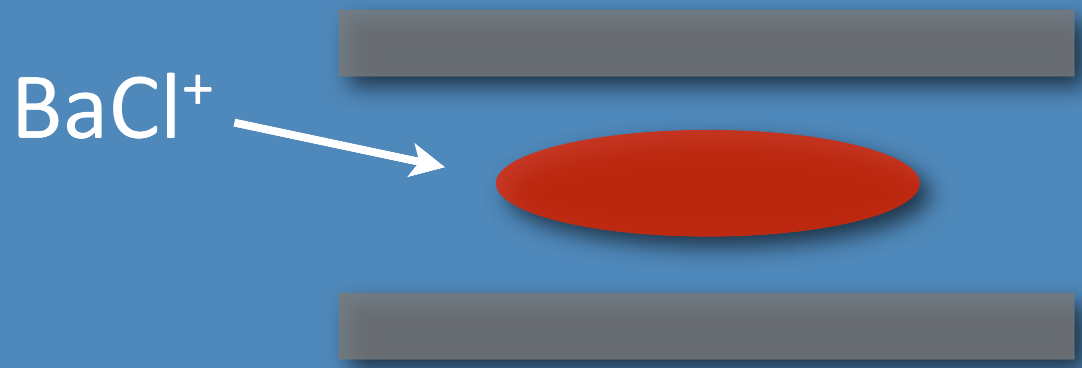
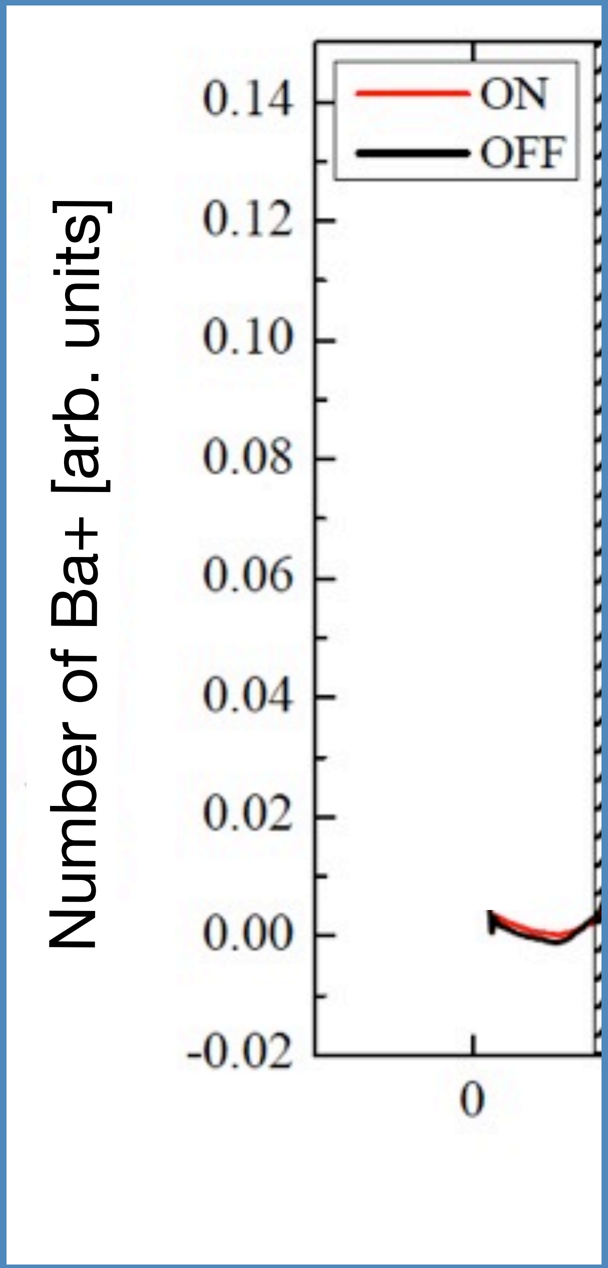


# Cold molecular ions?

## Internal State Thermometry



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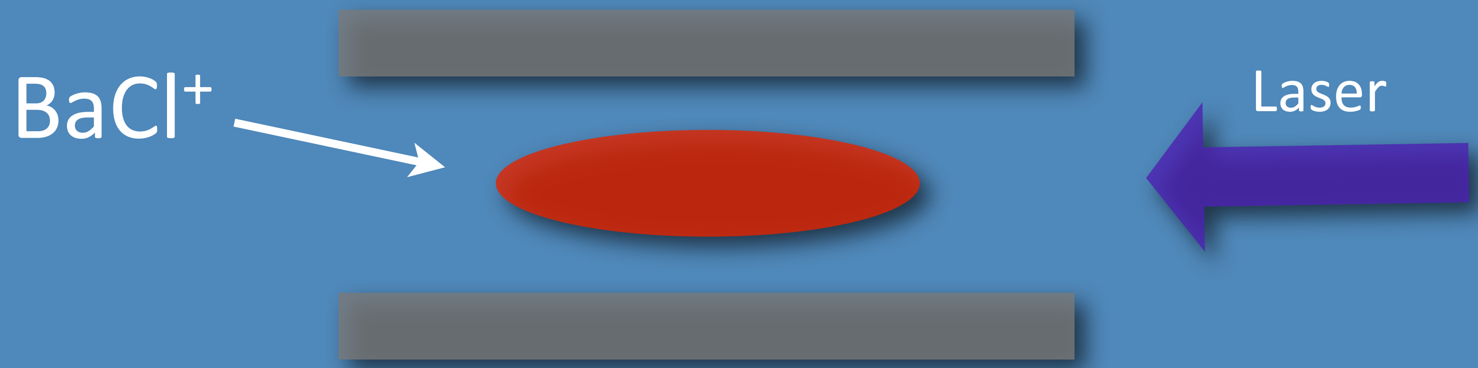


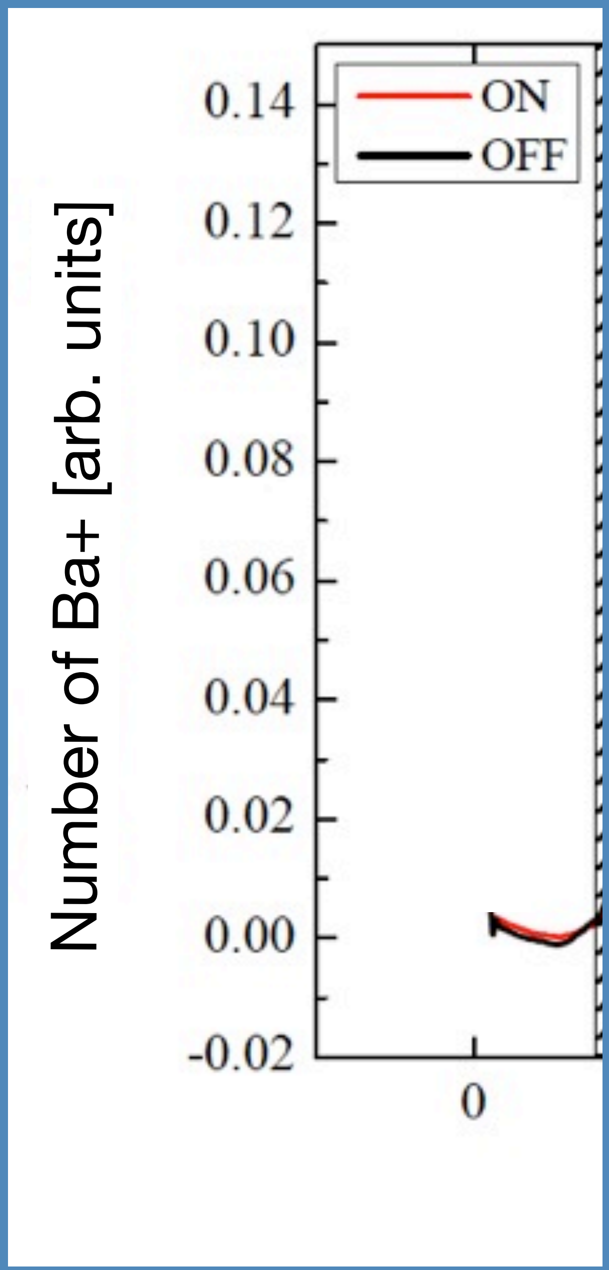

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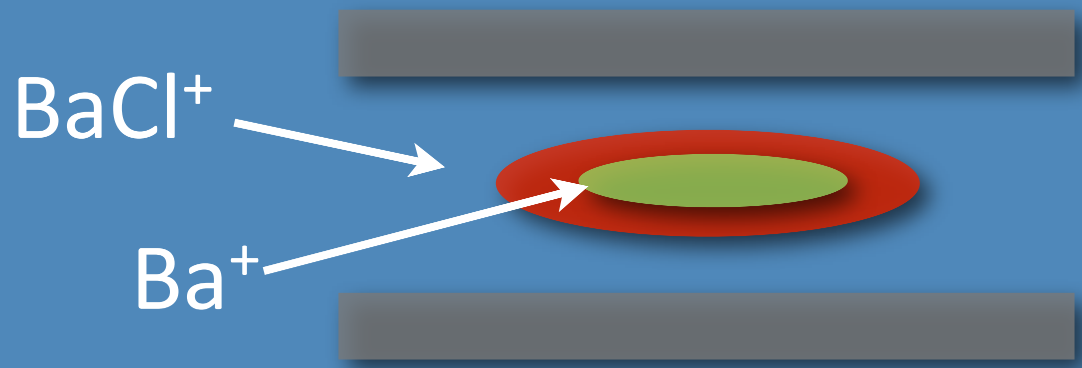


