Precision Laser Spectroscopy of Lithium

a) Li^+ 1s2s ^3S – 1s2p ^3P Transition

b) Li D Lines

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Why study Li^+/Li?

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}Li [3].

4. **Experimental Discrepancies**

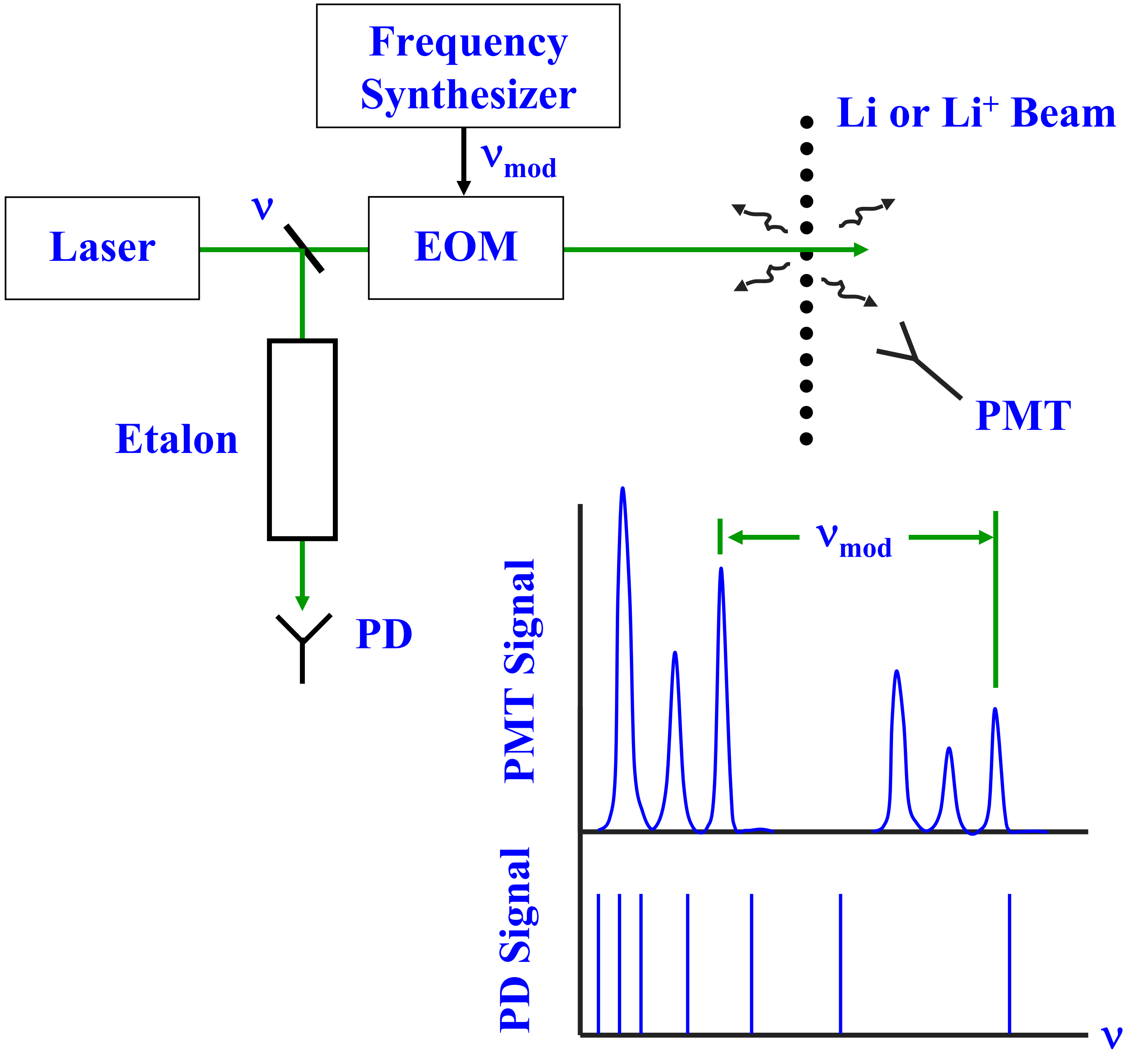
   a) **Li^+ 1s2p $^3P_{1,2}$ Fine Structure:** Two measurements with 1σ uncertainty ~0.65 MHz disagree by 11 MHz.

   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).

