

# Precision Laser Spectroscopy of Lithium

- a)  $\text{Li}^+ 1s2s \ ^3\text{S} - 1s2p \ ^3\text{P}$  Transition
- b) Li D Lines

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[www.yorku.ca/wlaser](http://www.yorku.ca/wlaser)



# Why study $\text{Li}^+/\text{Li}$ ?

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## 1. Test QED

Effects scale to lowest order as  $Z^4\alpha^3$  times Rydberg energy. QED effects order of magnitude greater than in H or He [1,2].

## 2. Theoretical Advances

Two & maybe 3 electron systems now “well understood” using Hylleraas Variational calculations [2].

## 3. Nuclear Probe

Measurements of isotope shifts plus theory yields relative nuclear charge radii with accuracies of less than 0.1 fm, more accurate than electron scattering. Ideal for studying halo neutrons  ${}^6,7,8,9,11\text{Li}$  [3].

## 4. Experimental Discrepancies

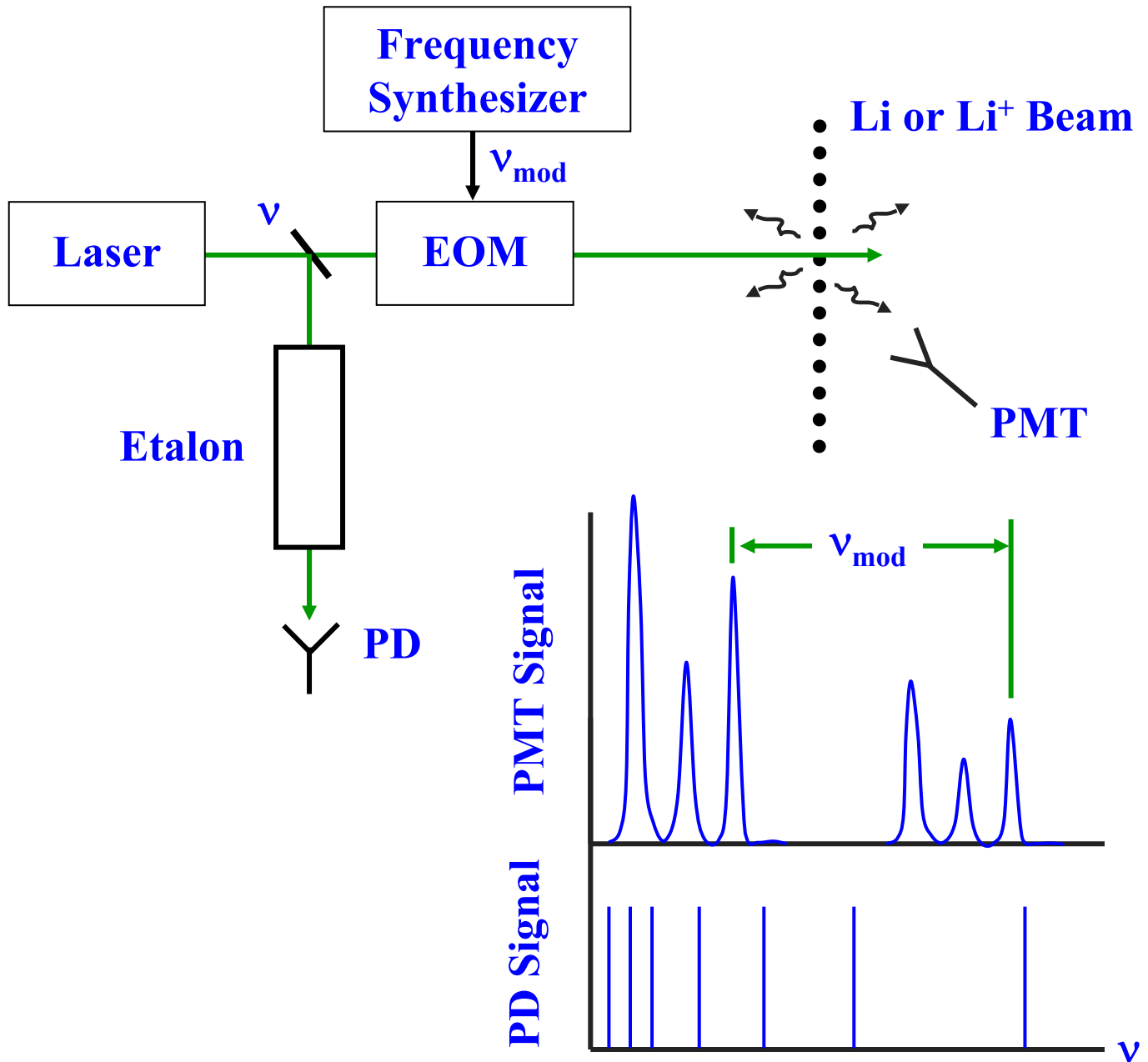
a)  $\text{Li}^+ 1s2p {}^3\text{P}_{1,2}$  Fine Structure: Two measurements with  $1\sigma$  uncertainty  $\sim 0.65$  MHz disagree by 11 MHz.

b) Neutral Li D Lines: Fabry Perot calibration problems.

1. W. A. van Wijngaarden, CJP **83**, 327 (2005).
2. G. Drake et al, CJP **83**, 311 (2005).
3. G. Ewald et al, PRL **94**, 039901 (2005).

