High Resolution Laser Spectroscopy

$$: National Science Council, Taiwan
國科會

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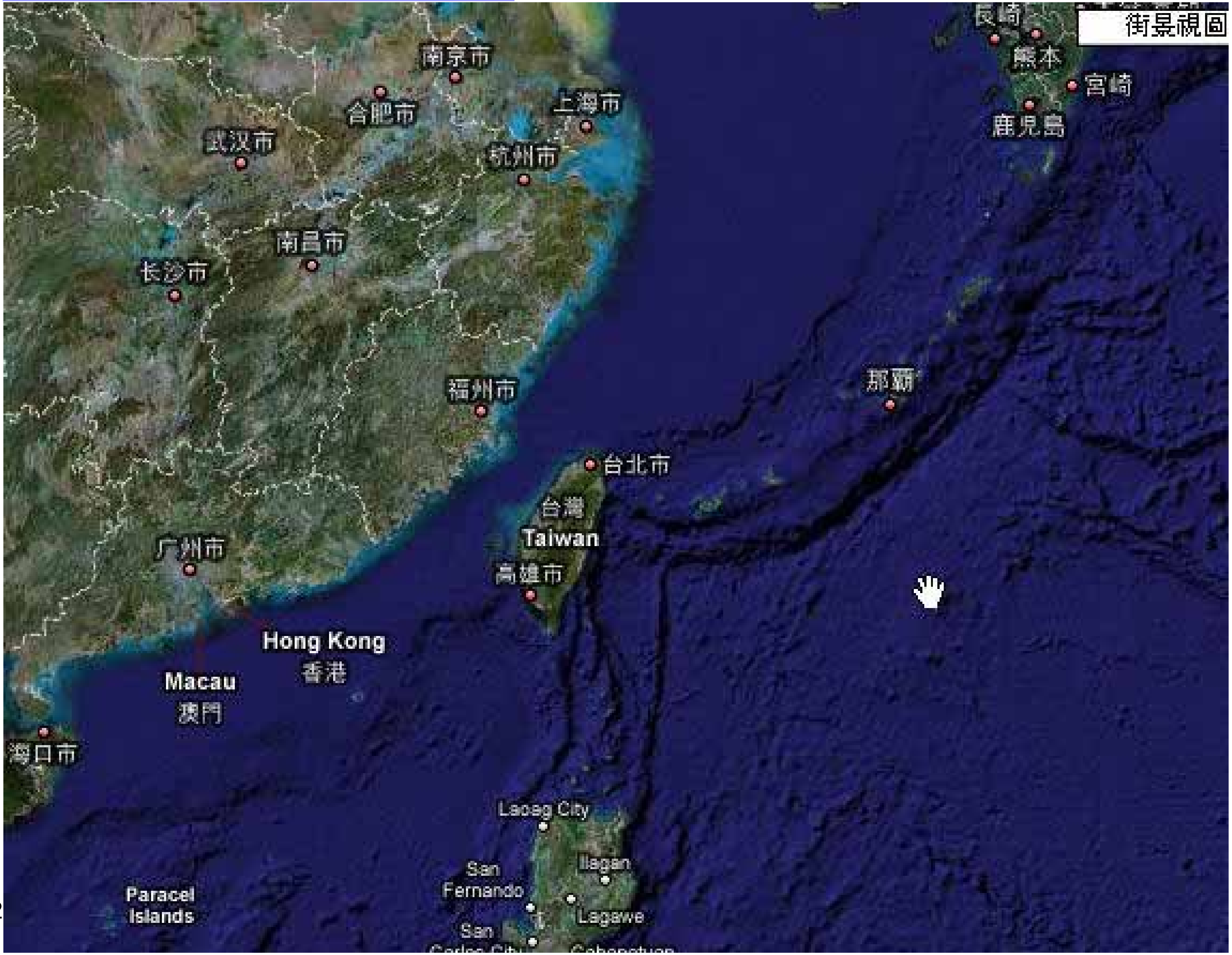
Taiwan from google map



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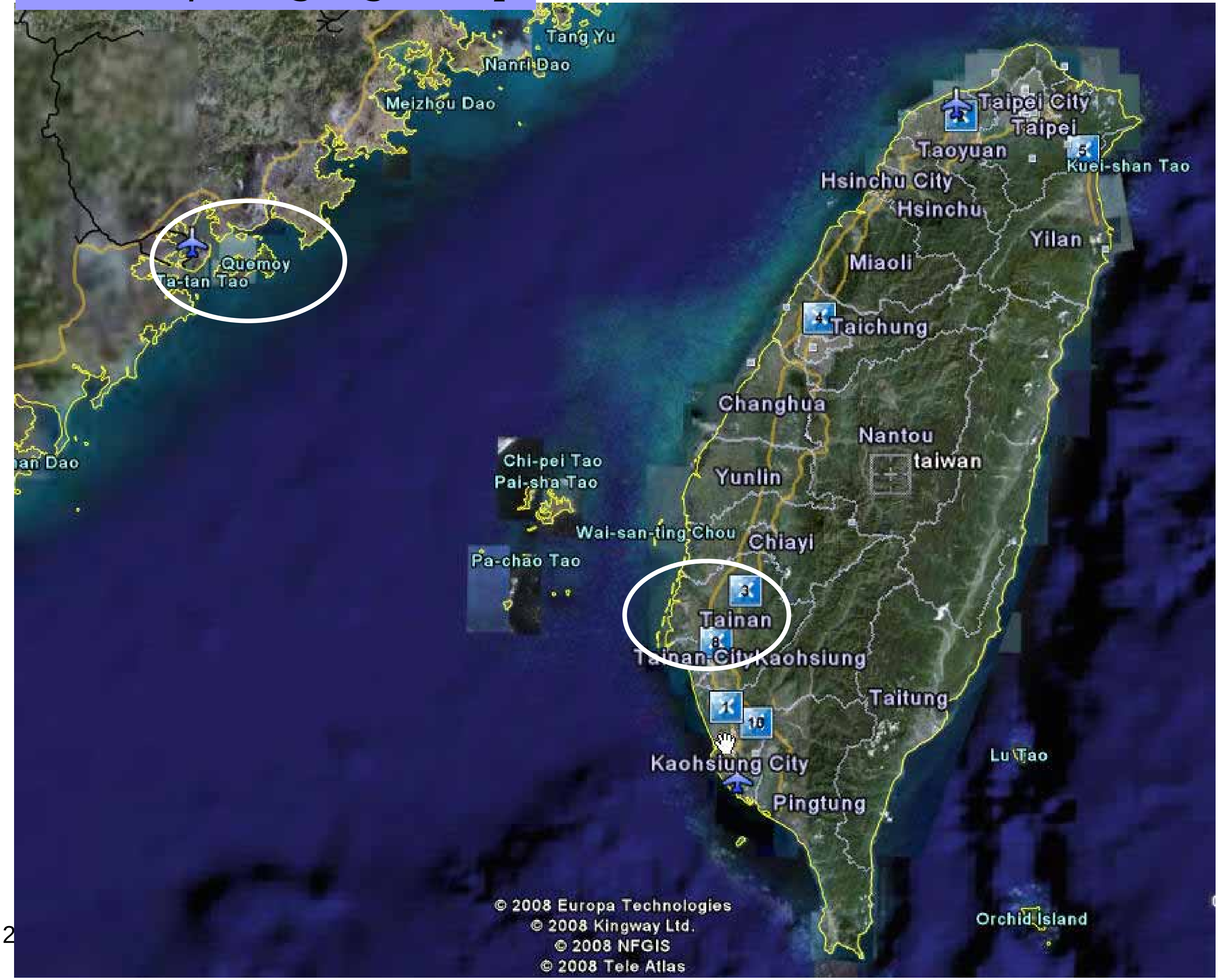
Taiwan from google map



Taiwan from google map

What is spectroscopy?

Intensity

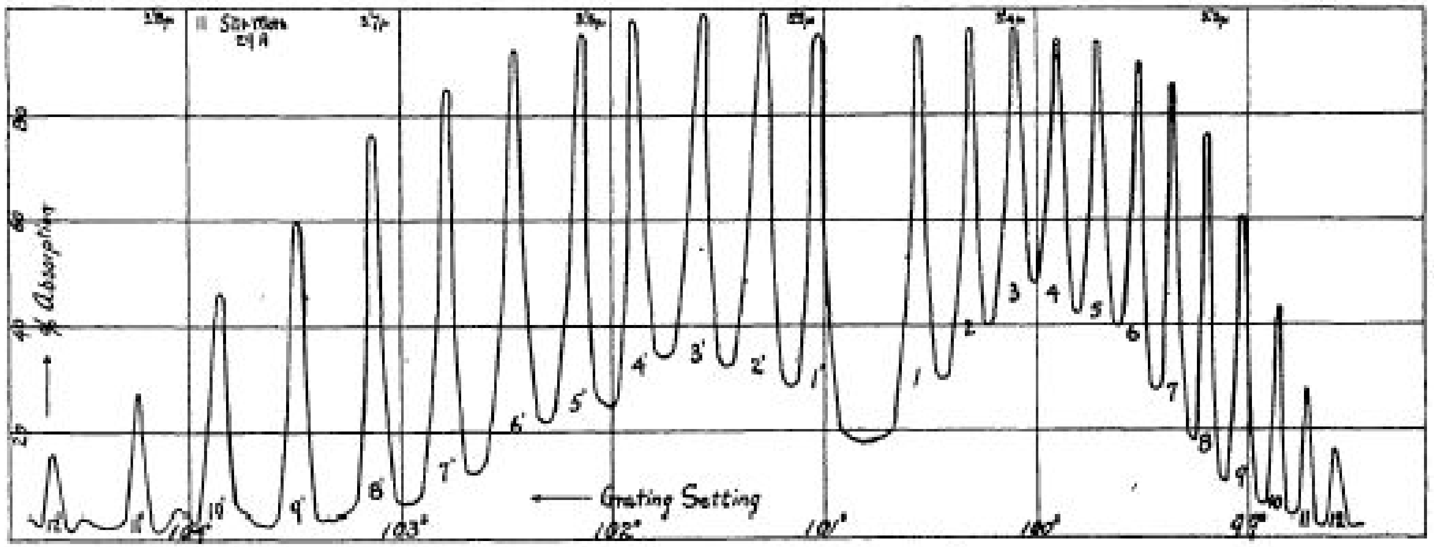


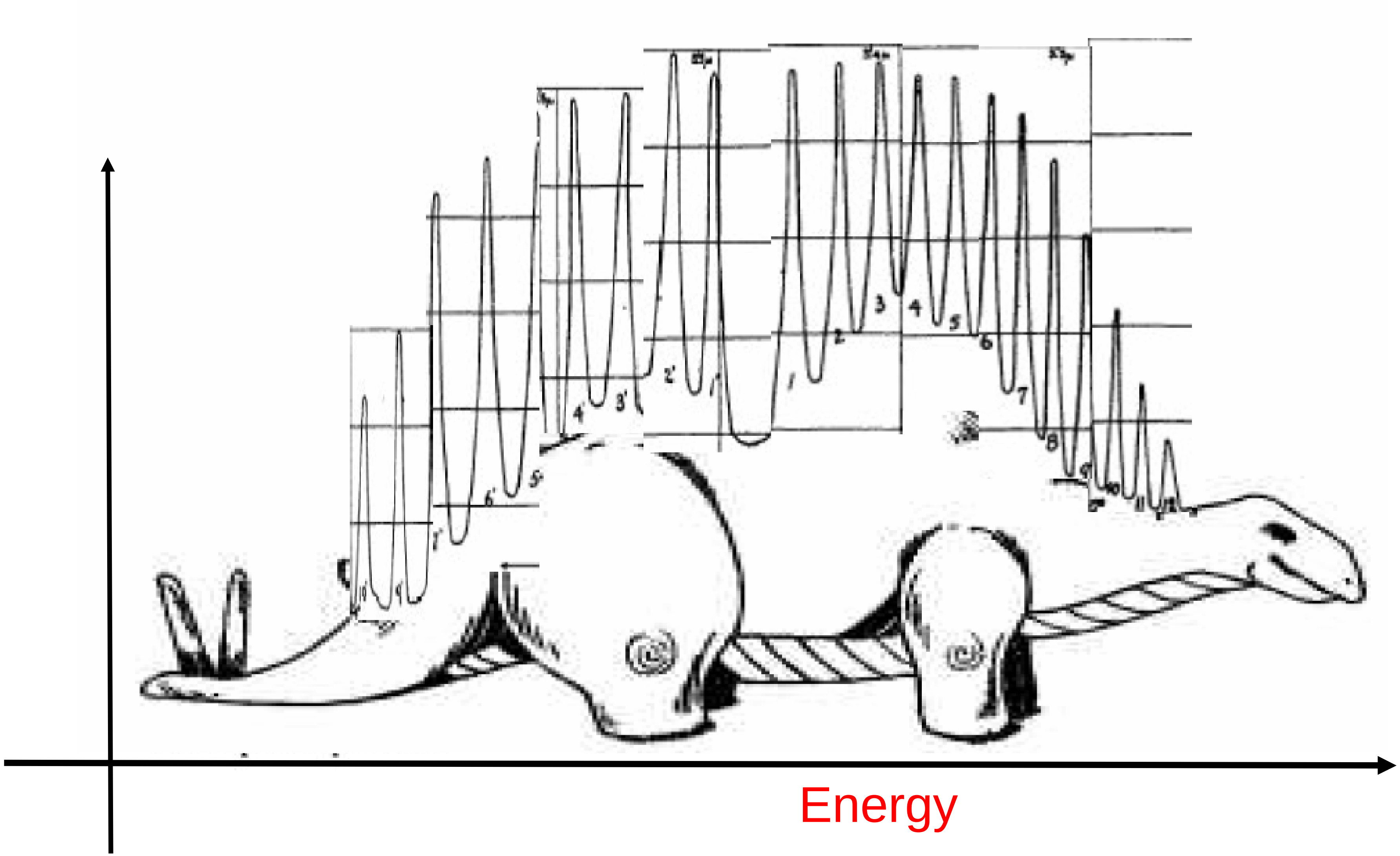
FIG. 3.—The HCl band at 3.46 μ, mapped with 7500-line grating. HCl at atmospheric pressure.

Energy

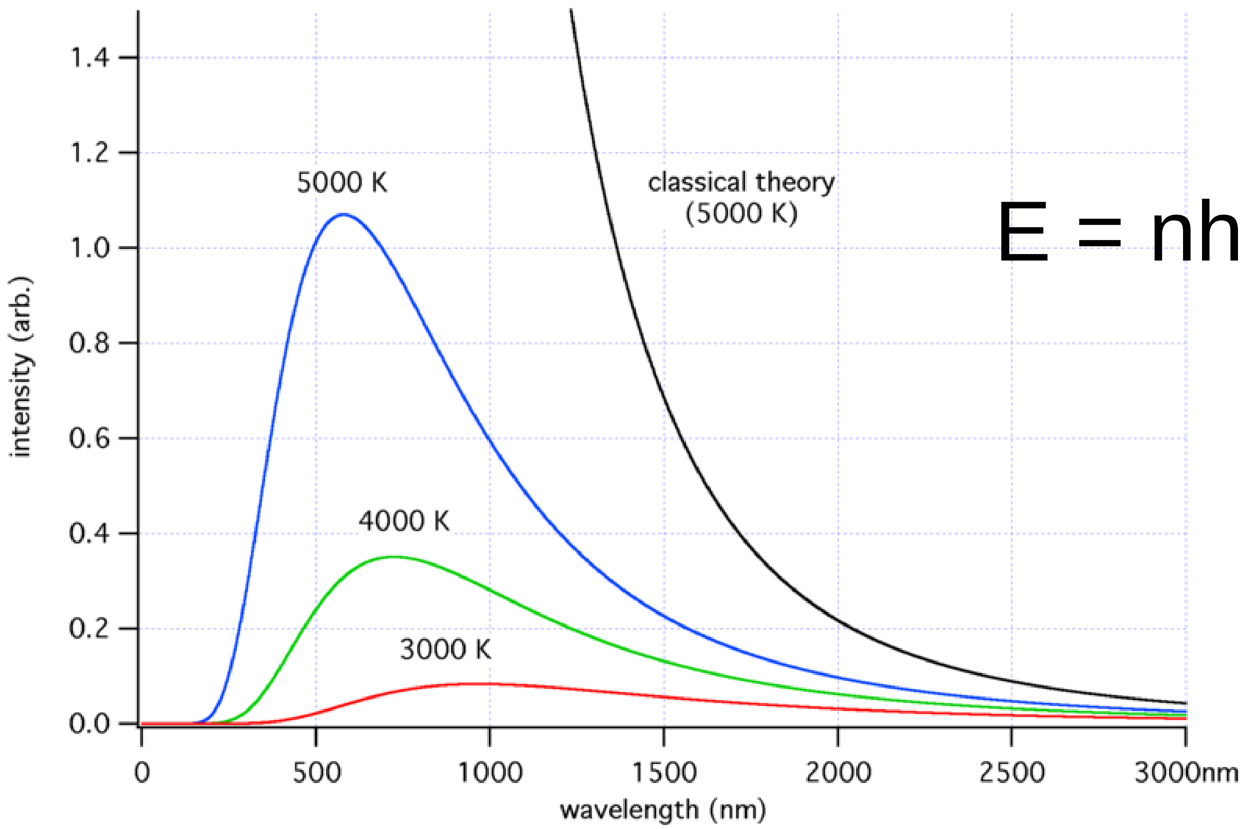
What are the important issues?

Line position, Intensity and Shape

When/Where does it start?



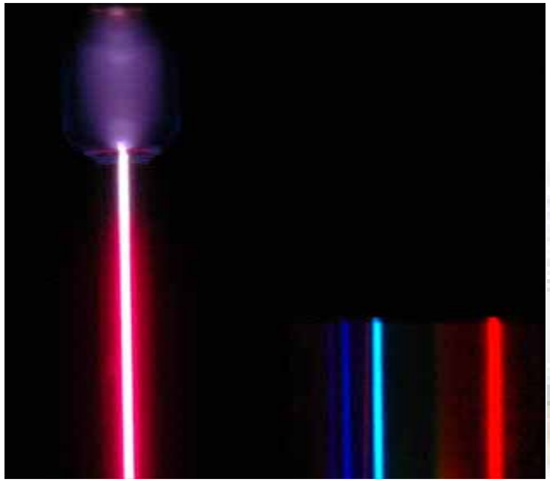
Black body radiation

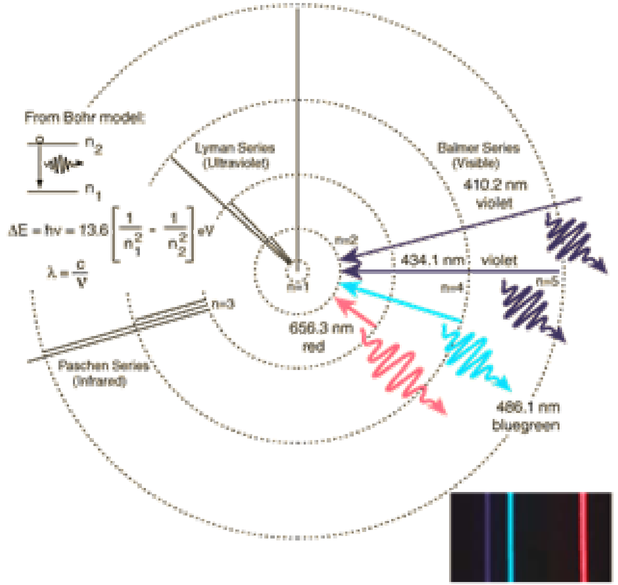


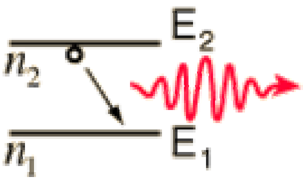
$E = nh\nu$

The dawn of Quantum Mechanics!

High resolution laser spectroscopy







A downward transition involves emission of a photon of energy:

$$E_{photon} = h\upsilon = E_2 - E_1$$

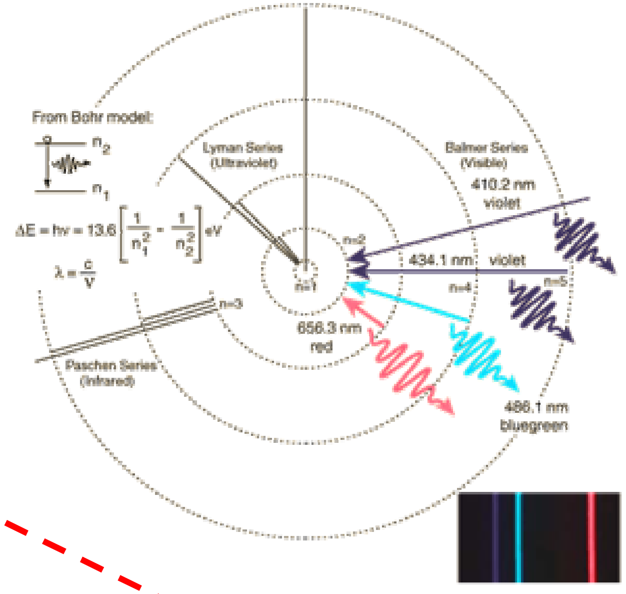
Given the expression for the energies of the hydrogen electron states:

$$h\upsilon = \frac{2\pi^2 me^4}{h^2}\left[\frac{1}{n_1^2} - \frac{1}{n_2^2}\right] = -13.6\left[\frac{1}{n_1^2} - \frac{1}{n_2^2}\right] \text{eV}$$

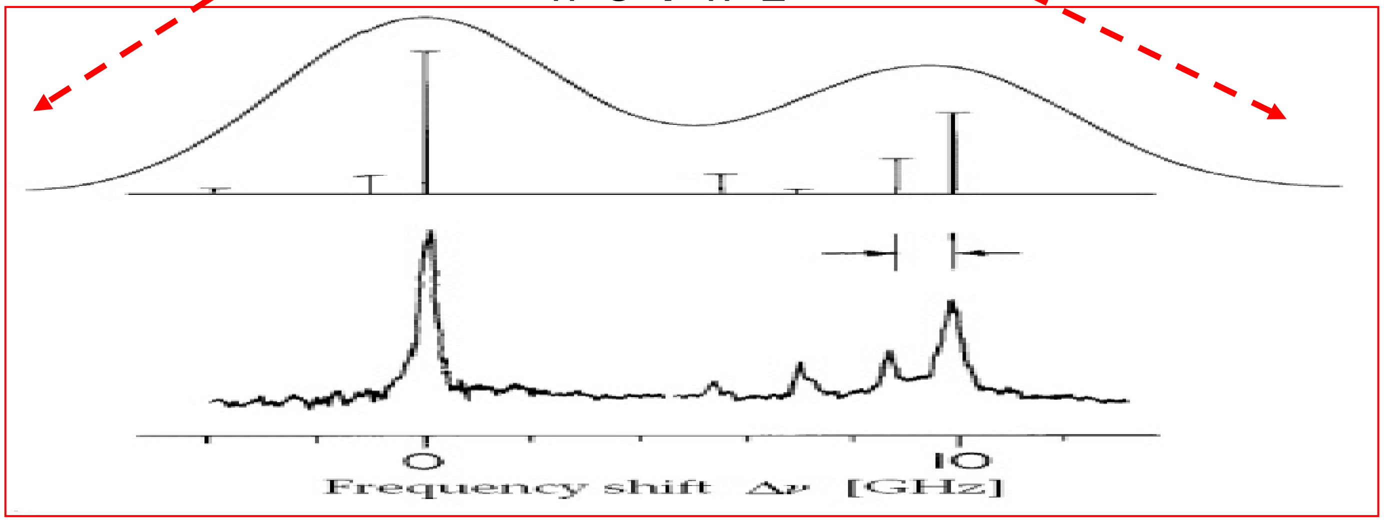
Bohr Model

$L = n\hbar$

High resolution laser spectroscopy



H a line
n=3 → n=2



Schrodinger Equation : $H\Psi = E\Psi$ → Wavefunctions and Eigenvalues

$$E = -\frac{Z^2 m e^4}{8 n^2 h^2 \varepsilon_0^2} = -\frac{13.6 Z^2}{n^2}\, eV$$

$$r = \frac{n^2 h^2 \varepsilon_0}{Z \pi m e^2} = \frac{n^2 a_0}{Z}$$

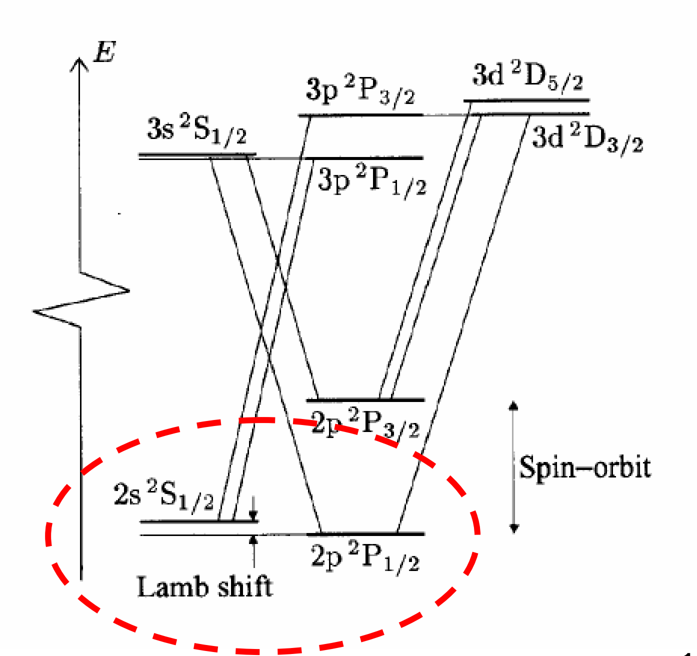
a_0 = 0.529 Å = Bohr radius

Spin Orbital interactions

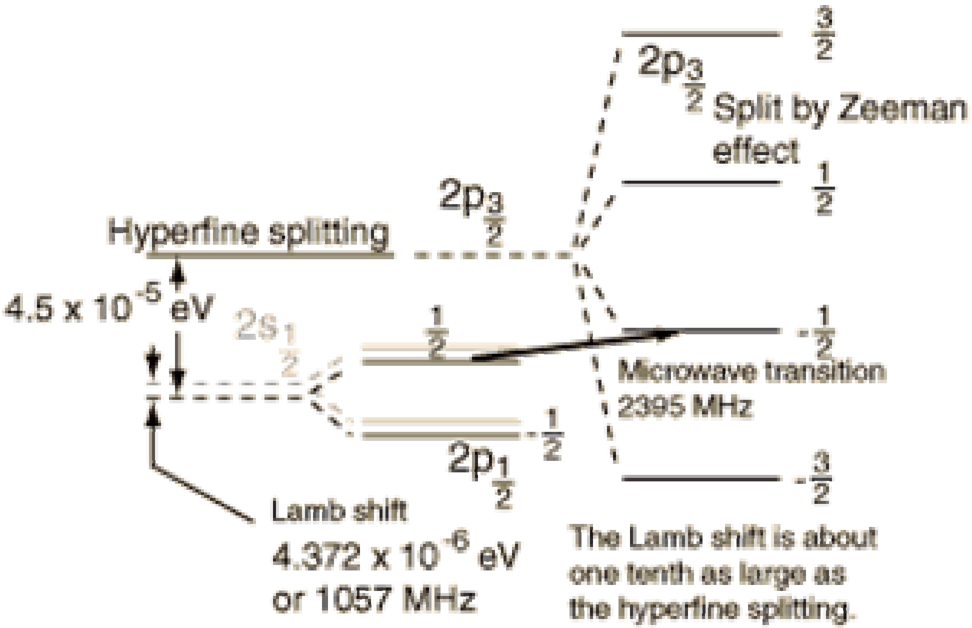
Transition Probabilities, Selection Rules

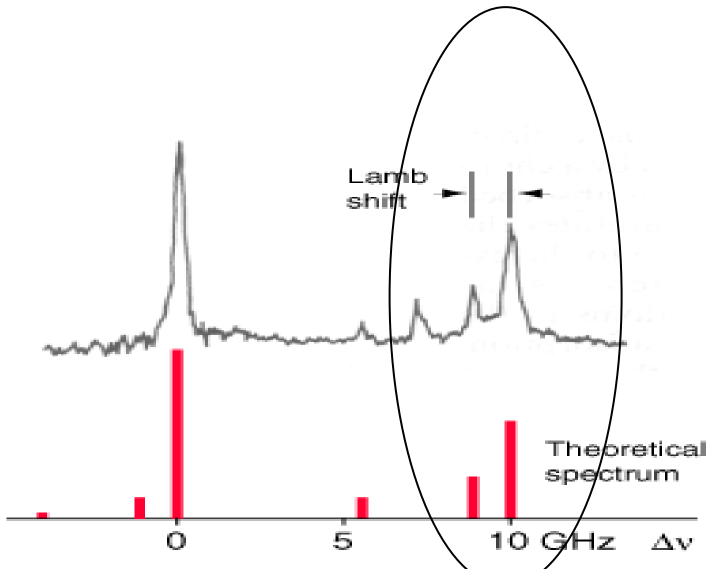
E (n,j)

2s $^2S_{1/2}$, 2p $^2P_{1/2}$ are degenerate.



