

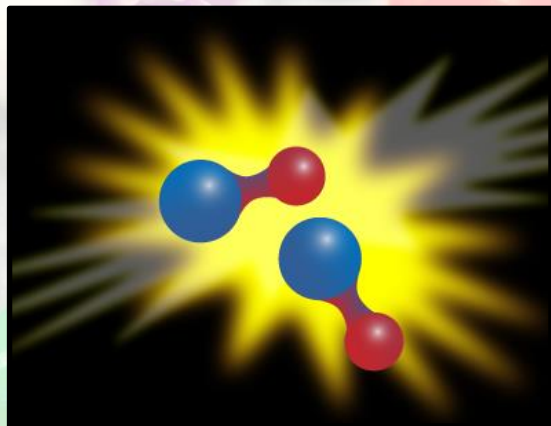
# Molecular Lattice Clocks



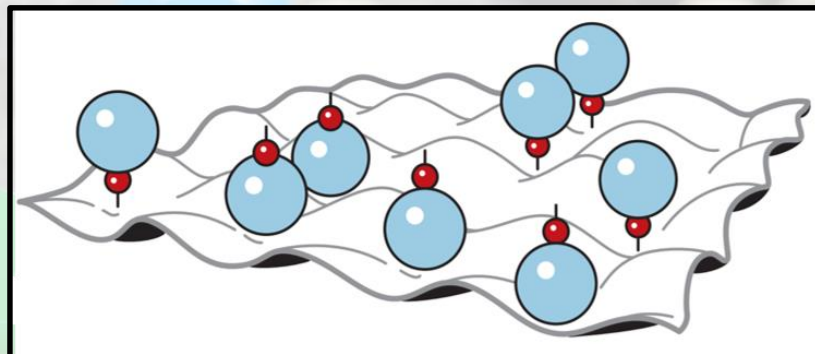
Tanya Zelevinsky  
*Columbia University, New York*



# Quantum science with molecules



Fundamental  
chemistry



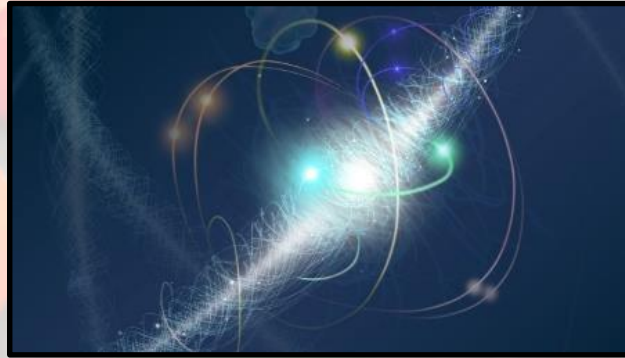
Quantum  
materials



**Fundamental  
physics & metrology**



# Quantum science with molecules

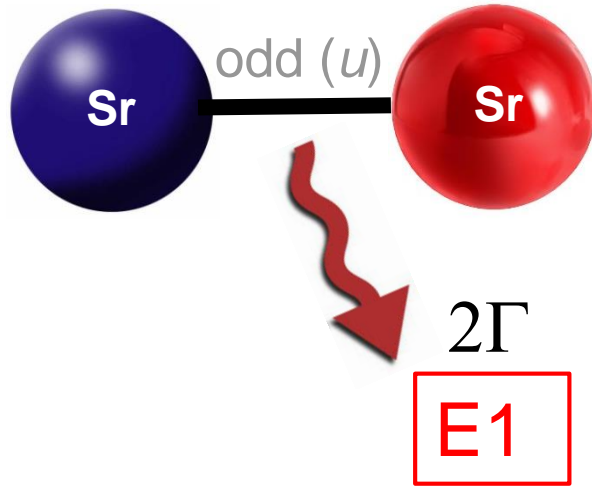


## **Molecular clocks: Limits of precision and coherence with molecular states**

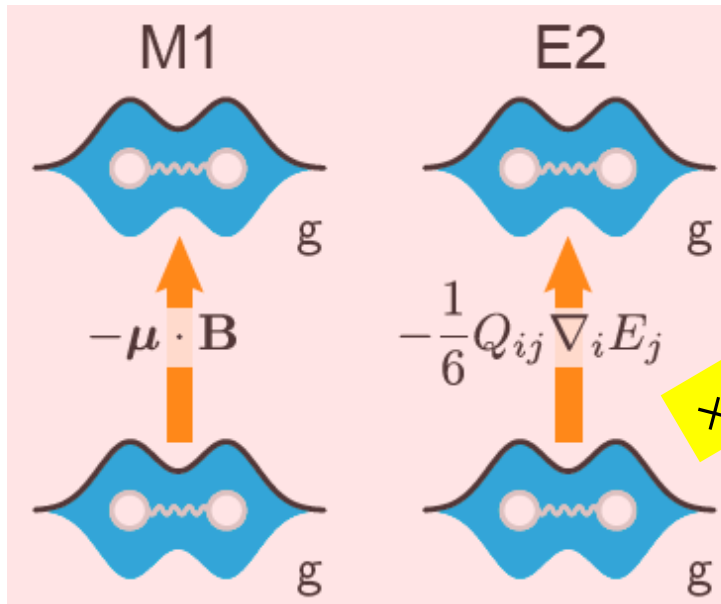
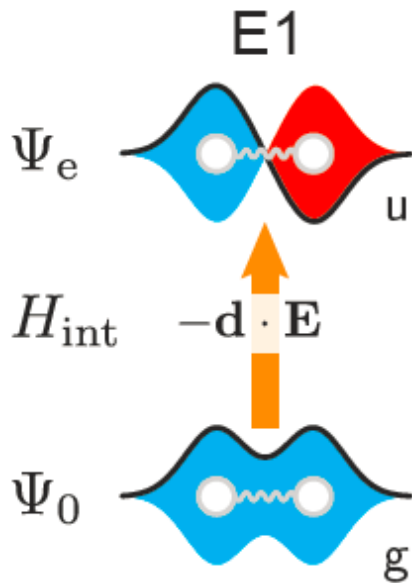
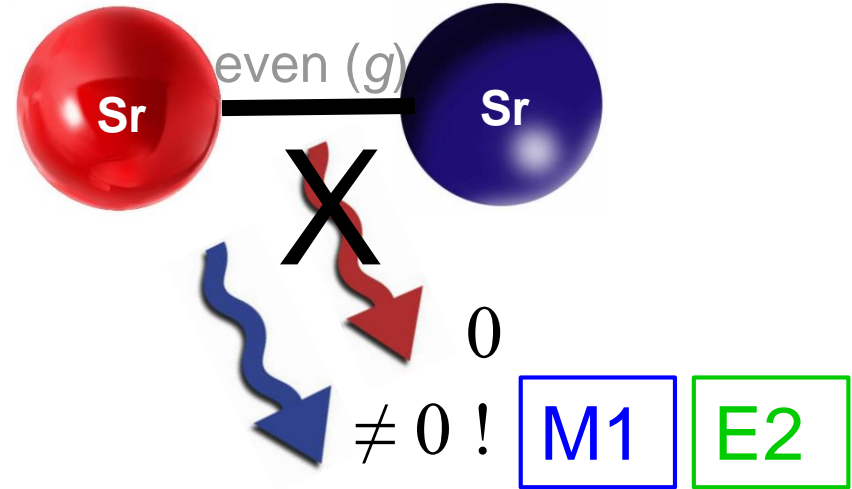
- **Metrology with atoms & molecules**
- **Precise measurements of fundamental phenomena**
- **Coherence in optical traps for clocks, qubits, cold chemistry**
- **New level of precision for molecular physics and QED**

# Two-body subradiant states

superradiant  
 $|S\rangle|P\rangle + |P\rangle|S\rangle$

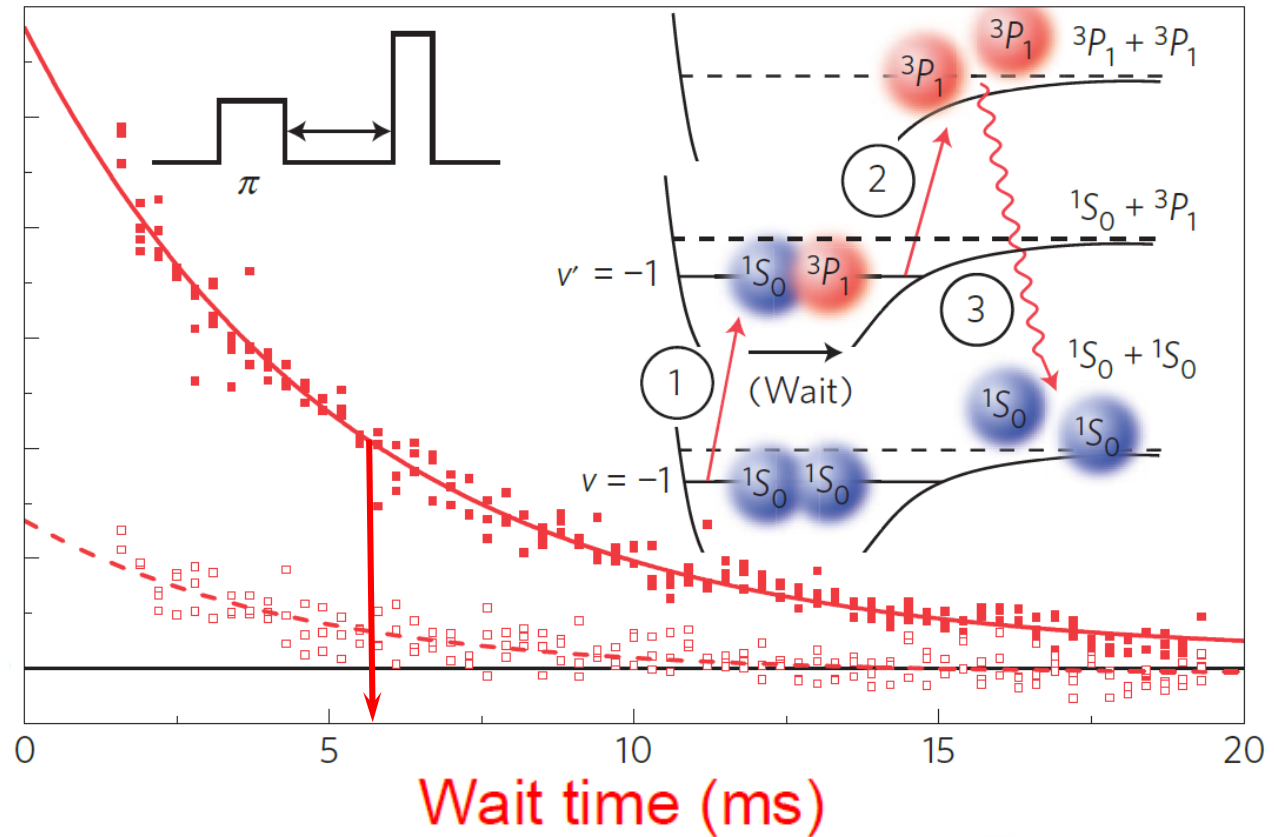
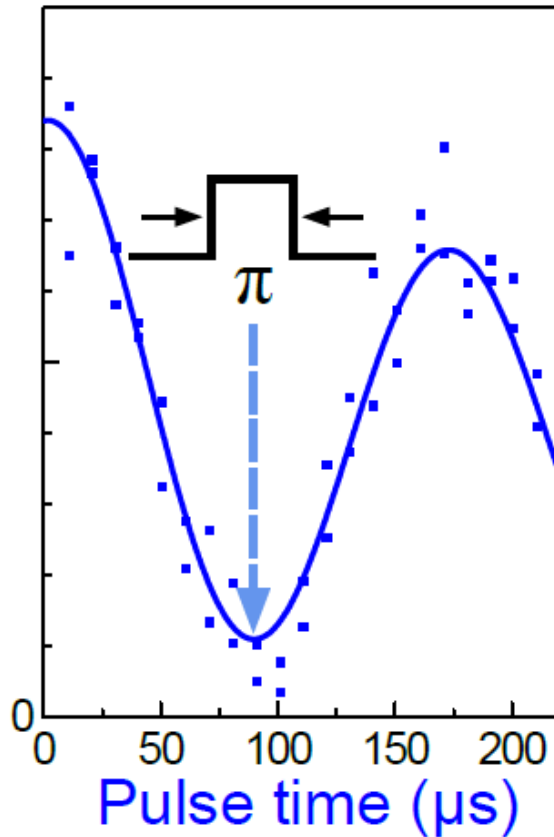