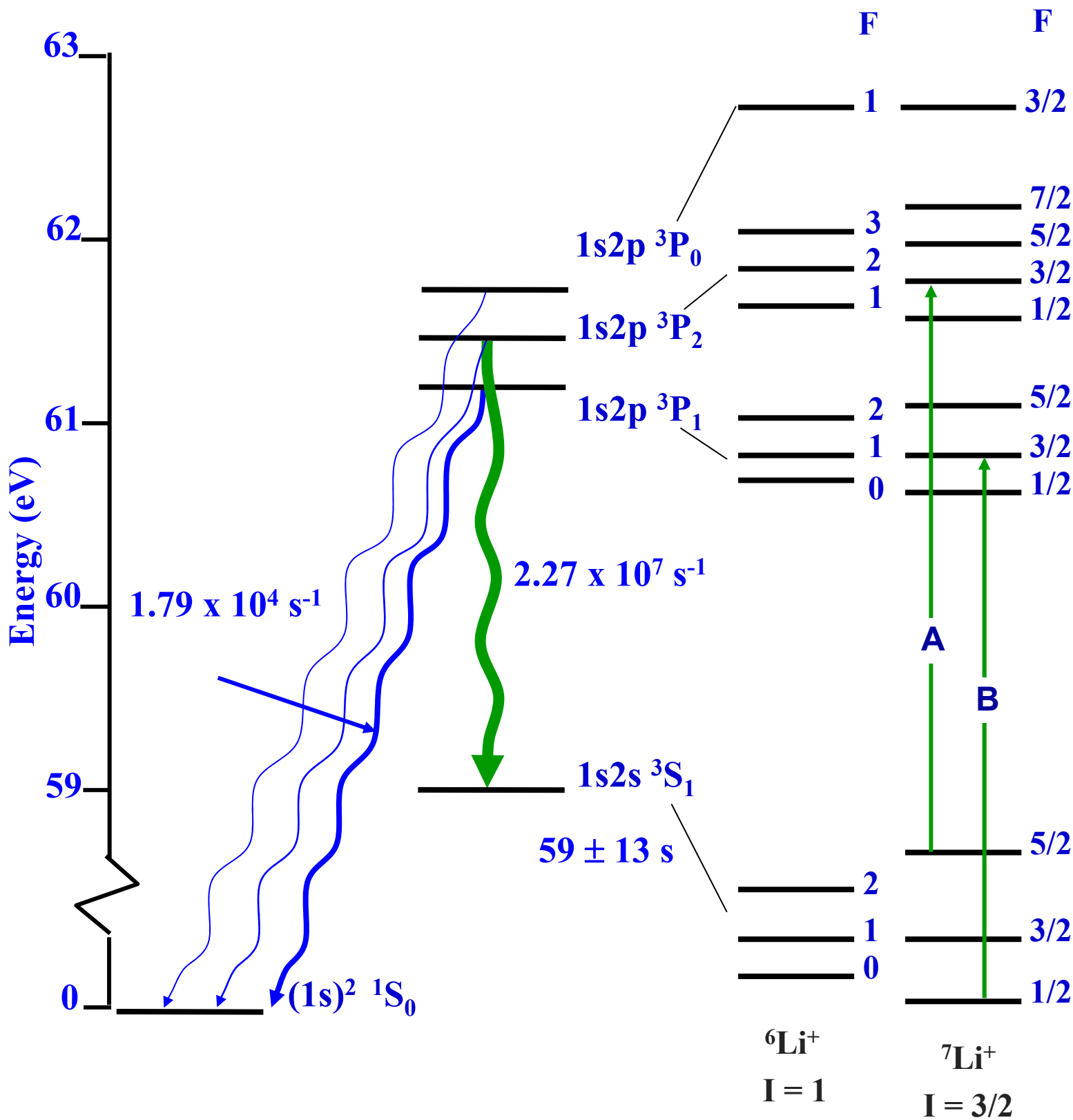
# Measurement Technique

W. van Wijngaarden, Adv. At. Mol. Opt. Phys. 36, 141 (1996)



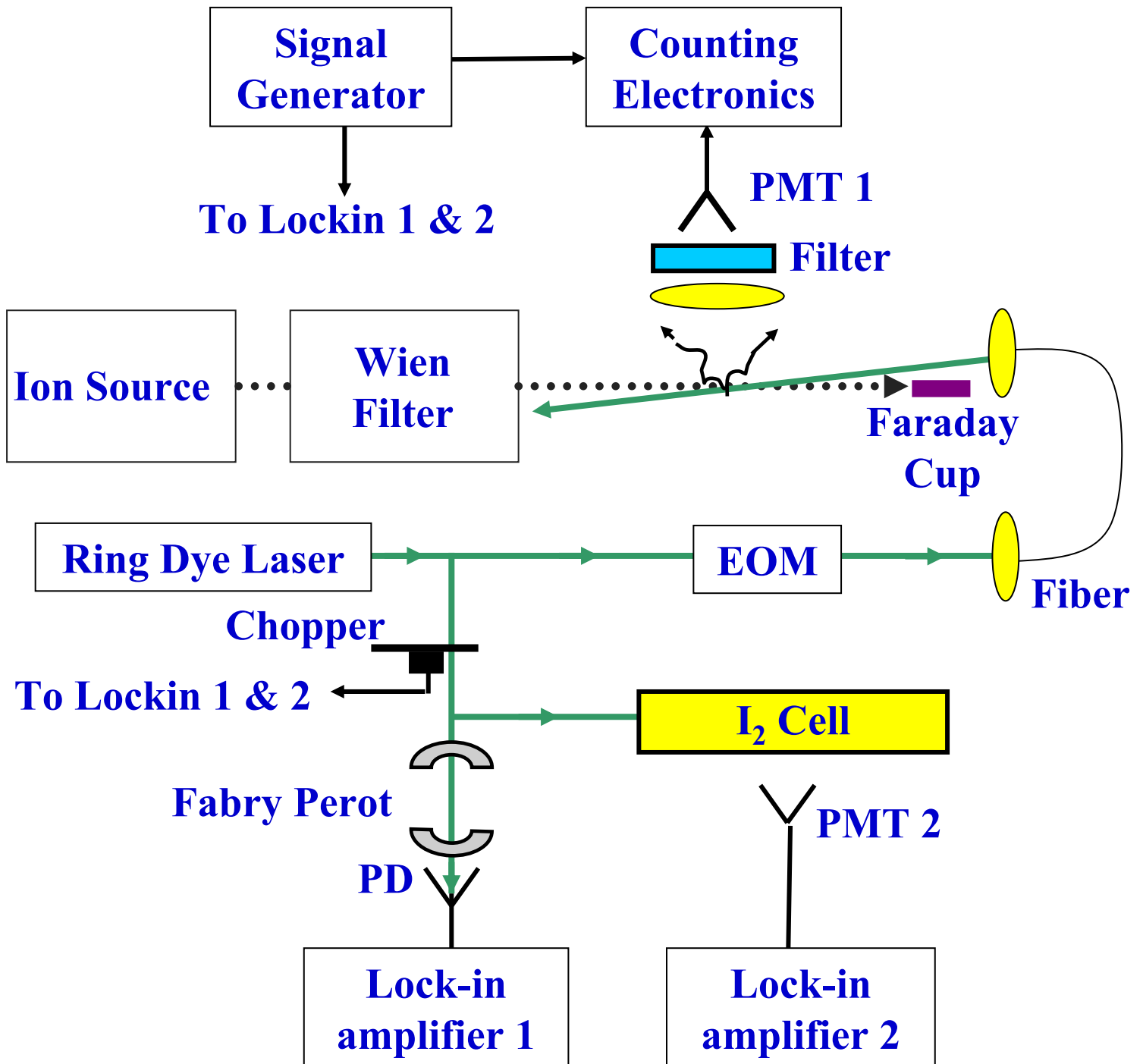
- Etalon peaks account for nonlinearity of laser scan
- Free spectral range found using EOM modulation frequency