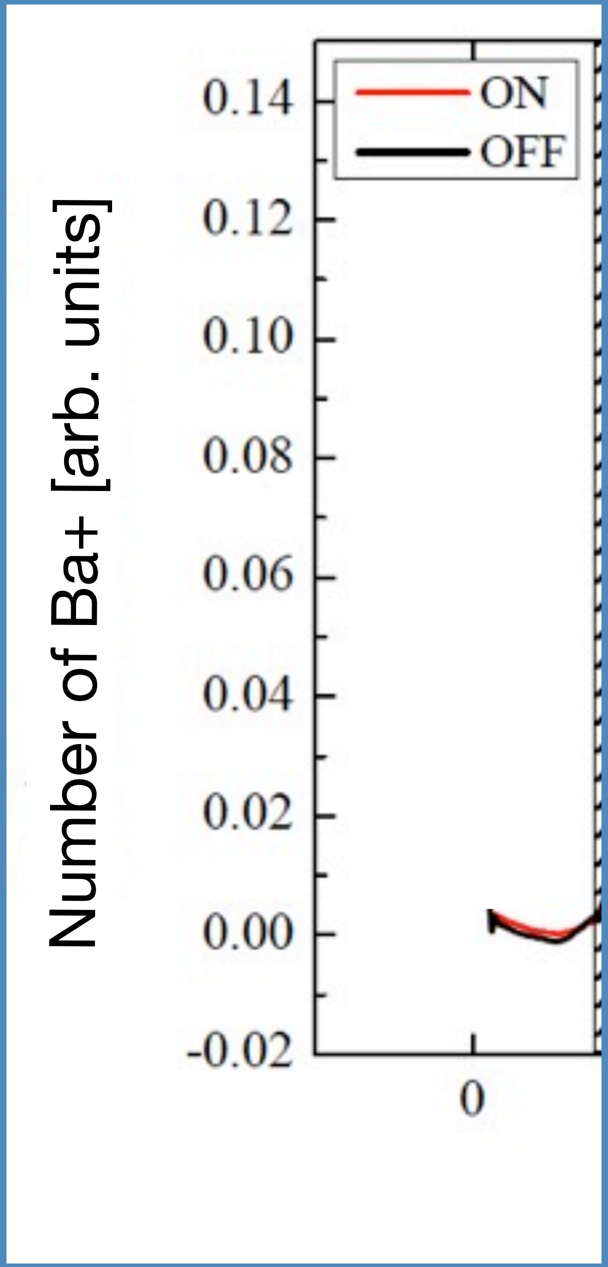

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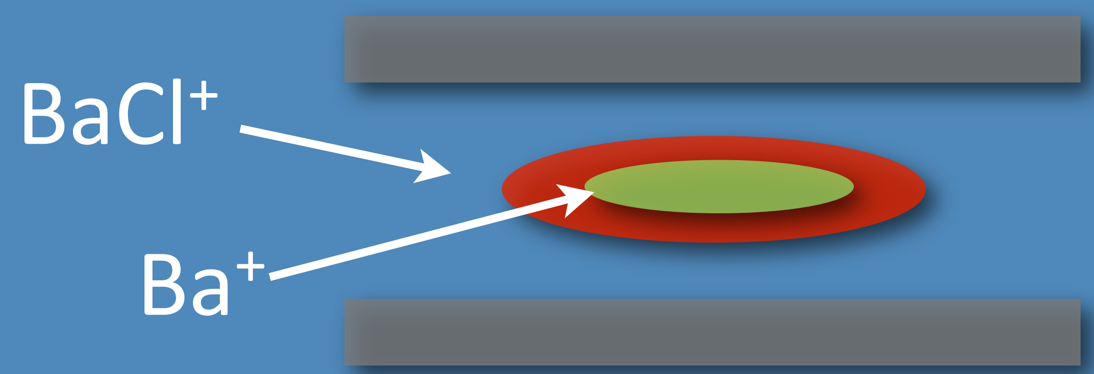