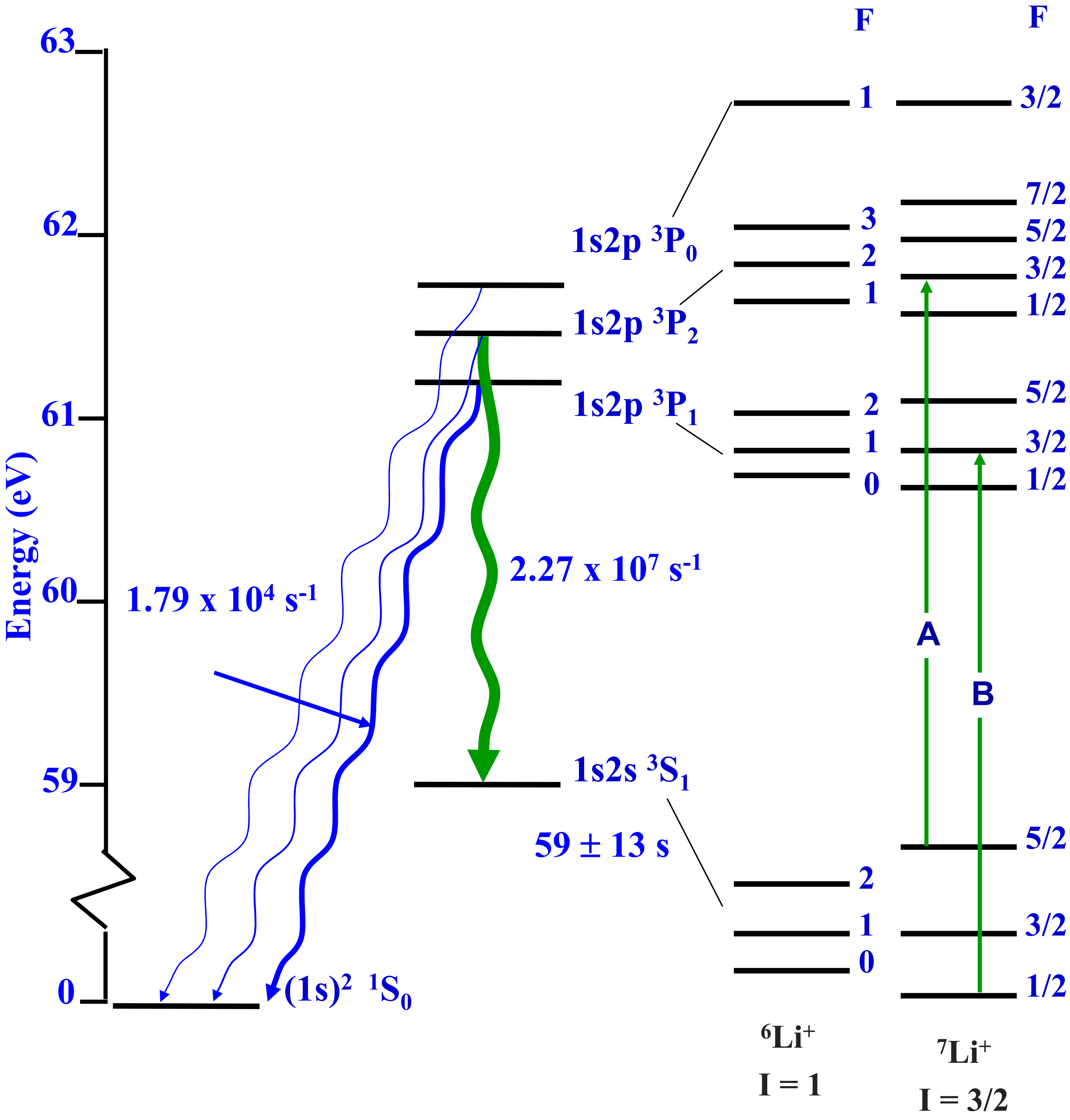
Measurement Technique

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



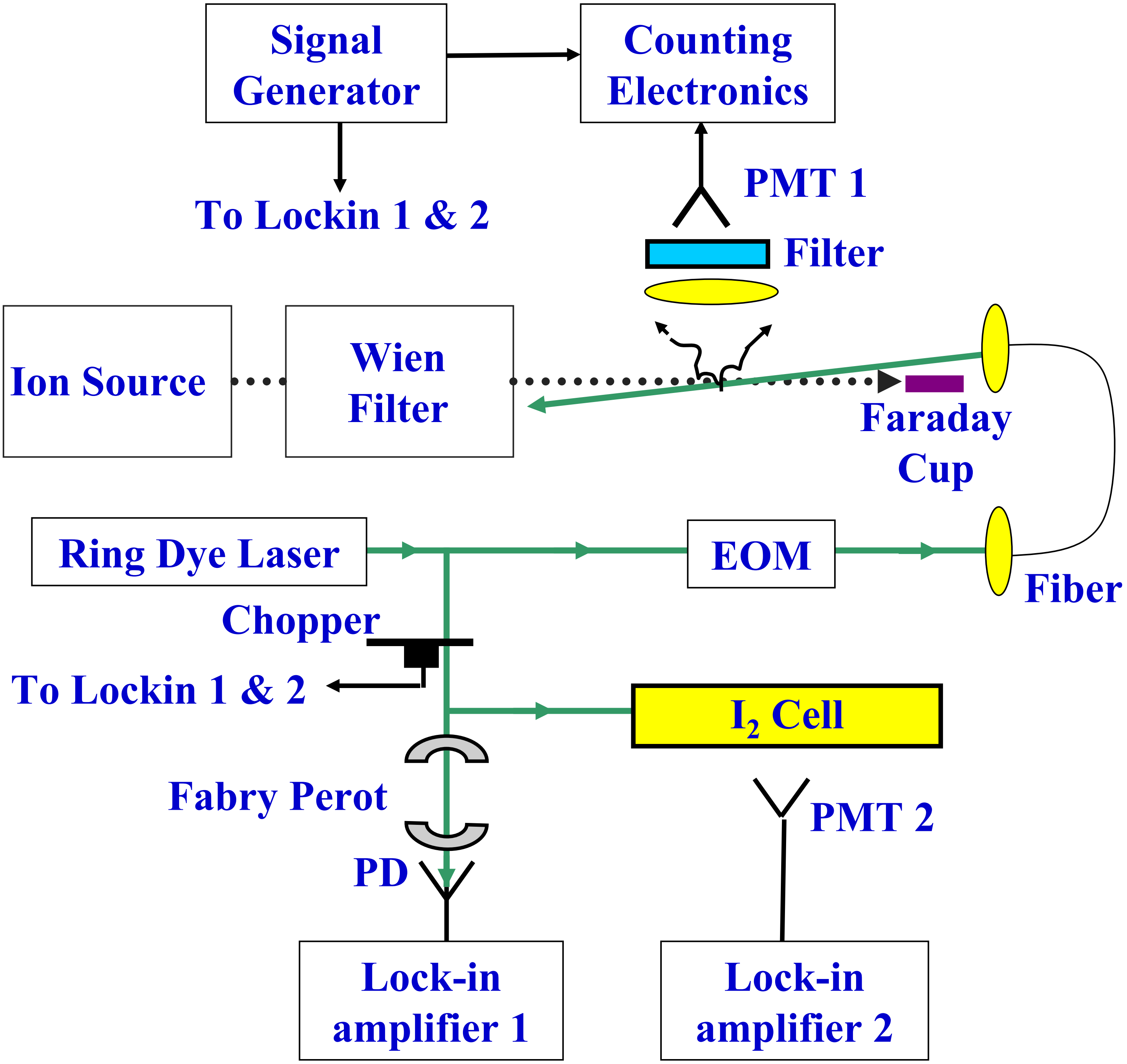
- Etalon peaks account for nonlinearity of laser scan
- Free spectral range found using EOM modulation frequency