# Relevant $\text{Li}^+$ States & Hyperfine Levels

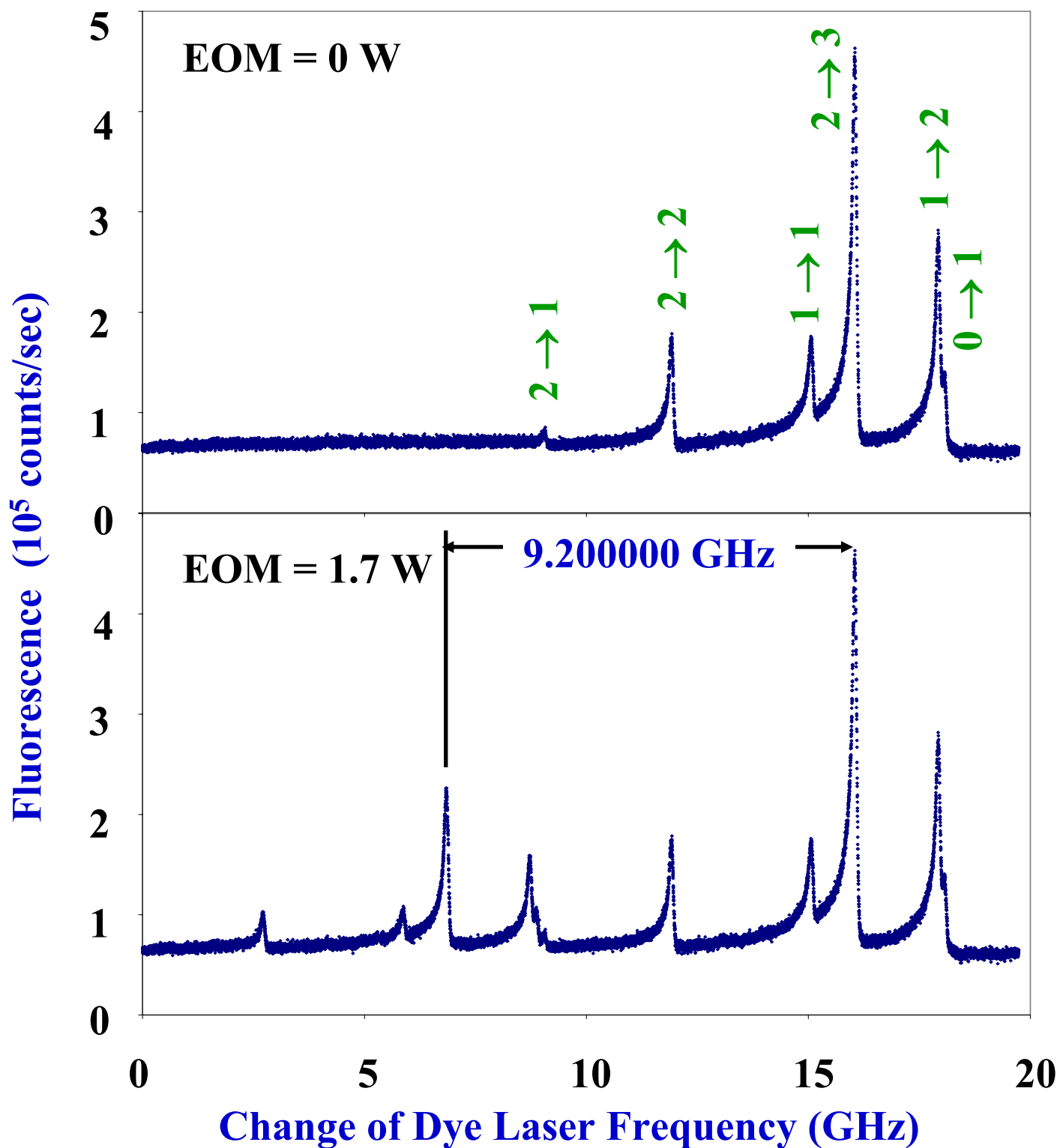


# Apparatus for studying $\text{Li}^+$

J. Clarke & W. van Wijngaarden, Phys. Rev. A 67, 012506 (2003)



# ${}^6\text{Li}^+ 1s2s {}^3S_1(\text{F}) \rightarrow 1s2p {}^3P_2(\text{F}')$ with 9.2 GHz EOM

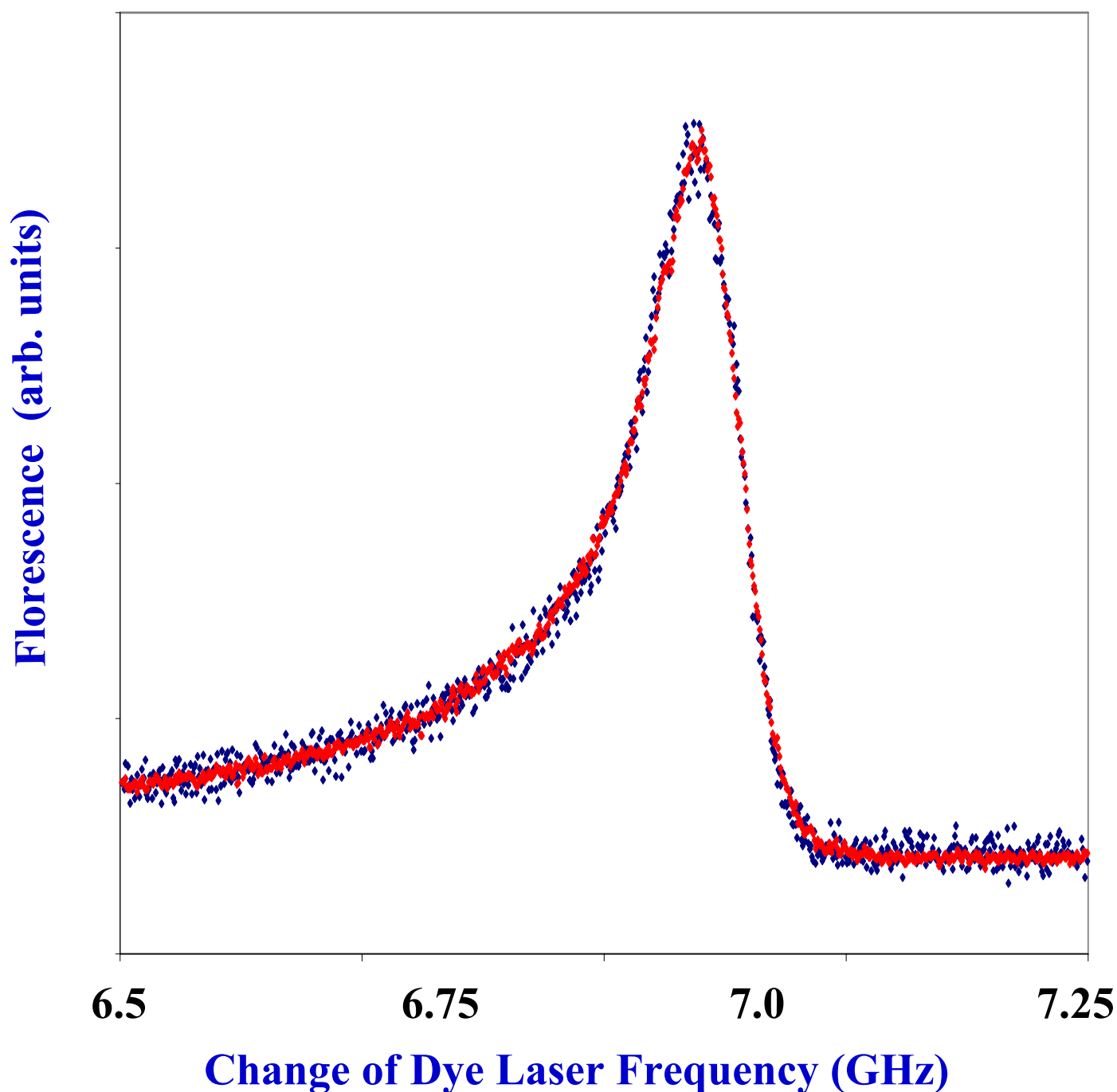


# Determination of Frequency Intervals

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**Red** peak excited by unshifted laser beam scaled & shifted to overlap **blue** peak produced by 9.2 GHz shifted laser beam.



# ${}^6\text{Li}^+$ Hyperfine Intervals

State	Interval F $\rightarrow$ F'	Interval (MHz)		Technique
$1s2s\ {}^3S_1$	2 $\rightarrow$ 1	5,993	$\pm 6$	Doppler Tuning <sup>1</sup>
		5,997	$\pm 4$	Laser Scan / Etalon <sup>2</sup>
		6,003.600	$\pm 0.050$	Microwave <sup>3</sup>
		6,003.66	$\pm 0.51$	Our Expt <sup>4</sup>
		6,003.614	$\pm 0.024$	H.V. Theory <sup>5</sup>
	1 $\rightarrow$ 0	2,998	$\pm 6$	Doppler Tuning <sup>1</sup>
		2,998	$\pm 4$	Laser Scan / Etalon <sup>2</sup>
		3,001.780	$\pm 0.050$	Microwave <sup>3</sup>
		3,001.827	$\pm 0.47$	Our Expt <sup>4</sup>
		3,001.765	$\pm 0.038$	H.V. Theory <sup>5</sup>
$1s2p\ {}^3P_1$	2 $\rightarrow$ 1	2,888.98	$\pm 0.63$	Our Expt <sup>4</sup>
		2,888.327	$\pm 0.029$	H.V. Theory <sup>5</sup>
	1 $\rightarrow$ 0	1,316.06	$\pm 0.59$	Our Expt <sup>4</sup>
		1,317.649	$\pm 0.046$	H.V. Theory <sup>5</sup>
$1s2p\ {}^3P_2$	3 $\rightarrow$ 2	4,127.16	$\pm 0.76$	Our Expt <sup>4</sup>
		4,127.882	$\pm 0.043$	H.V. Theory <sup>5</sup>
	2 $\rightarrow$ 1	2,857.00	$\pm 0.72$	Our Expt <sup>4</sup>
		2,858.002	$\pm 0.060$	H.V. Theory <sup>5</sup>

