

# Probing the Variation of Fundamental Constants with Polar Molecule Microwave Spectroscopy

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# OUTLINE

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- Background
  - Importance of variation
  - Why should fundamental constants vary?
  - Current state of affairs
- Our experiment
  - Producing cold, slow laboratory OH
  - Measuring  $\Lambda$ -doublet transition frequencies
- Future work
  - New experiment based on Th-229 nuclear transition

# Why should we care if the fundamental constants vary?

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- They're supposed to be constant
- Varying fundamental constants violate both Lorentz invariance and CPT symmetry.

# Why should we think the fundamental constants could vary?

- Attempts to unify gravity predict (or allow for) space-time varying fundamental constants

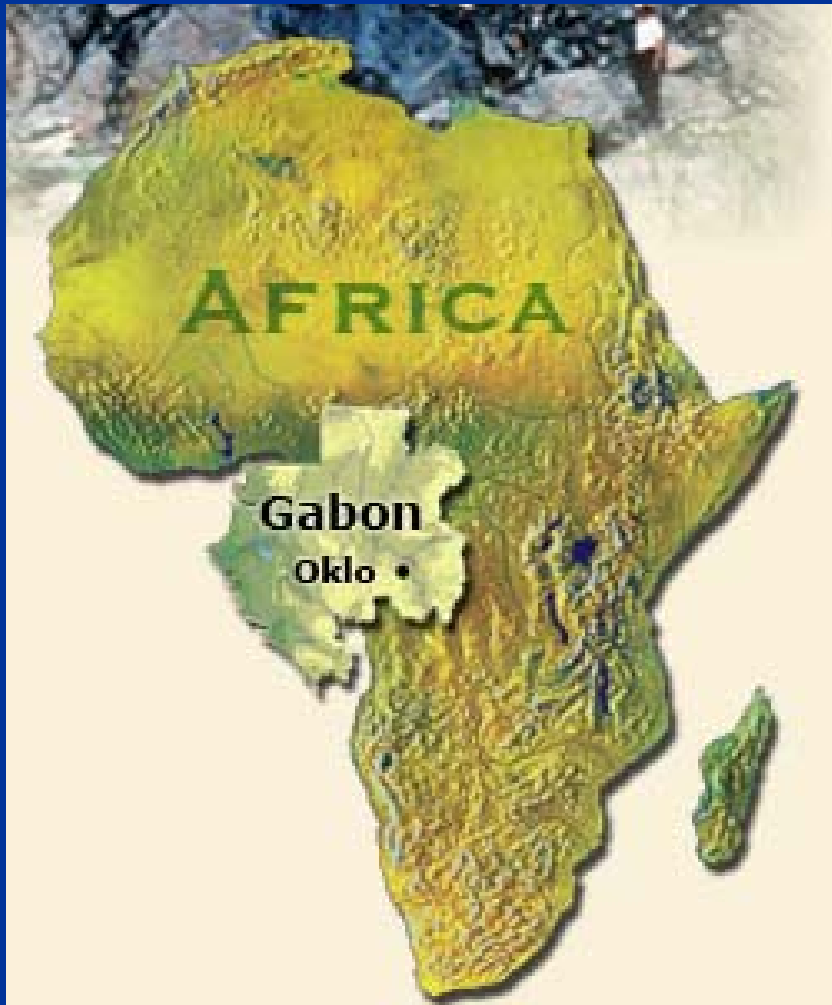
Why should they be constant?

- Interaction with dark energy quintessence field leads to varying fundamental constants.

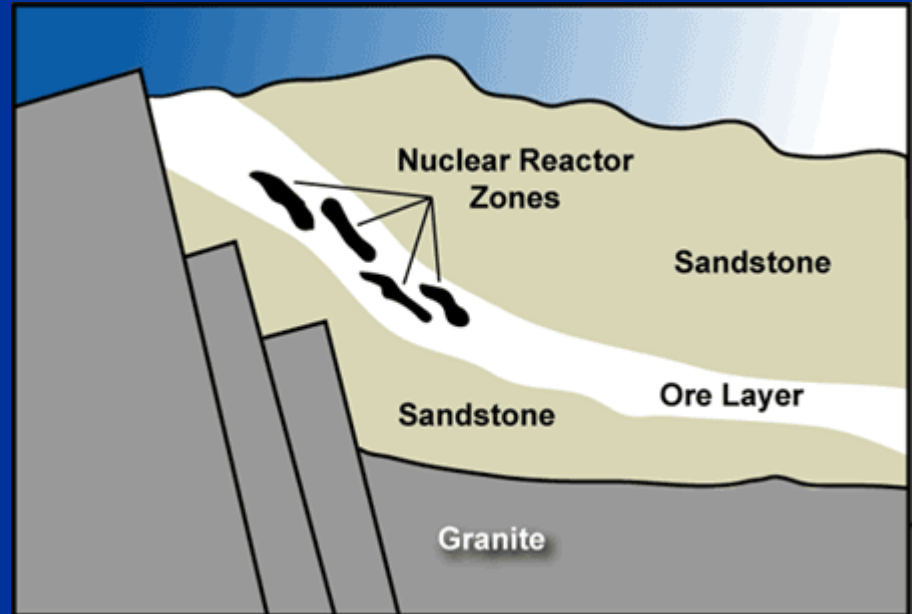
–Quintessence - photon interaction  $\rightarrow \delta_i \alpha \neq 0$

–Quintessence - electron interaction  $\rightarrow \delta_i m_e \neq 0$

# Oklo Natural Nuclear Reactor

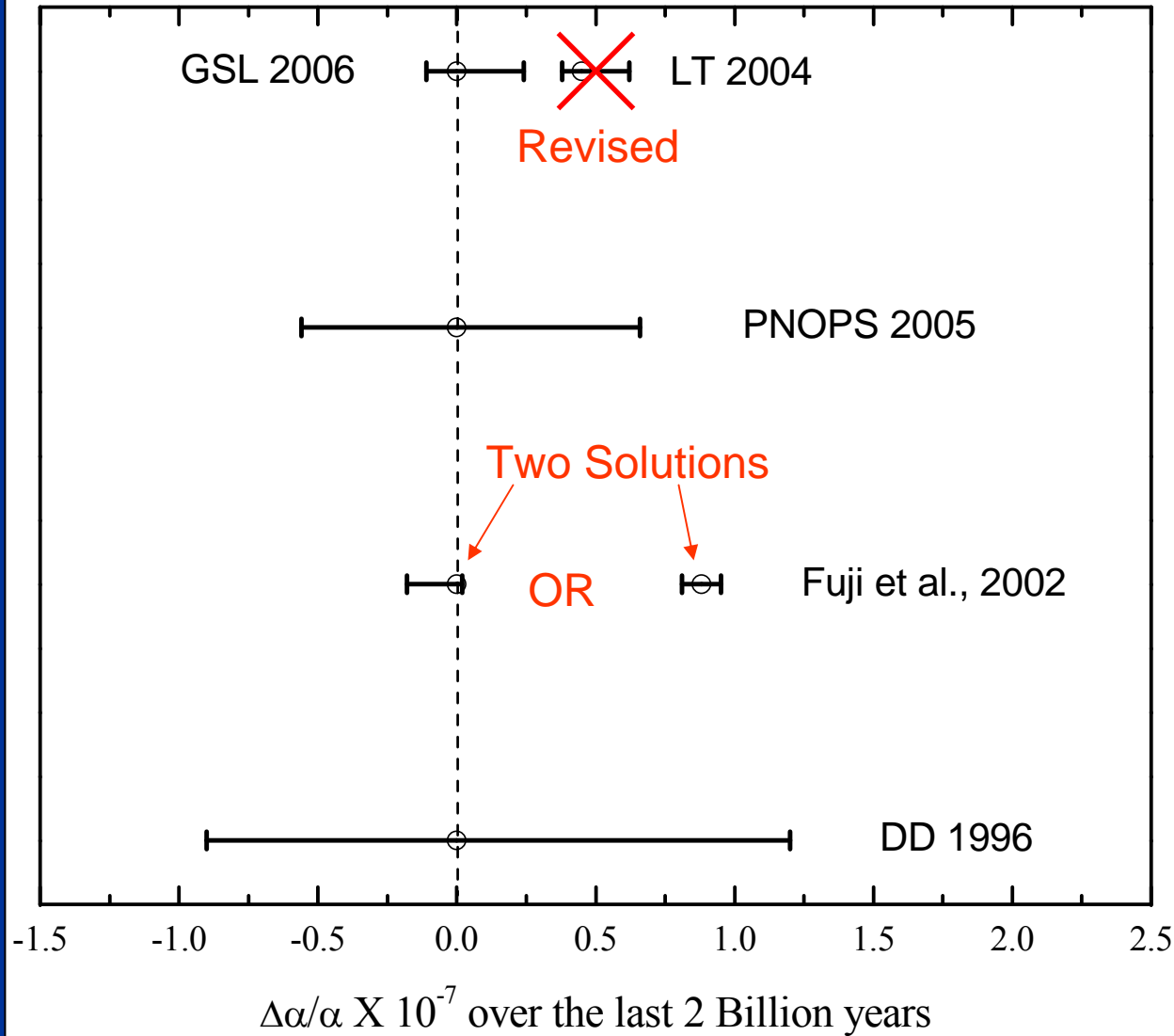


The uranium isotopes found at Oklo strongly resemble those in the spent nuclear fuel generated by today's nuclear power plants.



- Currently  $^{238}\text{U} \sim 99.3\%$  and  $^{235}\text{U} \sim 0.7\%$
- 2 billion years ago  $^{235}\text{U} \sim 4\%$  (typical reactor concentrations)
- Neutron capture cross-section for  $^{149}\text{Sm}$  sensitive to  $\Delta\alpha$  because of 97.3 meV resonance.

# Oklo Reactor



Consistent with zero:

$$\frac{\Delta\alpha}{\alpha} \leq 1(10^{-17})\text{yr}^{-1}$$

- (LT 2004) PRD 69 121701
- (PNOPS 2005) arXiv: hep-ph/0506186
- (DD 1996) Nucl. Phys. B480 37
- Fujii et al., AriXiv:hep-ph0205206
- (GSL 2006) PRC 74 024607

# Atomic clock measurements

## Simple Idea:

Measure atomic transition frequencies and see if they vary with time

## Problem:

To measure a frequency you need a 'clock' (i.e. another frequency)

## Solution:

Use Cs hyperfine transition as a reference

Optical transitions:

$$\nu_i \sim Ry F_i(\alpha)$$

Hyperfine transitions:

$$\nu_j \sim \alpha^2 (\mu_{Cs} / \mu_B) Ry F_j(\alpha)$$

Relativistic Correction:

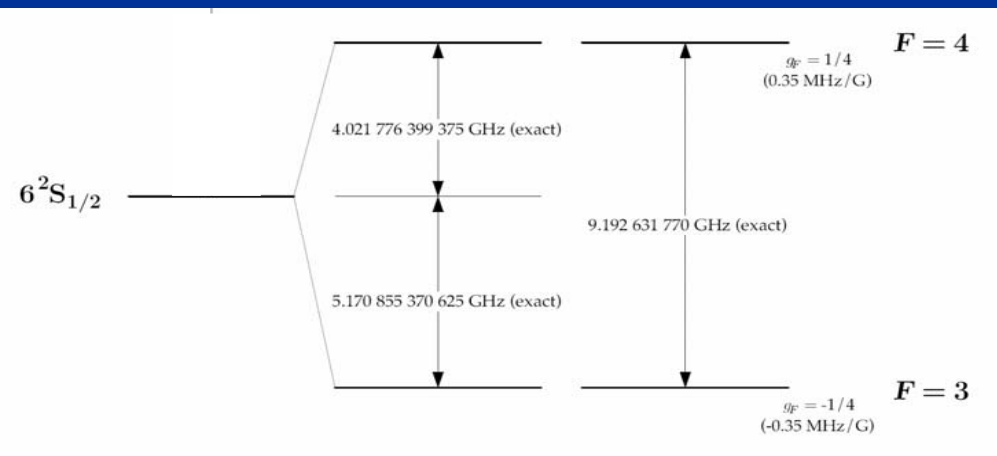
$$F(\alpha) \sim \alpha^N$$

Fractional variation:

$$\frac{d}{dt} \ln \left[ \frac{\nu_{Optical}}{\nu_{Cs}} \right] = (N_{Optical} - N_{Cs} - 2) \frac{\dot{\alpha}}{\alpha} - \frac{d}{dt} \ln \left[ \frac{\mu_{Cs}}{\mu_B} \right]$$

$$N_{Cs} = 0.8$$

$$N_{Hg+} = -3.2$$

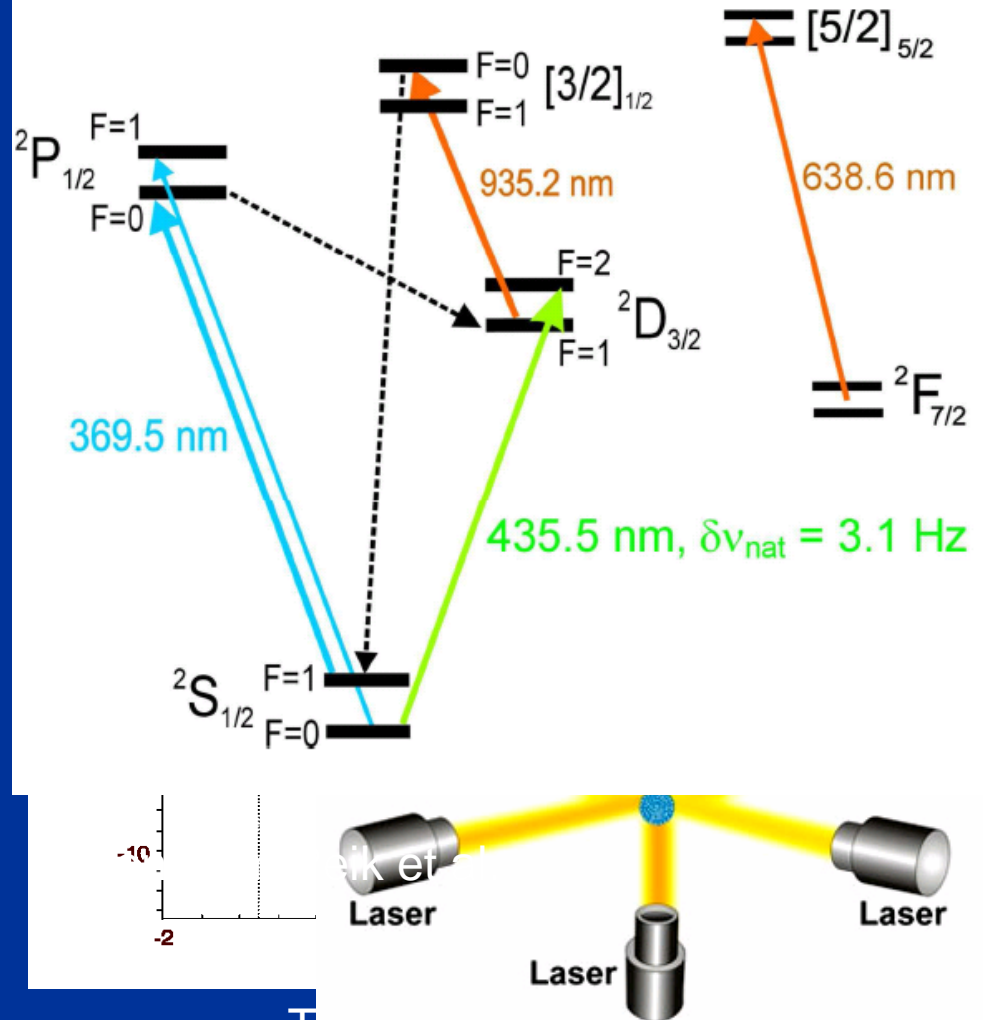


# Atomic clock measurements

## Several excellent experiments:

- $^{199}\text{Hg}^+$  vs. Cs
- H vs. Cs
- $^{171}\text{Yb}^+$  vs. Cs
- Rb vs. Cs
- Future: Sr, Yb, Ca, Al+...

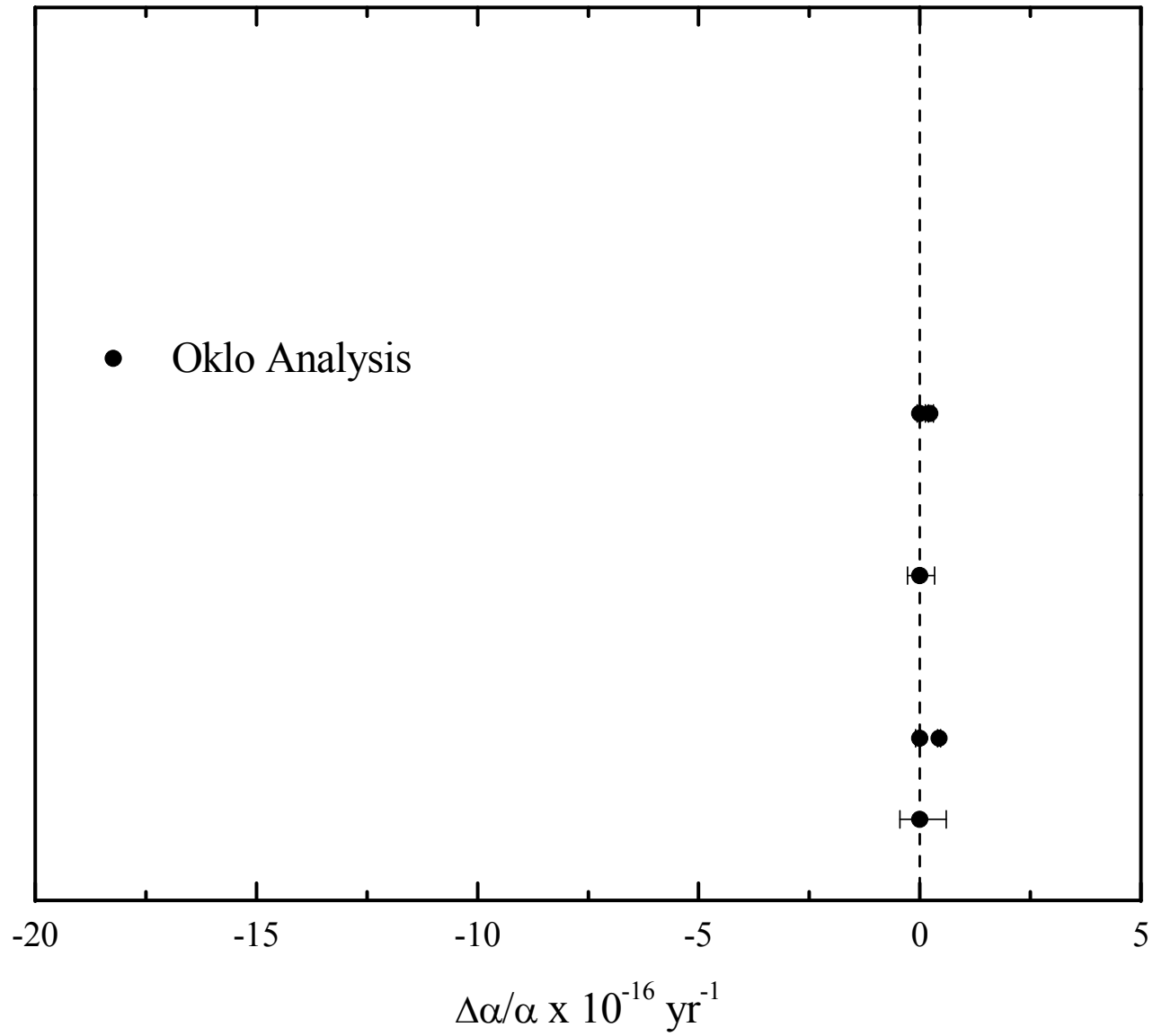
## $^{171}\text{Yb}^+$ level scheme



Taken from PRL 98 070801



# $\Delta\alpha/\alpha$ Status





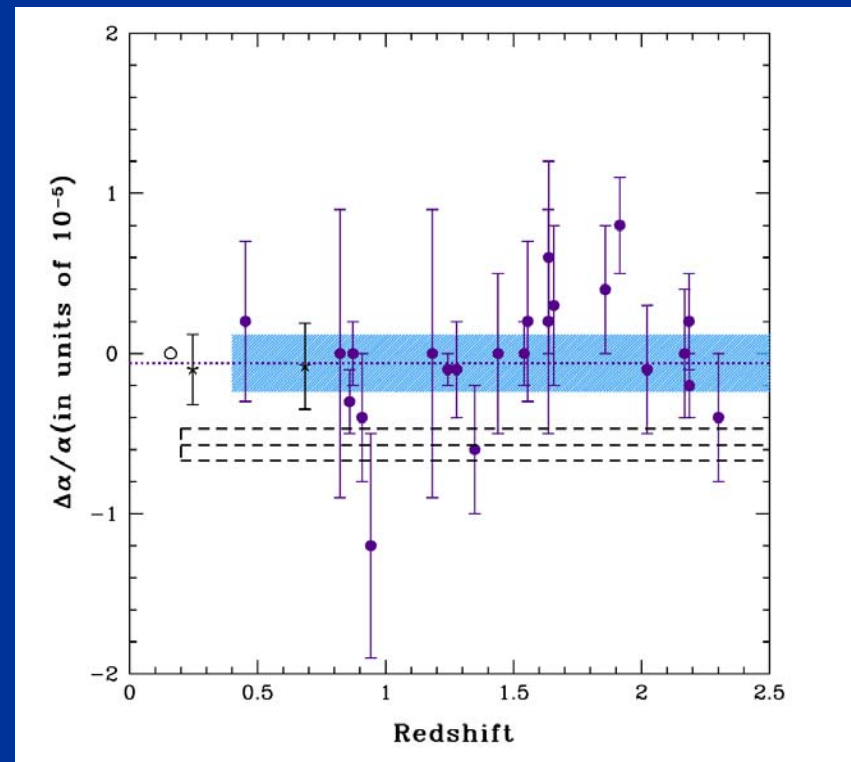
# Astrophysical measurements

## Quasar Absorption:

- Conceptually the same as atomic clock measurements.
- Quasars emit over a large spectrum
  - Look for absorption from gas between the quasar and us