Lamb Shift → QED

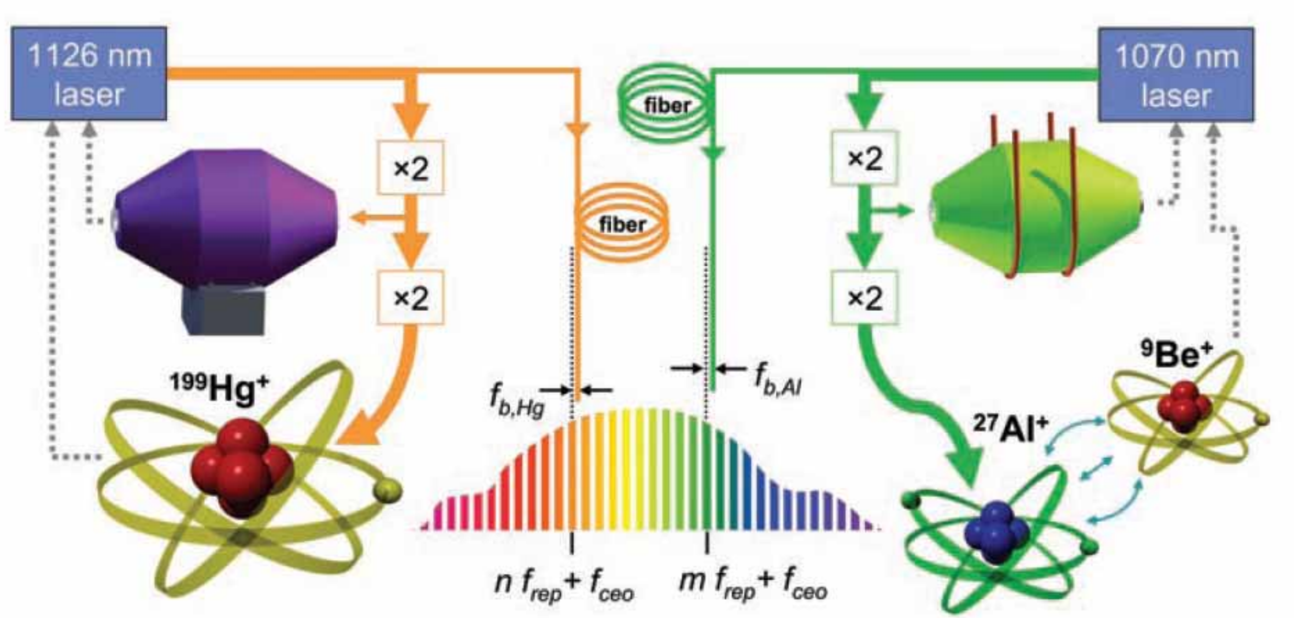


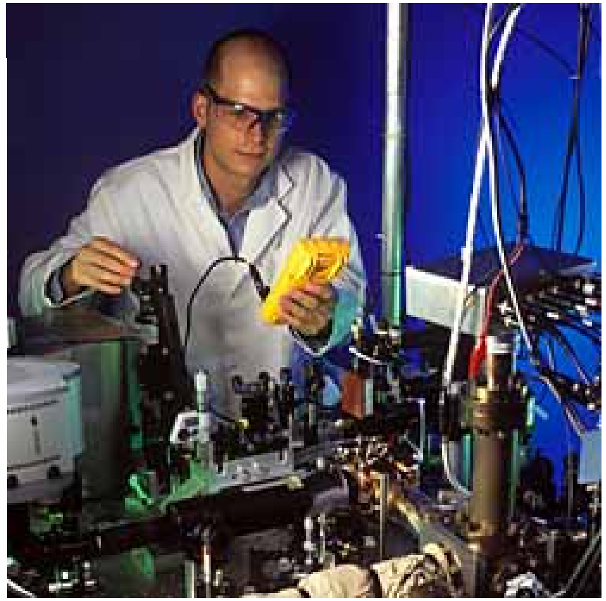


Hydrogen fine structure and hyperfine structure for the n=3 → 2 transition.
(After Ohanian, Modern Physics, Ch 7., spectrum from T. W. Hansch, Stanford Univ.)

It provided a high precision verification of theoretical calculations made with the quantum theory of electrodynamics.

NIST ‘Quantum Logic Clock’





The ratio of aluminum and mercury single-ion optical clock frequencies

ν_{Al^+} / ν_{Hg^+} is 1.052871833148990438(55),

5.2×10^{-17}

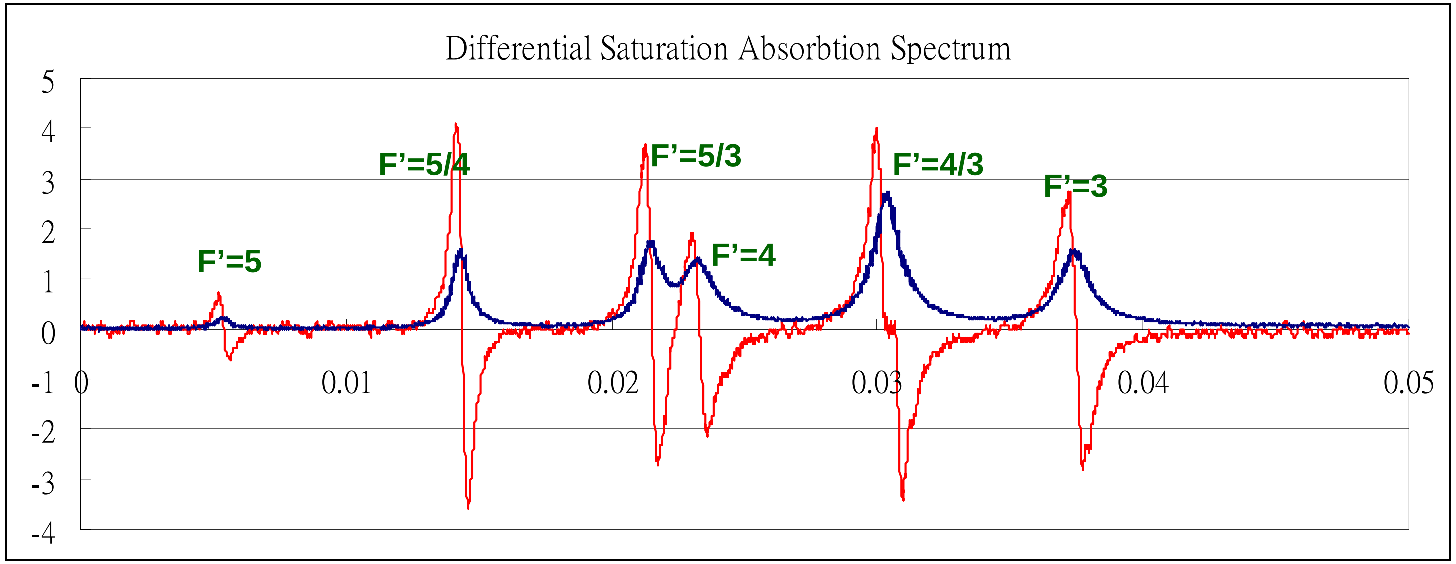
Quantum Phenomena of atoms

Atomic Spectroscopy

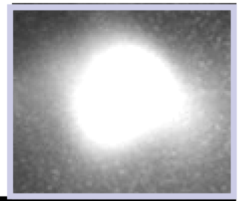
High-Precision and High-Resolution Laser Spectroscopy on Magneto-Optical Trap of Cesium Atoms

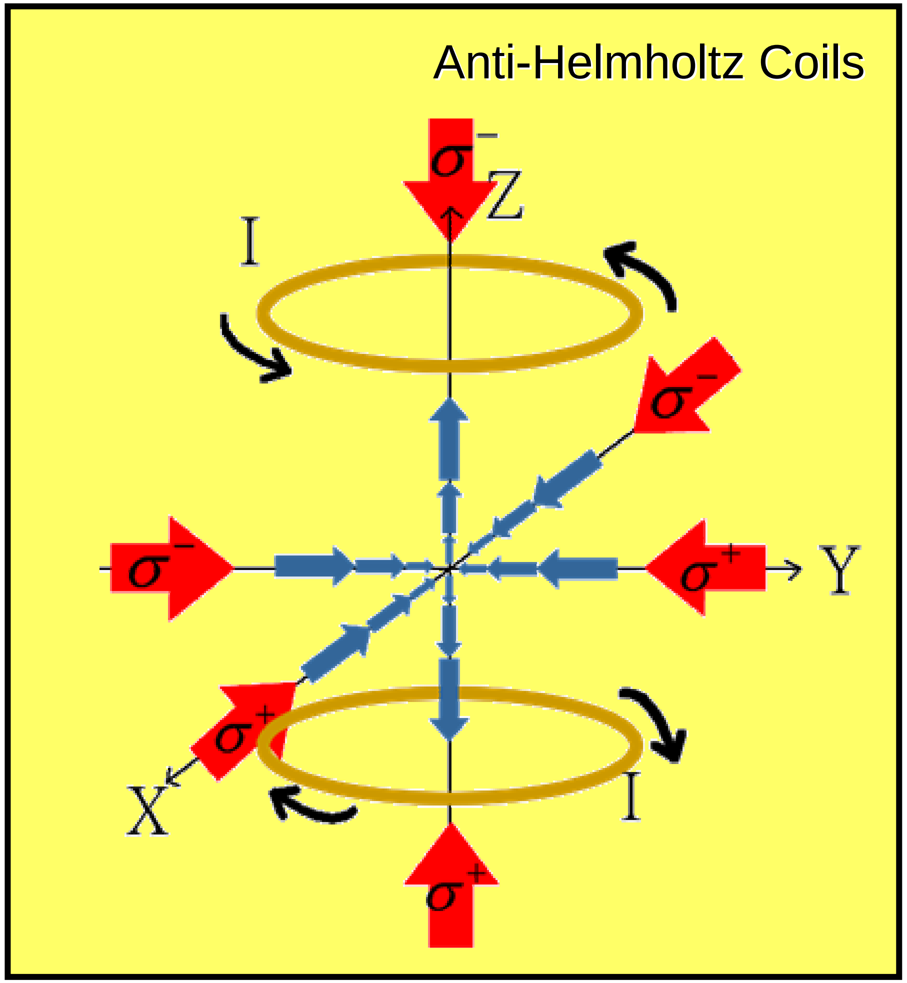
Atom number $4x10^9$, Cloud size 5 mm, Density $5x10^{10}/cm^3$

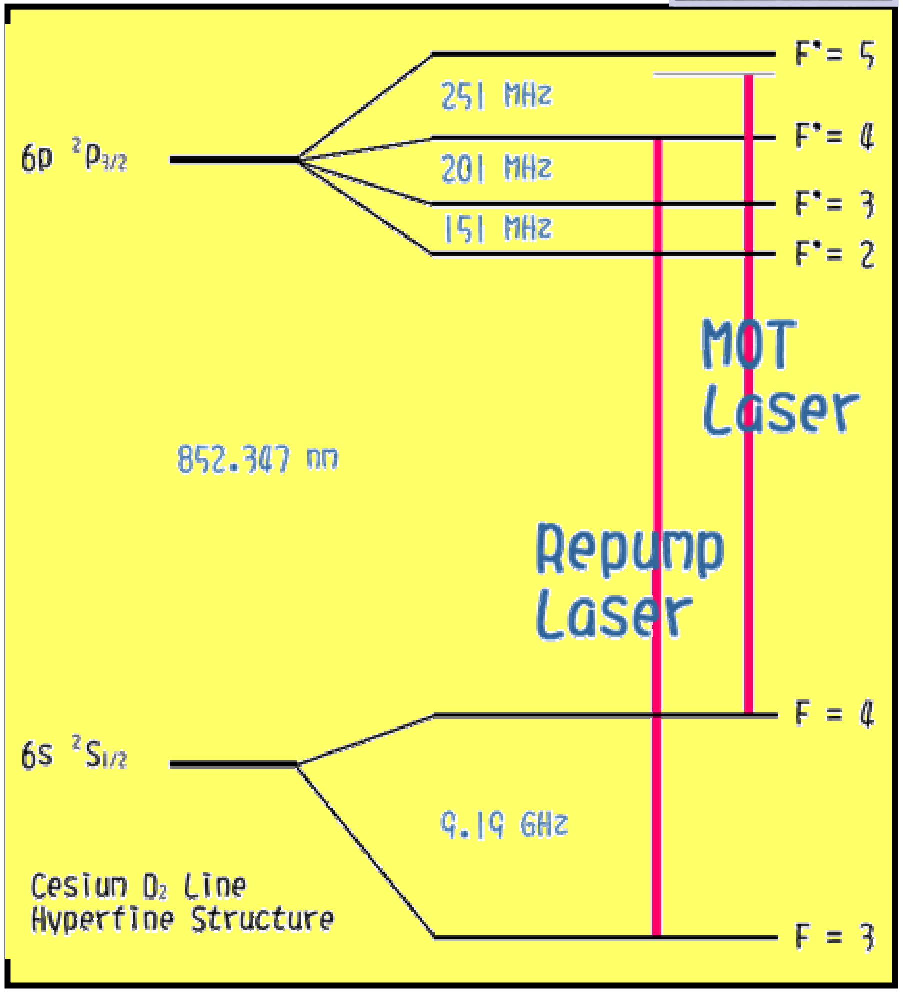
Atom temperature (Time of flight) : 100 μK



Magneto-Optical Trap of Cesium atoms

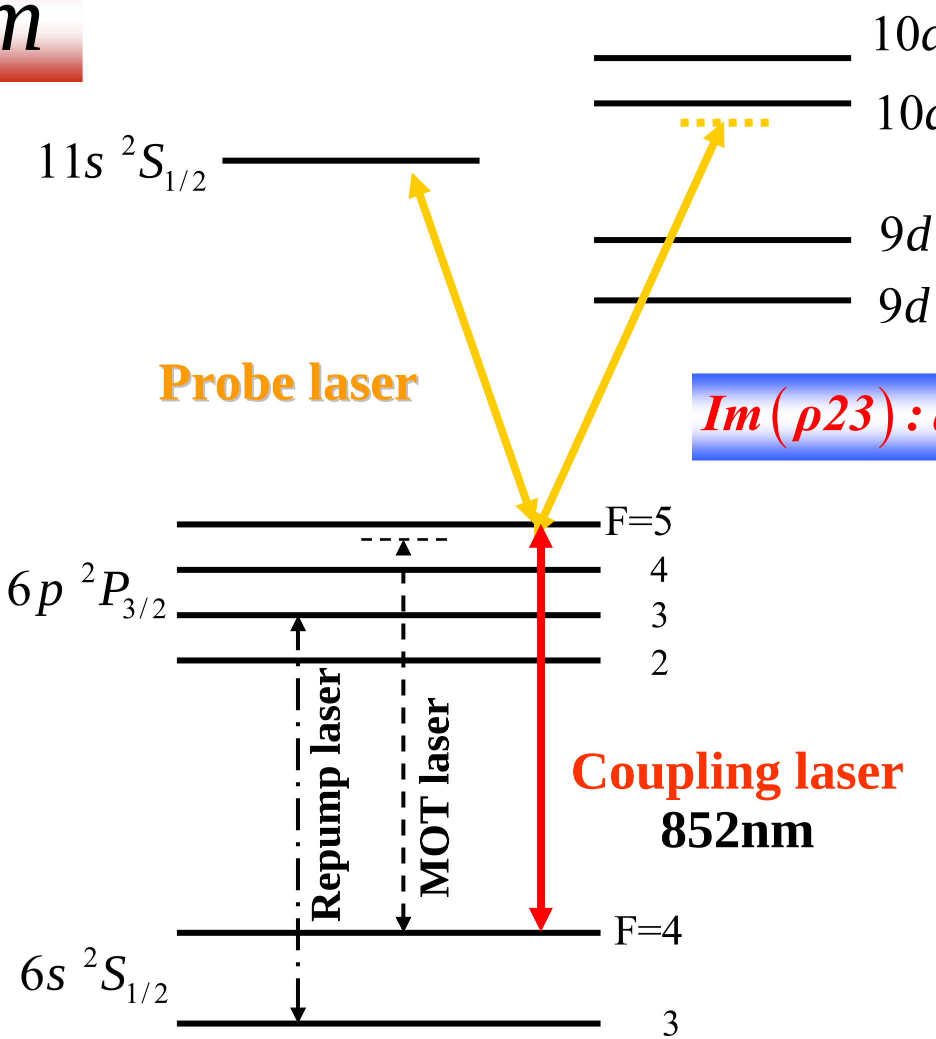






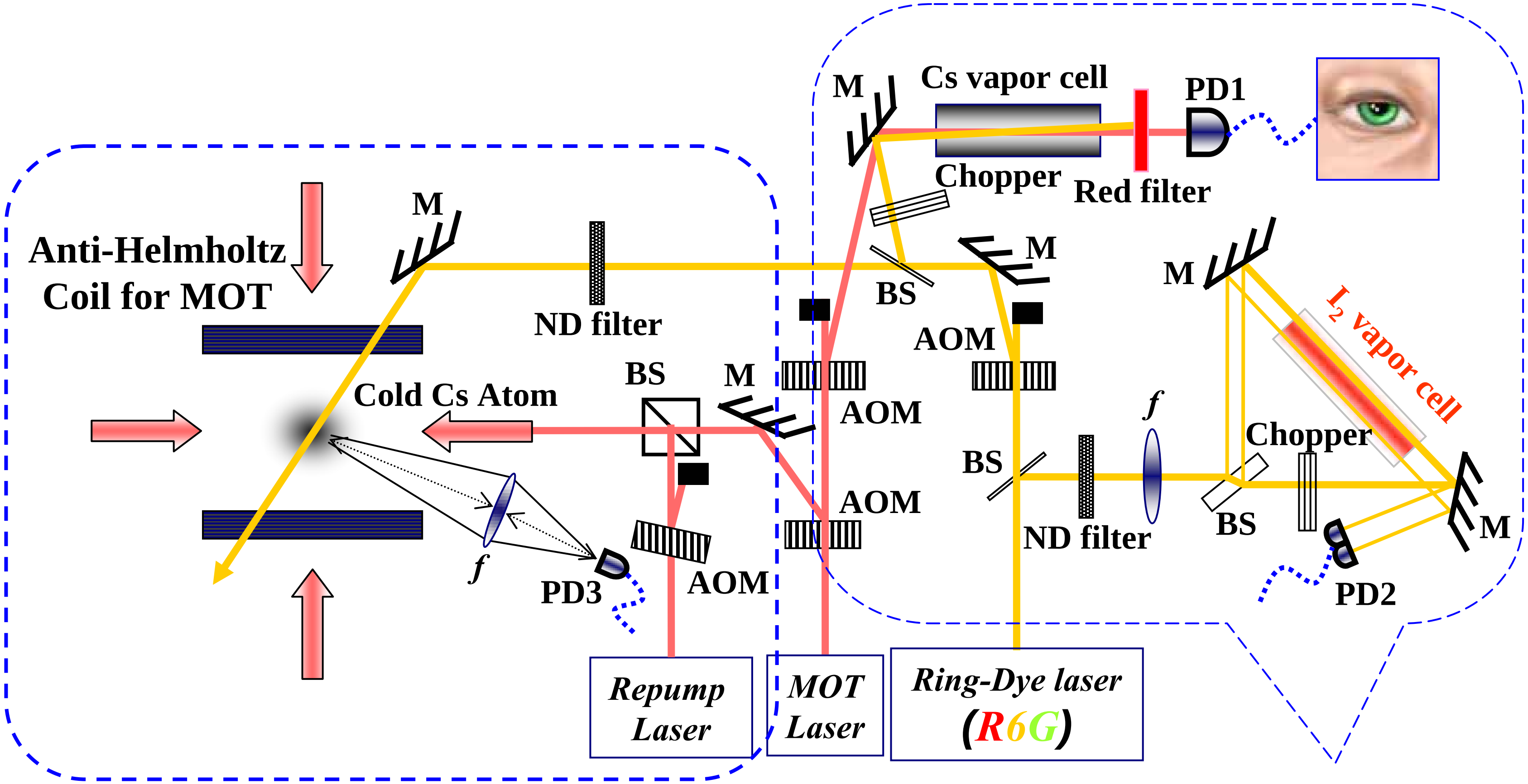
Energy level diagram

^{133}Cesium



Im(ρ23) : absorption

Experimental Setup



EIT setup

Precision measurement

Visible Cs MOT

Visible Cs MOT :

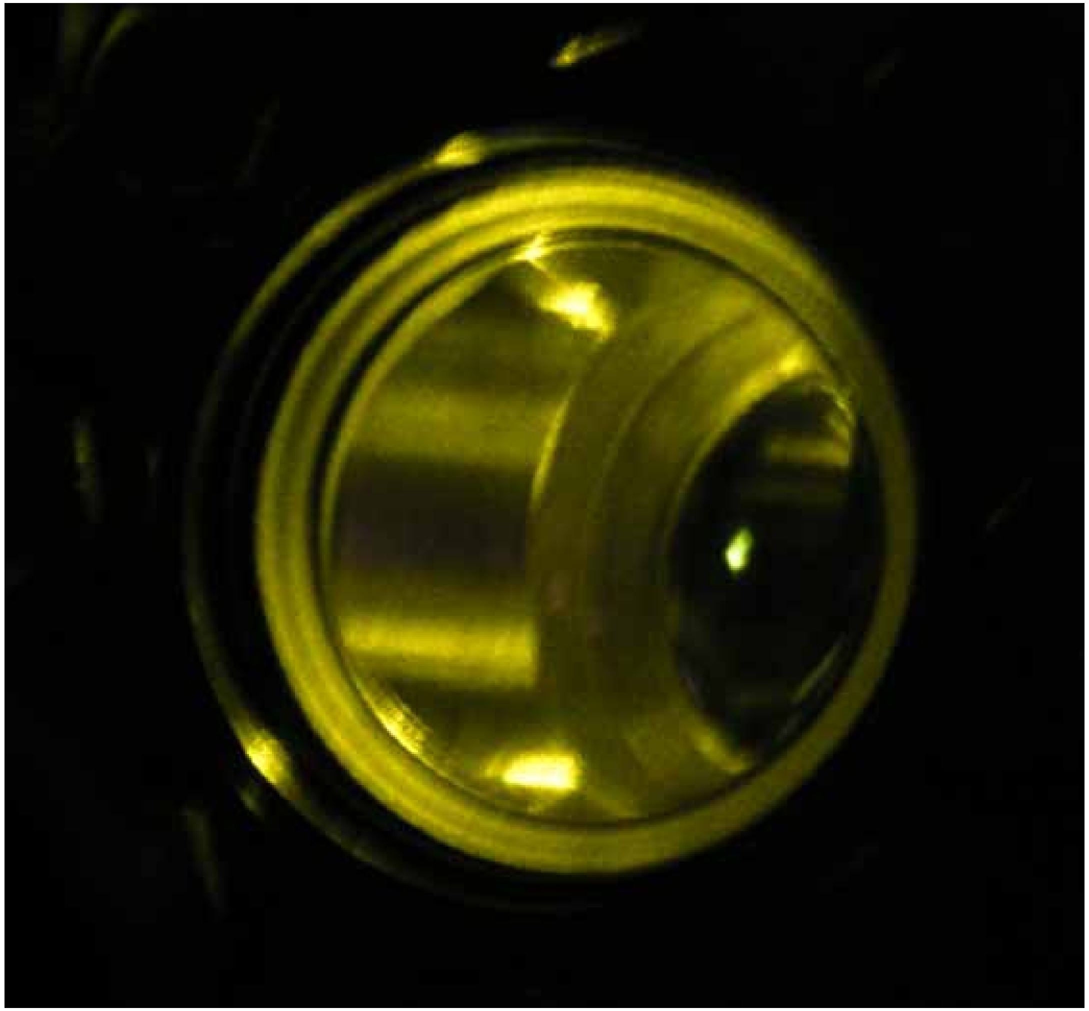
Probe laser transition

$\left|6p\ {}^2P_{3/2}\right\rangle \rightarrow \left|10d\ {}^2D_{5/2}\right\rangle$

