

New Opportunities for Fundamental Physics Research  
with Radioactive Molecules, Online, July. 2, 2021

# Measurement of the Variation of Electron-to-Proton Mass Ratio Using Ultracold Molecules Produced from Laser-Cooled Atoms

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# Motivation: variation of fundamental constants over time

Various theories postulates oscillating scalar field to account for **dark matter/dark energy**

"**Quintessence** is a hypothetical form of **dark energy**, more precisely **a scalar field**, postulated as an explanation of the observation of an accelerating rate of expansion of the universe."

"**Scalar field dark matter** is a classical, minimally coupled, scalar field postulated to account for the inferred dark matter."

(In both theories, frequency is set by the mass of the particle:  $\omega = \frac{mc^2}{\hbar}$ )

Coupling to EM fields and/or ordinary matter can cause variation of fundamental constants over time

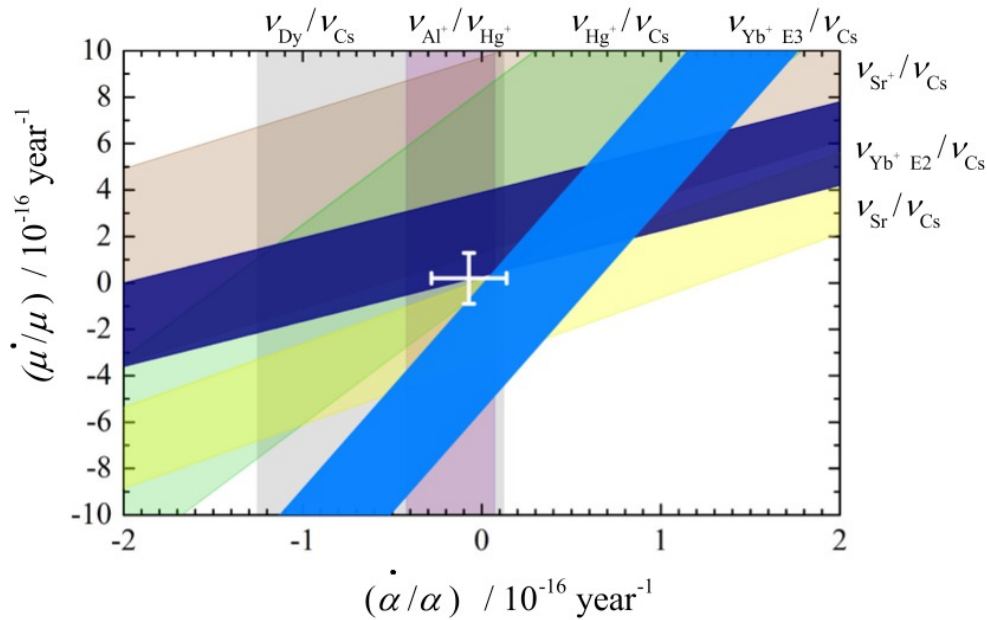
→Candidates for experimental tests:

$$\alpha, \mu = \frac{m_e}{M_p}, g - 2, \dots$$

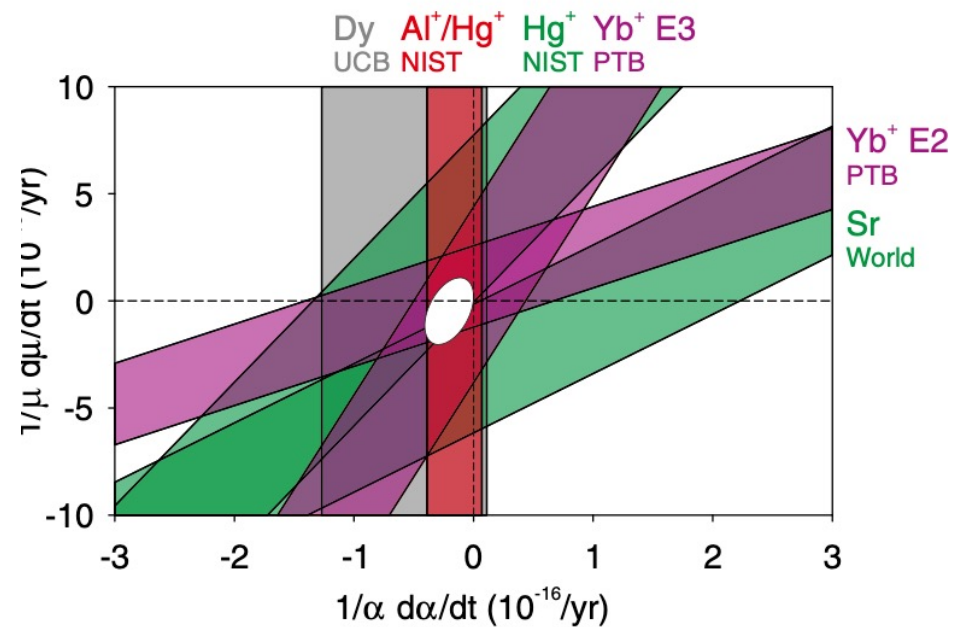
# high precision atomic spectroscopy

optical lattice clock + Cs frequency standards

Sensitive to both  $\alpha$  and  $\mu = \frac{m_e}{M_p}$



R. M. Godun et al.,  
PRL 113, 210801 (2014)

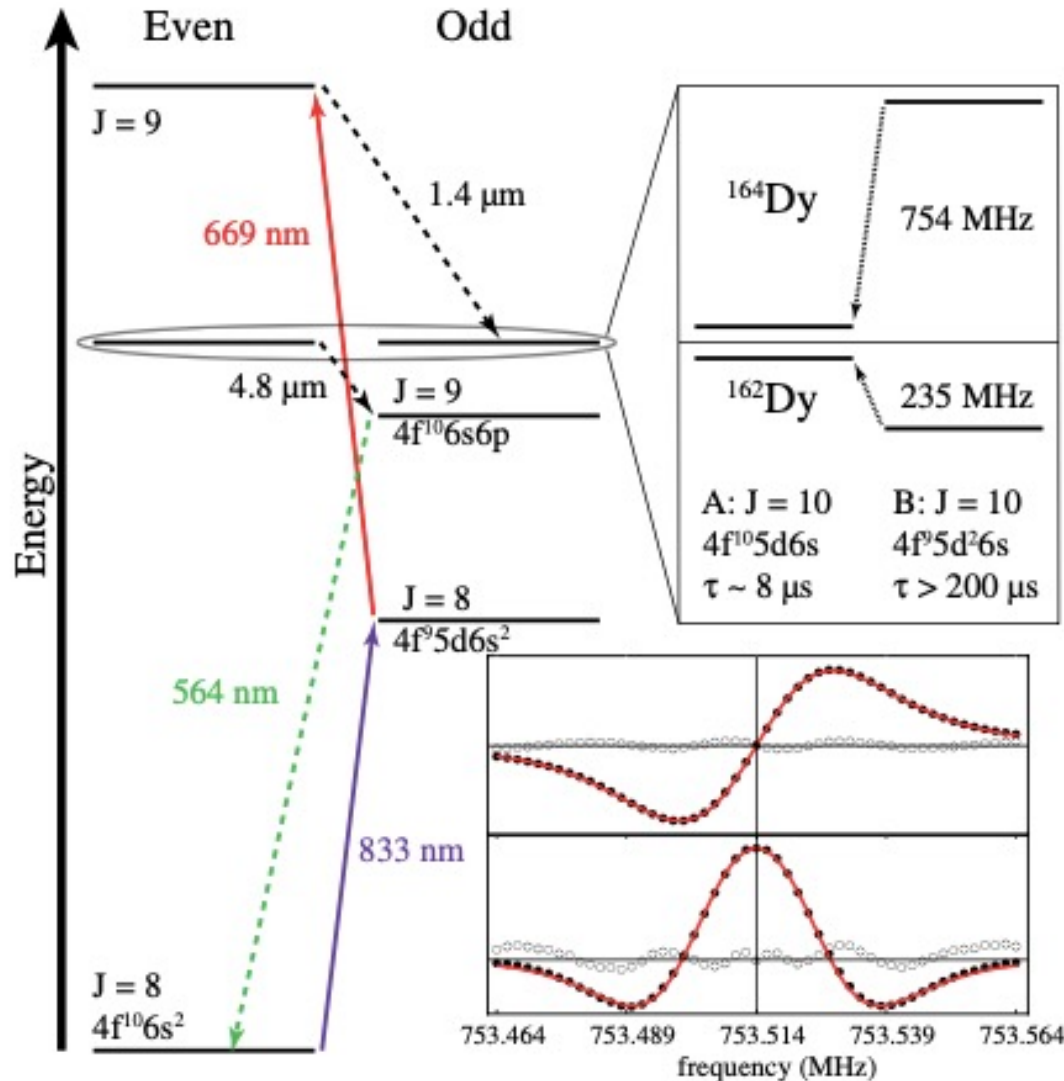


N. Huntemann et al  
PRL 113, 210802 (2014)



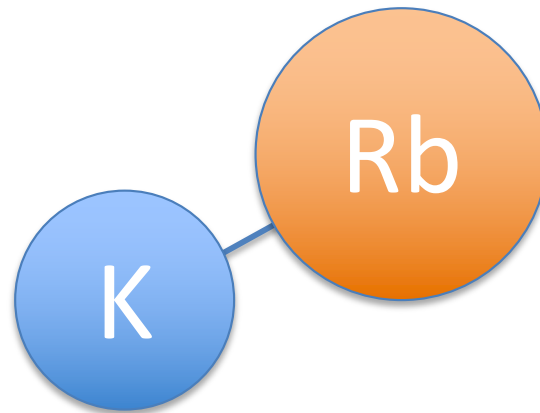
# New Limits on Variation of the Fine-Structure Constant Using Atomic Dysprosium

N. Leefer,<sup>1</sup> C. T. M. Weber,<sup>2</sup> A. Cingöz,<sup>3</sup> J. R. Torgerson,<sup>4</sup> and D. Budker<sup>1,5</sup>



almost complete  
degeneracy of  
opposite-parity  
excited states

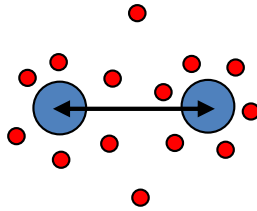
$$\dot{\alpha}/\alpha = (-5.8 \pm 6.9) \times 10^{-17} \text{ yr}^{-1}$$



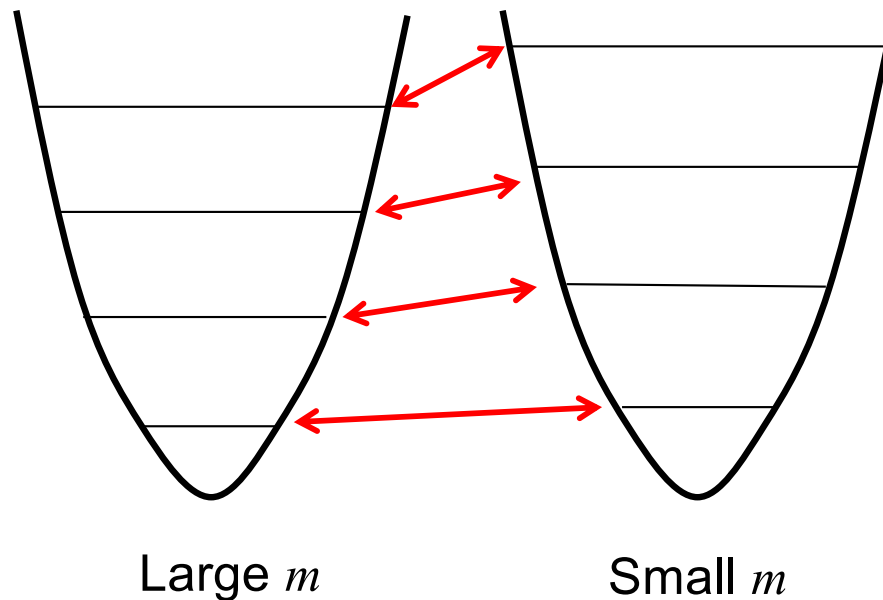
"Measurement of the variation of electron-to-proton mass ratio using ultracold molecules produced from laser-cooled atoms"

J. Kobayashi, A. Ogino and S. Inouye,  
Nature Communications 10, 3771 (2019)

molecular spectra are sensitive to  $\mu = \frac{m_e}{M_p}$



Molecular spectra are **directly** related to the inertial mass of nucleus.



