



Modeling of CO-LIF

Hem Wadhar
UCLA

Mentor: Thomas Settersten
Physical Sciences Institute



Motivation

- CO LIF is a sensitive technique that can be used to measure concentrations of CO.
- CO is an important intermediate species and a pollutant in combustion.
- Quantitative measurements of CO concentrations are essential for understanding of flame chemistry and to reduce emissions.

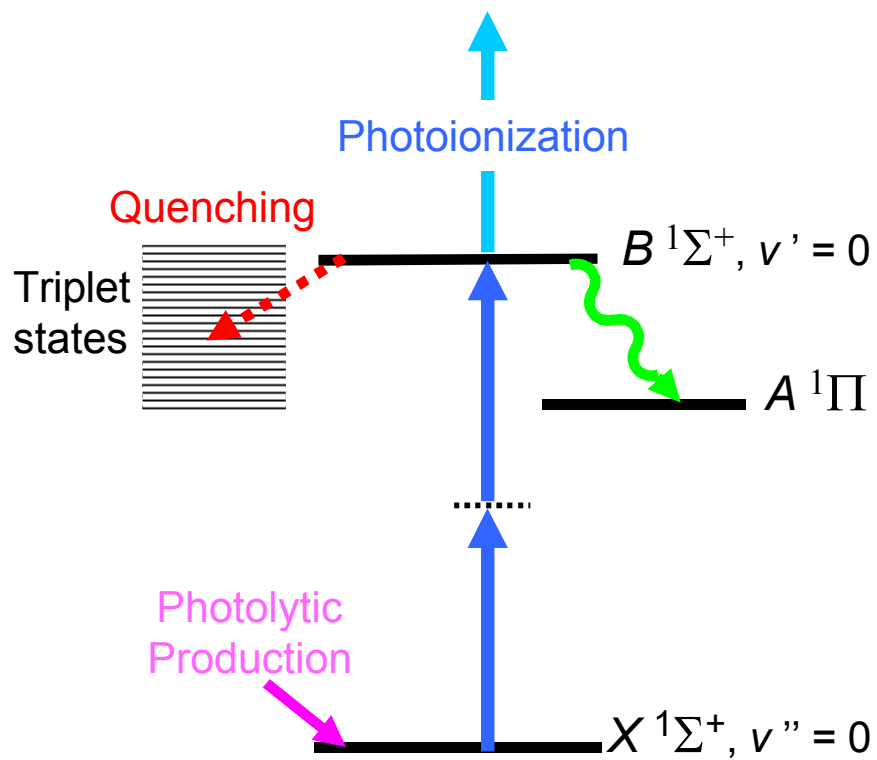
Background



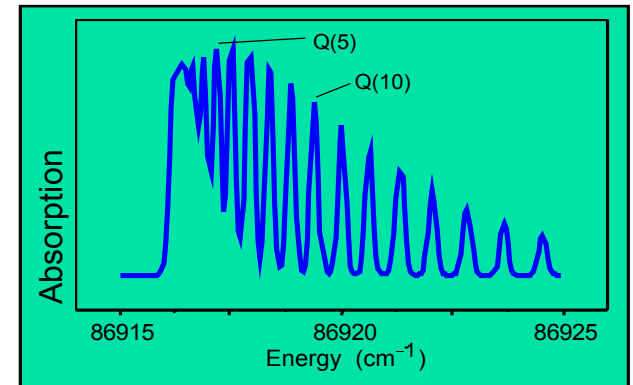
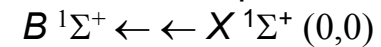
Planar LIF image of CO in flame
(CRF Advanced Imaging Laboratory)

- Two-Photon LIF is a potentially powerful CO diagnostic
 - “point” measurement
 - imaging
 - 100’s of ppm sensitivity
- Implementations at the CRF
 - single-point: TDF (Barlow)
 - line image: TCL (Barlow)
 - PLIF: AIL (Frank)

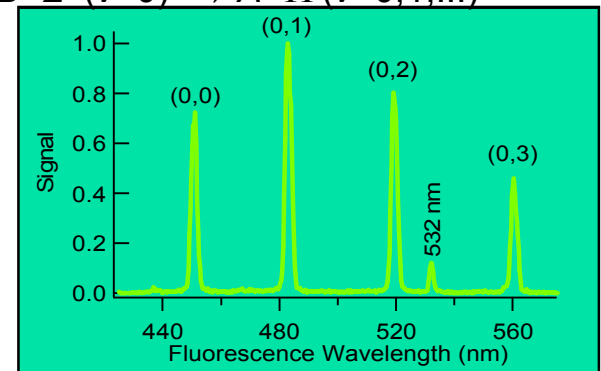
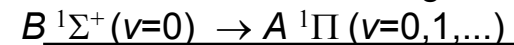
Background



TP Excitation via the Hopfield-Birge system



Fluorescence detection of the Angström bands





Predictive Modeling

- Develop computational simulations
- Considerations:
 - Photoionization
 - Quenching
 - Photolytic Production



Predictive Modeling

- Use Density Matrix Formalism to model the excitation and relaxation dynamics
- The population densities for rotational levels J in $X^1\Sigma^+(v=0)$ and J' in $B^1\Sigma^+(v'=0)$ are given by the diagonal density matrix elements ρ_J and $\rho_{J'}$
- Assume that coherences between levels such that $J \neq J'$ are zero.