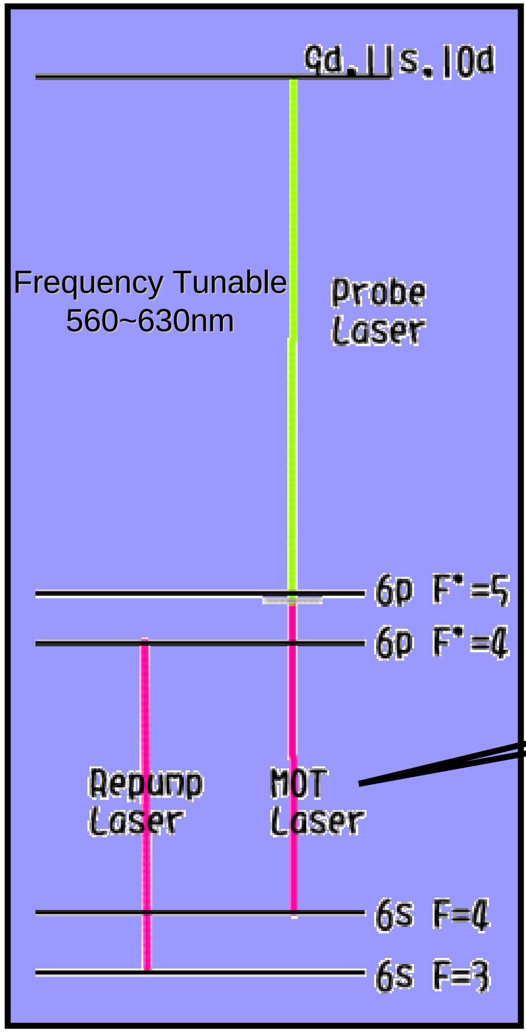
563.6nm

Atom number ~10^8

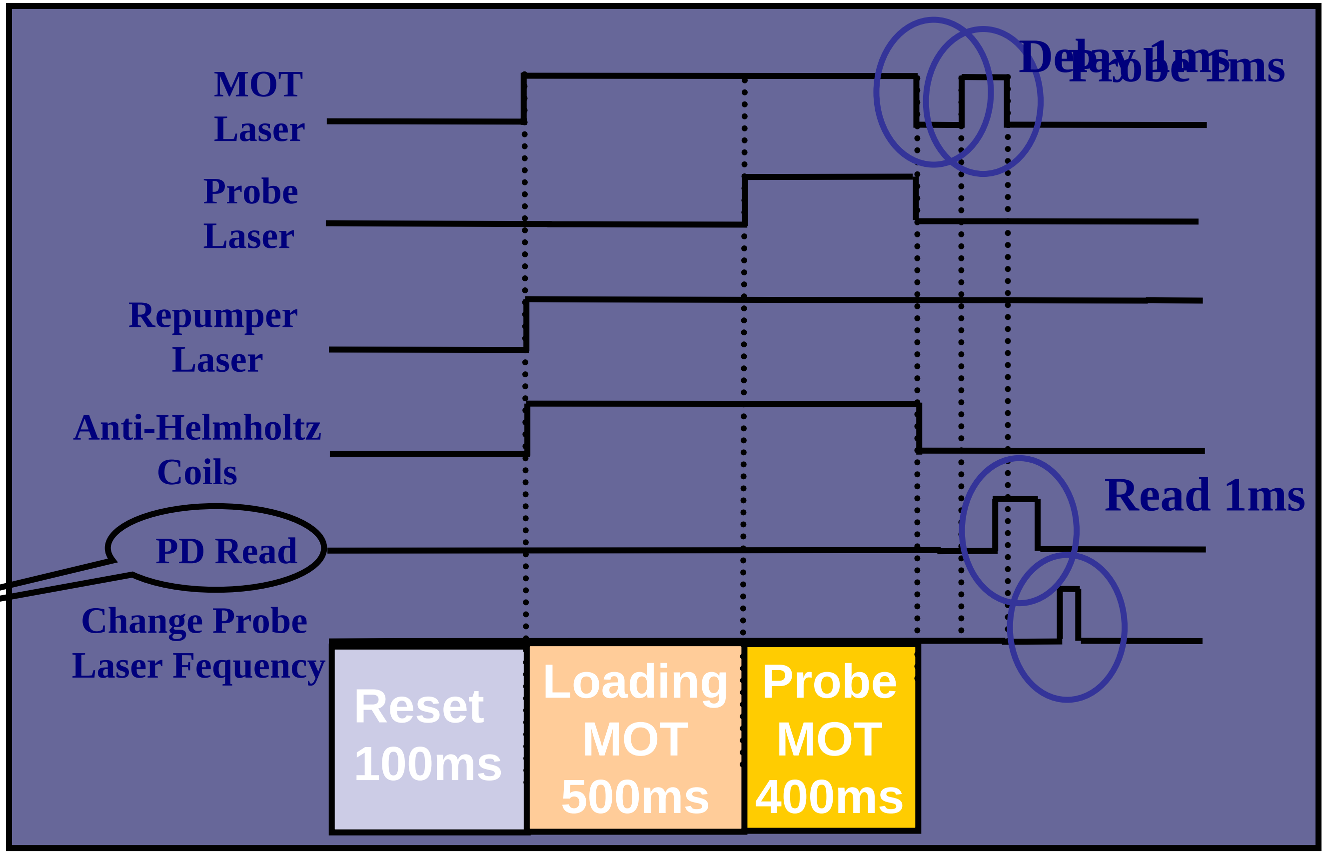
Temperature~200μK



Data Acquisition by External Scan

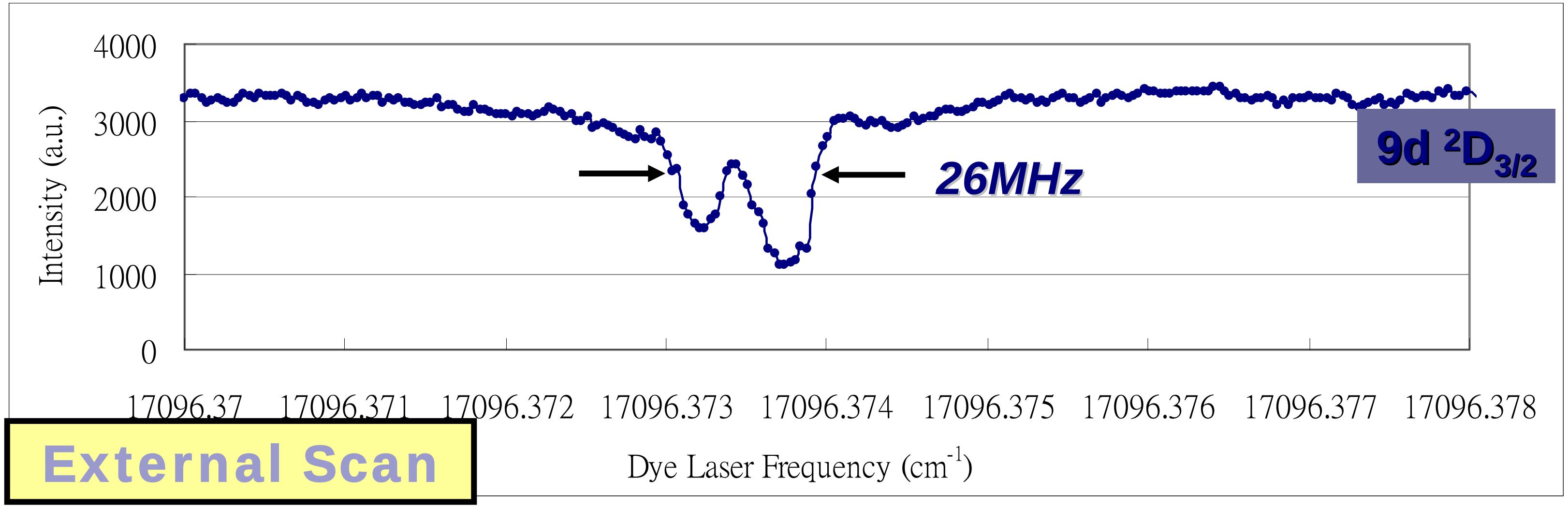


Trap Loss

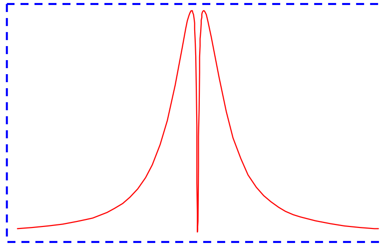


Atomic Transitions

$$\left|6s\ ^2S_{1/2}, F=5\right\rangle \rightarrow \left|6p\ ^2P_{3/2}, F=4\right\rangle \rightarrow \left|9d\ ^2D_{5/2}, F\right\rangle$$

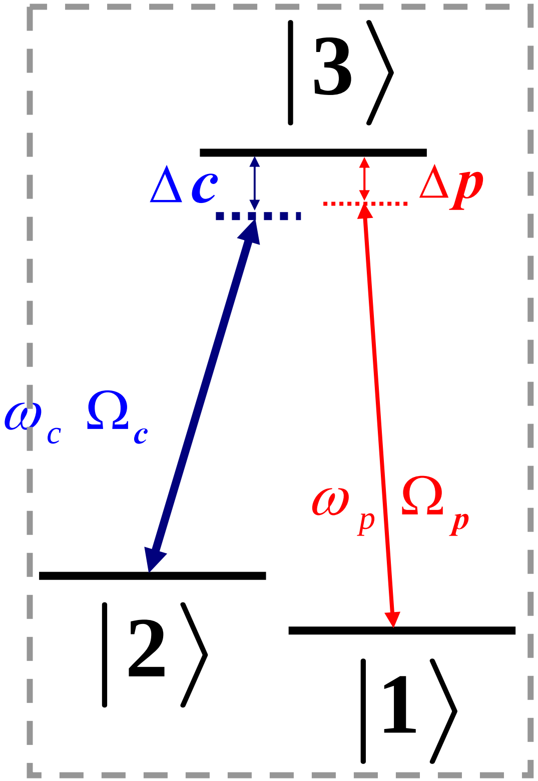


Electromagnetically Induced Transparency

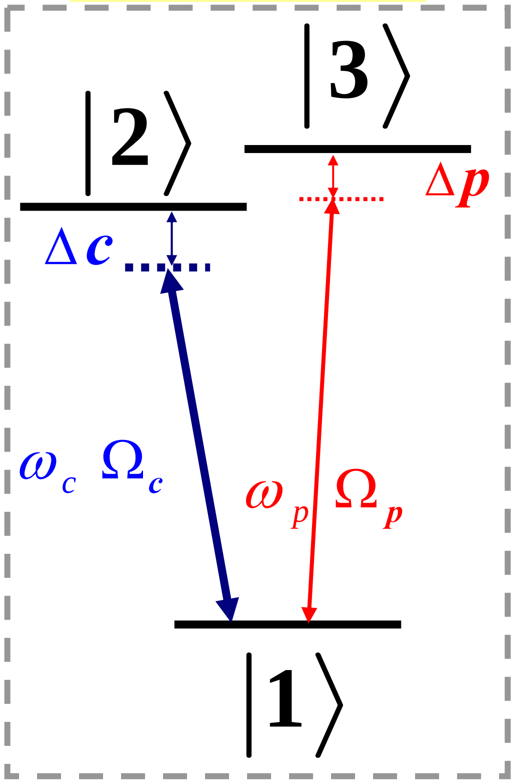


Quantum Interference

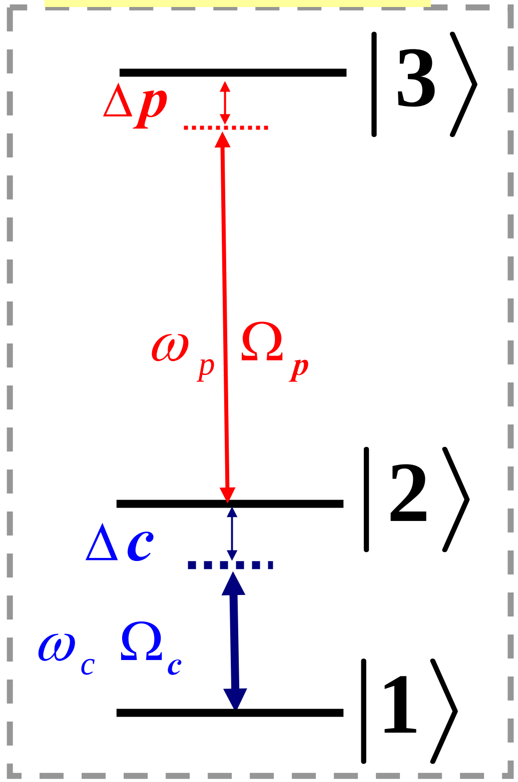
Λ - type



V - type



Cascade

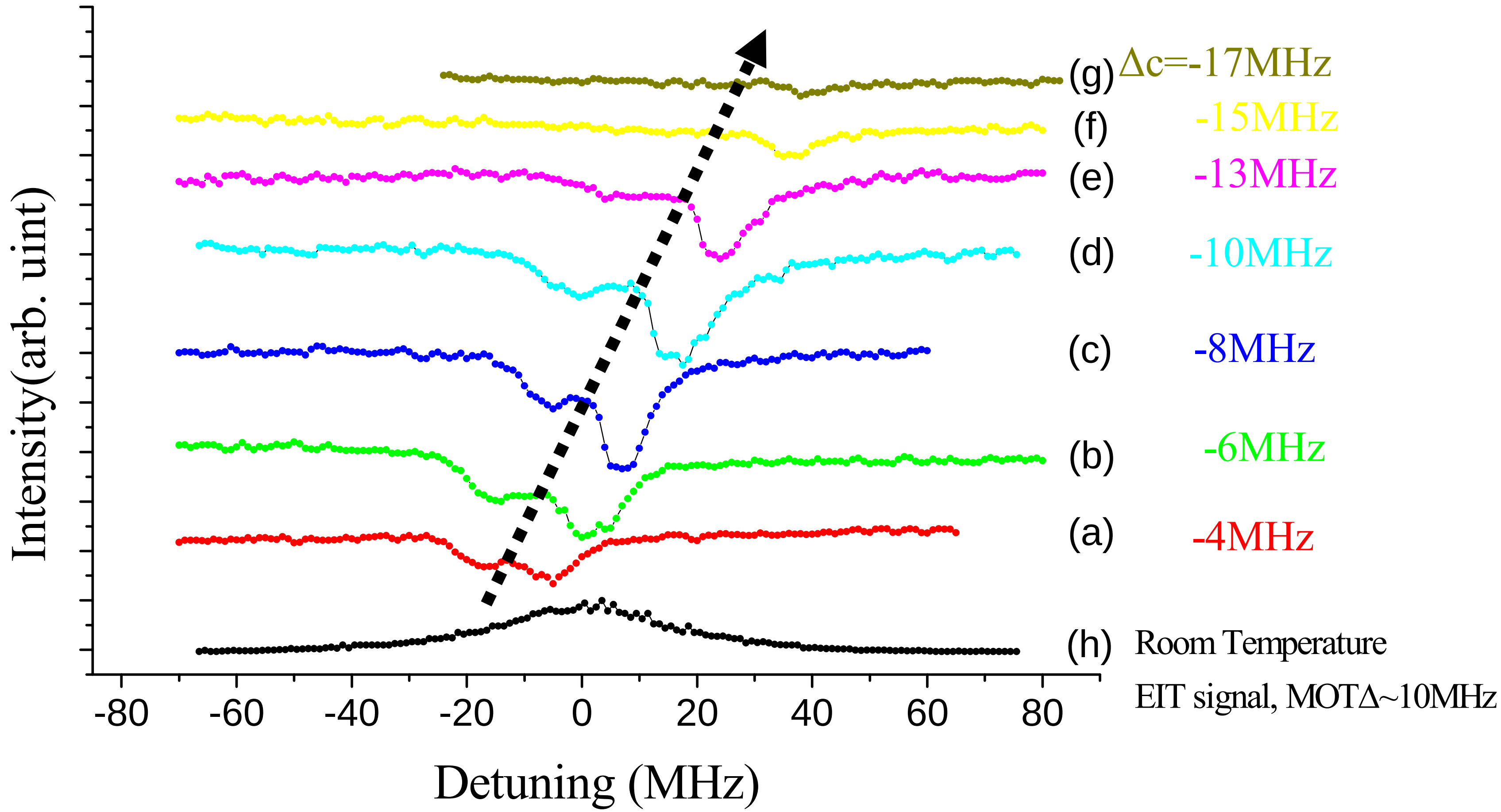


Results

Electromagnetically induced transparency (EIT) has been observed in a cascade system of laser-cooled Cs atoms.

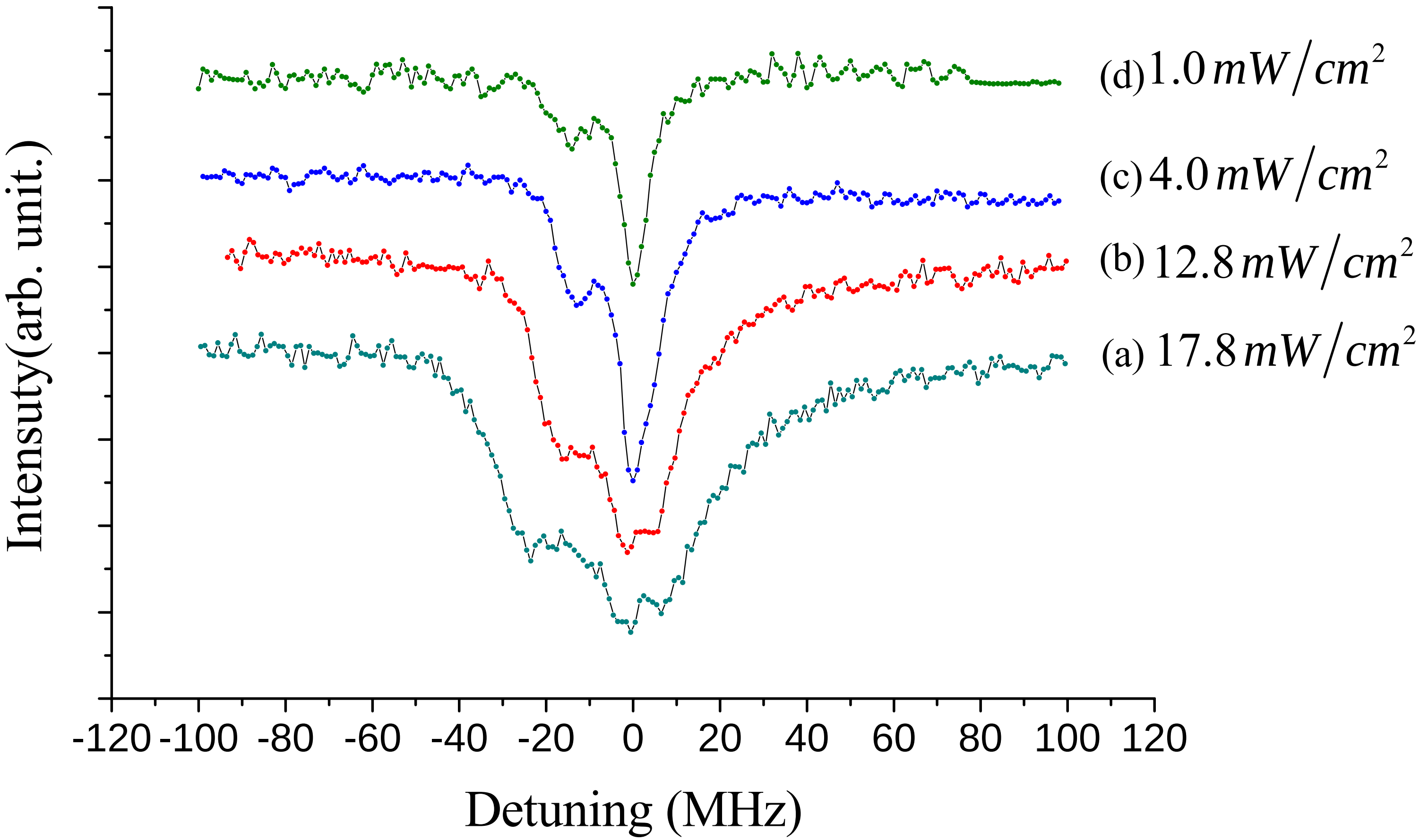
$$\left|6s\ ^2S_{1/2}, F=5\right\rangle \rightarrow \left|6p\ ^2P_{3/2}, F=4\right\rangle \rightarrow \left|11s\ ^2S_{1/2}, F\right\rangle$$

$wc: 10mW/cm^2$, $wp: 1.5mW/cm^2$



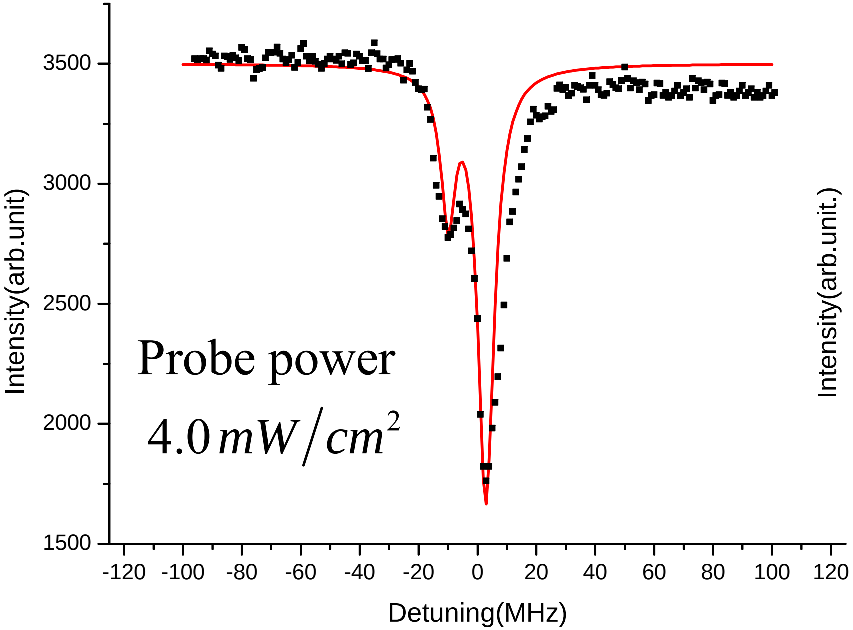
Probe power vs. Probe detuning

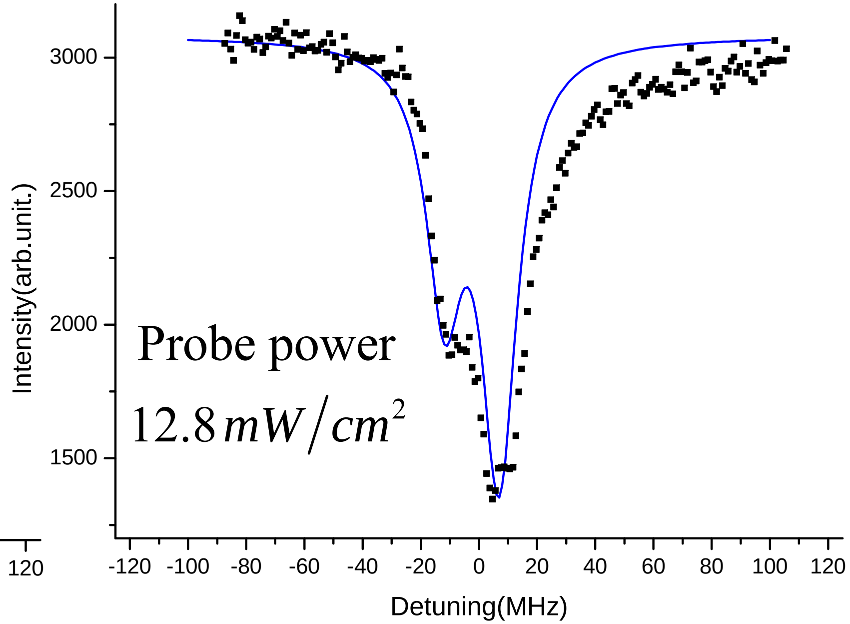
$$\left|6s\ ^2S_{1/2}, F=5\right\rangle \rightarrow \left|6p\ ^2P_{3/2}, F=4\right\rangle \rightarrow \left|11s\ ^2S_{1/2}, F\right\rangle, \text{ wc: 10mW/cm}^2$$



Numerical Simulation

$$\left|6s\ ^2S_{1/2}, F=5\right\rangle \rightarrow \left|6p\ ^2P_{3/2}, F=4\right\rangle \rightarrow \left|11s\ ^2S_{1/2}, F\right\rangle,\ \text{wc: } 10\text{mW/cm}^2$$