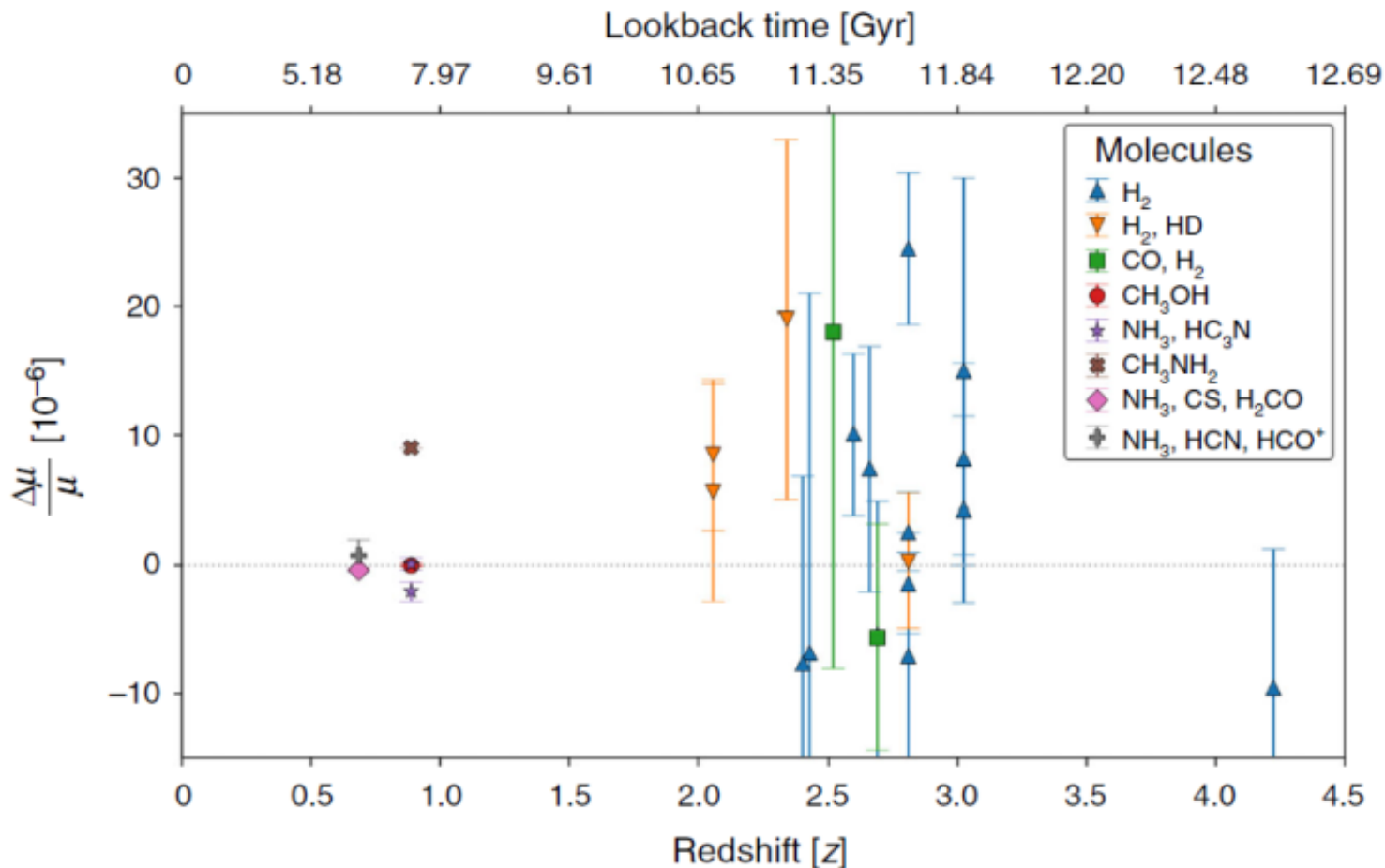
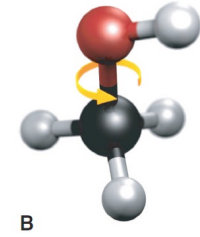
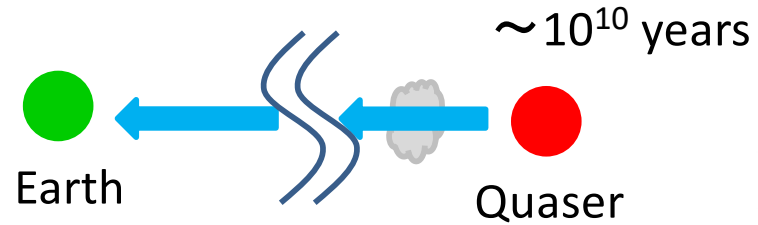
Relevant energy scales:

electronic  $\propto \mu^0$

vibrational  $\propto \frac{1}{\sqrt{\mu}}$

rotational  $\propto \frac{1}{\mu}$

# Radio-astronomical observations



$$\frac{10^{-6}}{10^{10} \text{ year}}$$

$$\sim 10^{-16} / \text{year}$$

No definitive answer yet

S. Anna-Maree et al., *Aust. J. Chem.* (2019)

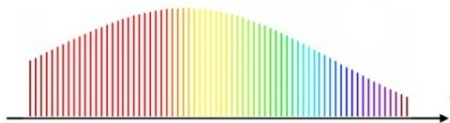
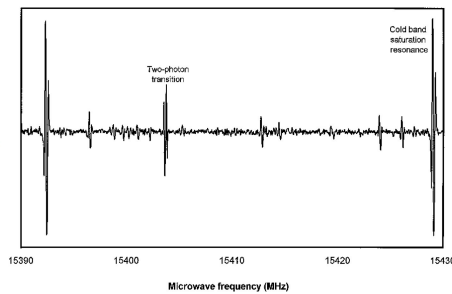
# The limit of time variation of $m_e/M_p (\equiv \mu)$

## Laboratory observations

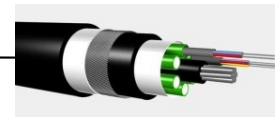
molecular spectroscopy of SF<sub>6</sub>

A. Shelkovernikov et al, PRL 100, 150801 (2008)

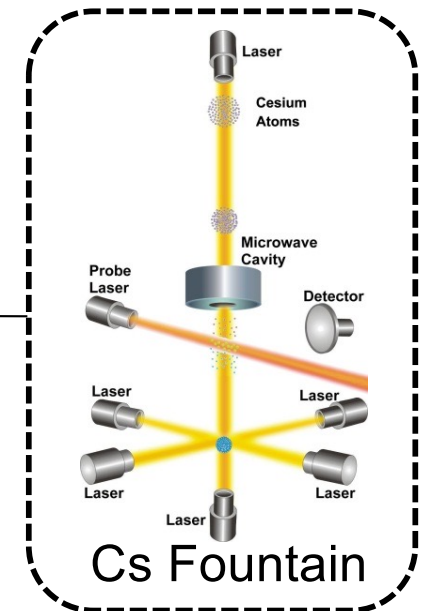
$$\dot{\mu} / \mu = (3.8 \pm 5.6) \times 10^{-14} / \text{year}$$



rovibrational transition in SF<sub>6</sub> + frequency comb



Optical Fiber link (~43km)





One can **enhance the sensitivity** to variation of  $\mu$  by using transitions between **nearly degenerate levels with different symmetries** (i.e. like **singlet** and **triplet** potentials of bi-alkali molecules).

### Enhanced Sensitivity to Variation of $m_e/m_p$ in Molecular Spectra

D. DeMille,<sup>1</sup> S. Sainis,<sup>1</sup> J. Sage,<sup>1</sup> T. Bergeman,<sup>2</sup> S. Kotochigova,<sup>3</sup> and E. Tiesinga<sup>4</sup>

<sup>1</sup>Department of Physics, Yale University, New Haven, Connecticut 06520, USA

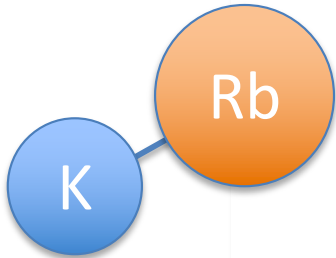
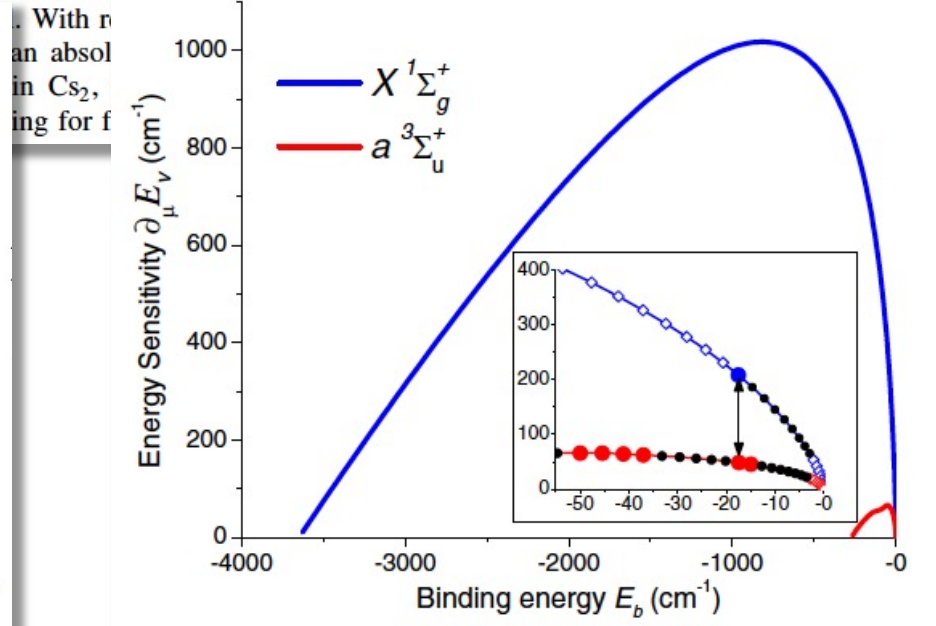
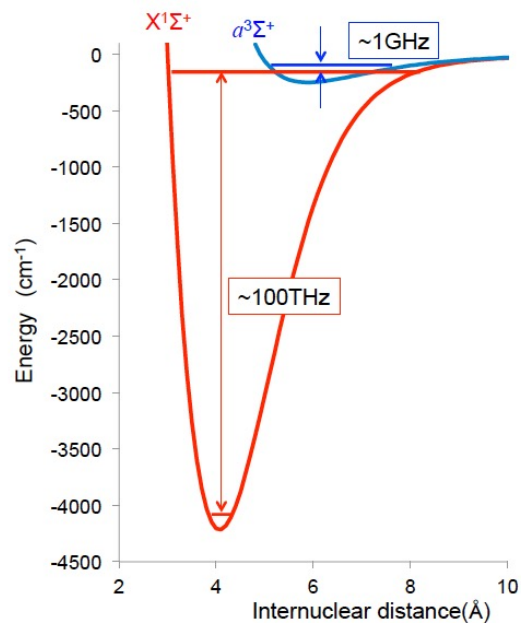
<sup>2</sup>Department of Physics and Astronomy, SUNY, Stony Brook, New York 11794, USA