## Other possibilities from astrophysics:

- Non-zero  $\Delta\alpha$  causes change to the CMB pattern. Look back to  $z \sim 1000$ ,  $\Delta\alpha \sim 10^{-3}$   
PRD 60, 023516
- Big Bang Nucleosynthesis



Taken from: R.Srianand et al PRL, 92 121302



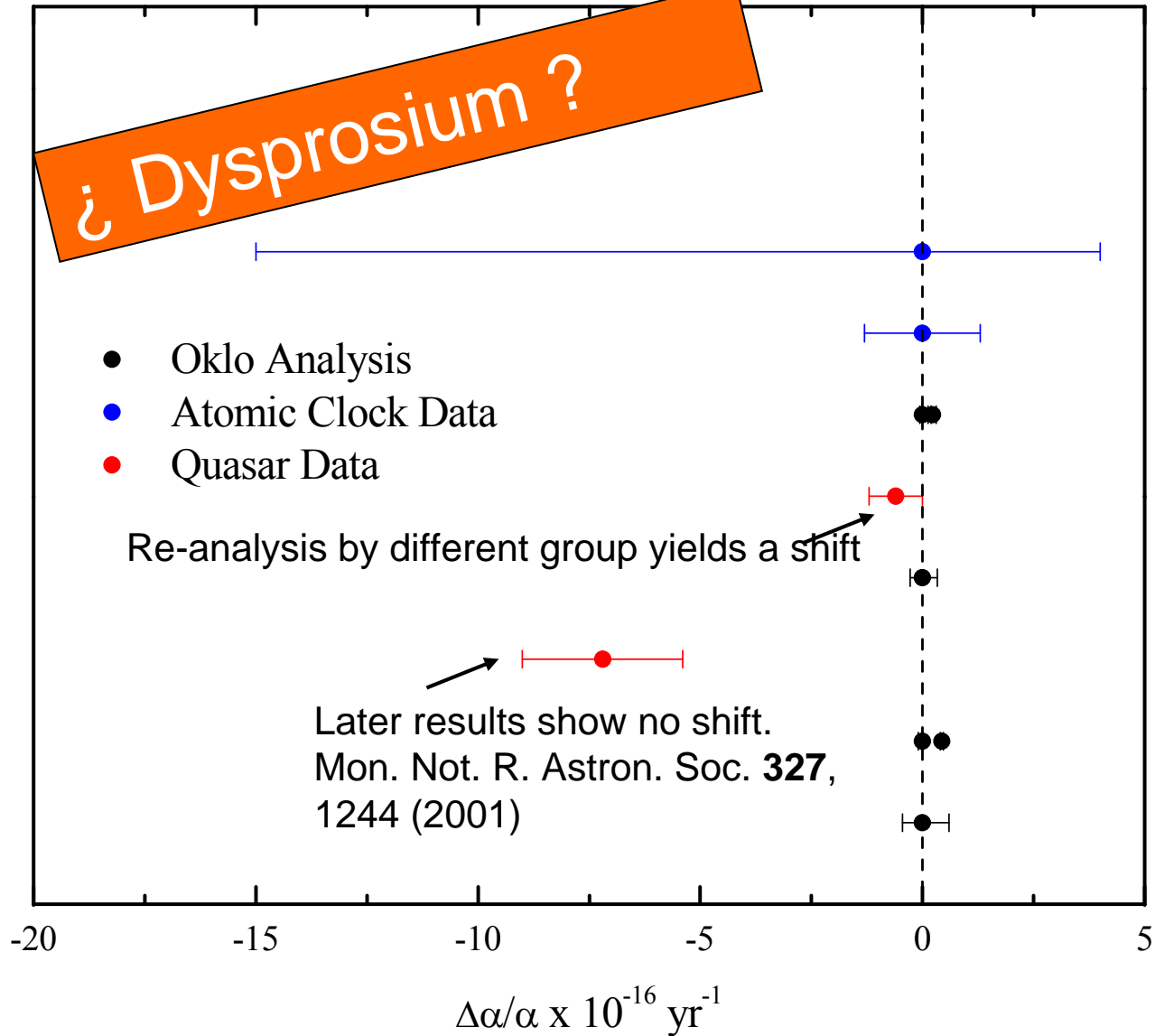
# $\Delta\alpha/\alpha$ Status

¿ Dysprosium ?

- Oklo Analysis
- Atomic Clock Data
- Quasar Data

Re-analysis by different group yields a shift

Later results show no shift.  
Mon. Not. R. Astron. Soc. **327**,  
1244 (2001)



Quasar Data:  
PRL 87 091301  
PRL 92 121302

Atomic Clock Data:  
PRL 98 070801

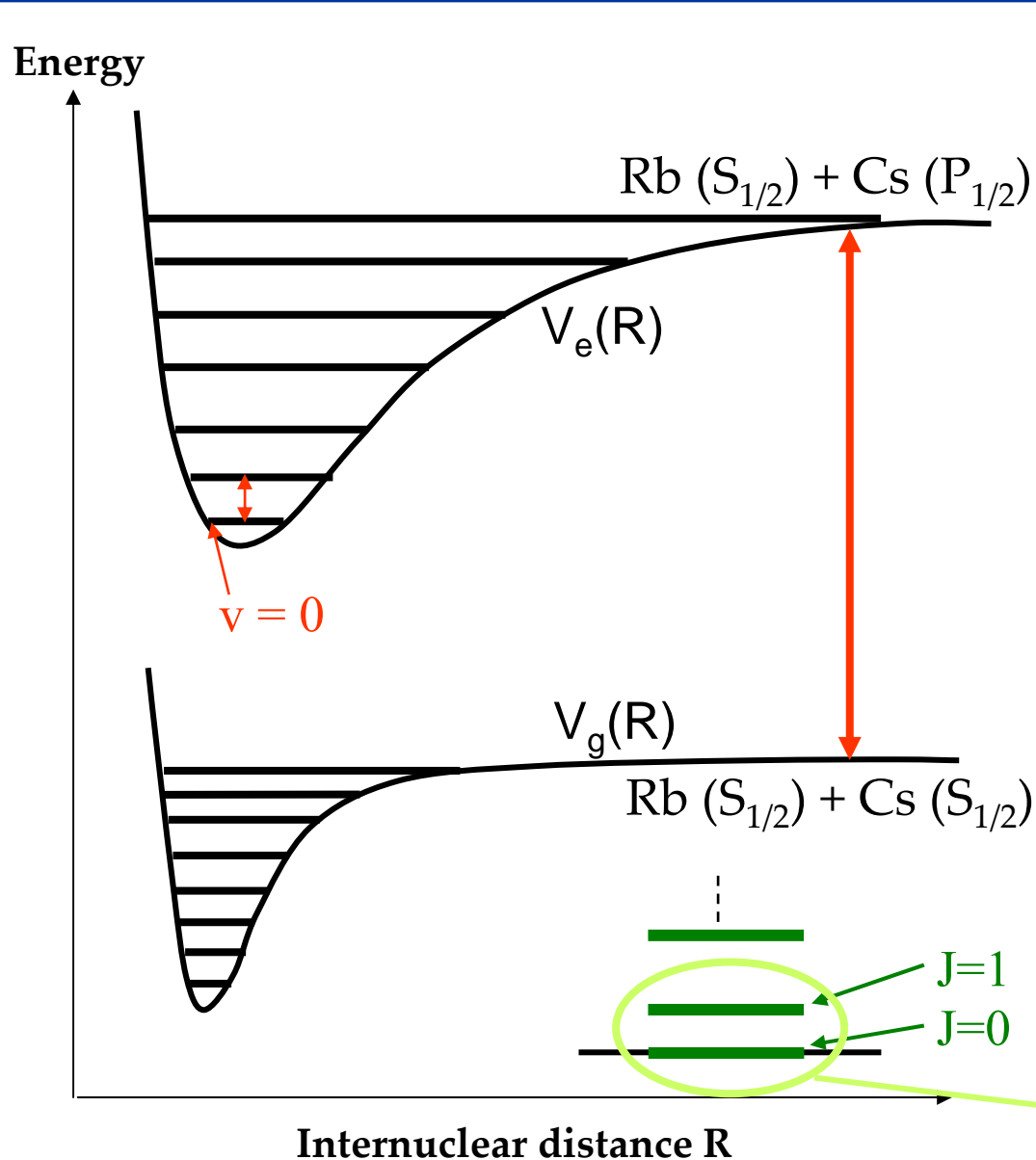
# Recap

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- Modern epoch (and then some) consistent with zero
  - Constraints are rapidly improving with no end in sight
- Early universe not so clear
  - Lack of control of systematics in astrophysical measurements necessitates the need for an 'ultimate' check

**OH Mega-masers allow interrogation of the early universe AND have an 'ultimate' check for systematics**

# Molecular structure: What is a molecule?



- Electronic potentials  
 $\sim 300$  THz ( $\sim 1.5$  eV)

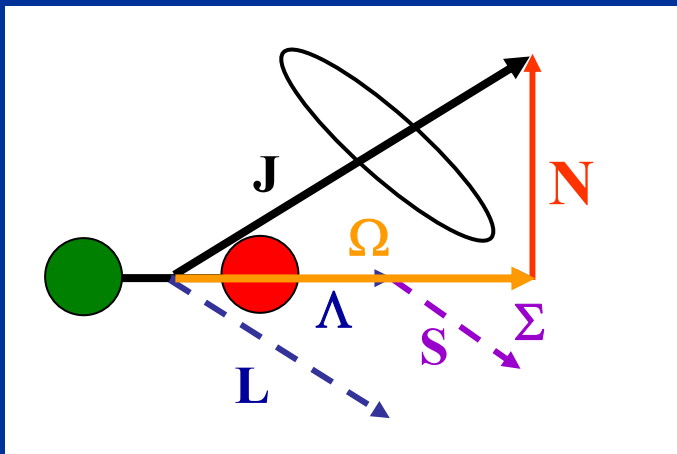
- Vibrational levels  
 $\sim 0.1 - 1$  THz

- Rotational levels  
 $\sim 0.1 - 1$  GHz

**Two levels for qubit**

# PA Primer: Electronic state labeling

- Heteronuclear diatomic molecules possess only axial symmetry
  - different good quantum numbers than for atoms

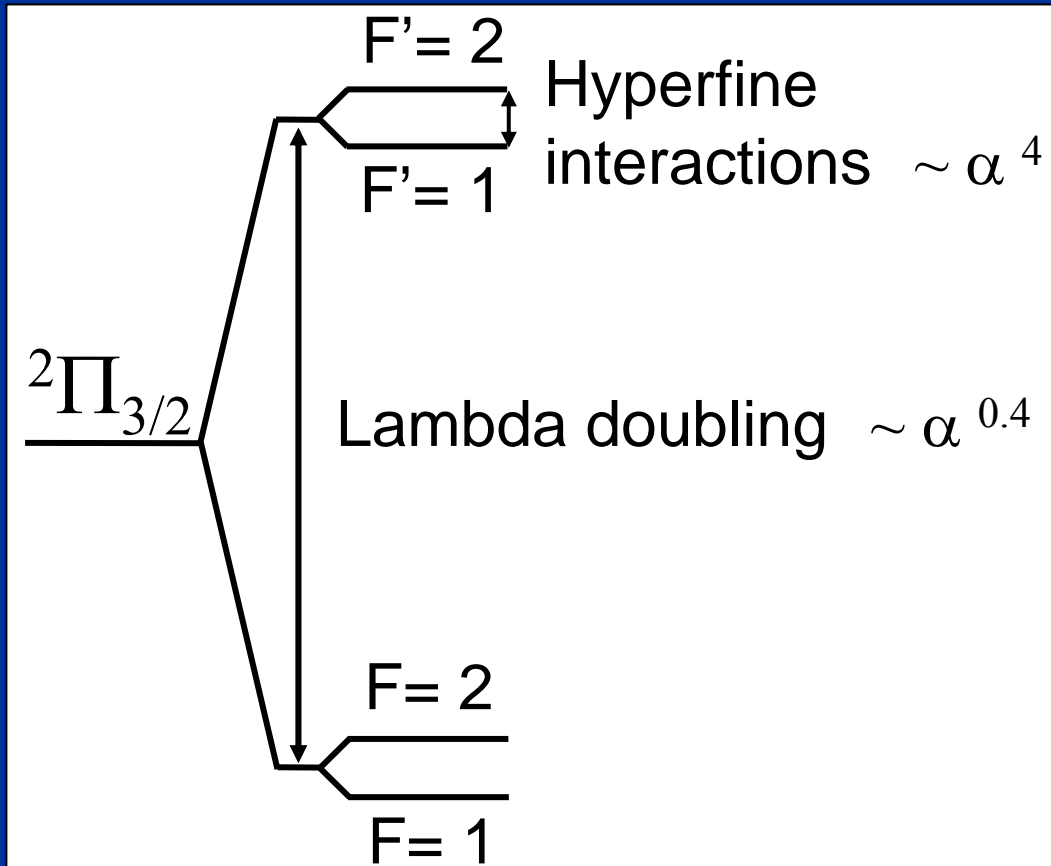


- $\Omega = |\Lambda + \Sigma|$
- $J = \Omega + N$

- Electronic potentials are labeled as  $^{2\Sigma+1}\Lambda_{\Omega}$ 
  - $\Sigma, \Pi, \Delta, \dots$  states for  $\Lambda = 0, 1, 2, \dots$   
(i.e.,  $^3\Sigma_1$  state has  $\Lambda=0, \Sigma=1, \Omega=1$ )
- Good quantum #'s are  $\Lambda, \Sigma, \Omega, J, m_J$  (or just  $\Omega, J, m_J$ )



# Using OH transitions to constrain $\alpha$



OH megamasers

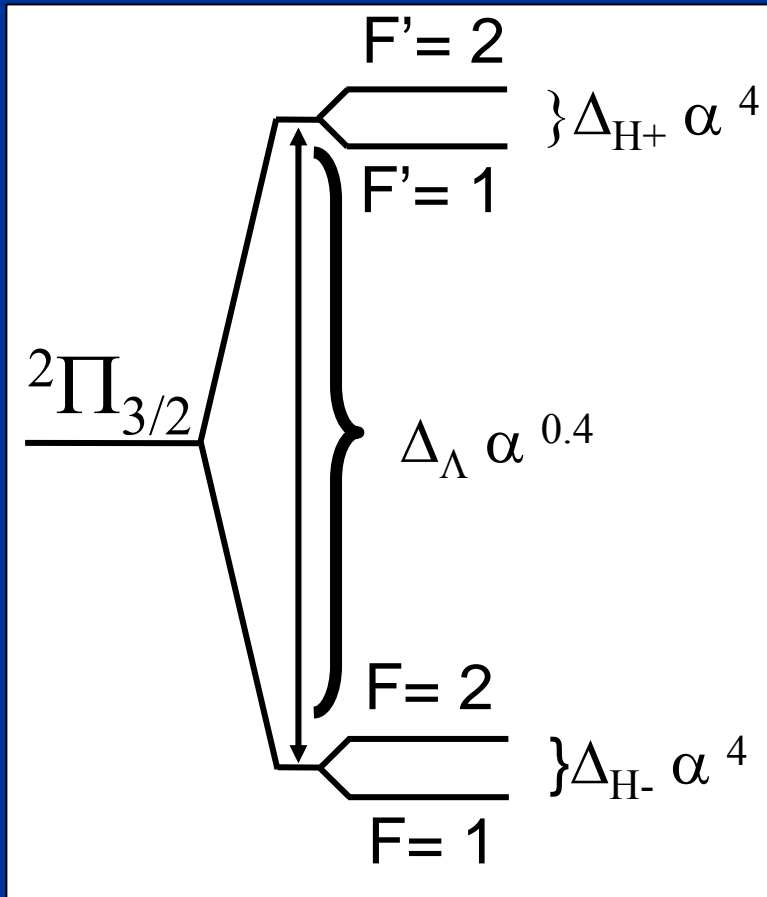


Darling, Phys. Rev. Lett **91**, 011301 (2003).  
Chengalur *et al.*, Phys. Rev. Lett. **91**, 241302 (2003).  
Kanekar *et al.*, Phys. Rev. Lett. **93**, 051302 (2004).

Allows for measurements of multiple transitions from the same gas cloud  
(Doppler shifts constrained and self check on systematics from closure)  
Previous uncertainty in laboratory based experiments is 100-200 Hz, which  
leads to  $\Delta\alpha/\alpha \sim 10^{-5}$

ter Meulen & Dymanus, *Astrophys. J.* **172**, L21(1972).