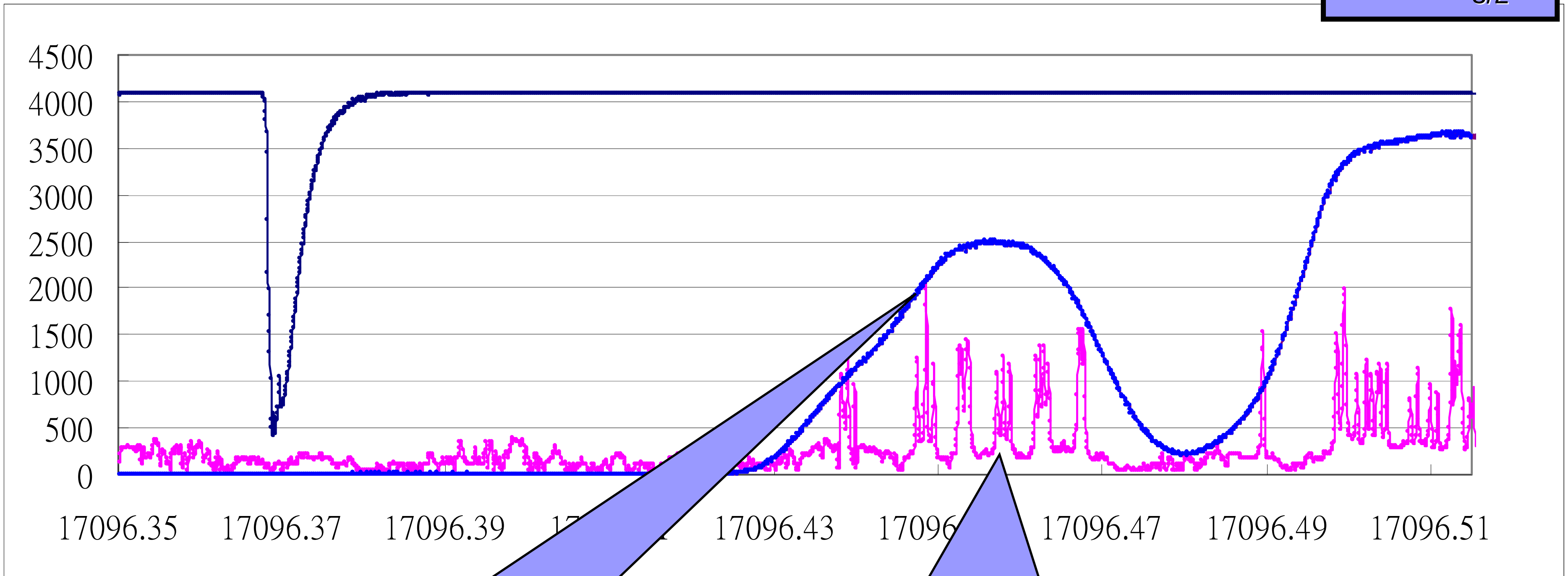




γ_2=5.2MHz, γ_3=2.5MHz, wc=3MHz, wp=1MHz, Δc=-10MHz Ωc=20MHz

Long Scan with Molecular Iodine Hyperfine Spectrum

9d $^2D_{3/2}$



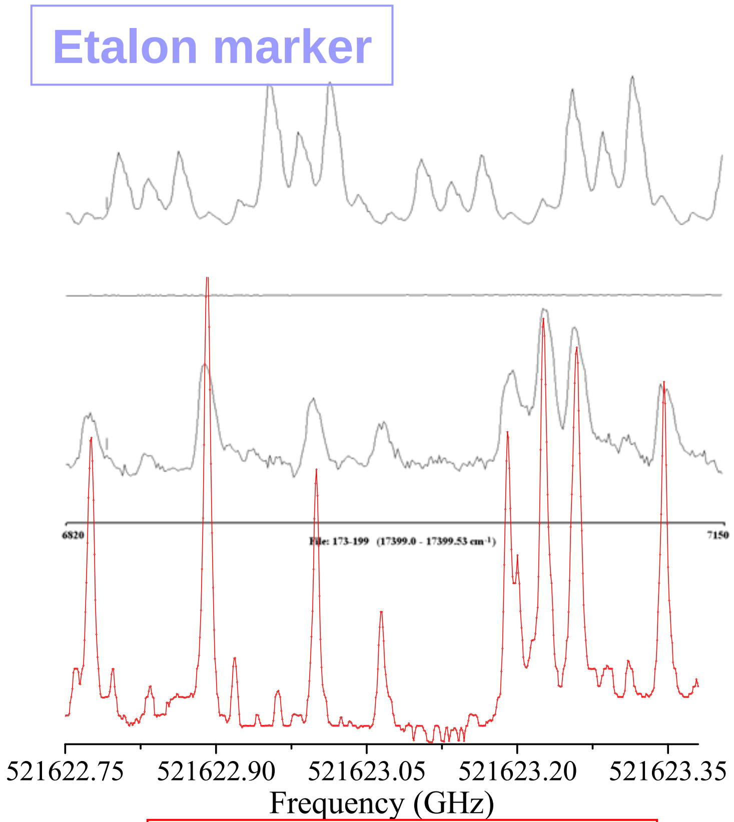
Iodine Excitation Spectrum
Absolute Frequency 200MHz

Iodine Excitation Spectrum
Absolute Frequency 3 MHz

Calibration procedure

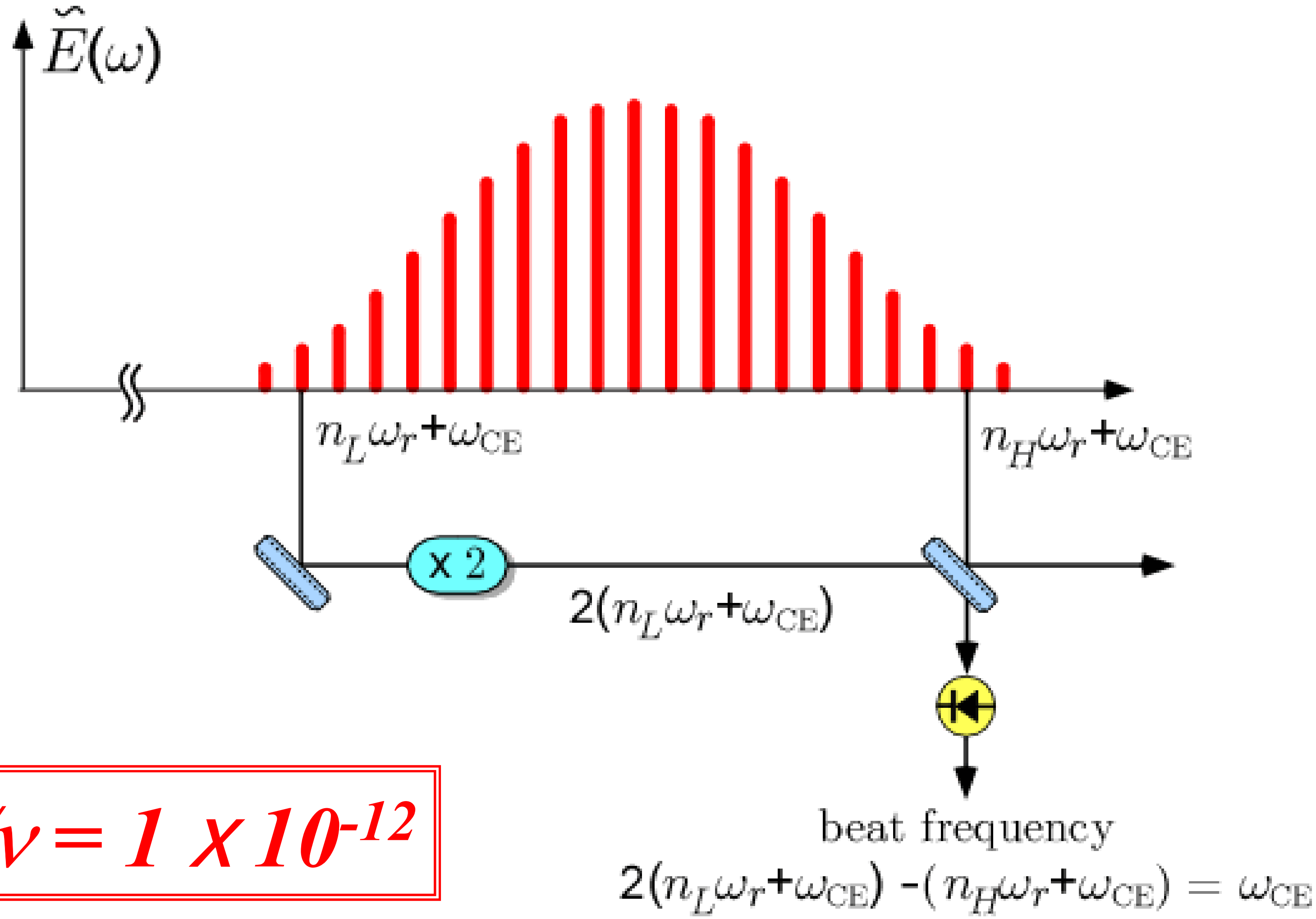
JSPS I_2 Hyperfine

		05100203.scn	5W/17.6mW	11s $^2S_{1/2}$
	cm^{-1}	Δ=-10MHz	(ν/c)	cm^{-1}
	I2_Reference	I_2_Experent(GHz)	C=299792458	deviation
peak1	17399.45226	521622.82650	17399.46462	0.01236
2	17399.45607	521622.94250	17399.46848	0.01241
3	17399.45968	521623.05150	17399.47212	0.01244
4	17399.46159	521623.11650	17399.47429	0.01270
5	17399.46625	521623.24150	17399.47846	0.01221
7	17399.46826	521623.30850	17399.48069	0.01243
8	17399.47123	521623.39950	17399.48373	0.01250
		Average(Δ)	0.01244	
		Stdev.(δ)	0.00015	*< 5MHz*
		EIT	521621.65660	17399.42559
		EIT-Δ		17399.41316
		MOT(Trap Loss)	521621.66980	17399.42603
		MOT-Δ		17399.41360



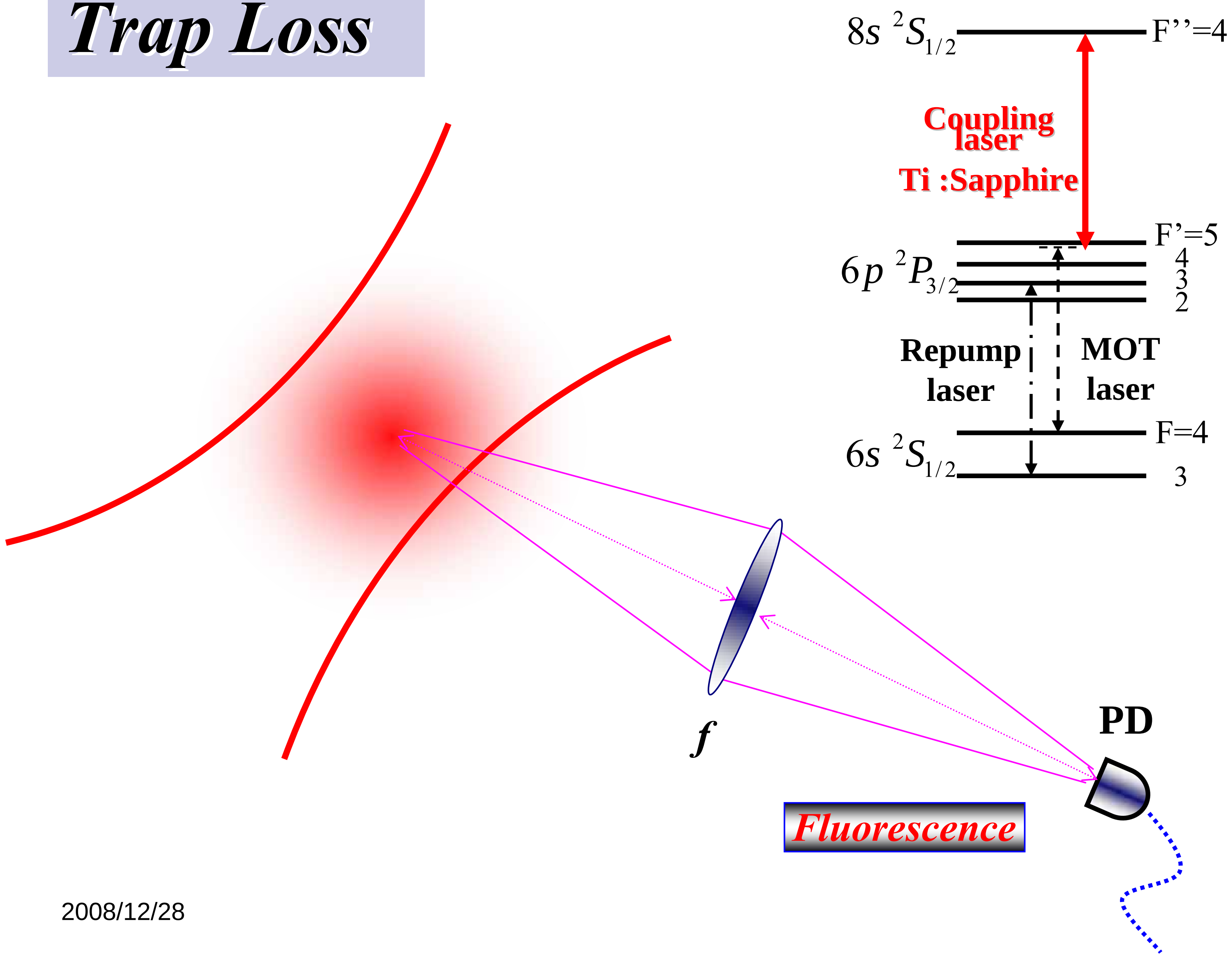
I_2 Hyperfine/This work

Frequency Comb



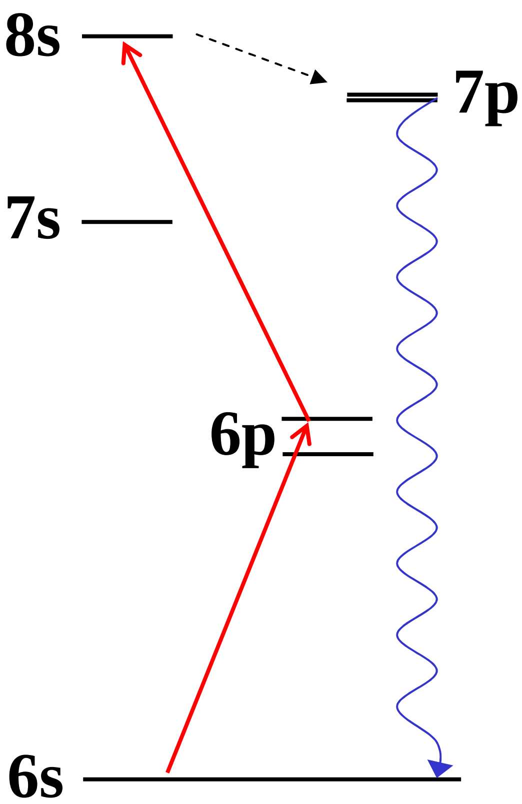
$\Delta\nu/\nu = 1 \times 10^{-12}$

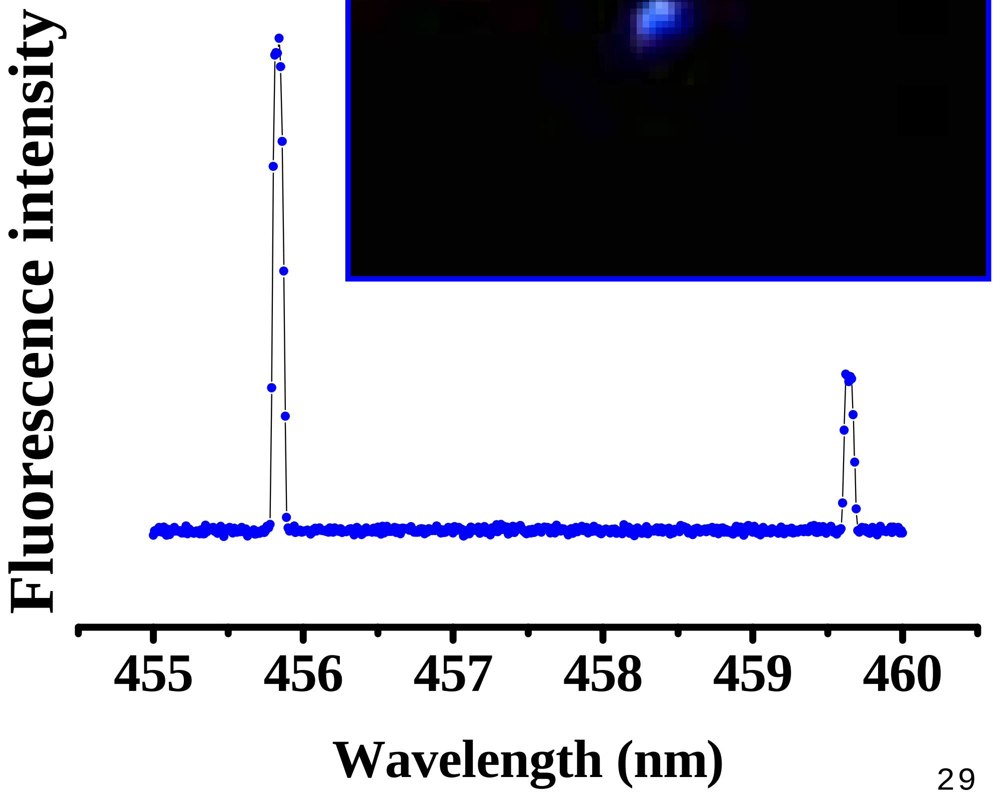
Trap Loss



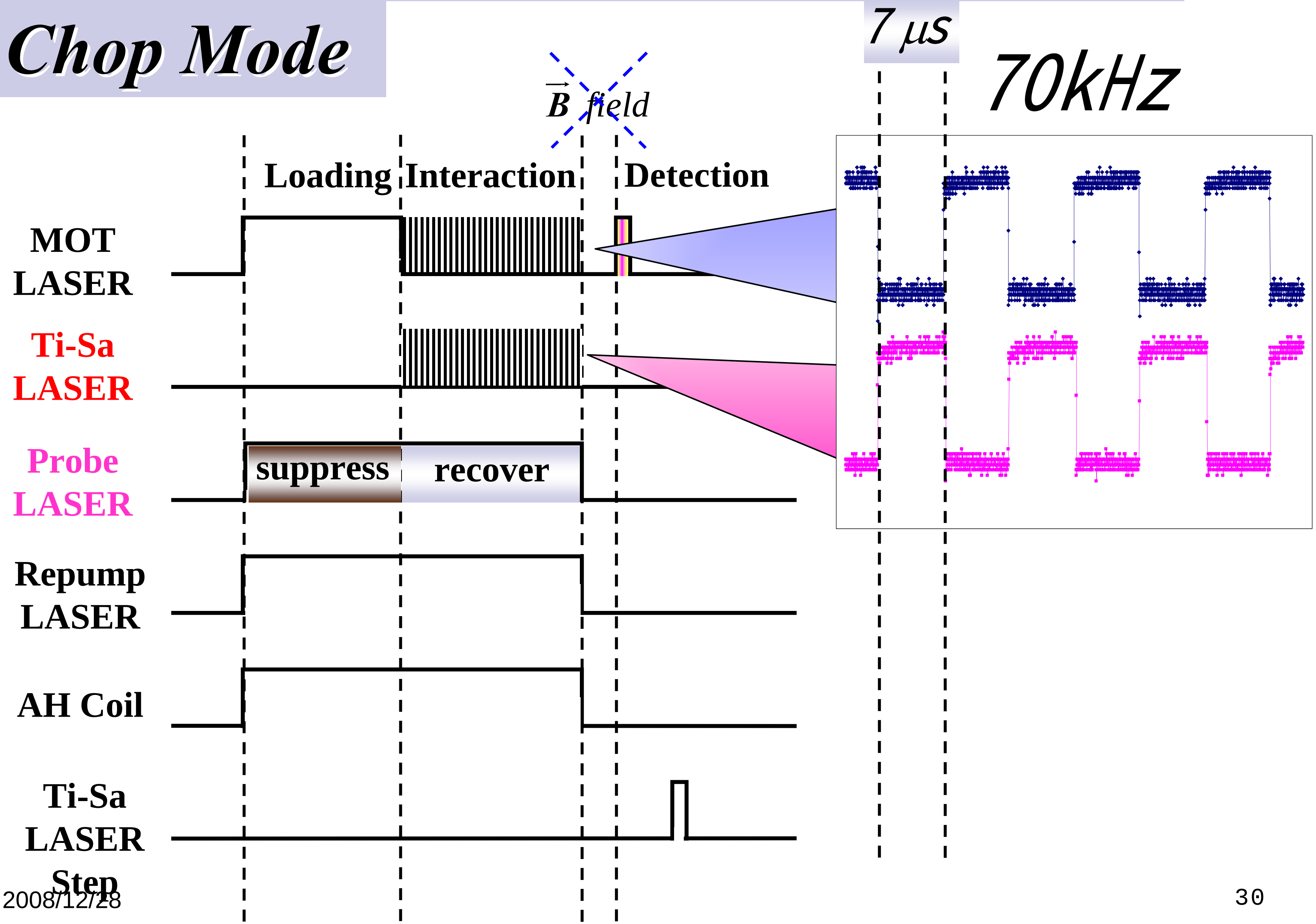
$8s\ ^2S_{1/2}$
F''=4
Coupling laser
Ti :Sapphire
F'=5
4
3
2
$6p\ ^2P_{3/2}$
Repump laser
MOT laser
F=4
3
$6s\ ^2S_{1/2}$
f
PD
Fluorescence

Decay fluorescence

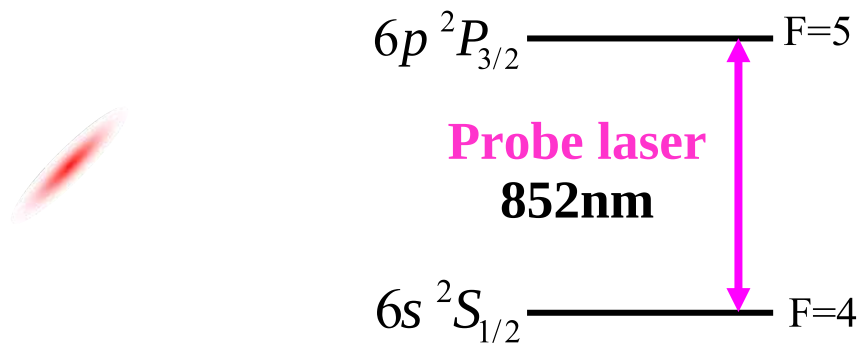




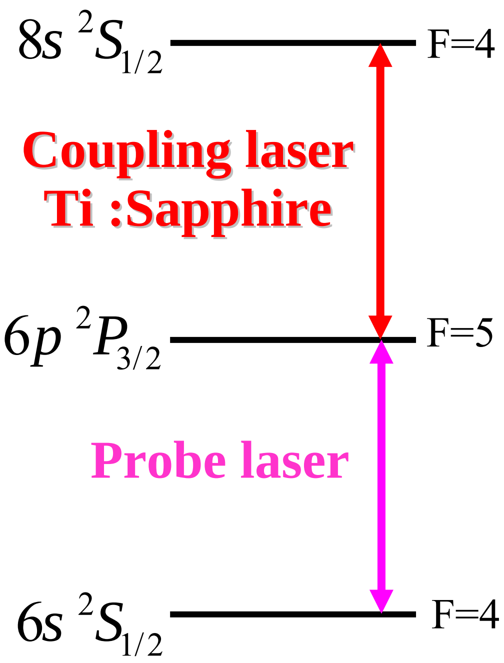
Chop Mode

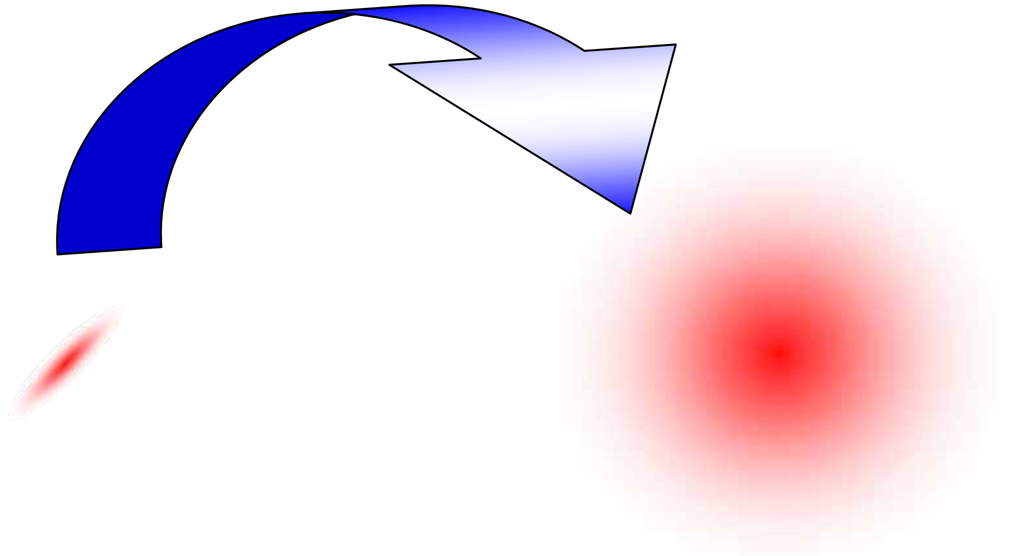

7 μs
70kHz
$\vec{B}$ field
Loading
Interaction
Detection
MOT LASER
Ti-Sa LASER
Probe LASER
suppress
recover
Repump LASER
AH Coil
Ti-Sa LASER Step