Relevant Li$^+$ States & Hyperfine Levels

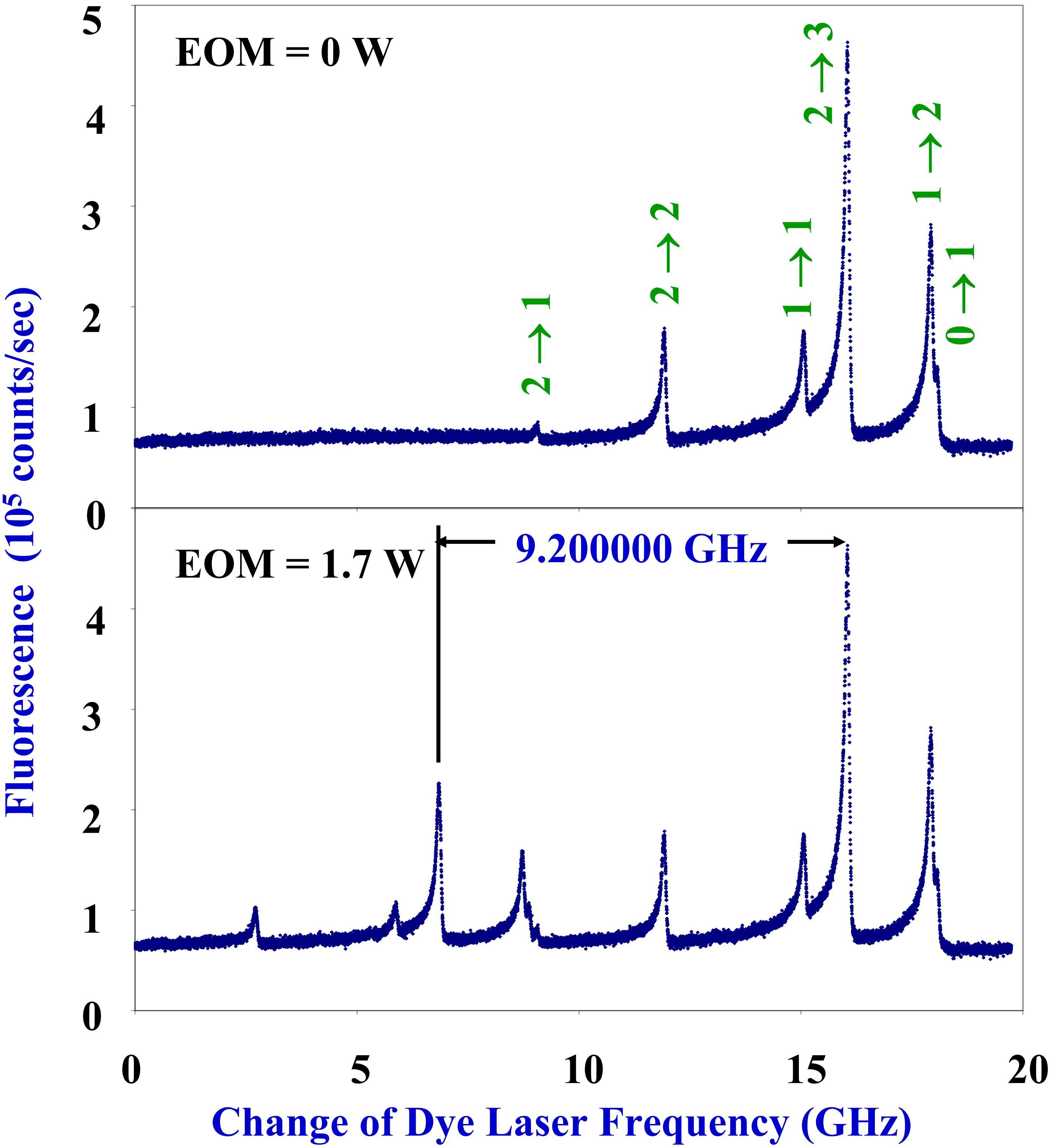


Apparatus for studying Li^+

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

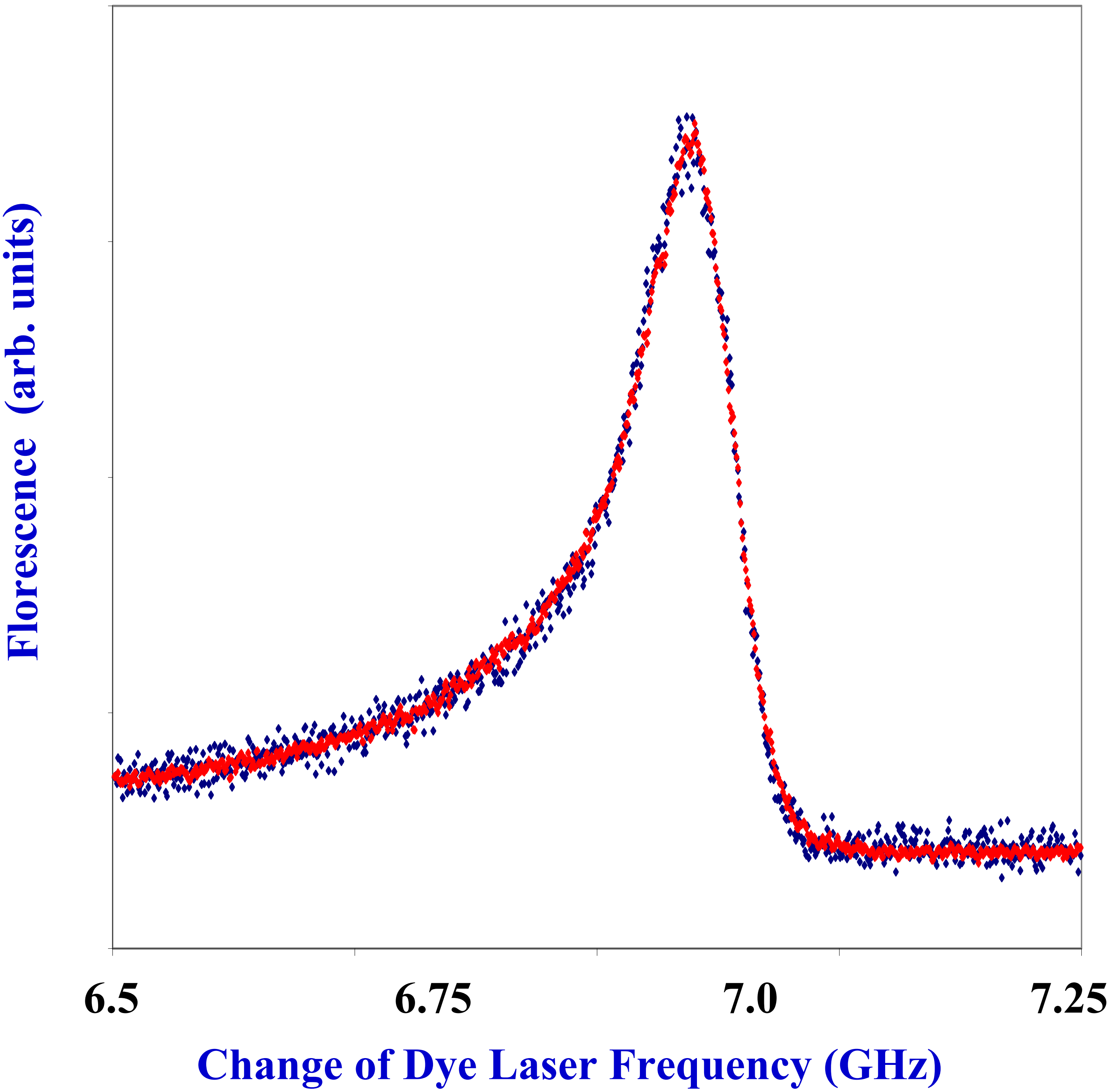


^{6}Li$^+$ 1s2s $^3S_1(F) \rightarrow$ 1s2p $^3P_2(F')$ with 9.2 GHz EOM



Determination of Frequency Intervals

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



^{6}Li$^+$ Hyperfine Intervals

State	Interval F → F'	Interval (MHz)	Technique
1s2s $^3S_1$	2 → 1	5,993 ± 6	Doppler Tuning[1]
		5,997 ± 4	Laser Scan / Etalon[2]
		6,003.600 ± 0.050	Microwave[3]
		6,003.66 ± 0.51	Our Expt[4]
		6,003.614 ± 0.024	H.V. Theory[5]
	1 → 0	2,998 ± 6	Doppler Tuning[1]
		2,998 ± 4	Laser Scan / Etalon[2]
		3,001.780 ± 0.050	Microwave[3]
		3,001.827 ± 0.47	Our Expt[4]
		3,001.765 ± 0.038	H.V. Theory[5]
1s2p $^3P_1$	2 → 1	2,888.98 ± 0.63	Our Expt[4]
		2,888.327 ± 0.029	H.V. Theory[5]
	1 → 0	1.316.06 ± 0.59	Our Expt[4]
		1,317.649 ± 0.046	H.V. Theory[5]
1s2p $^3P_2$	3 → 2	4,127.16 ± 0.76	Our Expt[4]
		4,127.882 ± 0.043	H.V. Theory[5]
	2 → 1	2,857.00 ± 0.72	Our Expt[4]
		2,858.002 ± 0.060	H.V. Theory[5]