# Using OH transitions to constrain $\alpha$



Astronomical observation:

$$\omega_{11} = \Delta_\Lambda \alpha_o^{0.4} - 1/2(\Delta_{H+} - \Delta_{H-}) \alpha_o^4 + RS_{11}$$

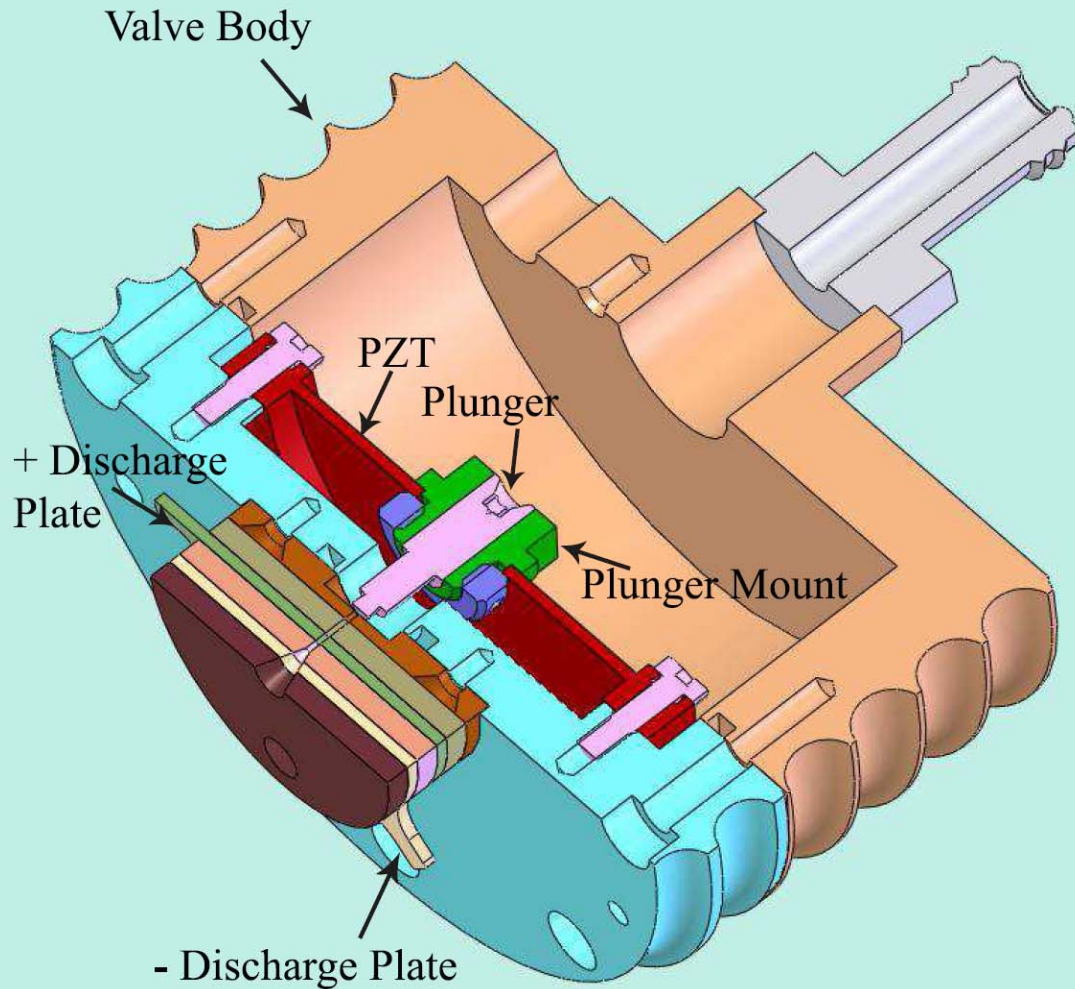
$$\omega_{22} = \Delta_\Lambda \alpha_o^{0.4} + 1/2(\Delta_{H+} - \Delta_{H-}) \alpha_o^4 + RS_{22}$$

Lab measurement:

$$\omega_{11} = \Delta_\Lambda (\Delta\alpha + \alpha_o)^{0.4} - 1/2(\Delta_{H+} - \Delta_{H-}) (\Delta\alpha + \alpha_o)^4$$

$$\omega_{22} = \Delta_\Lambda (\Delta\alpha + \alpha_o)^{0.4} + 1/2(\Delta_{H+} - \Delta_{H-}) (\Delta\alpha + \alpha_o)^4$$

# First make the molecules : Sourcery

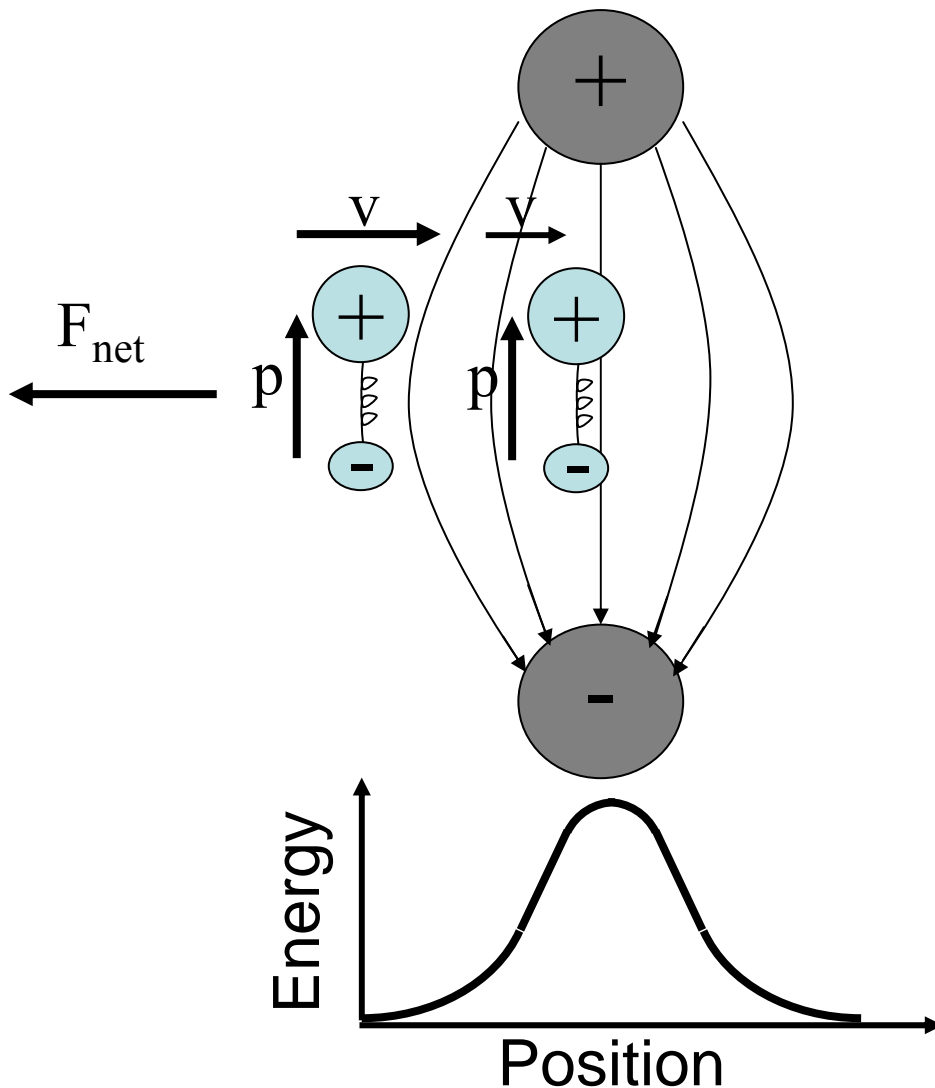


Supersonic  
Expansion:

-Cold molecules  
moving at a few 100  
m/s.

# Stark deceleration

Second step: slow the molecules in to the rest frame of the lab



**Conservative process, no cooling**

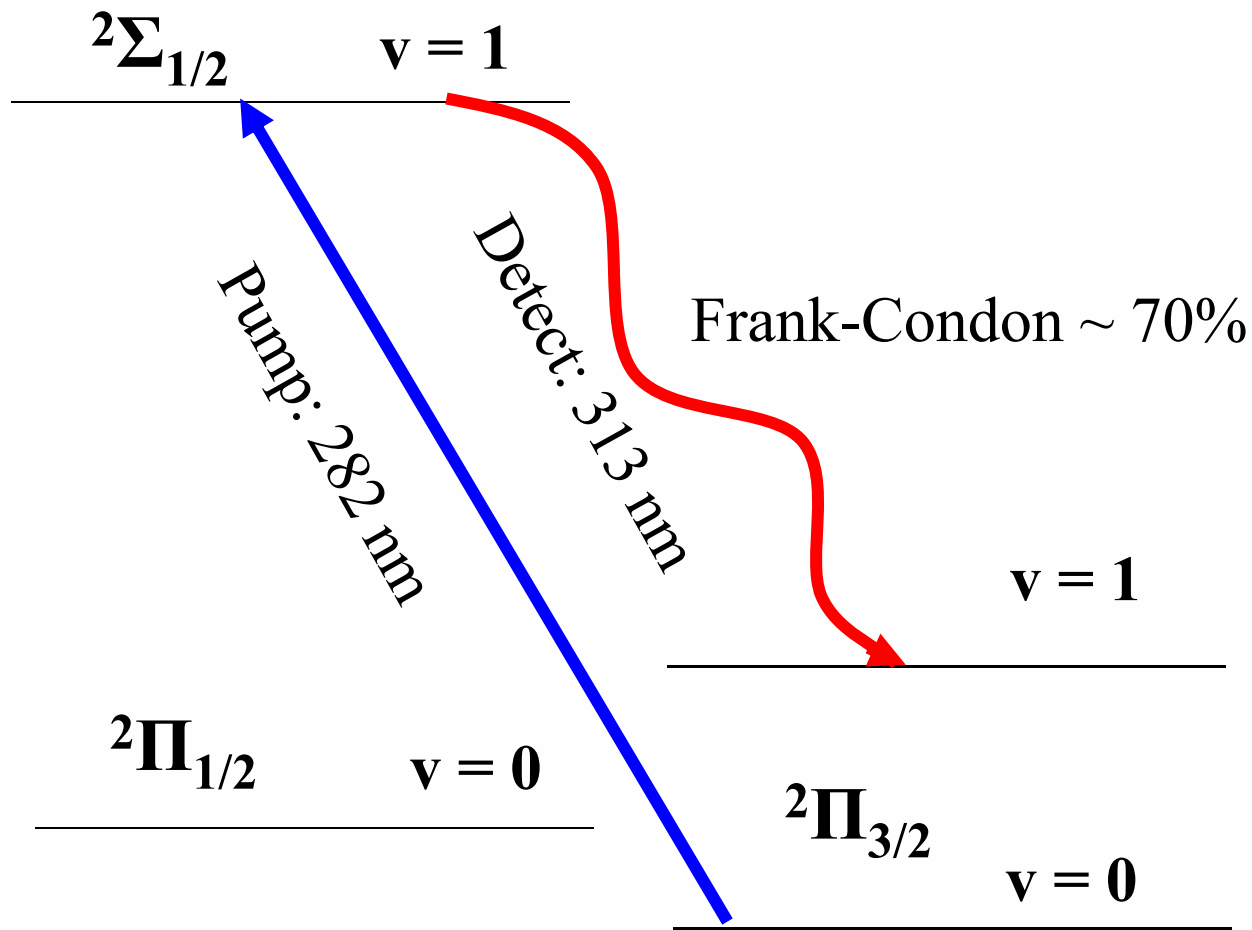
**Phase space selection**

**Phase space area linked to the deceleration angle ( $\phi_0$ )**

**Phase space rotation (constant density)**

**Resembles a pendulum driven by a constant torque**

# Basic energy structure of OH



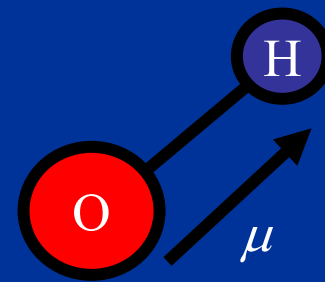
## Basics:

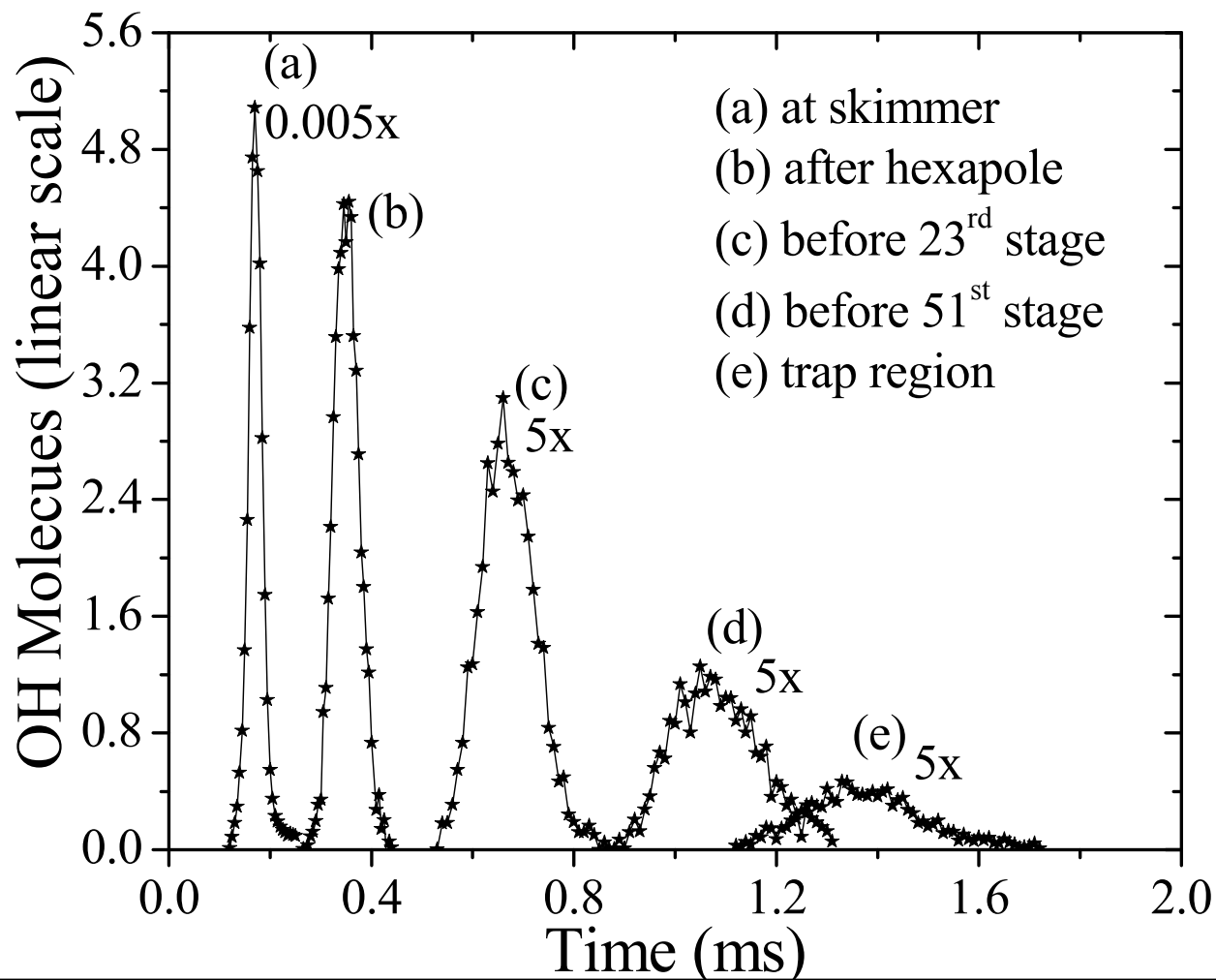
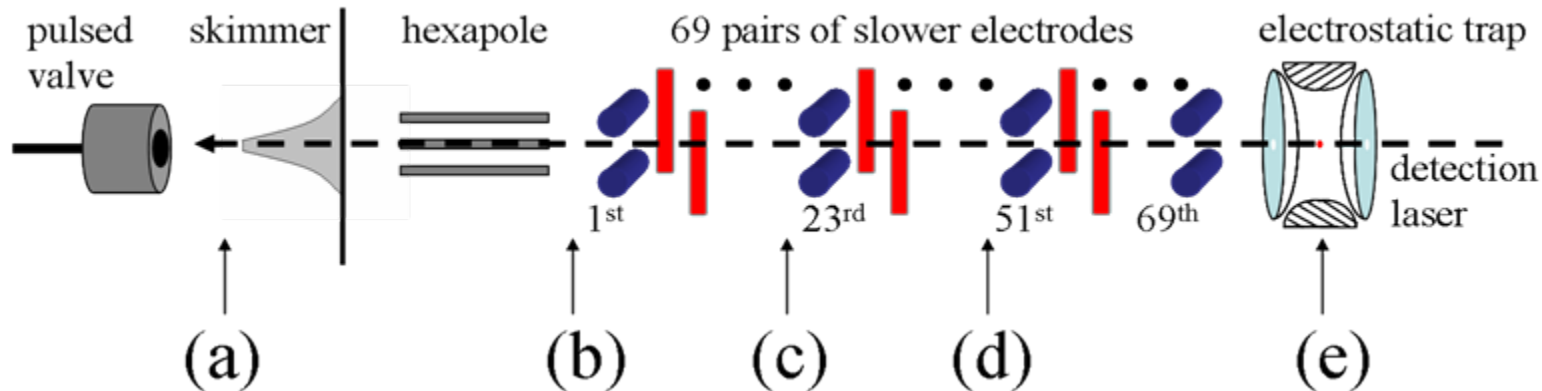
-Discharge H<sub>2</sub>O in Xe