<sup>3</sup>Physics Department, Temple University, Philadelphia, Pennsylvania 19122, USA

<sup>4</sup>Joint Quantum Institute and Atomic Physics Division, National Institute of Standards and Technology, Gaithersburg, Maryland 20899, USA

(Received 6 September 2007; published 29 January 2008)

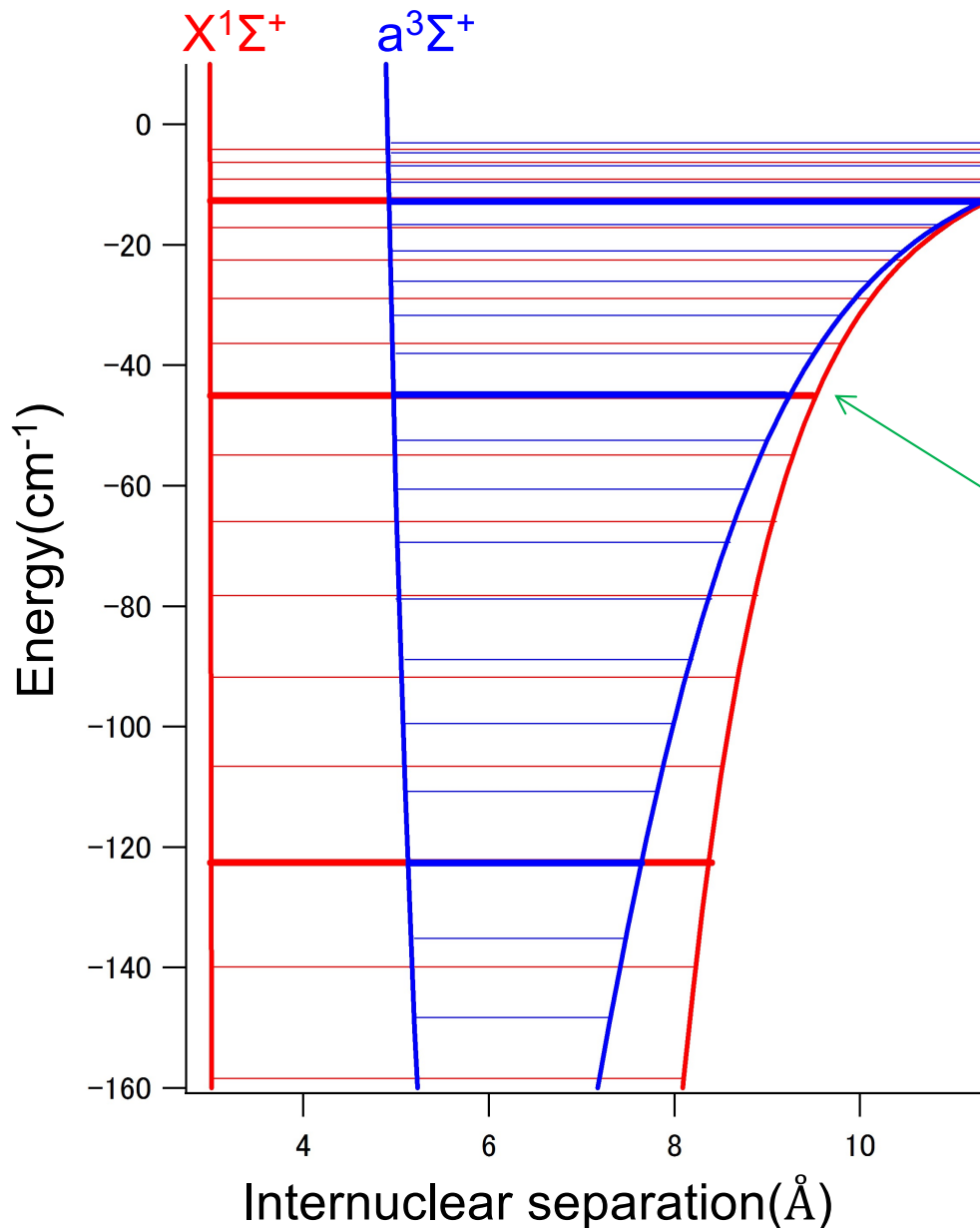
We propose new experiments with high sensitivity to a possible variation of the electron-to-proton mass ratio  $\mu \equiv m_e/m_p$ . We consider a nearly degenerate pair of molecular vibrational levels, each associated



# Candidates for “nearly degenerate levels” of $^{41}\text{K}^{87}\text{Rb}$

We need large sensitivity and non-vanishing mixing

$$\text{Sensitivity } S \equiv \frac{\partial f}{\partial \mu/\mu}$$



$v_X=91 \leftrightarrow v_a=22$   $\Delta\omega \sim 2.4\text{GHz}$   
 $S \sim 3.5\text{THz}$   
 Mixing  $\sim 9\%$

We selected this transition

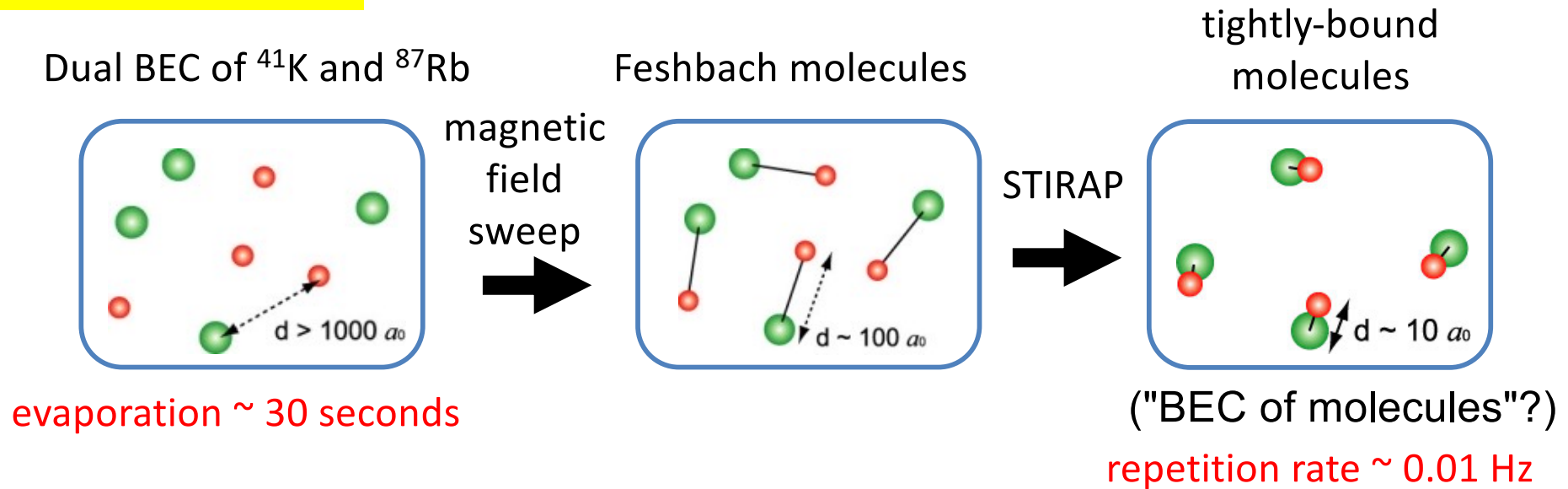
$v_X=86 \leftrightarrow v_a=16$   $\Delta\omega \sim 1\text{GHz}$   
 $S \sim 9.5\text{THz}$   
 Mixing  $\sim 2\%$

Enhancement:  $\frac{S}{\Delta\omega} = -14890(60)$

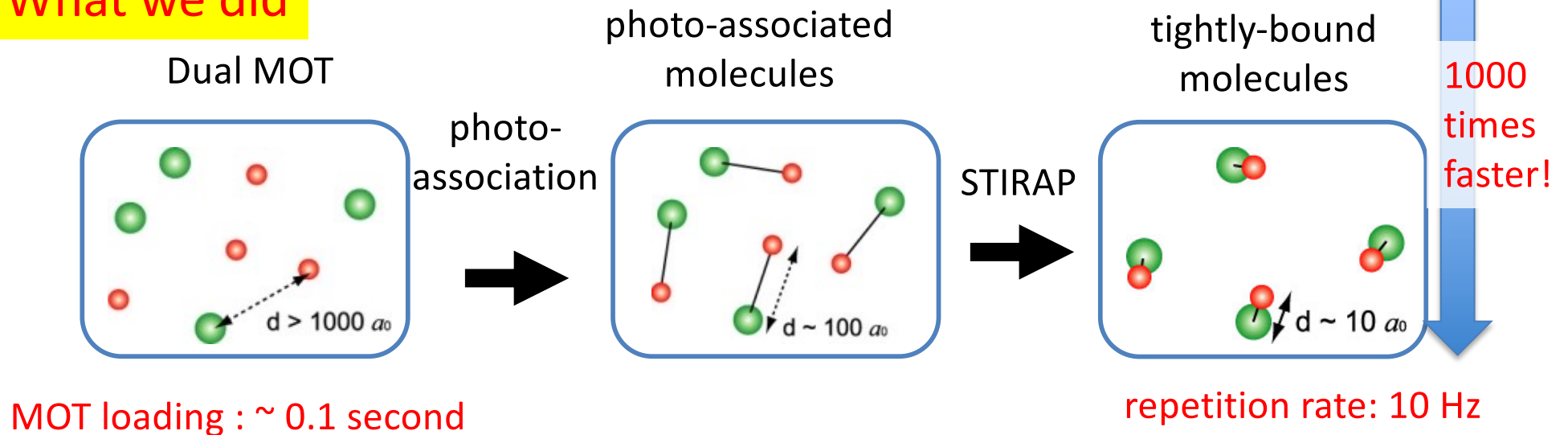
“optical sensitivity using microwave transition”

