

Title: Clocks and precision measurements with ultracold molecules

Speakers: Tanya Zelevinsky

Series: Colloquium

Date: March 13, 2024 - 2:00 PM

URL: <https://pirsa.org/24030111>

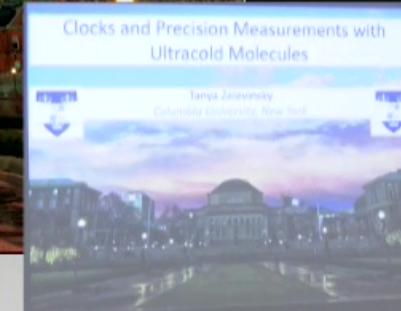
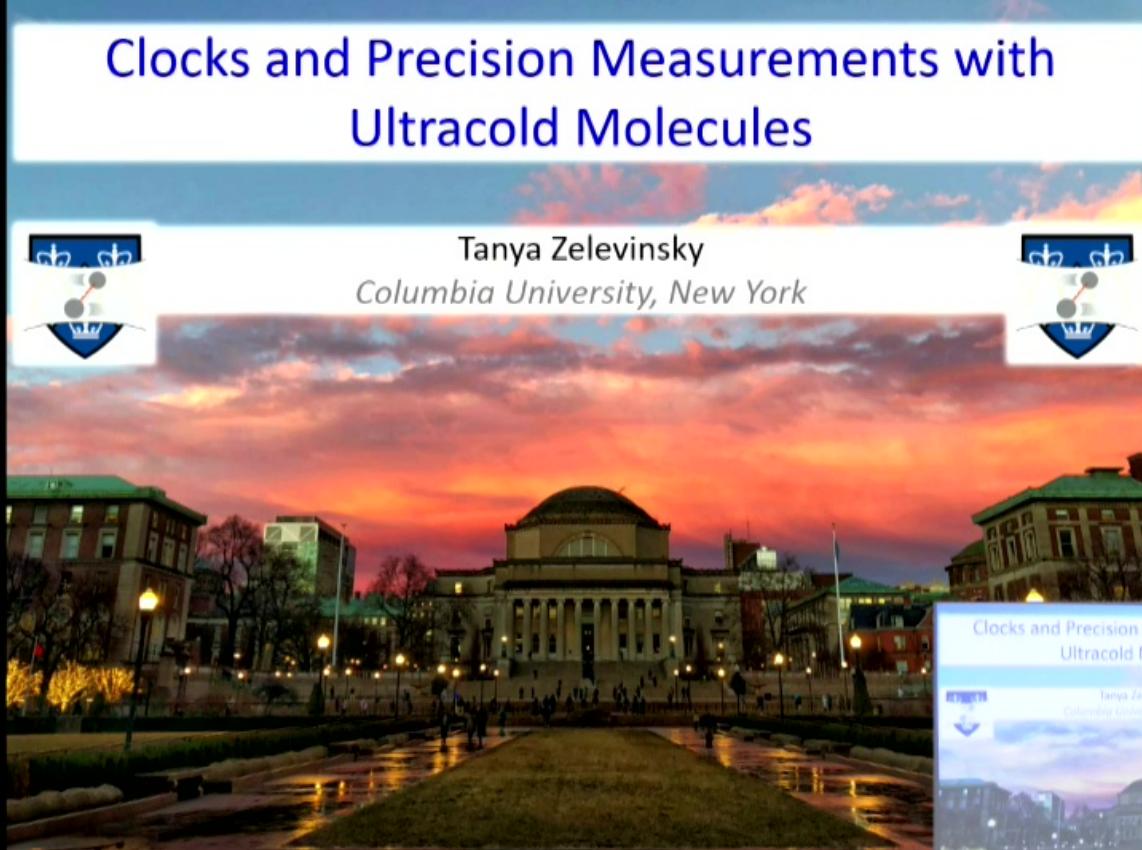
Abstract: Significant advancements in our understanding of the physical world have been driven by increasingly precise atomic spectroscopy. The level of accessible precision entered a new realm with the advent of laser cooling and trapping. Now we can extend the ultrahigh spectroscopic precision, or atomic clock technology, to more complex quantum particles like diatomic molecules. The ability to quantify molecular degrees of freedom, such as nuclear vibrations, with nearly atomic-clock precision illuminates their previously hidden properties. Moreover, it suggests possibilities to leverage this precision for probing fundamental aspects of physical interactions, including enhanced tests of Newtonian gravity at the nanometer scale.

Zoom link

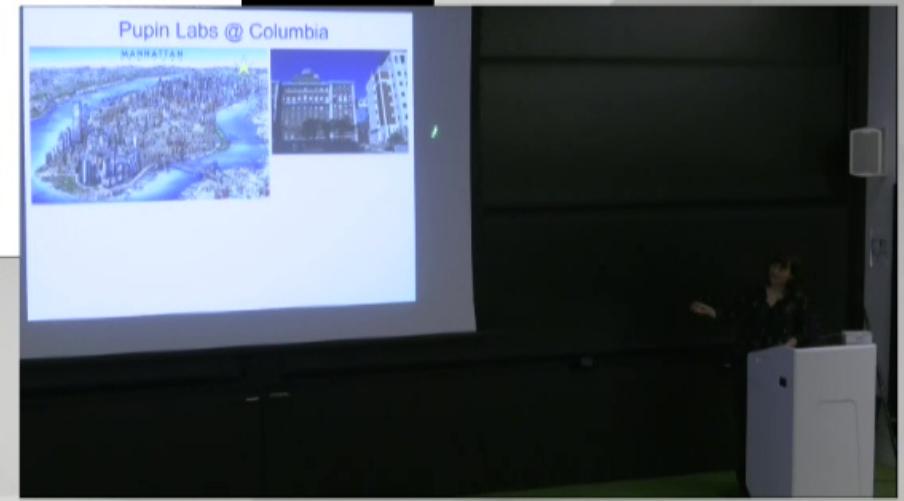
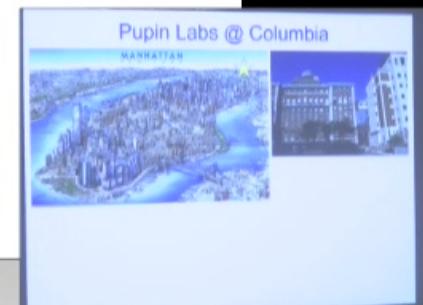
Clocks and Precision Measurements with Ultracold Molecules



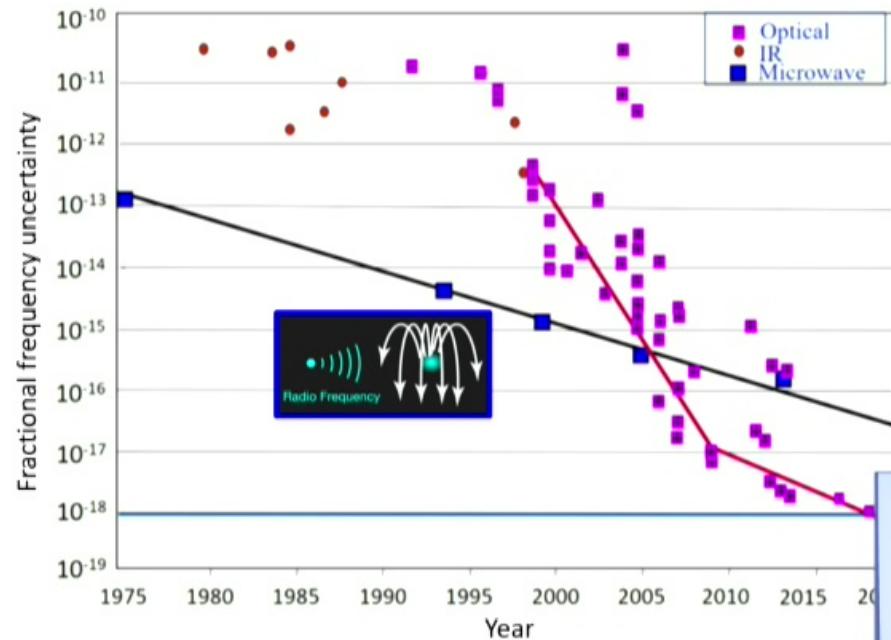
Tanya Zelevinsky
Columbia University, New York