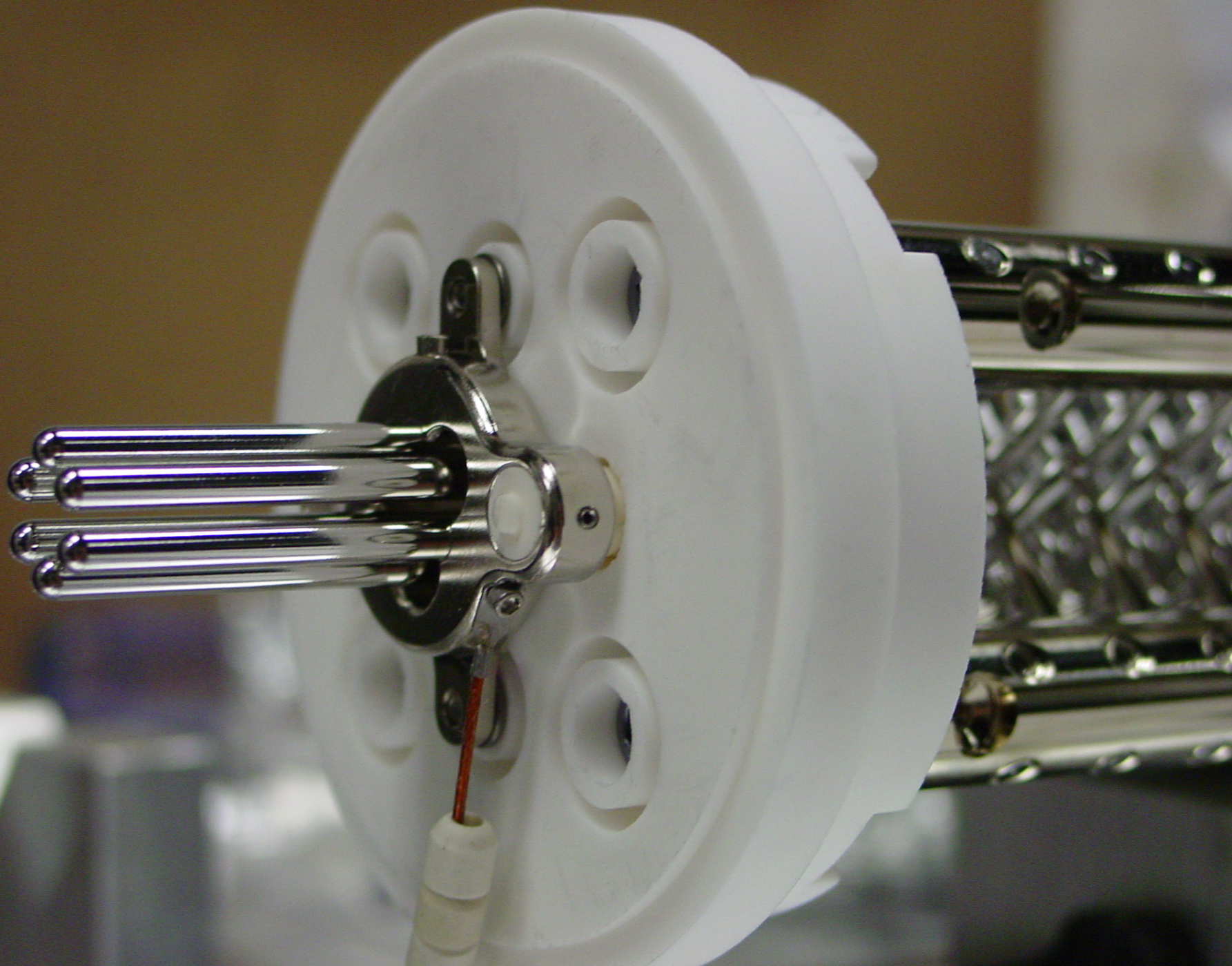
-Ground State  $2\Pi_{3/2}$

- $\lambda$  doublet spacing  
~1.7 GHz

- $\mu = 1.67D$

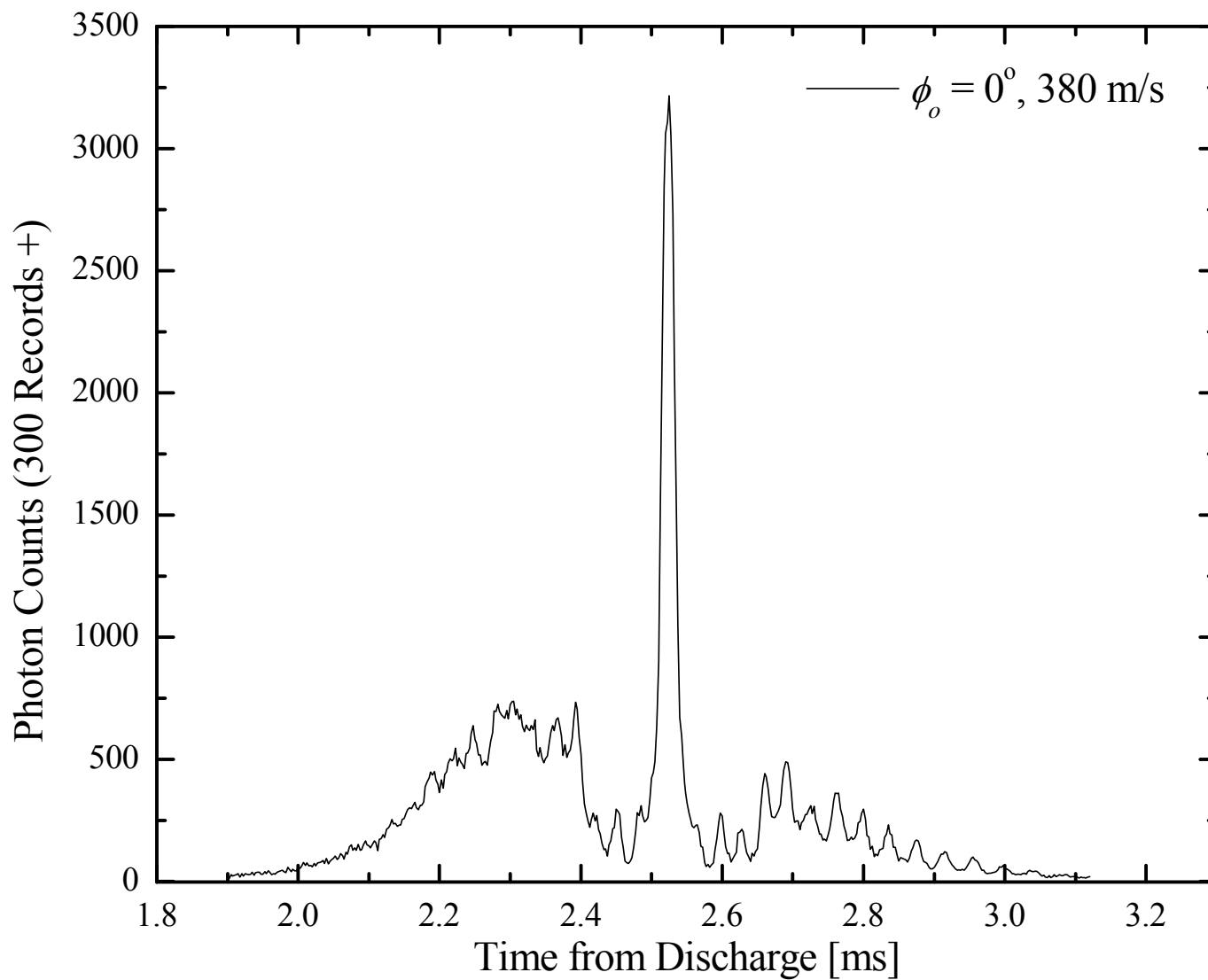


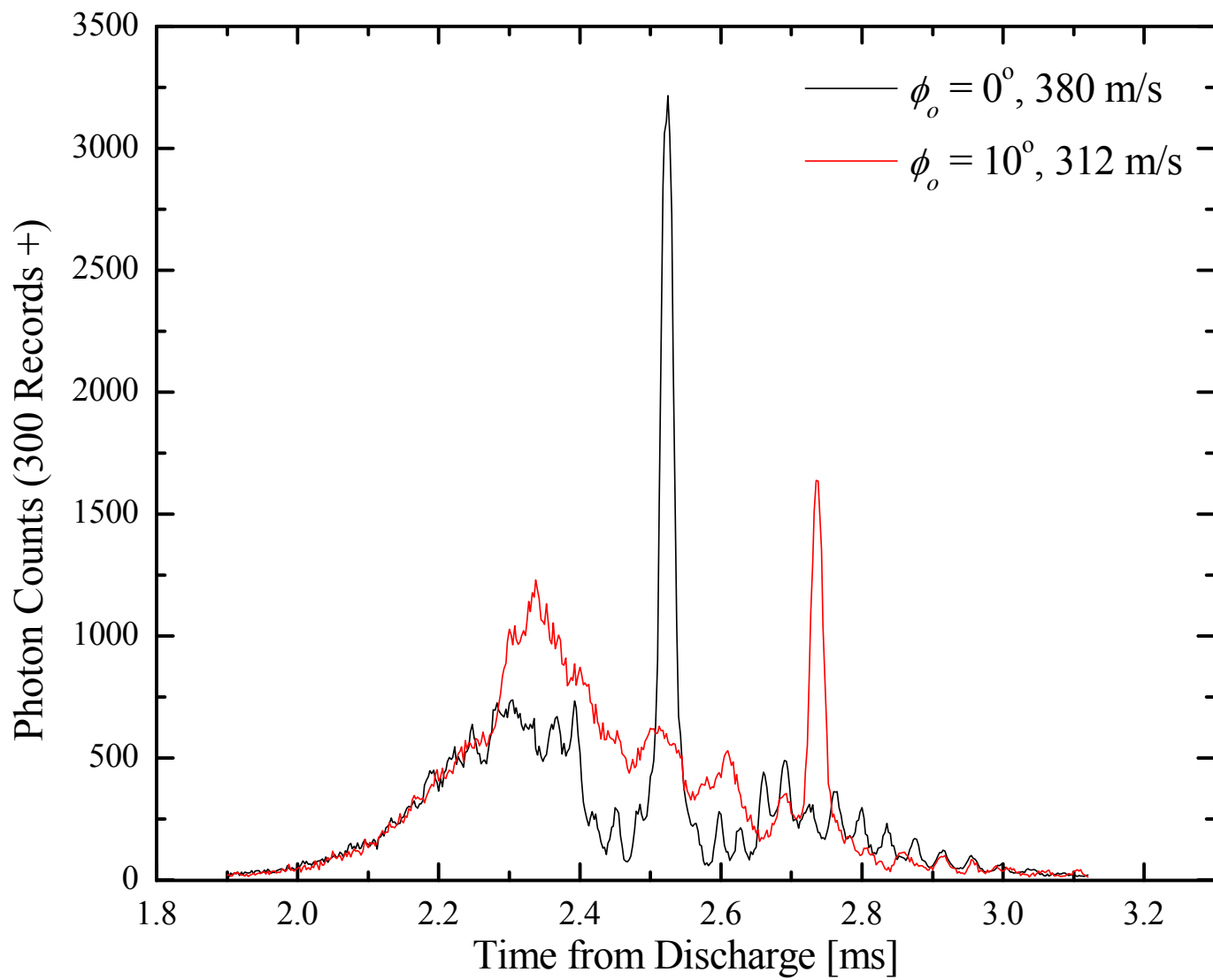


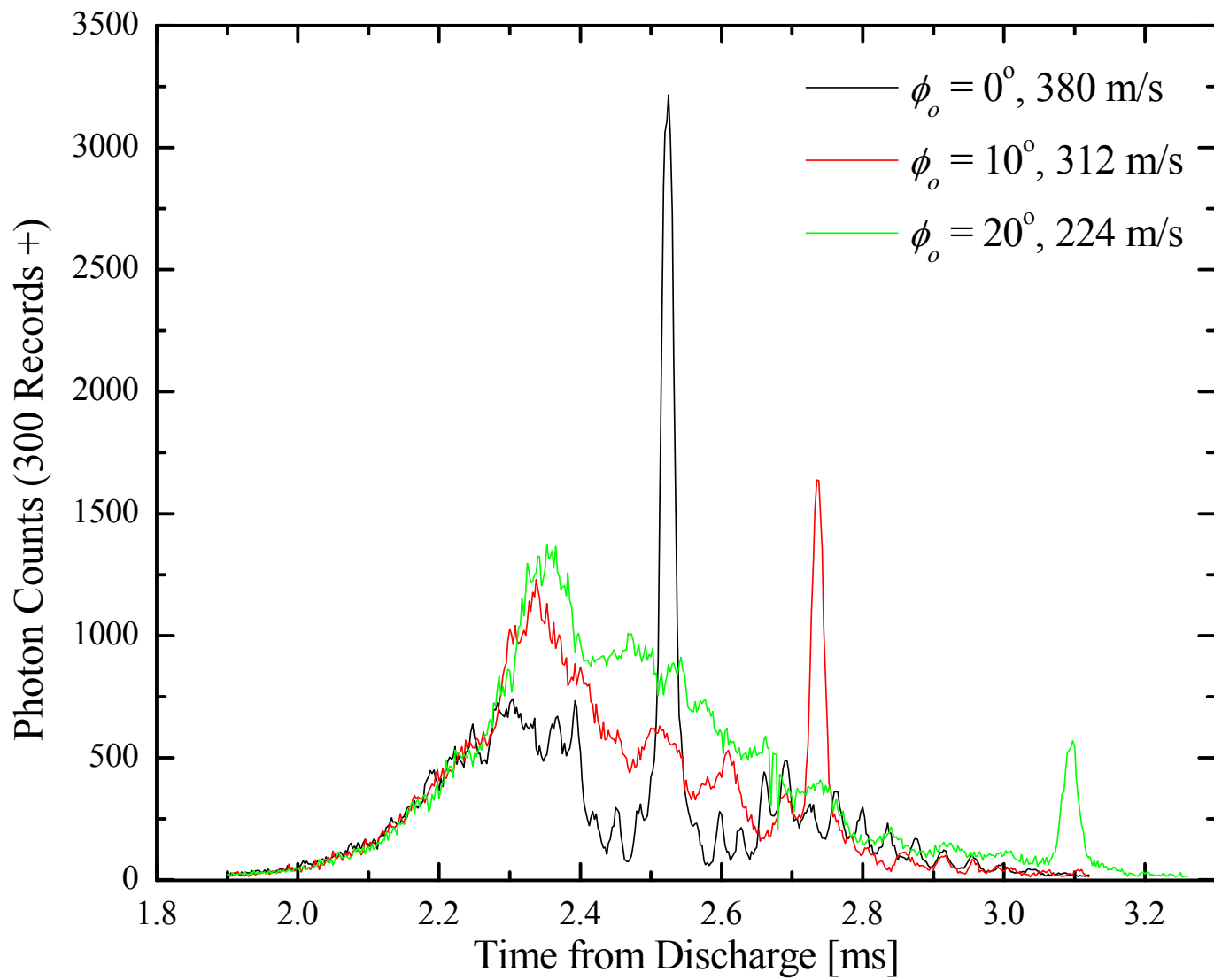




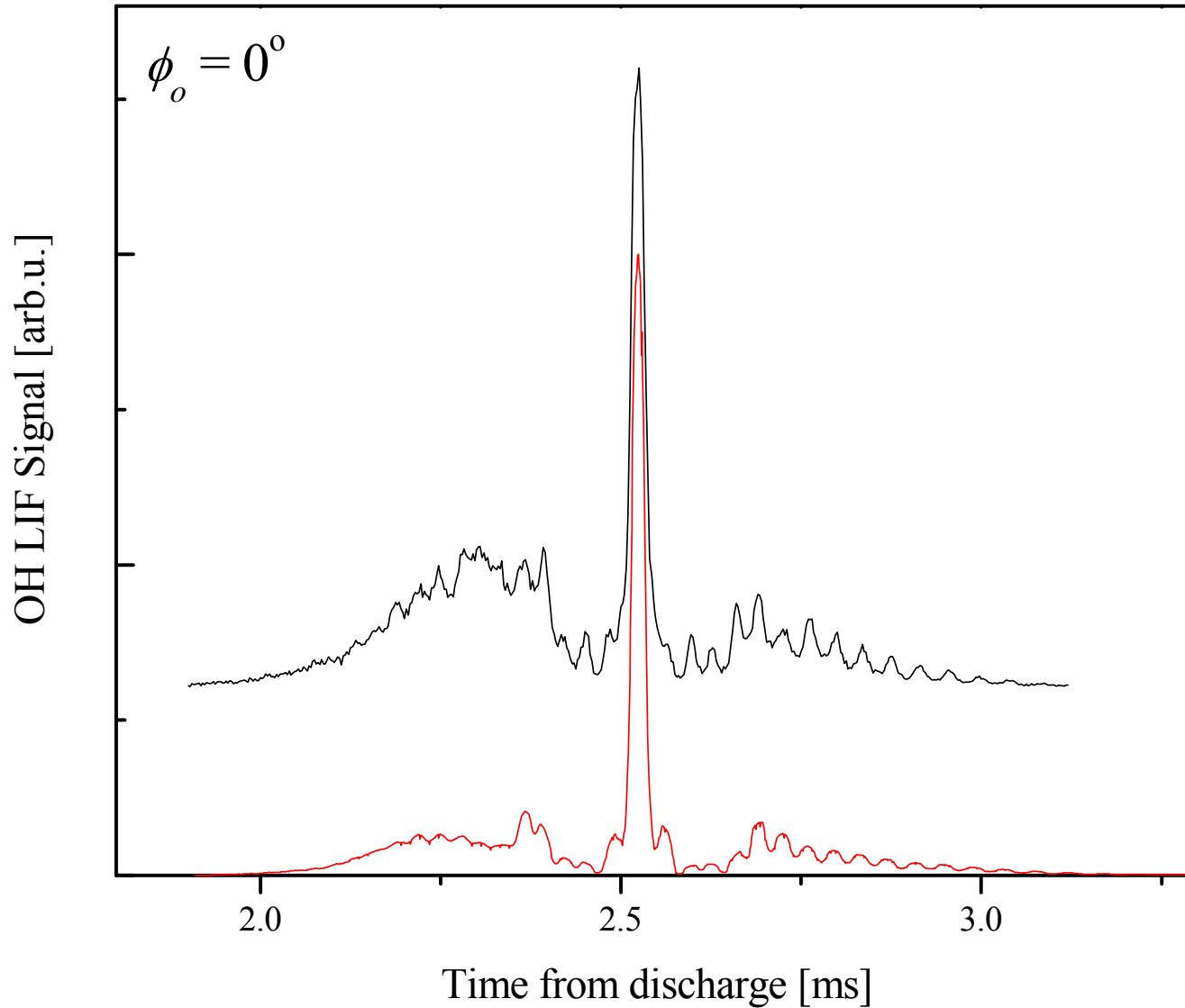






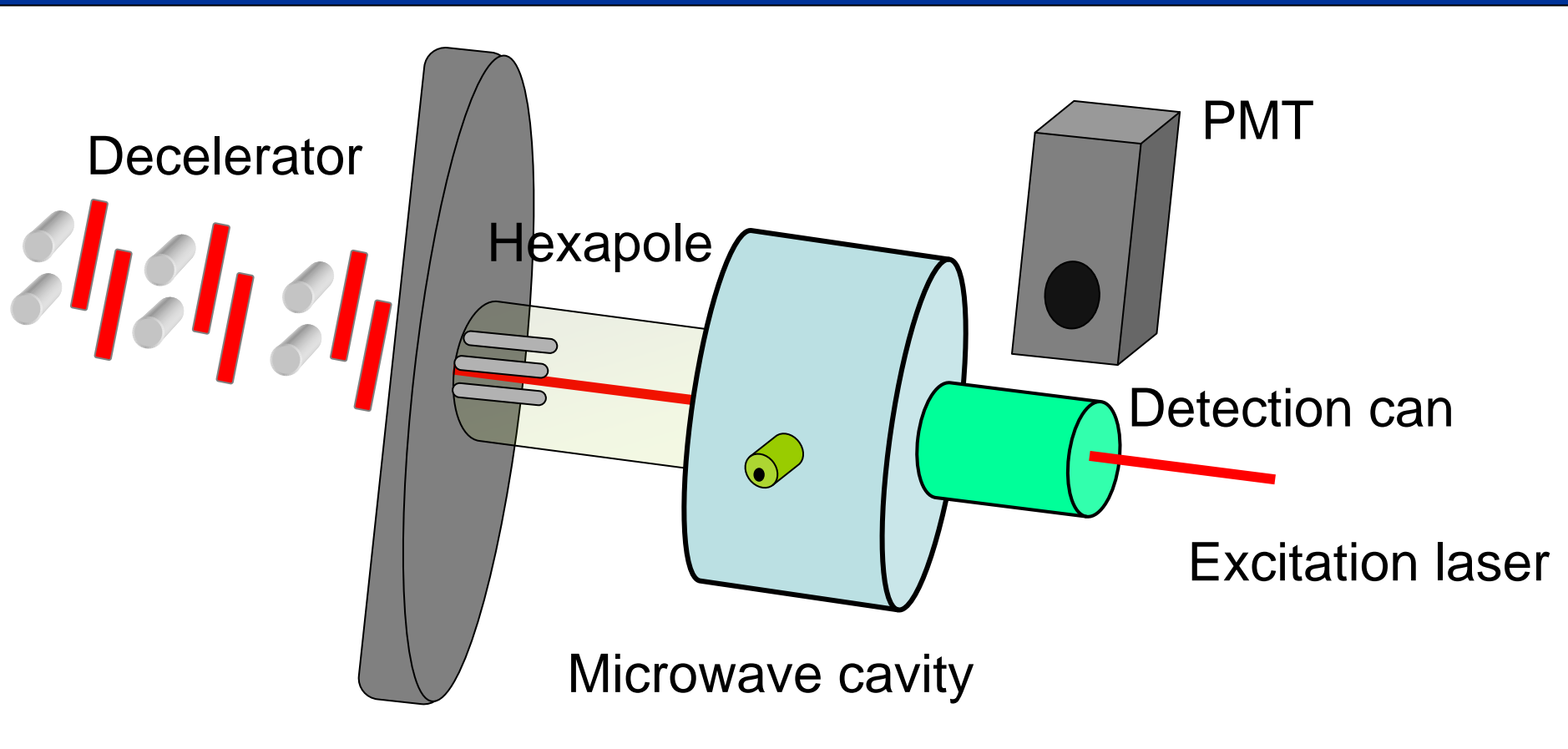


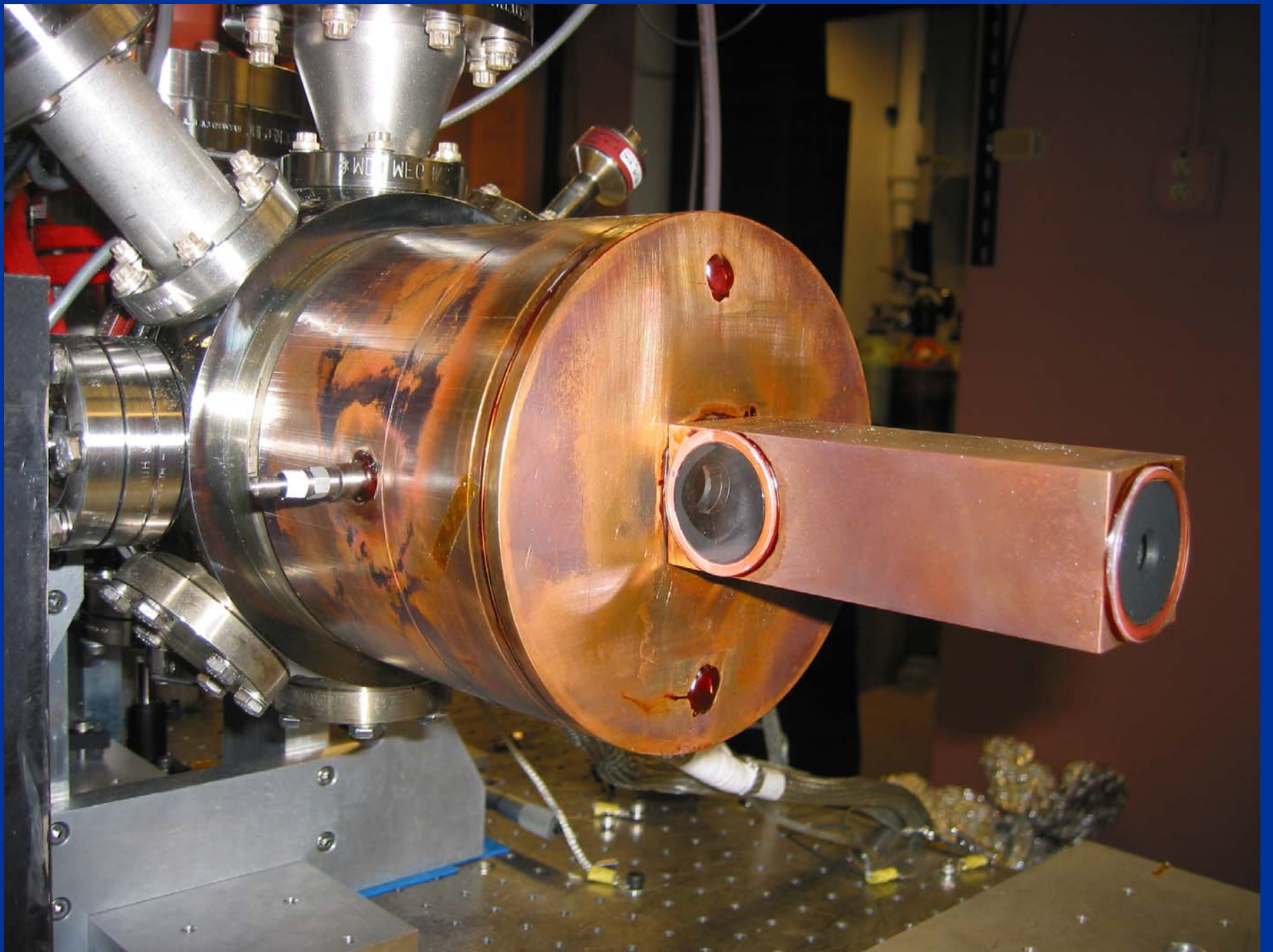
# 3D Monte Carlo Simulation Results



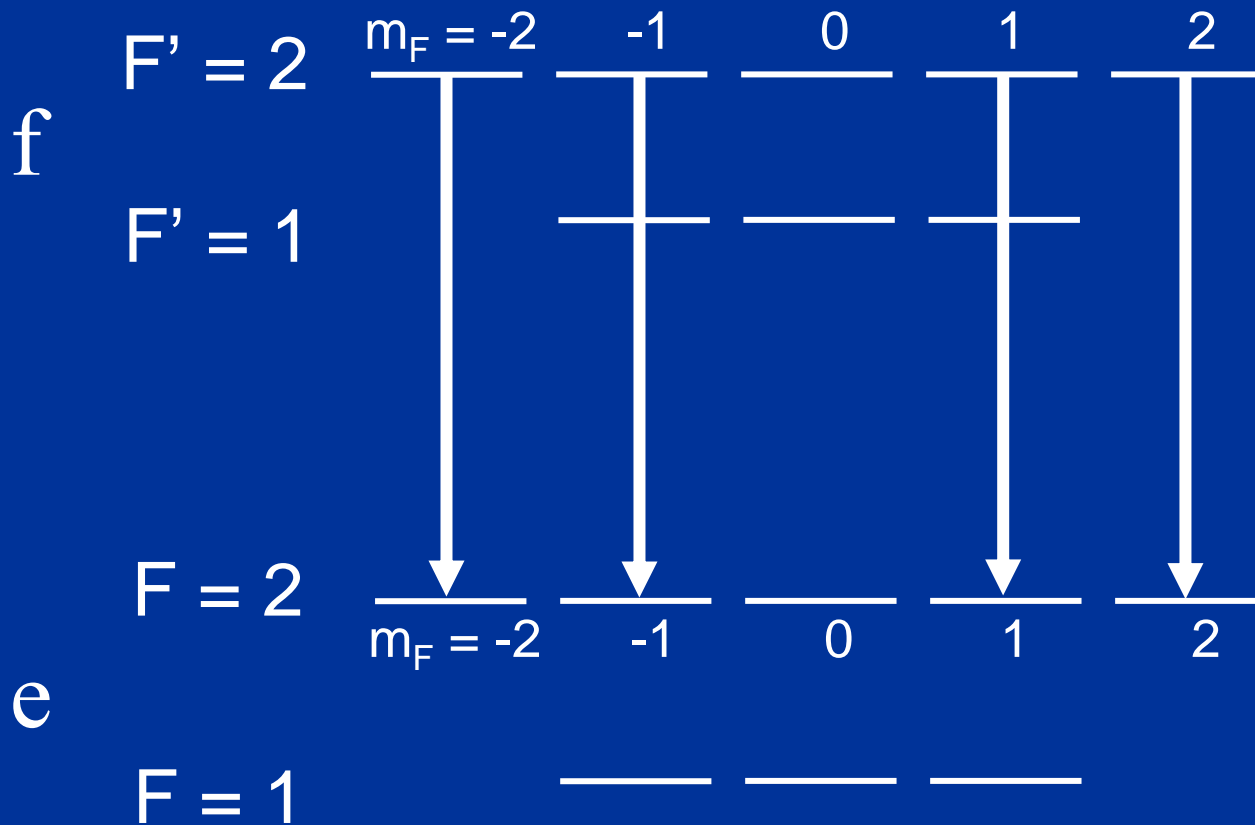