# "Indirect" method to produce ultracold molecules

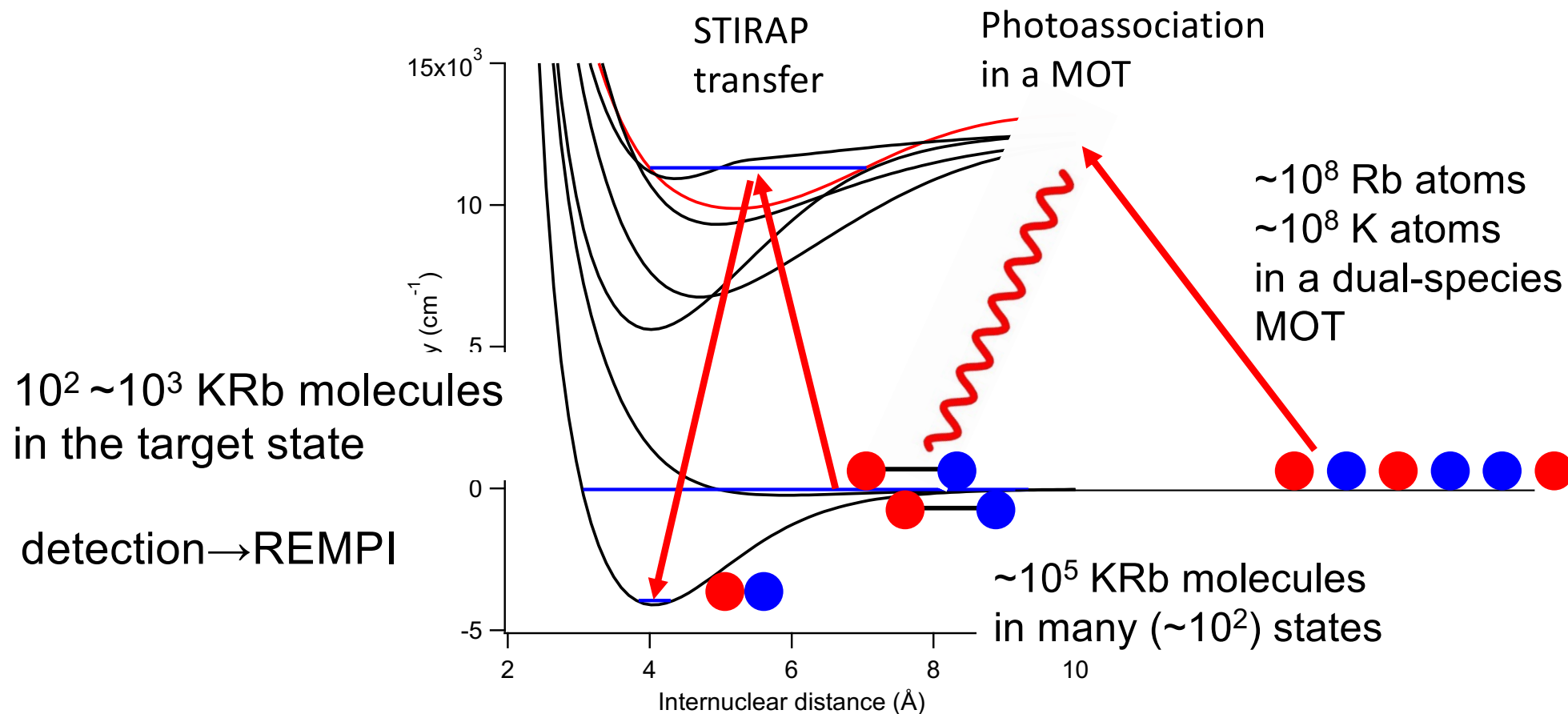
## Our initial plan



## What we did

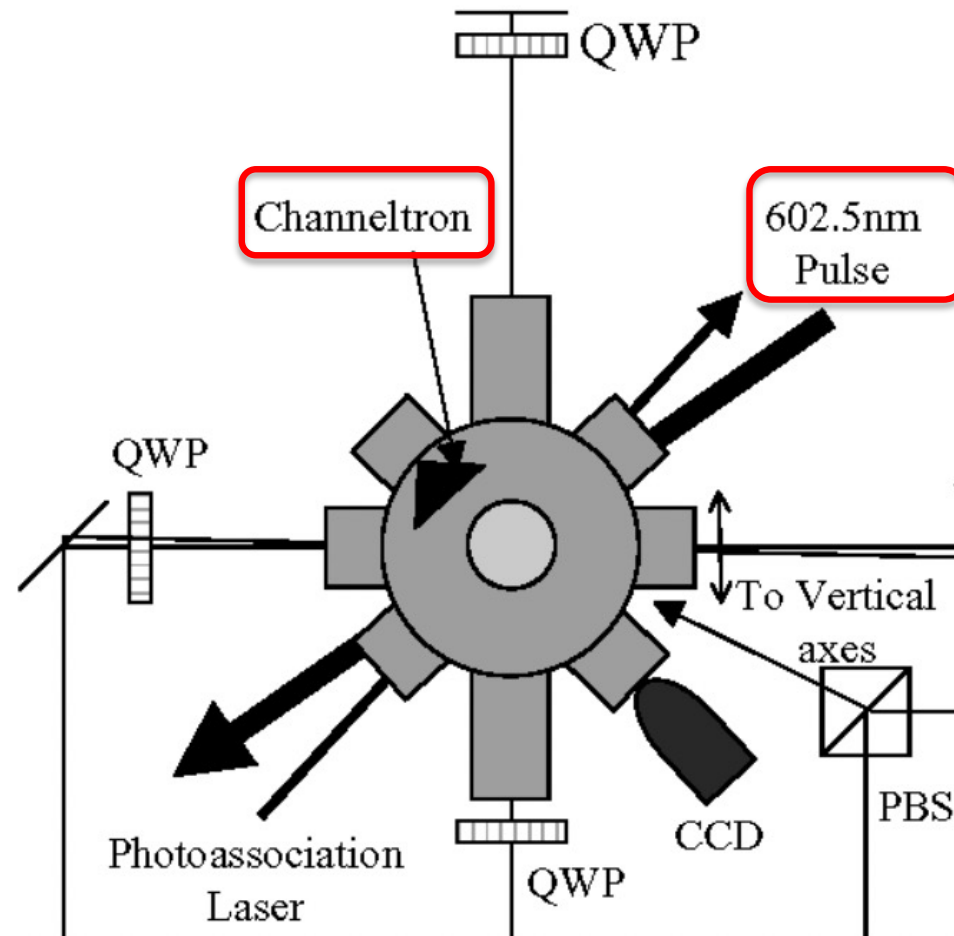


# Energy diagram



	PA+STIRAP	Feshbach + STIRAP
temperature	$\sim 100 \mu\text{K}$	$\sim 100 \text{nK}$
Number	$10^2 \sim 10^3?$	$10^5 \sim 10^6?$
Rate	$\sim 10 \text{ Hz}$	$\sim 0.01 \text{ Hz}$

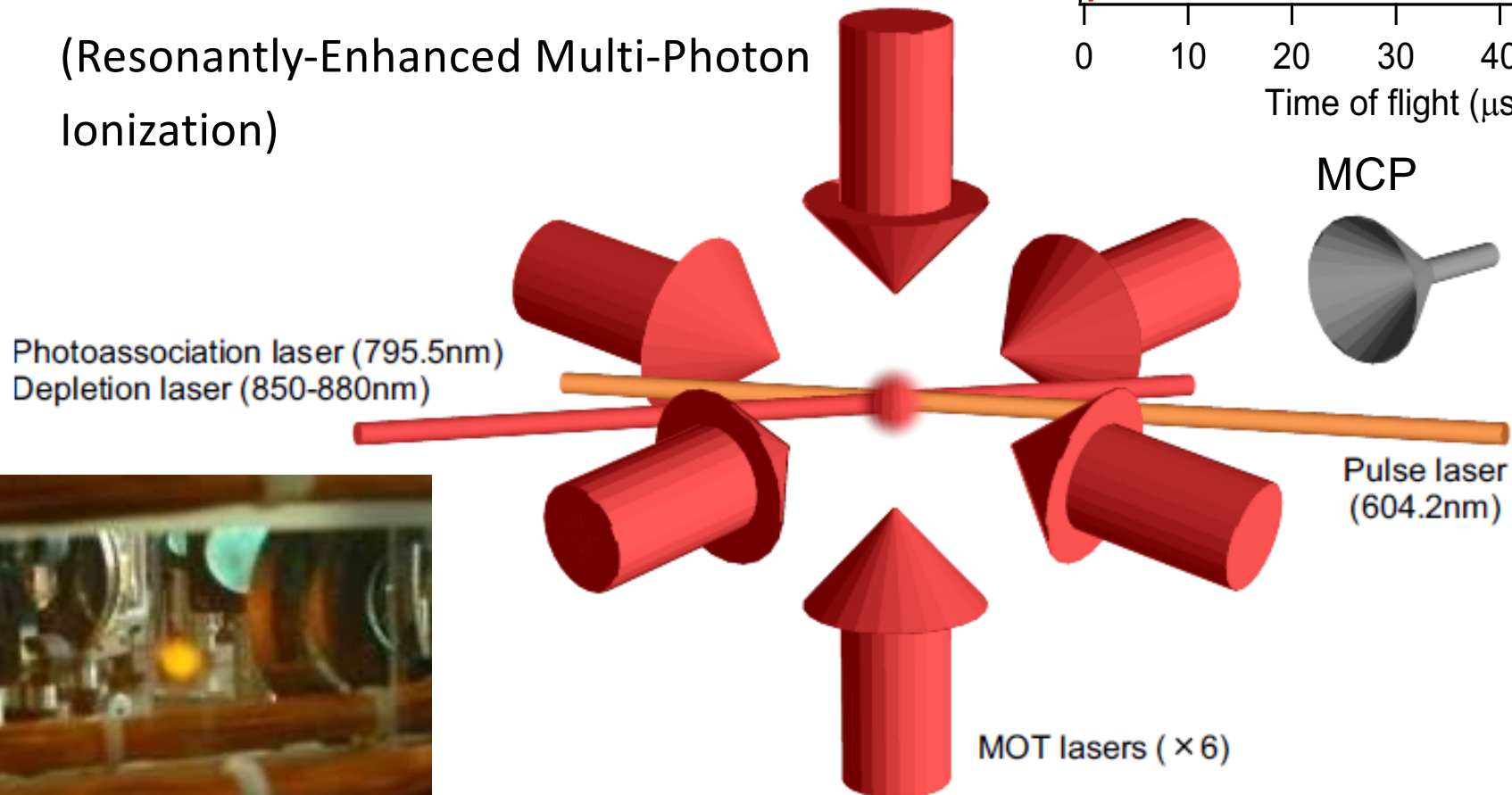
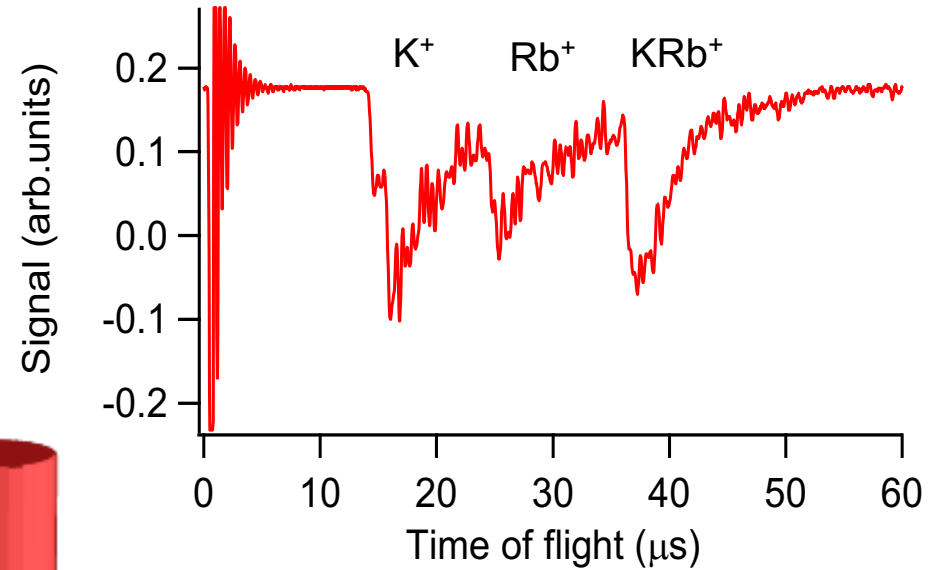
Detection of small number of molecules:  
Resonance-enhanced multiphoton ionization (REMPI)