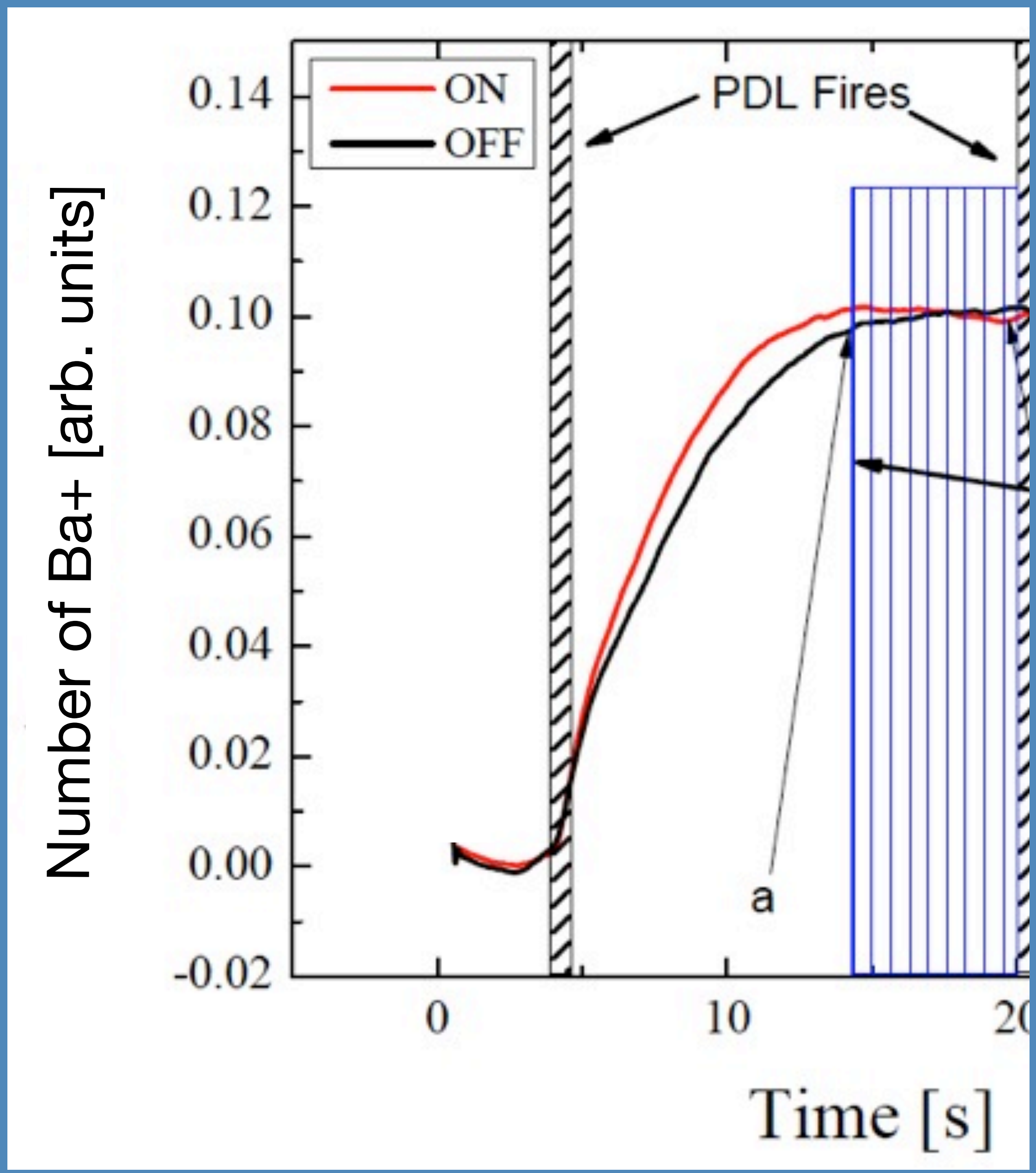

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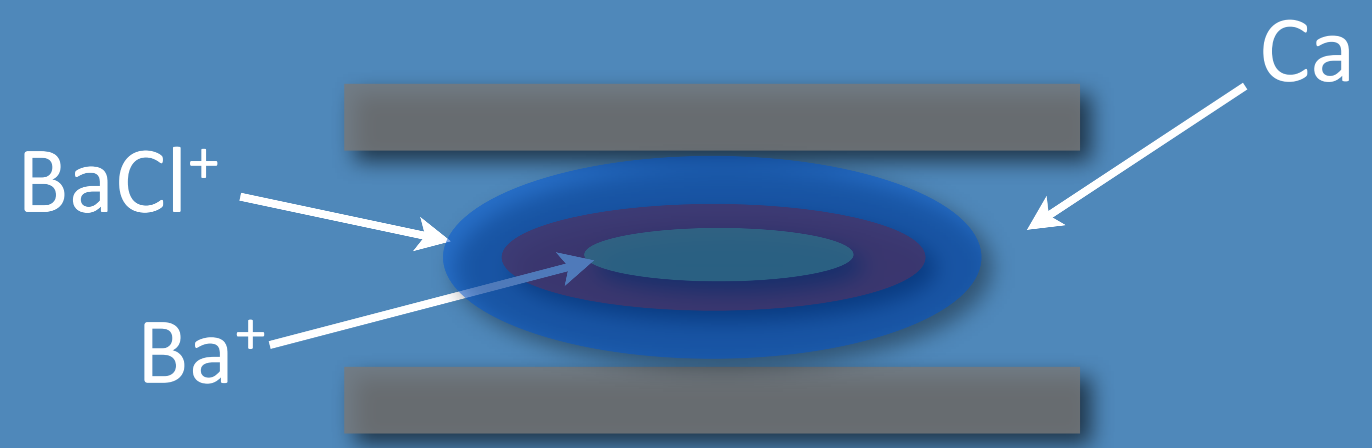


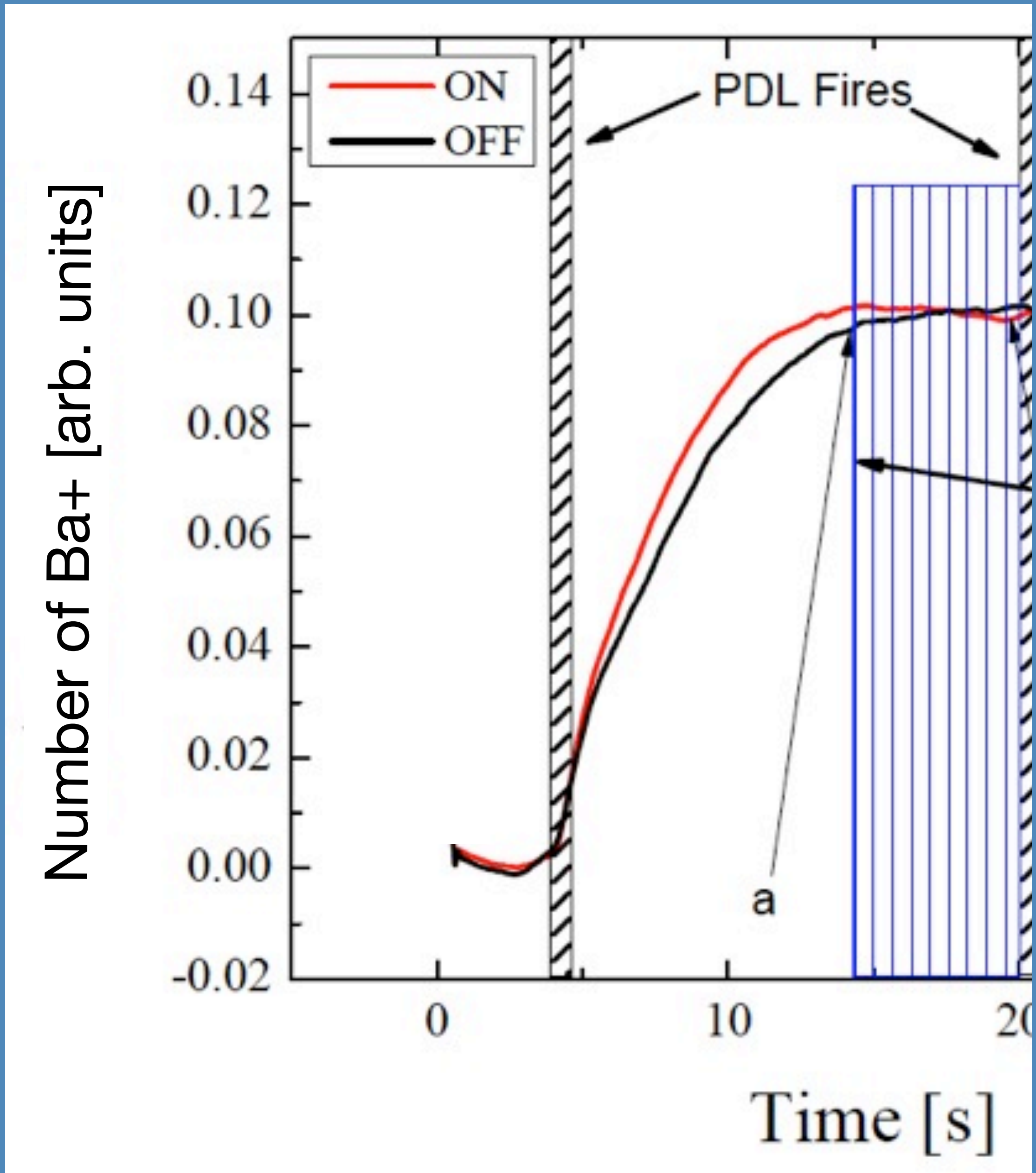

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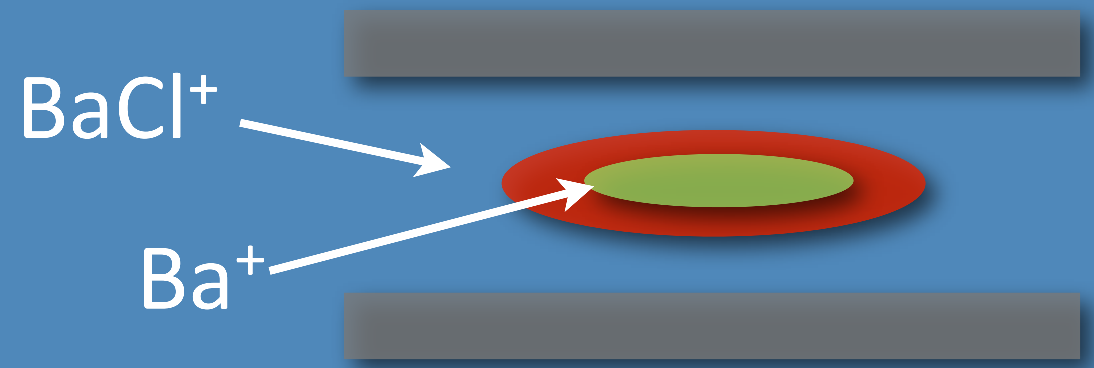