subradiant  
 $|S\rangle|P\rangle - |P\rangle|S\rangle$



# Two-body subradiant states

Coherent control



Molecule-light coherence,  
 $Q > 3 \times 10^{12}$

5.5 ms lifetime

# Ultracold photodissociation

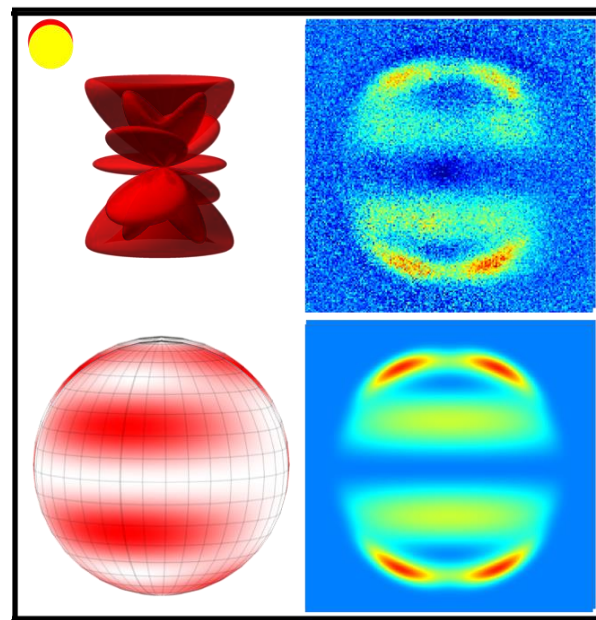
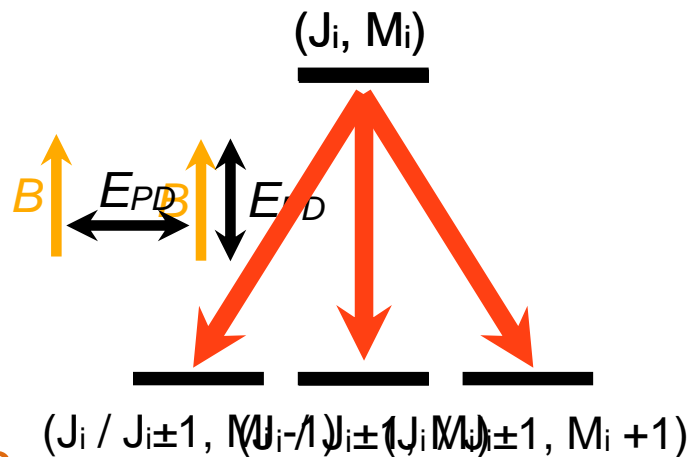
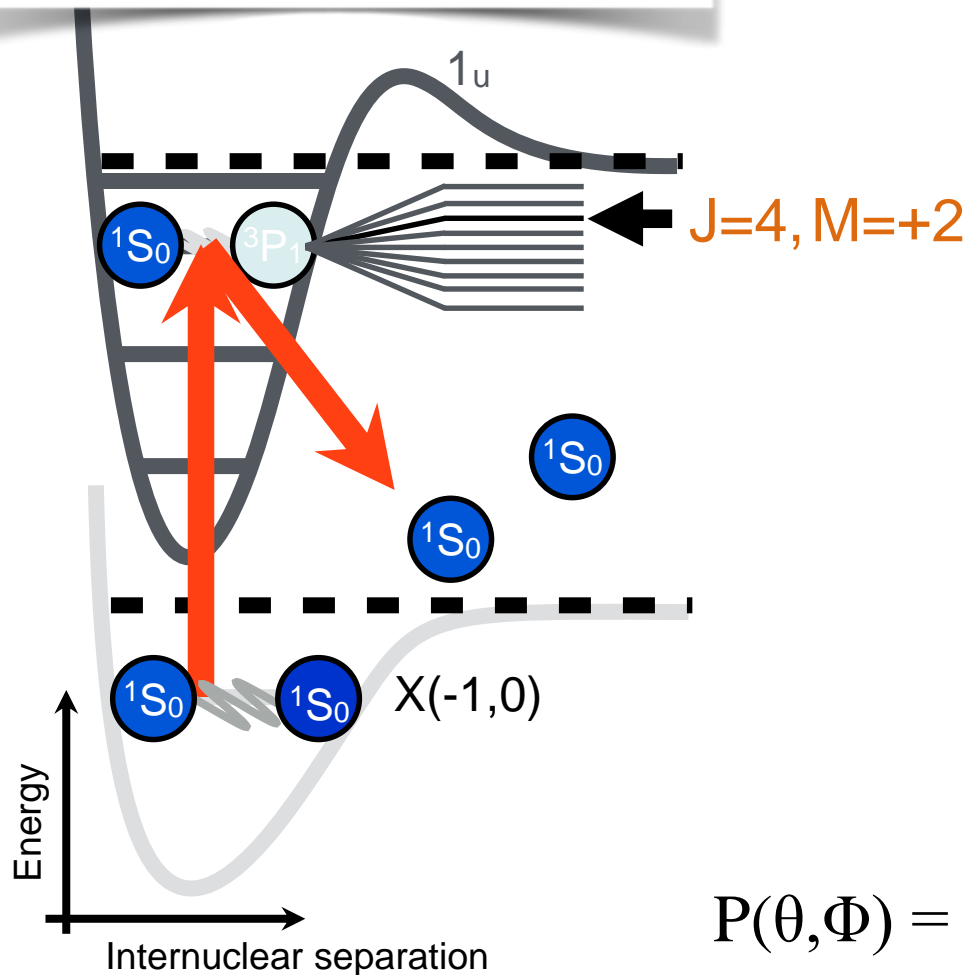
LETTER

doi:10.1038/nature18314

## Photodissociation of ultracold diatomic strontium molecules with quantum state control

M. McDonald<sup>1</sup>, B. H. McGuyer<sup>1</sup>, F. Apfelbeck<sup>1,4</sup>, C.-H. Lee<sup>1</sup>, I. Majewska<sup>2</sup>, R. Moszynski<sup>2</sup> & T. Zelevinsky<sup>1</sup>

122 | NATURE | VOL 535 | 7 JULY 2016



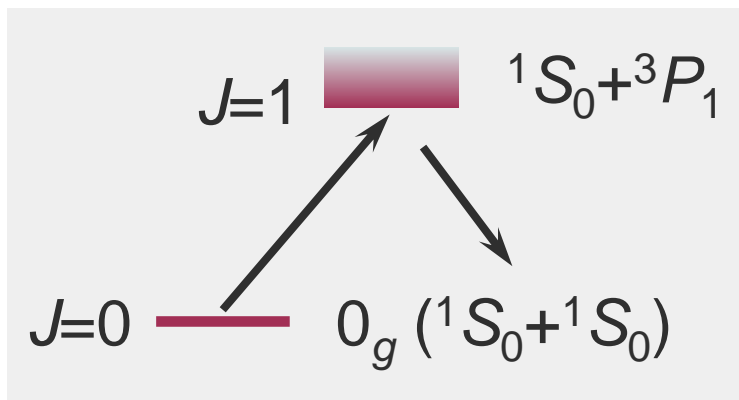
$$P(\theta, \Phi) = |\sqrt{R} \langle Y_4^3(\theta, \Phi) | Y_4^3(\theta, \Phi) \rangle|^2 |Y_4^3(\theta, \Phi)|^2$$

Credit: M. McDonald

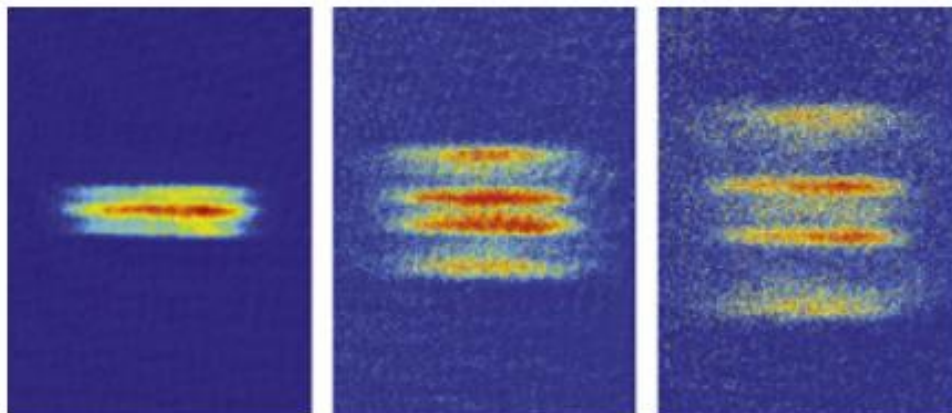


# Ultracold photodissociation

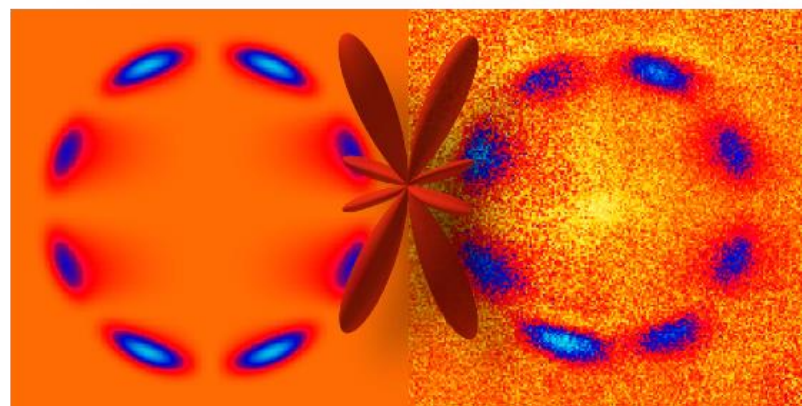
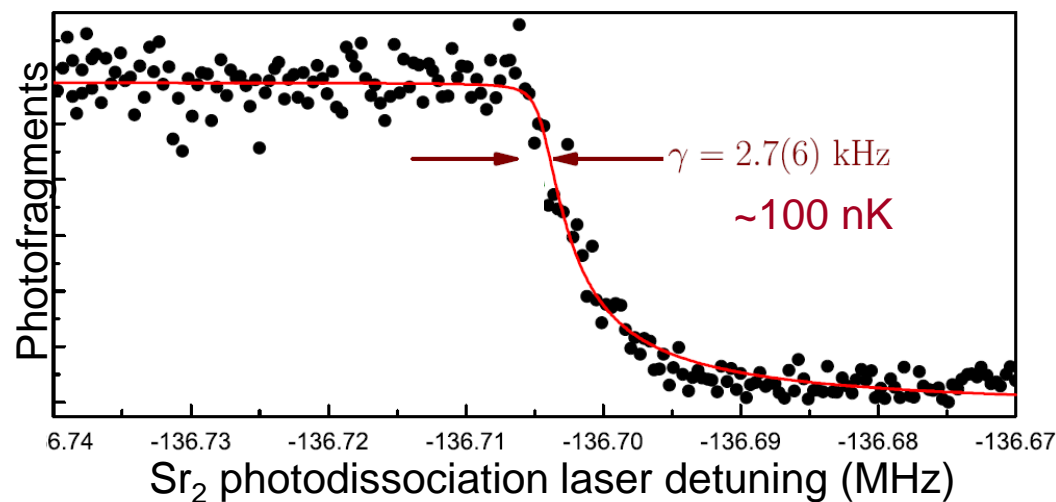
## State-resolved photochemistry



Side-view camera



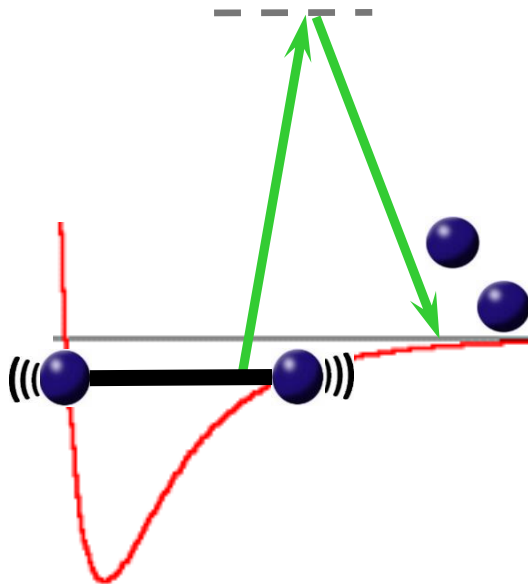
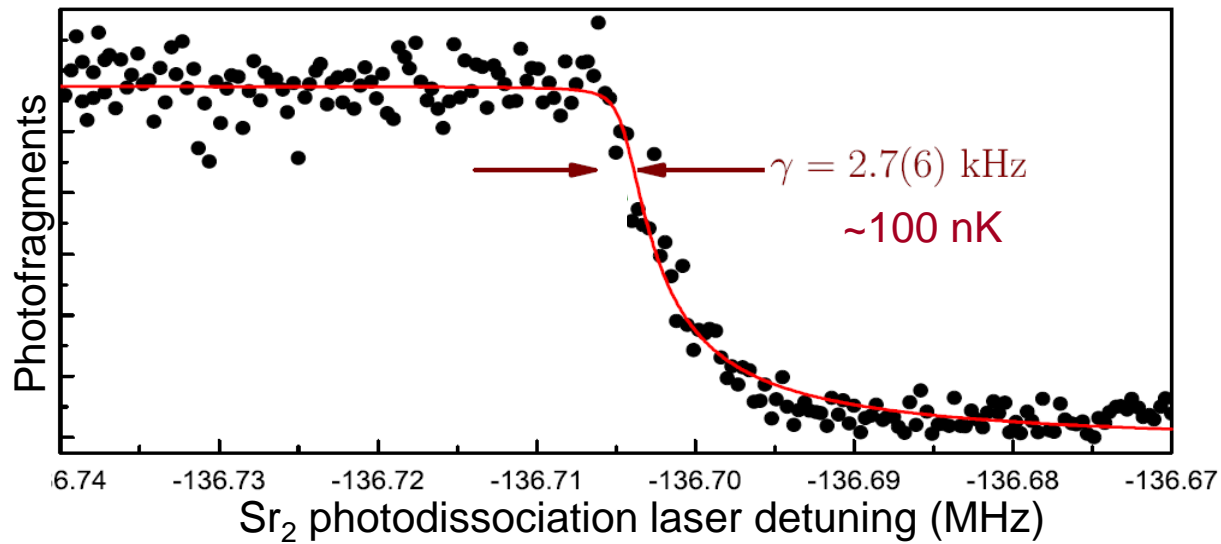
Line-of-sight camera: quantum interference