D. Wang et al., Eur. Phys. J. D 31, 165–177 (2004)

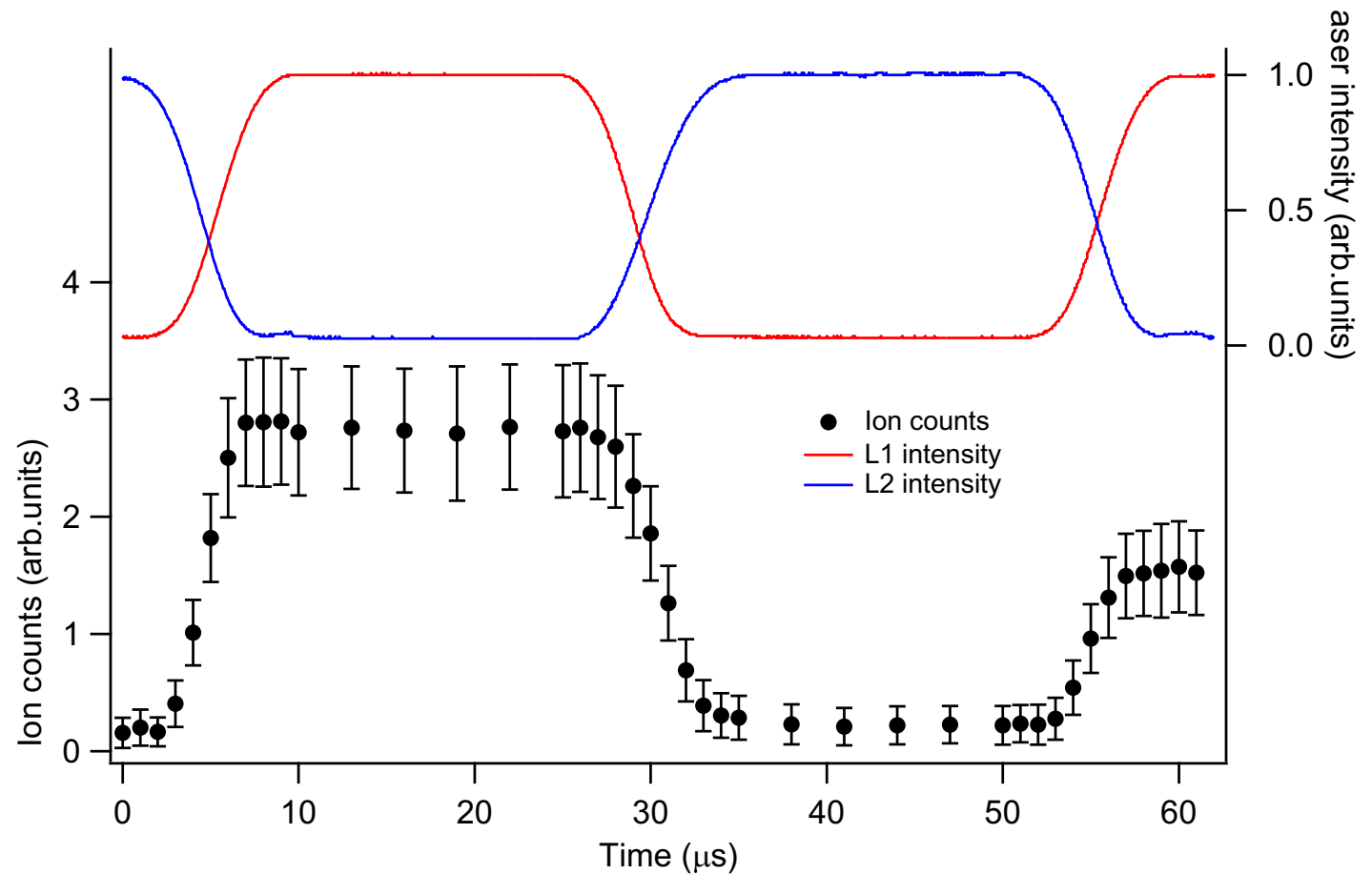
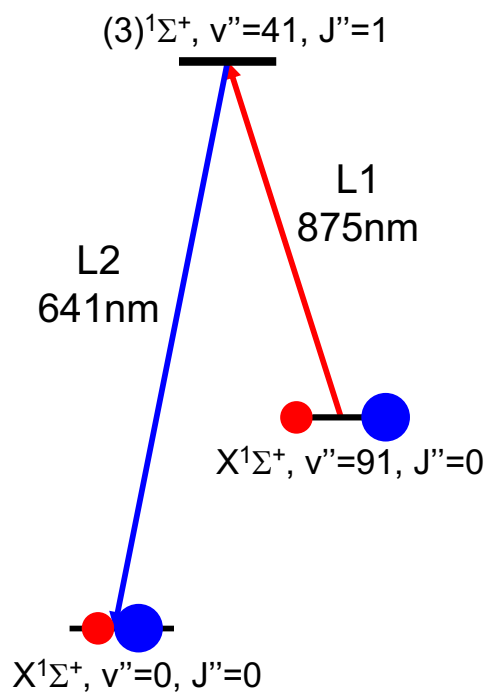
# Experimental setup

- MOT
- Photoassociation
- STIRAP
- rf - spectroscopy
- Detect molecules with REMPI  
(Resonantly-Enhanced Multi-Photon Ionization)

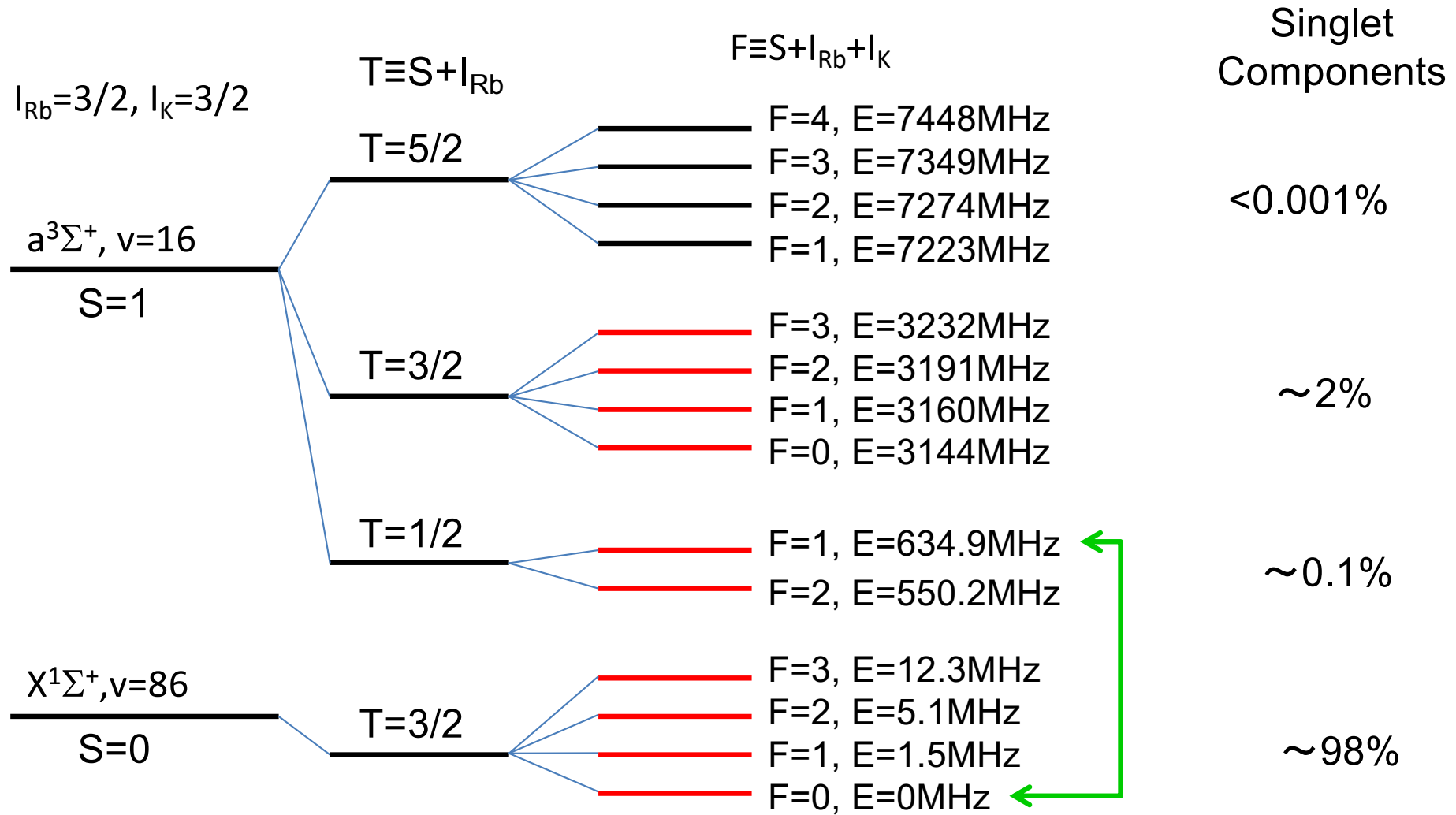


# STIRAP

- Monitored the ion counts during multiple transfers
- Recovered 53% after the double STIRAP
- One-way transfer efficiency: 73%



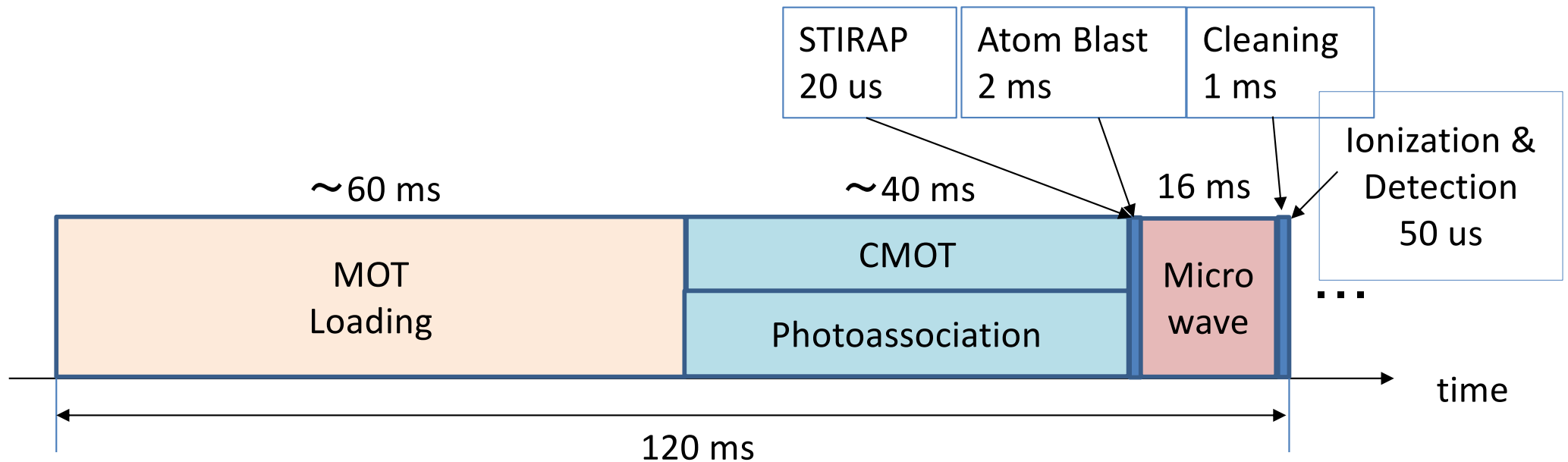
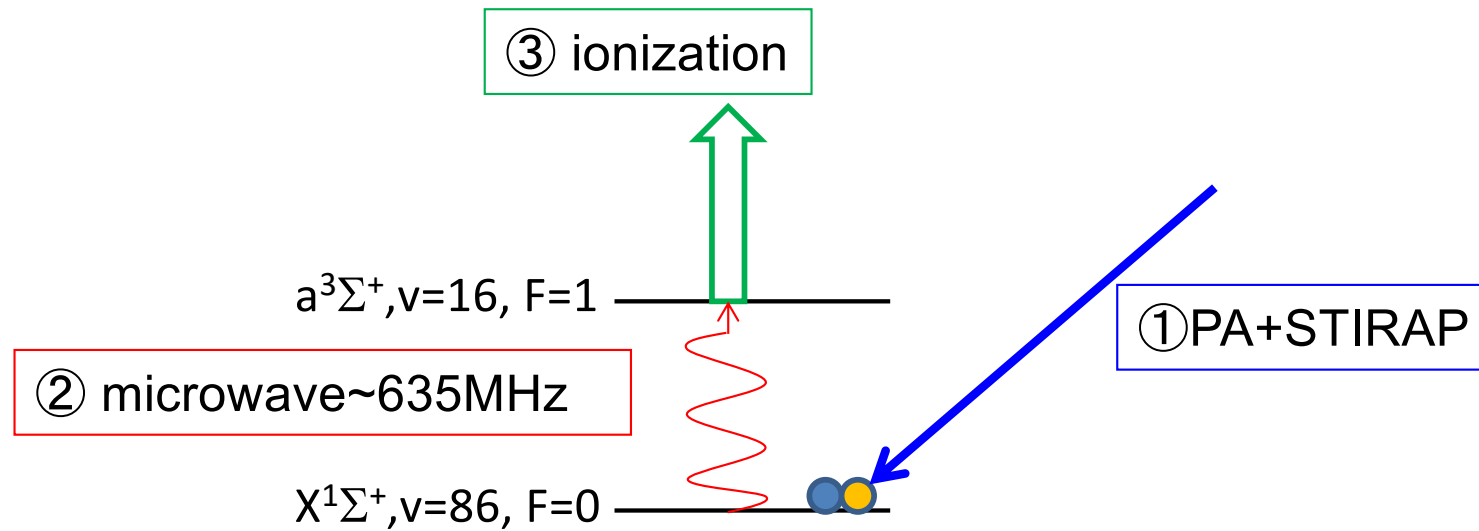
# Hyperfine structure of $X^1\Sigma^+ v=86$ and $a^3\Sigma^+ v=16$