Pupin Labs @ Columbia

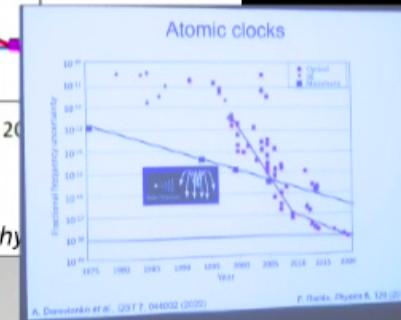


Atomic clocks

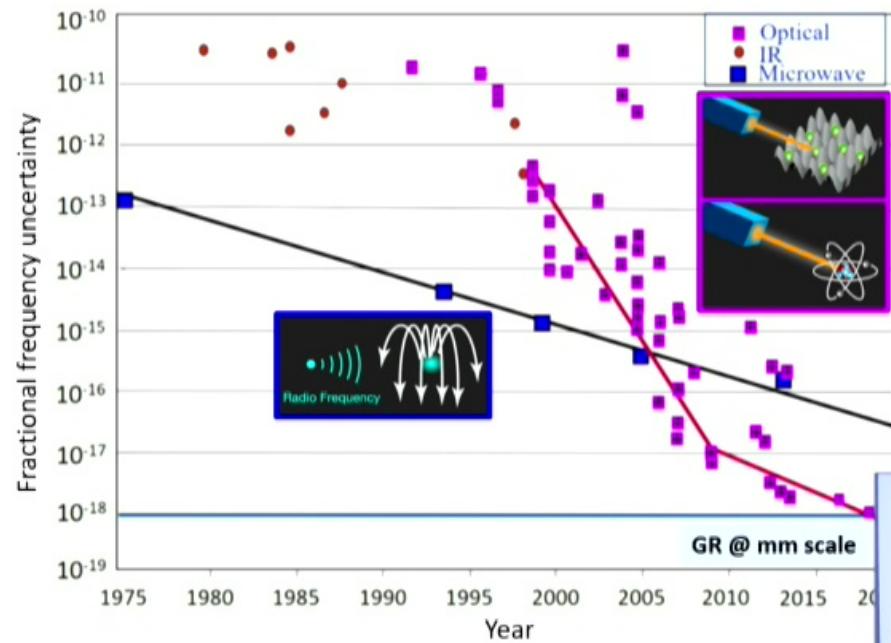


A. Derevianko *et al.*, QST 7, 044002 (2022)

F. Riehle, *Phys.*

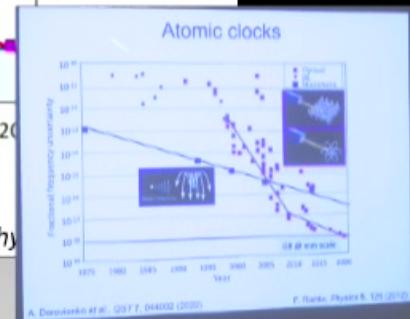


Atomic clocks

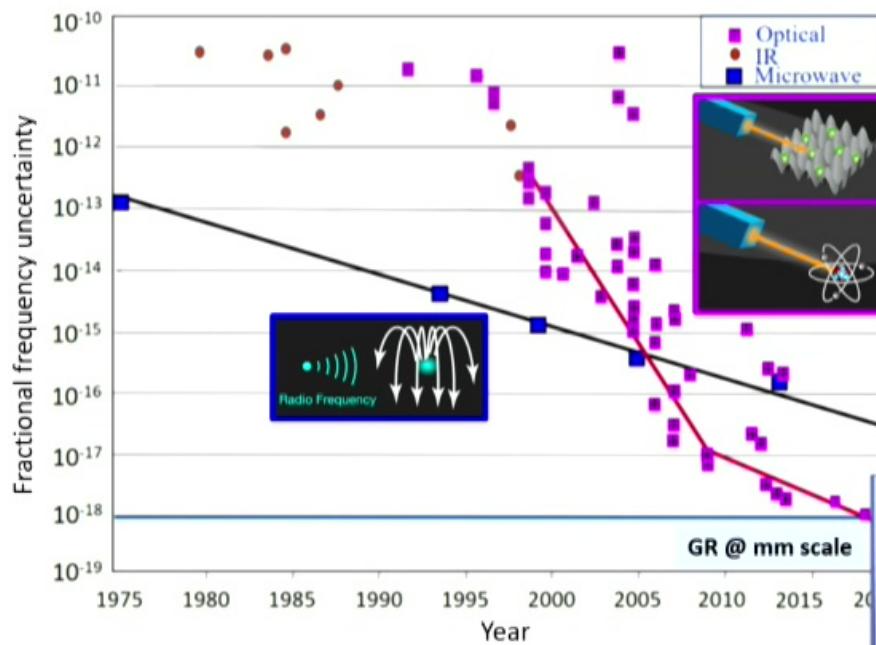


A. Derevianko et al., QST 7, 044002 (2022)

F. Riehle, Phys.

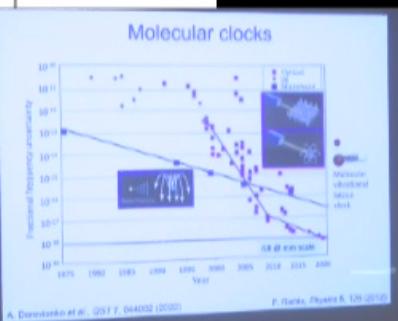


Molecular clocks

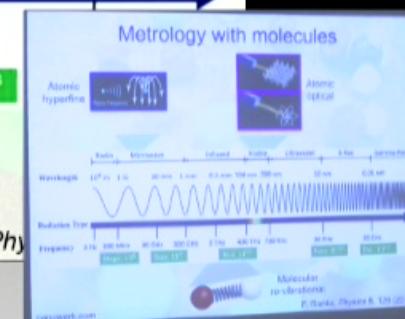
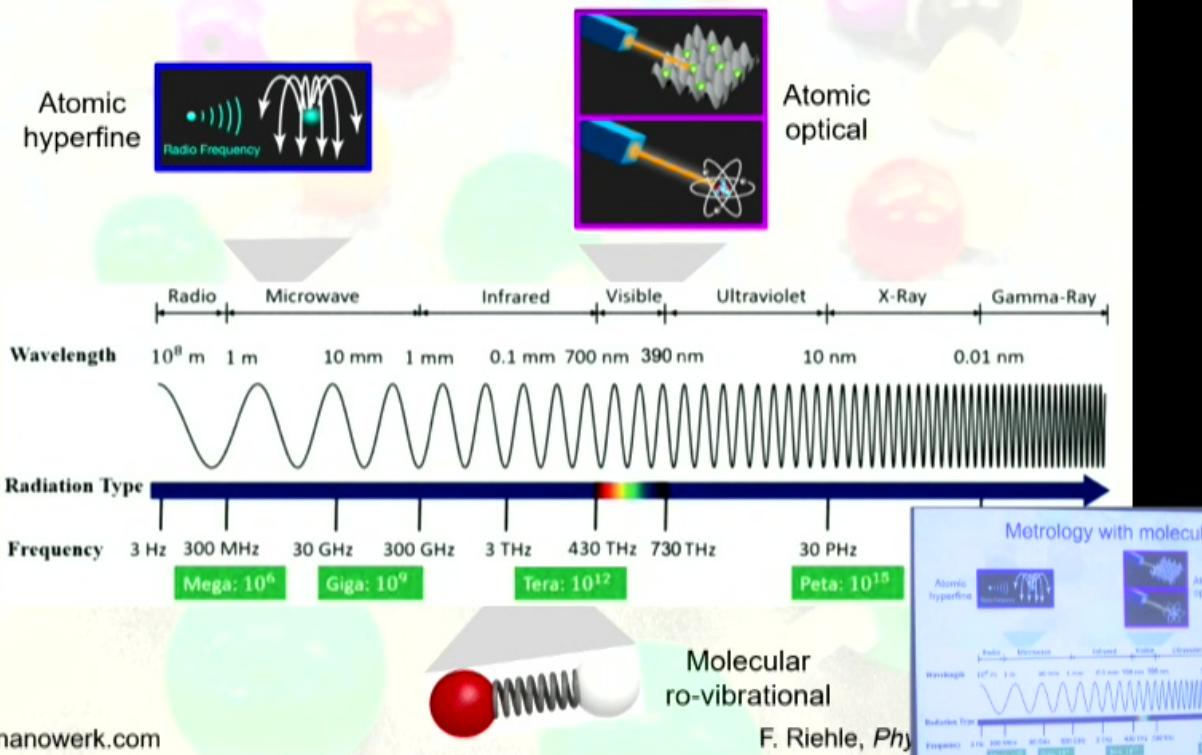


A. Derevianko et al., QST 7, 044002 (2022)

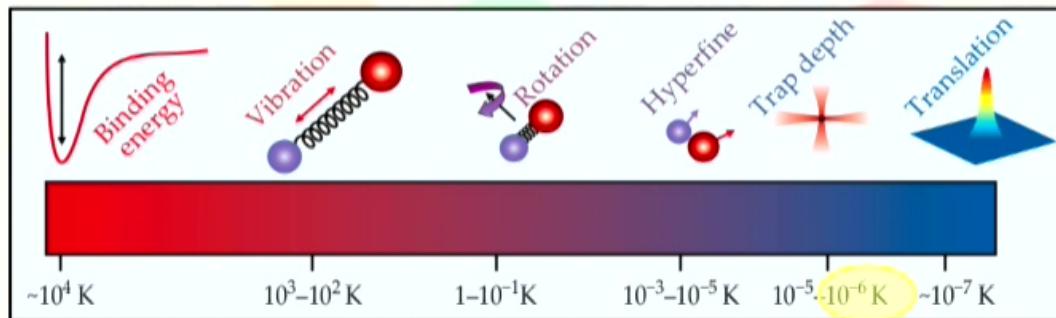
F. Riehle, Phys.



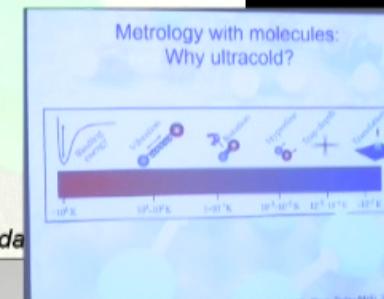
Metrology with molecules



Metrology with molecules: Why ultracold?

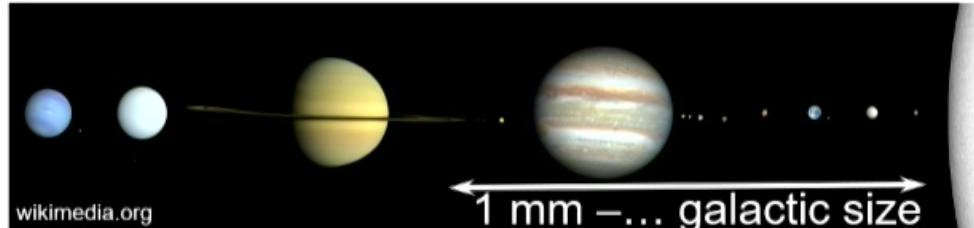


D. S. Jin and J. Ye, *Phys. Today* 64(5), 37 (2011)

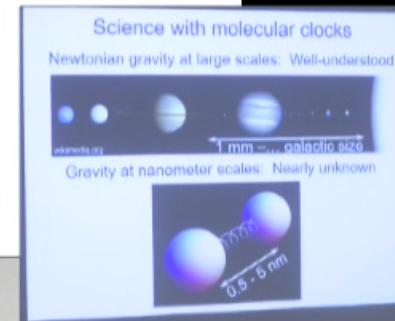
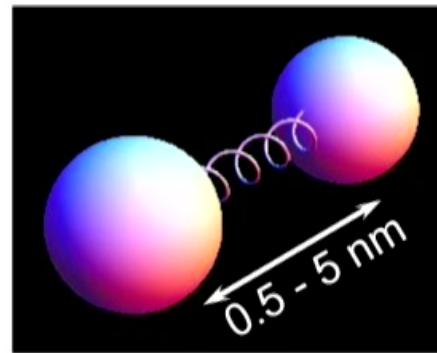


Science with molecular clocks

Newtonian gravity at large scales: Well-understood

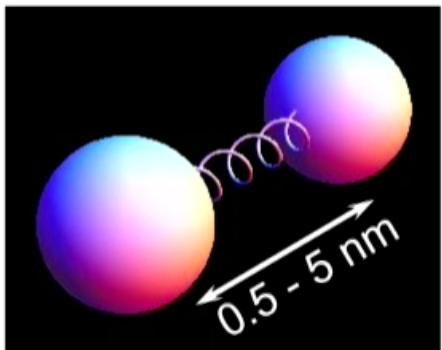


Gravity at nanometer scales: Nearly unknown

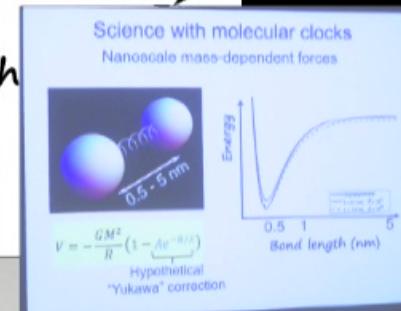
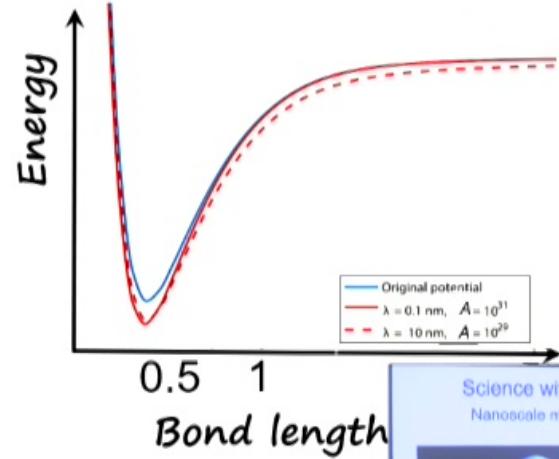


Science with molecular clocks

Nanoscale mass-dependent forces



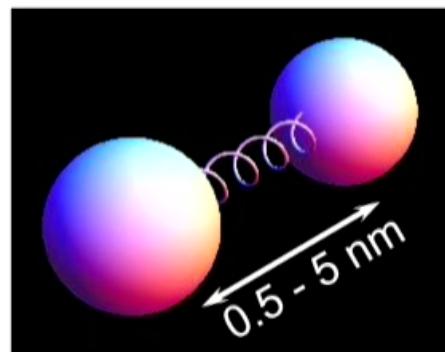
$$V = -\frac{GM^2}{R} \left(1 - \underbrace{Ae^{-R/\lambda}}_{\text{Hypothetical "Yukawa" correction}}\right)$$



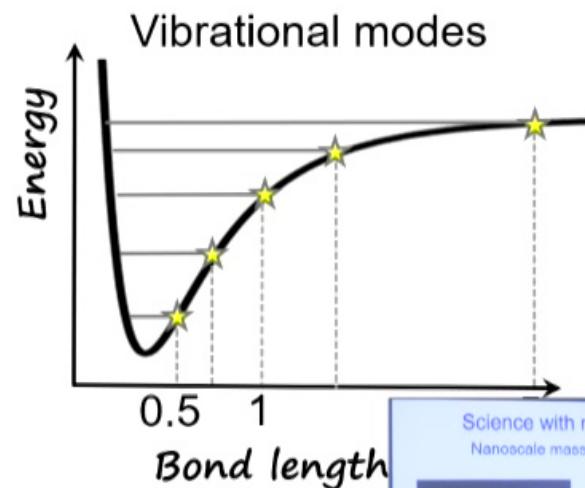
A person is standing at a podium, likely giving a presentation.

Science with molecular clocks

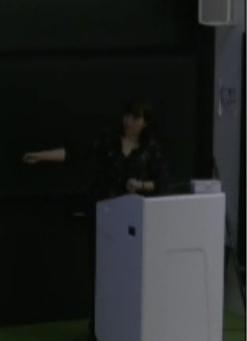
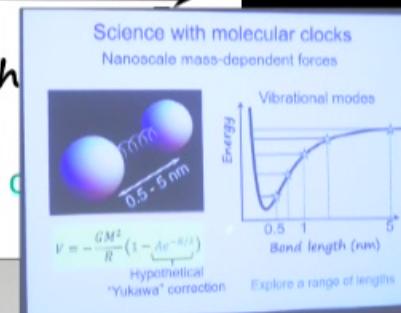
Nanoscale mass-dependent forces



$$V = -\frac{GM^2}{R} \left(1 - \underbrace{Ae^{-R/\lambda}}_{\text{Hypothetical "Yukawa" correction}}\right)$$

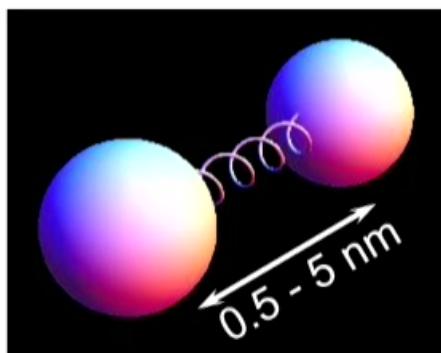


Explore a range of lengths



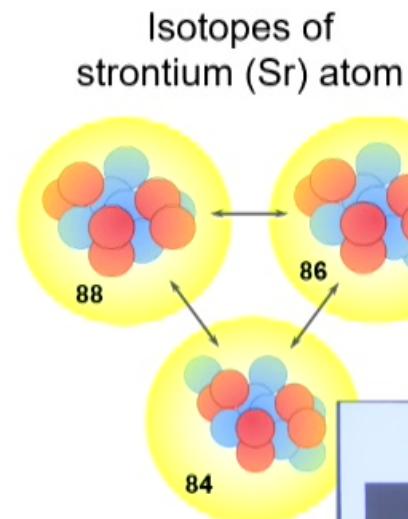
Science with molecular clocks

Nanoscale mass-dependent forces

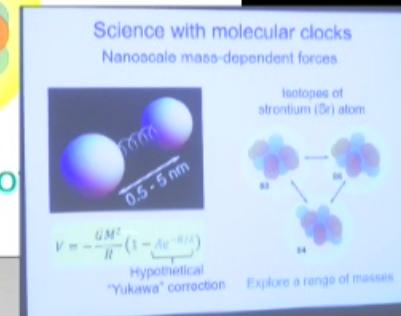


$$V = -\frac{GM^2}{R} \left(1 - \underbrace{Ae^{-R/\lambda}}_{\text{Hypothetical "Yukawa" correction}}\right)$$