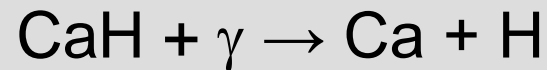
M. McDonald *et al.*, *Nature* **535**, 122 (2016)

B. McGuyer *et al.*, *NJP* **17**, 055004 (2015)

# Ultracold molecule dissociation



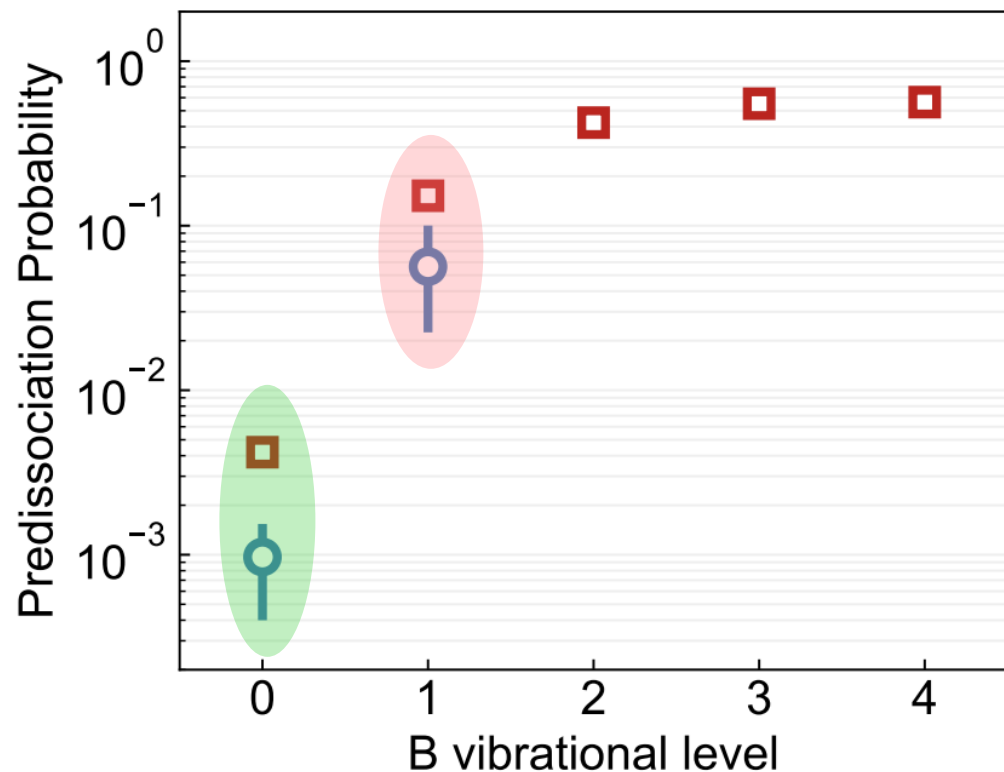
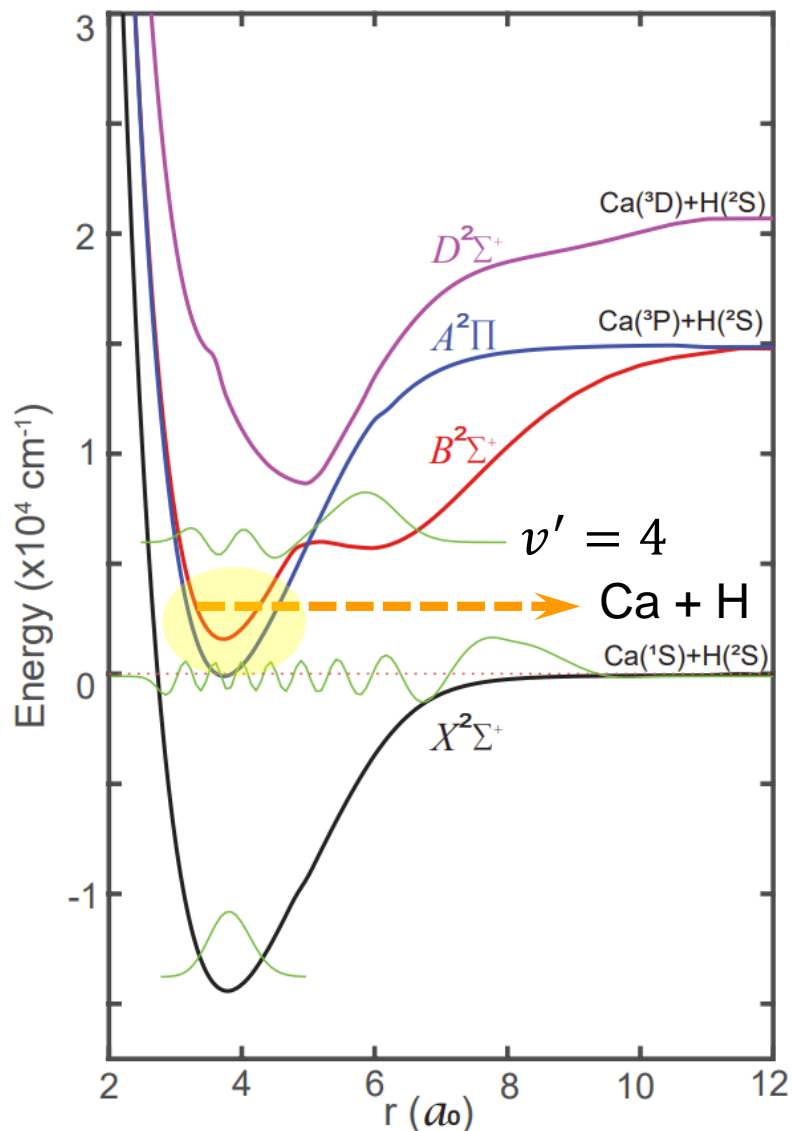
Produce ultracold gases  
of photofragments  
from precursor molecules?





# Dissociation of CaH molecules

Natural predissociation + 3D laser cooling



Anastassia Alexandrova



Claire Dickerson



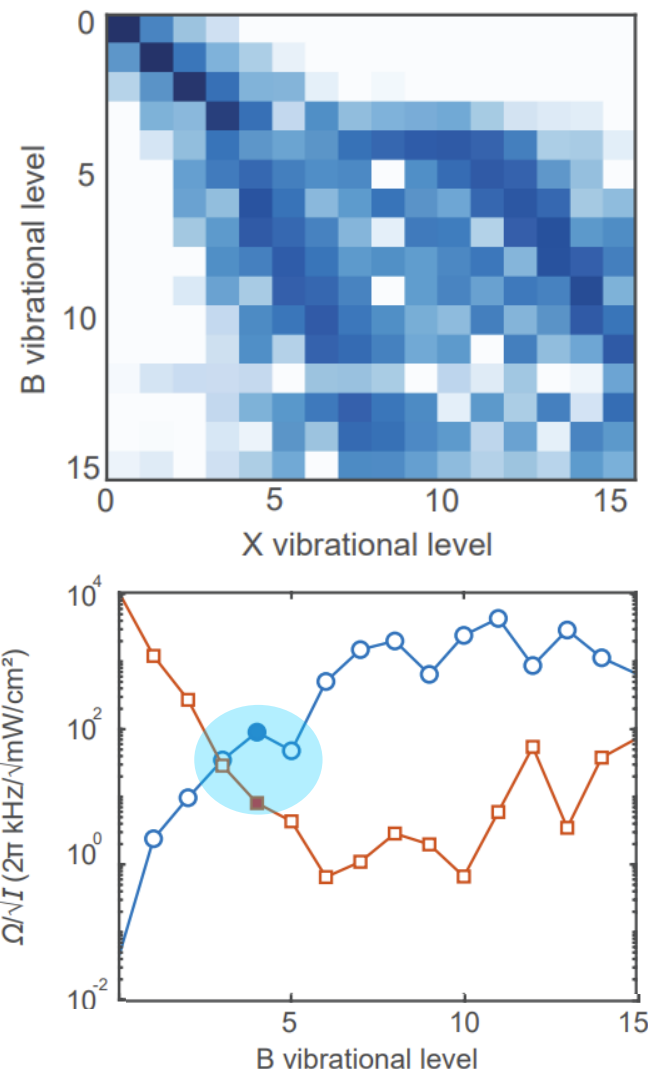
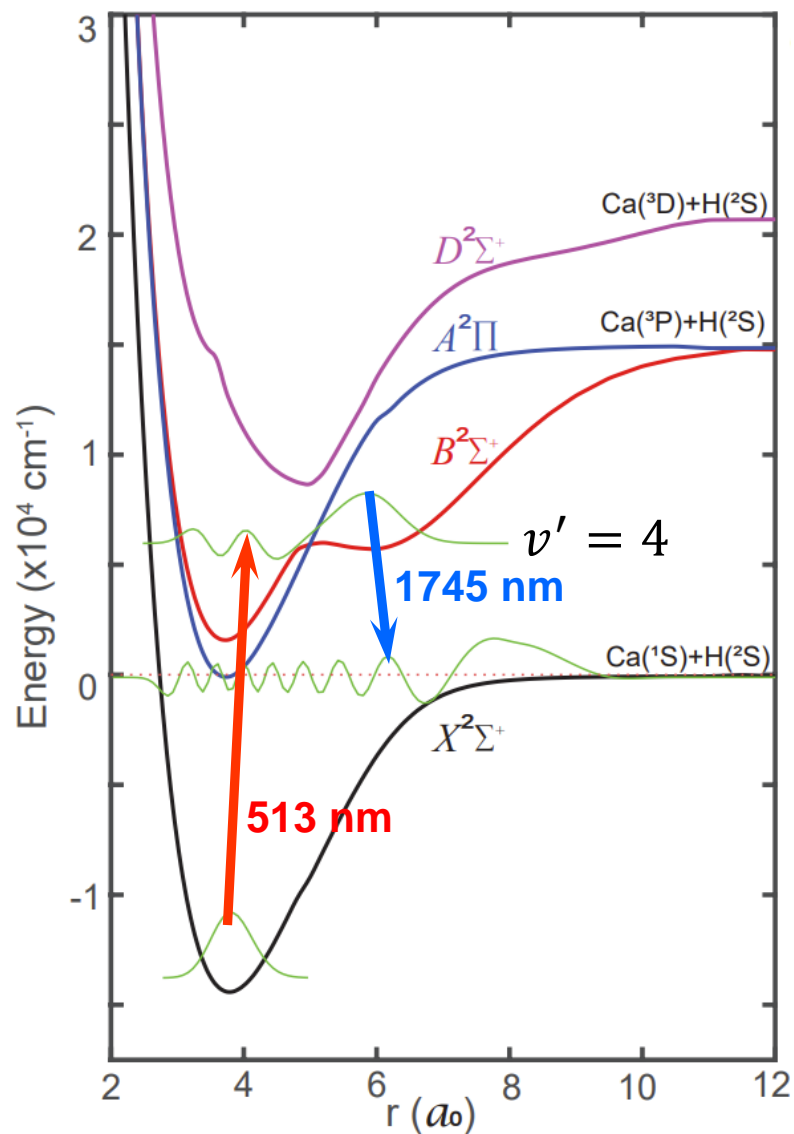
Daniel Neuhauser