1. B. Fan et al, Opt Lett **4** 233 (1979).

2. R. Bayer et al, Z Phys A **292**, 329 (1979)

3. J. Kowalski et al, Hyp Int **15** 159 (1983)

4. J. Clarke et al, PRA **67**, 12506 (2003)

5. E. Riis et al, PRA **49** 207 (1994)



# ${}^7\text{Li}^+ 1s2p \ ^3\text{P}_{1-2}$ Fine Structure

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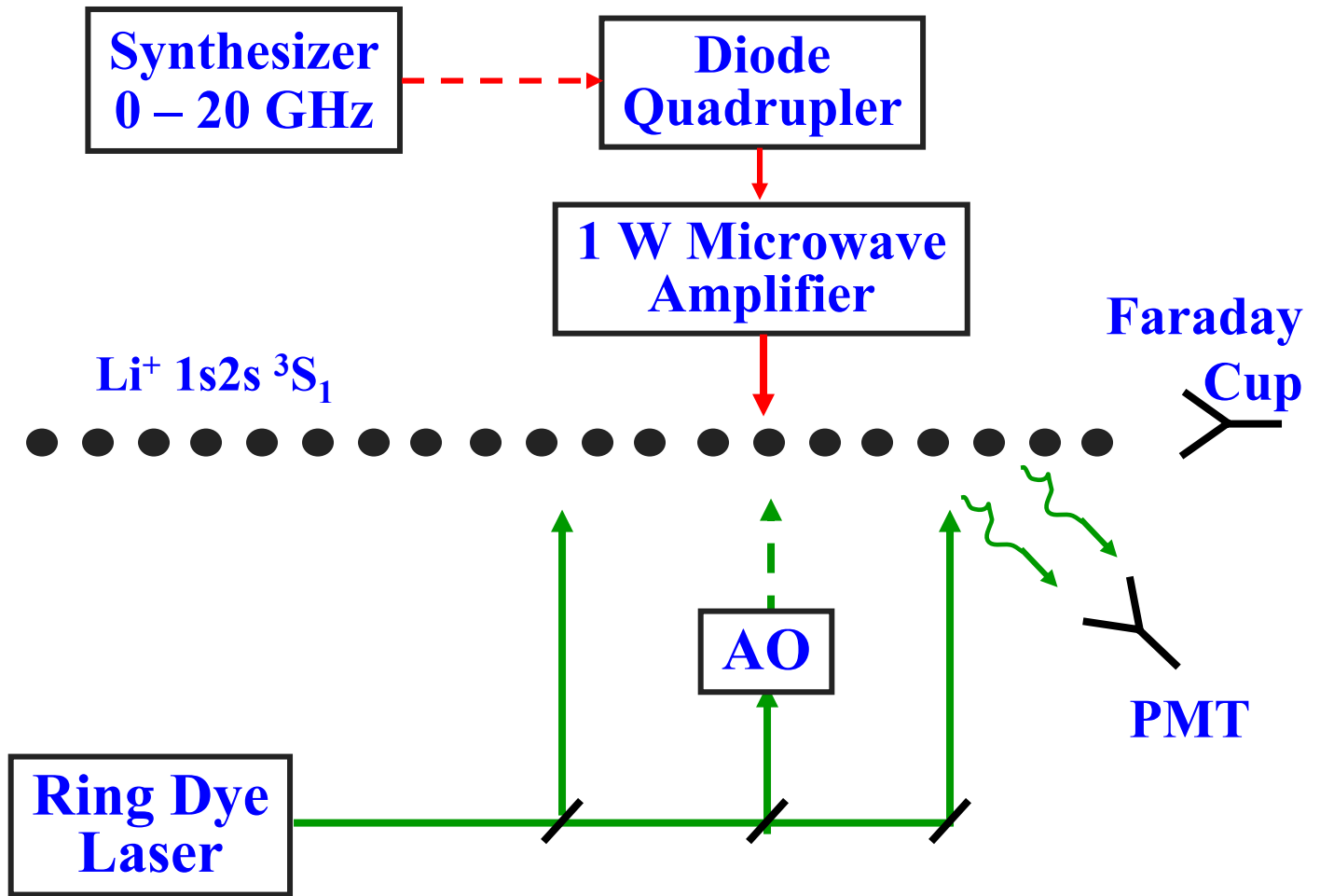
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Interval (MHz)	Technique
62,658 $\pm 28$	Laser Scan/Etalon <sup>1</sup>
62,678 $\pm 14$	Wavemeter <sup>2</sup>
62,682 $\pm 6$	Fast Beam <sup>3</sup>
62,667.4 $\pm 2.0$	Laser Heterodyne <sup>4</sup>
62,678.41 $\pm 0.65$	Fast Beam <sup>5</sup>
62,679.46 $\pm 0.98$	Our Expt <sup>6</sup>
62,679.4 $\pm 0.5$	H.V. Theory <sup>7</sup>

1. R. Bayer et al, Z. Phys. A **292**, 329 (1979)
2. R. Schwarzwald, Diplome Thesis, U. of Heidelberg (1982)
3. E. Riis et al, PRA **33**, 3023 (1986)
4. H. Rong et al, Z. Phys D **25**, 337 (1993)
5. E. Riis et al, PRA **49**, 207 (1994)
6. J. Clarke & WvW, PRA **67**, 12506 (2003)
7. T. Zhang et al, PRL, **77**, 1715 (1996)

# Ongoing Work

## Optical Double Resonance Expt.



**Objective: Measure 1s2p <sup>3</sup>P Fine Structure**

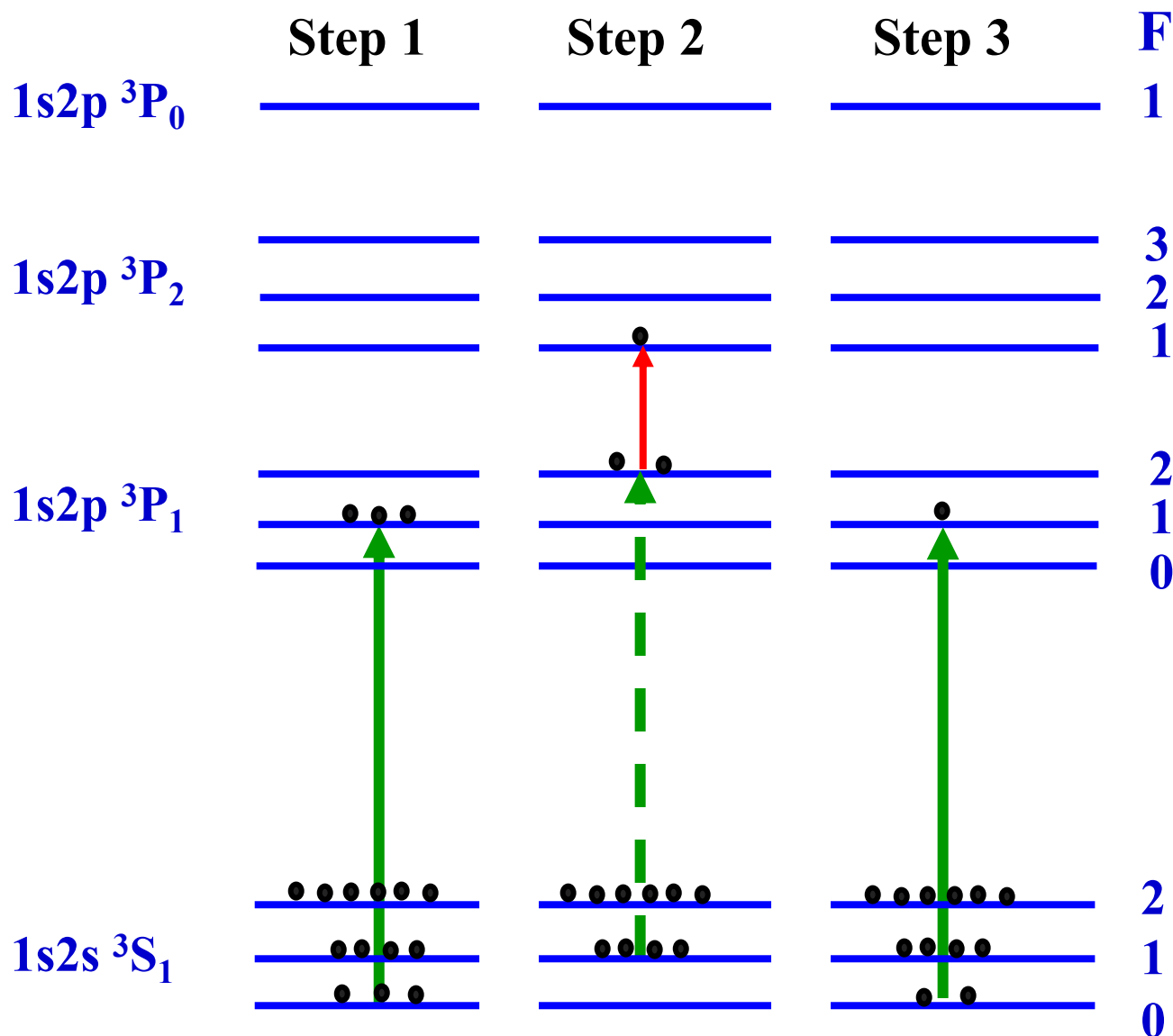
$$1s2p \ ^3P_{0-1} = 155.7 \text{ GHz}$$