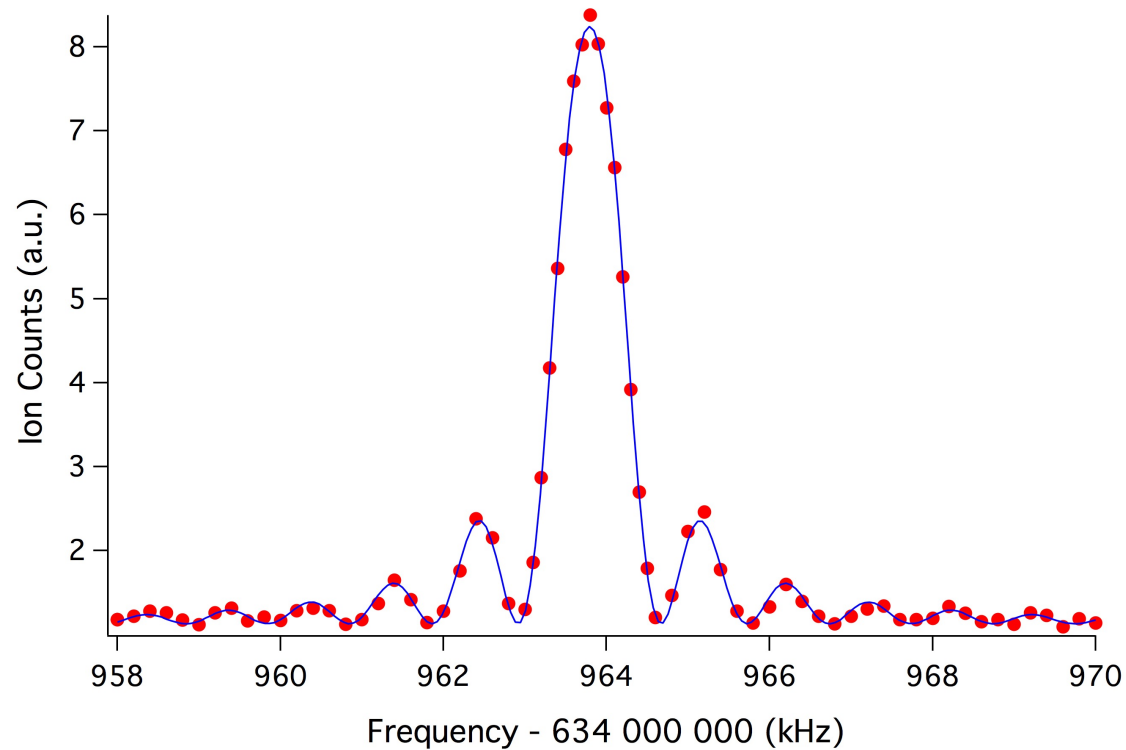
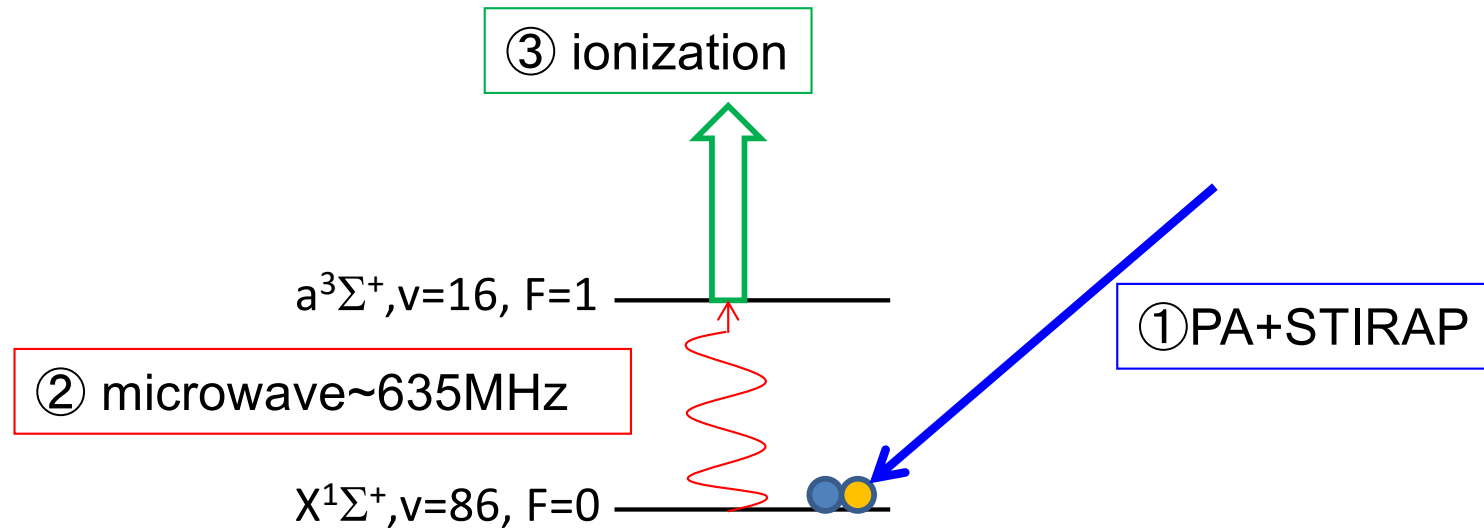
Use this microwave transition!



# Microwave transition between $X^1\Sigma^+, v=86, F=0 \rightarrow a^3\Sigma^+, v=16, F=1$



# Microwave transition between $X^1\Sigma^+, v=86, F=0 \rightarrow a^3\Sigma^+, v=16, F=1$

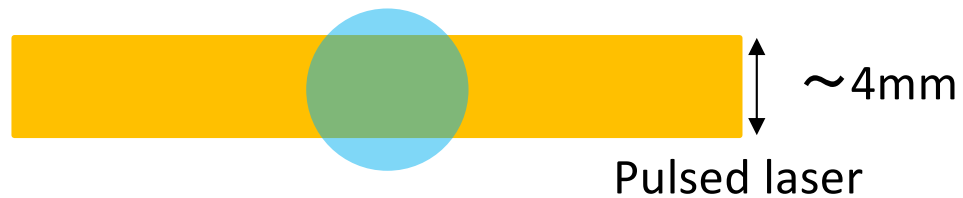


# Thermal diffusion of molecular cloud

Photoassociated KRb

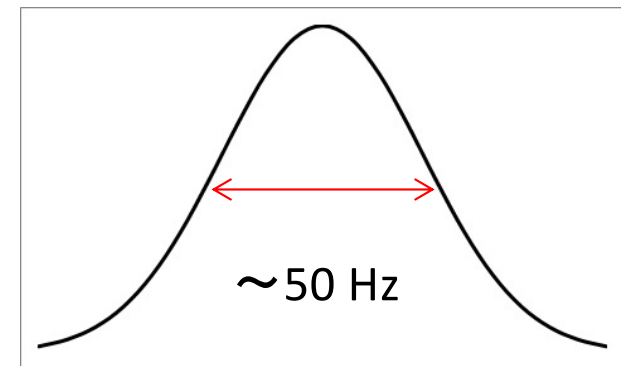
●  $T \sim 130 \mu\text{K}$ ,  $\bar{v} \sim 130 \text{ mm/s}$

↓ Thermal diffusion

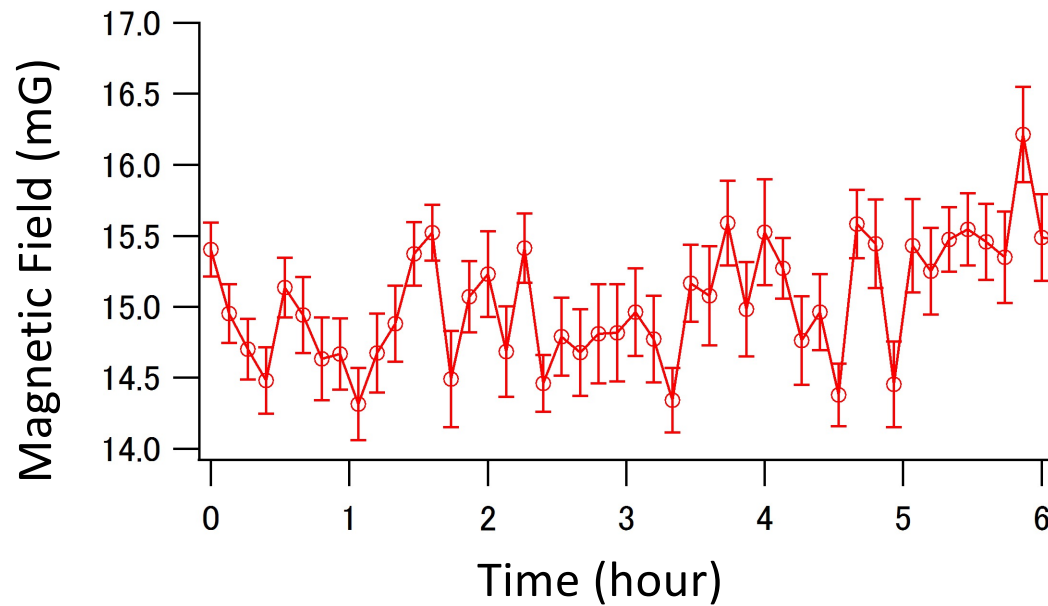
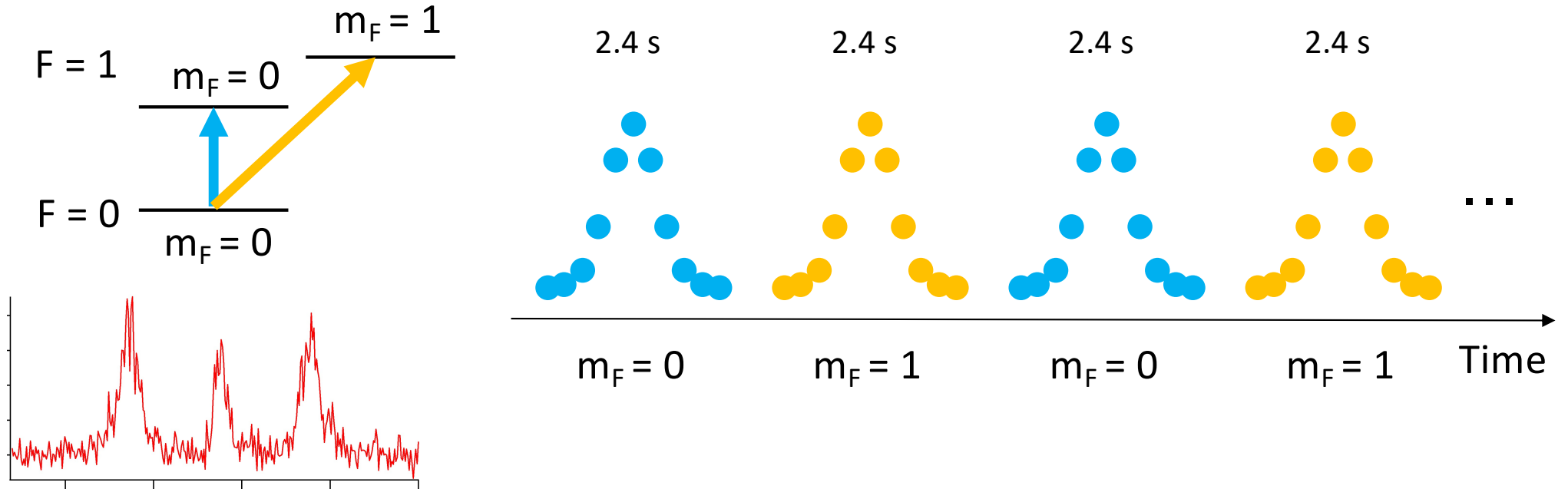