Suppression

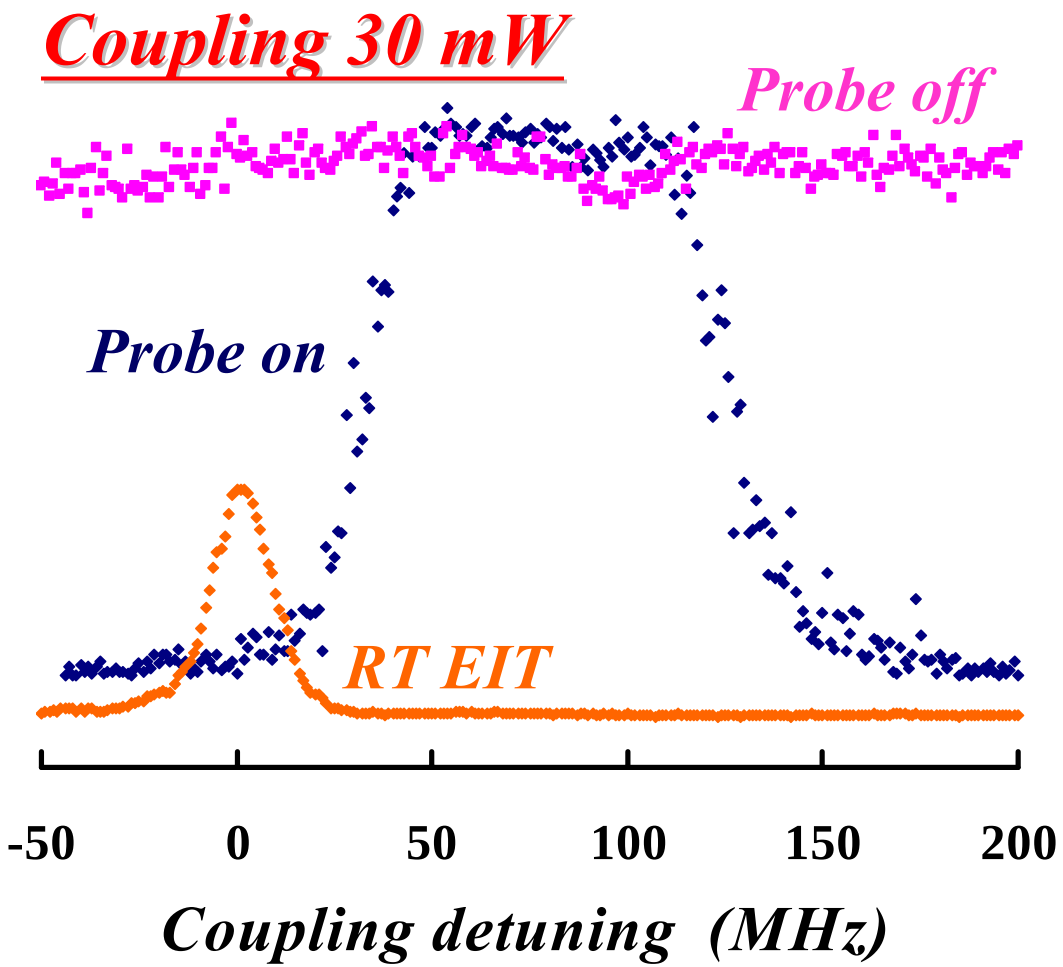


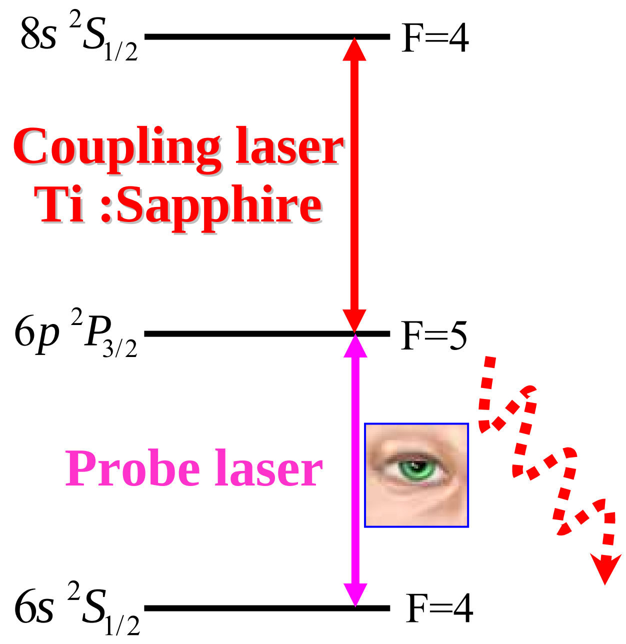
Suppression & Recovery



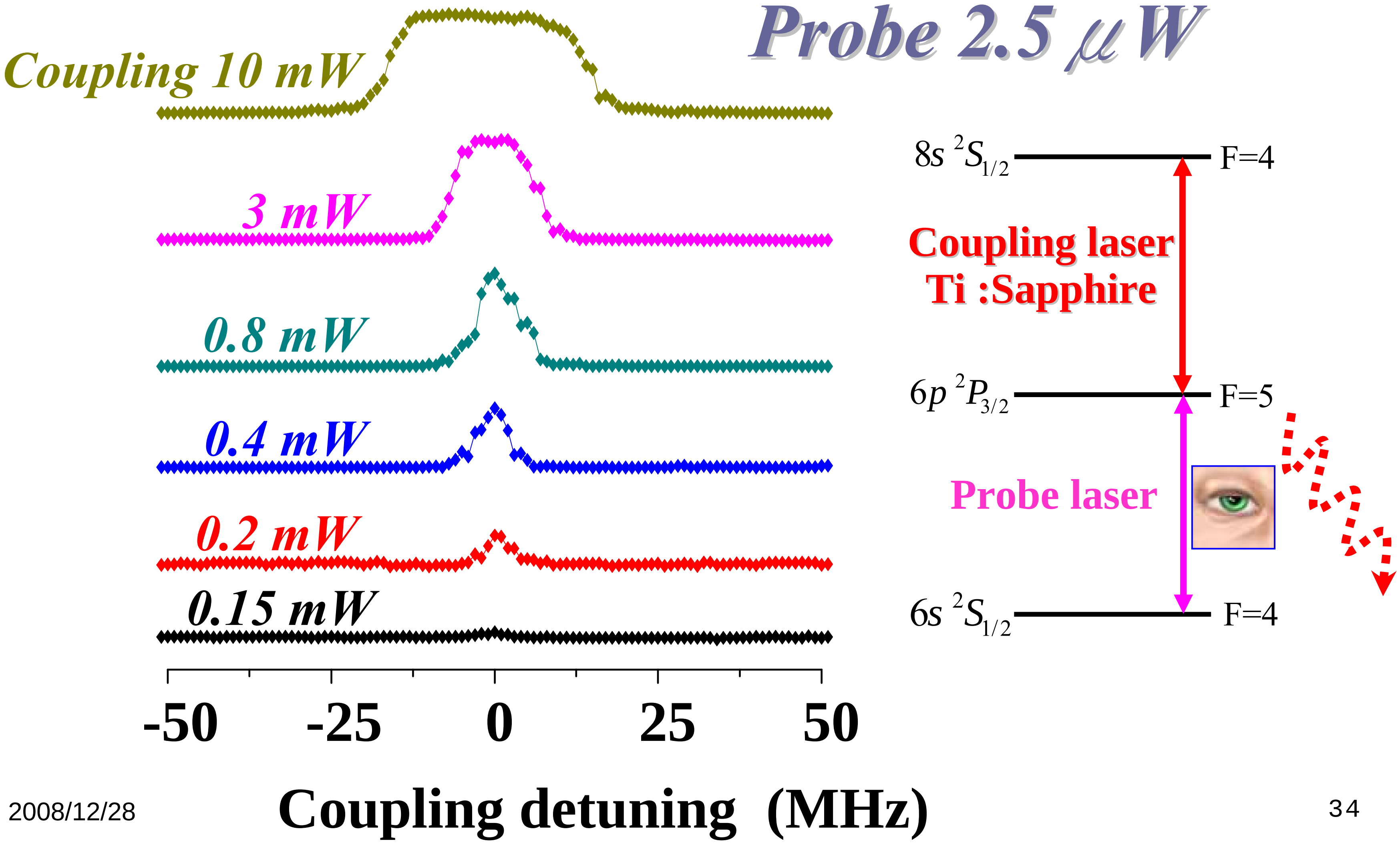


Suppression & Recovery

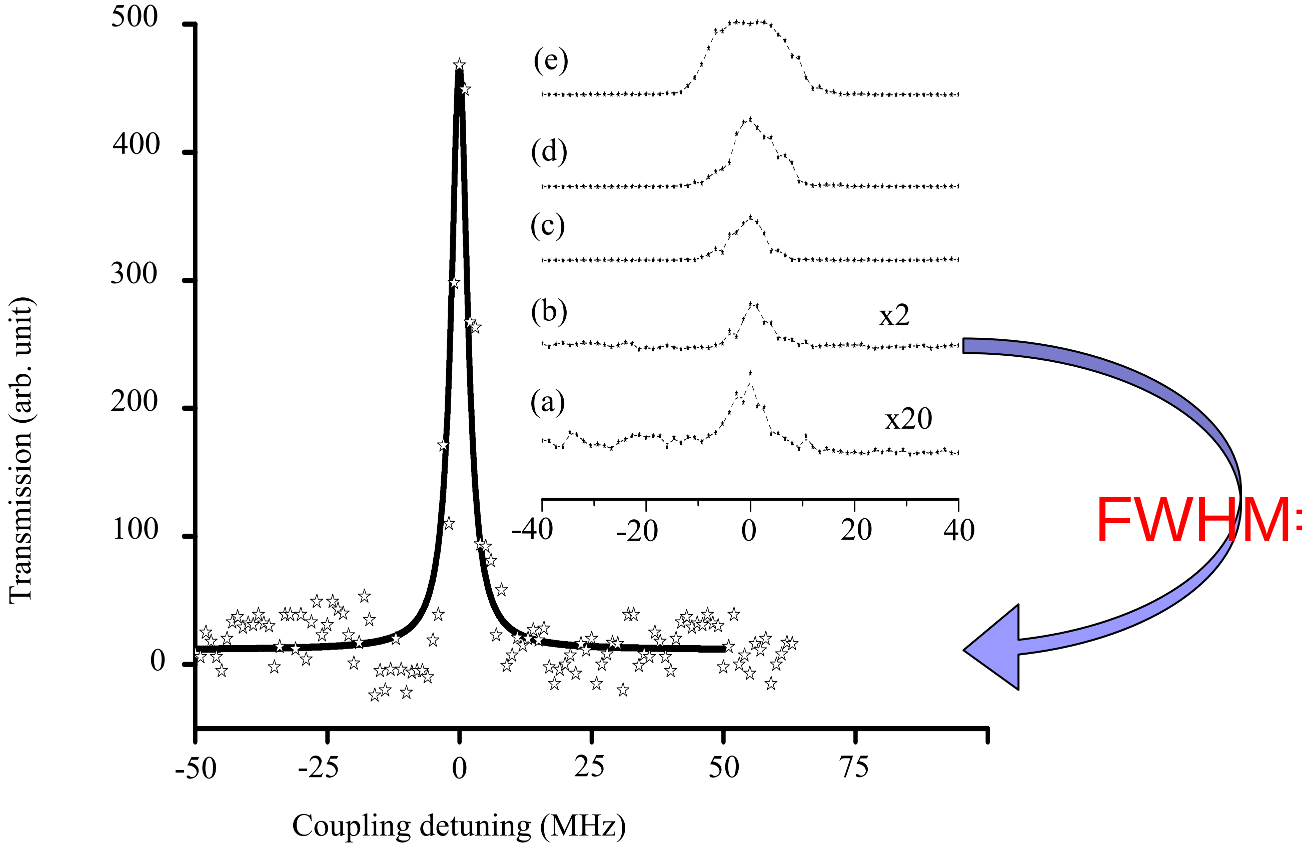




Power dependence

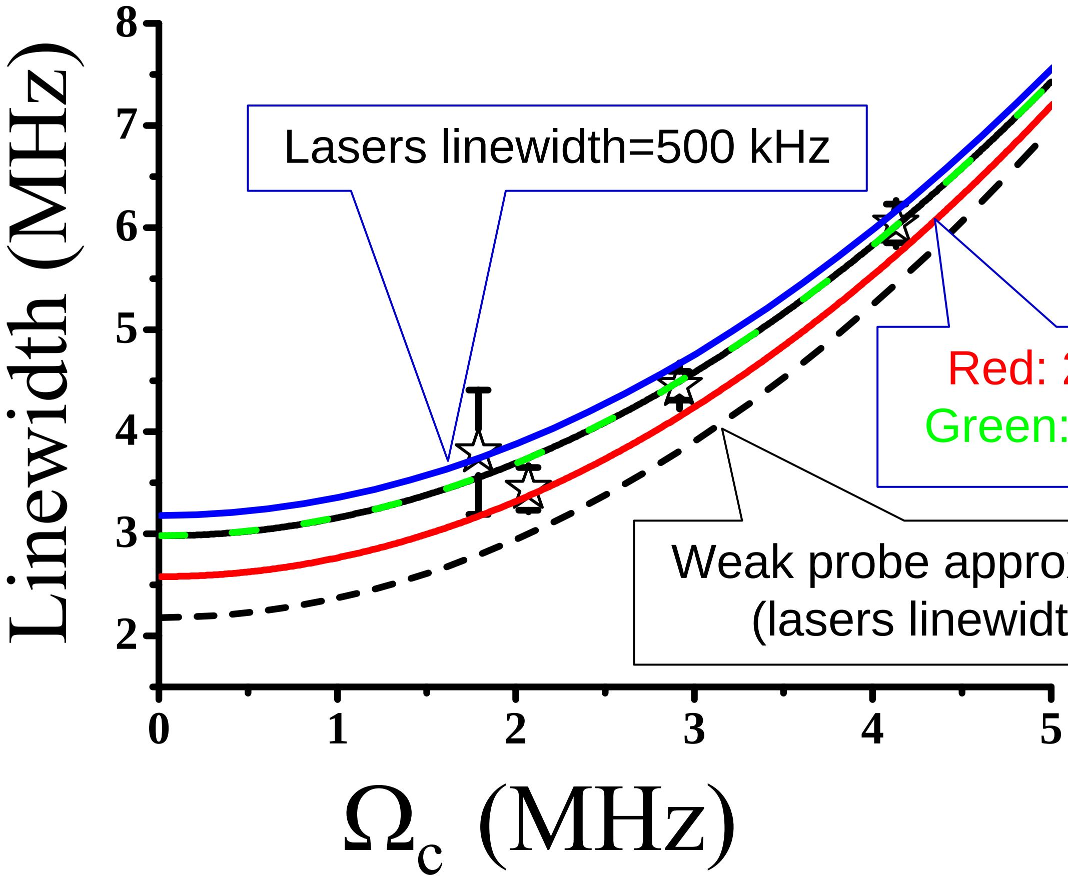


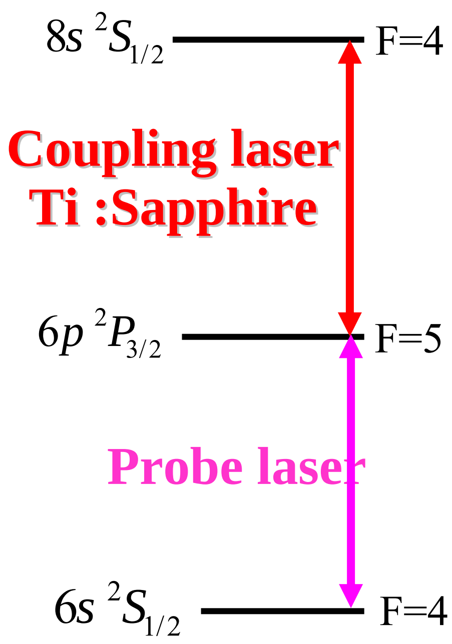
Power dependence



FWHM=3.44MHz

Linewidth vs. coupling Rabi frequency



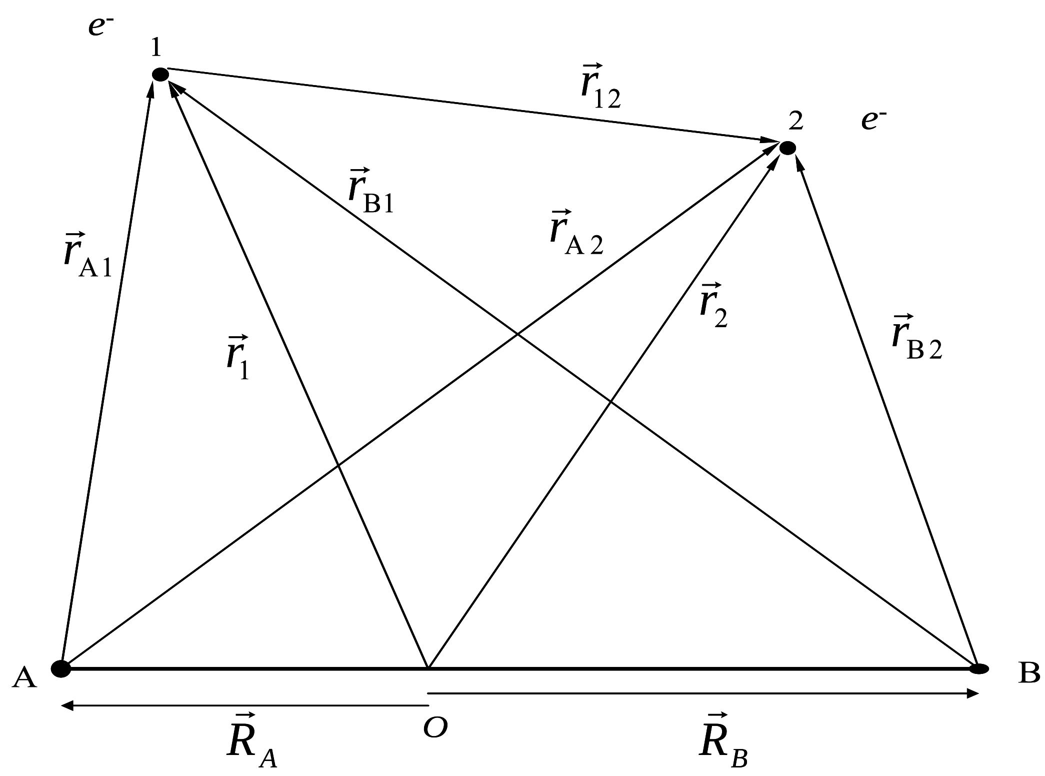


Laser linewidth serves as a de-coherence source.

Quantum Phenomena of atom-atom interactions

Molecular Spectroscopy

Diatomic molecule

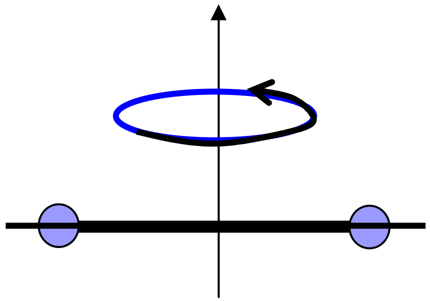


$$H_e\psi_q = \left(T_e + V\right)\psi_q = E_q(R)\psi_q$$

Vibrational Mode

Eigenvalues of Harmonic Oscillator

$$E_v = (v + \frac{1}{2})\hbar\omega$$



Rotational Mode

Eigenvalues as a Rigid Rotator

$$E_J = \frac{J^2}{2I} = \frac{J(J+1)\hbar^2}{2I}$$

Dunham Coefficients

$$T_{v,J} = \sum_{i=0}^{\infty}\sum_{j=0}^{\infty} Y_{ij}\left(v+\frac{1}{2}\right)^{i}[J(J+1)-\Lambda^2]^{j}$$

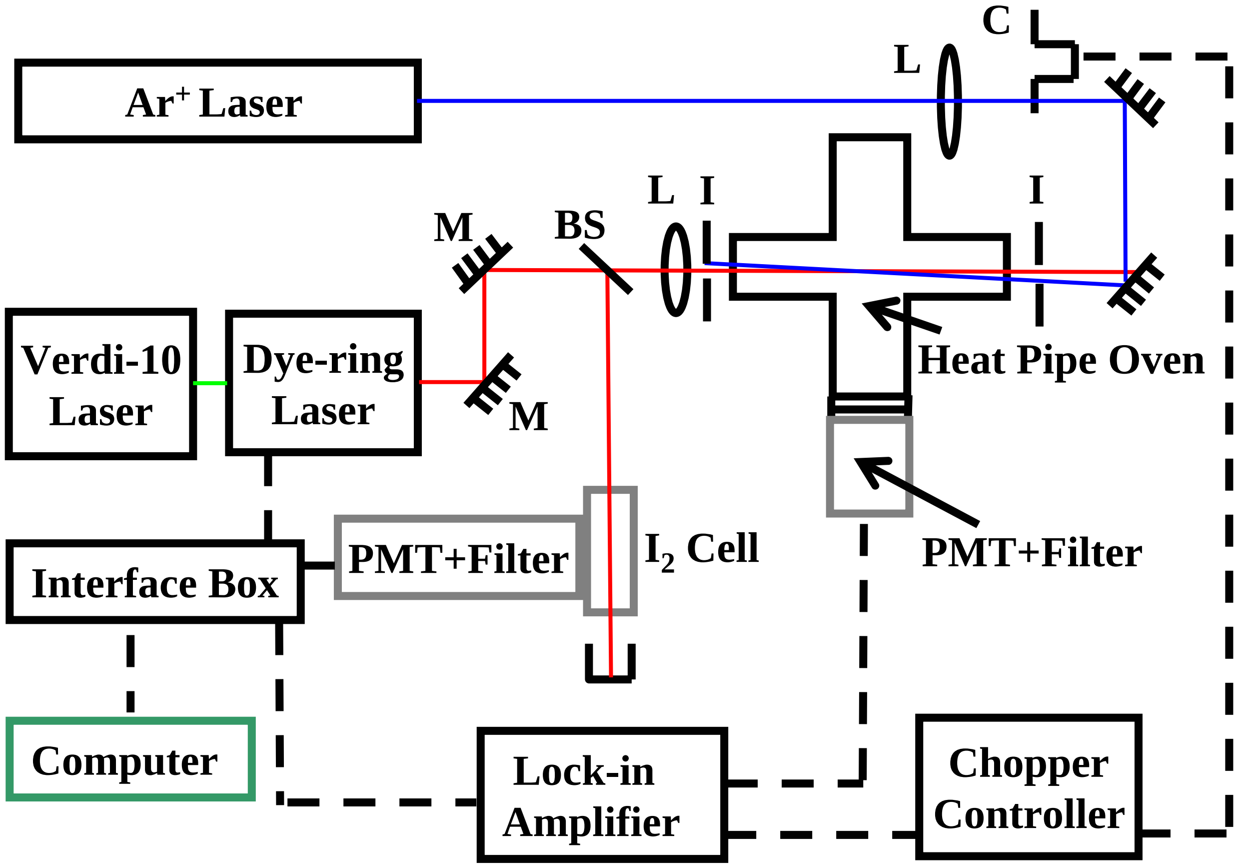
Lower terms of Dunham Coefficients (Y_{ij})

j\i	0	1	2	3	4	……..
0	T_e	ω_e	$-\omega_e x_e$	$\omega_e y_e$	$\omega_e z_e$	
1	B_e	$-\alpha_e$	γ_e	δ_e	..	
2	$-D_e$	$-\beta_e$	..	..	..	
3	H_e	..	..	..	..	
4	L_e	..	..	..	..	
..	..	..	..	..	..	

Harmonic Oscillator

Parts of Anharmonicity

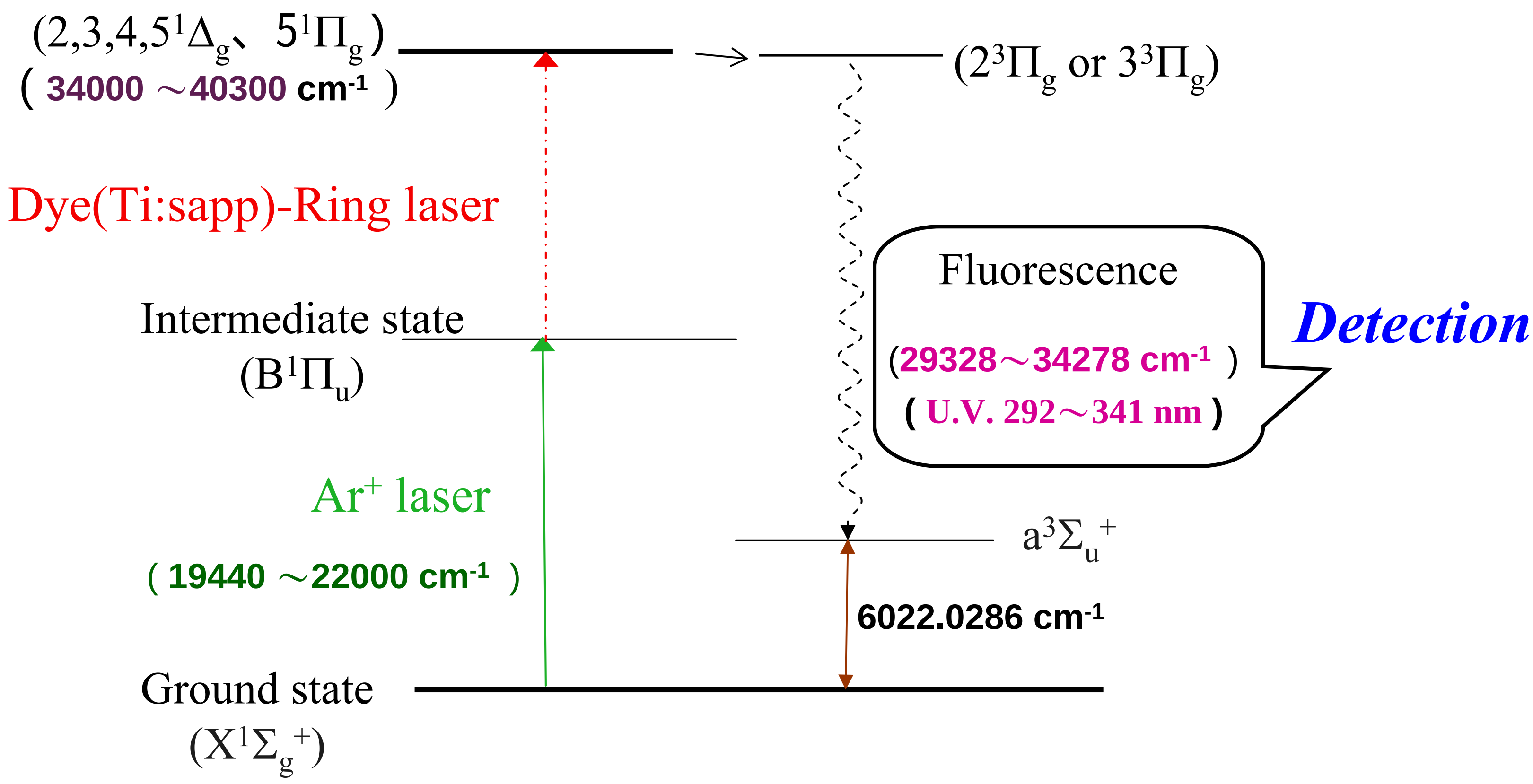
Experimental setup



System accuracy ~***0.03cm^{-1}***

Experimental setup

Probing energy range



Experimental energy range **34000 ～40300** cm^{-1}

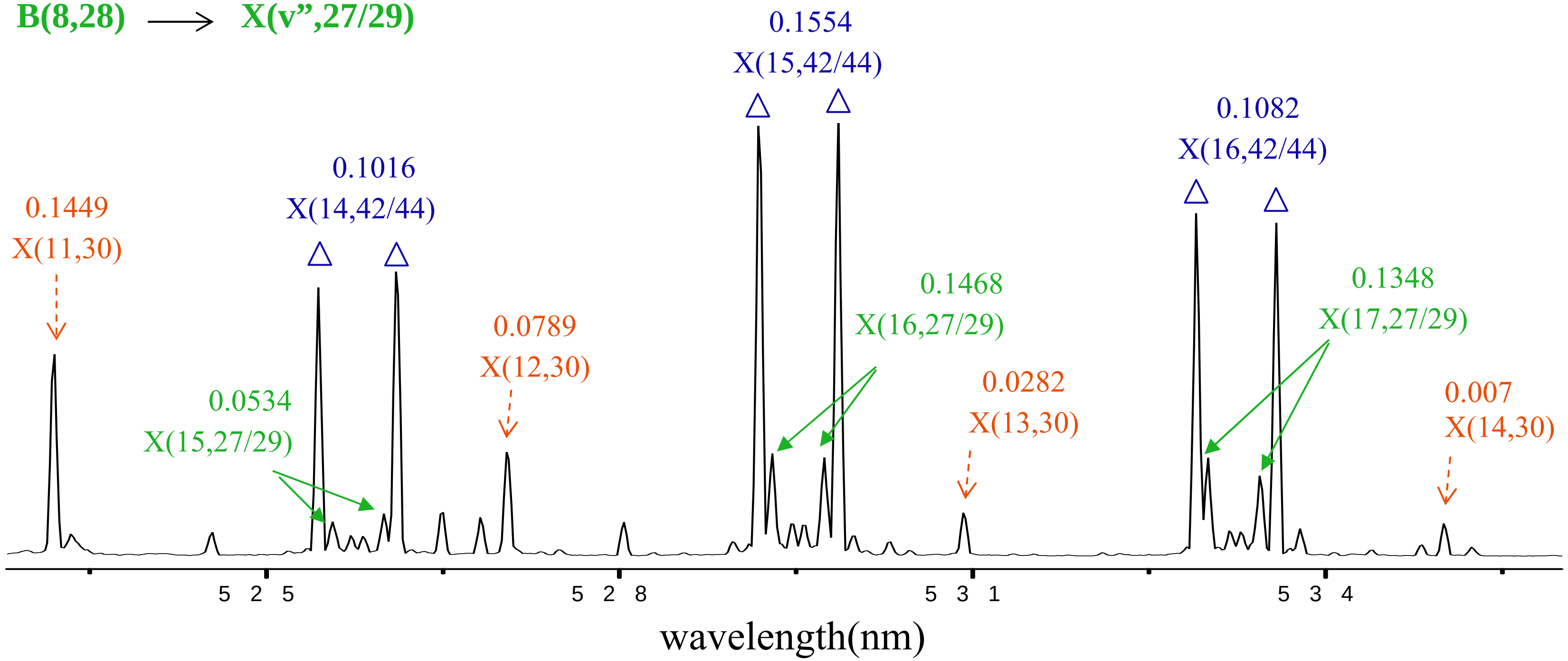
Laser Induced Fluorescence by Ar^+ Laser line at 496.5nm