$$1s2p \ ^3P_{1-2} = 62.7 \text{ GHz}$$

Natural linewidth of 1s2p <sup>3</sup>P state = 3.7 MHz

⇒ 0.1% measurement of <sup>3</sup>P<sub>1-2</sub> interval gives fine structure to one part in  $2 \times 10^7$

# ${}^6\text{Li}^+$ Hyperfine Level Populations

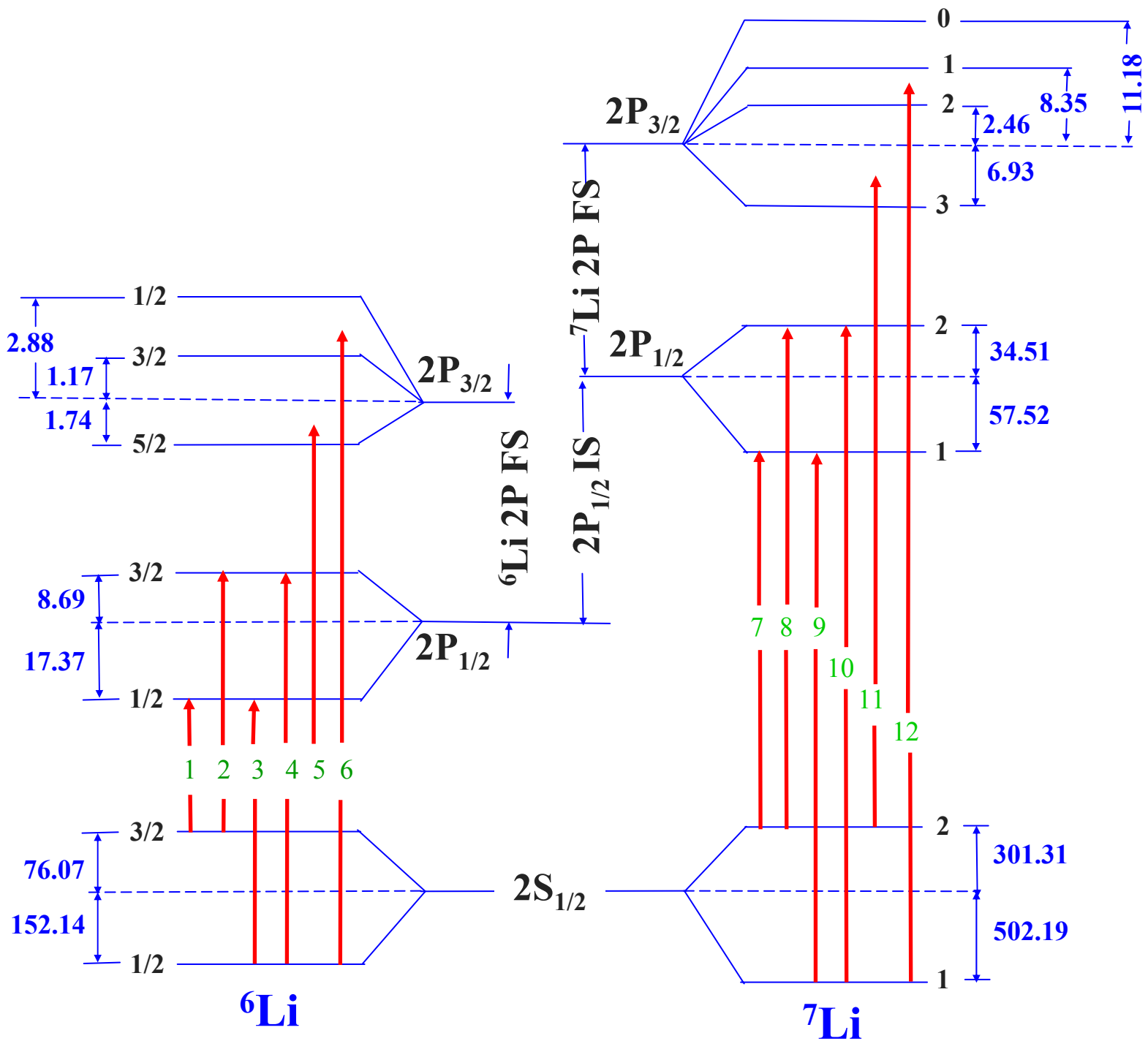


## Procedure

- 1) Optical Pumping depletes  $1s2s\ {}^3S_1$  ( $F=0$ ) level.
- 2) AO shifted laser excites  $1s2s\ {}^3S_1$  ( $F=1$ )  $\rightarrow$   $1s2p\ {}^3P_1$  ( $F=2$ ) & microwaves excite  $1s2p\ {}^3P_1$  ( $F=2$ )  $\rightarrow$   ${}^3P_2$  ( $F=1$ ) transition.
- 3) Excite  $1s2s\ {}^3S_1$  ( $F=0$ ) level & detect fluorescence to measure # transitions induced by microwaves

# Relevant Li Energy Levels

(units in MHz)



# Study of Li D Transitions

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Initial Expt. [1]	Later Expt. [2]
Diode Laser	Dye Laser >100 times more linear frequency scan
Etalon FSR = 300 MHz	Etalon FSR = 150 MHz
Old Digitizer 1 data pt = 0.5 MHz	New Digitizer 1 data pt = 12 kHz
Analysis of composite peaks requires modeling of optical pumping	Analysis independent of optical pumping modeling
	Helmholtz Coils cancel $B_{\text{Earth}}$