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)

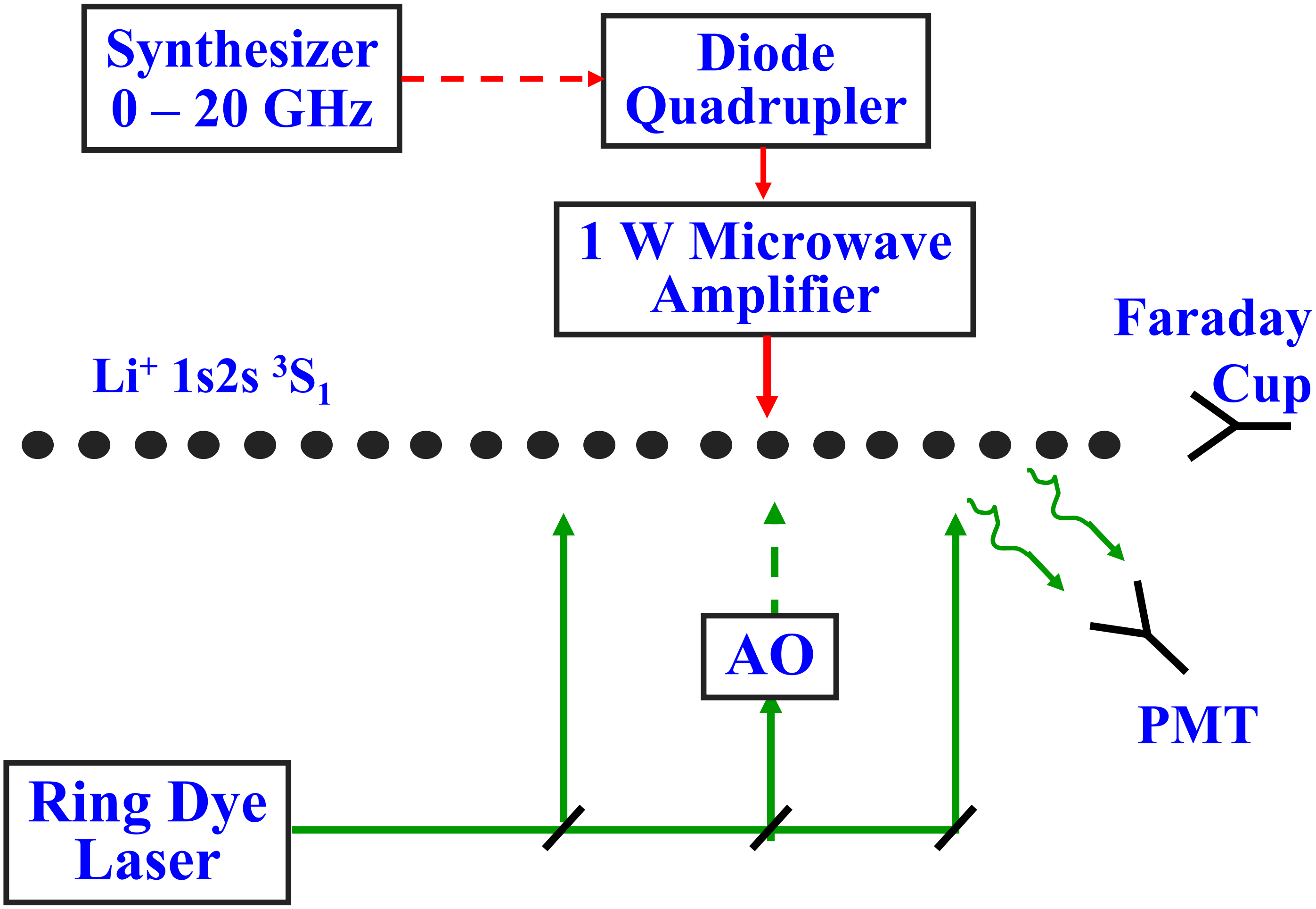
^{7}Li$^+$ 1s2p $^3P_{1\text{-}2}$ Fine Structure

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

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.