$B^1\Pi_u \rightarrow X^1\Sigma_g^+$

B(7,43) ⟶ X(v”,42/44)

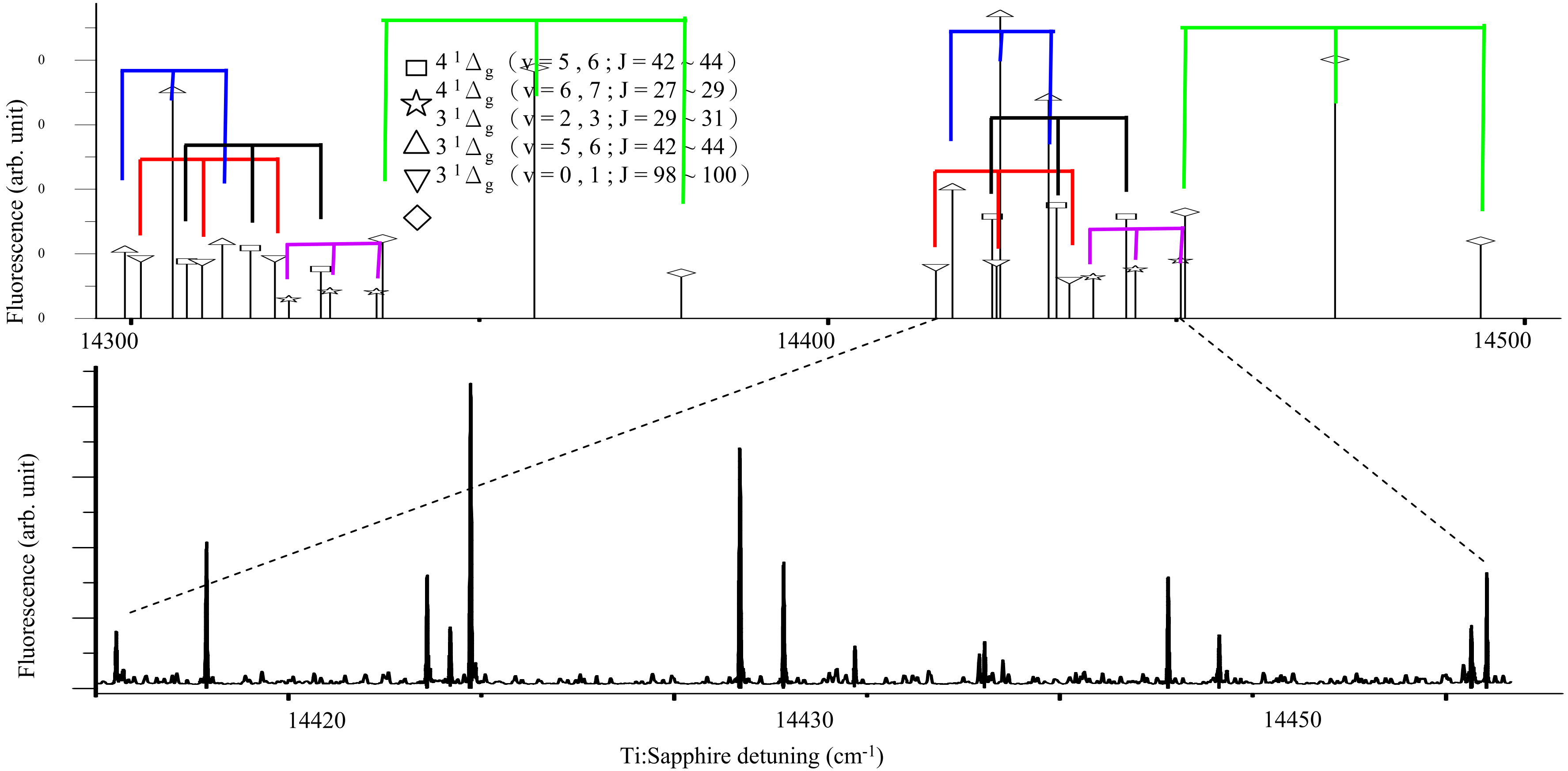
B(4,30) ⟶ X(v”,30)

B(8,28) ⟶ X(v”,27/29)



Experimental setup

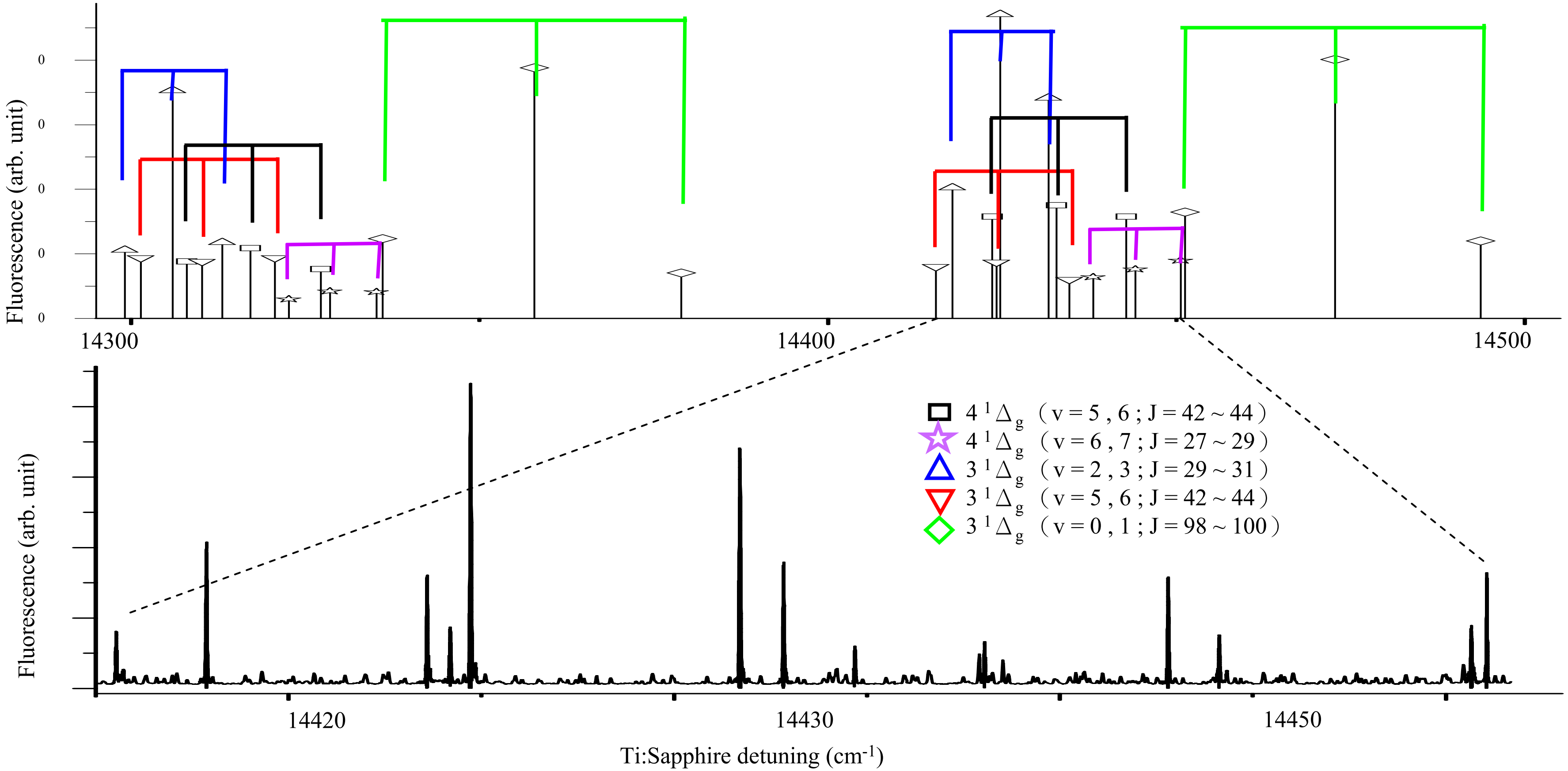
A typical part of fluorescence sticks spectrum converted from the raw fluorescence spectrum of the rovibrational progressions



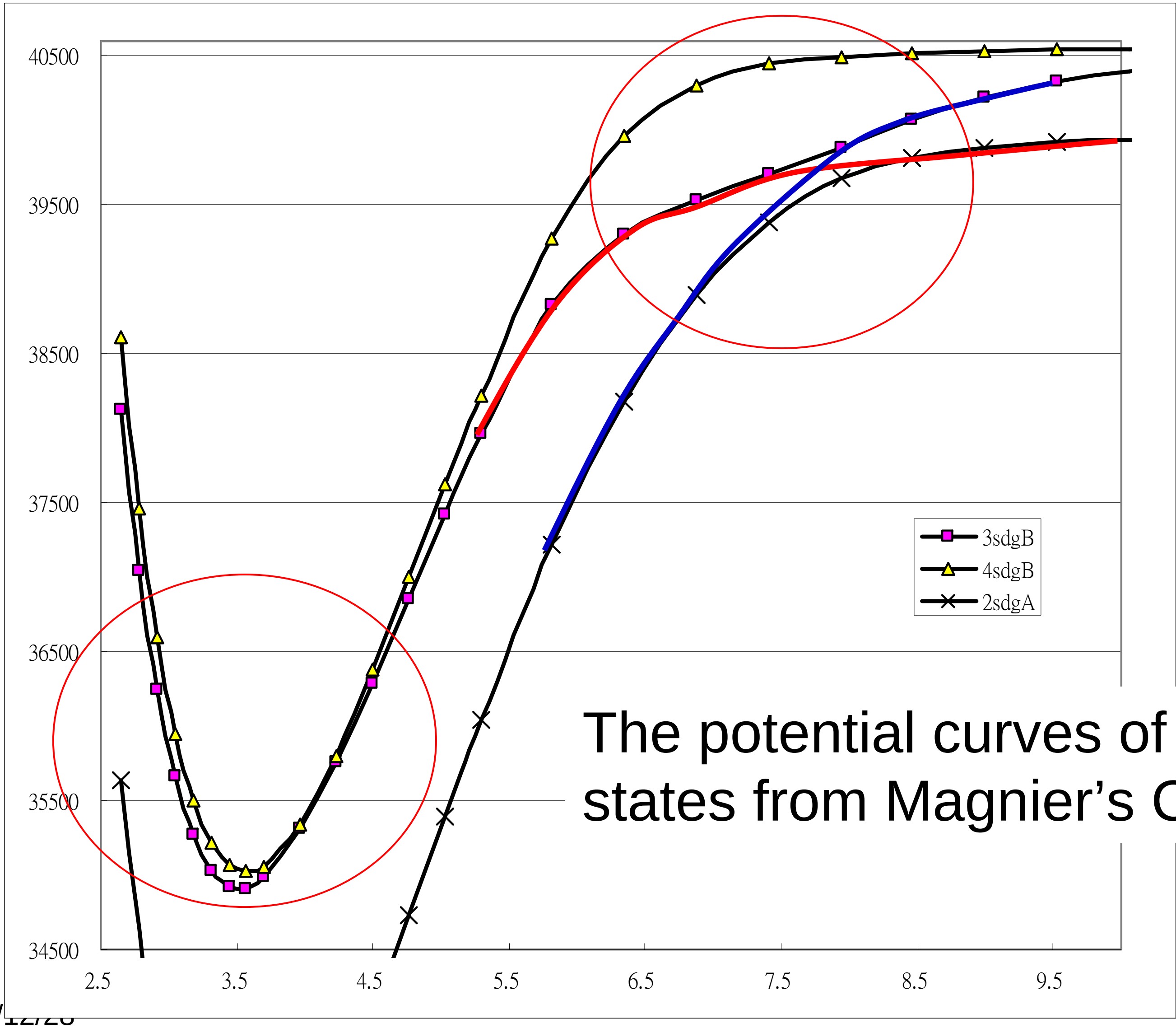
Laser Spectroscopy:

The third and fourth $^1\Delta_g$ states of Na_2 : a pair of twins

A typical part of fluorescence sticks spectrum converted from the raw fluorescence spectrum of the rovibrational progressions



The third and fourth $^1\Delta_g$ states of Na_2 : a pair of twins



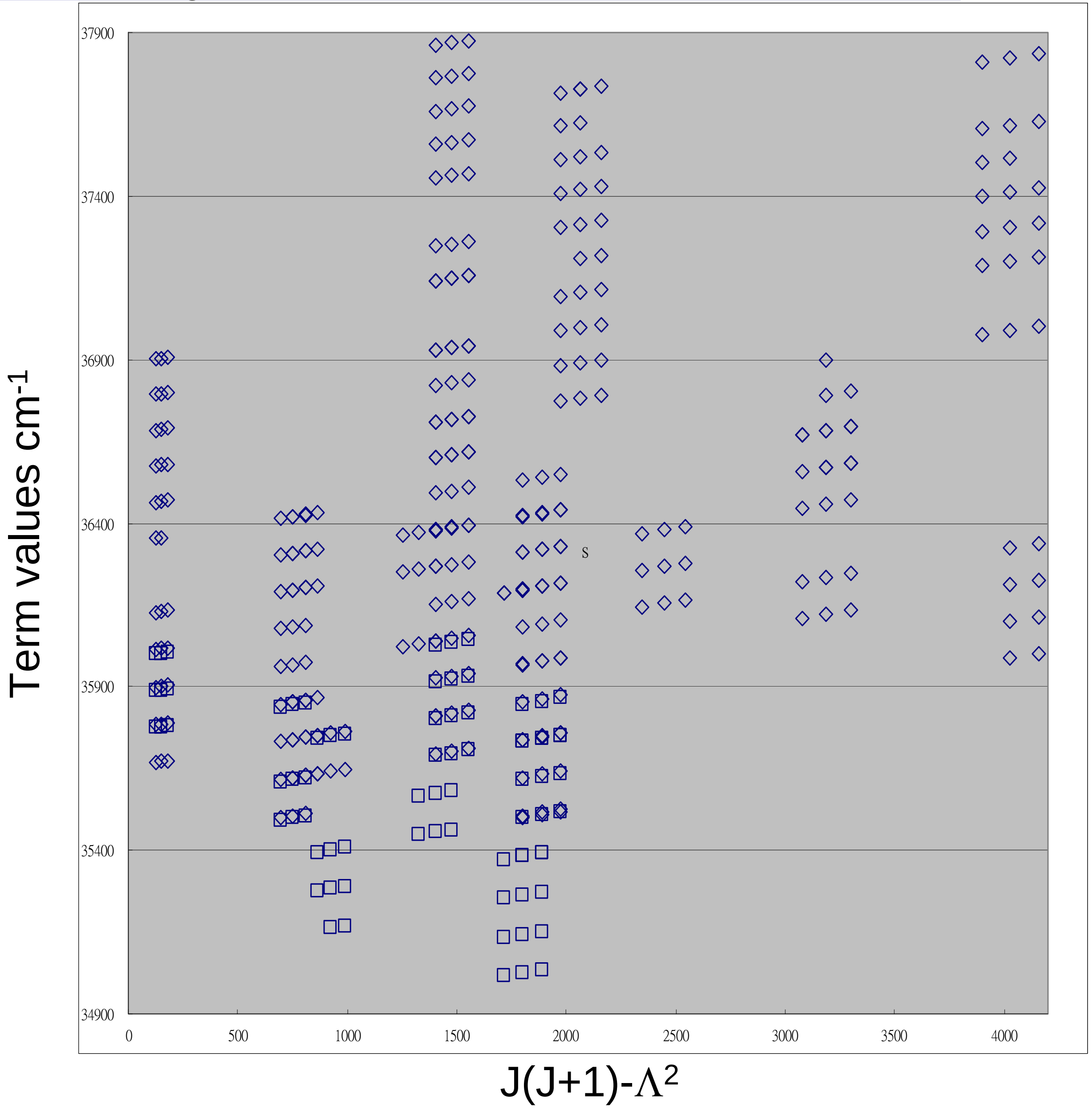
Na3s+Na4f

Na3s+Na4d

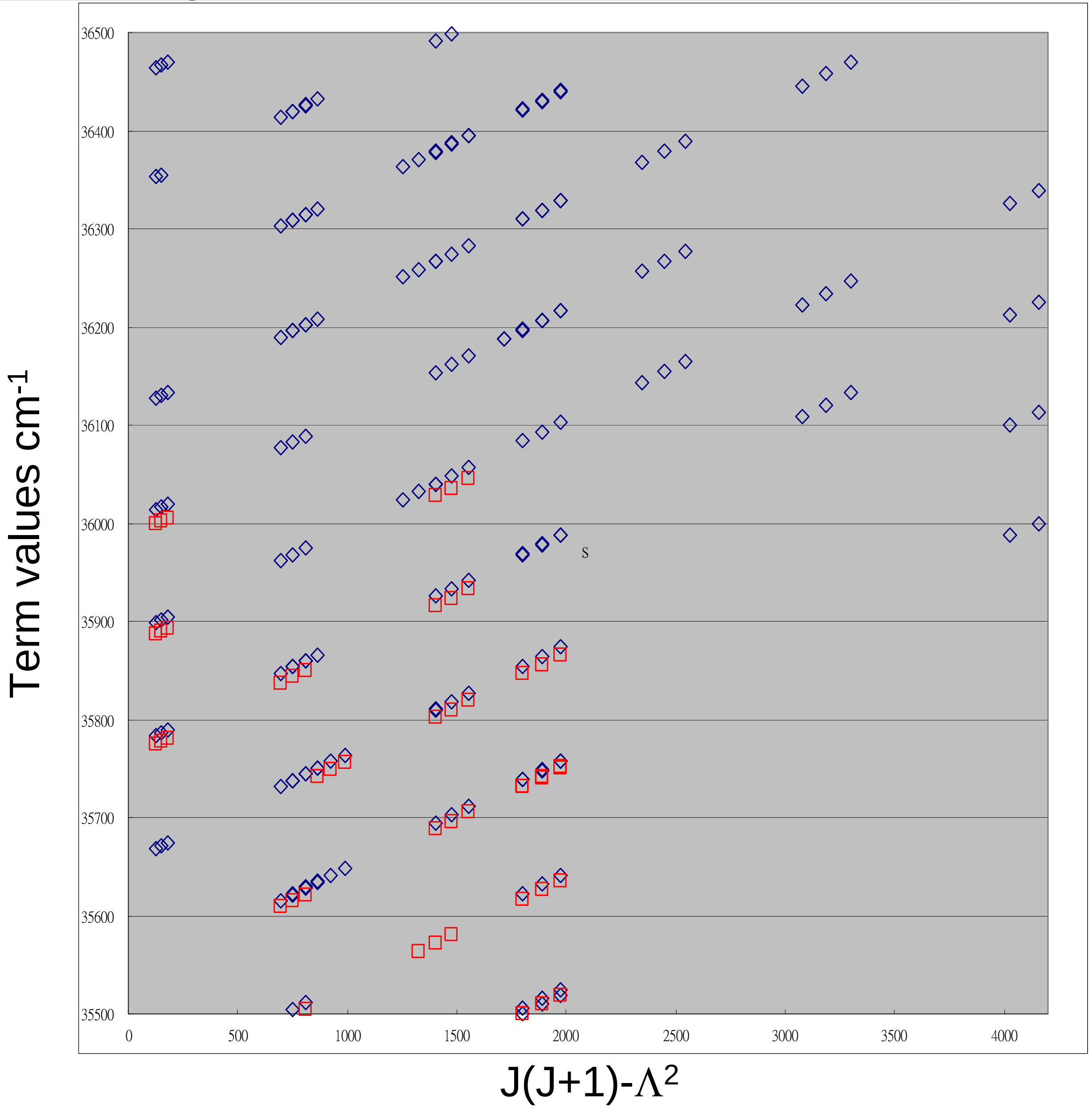
Na3p+Na3p

The potential curves of 2, 3, 4 $^1\Delta_g$ states from Magnier's Calculation.

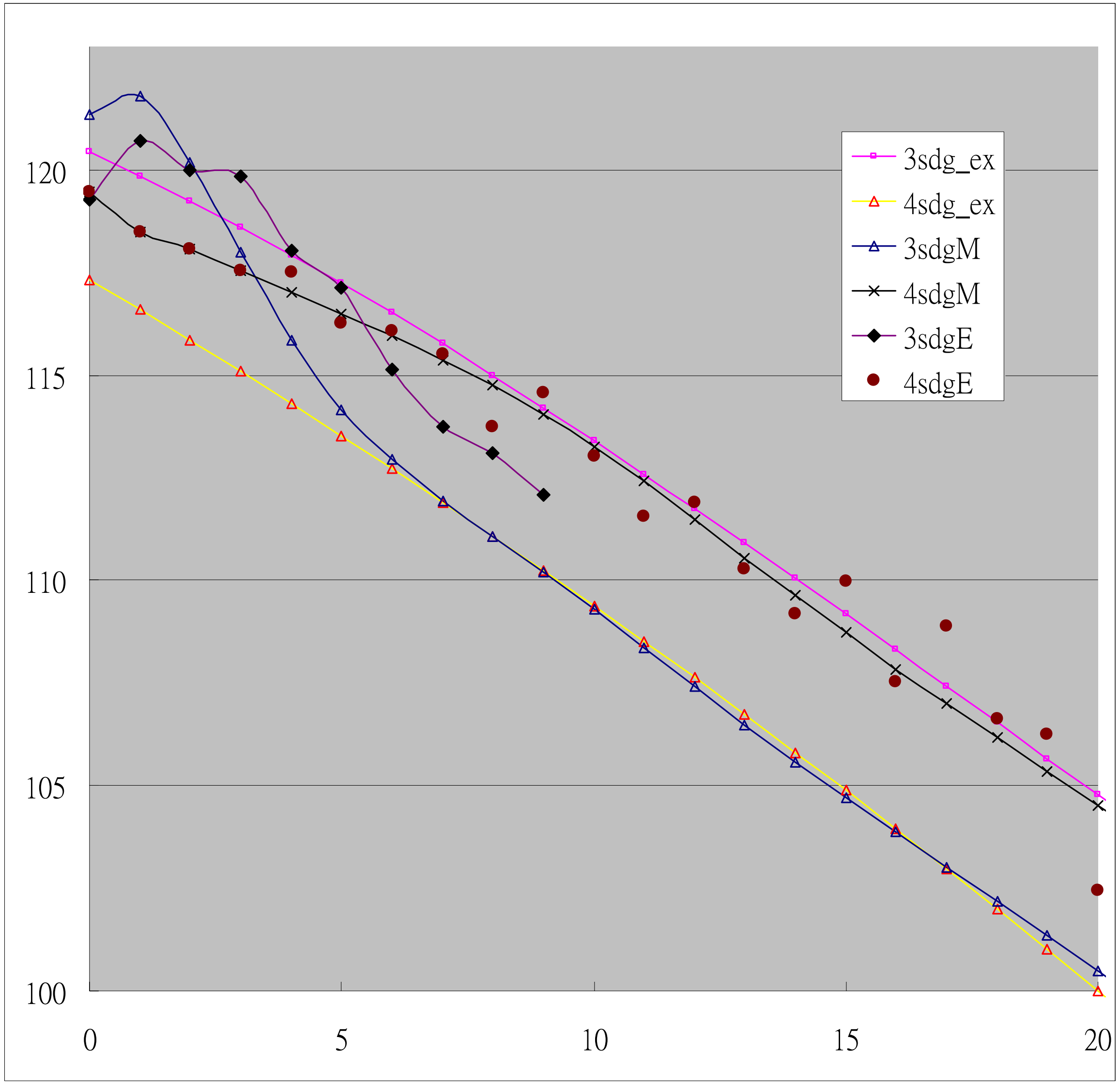
The third and fourth $^1\Delta_g$ states of Na_2 : a pair of twins