Hypothetical
“Yukawa” correction

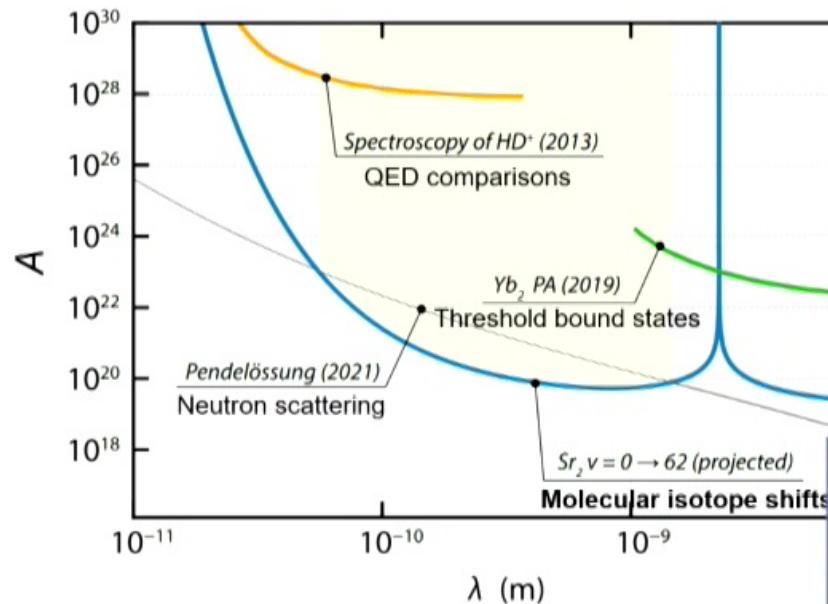


Explore a range of masses



“5th force” with molecular clock

New approach: Vibrational isotope shifts in nonpolar diatomic molecules

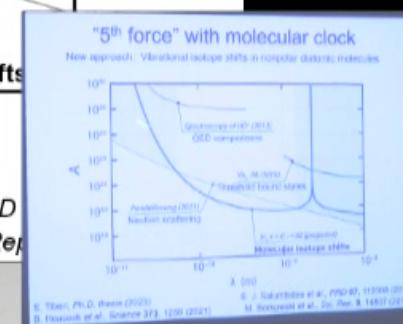


E. Tiberi, Ph.D. thesis (2023)

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

E. J. Salumbides et al., PRD

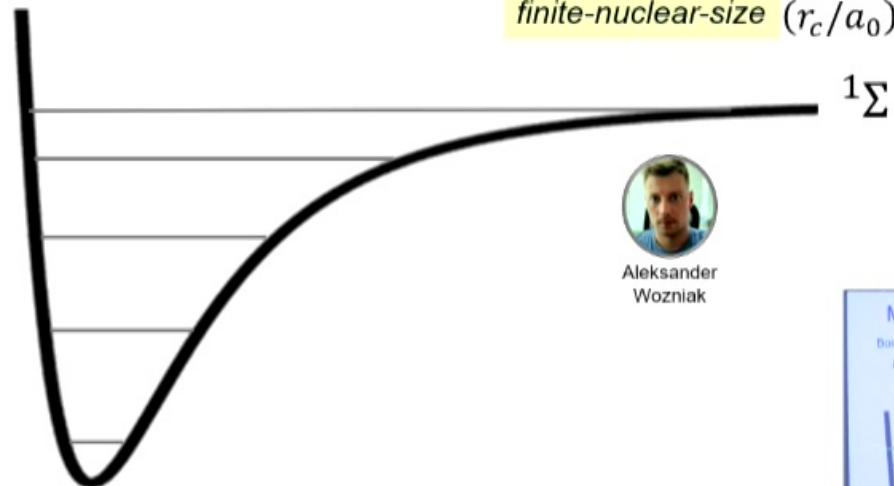
M. Borkowski et al., Sci. Rep.



Molecular clocks, QED, and 5th force

Born-Oppenheimer approximation

$$E_{\text{tot}} \approx E_{\text{el}} + E_{\text{vib}} + \cancel{E_{\text{rot}}}$$



Beyond B-O

adiabatic

$$\mu = \frac{m_e}{Am_p}$$

nonadiabatic

$$\mu^2$$

relativistic, QED

$$\alpha^2 \mu, \alpha^3 \mu$$

finite-nuclear-size

$$(r_c/a_0)^2$$



Aleksander
Wozniak

Molecular clocks, QED, and 5th force

Born-Oppenheimer approximation

$$E_{\text{tot}} \approx E_{\text{el}} + E_{\text{vib}} + \cancel{E_{\text{rot}}}$$

Beyond B-O

$$\mu = \frac{m_e}{Am_p}$$

adiabatic

$$\mu^2$$

nonadiabatic

$$\mu^3$$

relativistic, QED

$$\alpha^2 \mu, \alpha^3 \mu$$

finite-nuclear-size

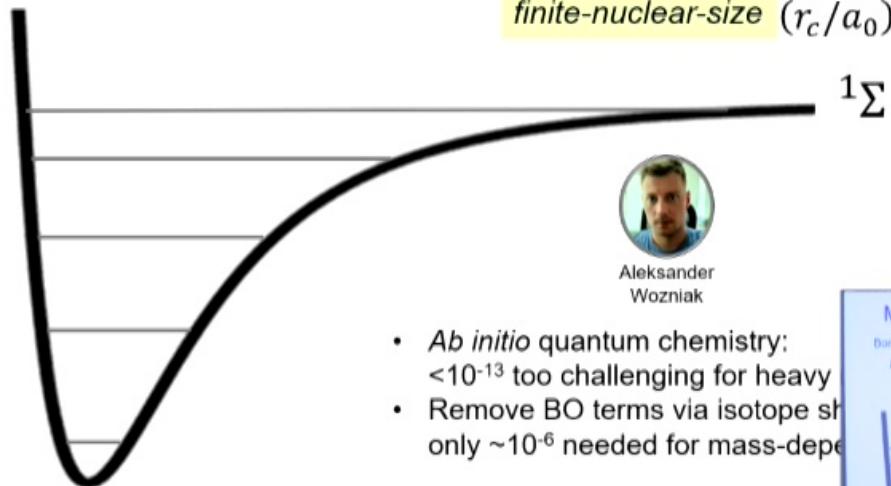
$$(r_c/a_0)^2$$



Molecular clocks, QED, and 5th force

Born-Oppenheimer approximation

$$E_{\text{tot}} \approx E_{\text{el}} + E_{\text{vib}} + \cancel{E_{\text{rot}}}$$



Beyond B-O

adiabatic

$$\mu = \frac{m_e}{Am_p}$$

nonadiabatic

$$\mu^2$$

relativistic, QED

$$\alpha^2 \mu, \alpha^3 \mu$$

finite-nuclear-size

$$(r_c/a_0)^2$$



Aleksander
Wozniak

- *Ab initio* quantum chemistry:
 $<10^{-13}$ too challenging for heavy molecules
- Remove BO terms via isotope shift:
 $\sim 10^{-6}$ needed for mass-dependent terms

J. J. Lutz and J. M. Hutson, *J. Chem. Phys.*

Molecular clocks, QED, and 5th force

Born-Oppenheimer approximation

$$E_{\text{tot}} \approx E_{\text{el}} + E_{\text{vib}} + \cancel{E_{\text{rot}}}$$

Beyond B-O

$$\mu = \frac{m_e}{Am_p}$$

adiabatic

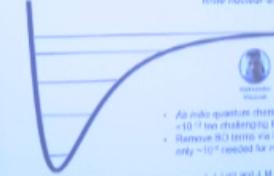
nonadiabatic

relativistic, QED

$\alpha^2 \mu, \alpha^3 \mu$

finite-nuclear-size

$$(r_c/a_0)^2$$

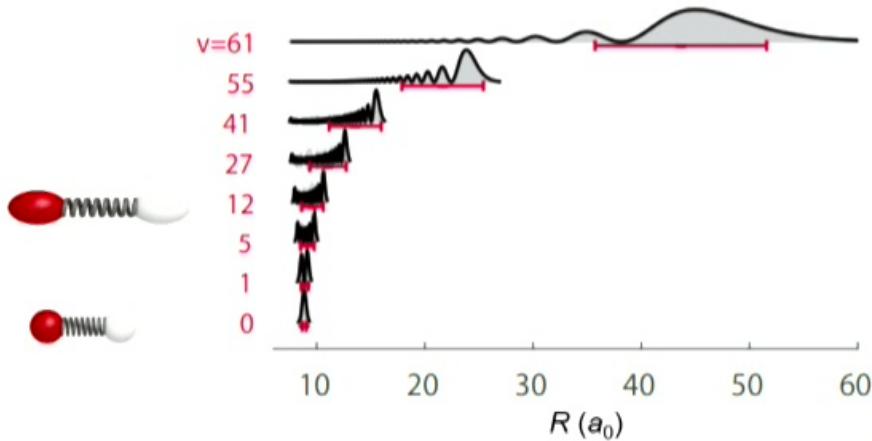


- *Ab initio* quantum chemistry:
 $<10^{-13}$ too challenging for heavy molecules
- Remove BO terms via isotope shift:
 $\sim 10^{-6}$ needed for mass-dependent terms

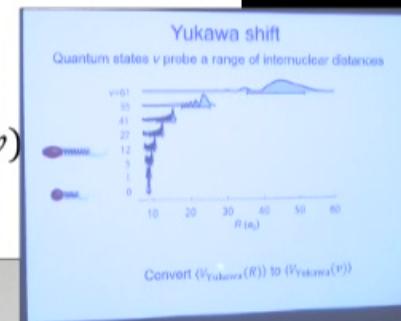
J. J. Lutz and J. M. Hutson, *J. Chem. Phys.*

Yukawa shift

Quantum states v probe a range of internuclear distances

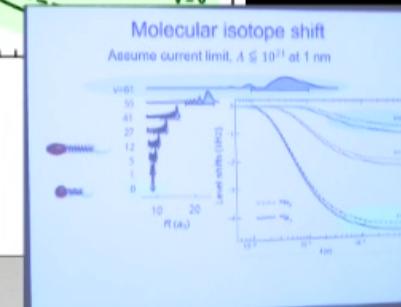
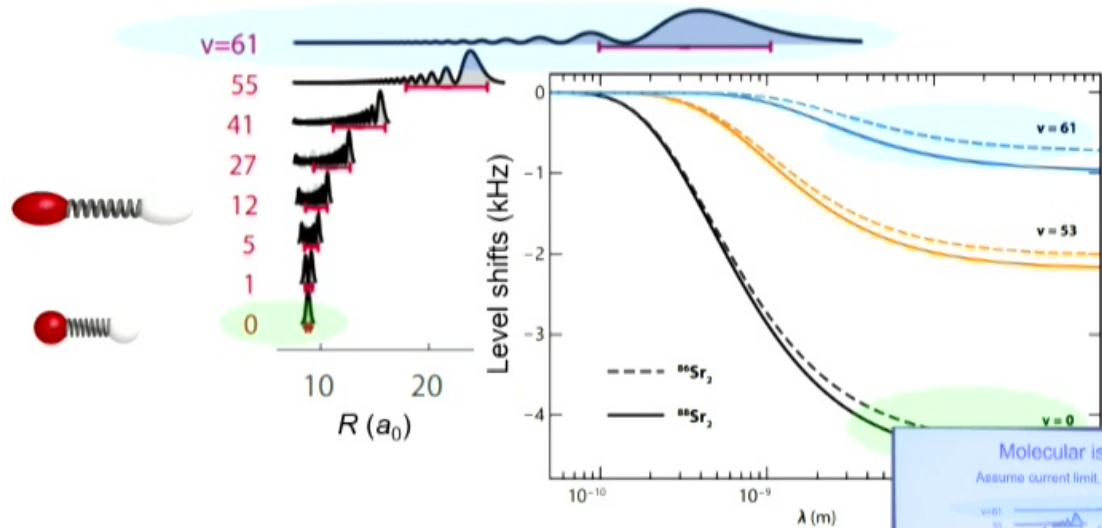


Convert $\langle V_{\text{Yukawa}}(R) \rangle$ to $\langle V_{\text{Yukawa}}(v) \rangle$



Molecular isotope shift

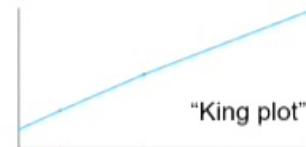
Assume current limit, $A \lesssim 10^{21}$ at 1 nm



Isotope shifts as test of fundamental interactions

Isotope shifts in atoms (e.g. Yb/Yb⁺) can point to new physics

- Isotope shift = "Mass shift" + "Field shift"(depends on $\Delta(r_c^2)$)
- With enough isotopes, can remove $\Delta(r_c^2)$
- Then plot of (Shifts 1) vs. (Shifts 2) is linear
- Any nonlinearities result from unknown atomic/nuclear physics or new electron-neutron interactions



I. Counts *et al.*, PRL 125, 123002 (2020), etc.

- Molecular isotope shifts: New way to test QED and search for sensitive to interaction between massive nuclei
- King-plot-like analysis methods need to be worked out