# Experimental Set-up

- All metal detection area
- Slowed OH beam





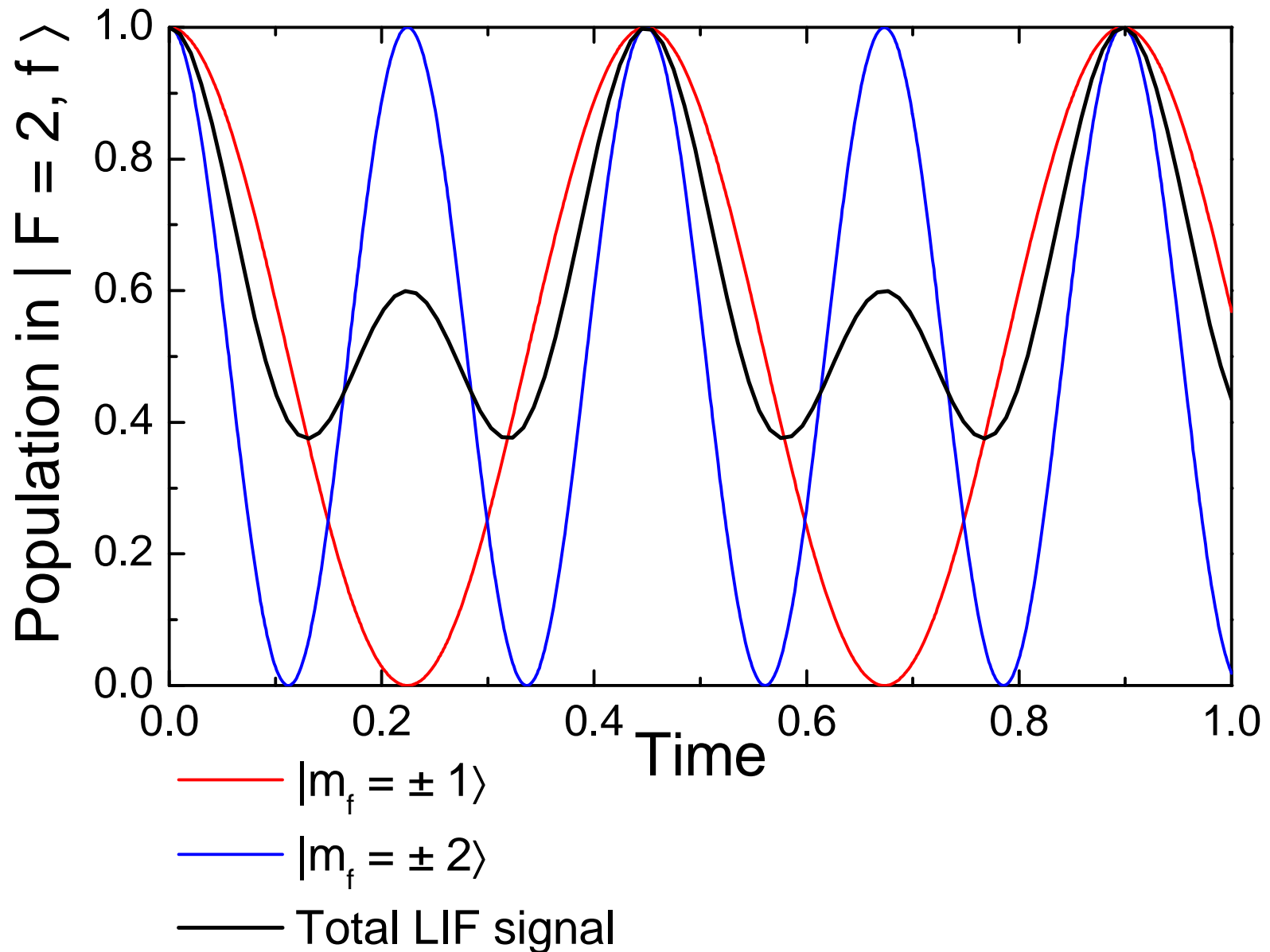
# Hyperfine structure



$$\langle \mu_e \rangle_{\pm 2} = 2 \times \langle \mu_e \rangle_{\pm 1}$$

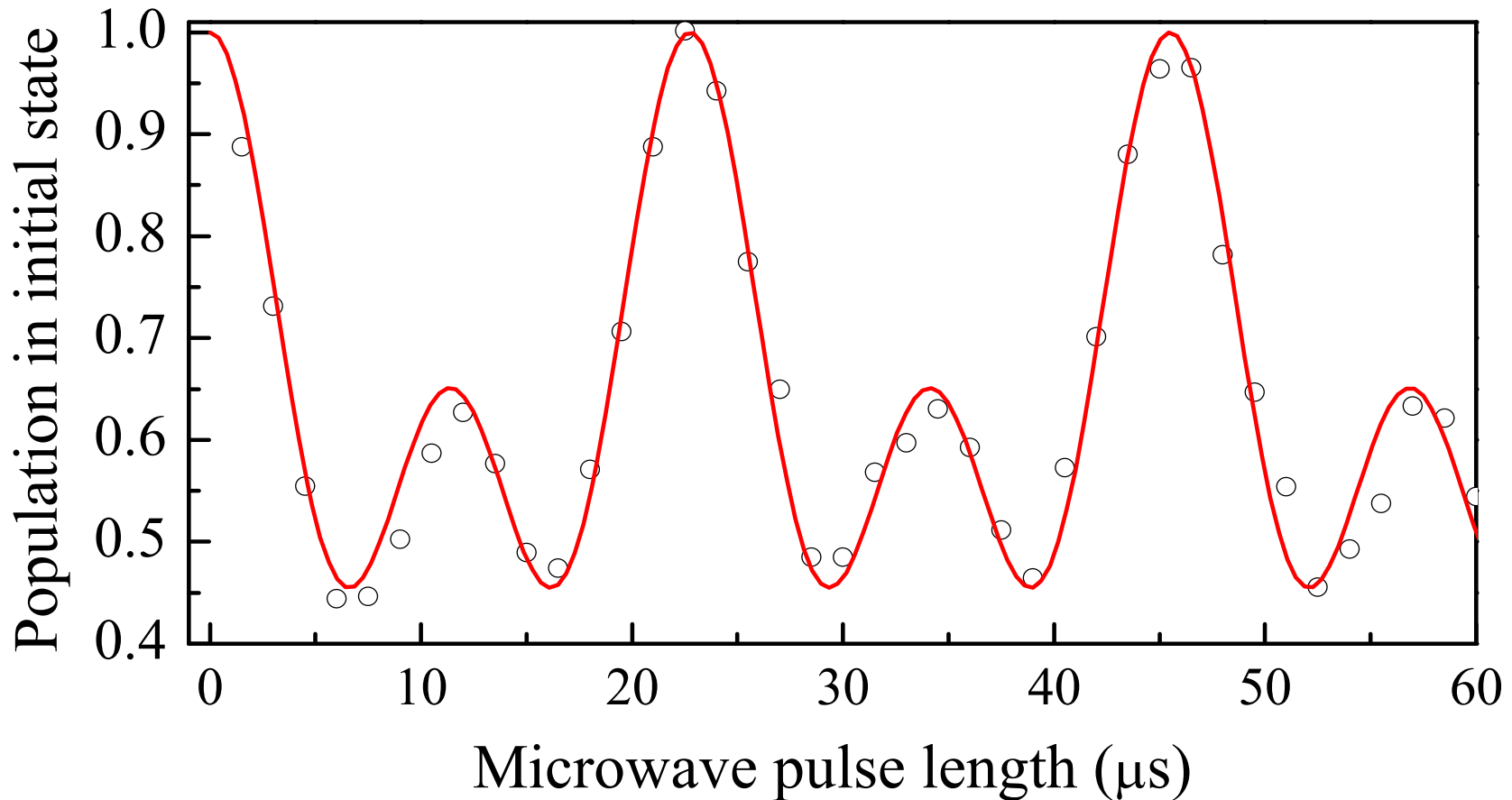
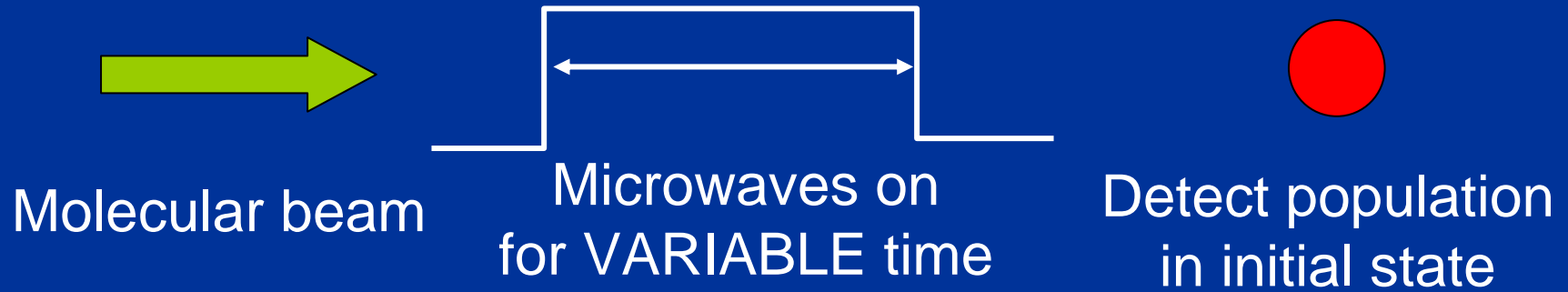
Transition dipoles are different by a factor of two.

# Double Rabi flopping

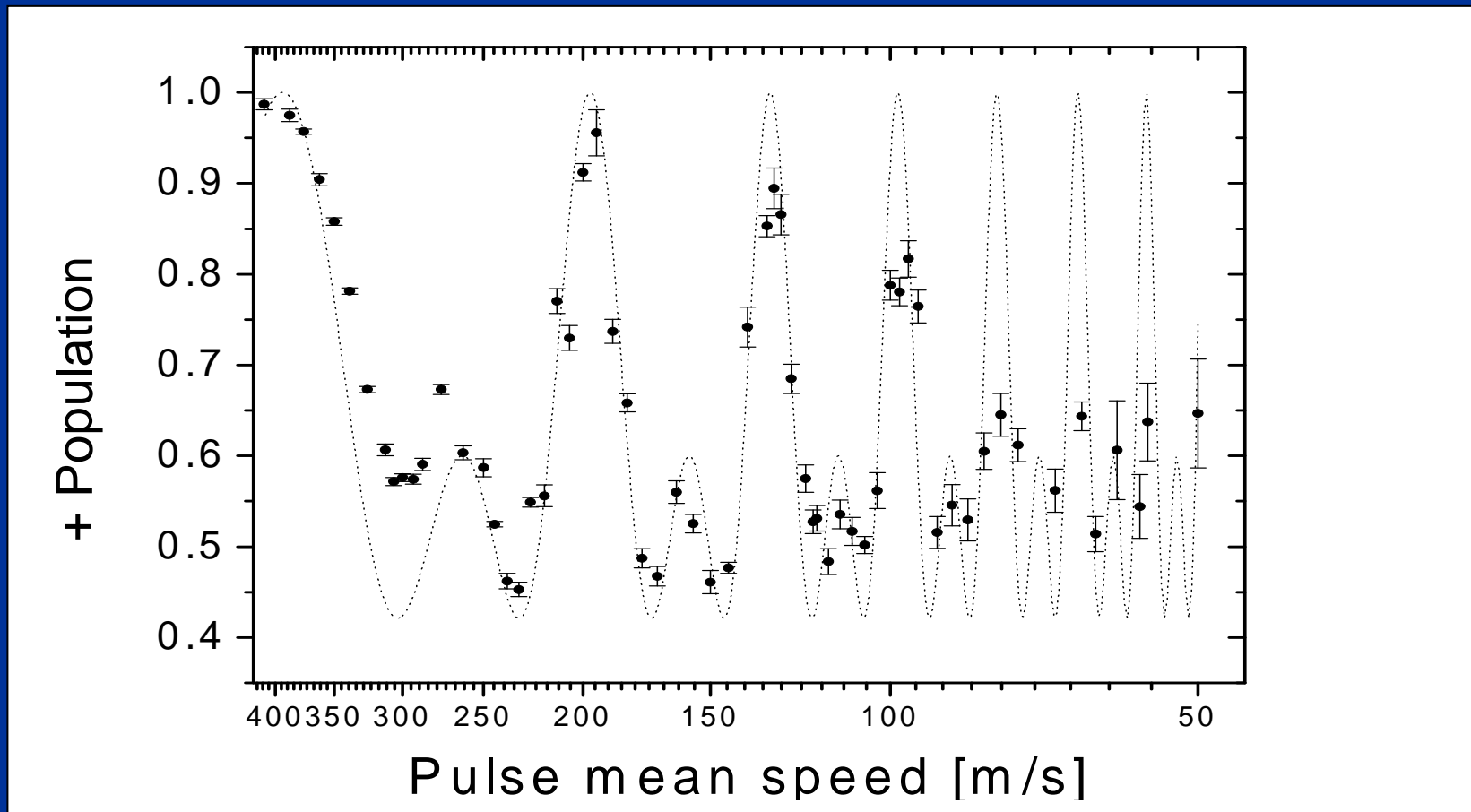
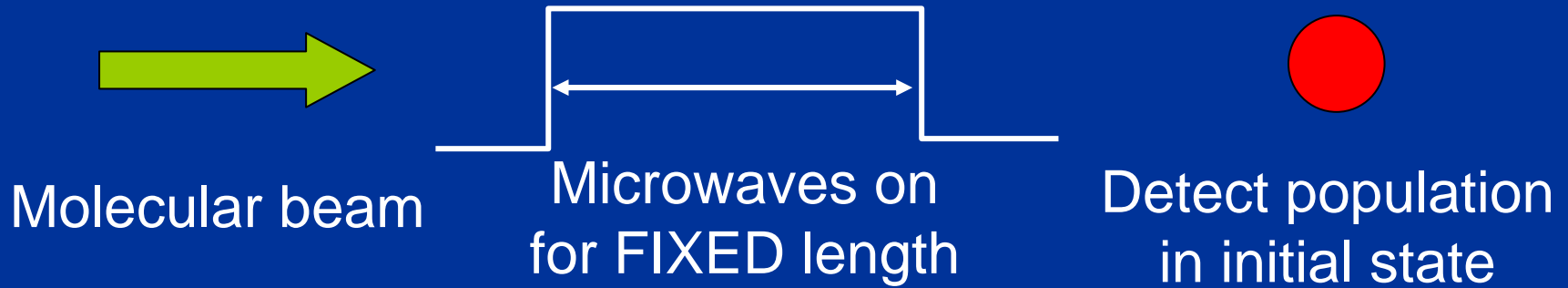