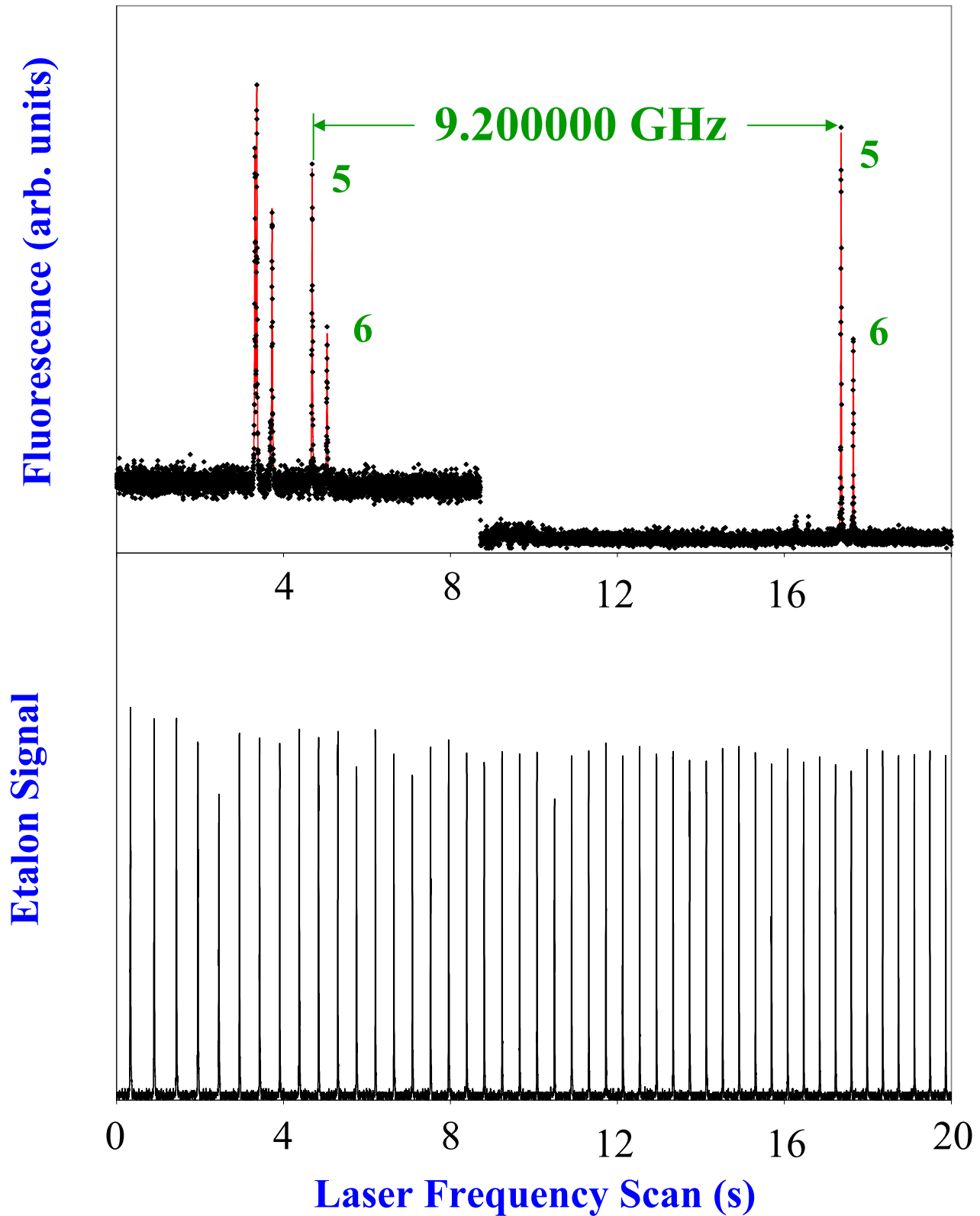
1. J. Walls et al, EPJ D **22**, 159 (2003)
2. G. Noble et al, PRA **74** 012502 (2006)

# Excitation of ${}^6\text{Li}$ D Lines

## Data from Initial Expt.

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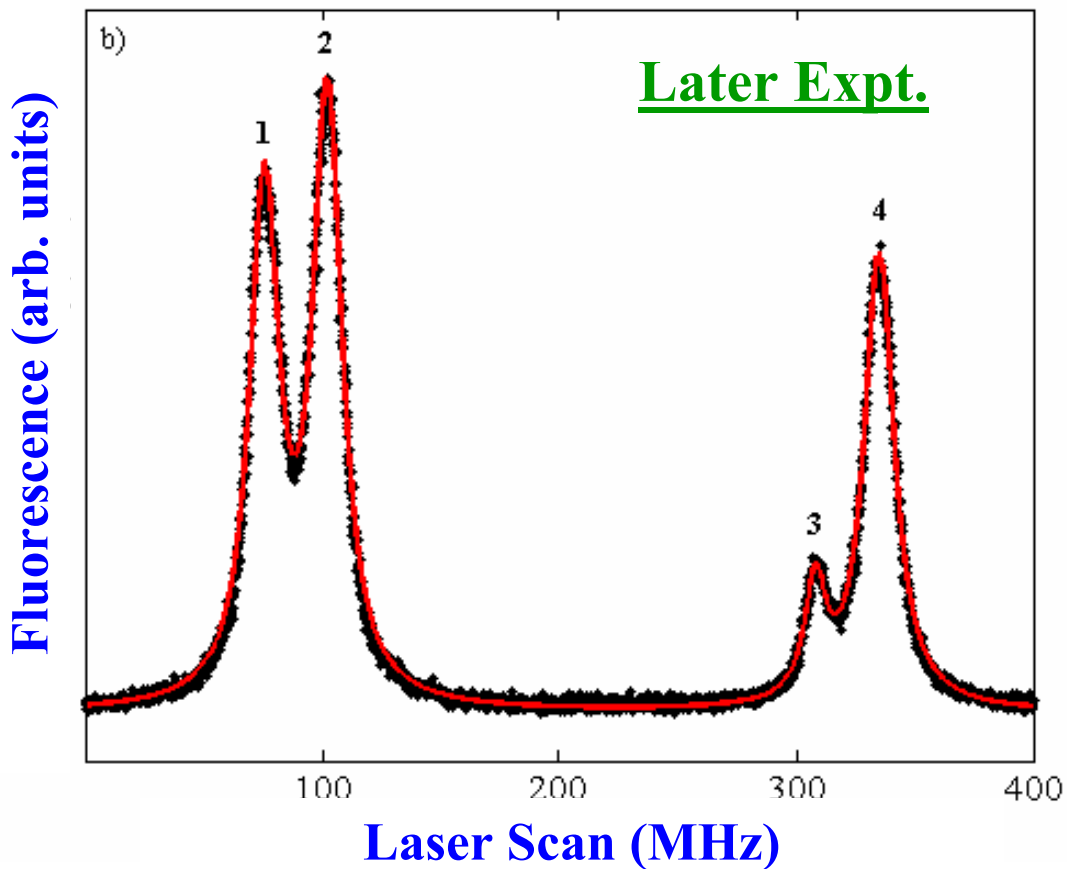
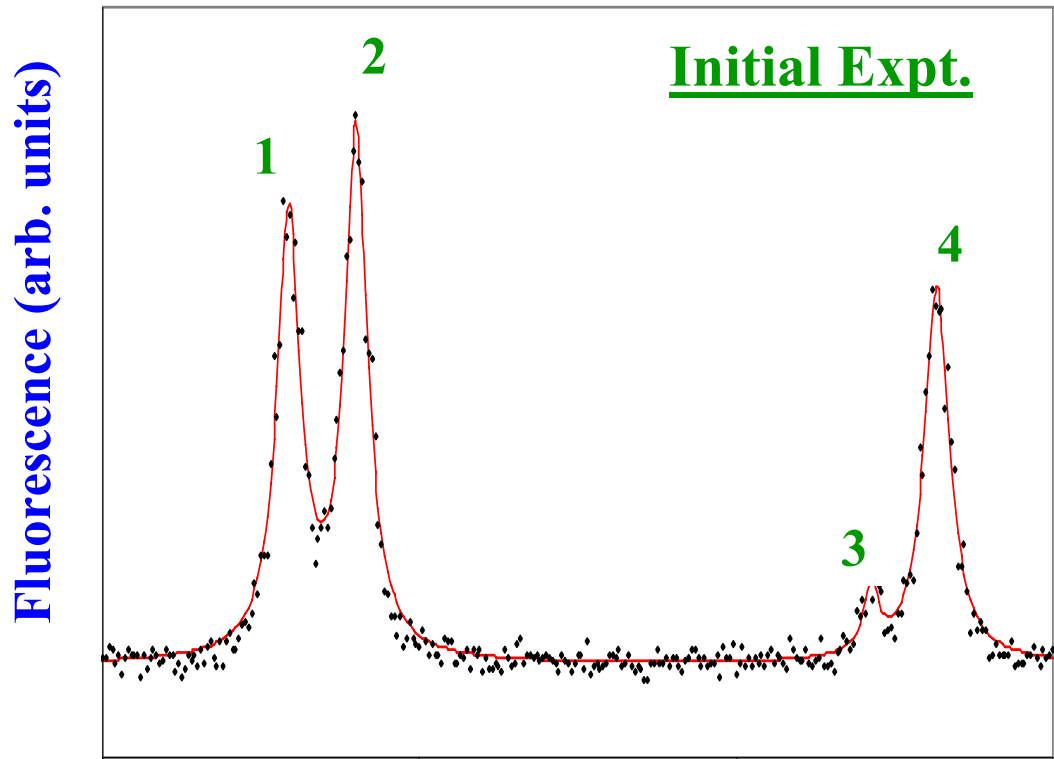
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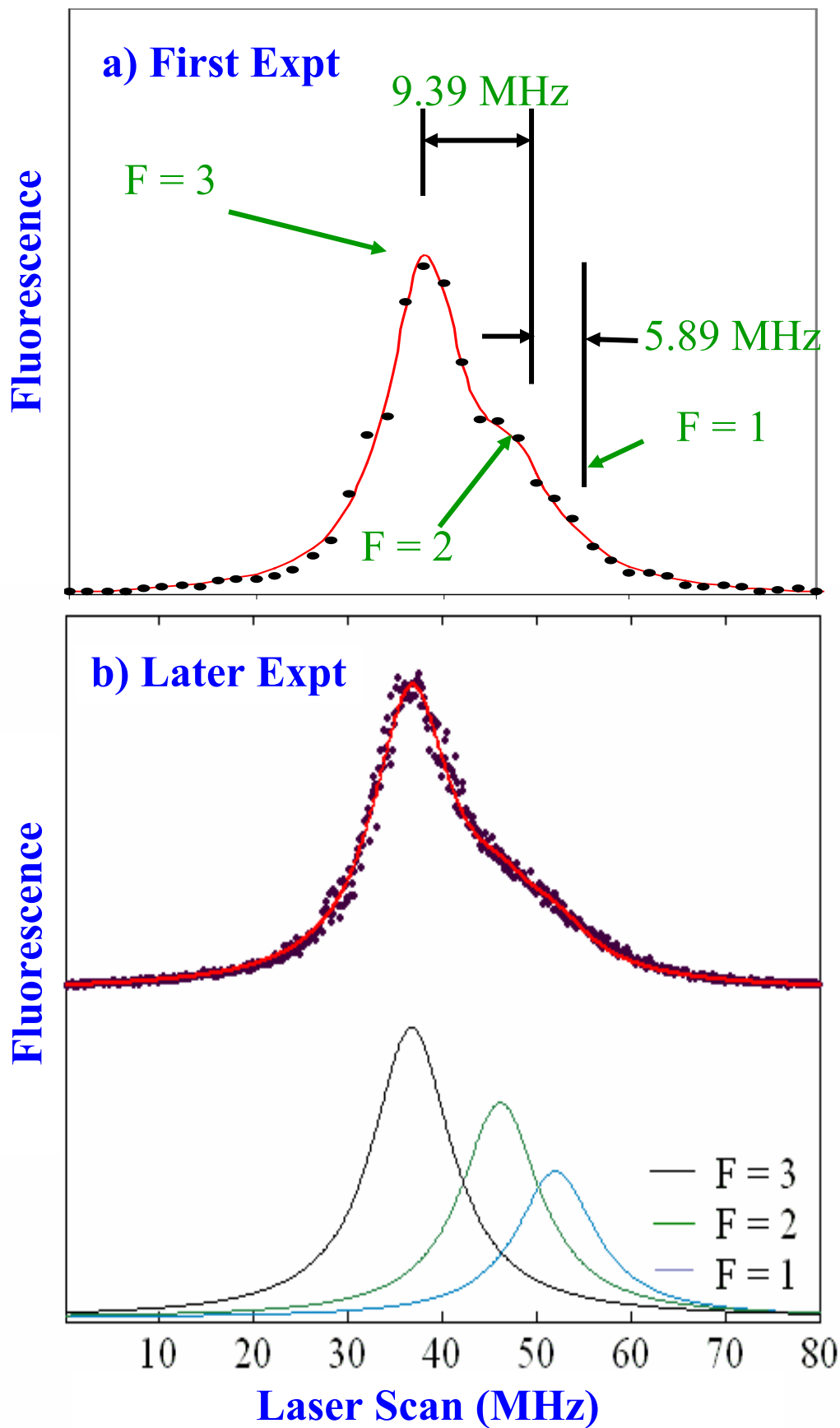
# Excitation of ${}^6\text{Li}$ D Lines