Isotope shifts as test of fundamental interactions

Isotope shifts in atoms (e.g. Yb/Yb⁺) can point to new physics

- Isotope shift = "Mass shift" + "Field shift"(depends on $\Delta(r_c^2)$)
- With enough isotopes, can remove $\Delta(r_c^2)$
- Then plot of (Shifts 1) vs. (Shifts 2) is linear
- Any nonlinearities result from unknown atomic/nuclear physics or new electron-neutron interactions

I. Counts *et al.*, PRL 125, 123002 (2020), etc.

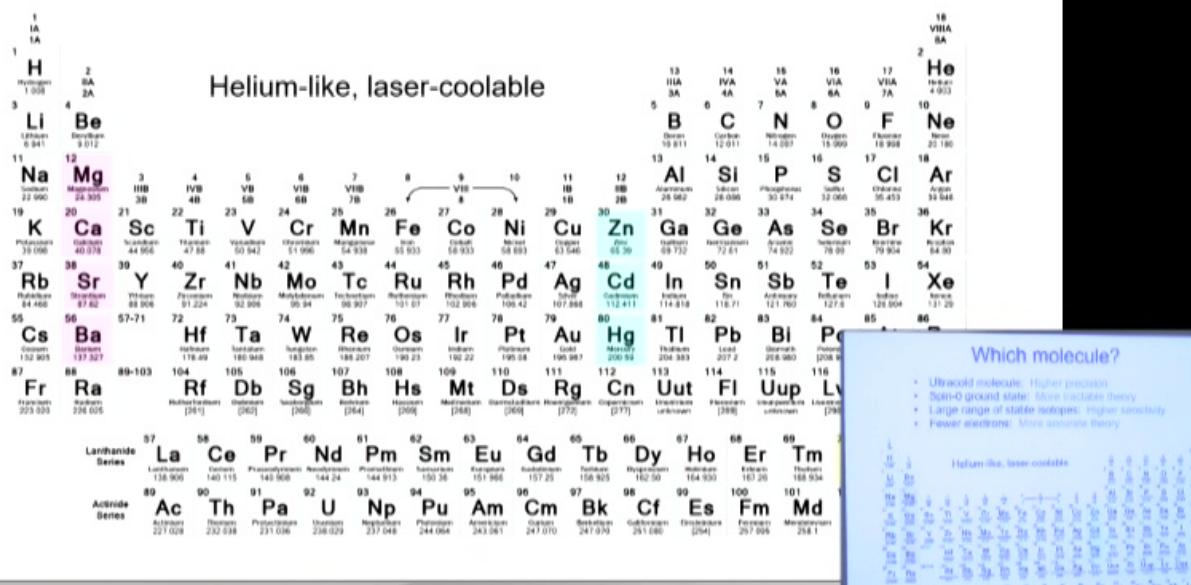
Molecular isotope shifts. New way to test QED and search for new physics; sensitive to interaction between massive nuclei

King-plot-like analysis methods need to be worked out



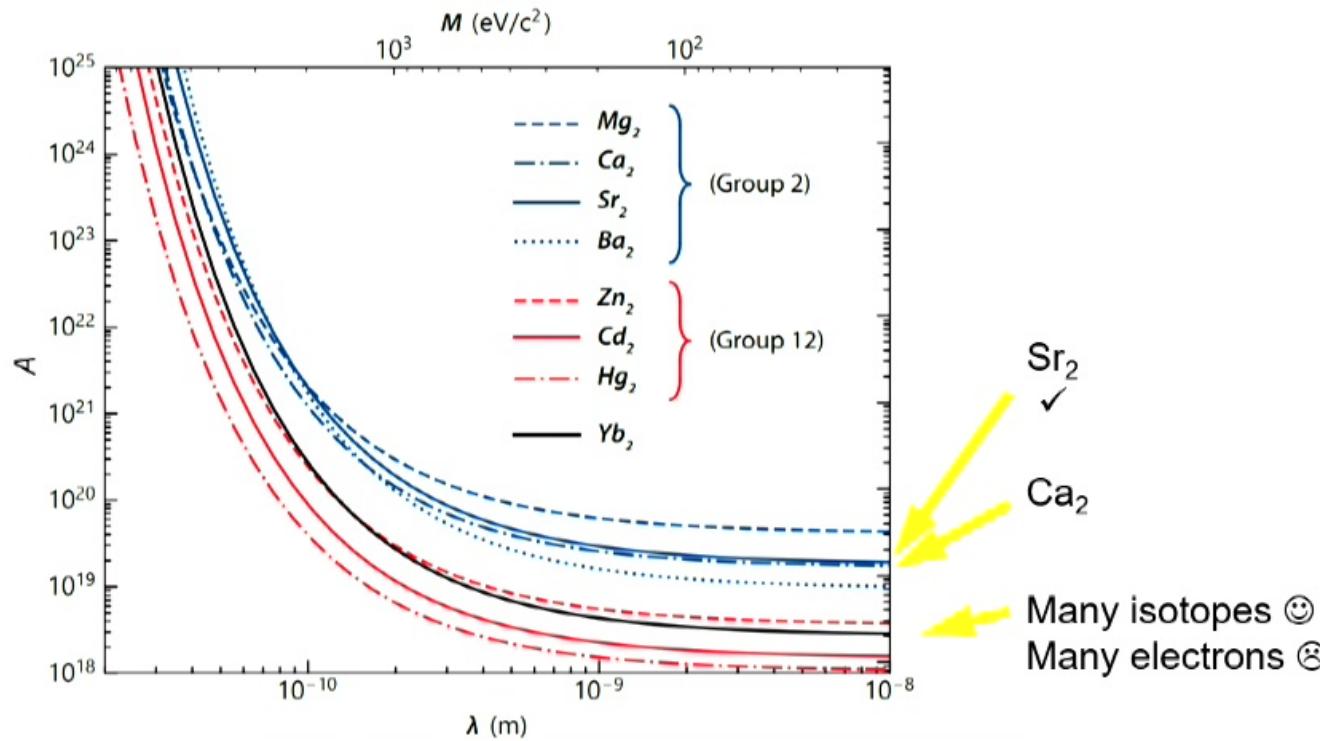
Which molecule?

- Ultracold molecule: Higher precision
 - Spin-0 ground state: More tractable theory
 - Large range of stable isotopes: Higher sensitivity
 - Fewer electrons: More accurate theory



Which molecule?

- Ultracold molecule: Higher precision
- Spin-0 ground state: More tractable theory
- Large range of stable isotopes: Higher sensitivity
- Fewer electrons: More accurate theory



Molecular clocks, QED, and 5th force

Born-Oppenheimer approximation

$$E_{\text{tot}} \approx E_{\text{el}} + E_{\text{vib}} + \cancel{E_{\text{rot}}}$$

Beyond B-O

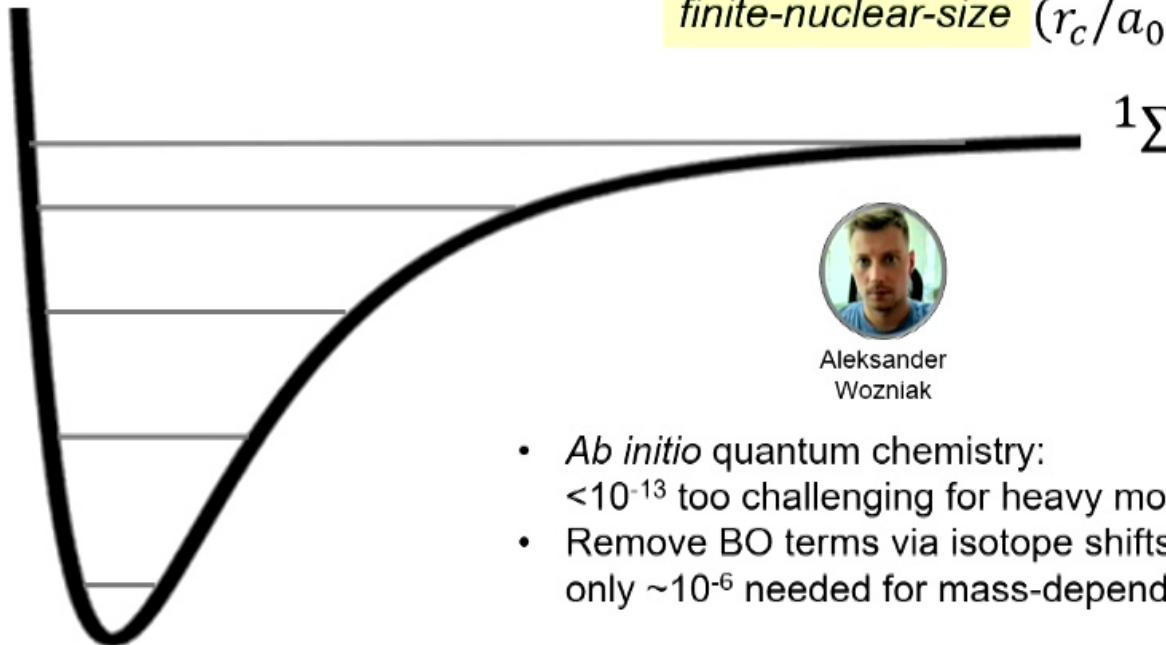
adiabatic

nonadiabatic

relativistic, QED

finite-nuclear-size $(r_c/a_0)^2$

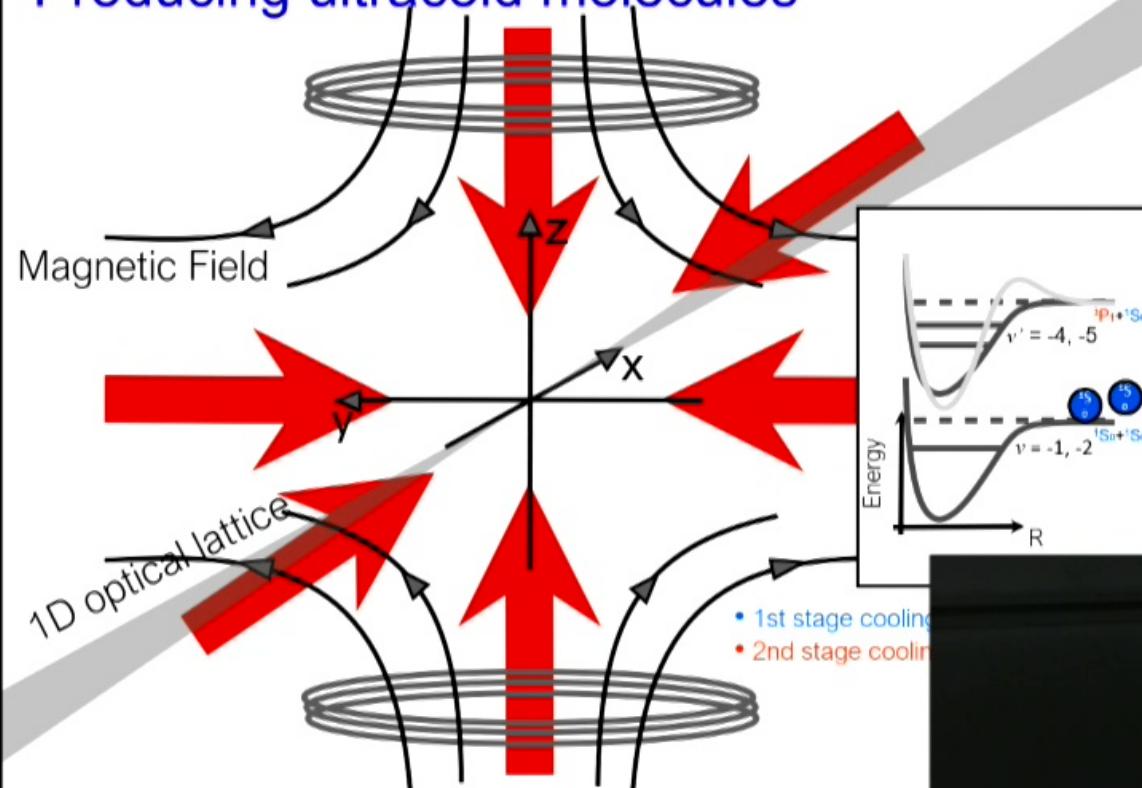
$$\mu = \frac{m_e}{Am_p}$$
$$\mu^2$$
$$\alpha^2 \mu, \alpha^3 \mu$$



- *Ab initio* quantum chemistry:
 $<10^{-13}$ too challenging for heavy molecule
- Remove BO terms via isotope shifts:
only $\sim 10^{-6}$ needed for mass-dependent terms