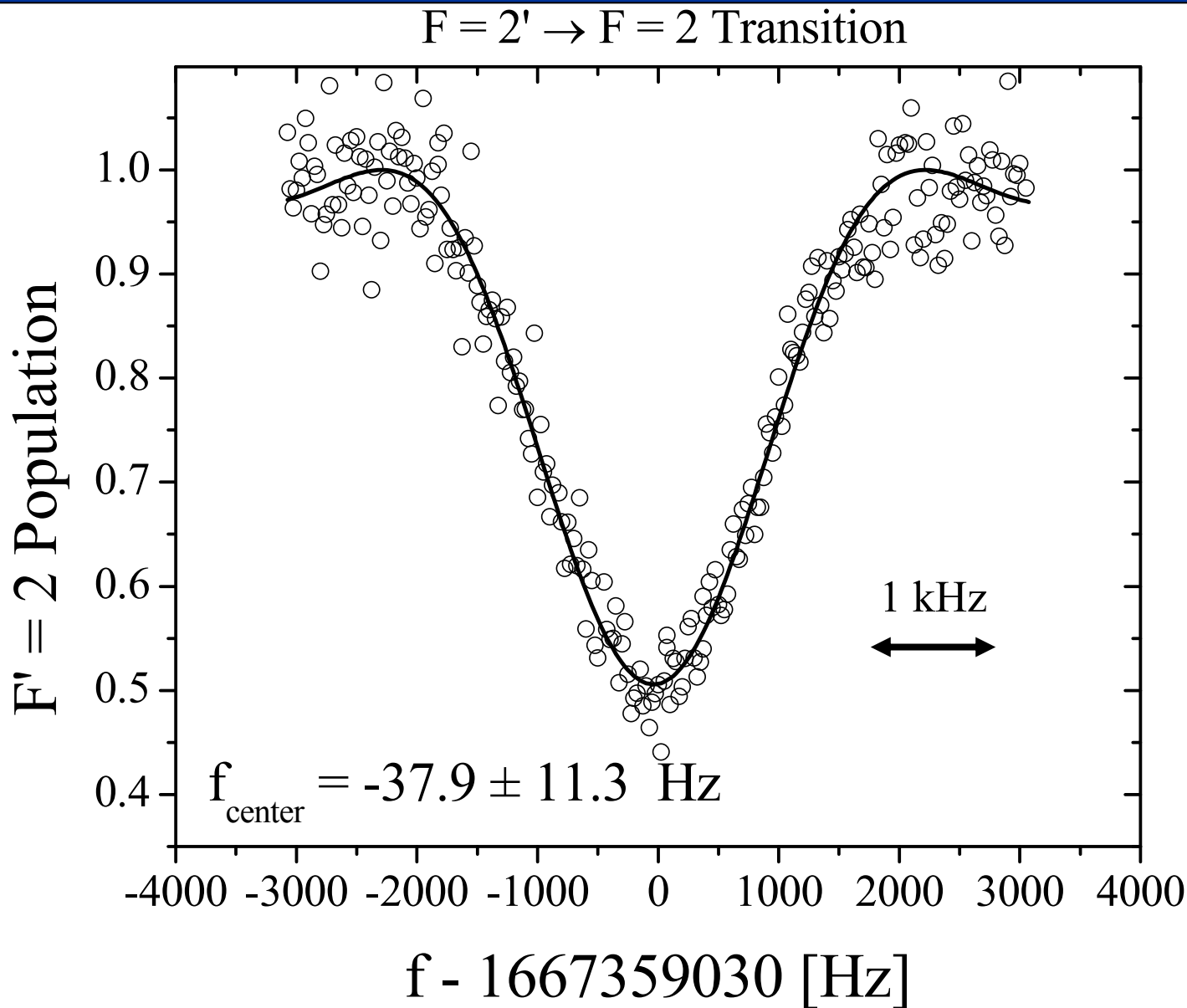
# Rabi flopping ( $F' = 2 \rightarrow F = 2$ )



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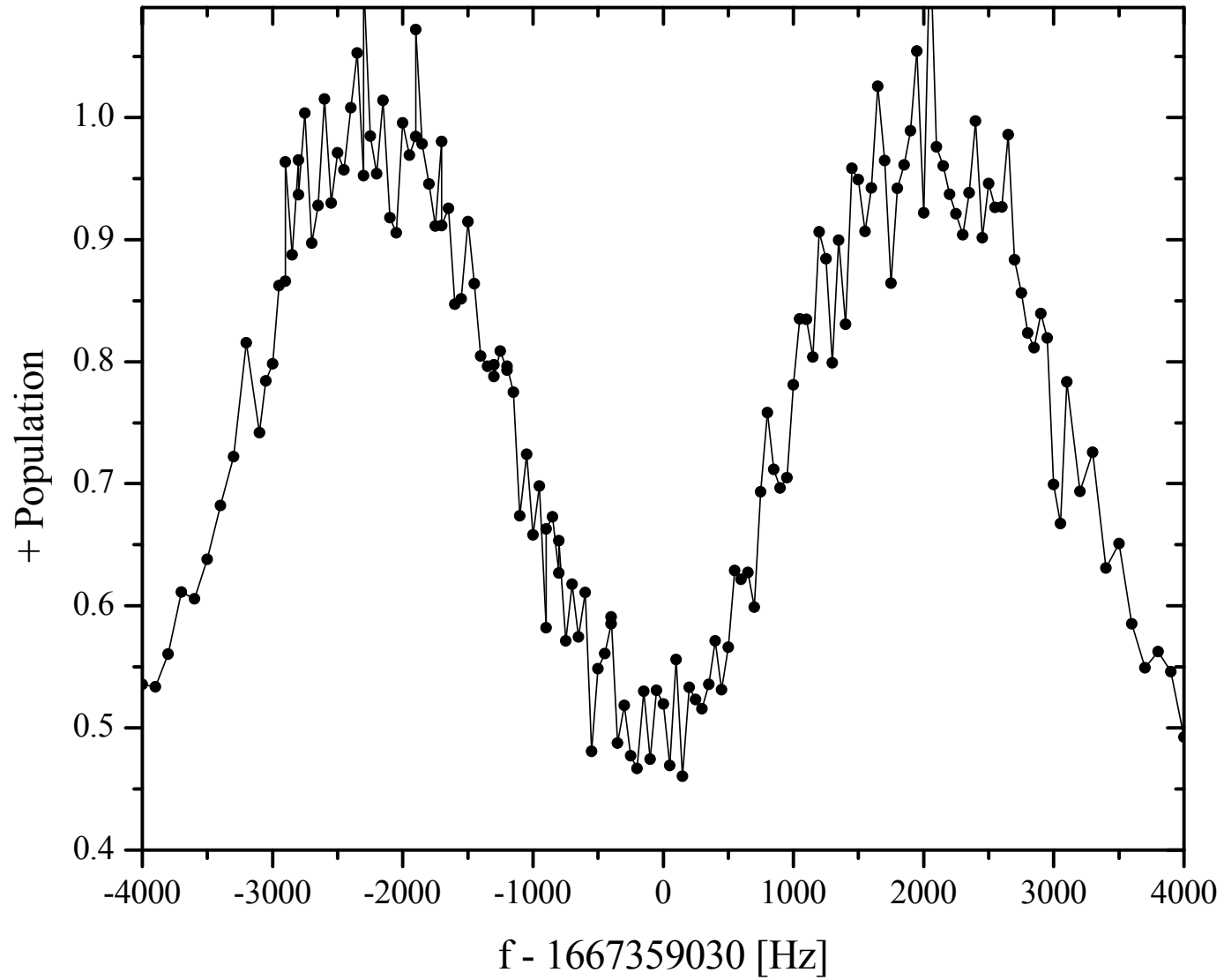
# Transition lineshape and center



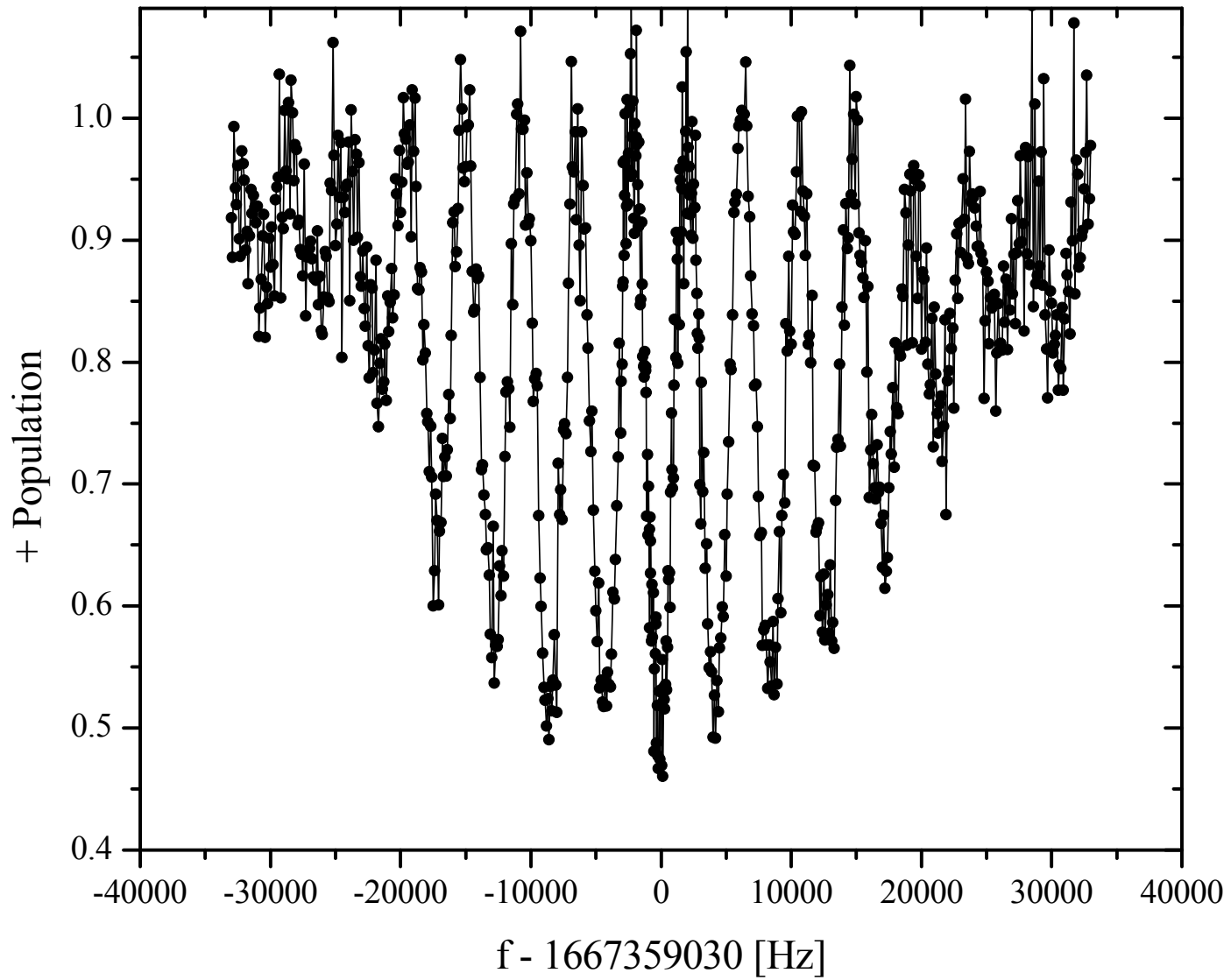
At fixed  
beam speed  
and pulse  
duration:

Vary  
detuning

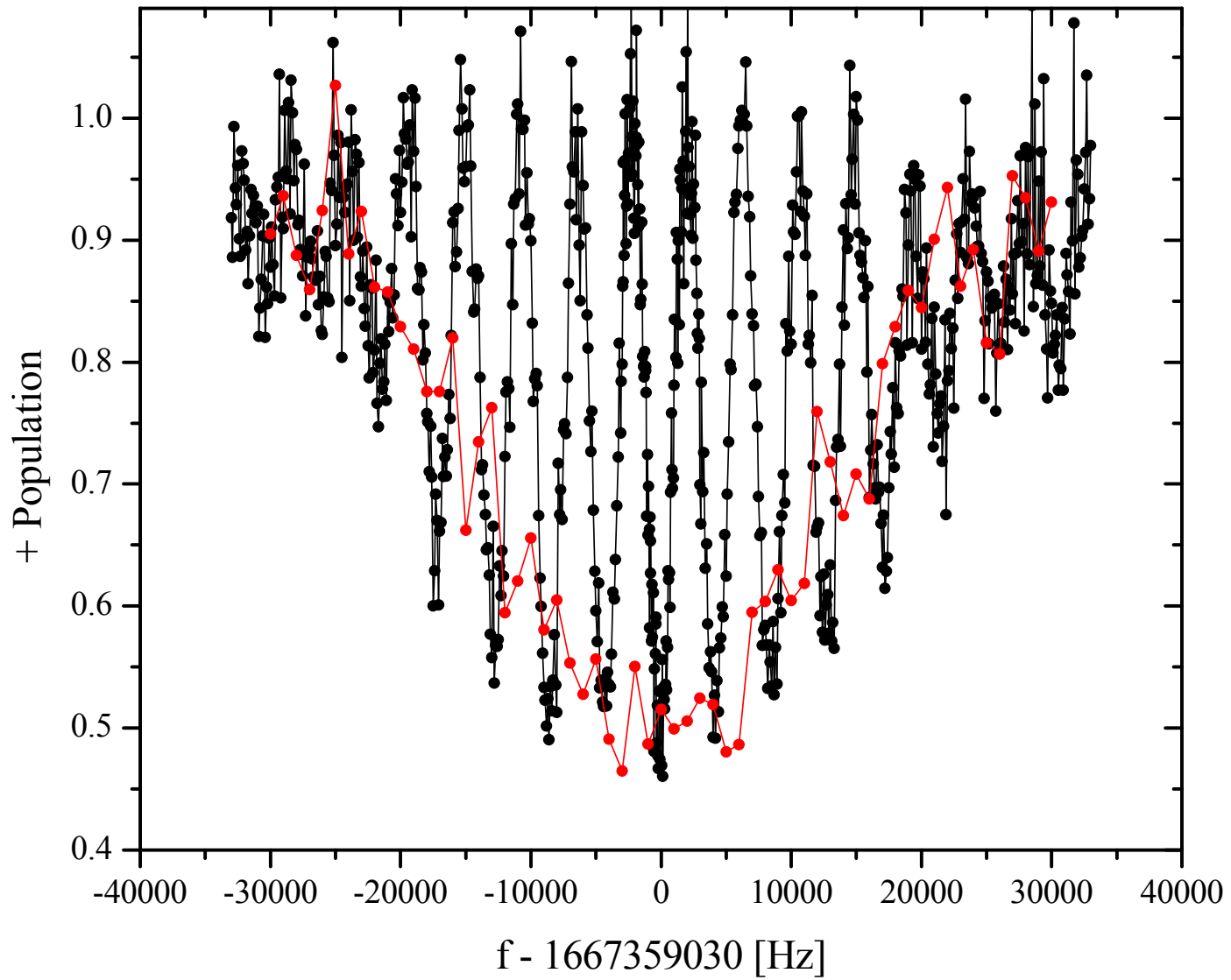
# Ramsey Spectroscopy, $2 \rightarrow 2$

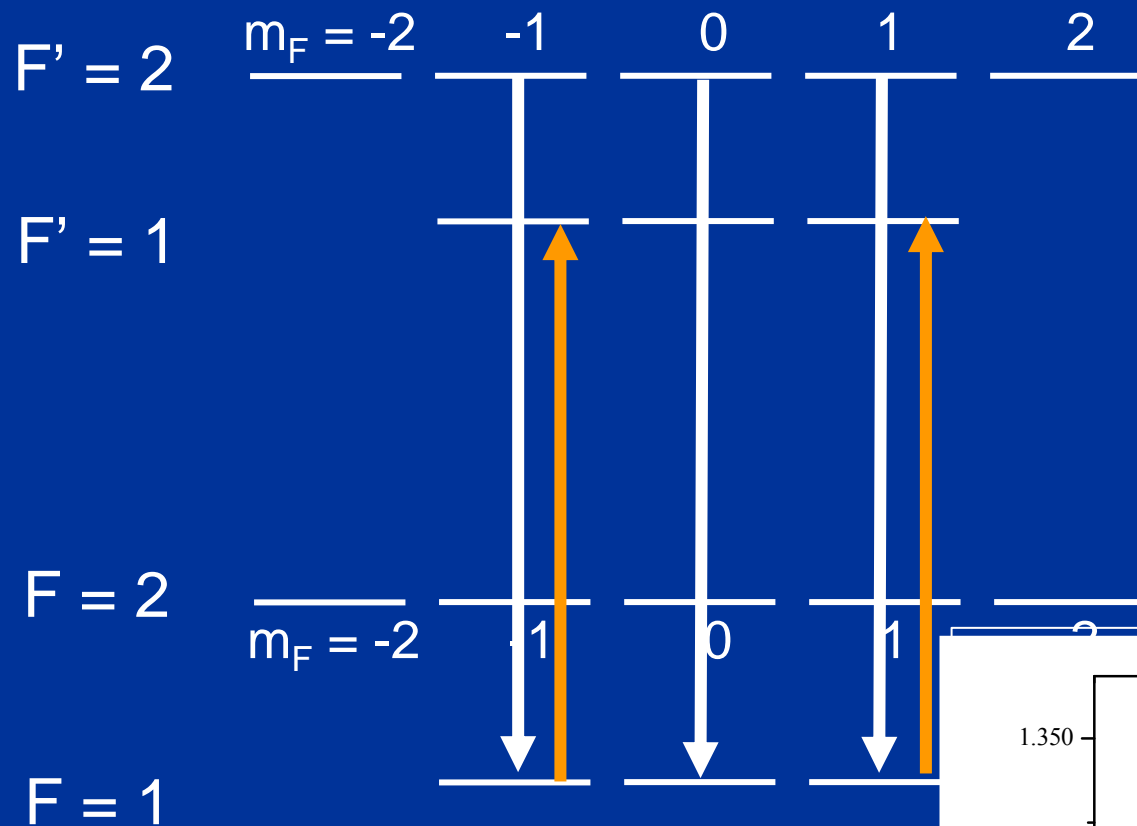


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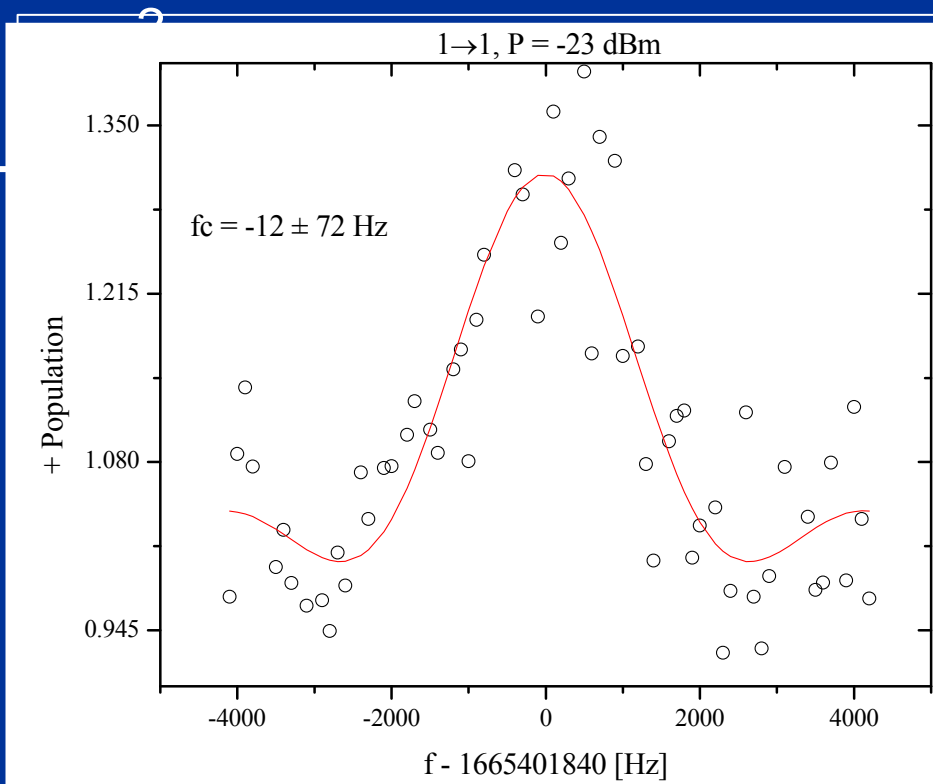