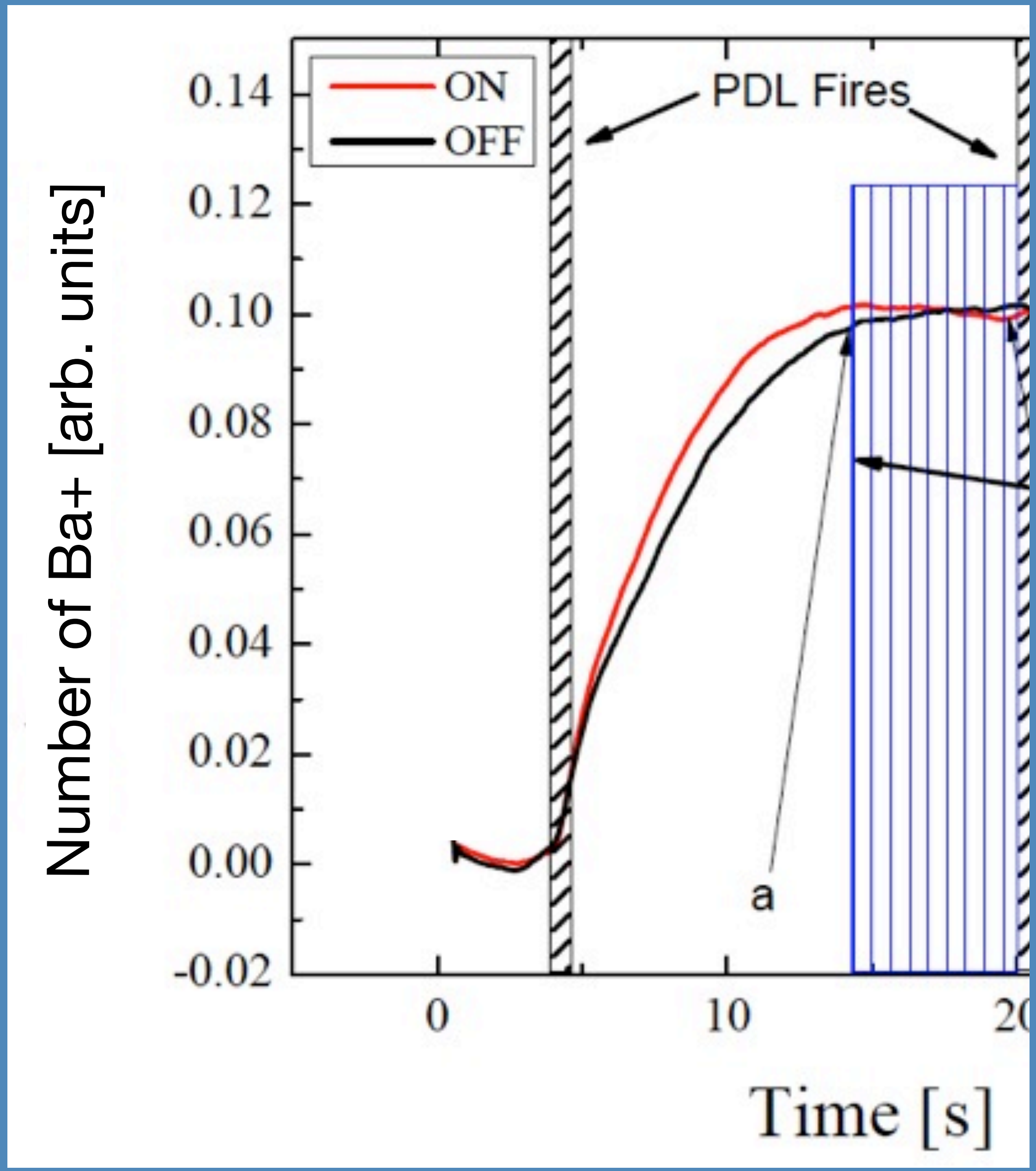

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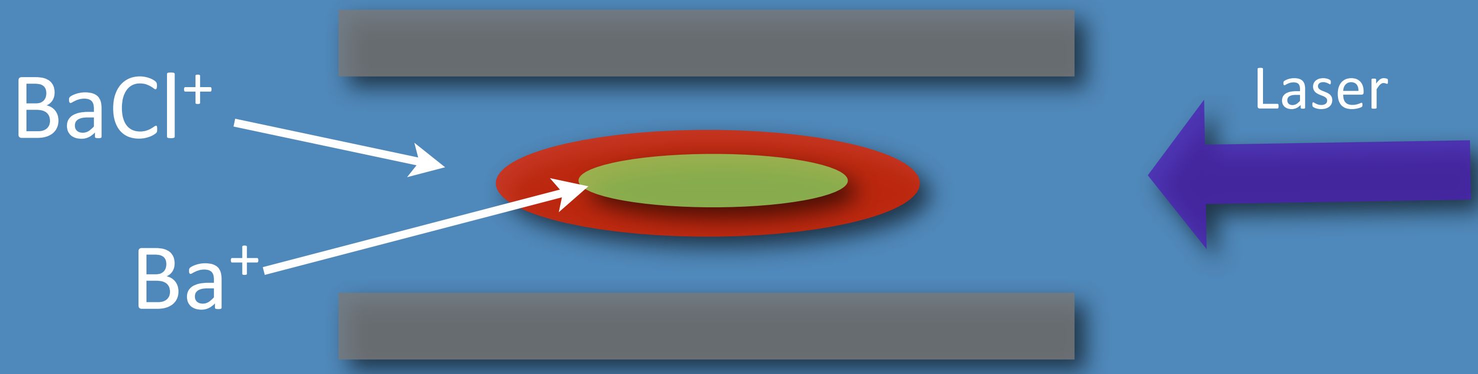