J. J. Lutz and J. M. Hutson, *JMS* **330**, 43 (2016)

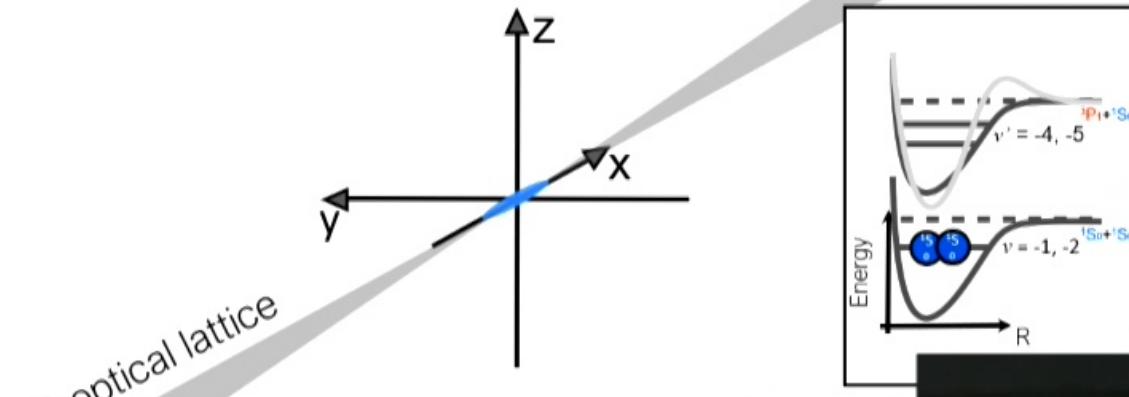
Producing ultracold molecules



- 1st stage cooling
- 2nd stage cooling



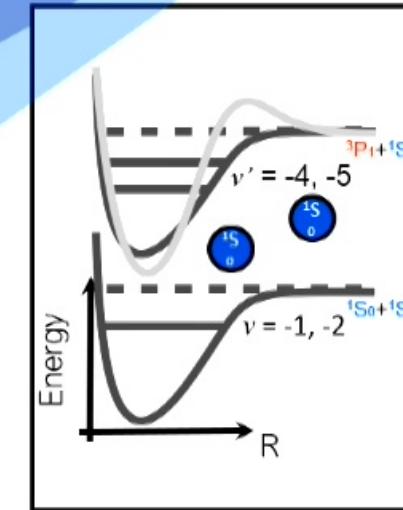
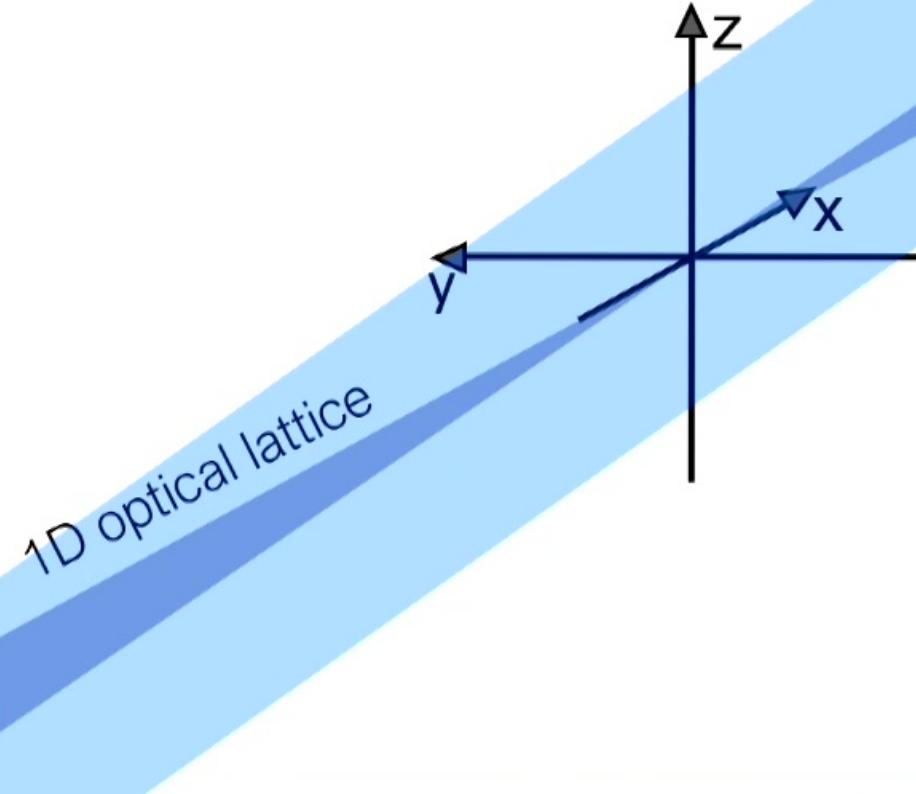
Producing ultracold molecules



- 1st stage cooling
- 2nd stage cooling
- Photoassociation



Producing ultracold molecules



- 1st stage cooling ($\sim \text{mK}$)
- 2nd stage cooling ($\sim \mu\text{K}$)
- Photoassociation / state prep
- Spectroscopy of excited states
- Recovery via photodissociation

Credit: M. McDonald

Ultracold photodissociation

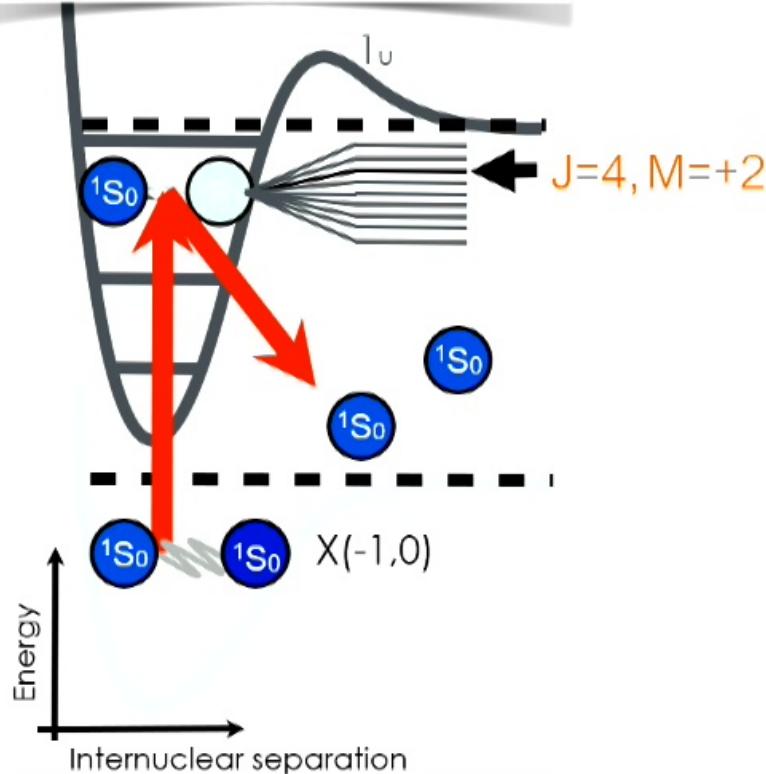
LETTER

DOI: 10.1038/nature18514

Photodissociation of ultracold diatomic strontium molecules with quantum state control

M. McDonald¹, B. H. McCay¹, F. Apelbeck¹, C. H. Low¹, I. Majewski², R. Moynihan² & T. Zelevinsky¹

122 | NATURE | VOL. 535 | 7 JULY 2016



Credit: M. McDonald

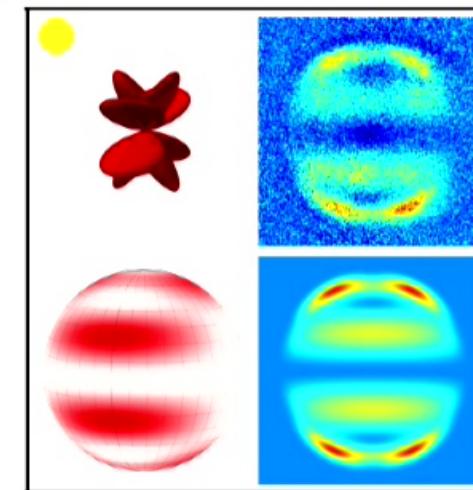
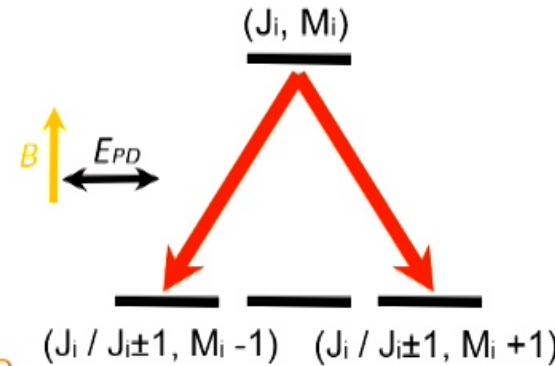
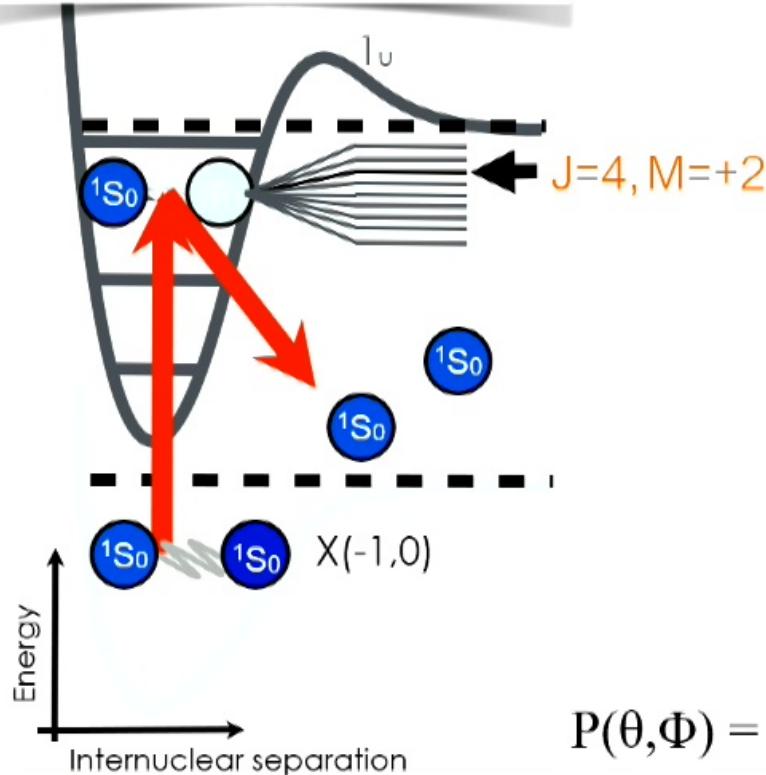
Ultracold photodissociation

LETTER

Photodissociation of ultracold diatomic strontium molecules with quantum state control

M. McDonald¹, B. H. McCay¹, F. Apelbeck¹, C. H. Low¹, I. Majewski², R. Moiseyev³ & T. Zelevinsky¹

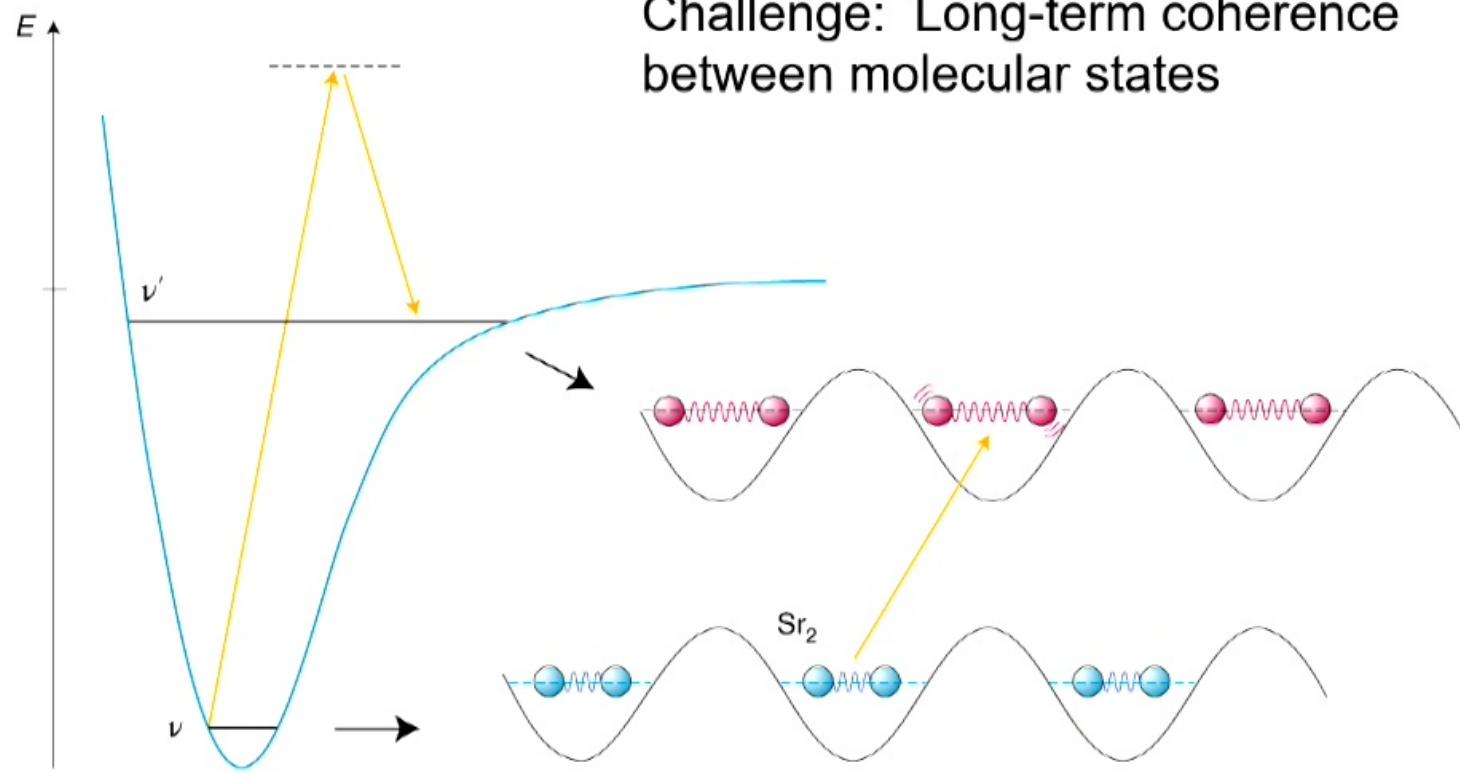
122 | NATURE | VOL 535 | 7 JULY 2016



$$P(\theta, \Phi) = |\sqrt{R} \cdot Y_4^1(\theta, \Phi) + \sqrt{1-R} \cdot e^{i\delta} \cdot Y_4^3(\theta, \Phi)|^2$$