Lan Cheng

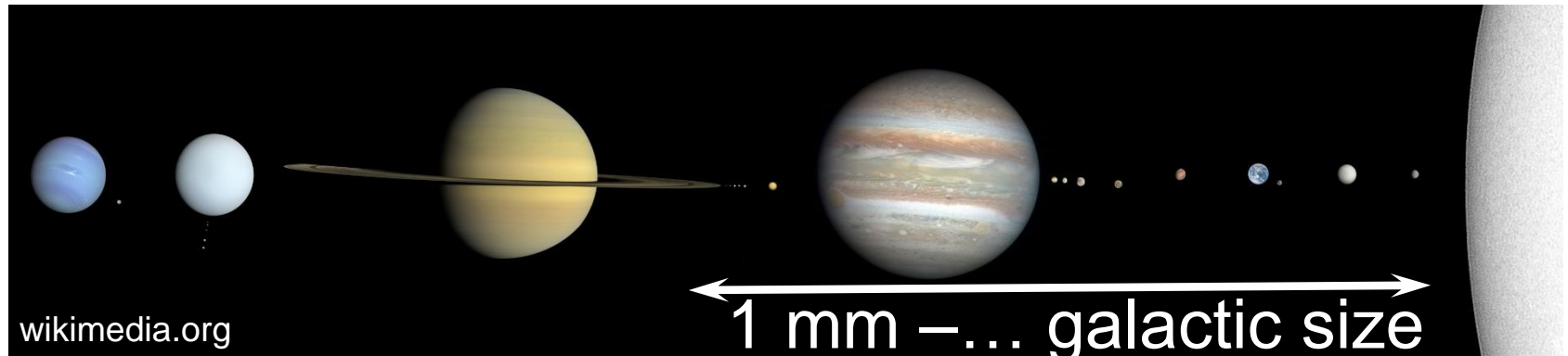
# Dissociation of CaH molecules

## Controlled dissociation

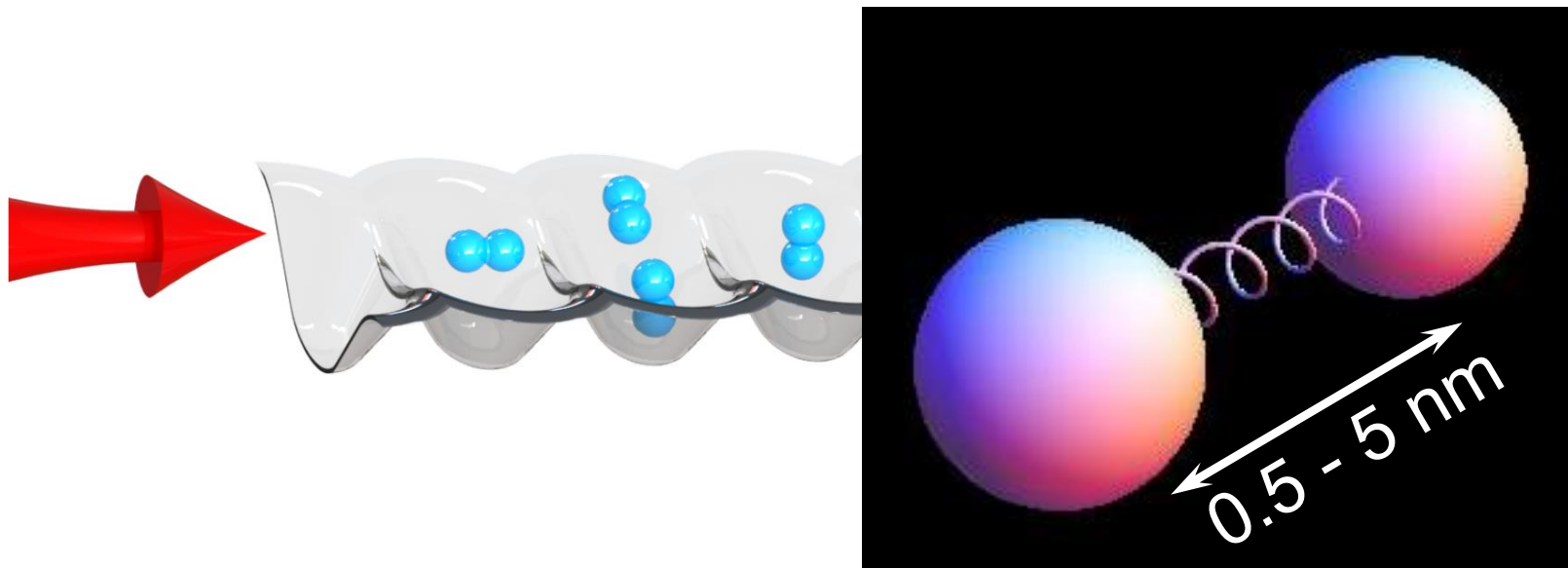


# Science with molecular clocks

Newtonian gravity at large scales: Well-understood



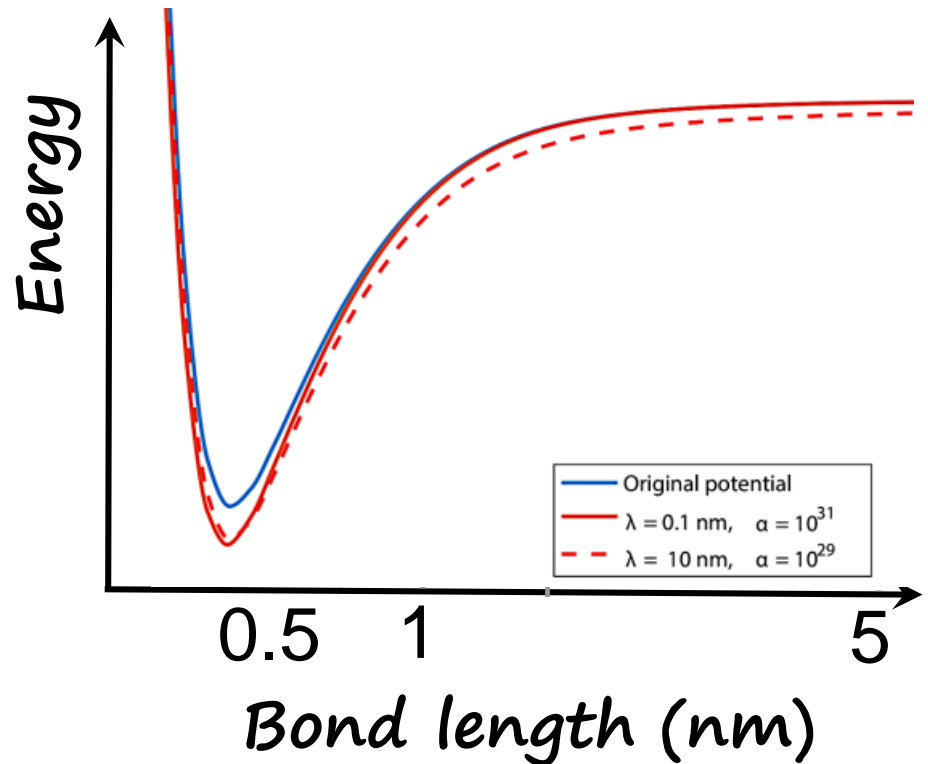
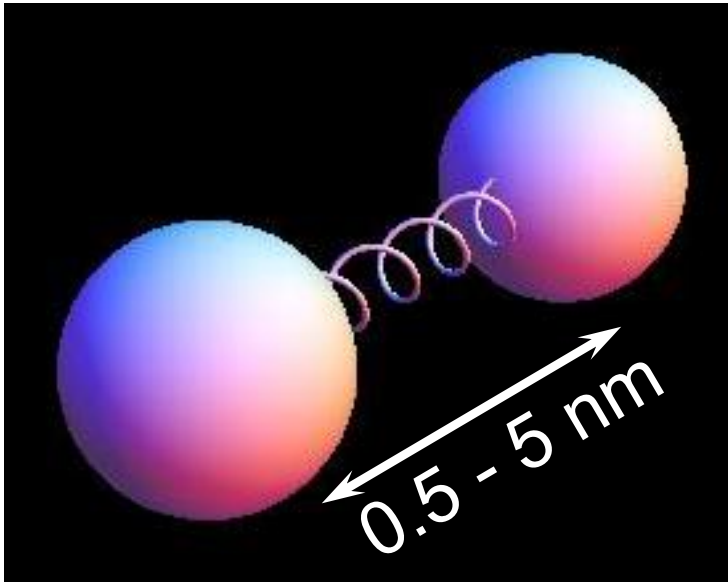
Gravity at nanometer scales: Nearly unknown





# Science with molecular clocks

## Nanoscale mass-dependent forces

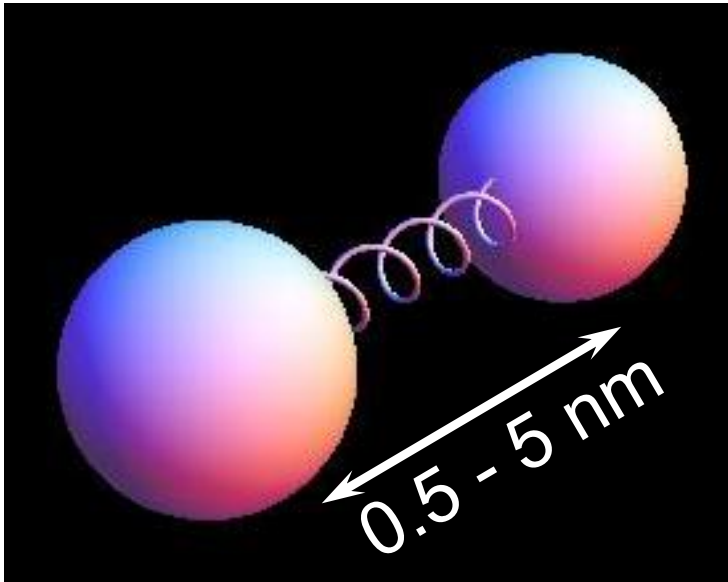


$$V = -\frac{GM^2}{r} \left( 1 + \underbrace{Ae^{-r/\lambda}} \right)$$

Hypothetical  
“Yukawa” correction

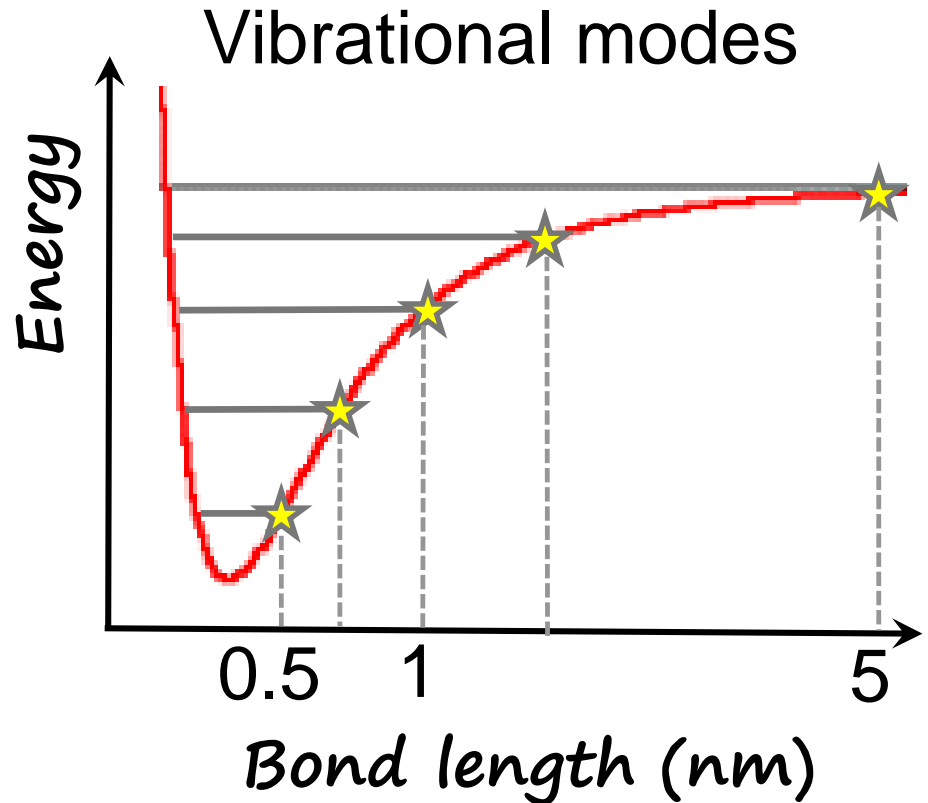
# Science with molecular clocks

## Nanoscale mass-dependent forces



$$V = -\frac{GM^2}{r} \left( 1 + \underbrace{Ae^{-r/\lambda}}_{\text{Hypothetical}} \right)$$