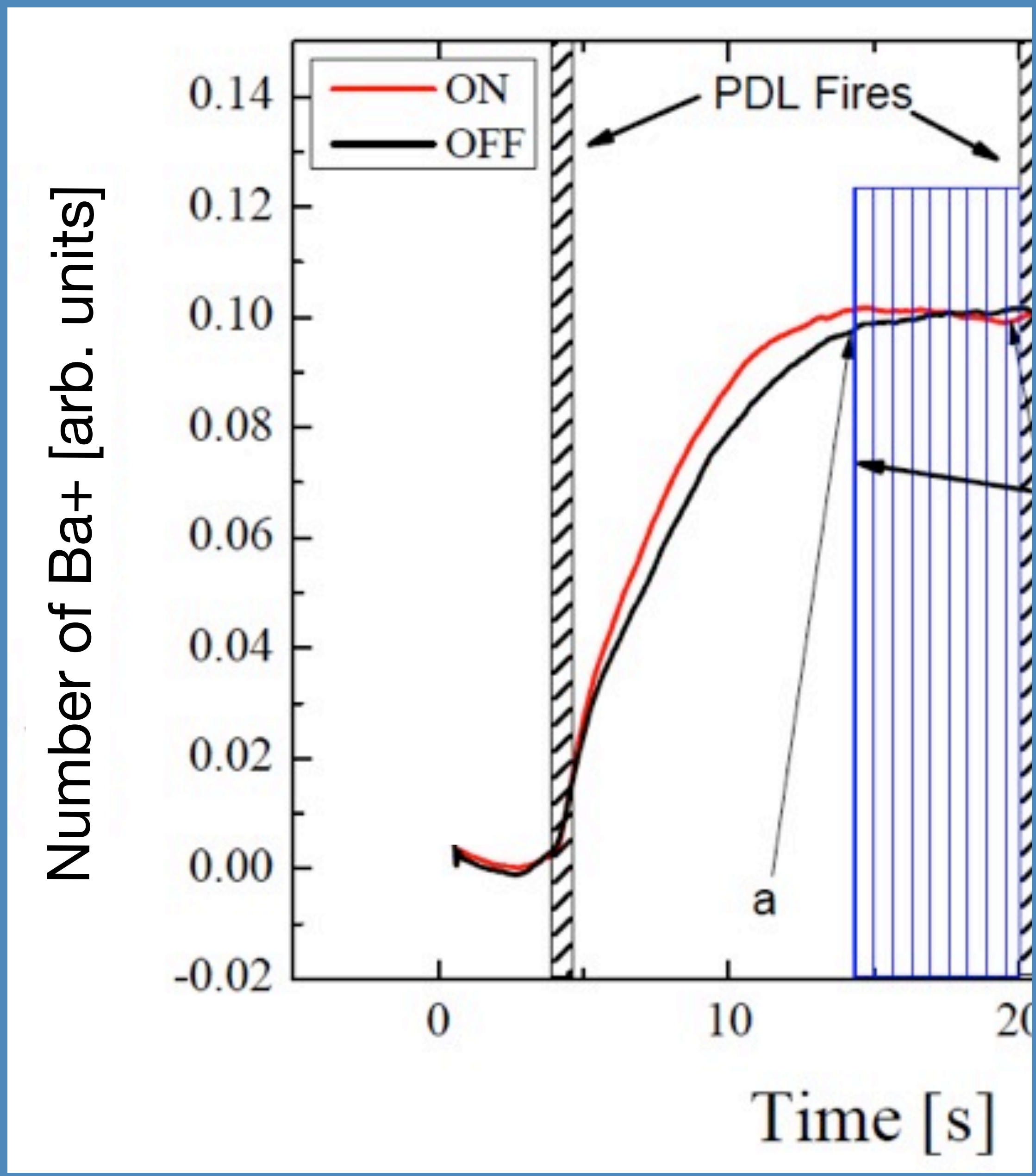

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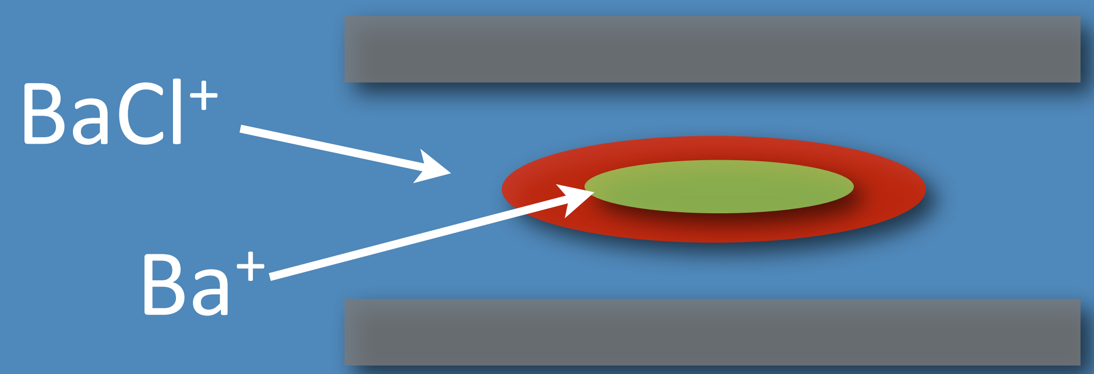
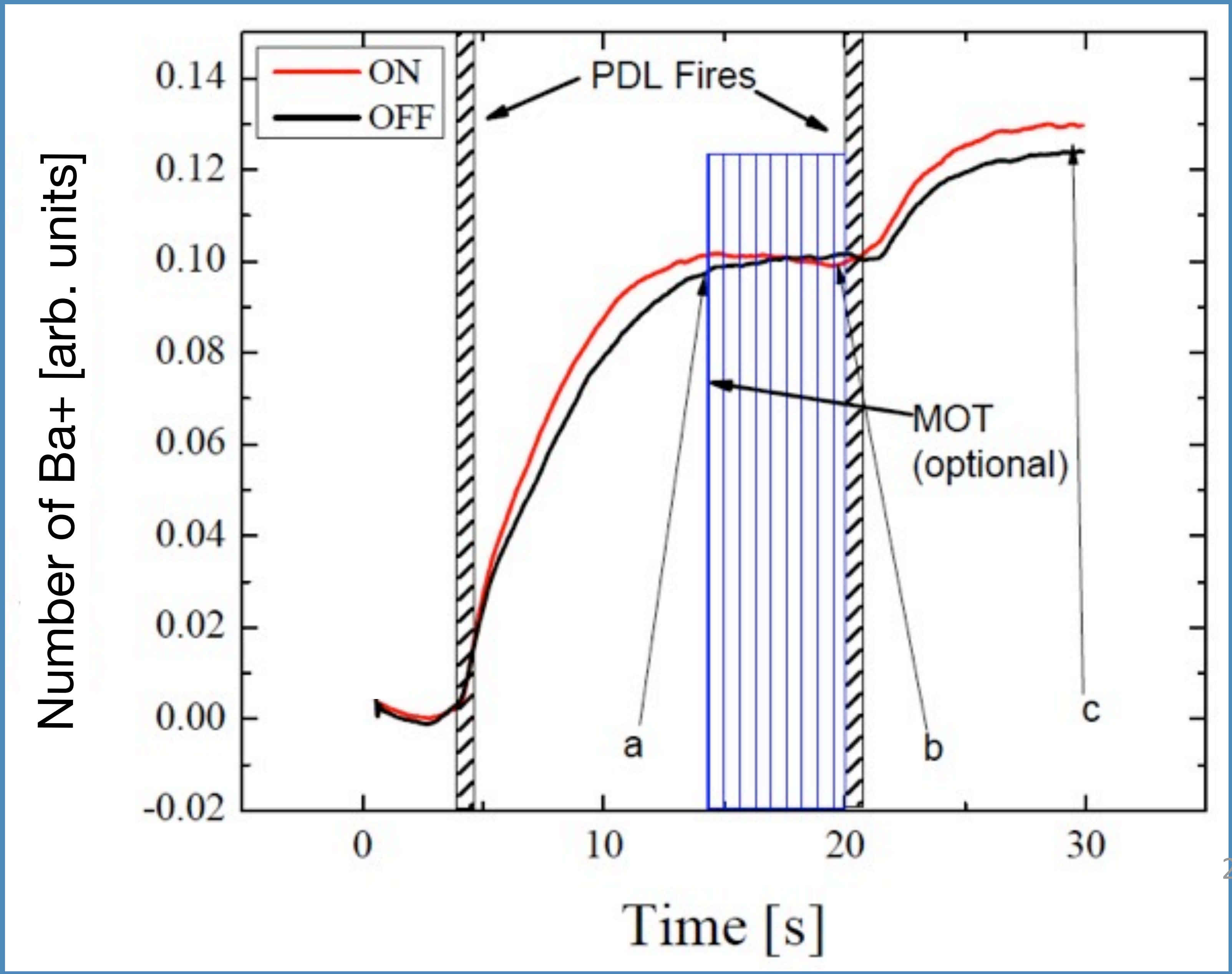
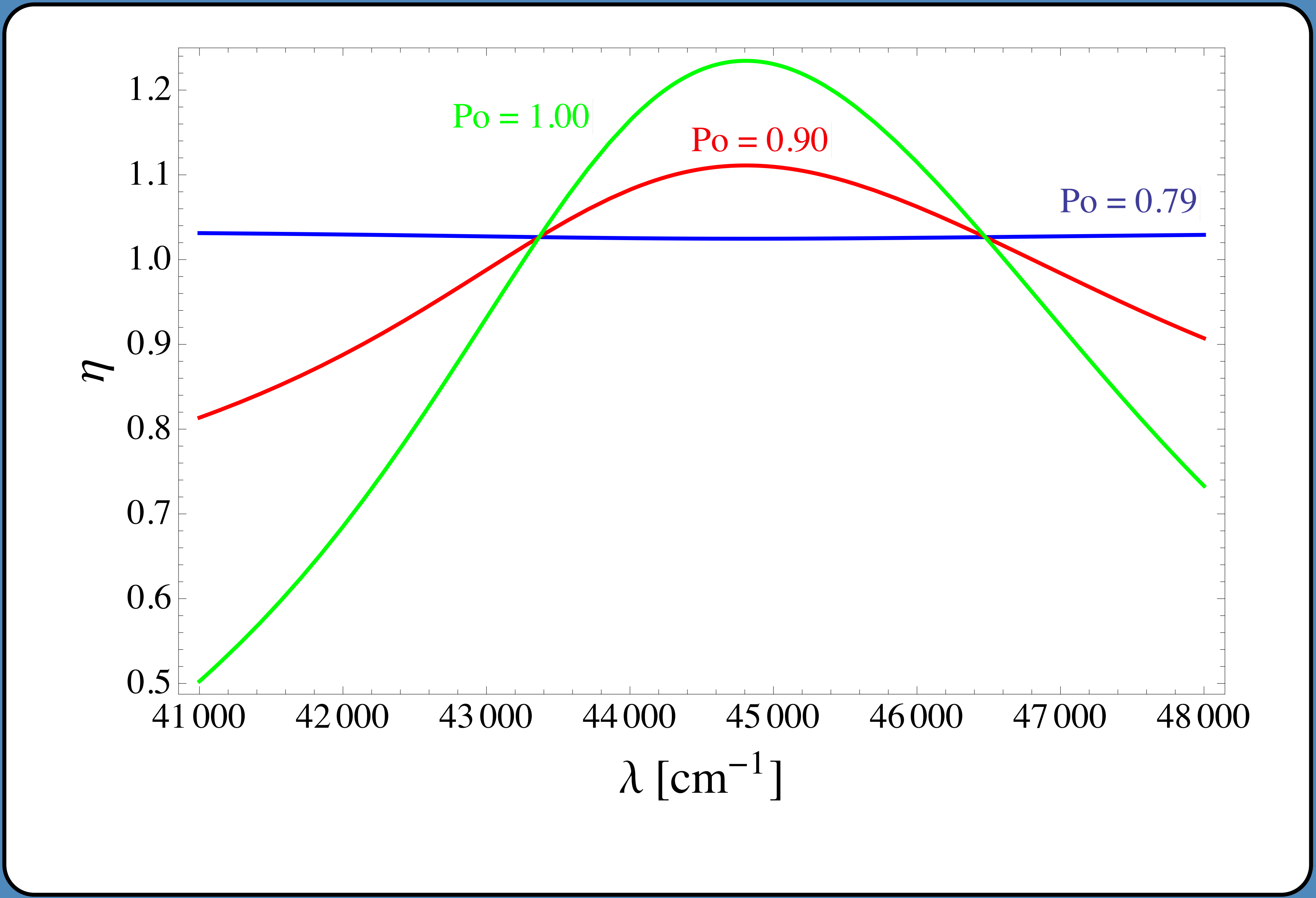