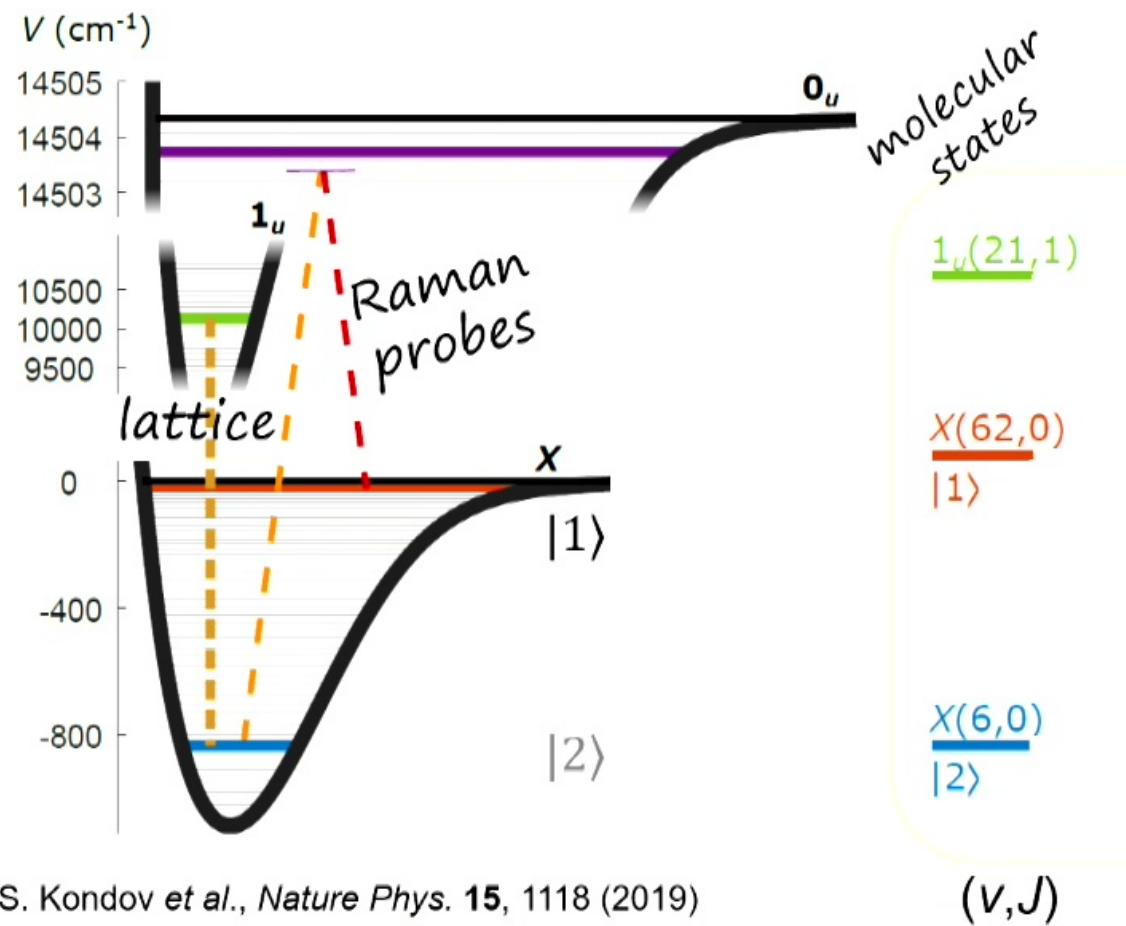
Credit: M. McDonald

Molecular lattice clock



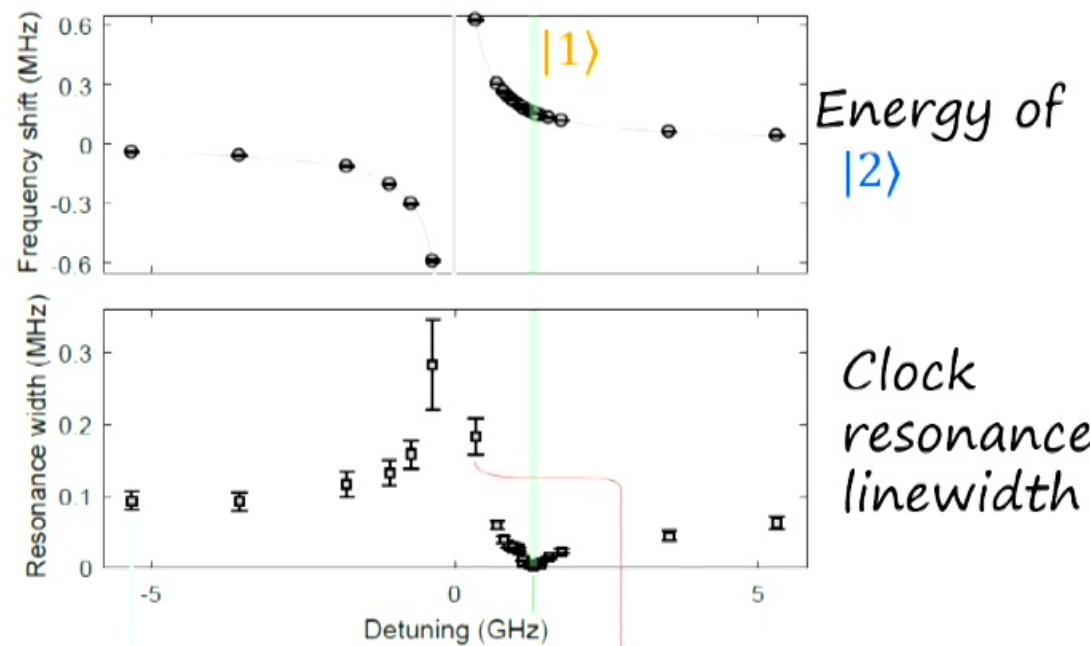
N. Poli, *Nature Phys.* **15**, 1106 (2019)

“Magic” lattice trap



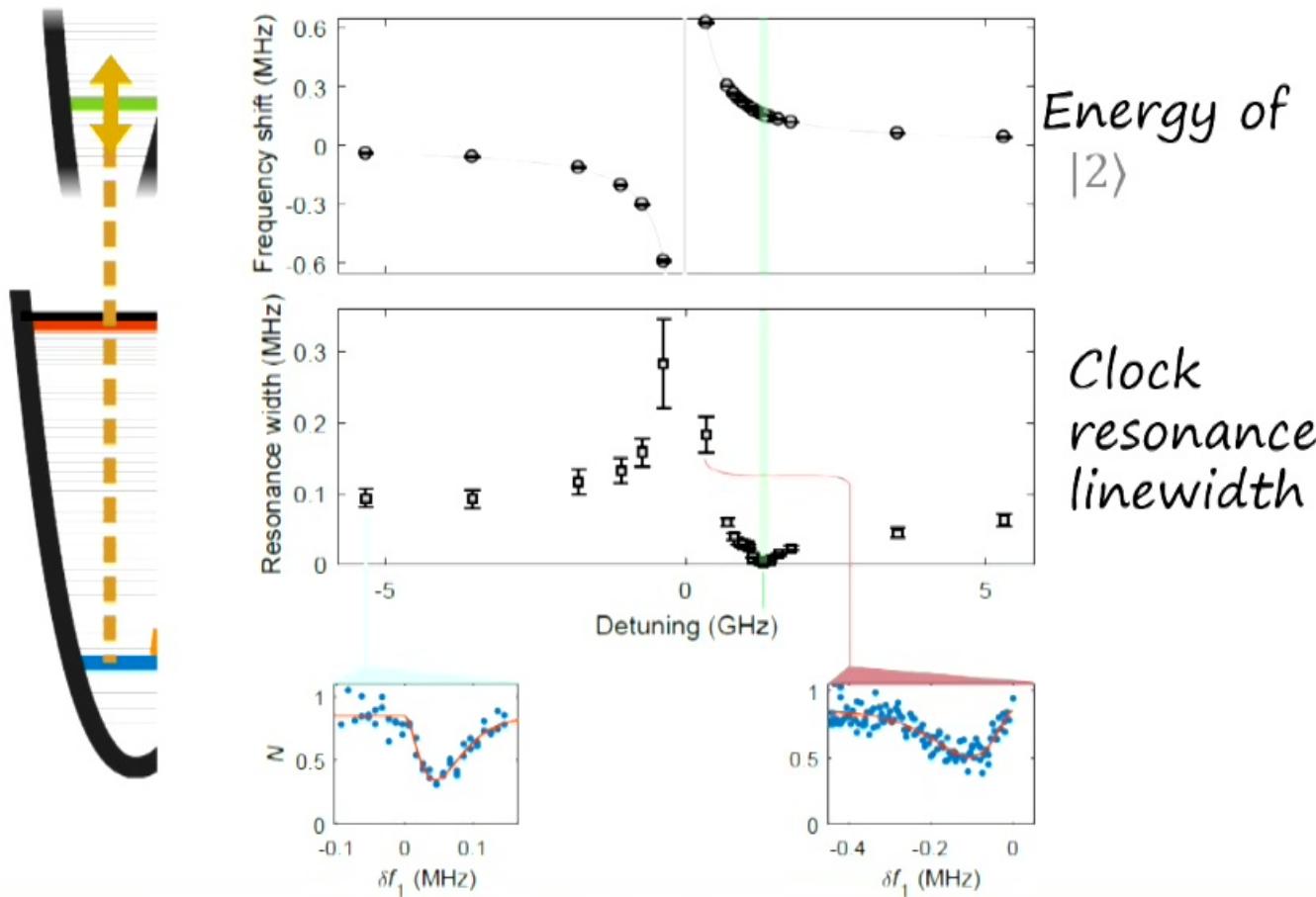
S. S. Kondov *et al.*, *Nature Phys.* **15**, 1118 (2019)