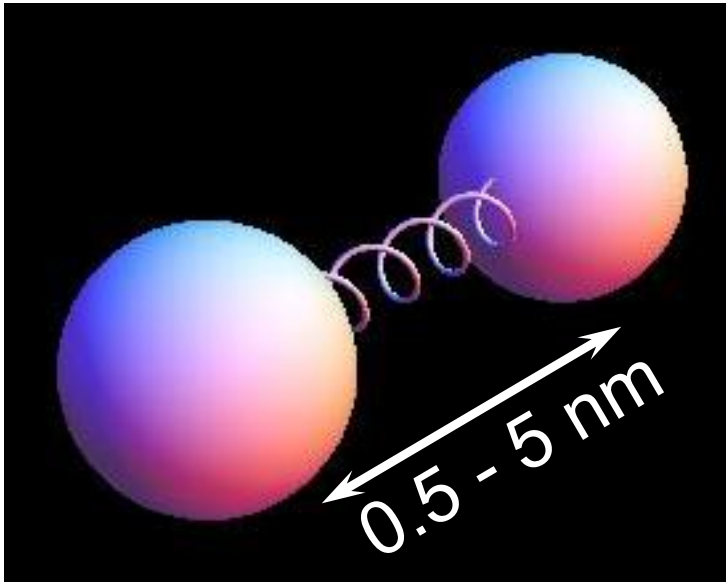
Hypothetical  
“Yukawa” correction



Explore a range of lengths

# Science with molecular clocks

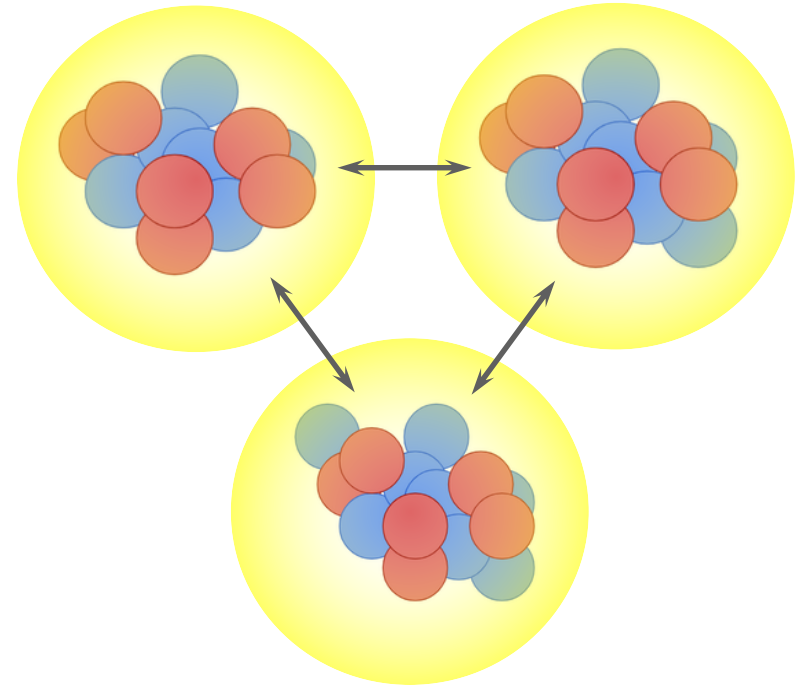
## Nanoscale mass-dependent forces



$$V = -\frac{GM^2}{r} \left( 1 + \underbrace{Ae^{-r/\lambda}} \right)$$

Hypothetical  
“Yukawa” correction

Isotopes  
of strontium atom

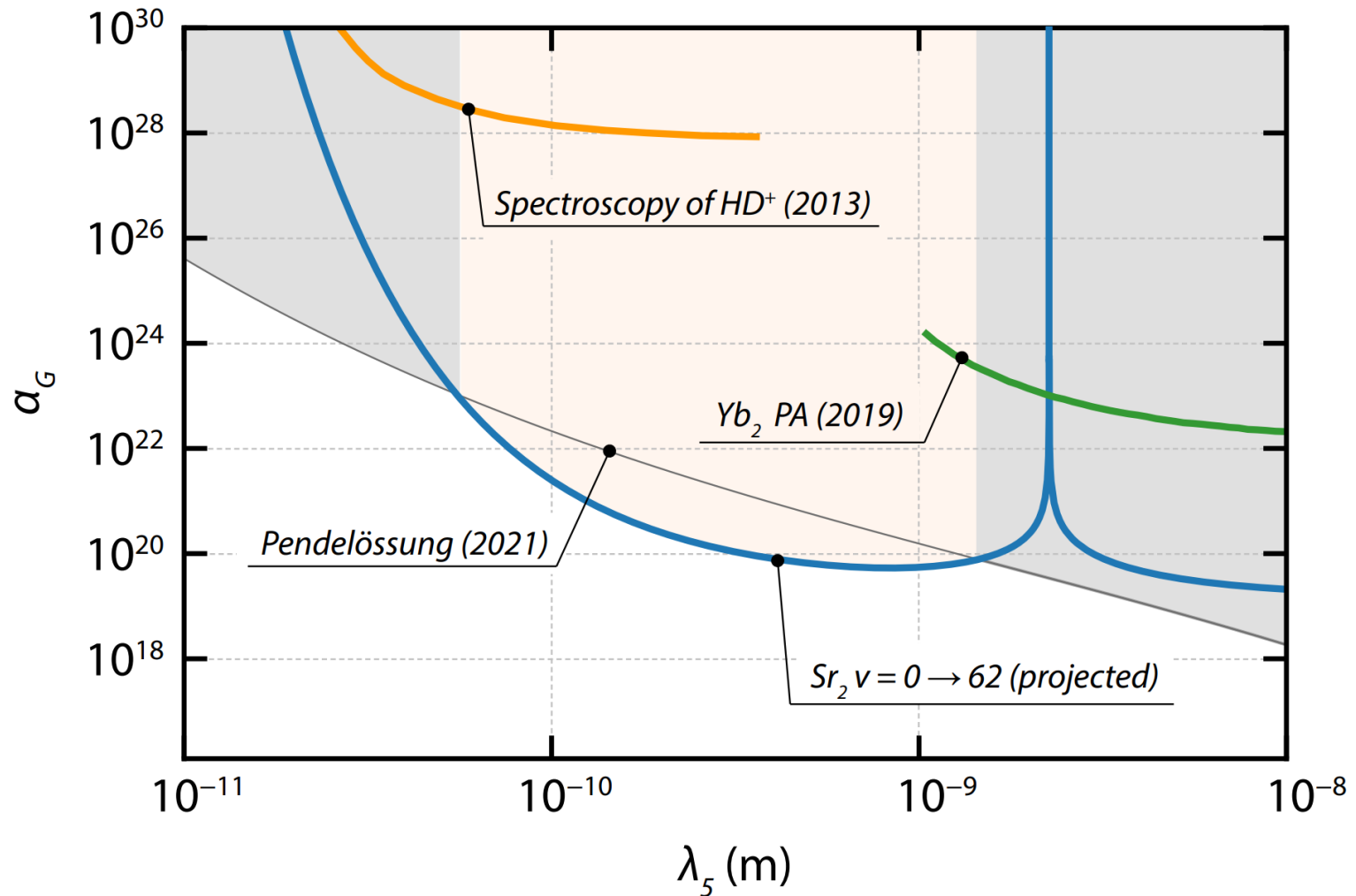


Explore a range of masses



# 5<sup>th</sup> force with molecular clock

*Current experimental capability*



E. Tiberi, *Ph.D. thesis* (2023)

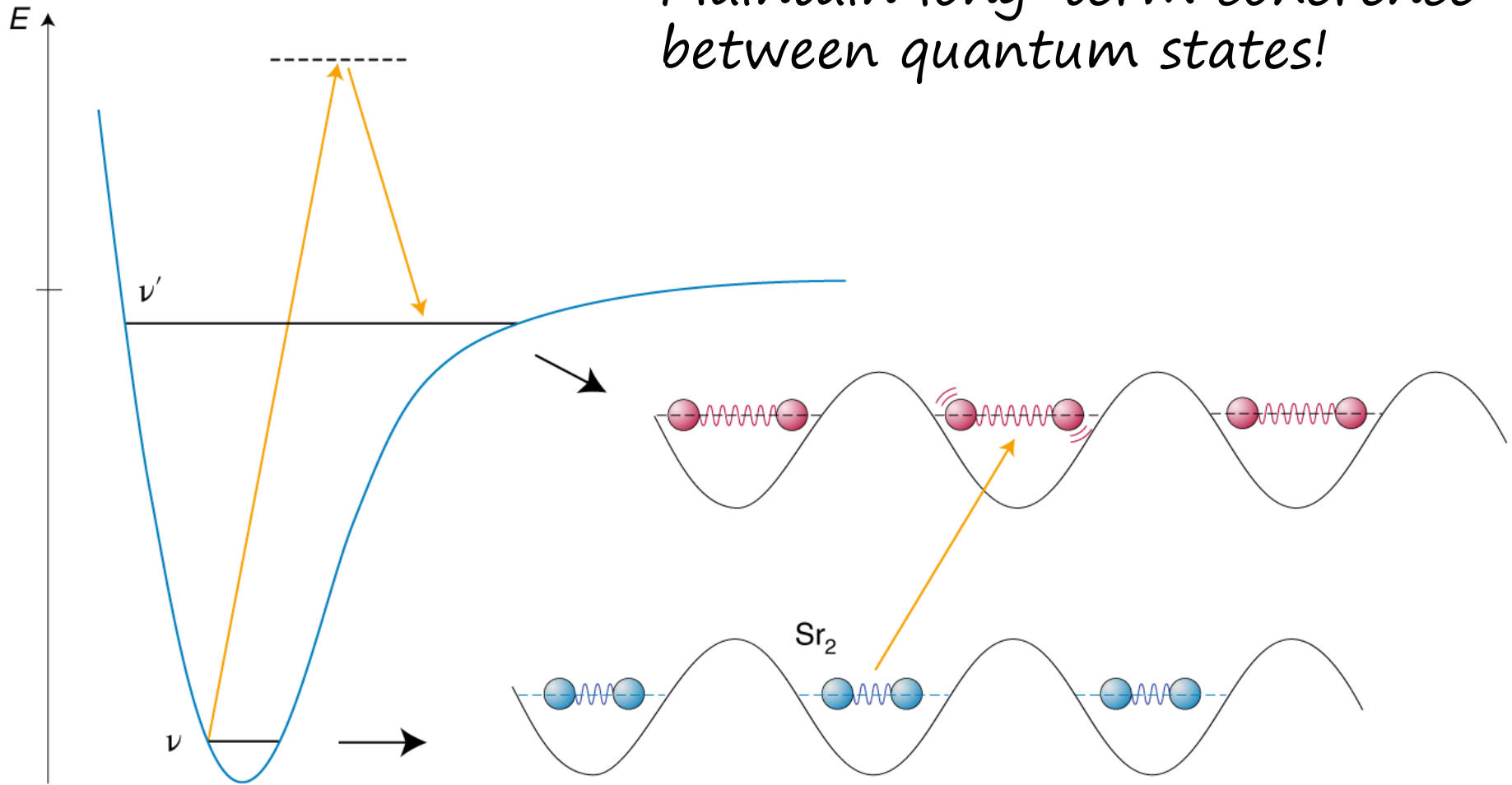
B. Heacock *et al.*, *Science* **373**, 1239 (2021)

E. J. Salumbides *et al.*, *PRD* **87**, 112008 (2013)

M. Borkowski *et al.*, *Sci. Rep.* **9**, 14807 (2019)

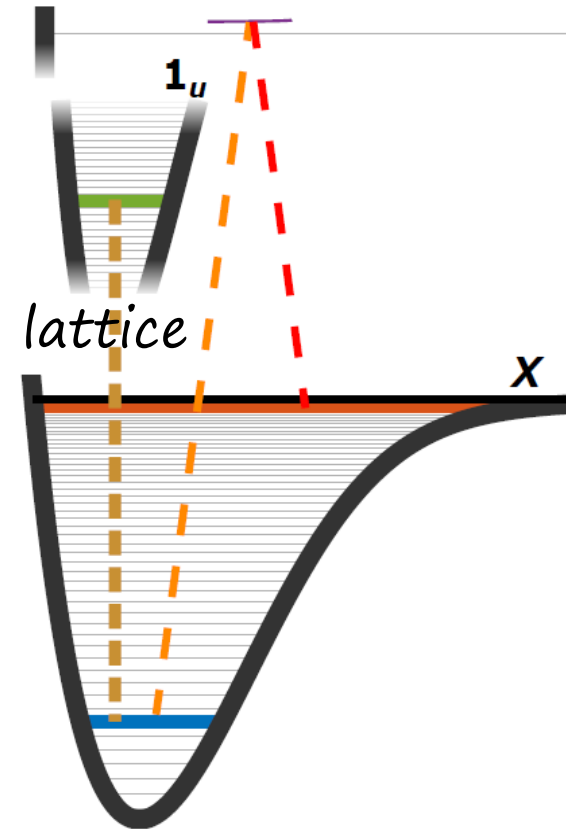
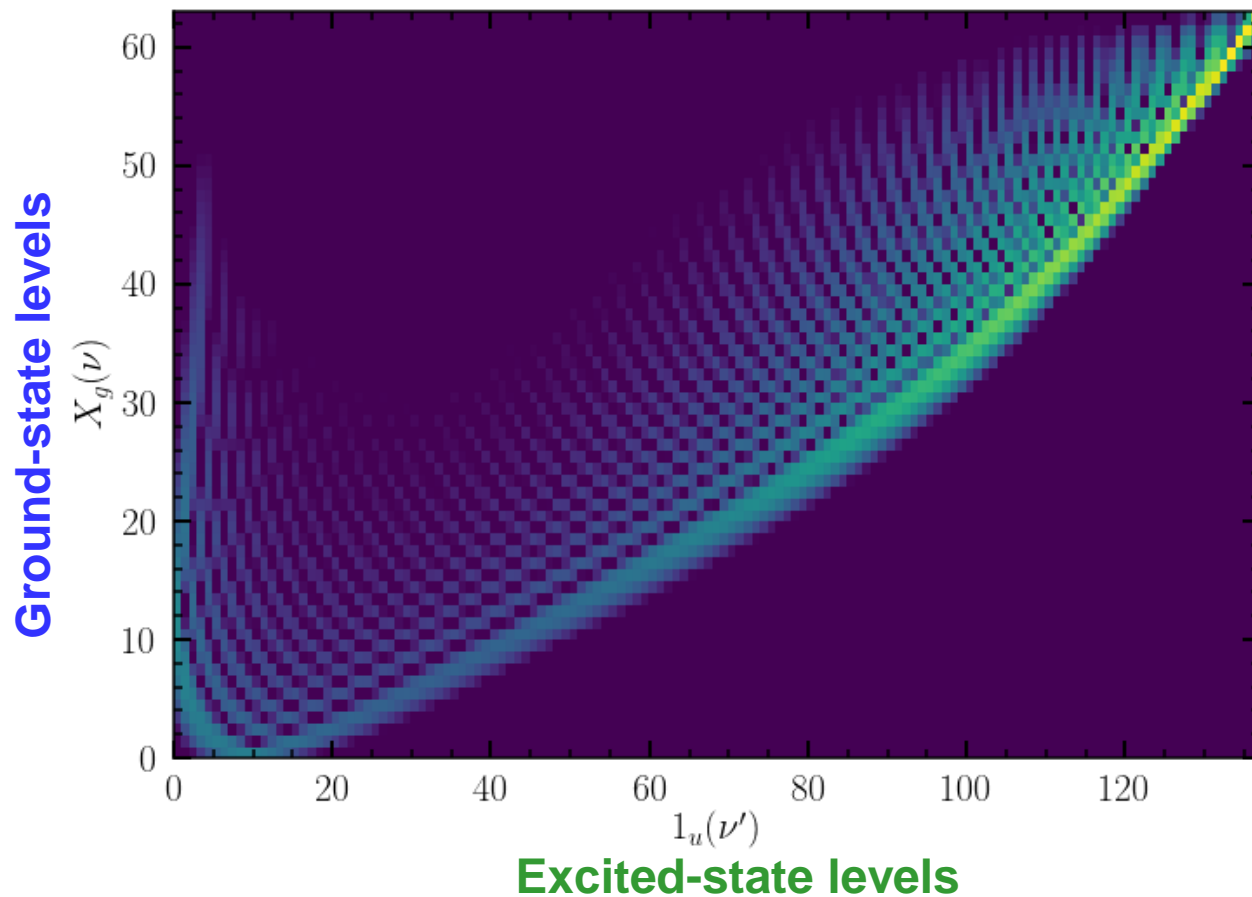
# Molecular lattice clock