The third and fourth $^1\Delta_g$ states of Na_2 : a pair of twins



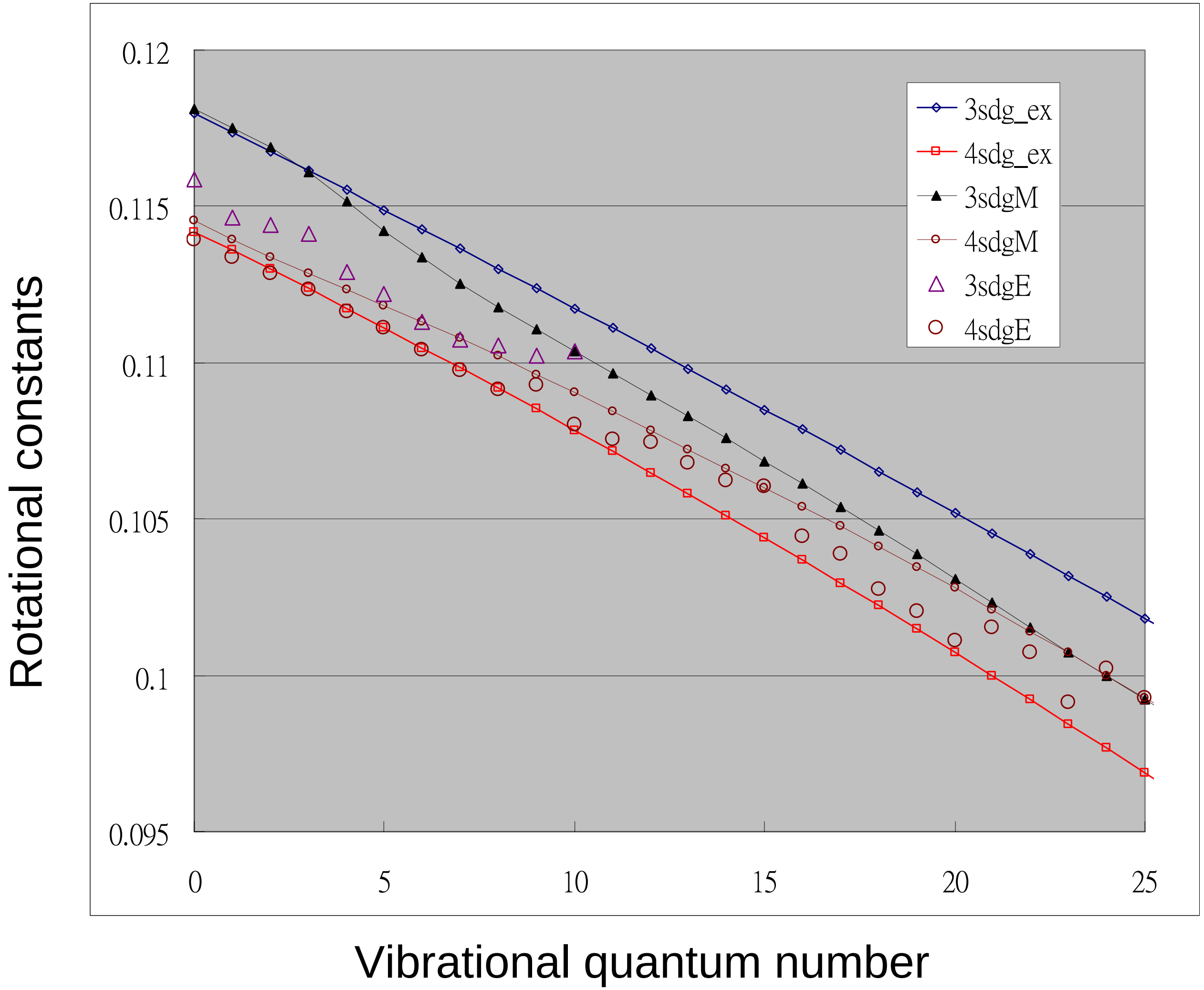
The third and fourth $^1\Delta_g$ states of Na_2 : a pair of twins



$\Delta G_{v+1/2}$

Vibrational quantum number

The third and fourth $^1\Delta_g$ states of Na_2 : a pair of twins

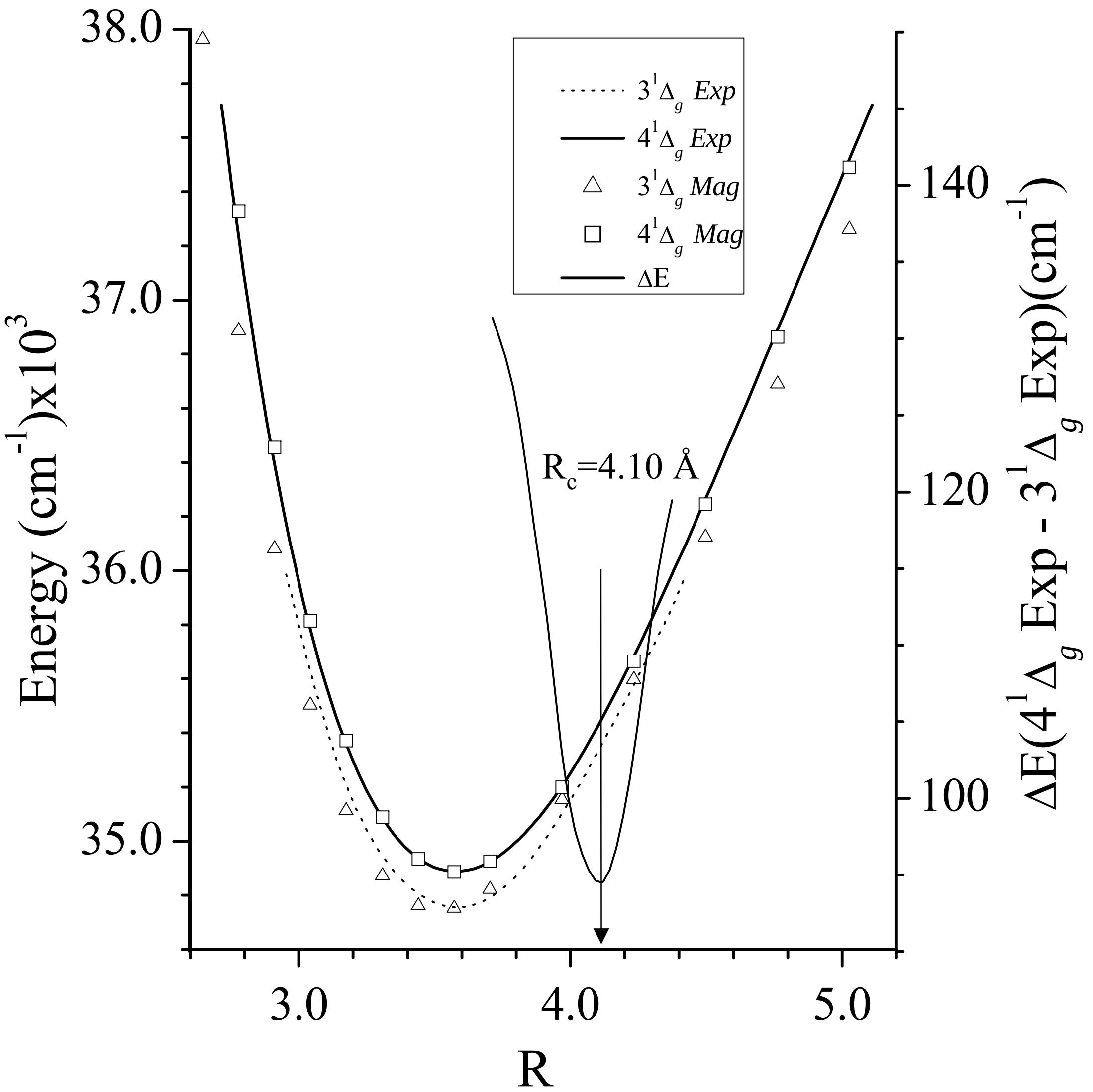


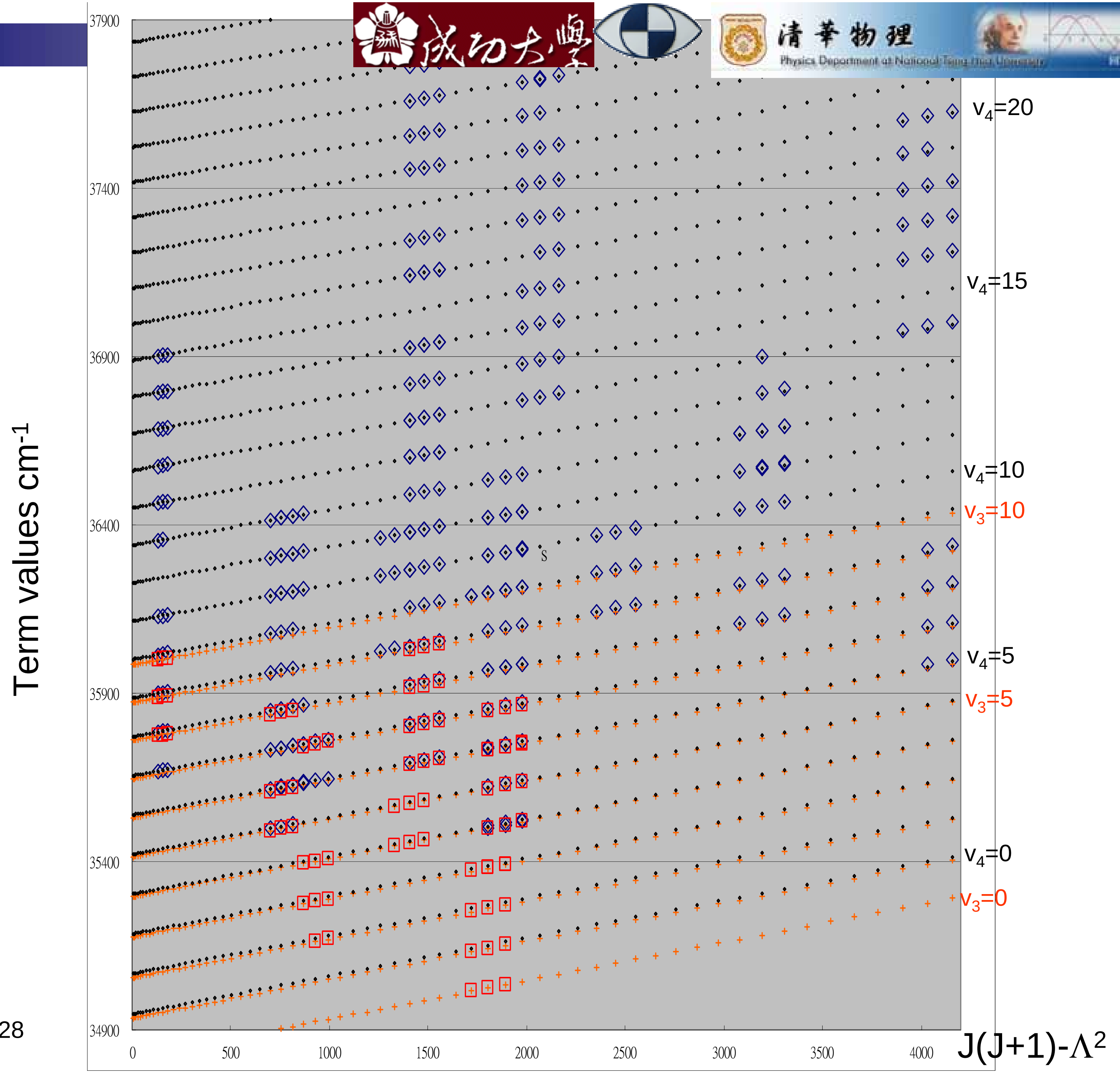
Wave-functions sharing

$$\psi_{3^1\Delta_g} = a\psi_{sd} + b\psi_{sf} \quad (3^1\Delta_g \; obseved \; in \; 0 \le v \le 9)$$
$$\psi_{4^1\Delta_g} = c\psi_{sd} + d\psi_{sf} \quad (4^1\Delta_g \; observed \; in \; 3 \le v \le 22)$$

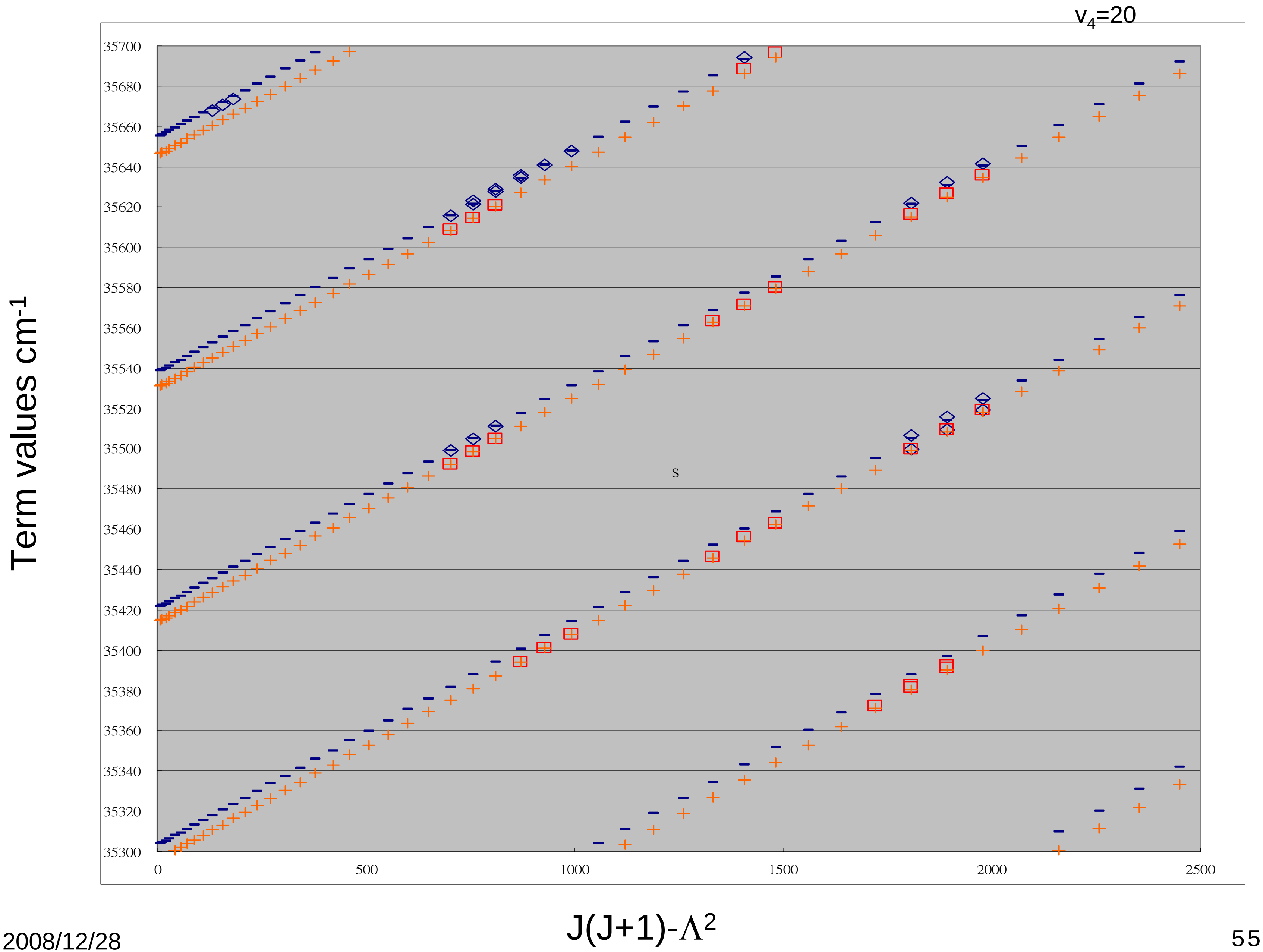
$$\begin{cases} |a|^2 > |b|^2, |d|^2 > |c|^2, & & E < E_c; \\ |b|^2 > |a|^2, |c|^2 > |d|^2, & \text{in the range of} & E > E_c; \\ |a|^2 \sim |b|^2, |c|^2 \sim |d|^2, & & E \sim E_c, \end{cases}$$

The third and fourth $^1\Delta_g$ states of Na_2 : a pair of twins





Term values cm-1
37900
37400
36900
36400
35900
35400
34900
0
500
1000
1500
2000
2500
3000
3500
4000
v4=20
v4=15
v4=10
v3=10
v4=5
v3=5
v4=0
v3=0
J(J+1)-Λ2



v_4=20
Term values cm-1
35700
35680
35660
35640
35620
35600
35580
35560
35540
35520
35500
35480
35460
35440
35420
35400
35380
35360
35340
35320
35300
0
500
1000
1500
2000
2500
s
J(J+1)-Λ2

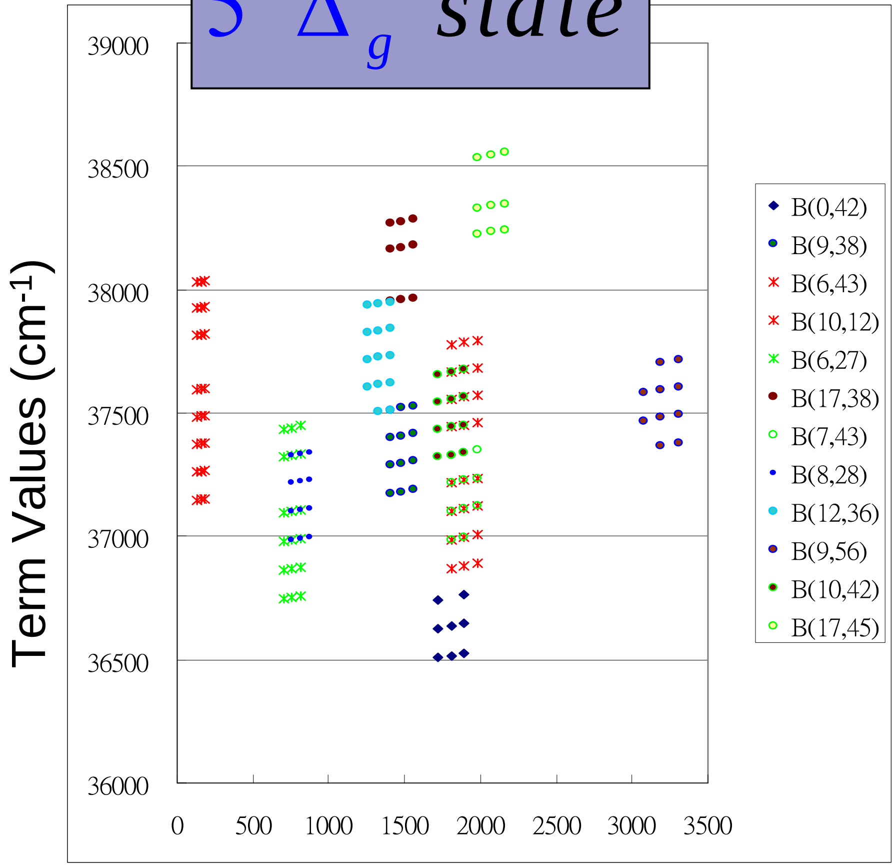
Analysis of

$5^1\Delta_g$ *state*

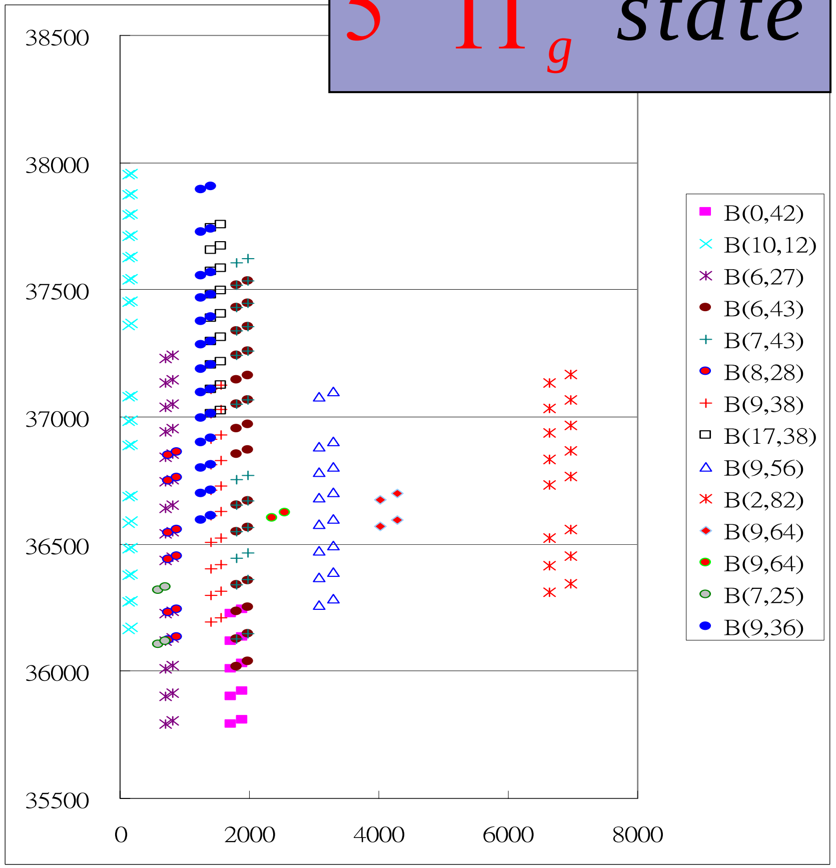
$5^1\Pi_g$ *state*

Data Fitting

$5\,^1\Delta_g$ *state*



$5\,^1\Pi_g$ *state*



$J(J+1) - \Lambda^2$

Least square fitting for Dunham Coefficients

The standard deviations excess the systematic accuracy*!!*

~0.03cm^{-1}

$5^1\Delta_g$ *state*	$5^1\Pi_g$ *state*
$\sigma = 0.27\ cm^{-1}$	$\sigma = 0.39\ cm^{-1}$

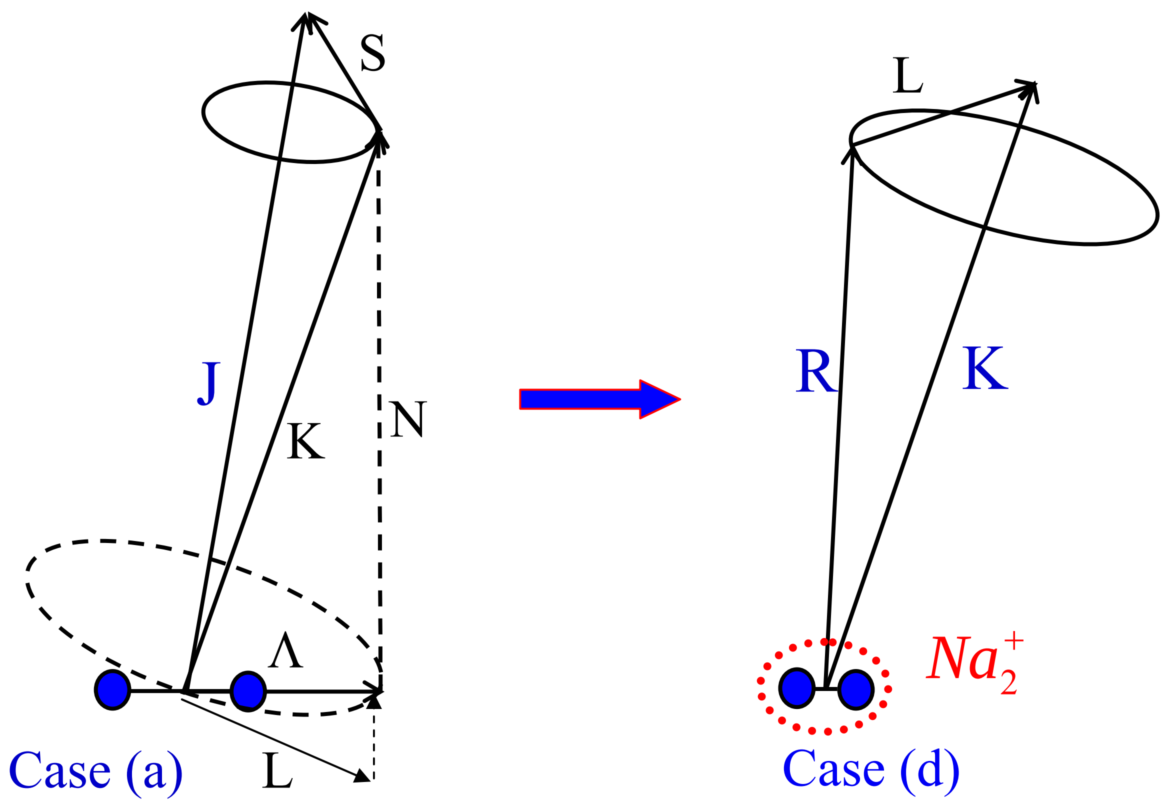
Is there something wrong!?