Measurement time  $\sim 16 \text{ ms}$

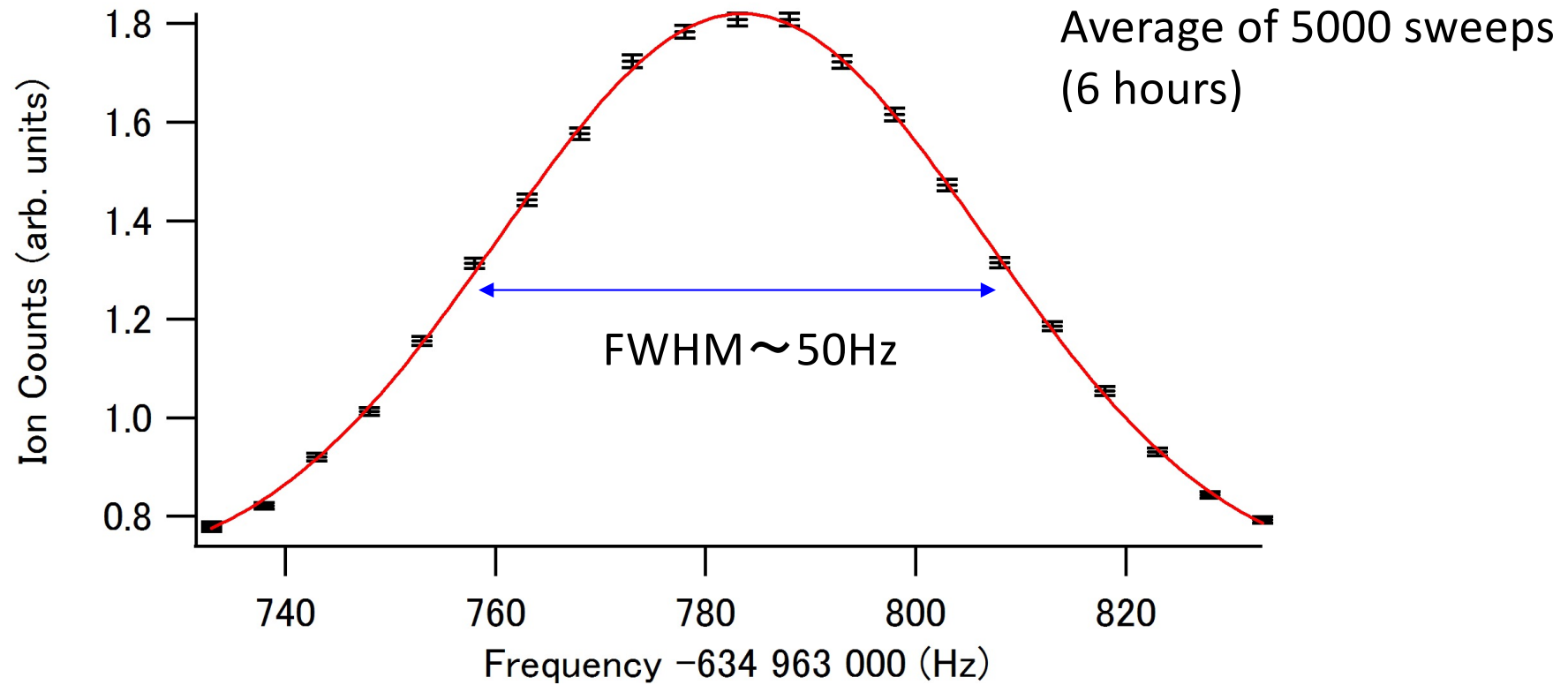
↓ ( $\pi$  pulse)



# Calibration of the magnetic field



# Obtained signal for $m_F = 0$



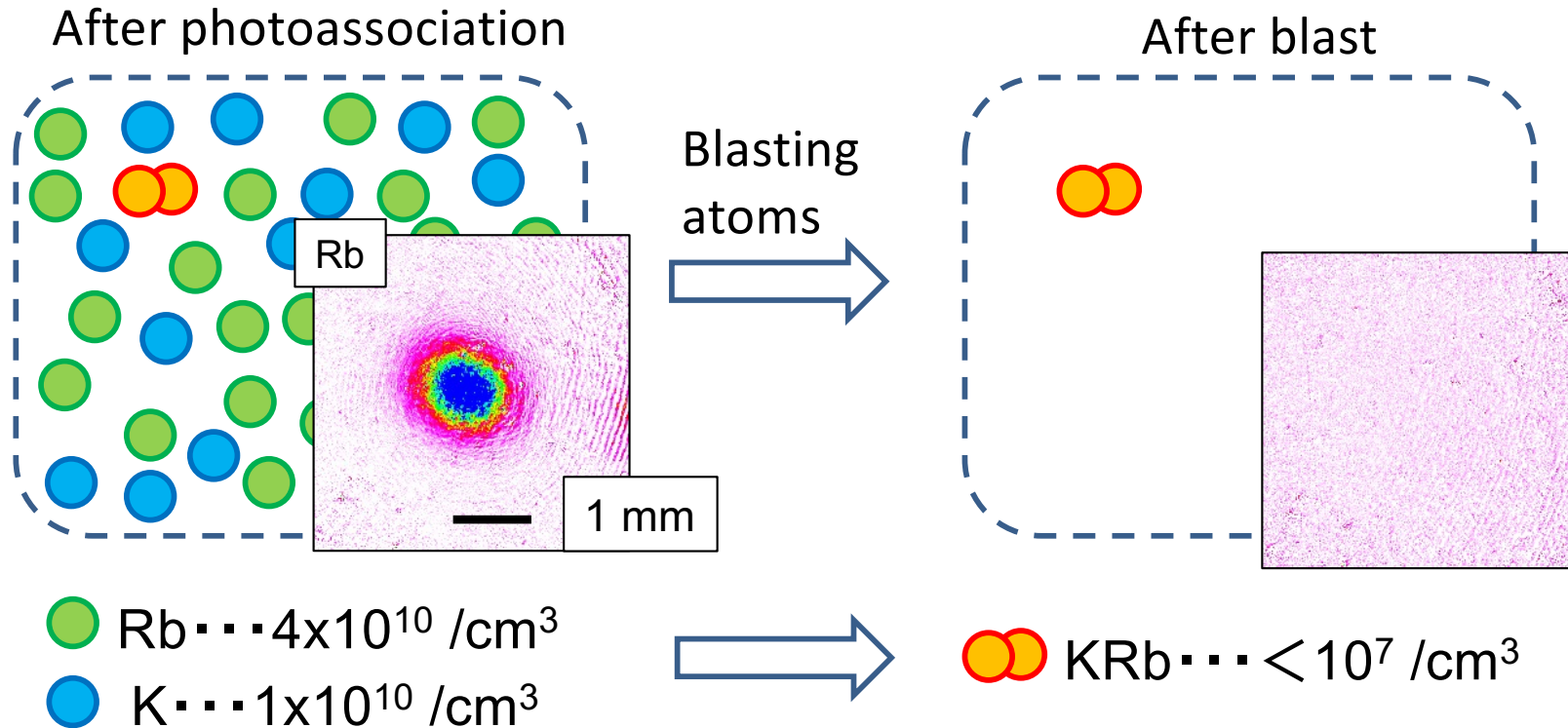
634 963 783.458  $\pm$  0.093 Hz

634 963 781.564  $\pm$  0.094 Hz

Zeeman shift  
compensation

S/N  $\sim 500$  (c.f. Number of molecules used  $\sim 10^6$ )

# Collisional shift



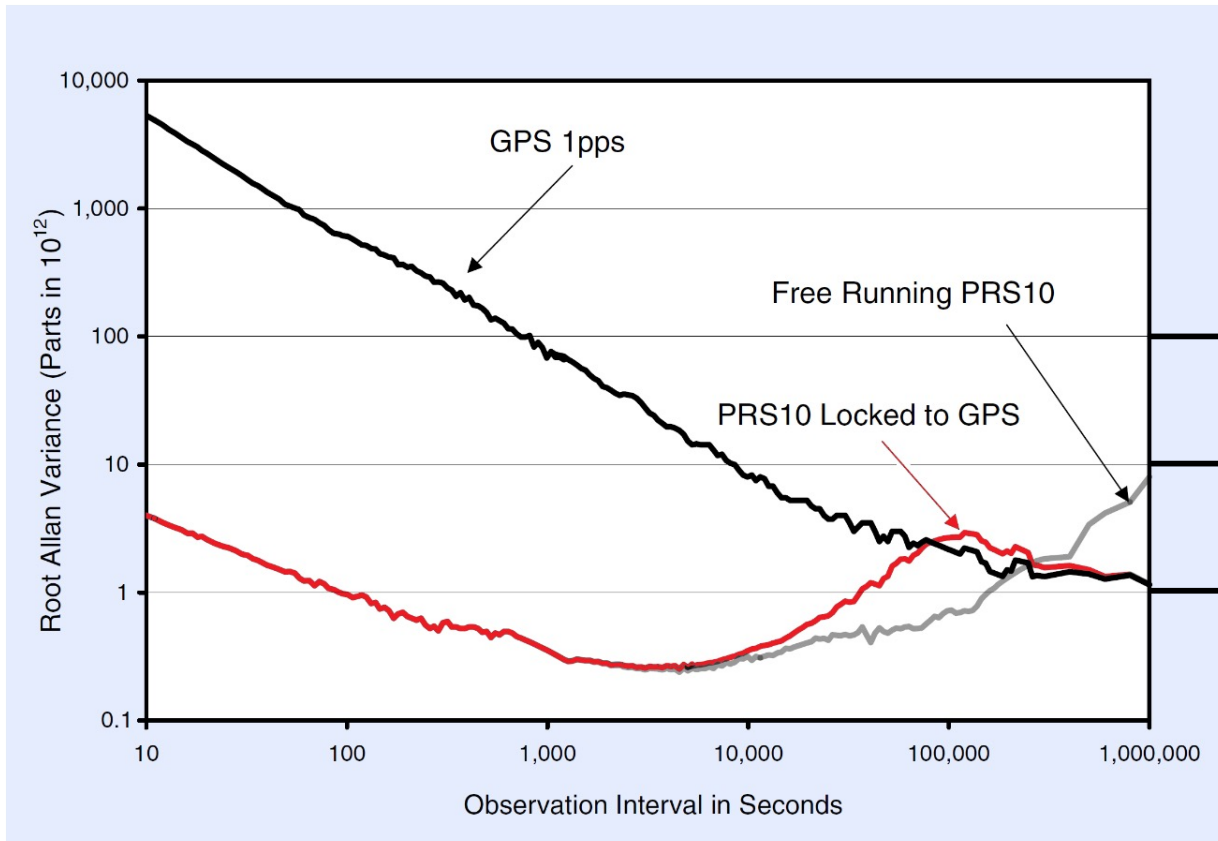
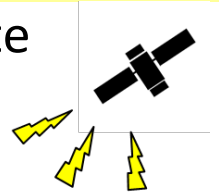
When we removed the blast, atomic density increased by more than factor of 100, but no shift observed