Synthesizer 0 – 20 GHz

Diode Quadrupler

1 W Microwave Amplifier

Faraday Cup

Li^+ 1s2s $^3S_1$

AO

PMT

Ring Dye Laser

Objective: Measure 1s2p ^3P Fine Structure

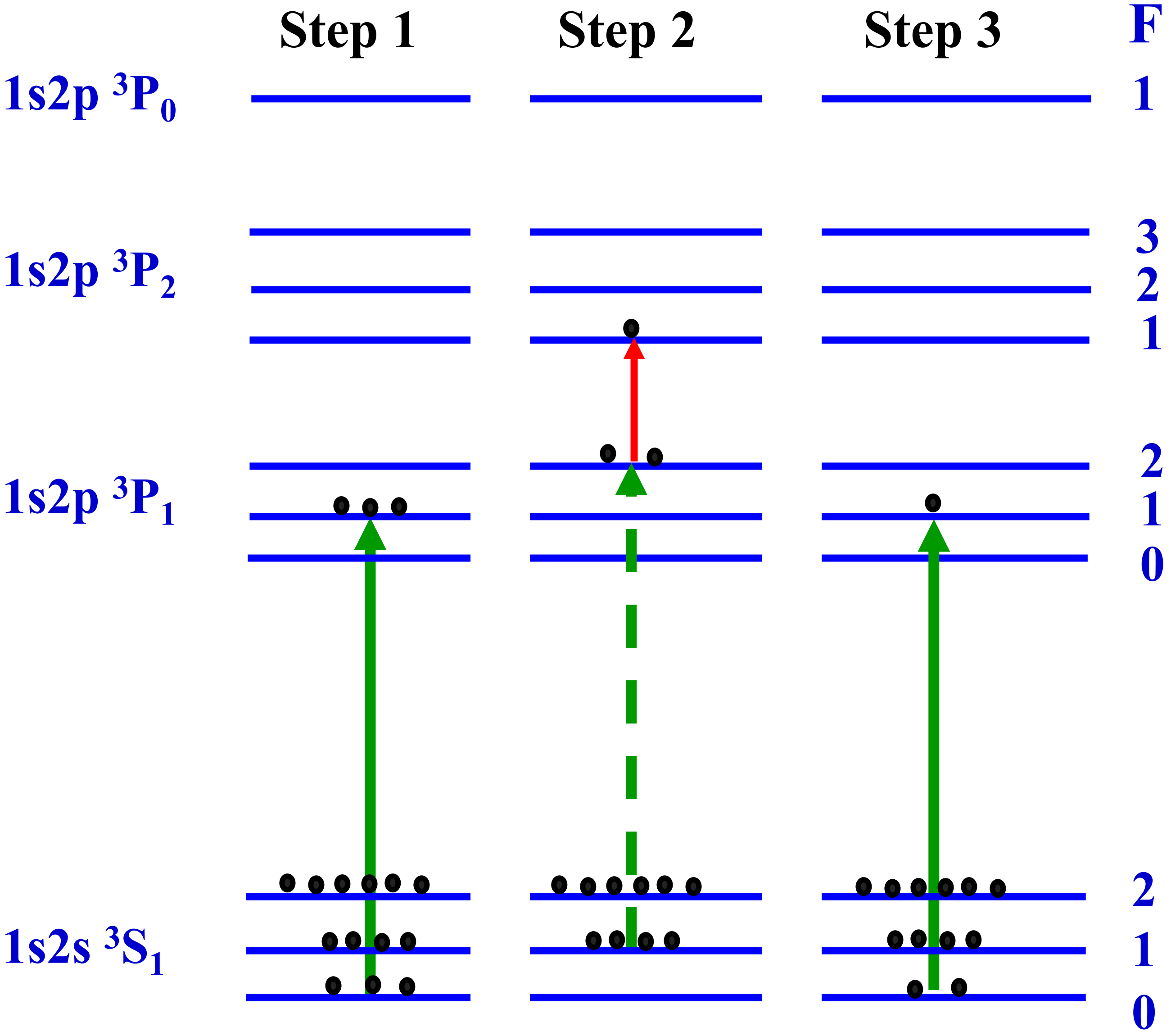
1s2p $^3P_{0\text{-}1}$ = 155.7 GHz

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

Natural linewidth of 1s2p ^3P state = 3.7 MHz

⇒ 0.1% measurement of $^3P_{1\text{-}2}$ interval gives fine structure to one part in 2×10^7

$^6Li^+$ Hyperfine Level Populations



Procedure

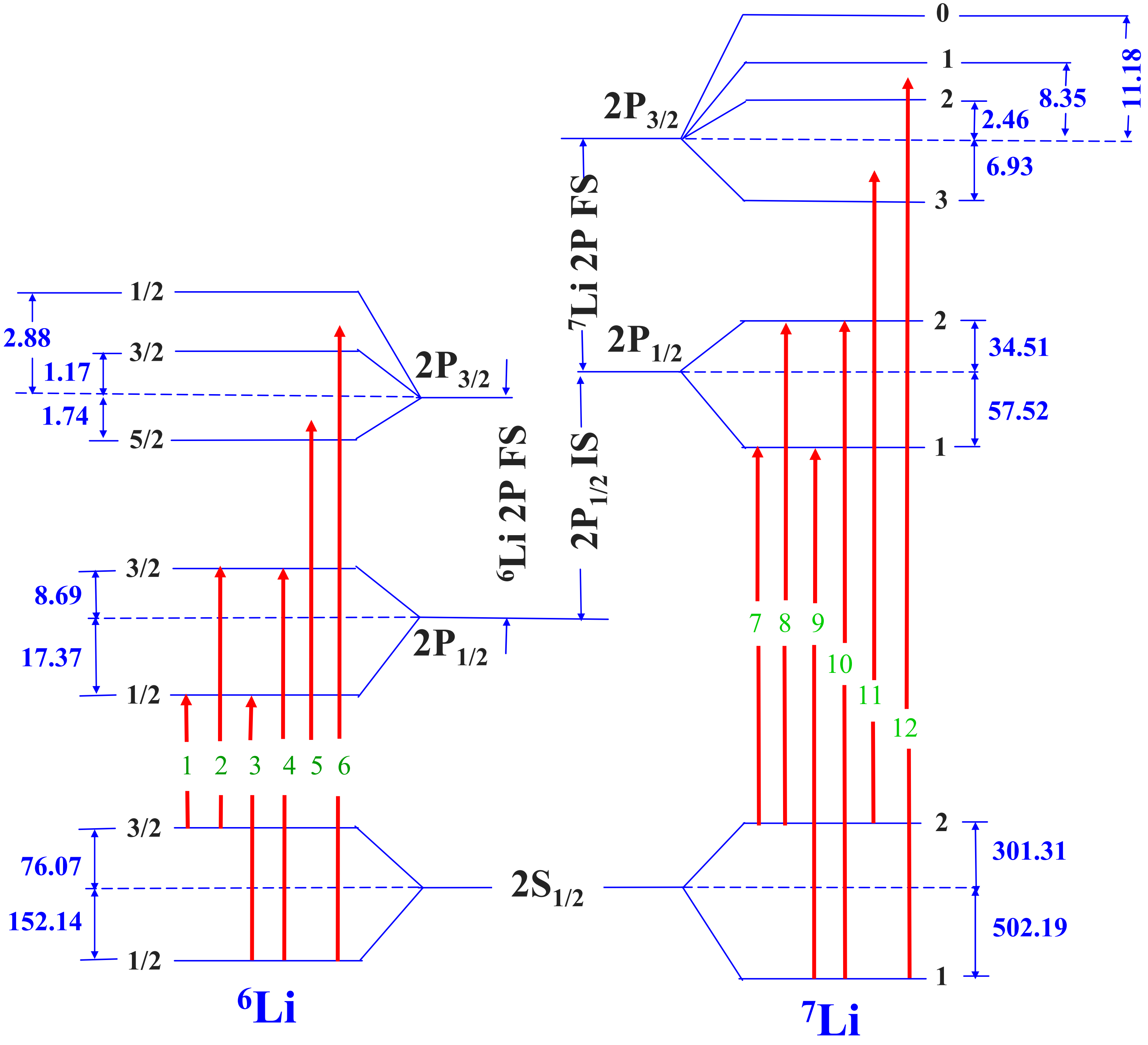
1) Optical Pumping depletes 1s2s $^3S_1$ (F=0) level.