L-uncoupling

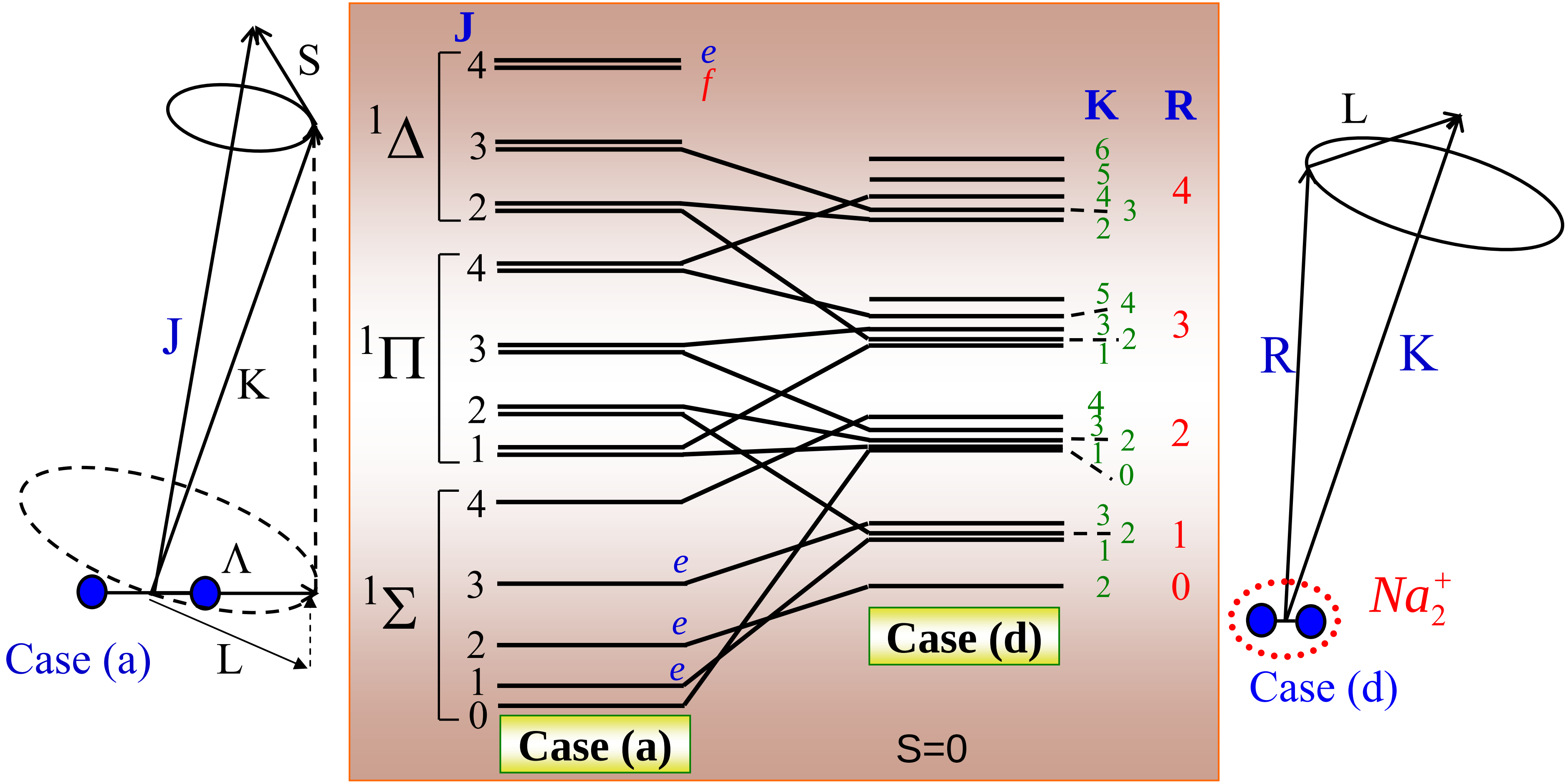
$5^1\Delta_g$ *state*

$5^1\Pi_g$ *state*

$3s+5d$



Coupling case transition



Separation of e/f levels

$$P, Q, R$$

$$\Delta J = -1, 0, 1$$

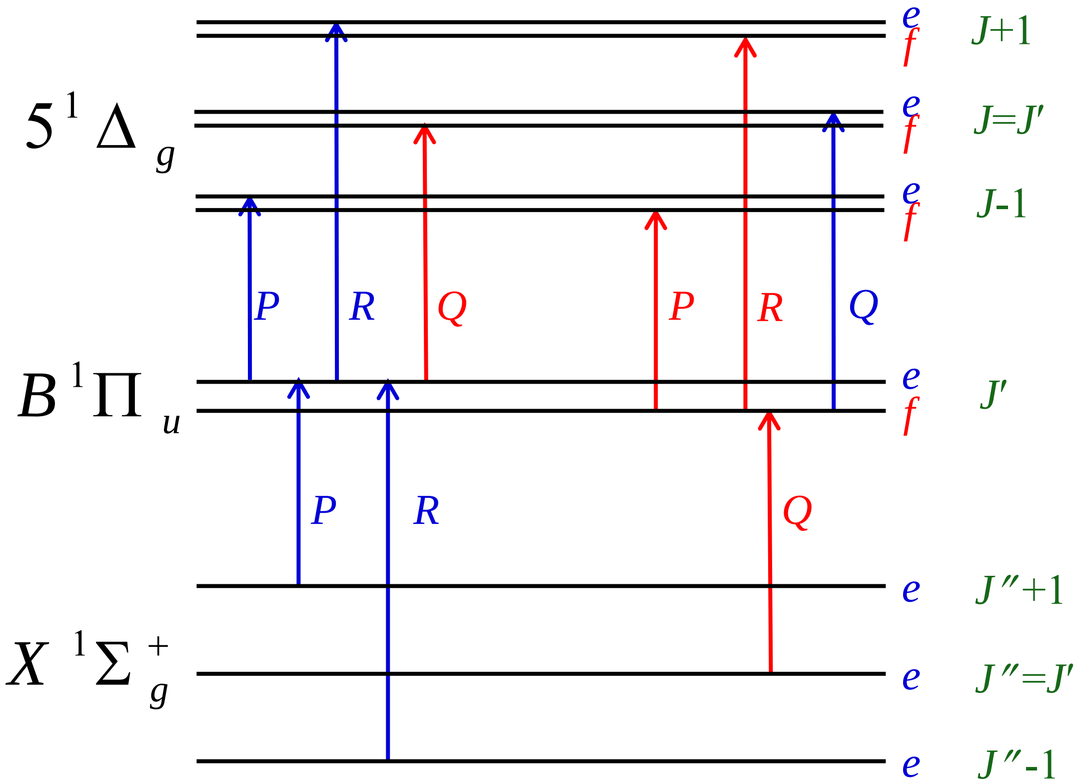
$$P, R$$

$$e \leftrightarrow e$$

$$f \leftrightarrow f$$

$$Q$$

$$e \leftrightarrow f$$

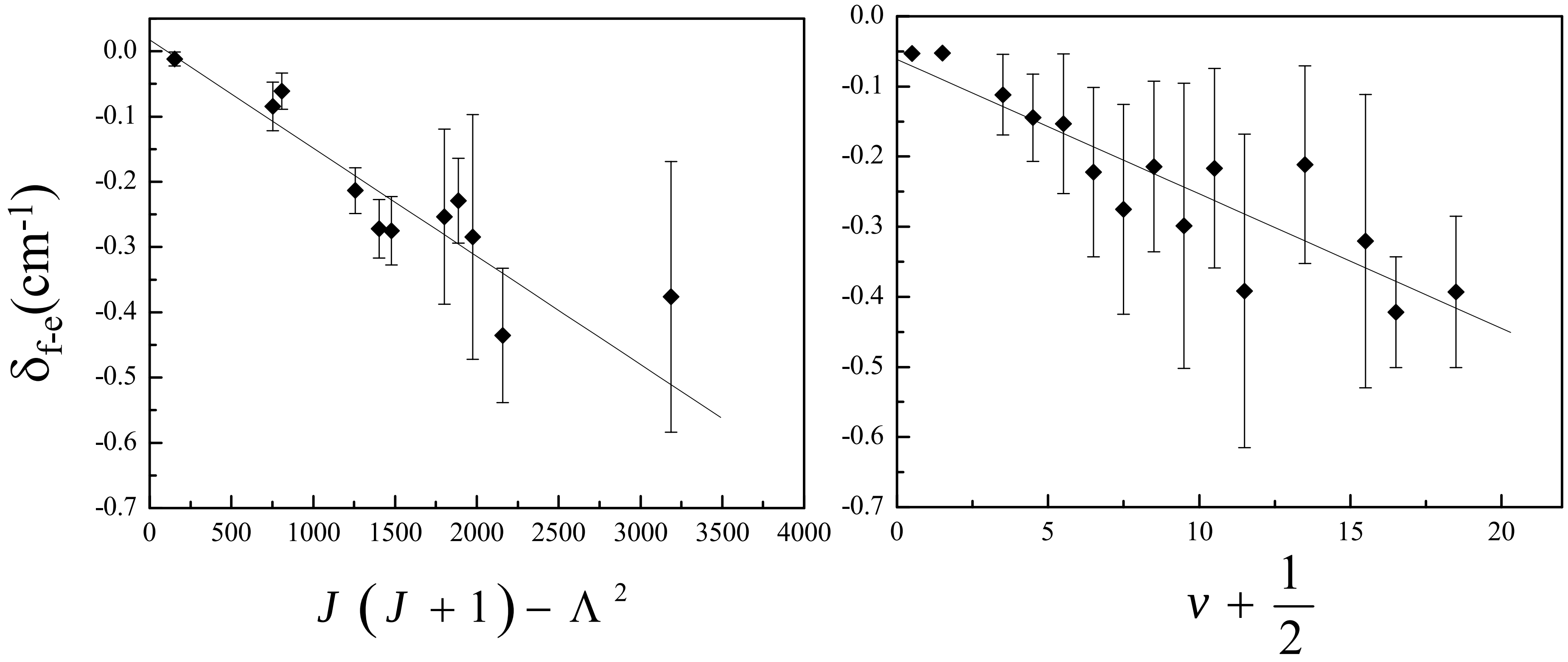


Case (a)

Dunham Coefficients for e-levels

$$T_{v,J} = \sum_{i=0}^{\infty} \sum_{j=0}^{\infty} Y_{ij} \left(v + \frac{1}{2} \right)^{i} [J(J+1) - \Lambda^{2}]^{j}$$

$5^1\Delta_g$ ： e/f level difference



Corrected Description

$$Term(v,J) = \sum Y_{ij}\left(v+\tfrac{1}{2}\right)^i [J(J+1)-\Lambda^2]^j$$
$$+\delta[J(J+1)-\Lambda^2]\left\{q_0 + q_v\left(v+\tfrac{1}{2}\right) + \mu[J(J+1)-\Lambda^2]\right\}$$

$$\delta = \begin{cases} 0 & \text{for } e \text{ parity} \\ -1 & \text{for } f \text{ parity} \end{cases}$$

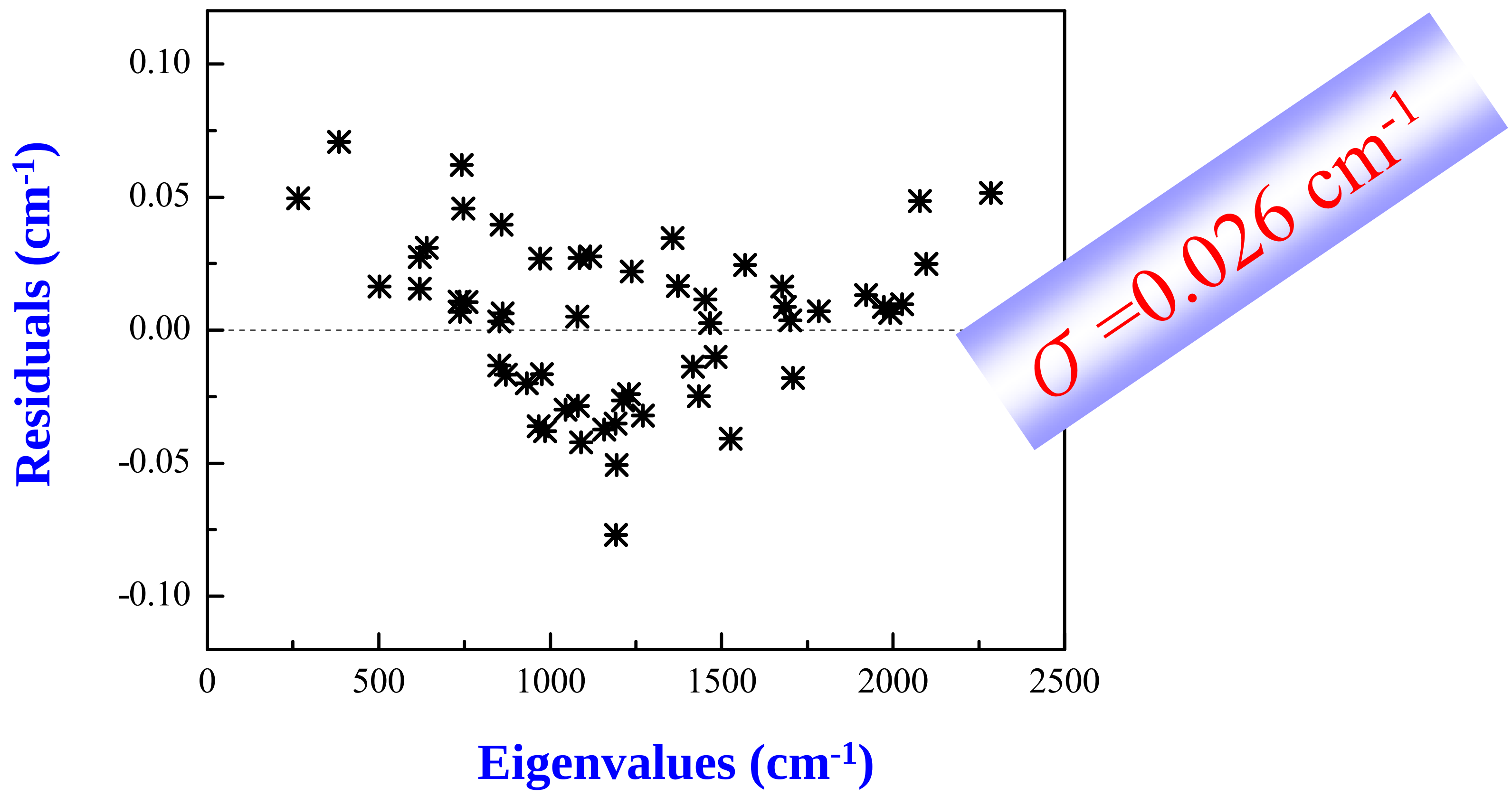
q_0 q_υ μ ⟹ **Corrected Constant**

$5^1\Delta_g$ ： Least Square Fitting

Y_{ij}	This work	Carlson	Mangier
Y_{00}	$0.3625049041\times10^{5}(0.12\times10^{0})$	$0.3625068\times10^{5}(0.2\times10^{0})$	35991
Y_{10}	$0.1208880160\times10^{3}(0.30\times10^{-1})$	$0.121014\times10^{3}(0.1\times10^{0})$	125.2
Y_{20}	$-0.4220256755\times10^{0}(0.25\times10^{-2})$	$-0.4185\times10^{0}(0.3\times10^{-1})$	
Y_{01}	$0.1151462792\times10^{0}(0.71\times10^{-4})$	$0.11386\times10^{0}(0.1\times10^{-3})$	
Y_{11}	$-0.5711260511\times10^{-3}(0.13\times10^{-4})$	$0.662\times10^{-3}(0.6\times10^{-4})$	
q_0	$-0.3764924100\times10^{-4}(0.90\times10^{-5})$		
q_v	$-0.1140212143\times10^{-4}(0.55\times10^{-6})$		
μ	$-0.7582958171\times10^{-8}(0.33\times10^{-8})$		

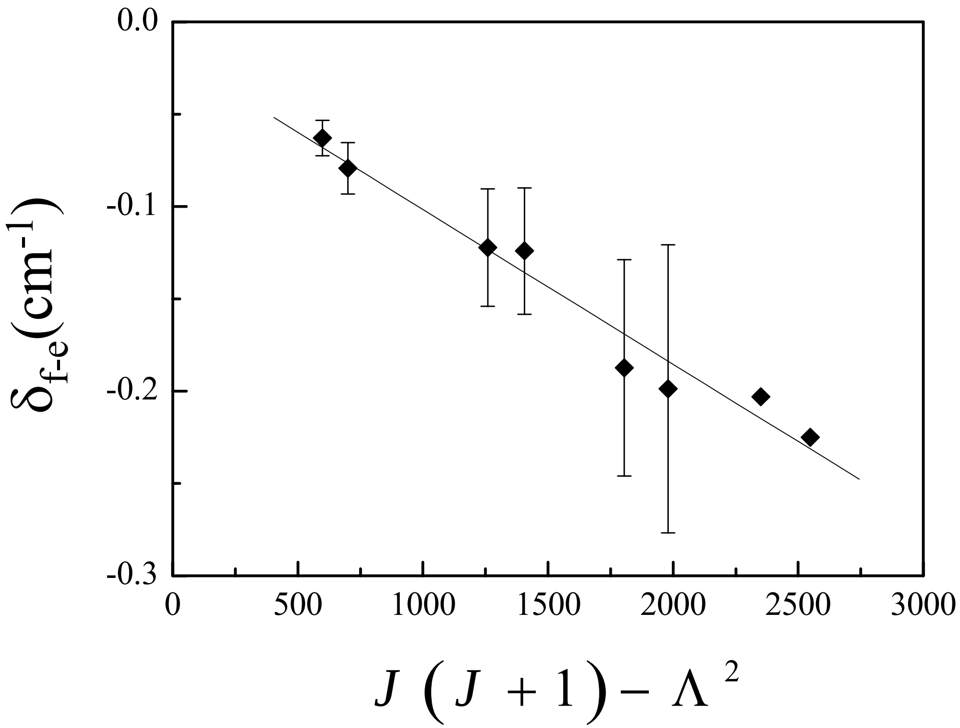
Splitting constants

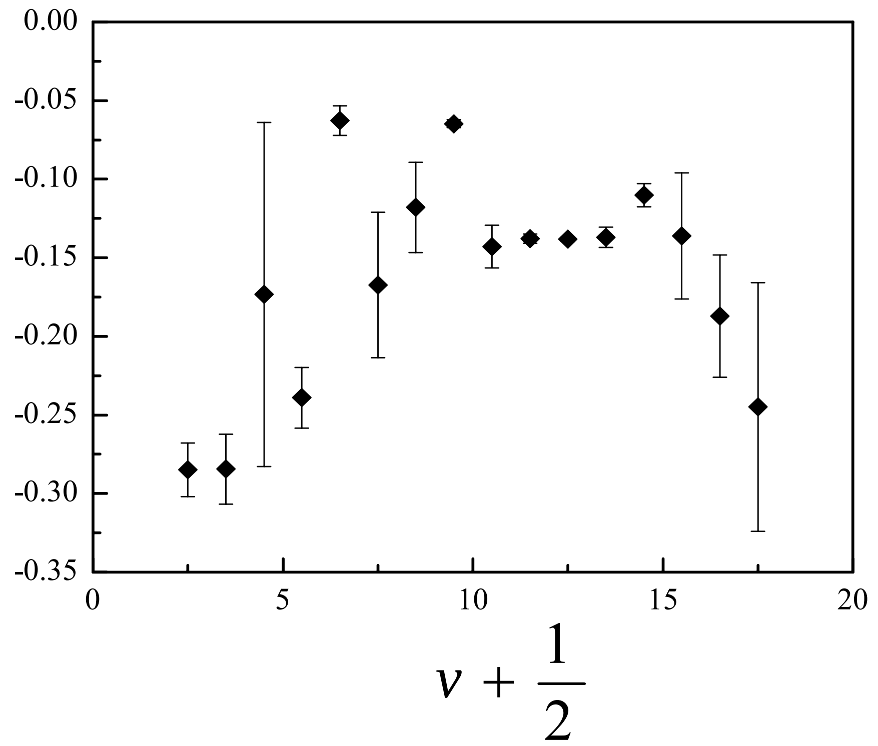
$5^1\Delta_g$ ： Regenerate the eigenvalue



R. Y. Chang, T. J. Whang, C. P. Cheng, and C. C. Tsai ,J. Chem. Phys. **123**, 224303 (2005).

$5^1 \Pi_g$ ： e/f level difference



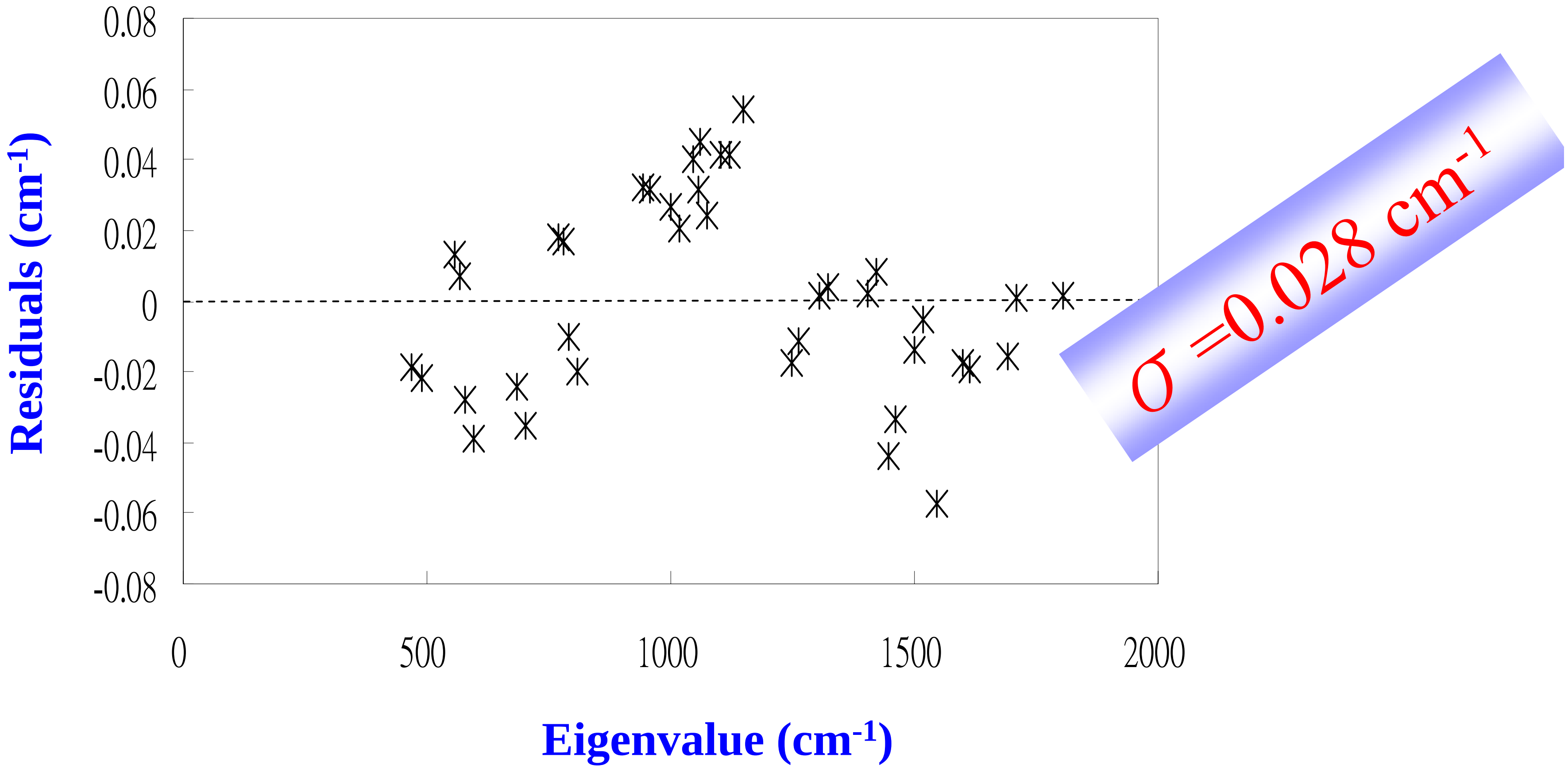


$5^1\Pi_g$ ： Least Square Fitting

Y_{ij}	This work	Carlson	Mangier
Y_{00}	$0.3555000951\times10^{5}(0.59\times10^{-1})$	$0.3555077\times10^{5}(0.3\times10^{0})$	35312
Y_{10}	$0.1120323209\times10^{3}(0.24\times10^{-1})$	$0.112128\times10^{3}(0.1\times10^{0})$	115.7
Y_{20}	$-0.4821718614\times10^{0}(0.38\times10^{-2})$	$-0.5112(0.1\times10^{-1})$	
Y_{01}	$0.1090586962\times10^{0}(0.20\times10^{-4})$	$0.10719(0.2\times10^{-3})$	
Y_{11}	$-0.5971091128\times10^{-3}(0.32\times10^{-5})$	$-0.541\times10^{-3}(0.5\times10^{-4})$	
q_0	$-0.1758320898\times10^{-3}(0.17\times10^{-4})$		
q_v	$0.7124318162\times10^{-5}(0.71\times10^{-6})$		
μ	$0.9608208561\times10^{-8}(0.80\times10^{-8})$		

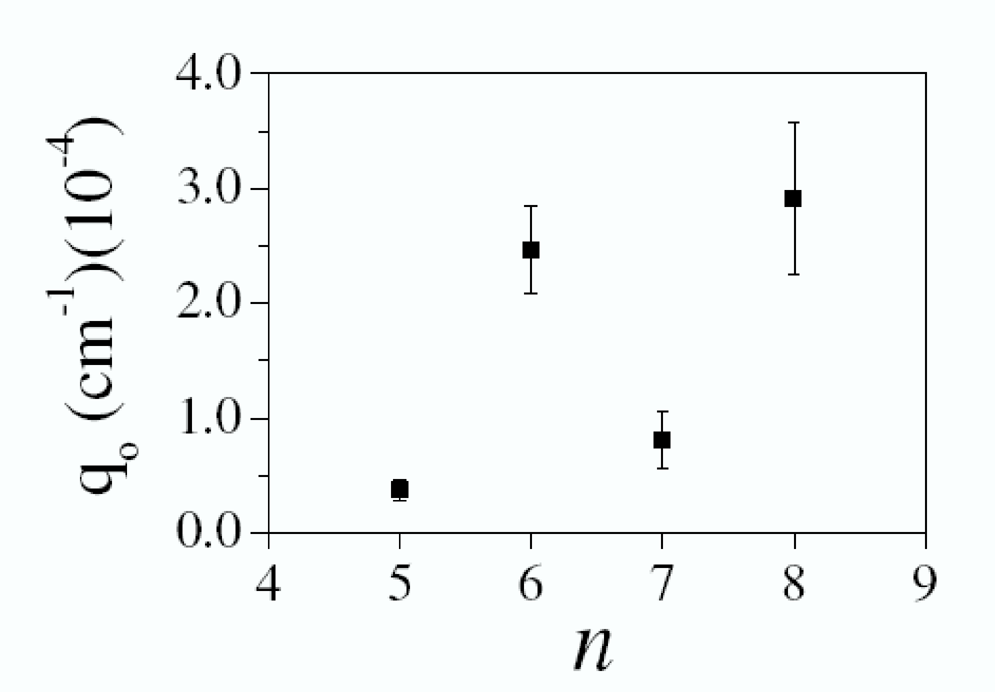
Splitting constants

$5^1\Pi_g$ ： *Regenerate the eigenvalue*



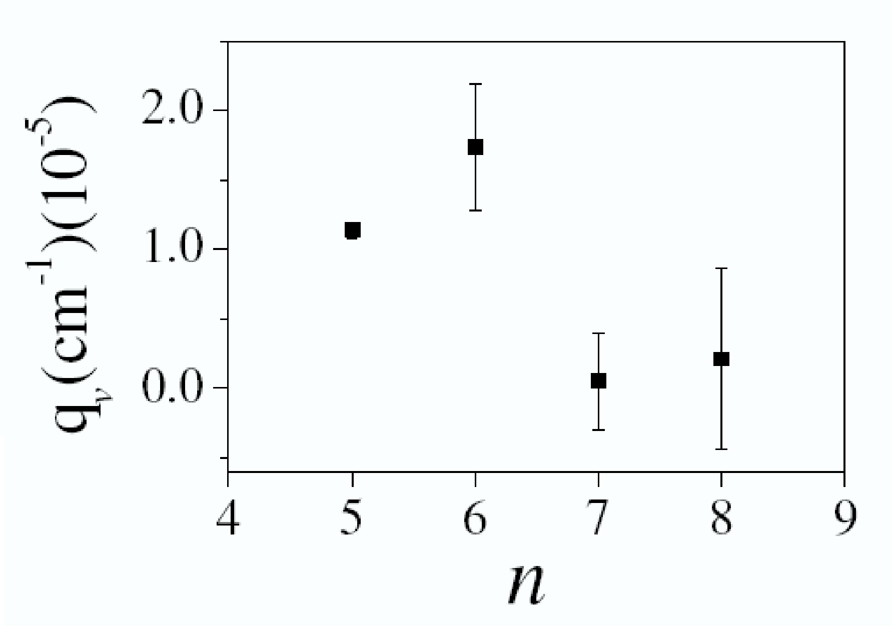
6d, 7d, 8d $^1\Delta_g$ *states*

Separation constants as a function of n in nd$^1\Delta_g$ states



$q_0[J(J+1)-\Lambda^2]$

$q_v[J(J+1)-\Lambda^2](v+1/2)$



Summary

Using high-resolution Laser spectroscopy

Electromagnetically induced transparency and de-coherence effects due to laser linewidth have been observed in Cesium atoms

Electronic orbital angular momentum *L* uncoupled from its internuclear axis and Molecular wavefunction-mixing and amplitude-sharing between the twins states (same symmetry) of Sodium dimer have been observed

Thanks for your attention!

Collaboration: Thou-Jen Whang, Chemistry, NCKU

Postdoctoral : Chanchal Chaudhuri

PhD Students: Ray-Yuan Chang 張瑞園
Yi-Chi Lee 李益志

And many master students……

我們的實驗室出現在 [成大簡介] 中的帥哥們…