Enhanced coherence in magic lattice



S. S. Kondov et al., *Nature Phys.* **15**, 1118 (2019)

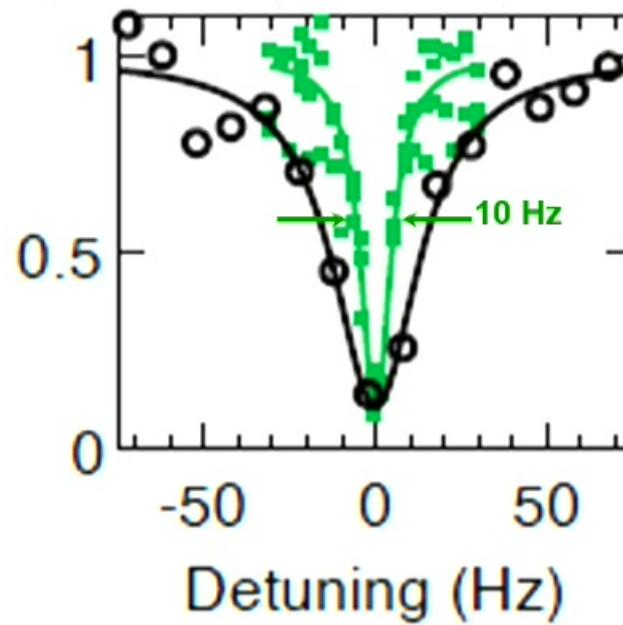
Enhanced coherence in magic lattice



S. S. Kondov et al., *Nature Phys.* **15**, 1118 (2019)

Narrow vibrational clock resonance

Coherence $10^4 \times$

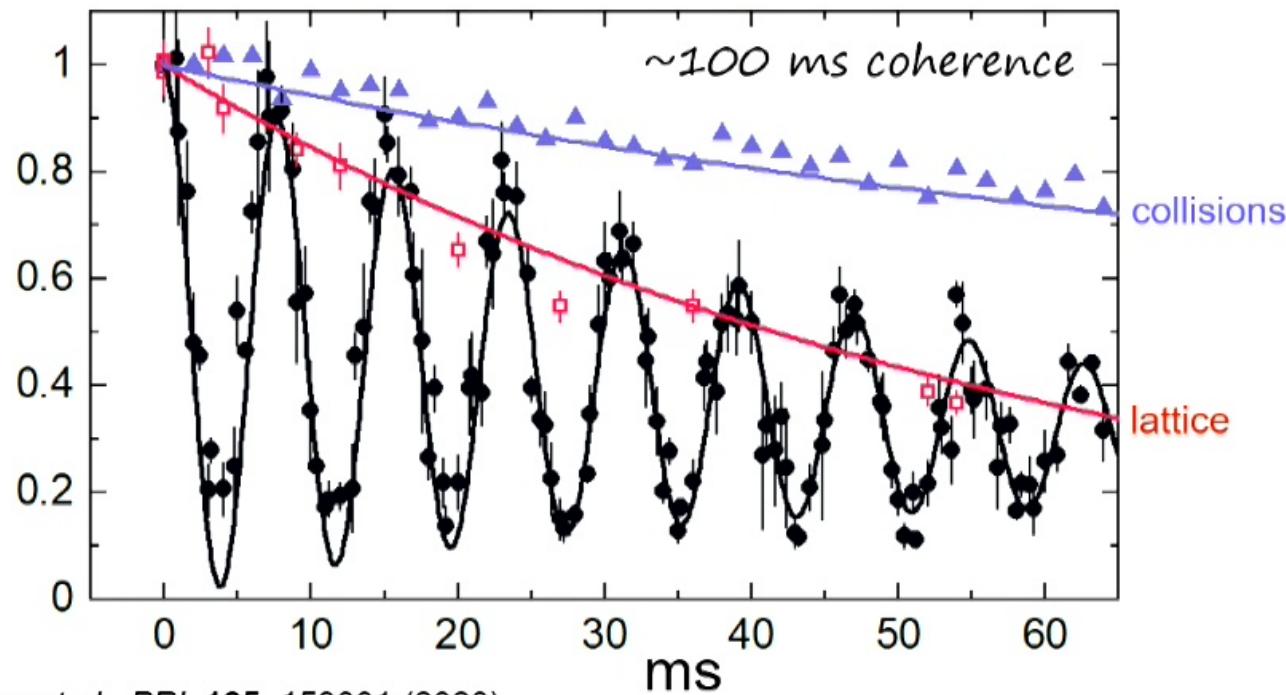
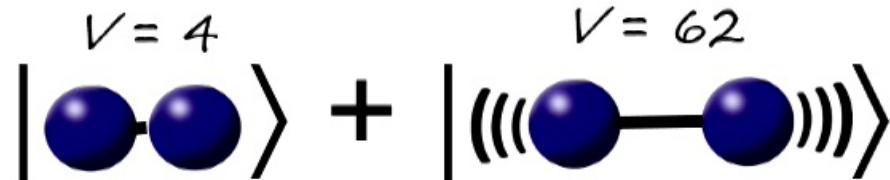


$$Q = 3 \times 10^{12}$$

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

Rabi oscillations

Tightly-to-weakly-bound molecules



K. H. Leung *et al.*, PRL 125, 153001 (2020)

Clock precision

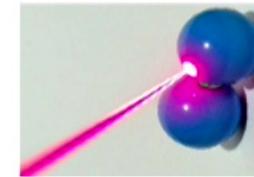
Systematic effects:

What can cause the clock frequency to shift?

- Lattice laser light



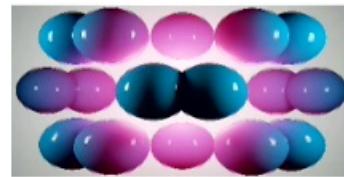
- Probe laser light



- Blackbody radiation



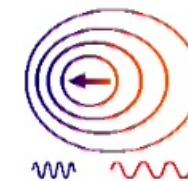
- Collisions



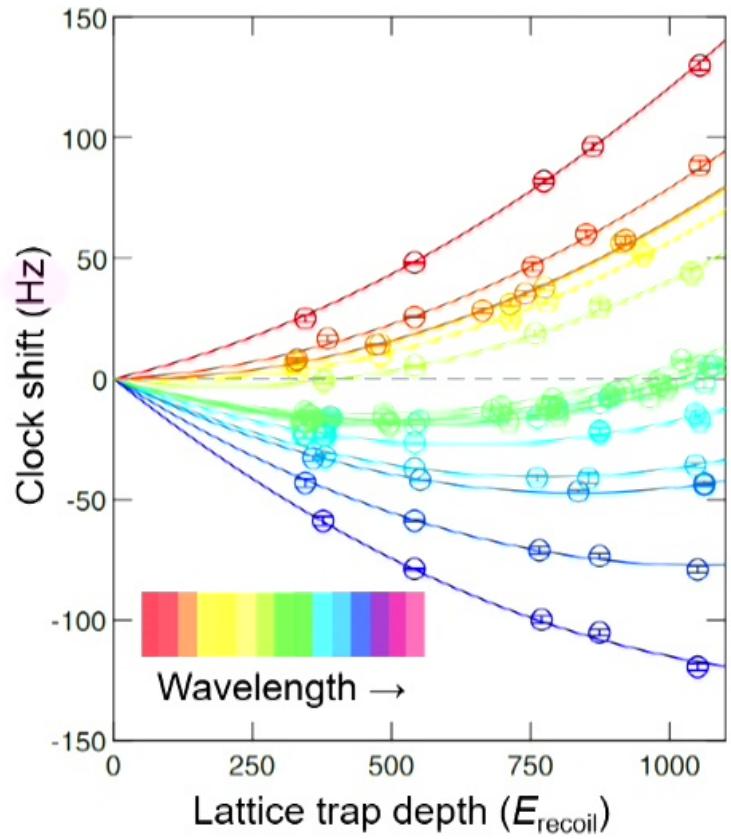
- Magnetic fields



- Doppler shifts

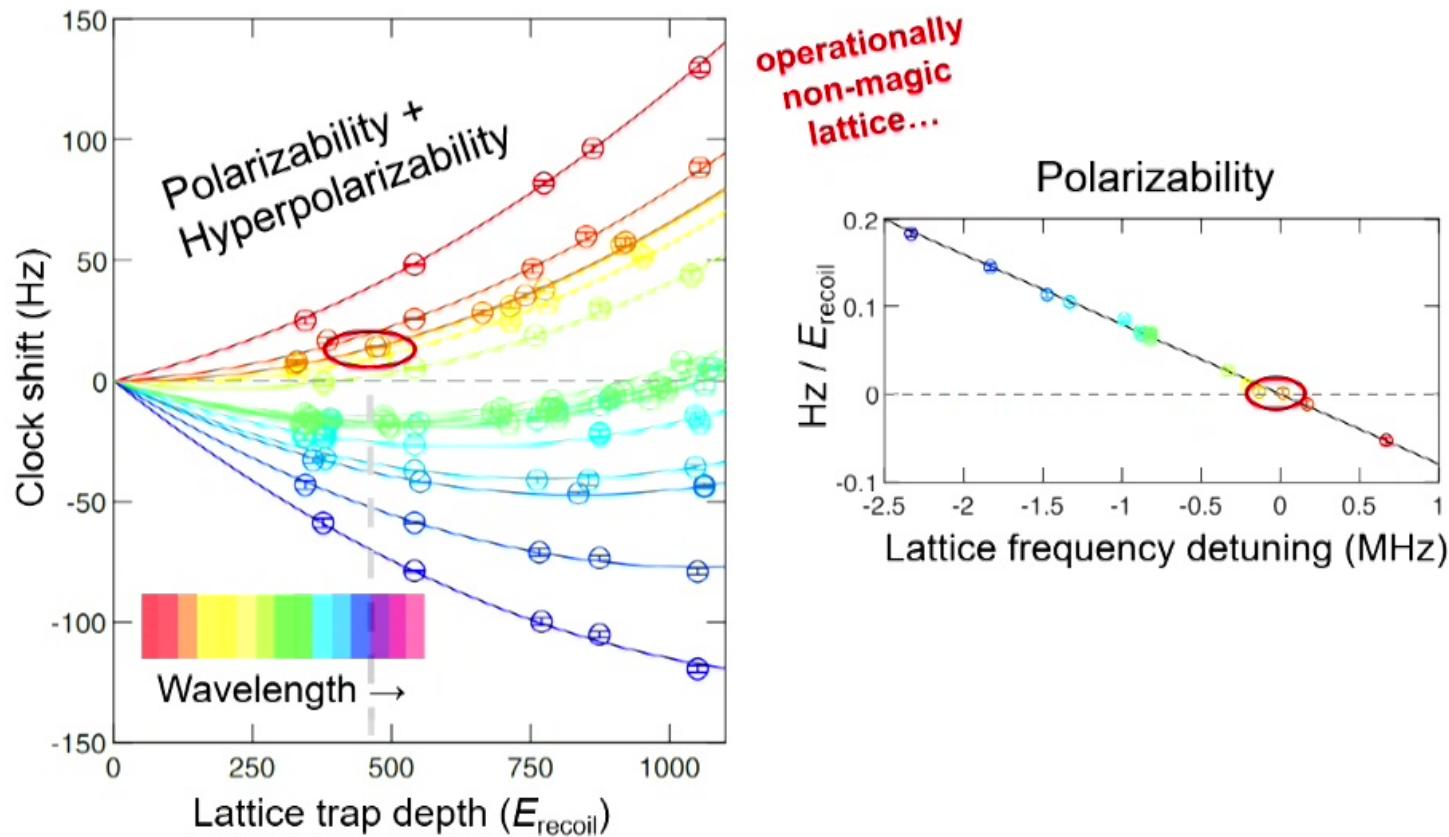


Lattice light shift: Zoom in



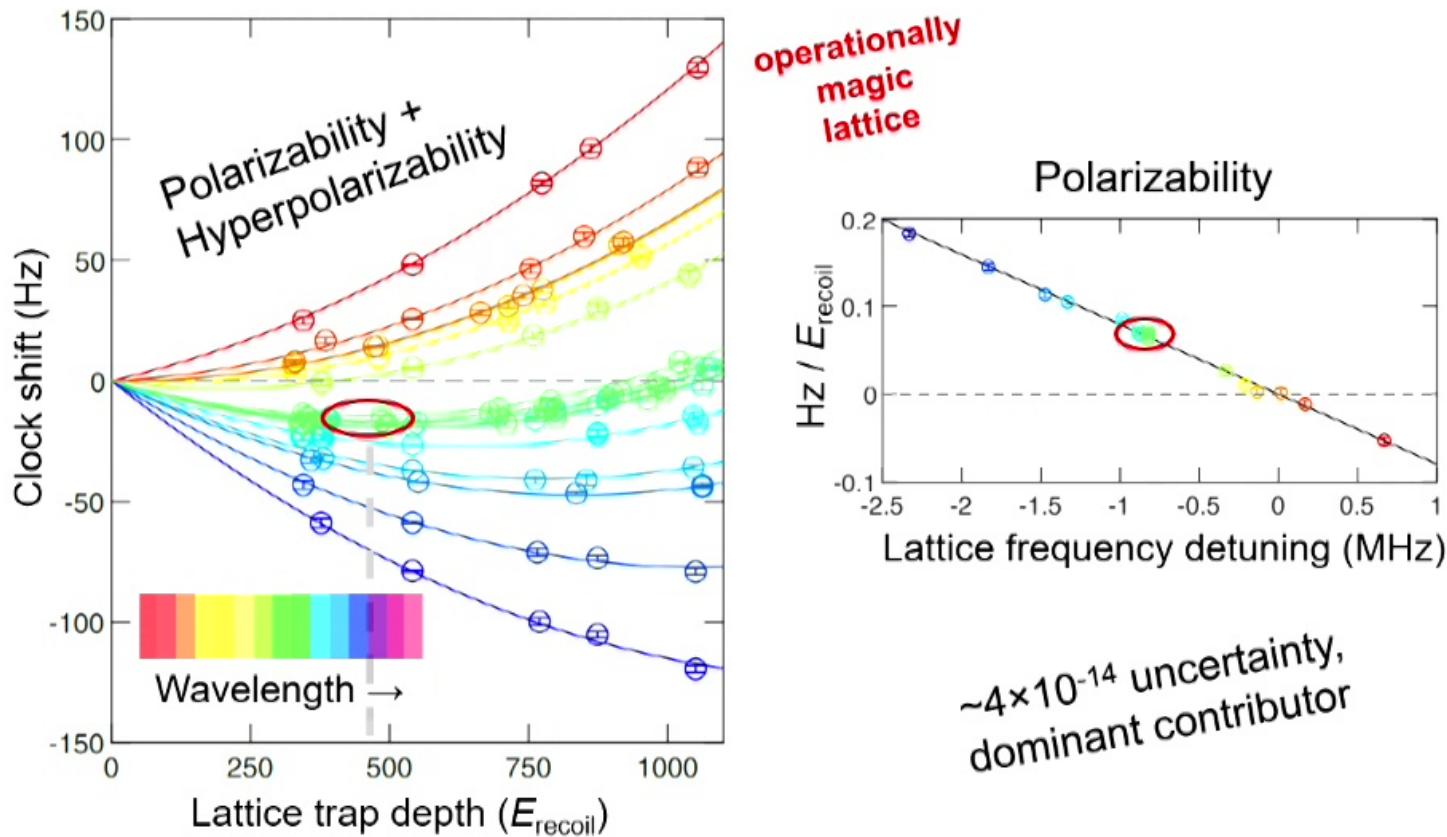
K. H. Leung *et al.*, *Phys. Rev. X* 13, 011047 (2023)

Observe vibrational hyperpolarizability



K. H. Leung *et al.*, *Phys. Rev. X* 13, 011047 (2023)

Effectively magic lattice



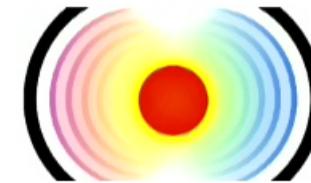
K. H. Leung *et al.*, *Phys. Rev. X* 13, 011047 (2023)