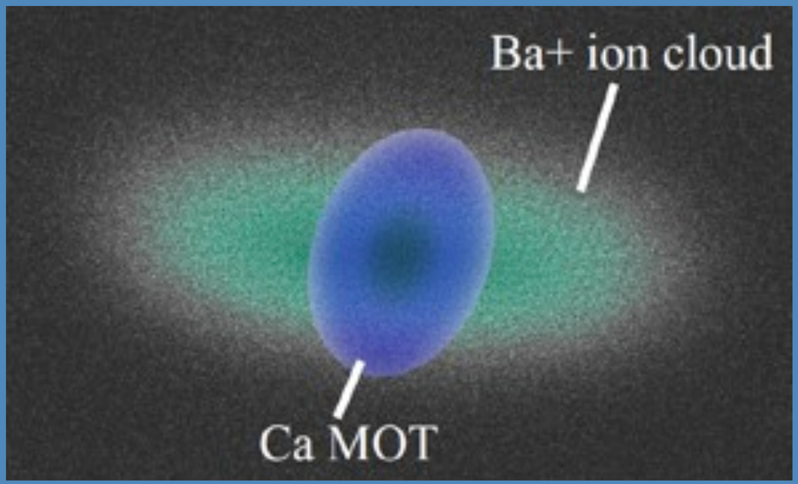
# Cold molecular ions

## First attempts at sympathetic cooling




External state cooling --> Ba+ ions.

Internal state cooling --> Ca MOT



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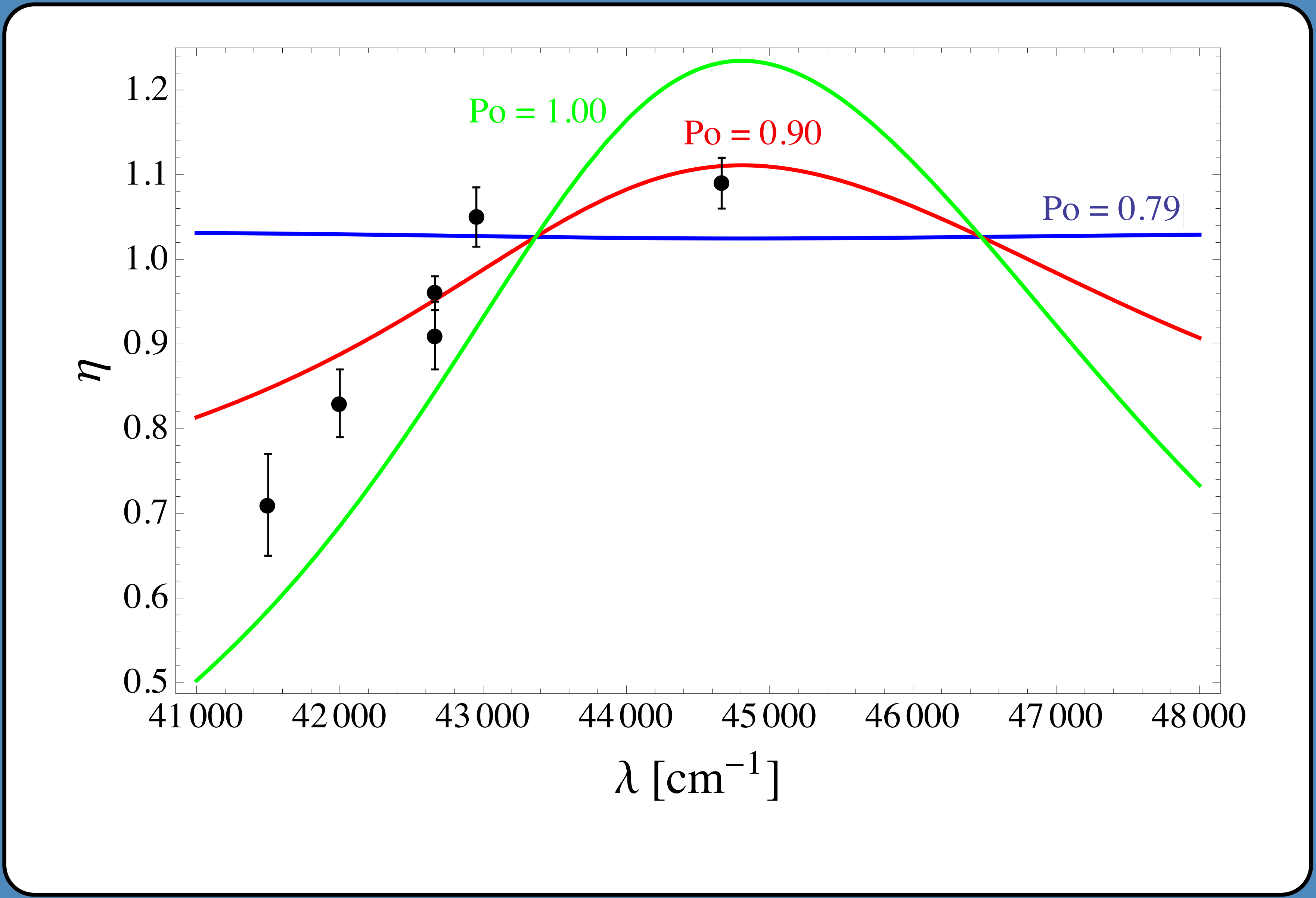


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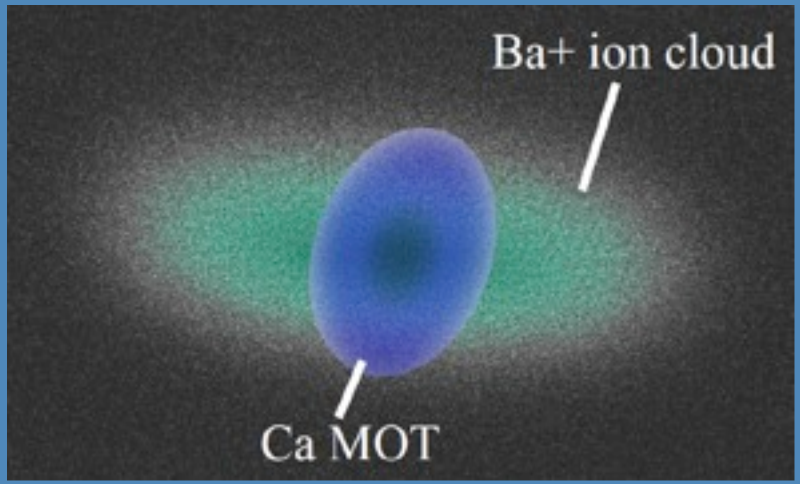

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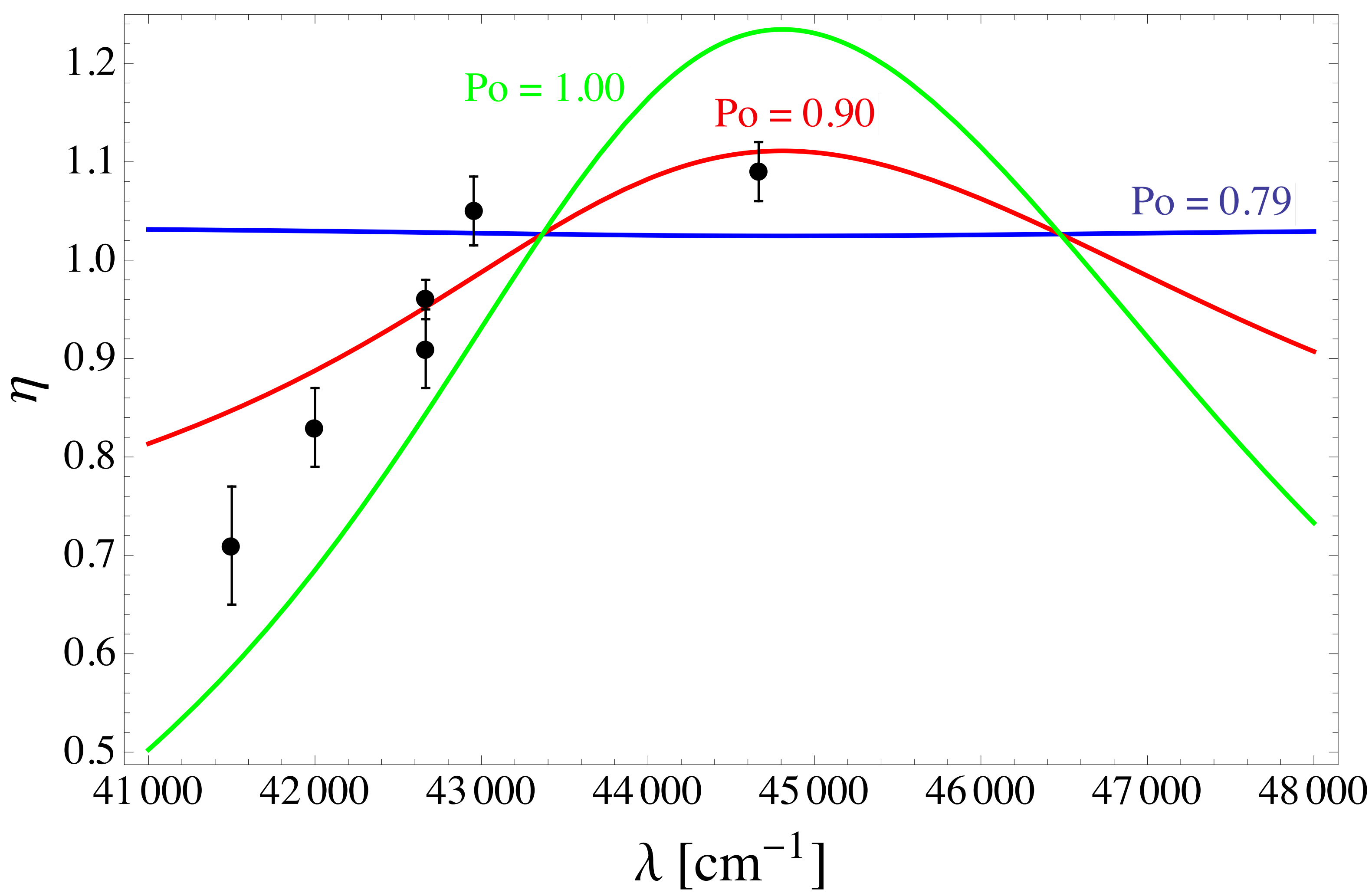