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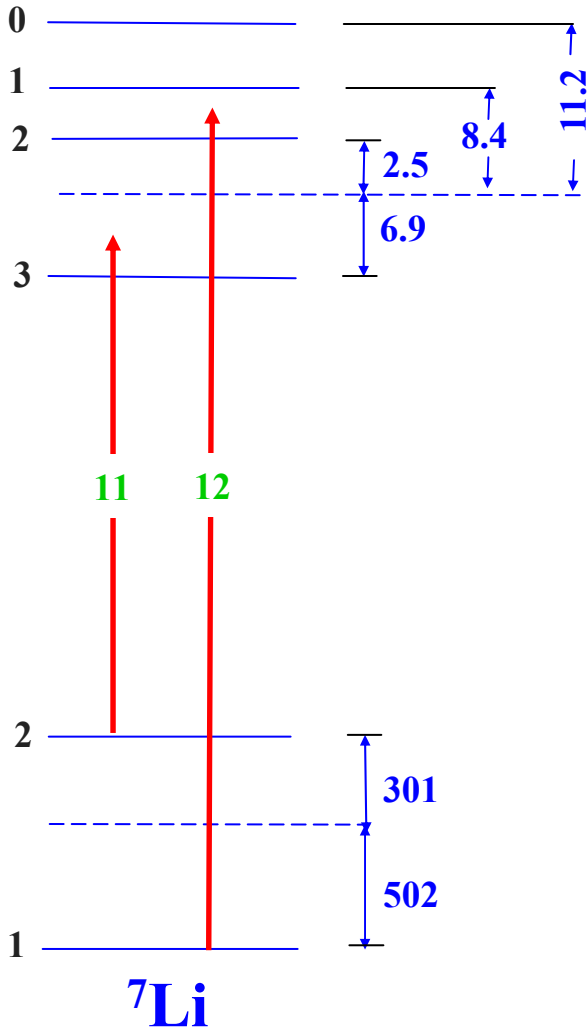


# Peak 11: ${}^7\text{Li } 2S_{1/2} (F=2) \rightarrow 2P_{3/2}$ Excitation



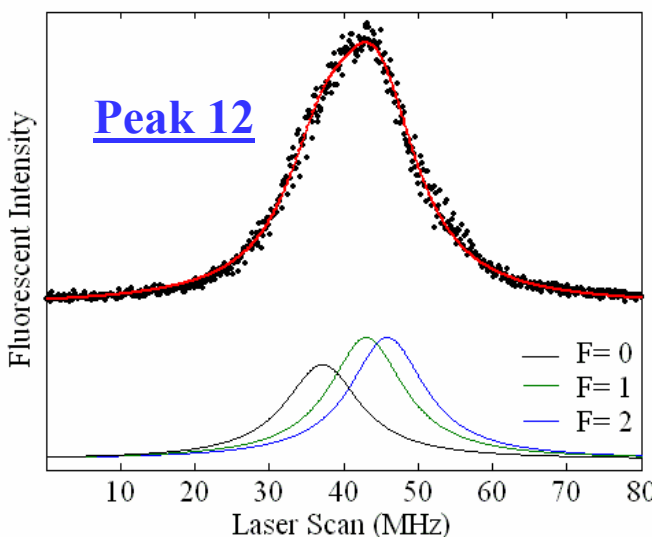


# Determining Line Center to better than Natural Linewidth



$2P_{3/2}$  Hyperfine Spacing  $\leq$  Natural Linewidth

- Optical pumping changes pops. of  $2S_{1/2}$   $m_F$  levels as atom passes through laser affecting contributions of  $2P_{3/2}$   $F = 0, 1, 2$  levels to peak 12
- Initial expt. relied on modeling to estimate fractions of fluorescence due to various excited state hyperfine levels
- Later expt. has higher resolution and fitting varies peak amplitudes