Clock precision

Systematic effects:

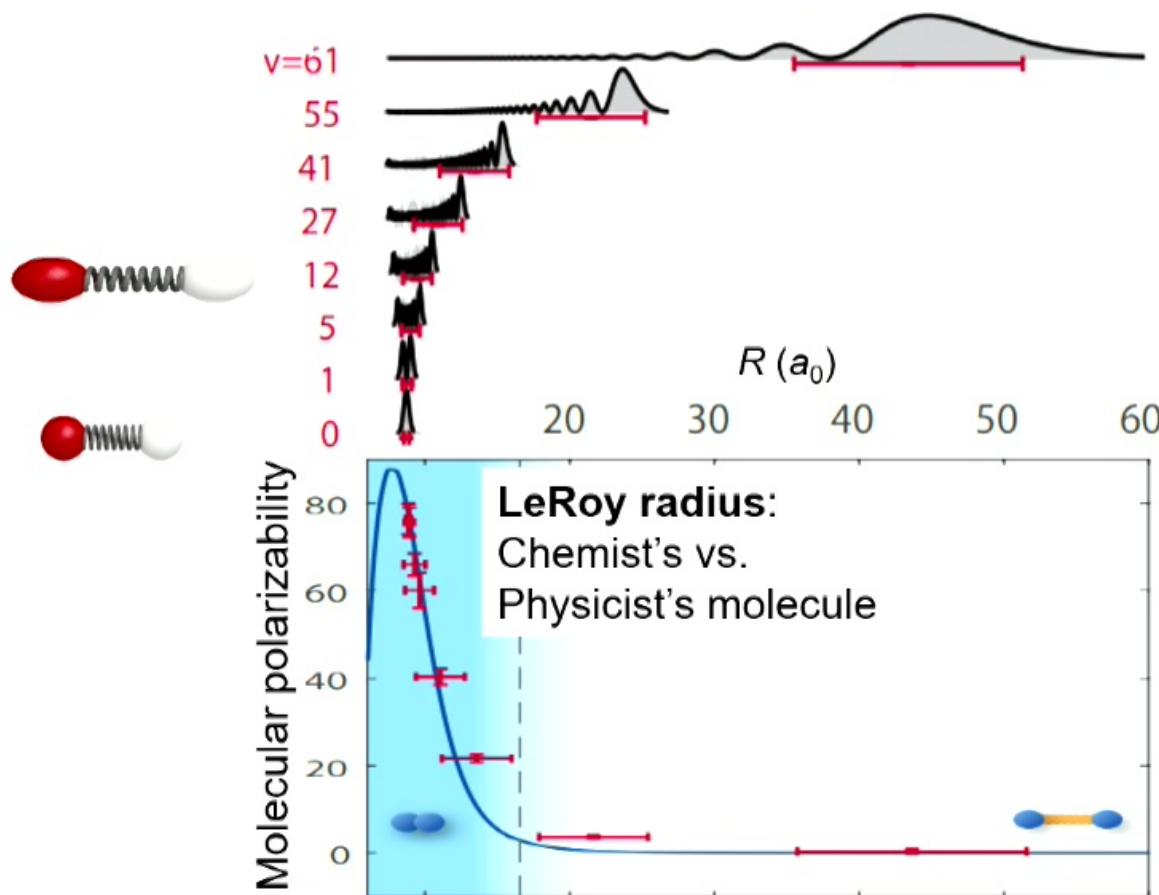
What can cause the clock frequency to shift?

- Blackbody radiation



Far-infrared polarizabilities

Response of vibrating molecule to BBR



Clock precision

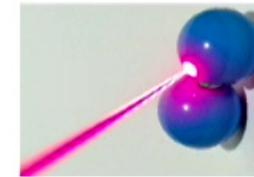
Systematic effects:

What can cause the clock frequency to shift?

- Lattice laser light



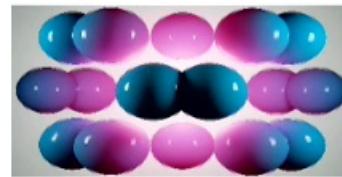
- Probe laser light



- Blackbody radiation



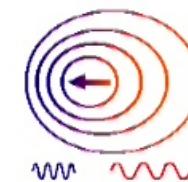
- Collisions



- Magnetic fields

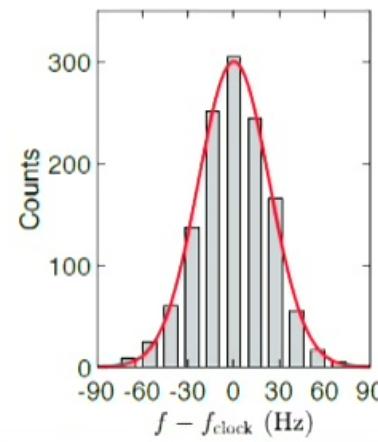
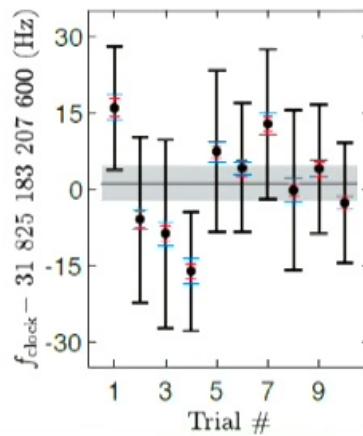
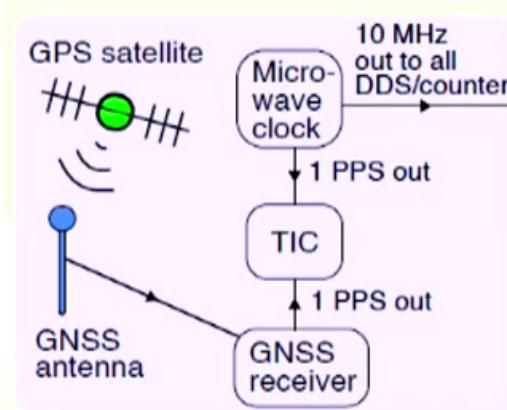


- Doppler shifts



Absolute clock measurement

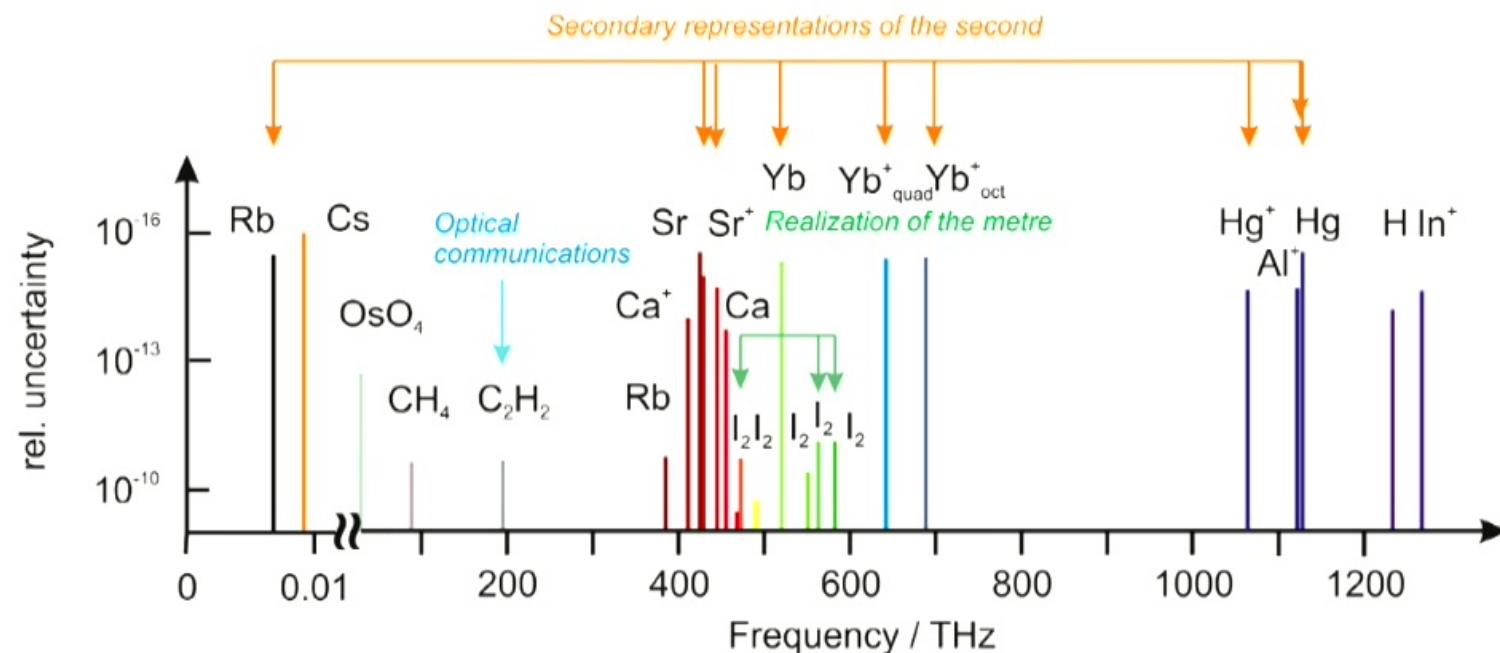
Comparison to Cs atomic time standard



1×10^{-13} uncertainty

Atomic and molecular frequency standards

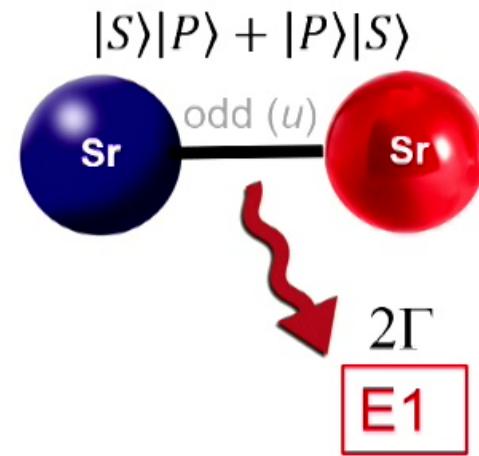
Primary and secondary



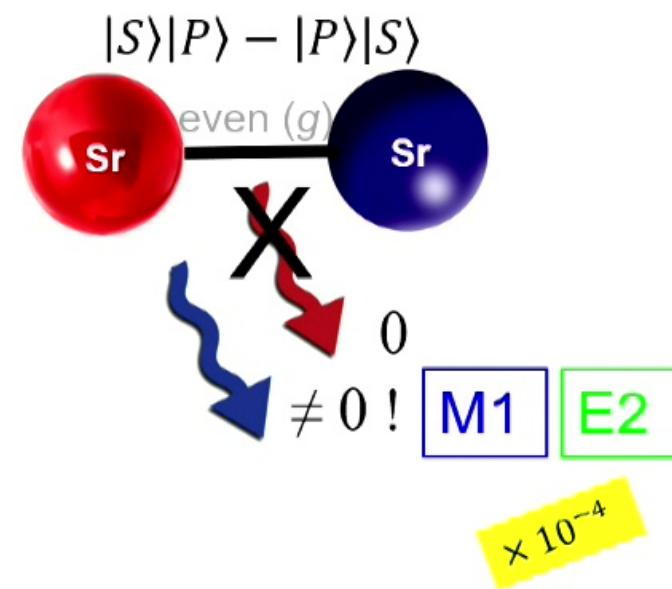
F. Riehle *et al.*, *Metrologia* **55**, 188 (2018)

Optical molecular clocks

Superradiant



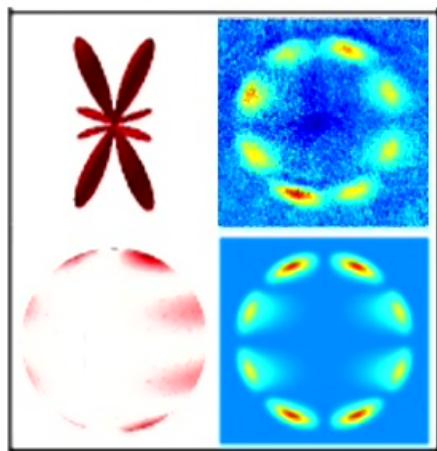
Subradiant



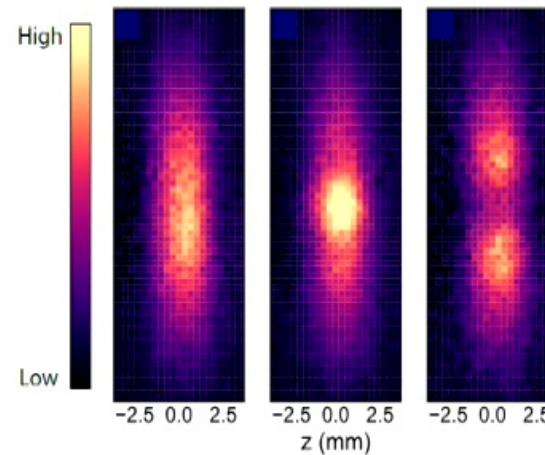
B. McGuyer *et al.*, *Nature Phys.* **11**, 32 (2015)

Other projects with cold molecules

Ultracold chemistry and quantum photodissociation



Laser cooling of new bosonic and fermionic molecules (CaH, CaD)



CeNTREX: Proton EDM & fundamental symmetries