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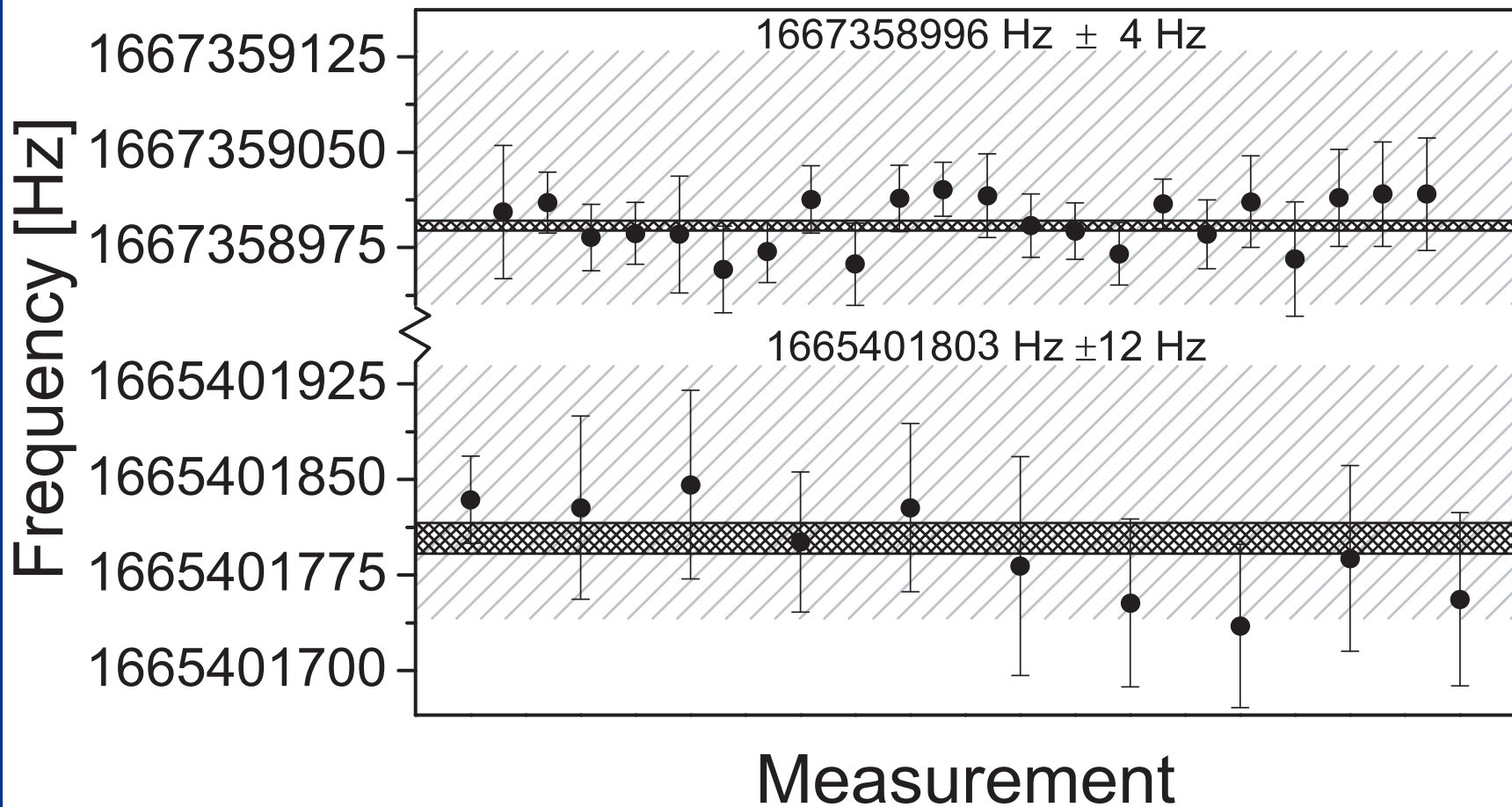




$F' = 1$  to  $F = 1$   
 Transition



# Line Center Summary





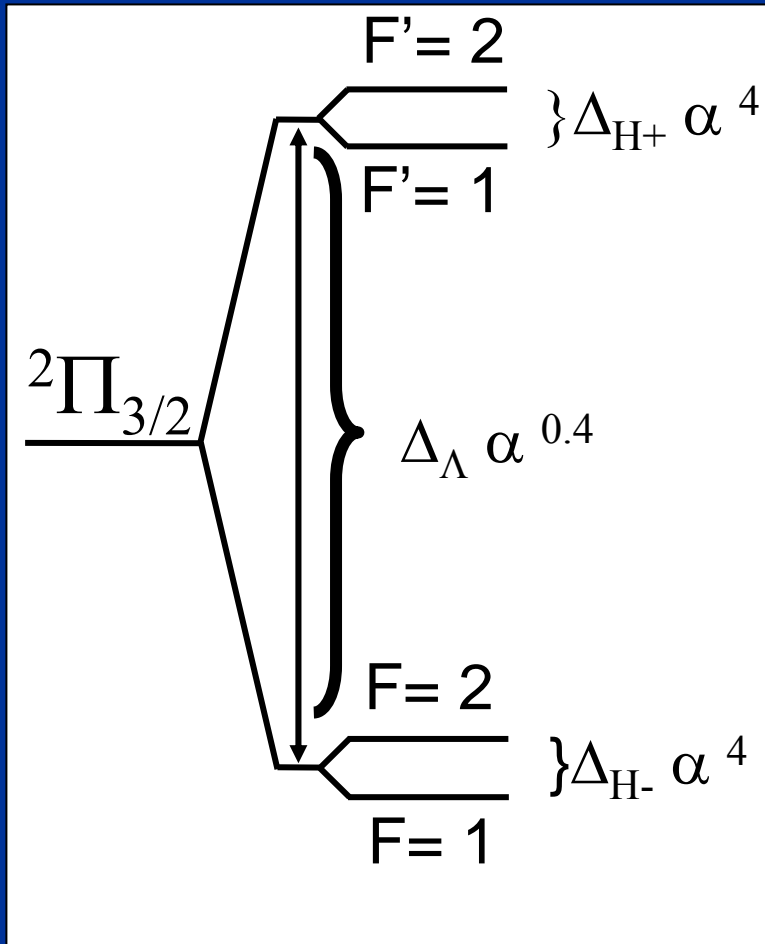
# $\Delta\alpha/\alpha?$

- This measurement will allow an order of magnitude improvement:

$$\Delta\alpha/\alpha = 1 \text{ ppm over } 10^{10} \text{ years}$$

- » Same as atomic clocks if assume time derivative is linear
- » Can probe spatial changes
- Still waiting on astrophysical result...
  - Over 100 hours of data taken on GBT, but analysis is bogged down by some terrestrial noise
  - New data set from Arecibo being analyzed (N. Kanekar)
- The future is bright
  - Because the frequency is in the cooled L-band no resolution limits yet
  - New telescopes coming on-line in the next 10 years can approach our resolution with only a few hours of data collection

# Using OH transitions to constrain $\alpha$



Astronomical observation:

$$\omega_{11} = \Delta_\Lambda \alpha_o^{0.4} - 1/2(\Delta_{H+} - \Delta_{H-}) \alpha_o^4 + RS_{11}$$

$$\omega_{22} = \Delta_\Lambda \alpha_o^{0.4} + 1/2(\Delta_{H+} - \Delta_{H-}) \alpha_o^4 + RS_{22}$$

Lab measurement:

$$\omega_{11} = \Delta_\Lambda (\Delta\alpha + \alpha_o)^{0.4} - 1/2(\Delta_{H+} - \Delta_{H-}) (\Delta\alpha + \alpha_o)^4$$

$$\omega_{22} = \Delta_\Lambda (\Delta\alpha + \alpha_o)^{0.4} + 1/2(\Delta_{H+} - \Delta_{H-}) (\Delta\alpha + \alpha_o)^4$$

Dirty secrets:

~~1.  $\Delta H_+ \approx \Delta H_-$  reduces the effect of the  $\alpha^4$  term~~

Use Satellite lines

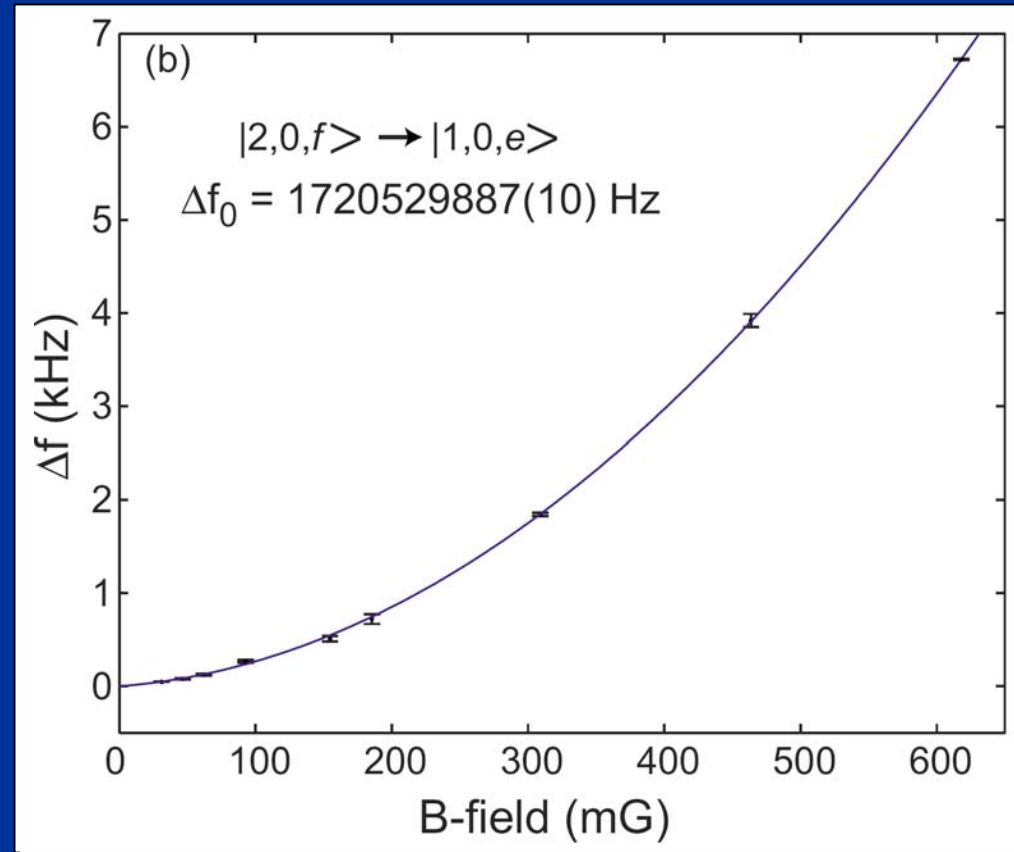
~~these transitions depend weakly on  $\alpha$ .~~

Use all 4 lines... really just want to see something first

~~2. What about the other constants?~~

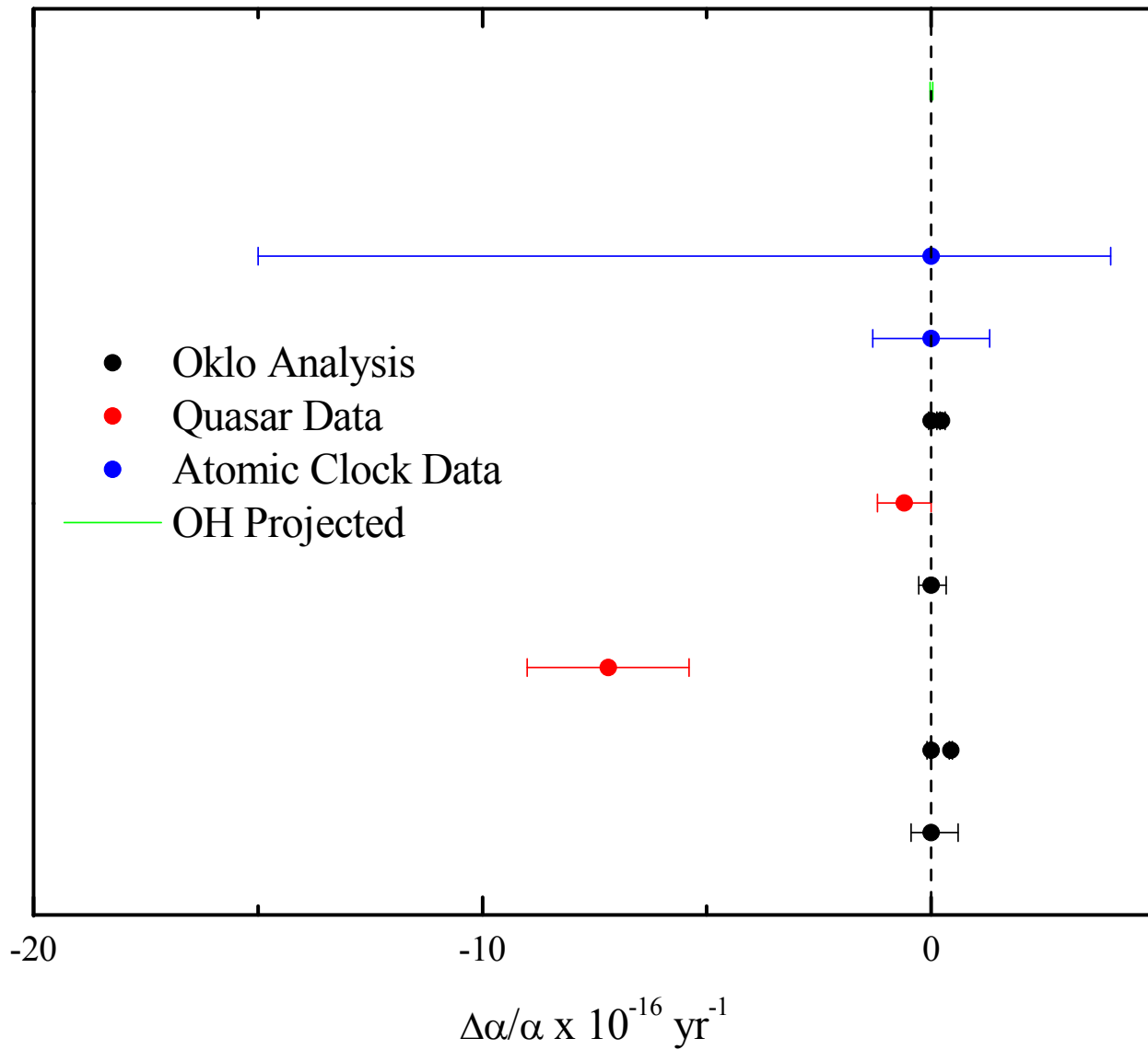
# Measuring the Satellite Lines

- Satellite lines are much more sensitive to magnetic fields
  - Main lines:  $\sim$  kHz/Gauss
  - Satellite lines:  $\sim$  MHz/Gauss
- In our experiment we could apply a very uniform field



$$\nu_{12} = 1\,612\,230\,825(15) \text{ Hz}$$
$$\nu_{21} = 1\,720\,529\,887(10) \text{ Hz}$$
$$\Delta\alpha/\alpha = 30 \text{ ppb over } 10^{10} \text{ years}$$