$2P_{3/2}$ Hyperfine Level	Predicted Contribution to Peak 12	Observed Contribution to Peak 12
$F = 0$	28%	29%
$F = 1$	36%	34%
$F = 2$	36%	37%

# Hyperfine Splitting Measurements

Atom	State	a (MHz)	Technique
${}^6\text{Li}$	$2S_{1/2}$	152.136839	Magnetic Resonance <sup>1</sup>
		152.109 ± 0.043	Our Expt <sup>2</sup>
	$2P_{1/2}$	17.375 ± 0.018	Opt. Double Resonance <sup>3</sup>
		17.8 ± 0.3	Level Crossing <sup>4</sup>
		16.8 ± 0.7	Laser Atomic Beam <sup>5</sup>
		17.386 ± 0.031	Our Expt <sup>2</sup>
${}^7\text{Li}$	$2S_{1/2}$	401.7520433	Magnetic Resonance <sup>1</sup>
		401.767 ± 0.039	Our Expt <sup>2</sup>
	$2P_{1/2}$	45.914 ± 0.025	Opt Double Resonance <sup>3</sup>
		46.05 ± 0.30	Laser Atomic Beam <sup>5</sup>
		46.010 ± 0.025	Our Expt <sup>2</sup>
		45.793	Full Core + Correlation <sup>6</sup>
	45.984 ± 0.007	MCHF <sup>7</sup>	
	45.945	MCHF + rel. corr. <sup>8</sup>	

1. A. Beckmann et al, Z. Phys. **270**, 173 (1974)
2. G. Noble et al, PRA **74** 012502 (2006)
3. E. Arimondo et al, Rev. Mod. Phys. **49**, 31 (1977)
4. W. Nagourney & W. Happer, PRA **17**, 1394 (1978)
5. L. Windholz et al, Z. Phys. D **16**, 41 (1990)
6. X. Guan, EPJ **2**, 21 (1998)
7. N. Yamanaka & Z. Wang, J. Phys. Soc. Japan **68**, 2561 (1999)
8. M. Goedfroid et al, J. Phys. B **34**, 1079 (2001)

# 2P Fine Structure Splitting

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	<b>Interval (MHz)</b>	<b>Technique</b>
${}^6\text{Li}$	10052.76 $\pm$ 0.22	Level Crossing <sup>1</sup>
	10051.62 $\pm$ 0.20	Laser Atomic Beam <sup>2</sup>
	10052.964 $\pm$ 0.050	Our Expt <sup>3</sup>
	10050.846 $\pm$ 0.012	H.V. Theory <sup>5</sup>
${}^7\text{Li}$	10053.24 $\pm$ 0.22	Level Crossing <sup>1</sup>
	10053.184 $\pm$ 0.058	Optical Double Resonance <sup>4</sup>
	10053.4 $\pm$ 0.2	Laser Atomic Beam <sup>2</sup>
	10053.119 $\pm$ 0.058	Our Expt <sup>3</sup>
	10051.214 $\pm$ 0.012	H.V. Theory <sup>5</sup>

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1. K. Brog et al, Phys. Rev. **153**, 91 (1966)
2. W. Scherf et al, Z. Phys. D **36**, 31 (1996).
3. G. Noble et al, PRA **74** 012502 (2006)
4. H. Orth et al, Z. Phys. A **273**, 221 (1975).
5. Z. Yan & G. Drake, PRA **66**, 042504 (2002).

# ${}^6,{}^7\text{Li}$ Relative Nuclear Charge Radius

$$\Delta r_c^2 = r_c^2({}^6\text{Li}) - r_c^2({}^7\text{Li}) = (\text{ISO}_{\text{meas}} - E_{\text{calcul}}) / C$$

Reference	Transition	$\delta\nu$ (MHz)	$\Delta r_c^2$ (fm <sup>2</sup> )
E. Riis et al <sup>1</sup>	$\text{Li}^+ 2\ {}^3\text{S}_1 - 2\ {}^3\text{P}_0$	$34747.73 \pm 0.55$	0.78
	$\text{Li}^+ 2\ {}^3\text{S}_1 - 2\ {}^3\text{P}_1$	$34747.46 \pm 0.67$	0.78
	$\text{Li}^+ 2\ {}^3\text{S}_1 - 2\ {}^3\text{P}_2$	$34748.91 \pm 0.62$	0.64
W. Scherf et al <sup>2</sup>	$\text{Li } 2\ {}^2\text{S}_{1/2} - 2\ {}^2\text{P}_{1/2}$	$10533.13 \pm 0.15$	0.39
	$\text{Li } 2\ {}^2\text{S}_{1/2} - 2\ {}^2\text{P}_{3/2}$	$10534.93 \pm 0.15$	0.96
GSI Group <sup>3,4</sup>	$\text{Li } 2\ {}^2\text{S}_{1/2} - 3\ {}^2\text{S}_{1/2}$	$11453.95 \pm 0.13$	0.60
	$\text{Li } 2\ {}^2\text{S}_{1/2} - 3\ {}^2\text{S}_{1/2}$	$11453.734 \pm 0.030$	0.46
	$\text{Li } 2\ {}^2\text{S}_{1/2} - 2\ {}^2\text{P}_{1/2}$	$10533.160 \pm 0.068$	0.40
D. Das et al <sup>5</sup>	$\text{Li } 2\ {}^2\text{S}_{1/2} - 2\ {}^2\text{P}_{1/2}$	$10534.215 \pm 0.039$	0.83
	$\text{Li } 2\ {}^2\text{S}_{1/2} - 2\ {}^2\text{P}_{3/2}$	$10533.352 \pm 0.068$	0.32
This Work <sup>6</sup>	$\text{Li } 2\ {}^2\text{S}_{1/2} - 2\ {}^2\text{P}_{1/2}$	$10534.039 \pm 0.070$	0.76
	$\text{Li } 2\ {}^2\text{S}_{1/2} - 2\ {}^2\text{P}_{3/2}$	$10534.194 \pm 0.104$	0.66
Nuclear Theory <sup>7</sup>			0.74
Elect. Scatt. <sup>8</sup>			0.84

1. Riis et al, PRA **49**, 207 (1994)
2. Scherf et al, Z. Phys. D **36**, 31 (1996)
3. Ewald et al, PRL **94**, 039901 (2005)
4. Bushaw et al, PRL **91**, 043004 (2003)

5. Das et al, PRA **75**, 052508 (2007)
6. Noble et al, PRA **74** 012502 (2006)
7. Pieper et al, PRC **64**, 014001 (2001)
8. de Jager, At.Nucl. Data **14**, 479 (1974)

# Conclusions

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## Li<sup>+</sup> 1s2s <sup>3</sup>S - 1s2p <sup>3</sup>P Transition

- Hyperfine intervals of <sup>6</sup>Li<sup>+</sup> 1s2p <sup>3</sup>P state order of magnitude more accurate than previous work.
- Discrepancy of 1s2p Li<sup>+</sup> fine structure resolved.

## Li D Lines

- Our expt. simultaneously yields <sup>6,7</sup>Li isotope shift, hyperfine & fine structure of 2S<sub>1/2</sub> & 2P<sub>1/2</sub> states.
- Hyperfine & fine splittings agree with best existing data
- Theoretical fine structure estimates lower by a few MHz.
- D1 & D2 isotope shifts give  $r_c(^6\text{Li}) = 2.544$  &  $2.524$  fm using  $r_c(^7\text{Li})$ , a difference of only  $2 \times 10^{-17}$  m

## Ongoing Work

- Improve 1s2p <sup>3</sup>P Li<sup>+</sup> fine structure by order of magnitude in optical double resonance expt.