Maintain long-term coherence between quantum states!



# Magic wavelengths

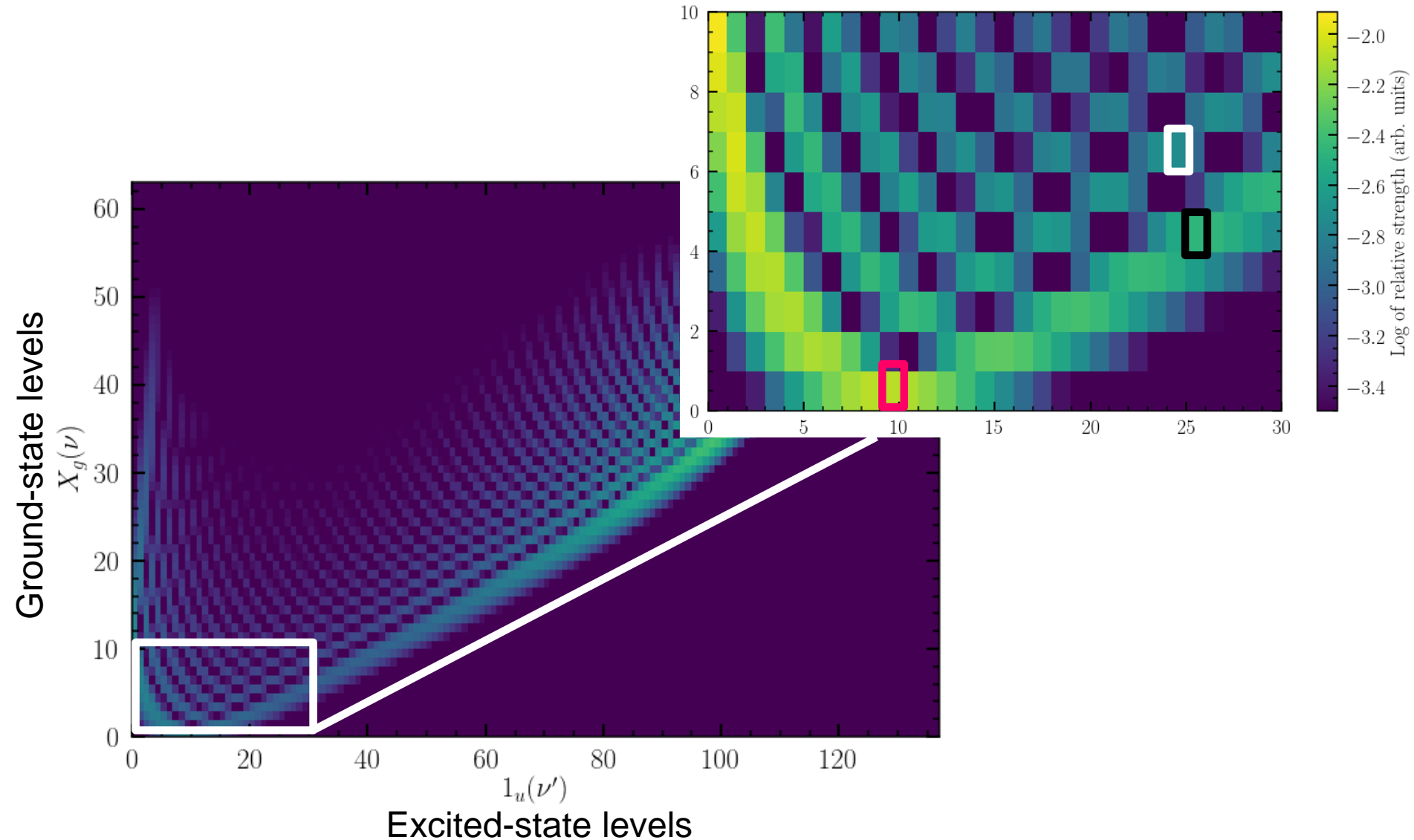
Trap light scattering limits coherence: Choose best  $\lambda$ !





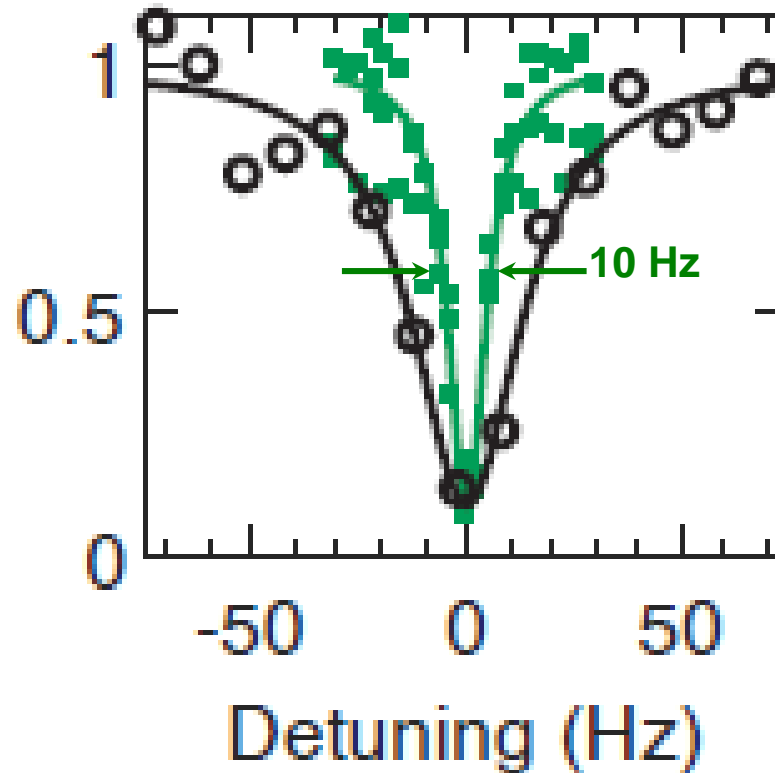
# Magic wavelengths

Trap light scattering limits coherence: Choose best  $\lambda$ !



# Molecular lattice clock

Coherence in magic lattice :  $\times 10^4!$



$$Q = 3 \times 10^{12}$$

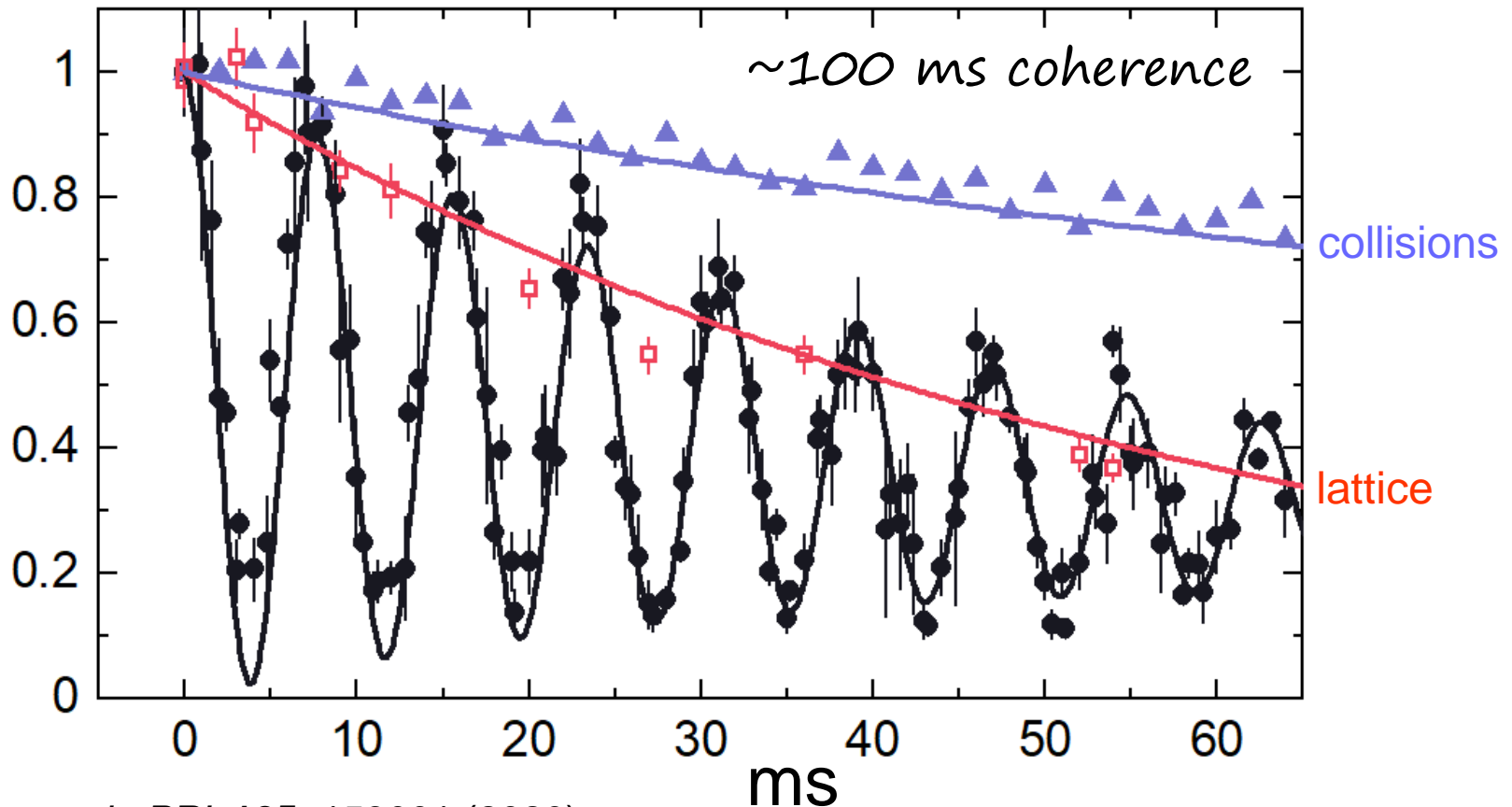
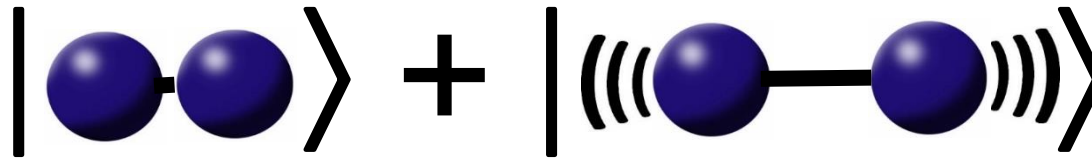
$$Q \text{ (intrinsic)} > 10^{26}$$