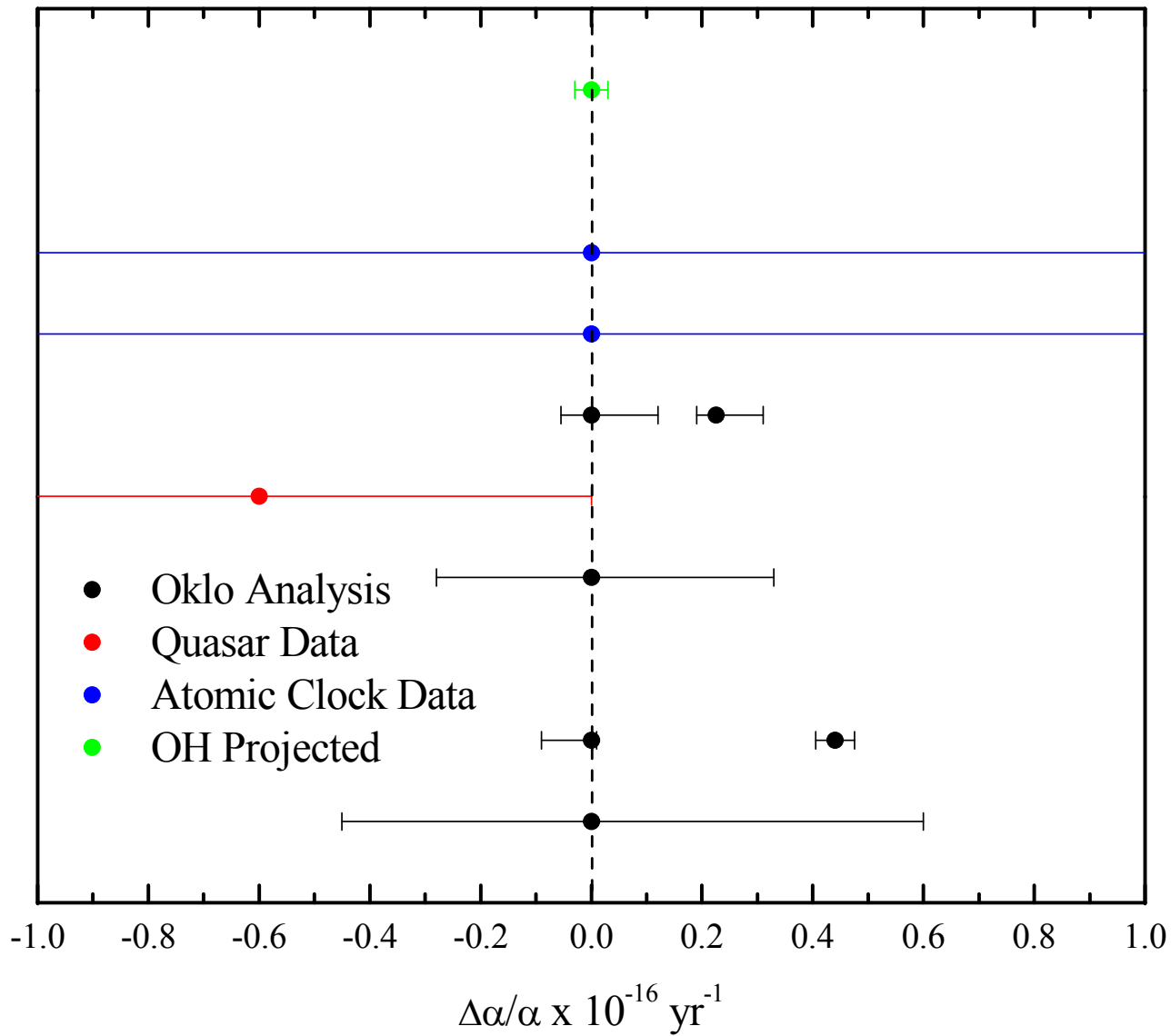
## $\Delta\alpha/\alpha$ Status



Quasar Data:  
PRL 87 091301  
PRL 92 121302

Atomic Clock Data:  
PRL 98 070801

# $\Delta\alpha/\alpha$ Status



Quasar Data:  
PRL 87 091301  
PRL 92 121302

Atomic Clock Data:  
PRL 98 070801

# Th-229 (Yale)

- Transition frequency is (probably) fantastically sensitive to constant variation:

$$\frac{\delta\omega}{\omega} \approx 10^5 \left( 4 \frac{\delta\alpha}{\alpha} + \frac{\delta X_q}{X_q} + \frac{\delta X_s}{X_s} \right)$$

$$X_q = \frac{m_q}{\Lambda_{QCD}}, X_s = \frac{m_s}{\Lambda_{QCD}}$$

V. Flambaum, arXiv:physics/0604188

- Linewidth is ridiculously small:  
 $< 100 \mu\text{Hz}$
- The greatest clock ever

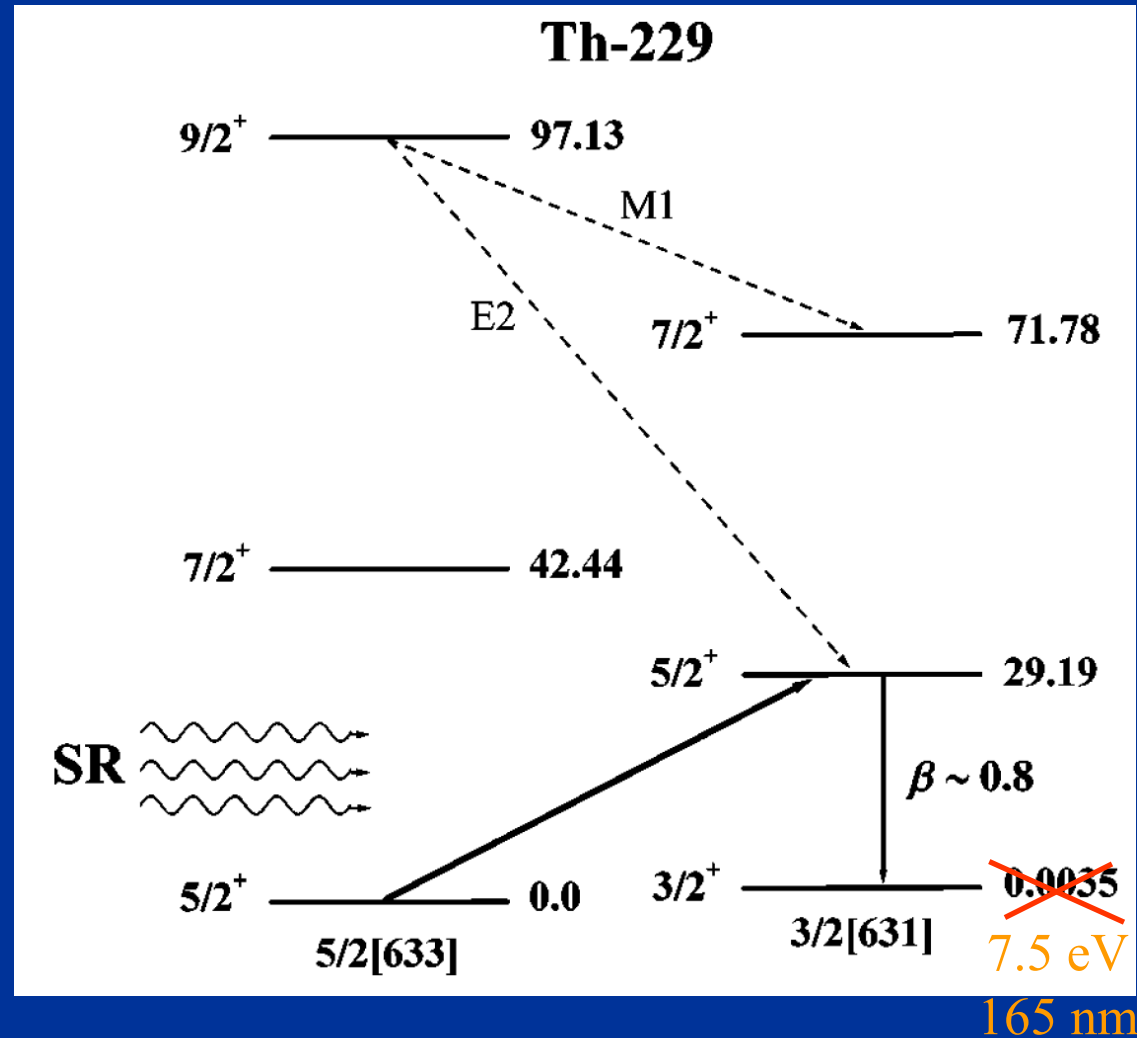


Figure taken from PRC 61 064308

# Th-229

How do you optically observe a nuclear transition?

Previous proposals (incomplete):

~~Vapor cell-like experiments~~

Having Discharge Cyclic Excite the nuclear transition leaves a unique.  
What are the properties of this state and how to look for transition of the state:  
photons and alphas.

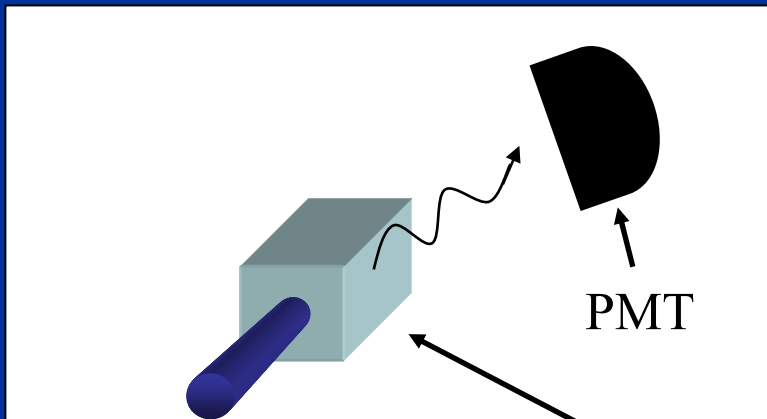
1. **Less sensitive to its environment than electronic transitions.**  
Inamura et al, Hyperfine Interactions (2005)
2. **Traps the nucleus in a metastable state by changing the local spin of the atom.**  
Chang et al, Phys Rev Lett 89, 173001 (2002)
3. **of Decay products**  
Chang et al, Phys Rev Lett 89, 173001 (2002)

E. Peik, Europhys. Lett 61 181 (2003).

1 + 2 + 3 → Put it in a solid to look for transitions (and do NMR spectroscopy or look for decay products)

# Th-229

General Idea (first we just need to see the state):



VUV transmissive material  
doped with Th-229

Tunable laser @ 165 nm  $\pm$  10 nm

- H<sub>2</sub> Raman Cell
- Eventually you'll want a nice CW laser
- VUV comb

**A few notes about detection:**

1. TIR makes solid angle of detection  $\sim 4\pi$
2. PMTs are excellent here (QE  $\sim 40\%$ )
3. Use of monochromator and exploiting the long time scale should give excellent background discrimination.
4. NMR detection of the change in **I** is potentially background free. (Th<sup>232</sup> has **I** = 0)
5. Also look at decay spectrum.



# Th-229

What are the possibilities for VUV transmissive materials?

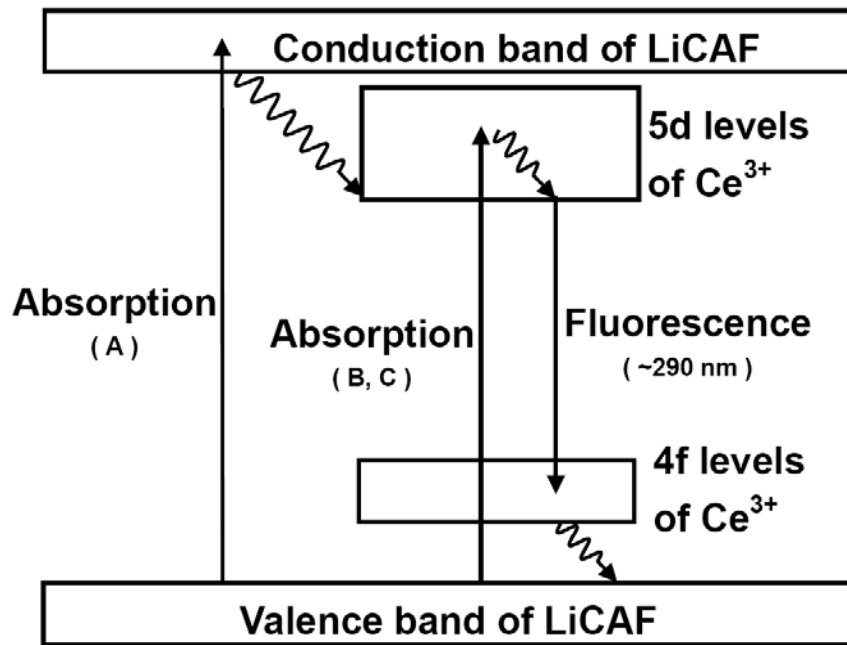
## Readily available

1.  $\text{CaF}_2$
  2.  $\text{MgF}_2$
  3.  $\text{LiF}$
  4. Modified Fused Silica (157 nm photo-lithography)?
- } Th,  $\text{ThF}_3$ , or  $\text{ThF}_4$ ?

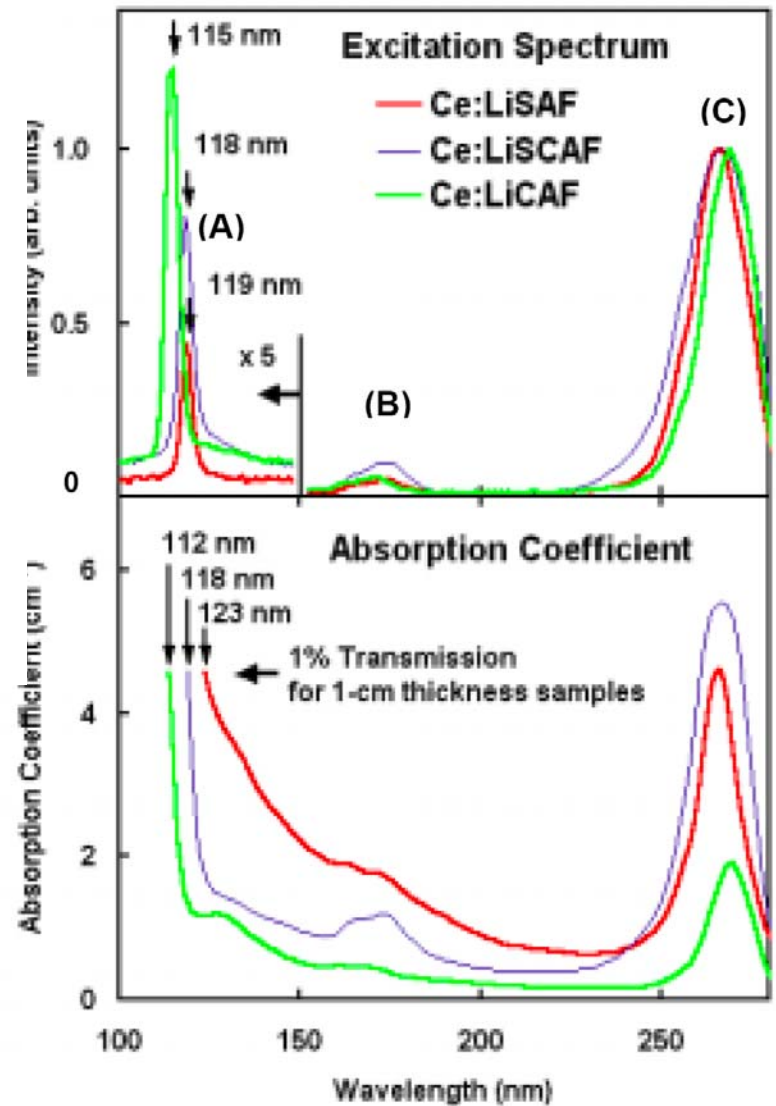
	Ionization Energy
Th	6.1 eV
$\text{Th}^{+1}$	11.5 eV
$\text{Th}^{+2}$	20 eV
$\text{Th}^{+3}$	28.8 eV

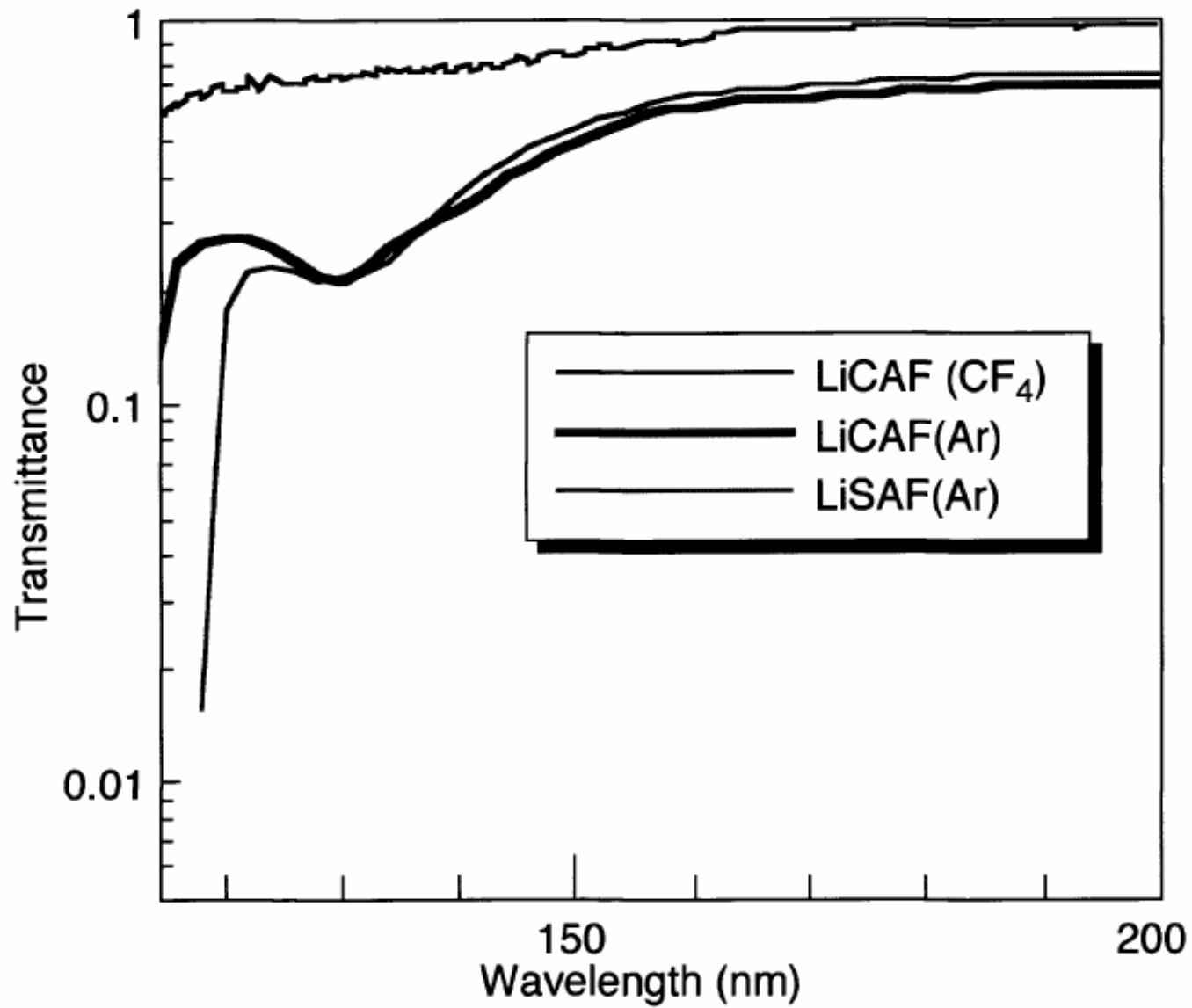
## Not so easy

1. **Ce:LiSAF**
2. **Ce:LiCAF**
  - High transmission down to 110 nm
  - Developed for tunable UV lasers around 300 nm based on  $\text{Ce}^{3+}$ .
  - Crystal developed specifically to handle large amounts of UV power AND to maintain the  $\text{Ce}^{3+}$  level structure!
  - Would expect  $\text{Th}^{3+}$  to not be modified so could verify its presence by strong absorption on the  $d \rightarrow f$  line ( $\tau \sim 10$  ns).



- 5s and 5p level shield 4f electron from crystal field
- Seems band gap is large





# Th-229

Back of the envelope signal-to-noise calculation:

Resonant cross-section:

Parameters:

$$\sigma = \frac{\lambda^2}{2\pi} \frac{\Gamma}{\Delta\omega_L}$$

$$\Gamma = 2\pi (10 \mu\text{Hz})$$

$$\lambda = 165 \text{ nm}$$

For  $\Delta\omega_L = 1 \text{ cm}^{-1}$ ,  $P = 10 \mu\text{J}$ , 10 ns pulse, 1 mm X 1 mm XTAL:

$$N_{\text{Excited}} = N_{\text{Total}} \sigma N_{\text{photons}}$$

	$N_{\text{Total}}$	After one pulse: $\Gamma N_{\text{Excited}}(t=0)$	After 100 ns: $\Gamma N_{\text{Excited}}(t=100 \text{ ns})$
Th-229 Specific Activity: 0.161 $\mu\text{g}/\mu\text{Ci}$	1 $\mu\text{Ci}$	0.3	24 (ppb)
	10 $\mu\text{Ci}$	3	200 (ppb)
	100 $\mu\text{Ci}$	30	1240 (ppb)
	1 mCi	300	2.4 (ppm)

# Th-229

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## Possible “flies”:

1. Th-229: \$50, 000 per mg
2. Fabrication of XTAL, Thorium is radioactive (7900 yr half life)
3. 165 nm laser system
4. Electron Bridge mechanism
5. Background from long-time scale fluorescence in crystal (?)
6. Forming of color centers, leading to more of #5.
7. Broadening due to Hyperfine coupling to electronic cloud and/or nuclear electric quadrupole moment