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)



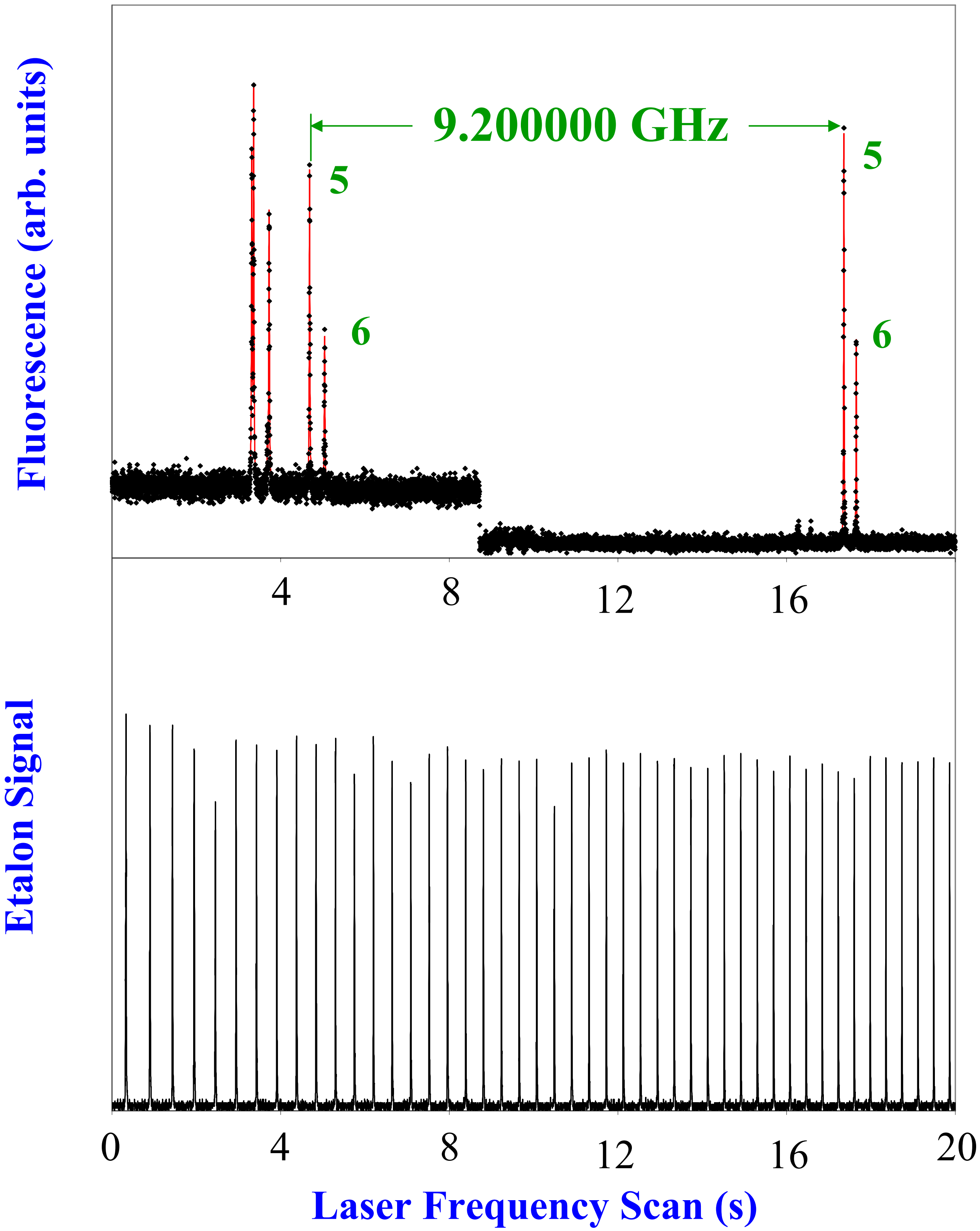
Study of Li D Transitions

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_{Earth}

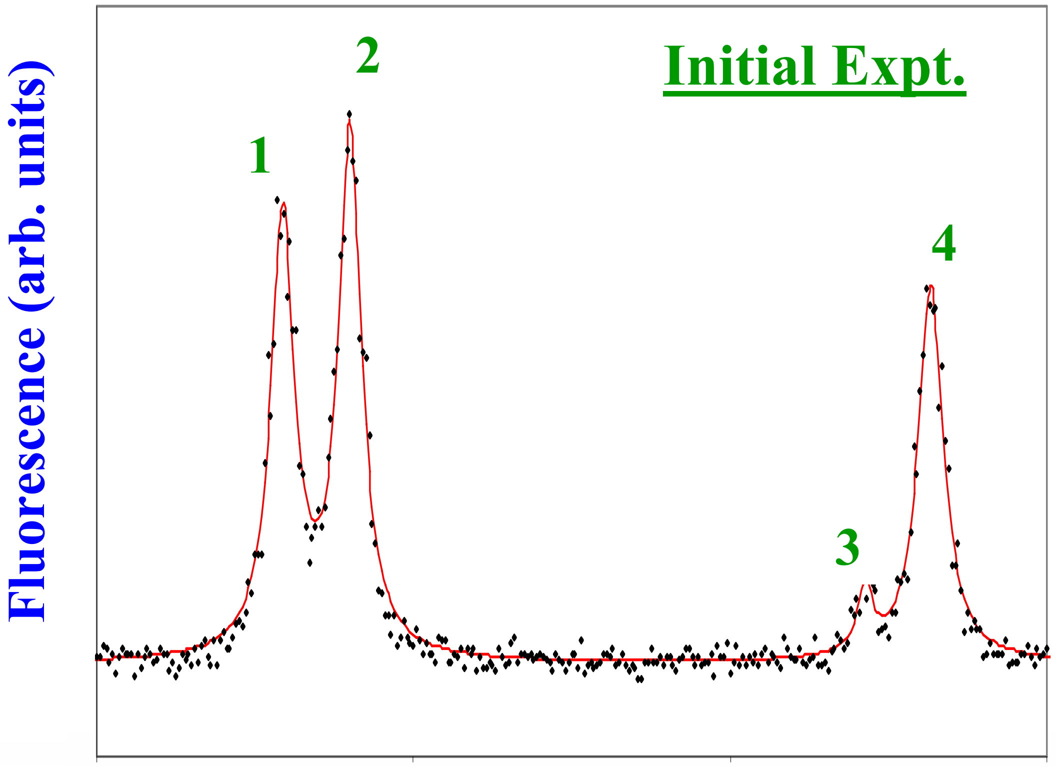
1. J. Walls et al, EPJ D **22,** 159 (2003)
2. G. Noble et al, PRA **74** 012502 (2006)