$\Rightarrow$  Collisional shift should be negligible

# Reference Clock

SRS FS725 locked to GPS signal

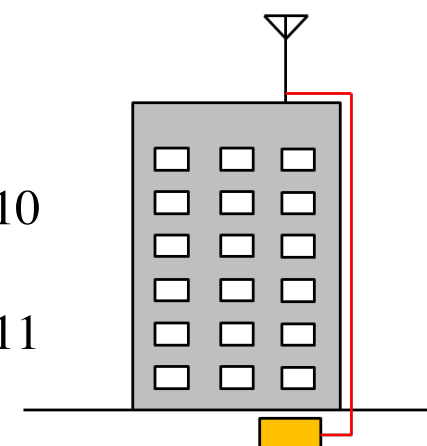
GPS satellite



$10^{-10}$

$10^{-11}$

$10^{-12}$



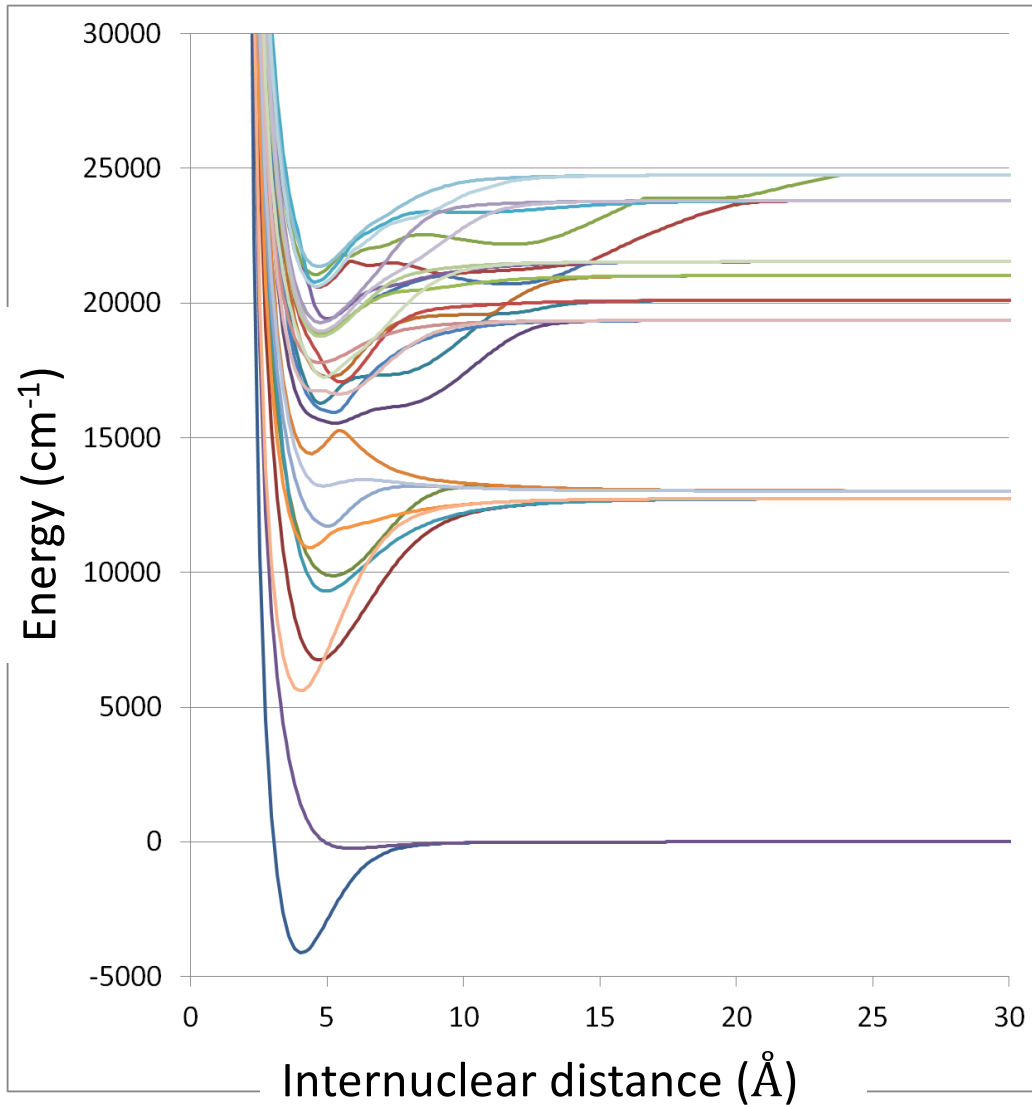
Laboratory

$$\frac{\delta f}{f} < 3 \times 10^{-12}$$

$$\delta f_{\text{sys}} = \pm 1 \text{ mHz}$$

# Black Body Radiation shift

Potential Energy curves for KRb



Ab-initio calculation of the transition dipole moment  
[R.Beuc et al., J. Phys. B 39 S1191 (2006)]

BBR shift @ 300K

$a^3\Sigma^+, v=16$  -2.89(14) Hz

$X^1\Sigma^+, v=86$  -2.81(14) Hz

$T = 26.8 \pm 2.0 \text{ }^\circ\text{C}$

$\Rightarrow \delta f_{\text{sys}} = \pm 10 \text{ mHz}$



# Uncertainties (Error Budget)

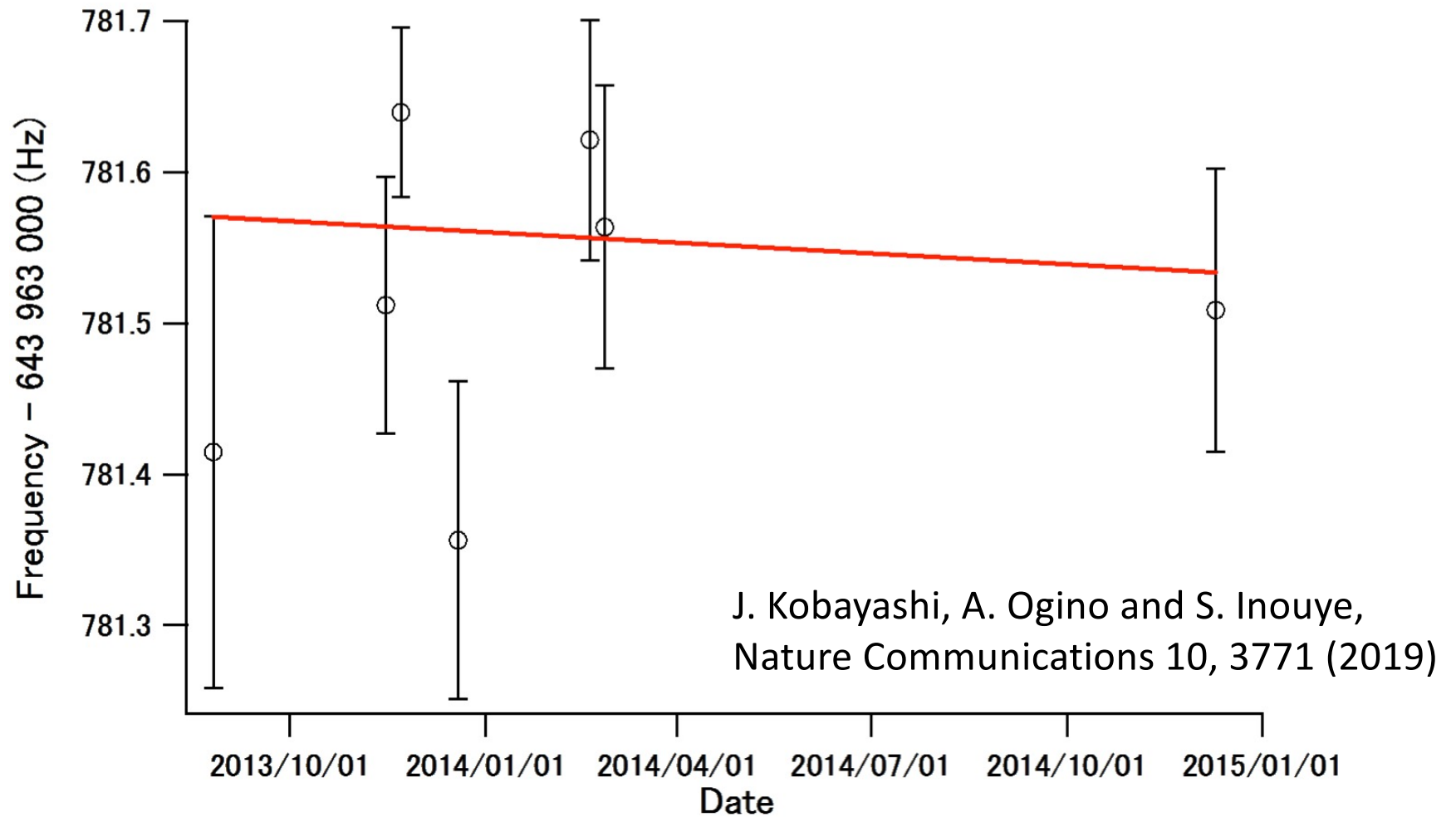
$^{41}\text{K}^{87}\text{Rb}$  ( $X^1\Sigma^+, v=86, N=0, T=3/2, F=0, m_F=0$ )  
 $\rightarrow a^3\Sigma^+, v=16, N=0, T=1/2, F=1, m_F=0$ )

	<b>Statistic</b>	<b>Systematic</b>
S/N of the signal	93 mHz	-
Zeeman shift	15 mHz	1 mHz
Stark shift	-	1 mHz
Reference clock	-	< 1 mHz
BBR shift	-	10 mHz
Density shift		< 1 mHz
<b>Total</b>	<b>94 mHz</b>	<b>10 mHz</b>

$$\delta f_{\text{total}} = 95 \text{ mHz}$$

$$\frac{\delta\mu}{\mu} = 1.0 \times 10^{-14}$$

# Final result



$$\frac{1}{\mu} \frac{\partial \mu}{\partial t} = \left( 0.30 \pm 1.00_{\text{Stat}} \pm 0.16_{\text{Sys}} \right) \times 10^{-14} / \text{year}$$

## Conclusion

- We measured the variation of electron-to-proton mass ratio using ultracold molecules produced from laser-cooled atoms
- Obtained result was  $\frac{1}{\mu} \frac{d\mu}{dt} = \left(0.30 \pm 1.00_{St} \pm 0.16_{Sys}\right) \times 10^{-14} / \text{year}$ , which was consistent with zero (Nat. Comm. 10, 3771 (2019))

## Outlook

Dr. Kobayashi and the machine moved to Hokkaido (North of Japan). Current plan for the 2<sup>nd</sup> generation is:

- increase the observation time by trapping the molecules
- increase number of molecules by creating from a BEC trapped in a 3D optical lattice
- keep the repetition rate by creating BEC via direct laser cooling

A. Ogino     Dr. J. Kobayashi