Differential Equations

$$\dot{\rho}_J = -\Omega_J \beta_J - \rho_J \Gamma_J + \sum_i \rho_{J_i} R_{J_i J}$$

$$\dot{\rho}_{J'} = +\Omega_J \beta_J - \rho_{J'} \Gamma_{J'} + \sum_i \rho_{J'_i} R_{J'_i J'}$$

$$\dot{\alpha}_J = -\Delta \beta_J - (\gamma_J + \gamma_L) \alpha_J$$

$$\begin{aligned} \dot{\beta}_J = & +\Delta \alpha_J - (\gamma_J + \gamma_L) \beta_J \\ & +\Omega_J (\rho_J - \rho_{J'})/2 \end{aligned}$$

- α_j, β_j are the real and imaginary parts of the coherence.
- Ω_j is the time dependent Rabi frequency. It is a function of laser intensity.
- Δ is the two-photon laser detuning.
- Γ_j is the total collisional population loss rate.
- $\Gamma_{j'}$ is the total collisional population loss rate in addition to spontaneous emission, quenching and photoionization.
- γ_j is the coherence dephasing rate.
- R_{jj} is the rotational energy transfer rate.



Differential Equations

$$\dot{\rho}_{\text{ion}} = \left(\rho_B + \sum_i \rho_{J_i} \right) \sigma_{\text{ion}} \frac{I(t)}{\hbar \omega_L} ,$$

$$\dot{\rho}_{\text{lost}} = \left(\rho_B + \sum_i \rho_{J_i} \right) (Q + A) ,$$

$$\dot{\rho}_X = -\rho_X \Gamma_X + \sum_i \rho_{J_i} R_{J_i X} ,$$

$$\dot{\rho}_B = -\rho_B \Gamma_B + \sum_i \rho_{J'_i} R_{J'_i B} .$$

$$\dot{\rho}_1 = -W_{\text{ph}} \rho_1$$

$$\dot{S}_{\text{LIF}} = \left(\rho_B + \sum_i \rho_{J'_i} \right) A$$

- In addition we have to track the
 - CO- ion population
 - Population lost
 - "Bath" levels for X and B states.
- We also have CO₂ molecules photolytically creating CO (in the ground state)
- Finally S is the LIF signal generated by all of these equations.



Numerical Considerations

- In total we have
 - #Equations = 4(#Levels) + 7
 - Our system size can become large
- We use the Adams-Bashforth Method implemented by SUNDIALS

$$y_{n+1} = y_n + h \sum_{j=0}^s b_j f(t_{n-j}, y_{n-j}), \quad 0 \leq r \leq n$$

$$b_j = \frac{(-1)^j}{j!(s-j)!} \int_0^1 \prod_{i=0, i \neq j}^s (u+i) du, \quad j = 0, \dots, s.$$

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~/Desktop/test

hwadhar@bpatter-intern ~/Desktop/test
$ ./RET

=====
Photolysis Rate Coefficient: 0.040878 cm2/J
Photolysis Yield : 0.000000
Accessible CO2 fraction : 1.000000

Temperature : 1990.000000 K
Pressure : 10.000000 ATM
%CO Total : 0.000979
%CO2 Total : 0.075630
Mole Fraction in CO X(v=0) : 0.000770

Absorption Line Width : 1.727119 cm-1
-----> 2-gamma : 325.329066 /ns
2-gamma (pure dephasing) : 401.266437 /ns
RET% (J=0) : 73.186510 /ns
Q : 2.796060 /ns
R : 0.045200 /ns

using : 1 velocity groups
Laser tau : 5.000000 ns
Laser G(2) : 1.000000
Laser gamma (FWHM) : 0.000000 cm-1