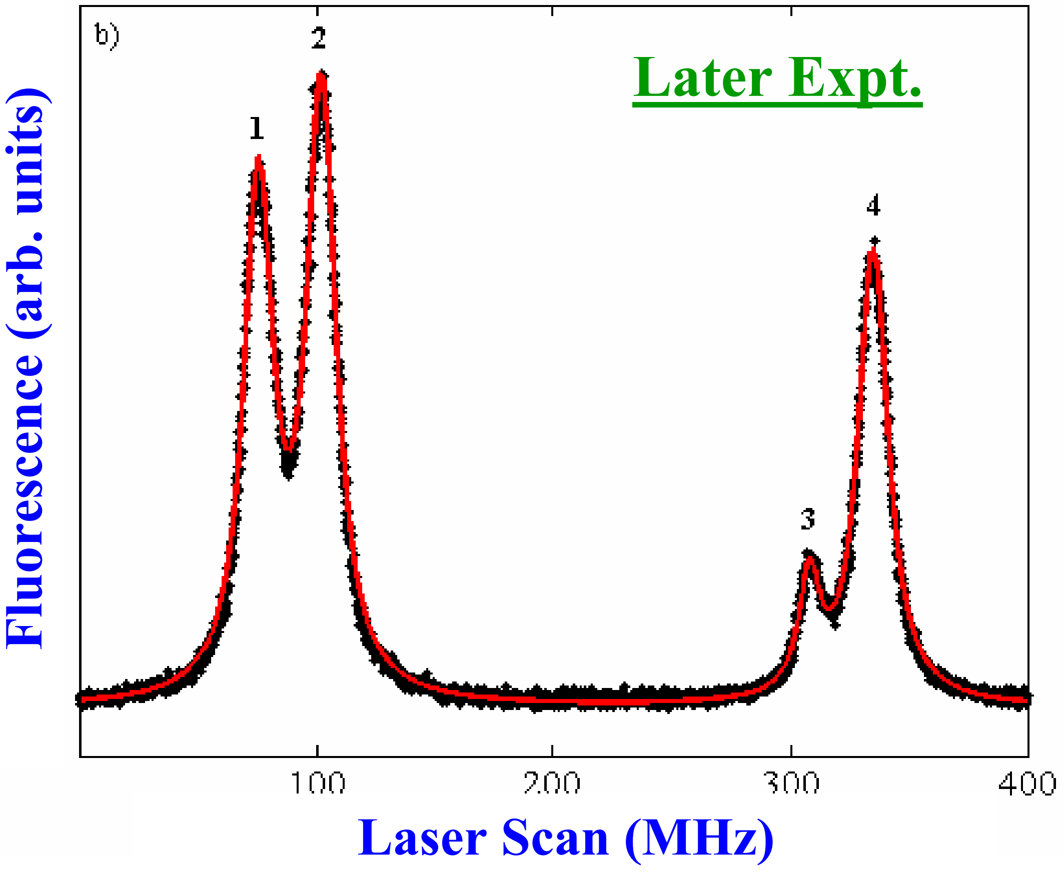
Excitation of ^{6}Li D Lines

Data from Initial Expt.