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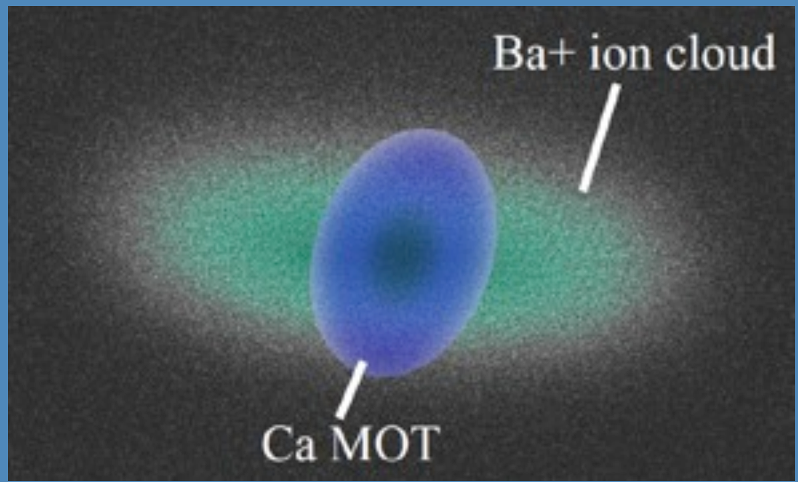


$K_Q \sim 1 \times 10^{-9}\ cm^3 s^{-1}$

~1/5 of the Langevin rate

# • Cold molecular ions

Vibrational quenching rate is ~1/5 of the Langevin rate (classical upper limit).

Sympathetic cooling extremely efficient

Neutral atom -- Neutral molecule vibrational quenching rates are $\gg 10^6$ times slower!

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SYMPATHETIC COOLING EXTREMELY EFFICIENT

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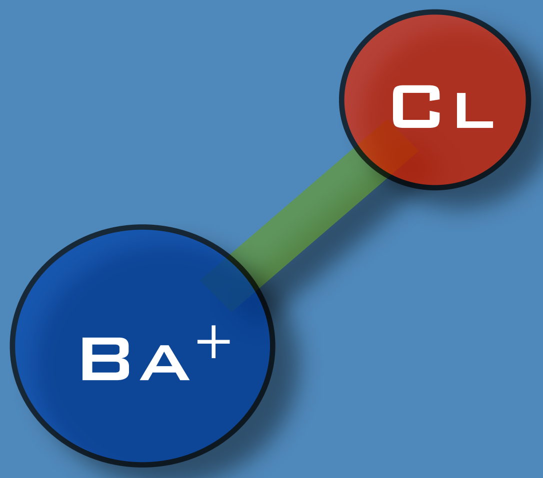

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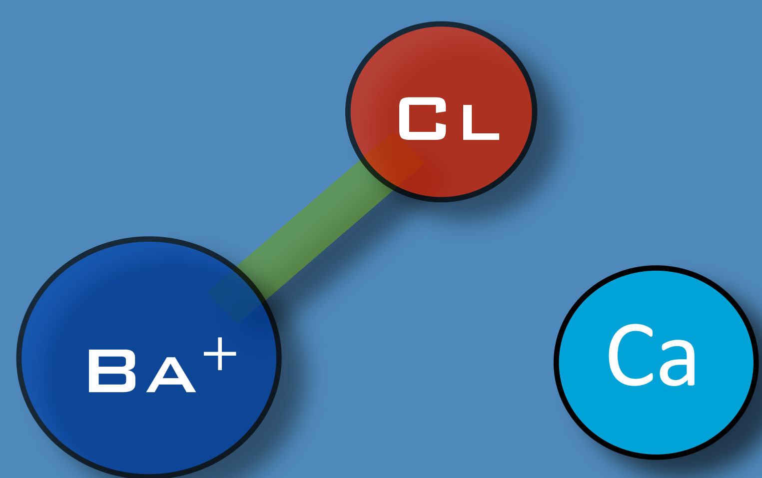

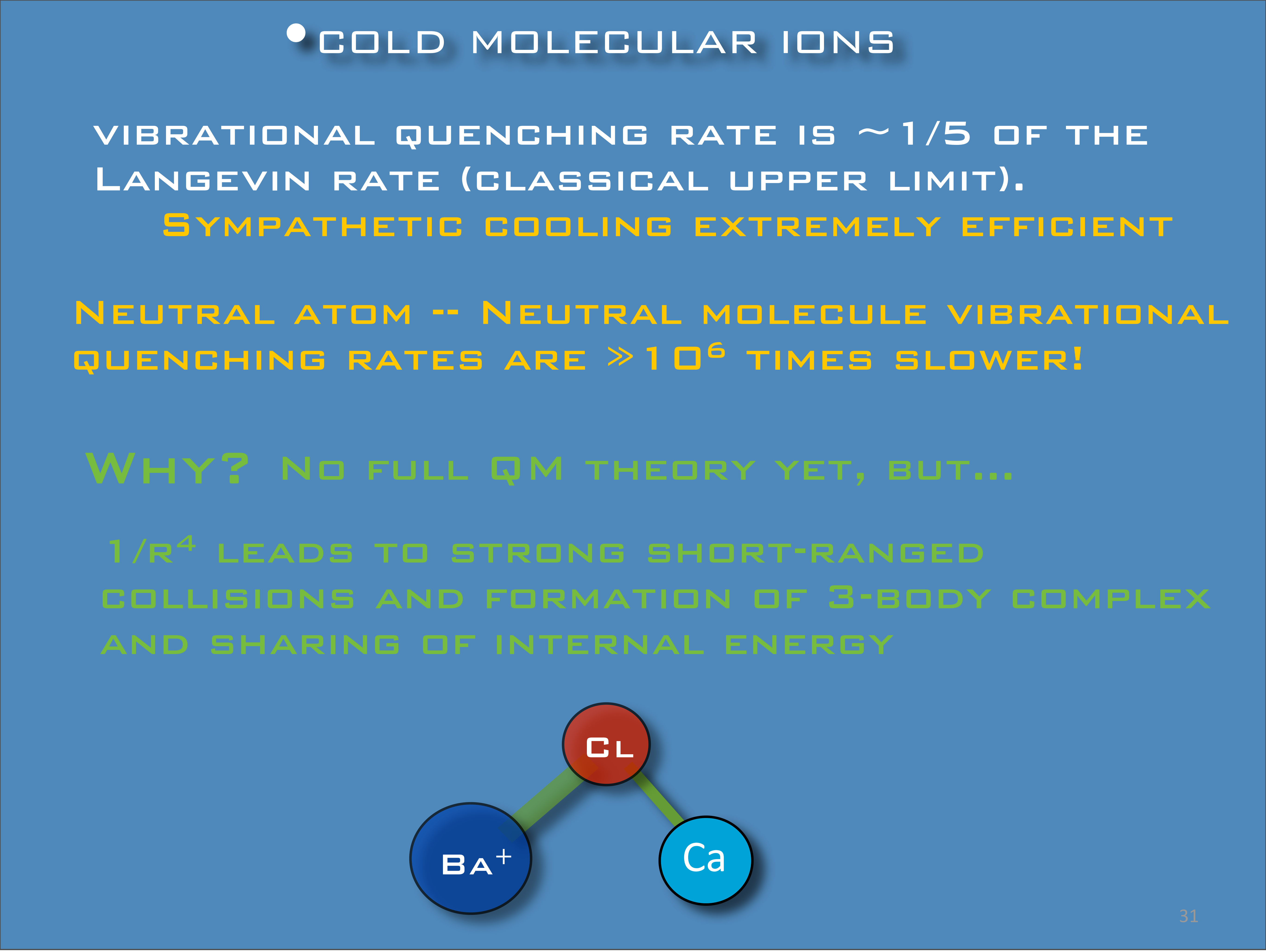
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Ba+
Ca
31