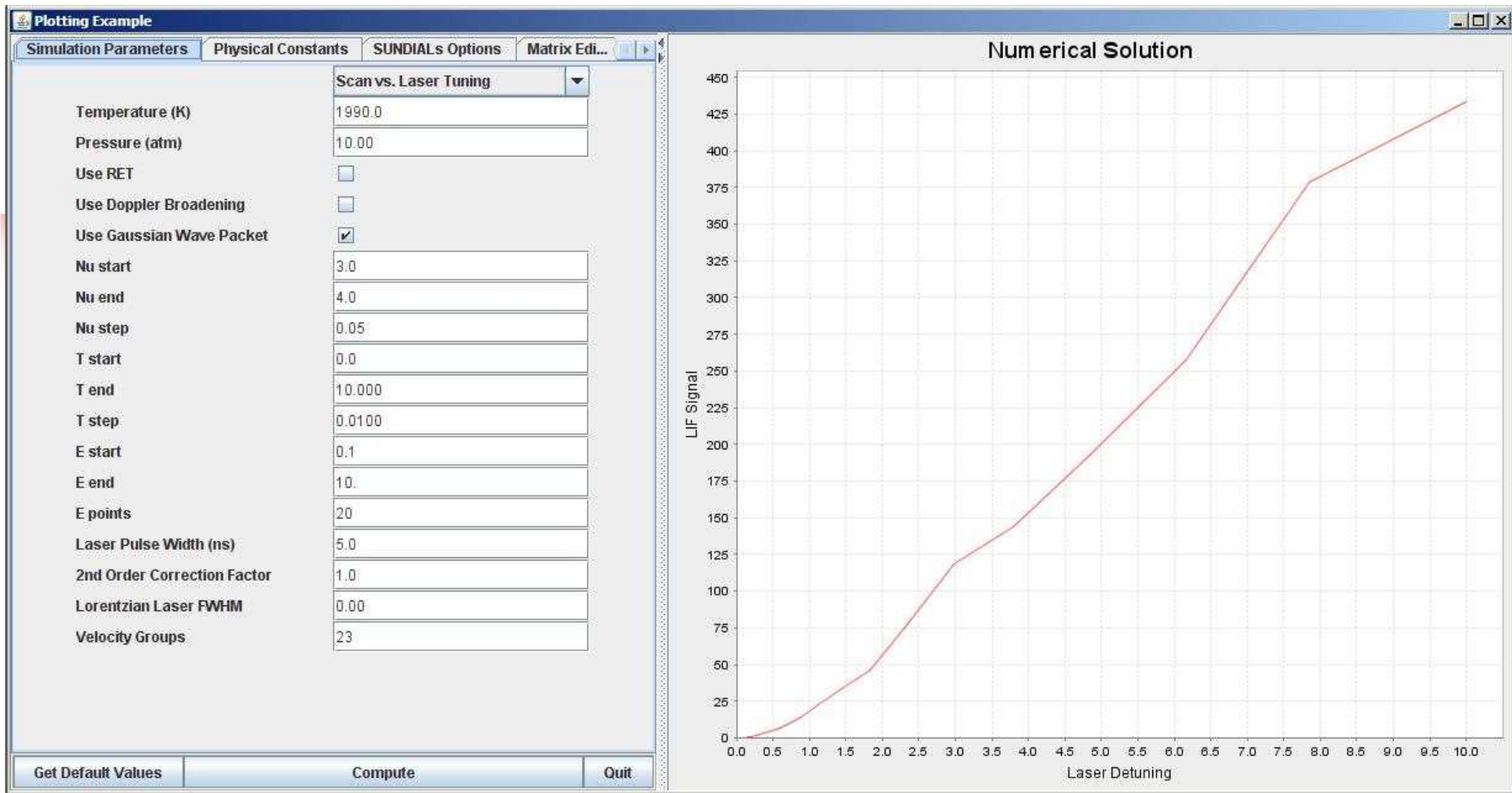
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Frequency scan with 20 points
i      detL      I0      LIF
=====
1      3.00      0.10000000  2.47889e-008
2      3.05      0.10000000  2.4792791e-008
3      3.10      0.10000000  2.4793735e-008
4      3.15      0.10000000  2.4793568e-008
5      3.20      0.10000000  2.4790697e-008
6      3.25      0.10000000  2.4786929e-008
7      3.30      0.10000000  2.4780465e-008
8      3.35      0.10000000  2.4773945e-008
9      3.40      0.10000000  2.476546e-008
10     3.45      0.10000000  2.4754506e-008
11     3.50      0.10000000  2.474317e-008
12     3.55      0.10000000  2.4729714e-008
13     3.60      0.10000000  2.471628e-008
14     3.65      0.10000000  2.4700054e-008
15     3.70      0.10000000  2.468418e-008
16     3.75      0.10000000  2.4665441e-008
17     3.80      0.10000000  2.4647828e-008
18     3.85      0.10000000  2.4627858e-008
19     3.90      0.10000000  2.4606903e-008
20     3.95      0.10000000  2.4584804e-008

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$

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- Terminal Window Program
 - Programmed in C
 - Runs very fast



- JAVA GUI and Plotting Routines
- Platform Independent and portable
- Uses the Java native interface (JNI) to link with the C program to produce data