# Molecular lattice clock

*Coherence in magic lattice*

$V = 4$

$V = 62$



# Clock precision & accuracy

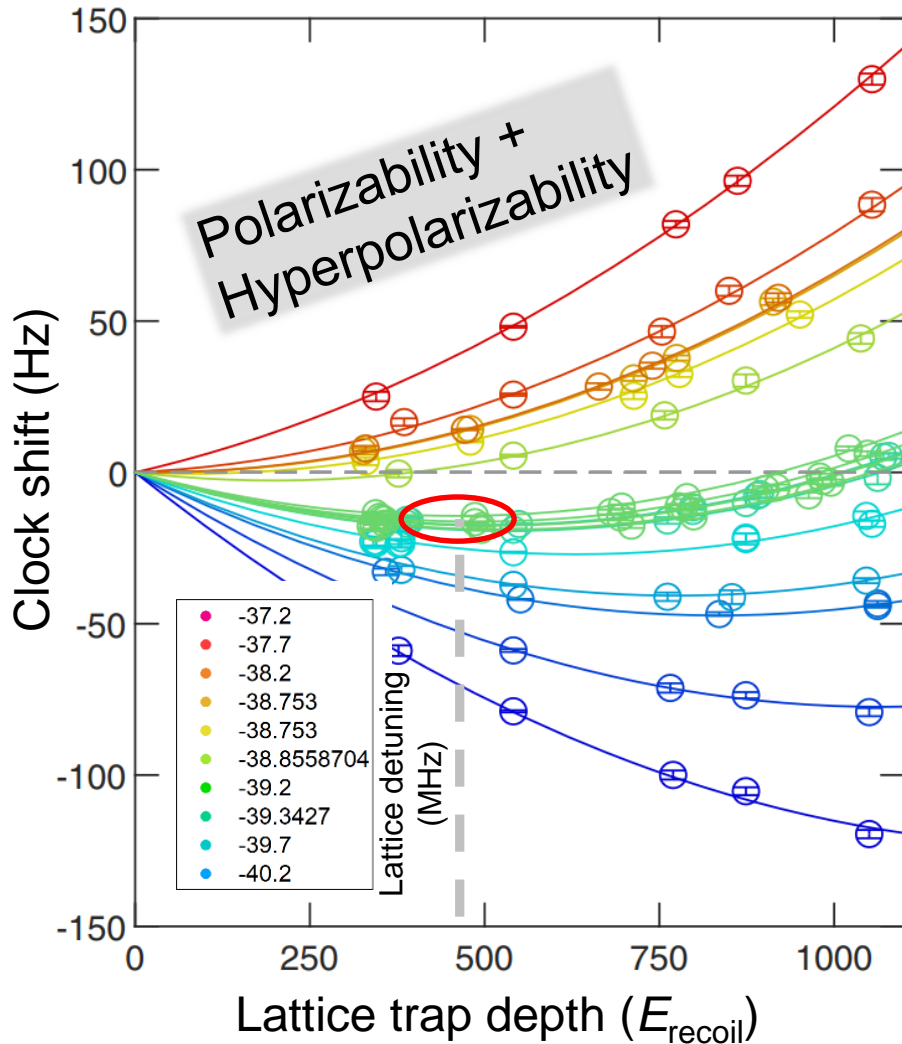
*Systematic effects:*

*What can cause the clock frequency to shift?*

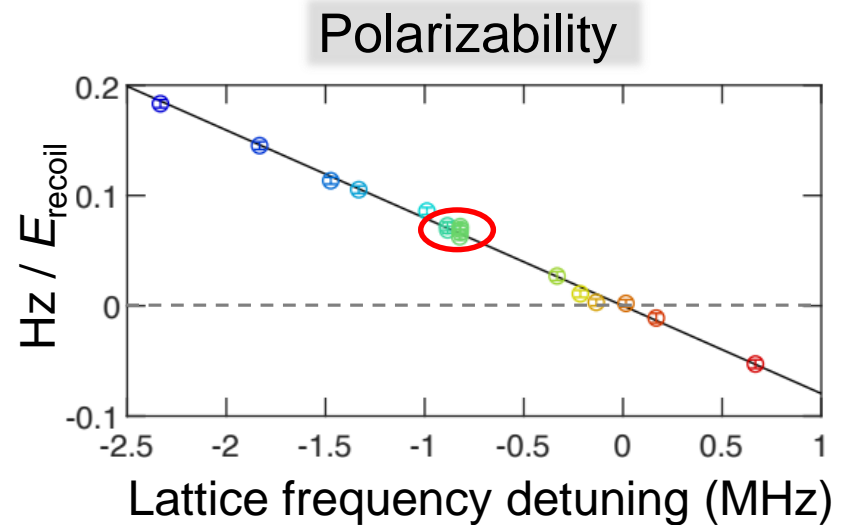
$\times 10^{-14}$

Systematic	Correction	Uncertainty
● Lattice Stark ( $E1, M1, E2$ )	100.1	3.4
● Lattice Stark (hyperpolarizability)	-50.8	1.9
● Probe Stark (total)	31.5	2.2
● BBR	-2.2	0.4 $\times$
● Density	-0.6	0.3
Quadratic Zeeman	0	0.05
dc Stark	0	< 0.1
Doppler and phase chirps	0	< 1
Lattice tunneling	0	< 0.1
Line pulling	0	< 0.1
Scan-and-fit	0	< 0.6
<b>Total</b>	77.9	<b>4.6</b>

# Clock shifts: Lattice light intensity



operationally  
magic  
lattice

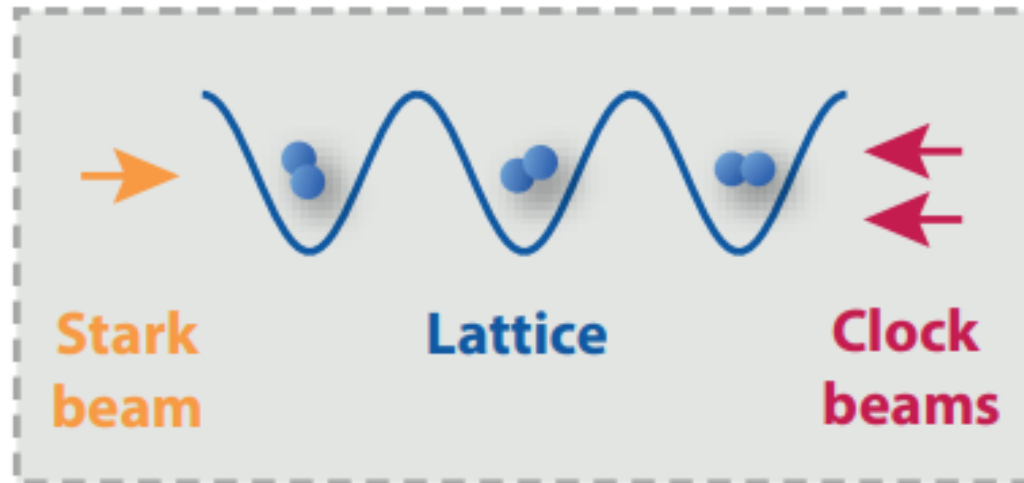


$\sim 4 \times 10^{-14}$  uncertainty



# Clock shifts: BBR

*Purely vibrational BBR shifts?*

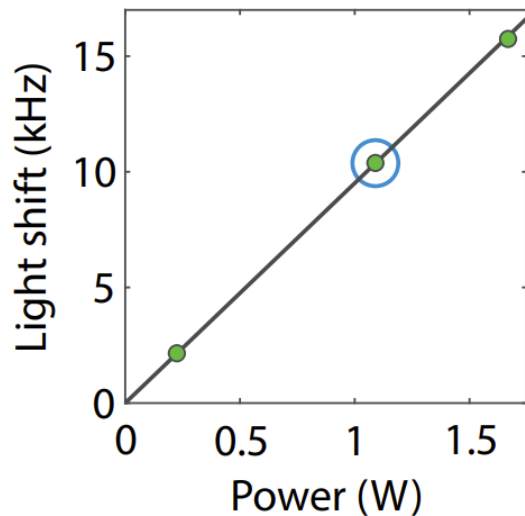
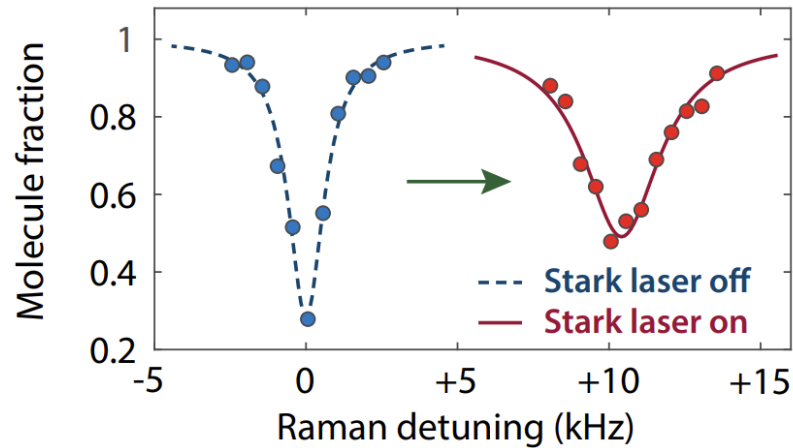


Wojtek Skomorowski

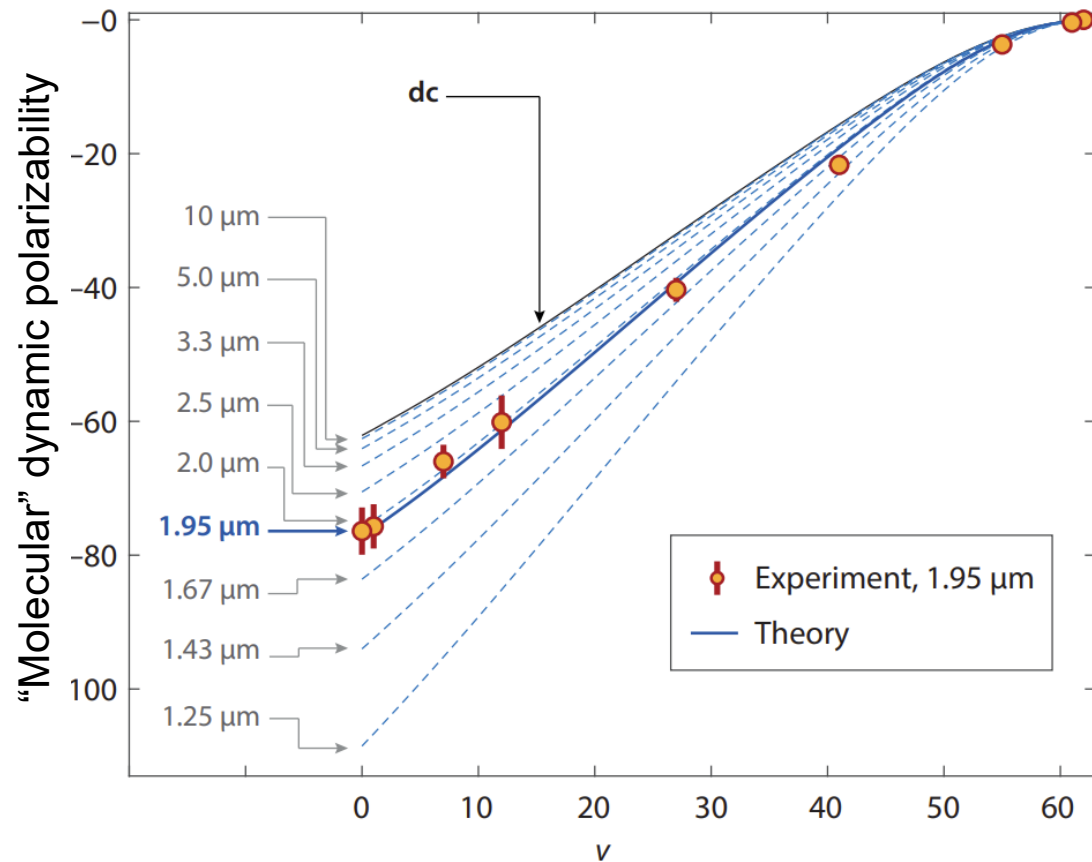
# Molecular polarizabilities

*DC to infrared*

IR Stark shift measurement (2  $\mu\text{m}$ )  
using clock transitions



IR Stark shifts, all vibrational states:  
Measurement & calculations



# Molecular polarizabilities

*DC to infrared*

IR Stark shifts, all vibrational states:  
Measurement & calculations

Net BBR shift:

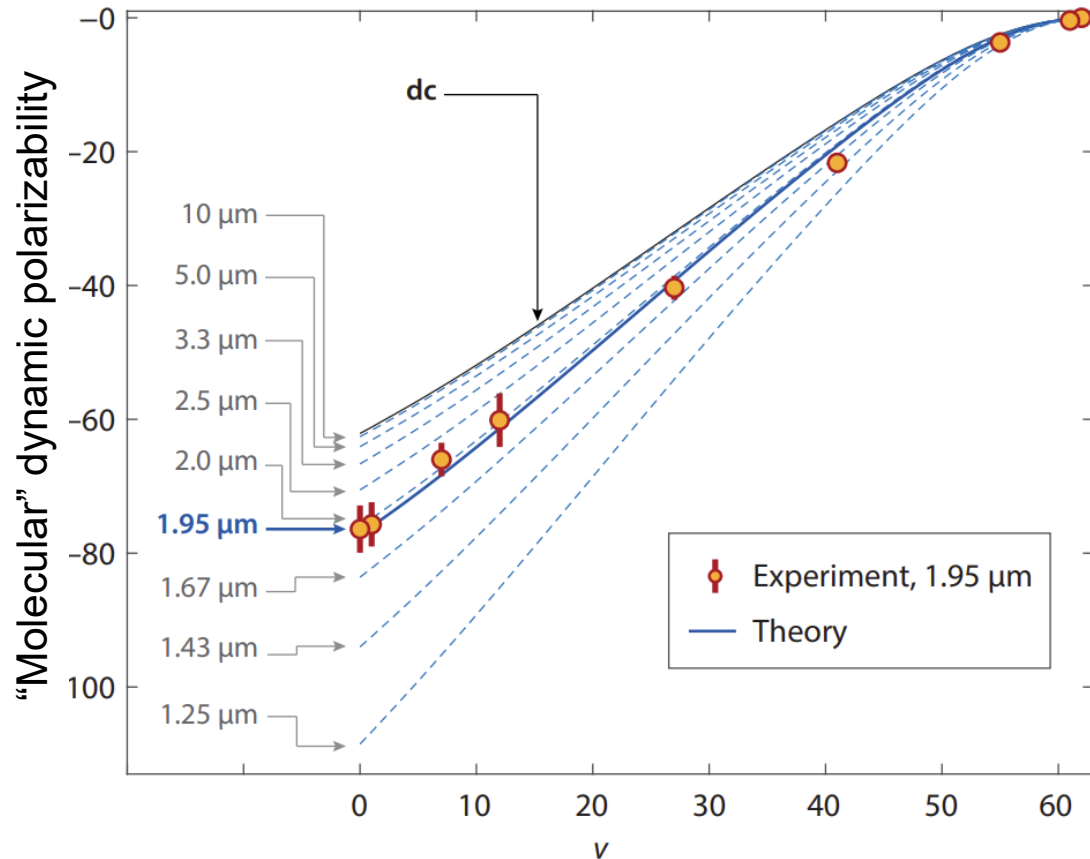
$$\mathcal{O}(10^{-14})$$

BBR shift uncertainty:

$$< 5 \times 10^{-16}$$

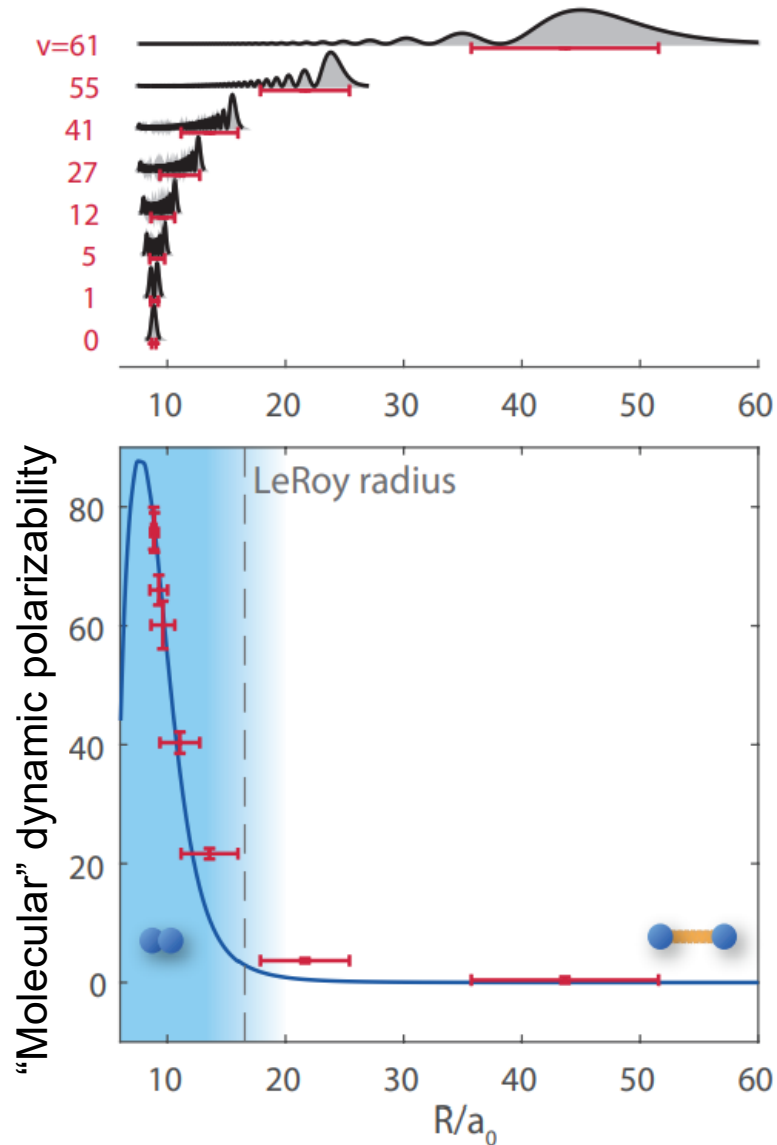
Dynamic correction:

$$\eta \sim 0.5\%$$



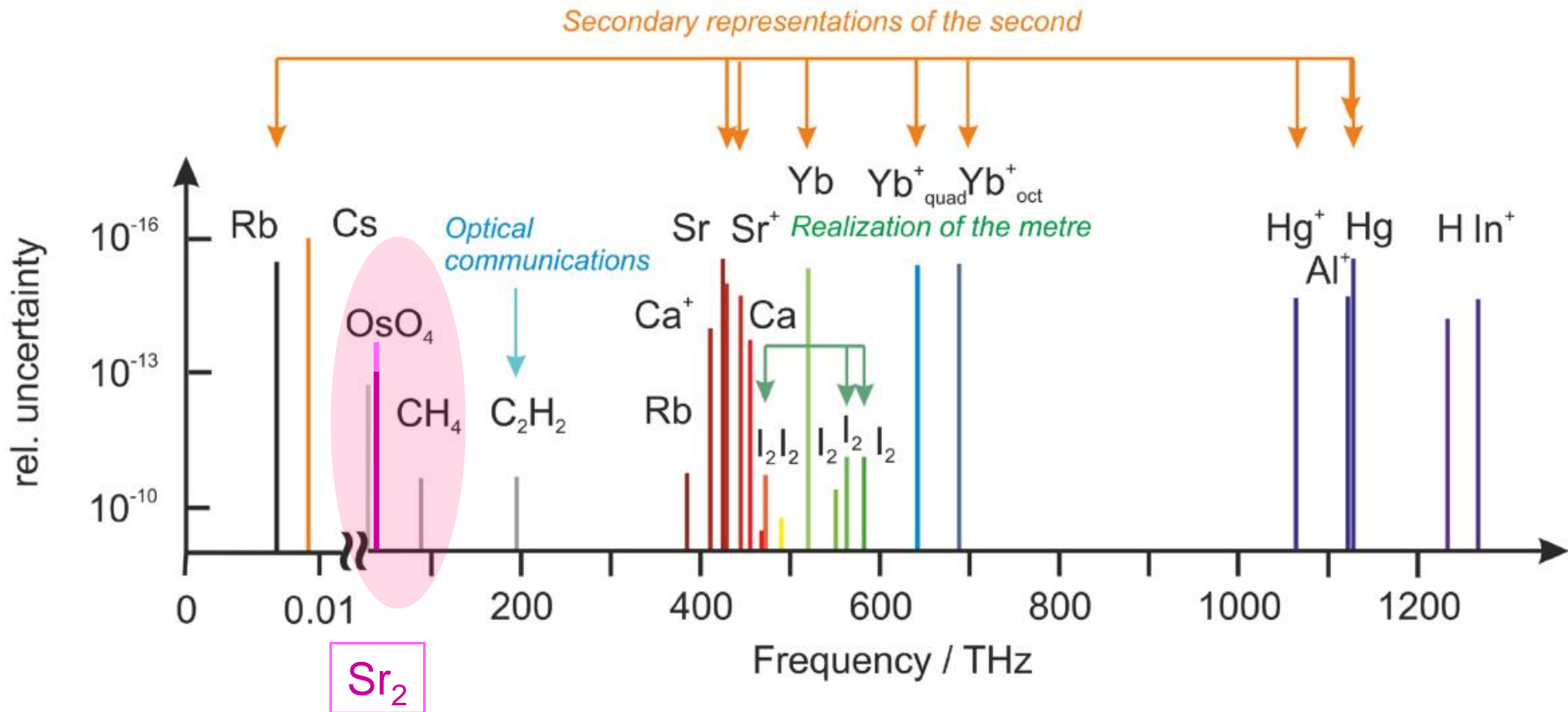
# Molecular polarizabilities

*Physicists' vs. chemists' molecules*



# Frequency standards

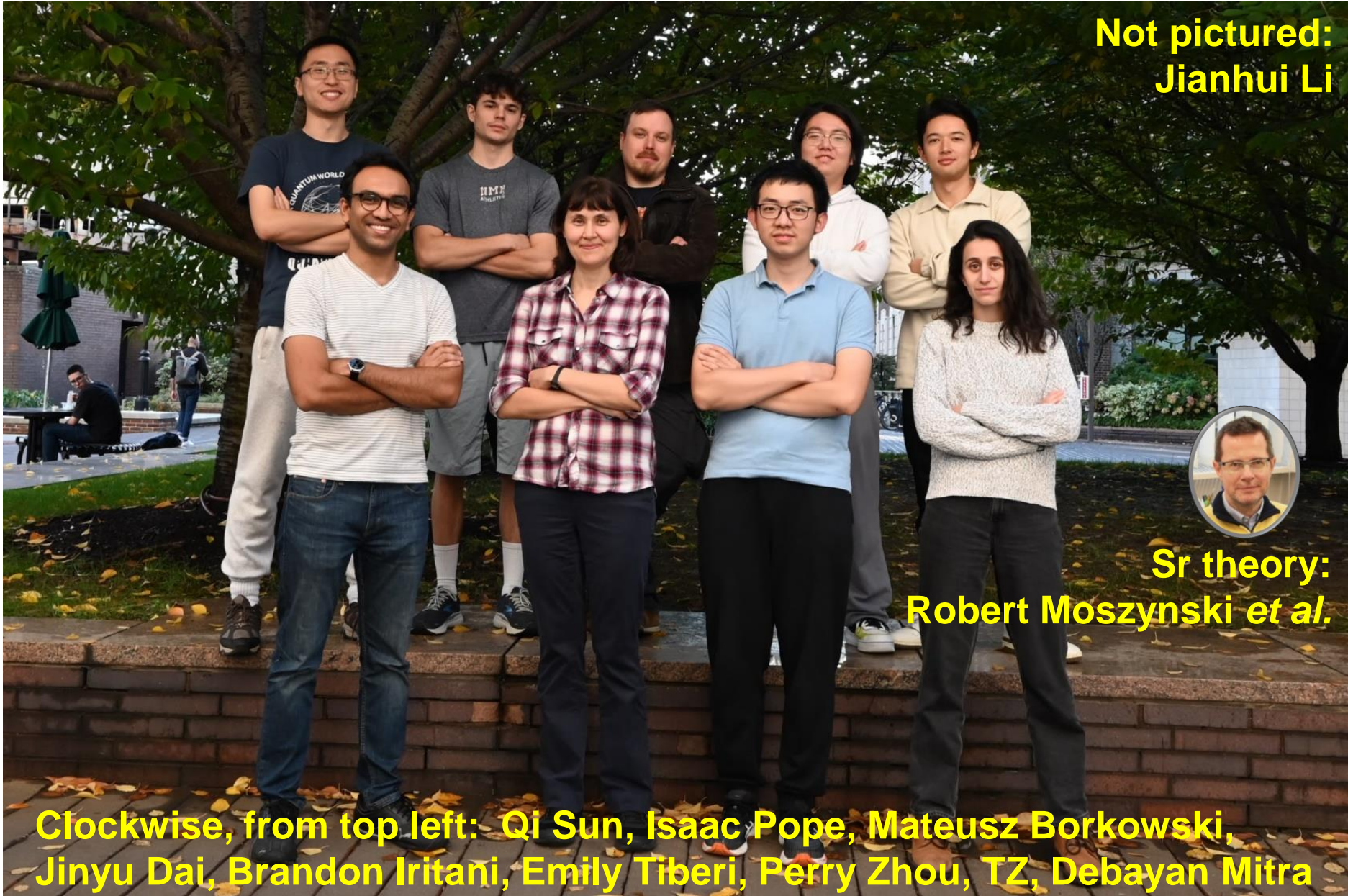
2017 recommendations





# ZLab

Not pictured:  
Jianhui Li



Sr theory:  
Robert Moszynski *et al.*

Clockwise, from top left: Qi Sun, Isaac Pope, Mateusz Borkowski,  
Jinyu Dai, Brandon Iritani, Emily Tiberi, Perry Zhou, TZ, Debayan Mitra

# Poster presenters

Mateusz Borkowski



**Sr<sub>2</sub> lattice clock  
(Monday #38)**

Eliot Bohr



**Superradiance-enhanced  
Ramsey spectroscopy  
(Tuesday #50)**

Sofus L. Kristensen



**Superradiant optical line narrowing  
(Thursday #108)**

Stefan A. Schäffer



**Coherent control of noise  
in collective emission  
(Thursday #109)**