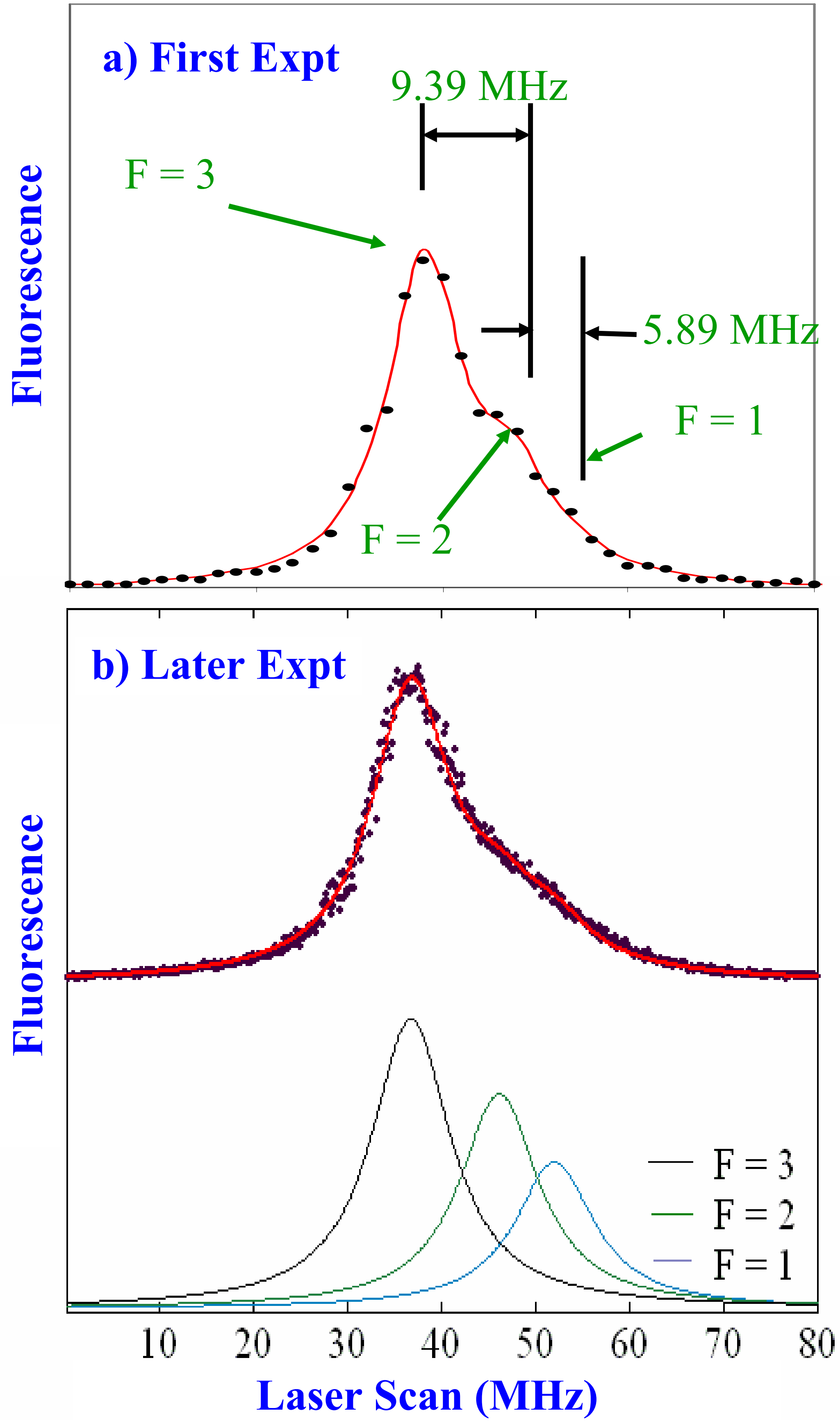


Excitation of ^{6}Li D Lines

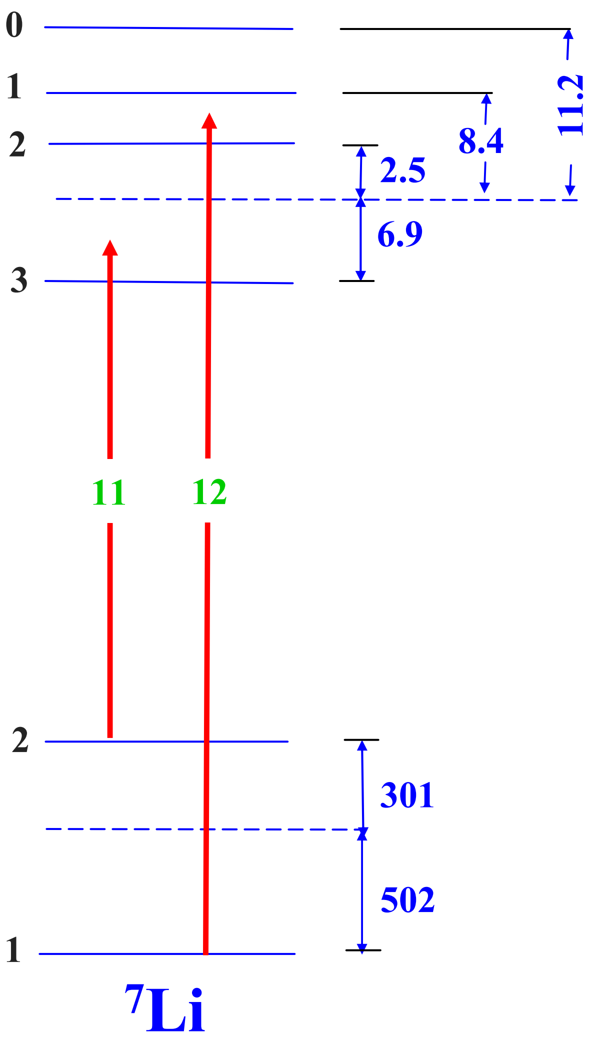




Peak 11: ^{7}Li 2S$_{1/2}$ (F=2)$\rightarrow$2P$_{3/2}$ Excitation

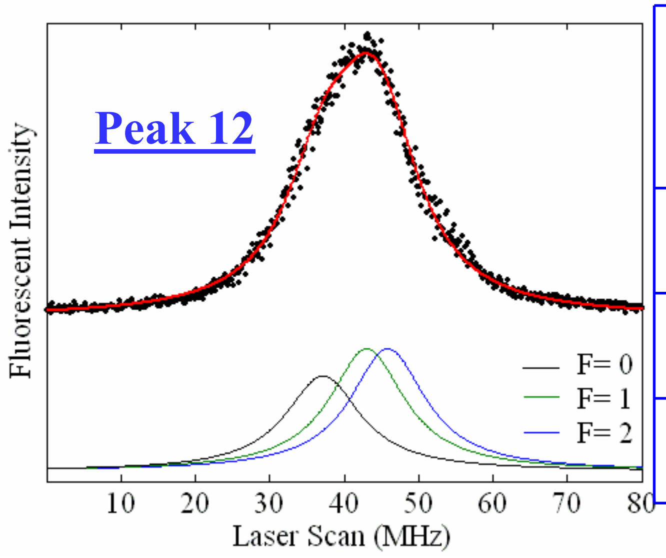


Determining Line Center to better than Natural Linewidth



$2P_{3/2}$ Hyperfine Spacing ≤ 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}Li	$2S_{1/2}$	152.136839	Magnetic Resonance[1]
		152.109 ± 0.043	Our Expt[2]
	$2P_{1/2}$	17.375 ± 0.018	Opt. Double Resonance[3]
		17.8 ± 0.3	Level Crossing[4]
		16.8 ± 0.7	Laser Atomic Beam[5]
		17.386 ± 0.031	Our Expt[2]
^{7}Li	$2S_{1/2}$	401.7520433	Magnetic Resonance[1]
		401.767 ± 0.039	Our Expt[2]
	$2P_{1/2}$	45.914 ± 0.025	Opt Double Resonance[3]
		46.05 ± 0.30	Laser Atomic Beam[5]
		46.010 ± 0.025	Our Expt[2]
		45.793	Full Core + Correlation[6]
		45.984 ± 0.007	MCHF[7]
		45.945	MCHF + rel. corr.[8]

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

	Interval (MHz)	Technique
^{6}Li	10052.76 ± 0.22	Level Crossing[1]
	10051.62 ± 0.20	Laser Atomic Beam[2]
	10052.964 ± 0.050	Our Expt[3]
	10050.846 ± 0.012	H.V. Theory[5]
^{7}Li	10053.24 ± 0.22	Level Crossing[1]
	10053.184 ± 0.058	Optical Double Resonance[4]
	10053.4 ± 0.2	Laser Atomic Beam[2]
	10053.119 ± 0.058	Our Expt[3]
	10051.214 ± 0.012	H.V. Theory[5]

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}Li Relative Nuclear Charge Radius

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

Reference	**Transition**	**$\delta\nu$ (MHz)**	**Δr_c^2 (fm^2)**
E. Riis et al[1]	$\mathrm{Li}^+\ 2\,^3S_1 - 2\,^3P_0$	34747.73 ± 0.55	0.78
	$\mathrm{Li}^+\ 2\,^3S_1 - 2\,^3P_1$	34747.46 ± 0.67	0.78
	$\mathrm{Li}^+\ 2\,^3S_1 - 2\,^3P_2$	34748.91 ± 0.62	0.64
W. Scherf et al[2]	$\mathrm{Li}\ 2\,^2S_{1/2} - 2\,^2P_{1/2}$	10533.13 ± 0.15	0.39
	$\mathrm{Li}\ 2\,^2S_{1/2} - 2\,^2P_{3/2}$	10534.93 ± 0.15	0.96
GSI Group[3,4]	$\mathrm{Li}\ 2\,^2S_{1/2} - 3\,^2S_{1/2}$	11453.95 ± 0.13	0.60
	$\mathrm{Li}\ 2\,^2S_{1/2} - 3\,^2S_{1/2}$	11453.734 ± 0.030	0.46
	$\mathrm{Li}\ 2\,^2S_{1/2} - 2\,^2P_{1/2}$	10533.160 ± 0.068	0.40
D. Das et al[5]	$\mathrm{Li}\ 2\,^2S_{1/2} - 2\,^2P_{1/2}$	10534.215 ± 0.039	0.83
	$\mathrm{Li}\ 2\,^2S_{1/2} - 2\,^2P_{3/2}$	10533.352 ± 0.068	0.32
This Work[6]	$\mathrm{Li}\ 2\,^2S_{1/2} - 2\,^2P_{1/2}$	10534.039 ± 0.070	0.76
	$\mathrm{Li}\ 2\,^2S_{1/2} - 2\,^2P_{3/2}$	10534.194 ± 0.104	0.66
Nuclear Theory[7]			0.74
Elect. Scatt.[8]			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

Li^+ 1s2s ^3S - 1s2p ^3P Transition

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

Li D Lines

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

Ongoing Work

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