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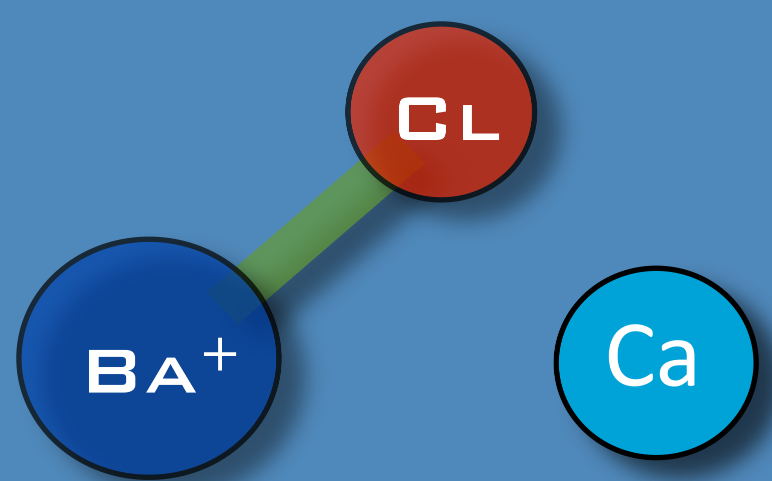

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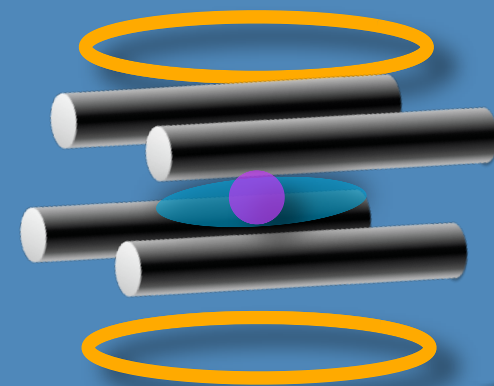
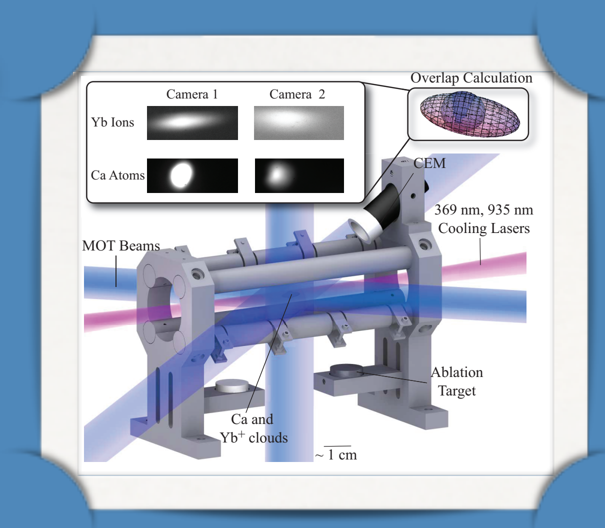
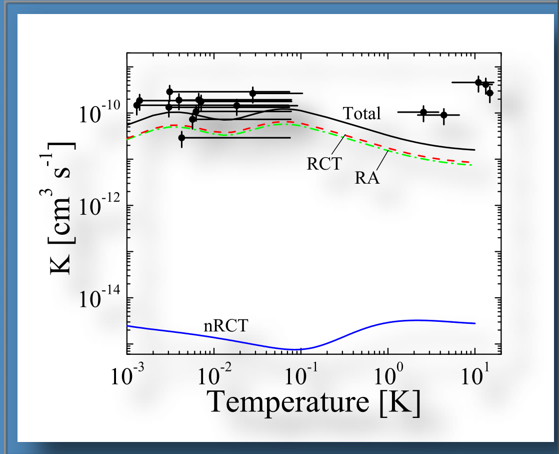
# Summary

- MOTION Trap

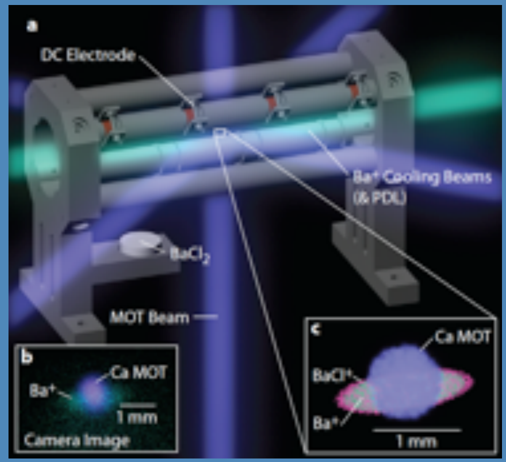


- Some surprising chemistry




- Making cold molecular ions

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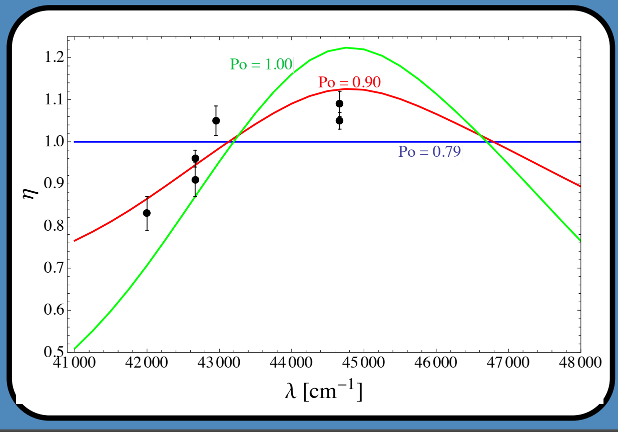
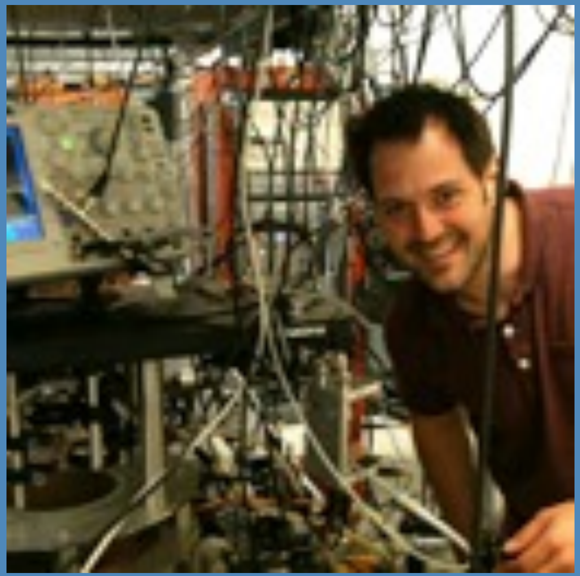


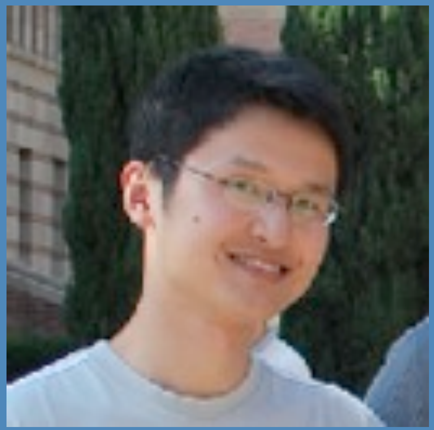
# Acknowledgements



Res. Professor: Wade G. Rellergert
Everything

Prof. Svetlana Kotochigova
Temple University
Theory

Scott T. Sullivan
MOTION Trap Implementation



Kuang Chen Steven Schowalter
Spectroscopy/integrated MS