Noise-Immune Cavity-Enhanced Optical Heterodyne Molecular Spectrometry: Modeling under Saturated Absorption

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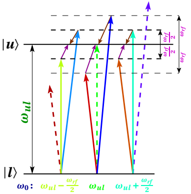


Outline

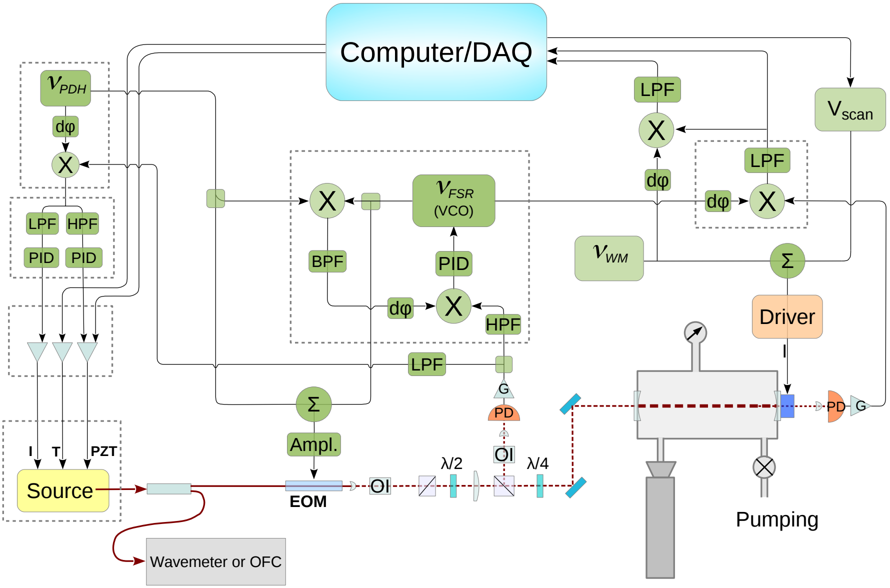
Introduction

- **Noise-Immune Cavity-Enhanced Optical Heterodyne Molecular Spectrometry vs Cavity RingDown Spectroscopy**
- Both Absorption-based Techniques (in High Finesse Cavity)
  - Heterodyne (Absorbance) vs. Response Time (Absorption) Detection
  - Continuous vs. Discontinuous Acquisition (LOD),
  - “Trichromatic” vs. “Monochromatic” EMF
  - Ultimate Sensitivity: Photon-Shot-Noise Limited
  - Lockings (Cavity, ...)
- “Weak” Absorption (Optically Thin Medium)
- Saturation: Lamb Dip, Crossover Resonances vs. Cross-Sideband Resonances, etc...

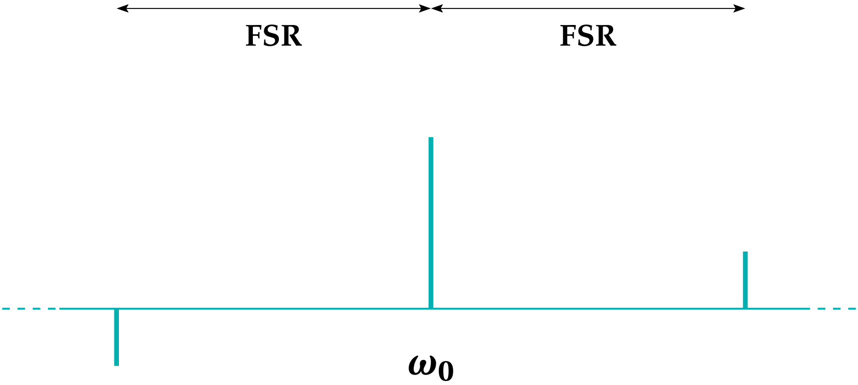
NICE-OHMS Fundamentals

- **Combining both advantages:**
  - **Radio-Frequency Modulation: Noise Immunity and Photon Shot Noise Ultimate Limit**
  - **High Finesse Cavity: i.e., Long Equivalent Absorption Length (> km), Narrow Bandwidth**
- **High EMF Trapped**
  - **NonLinear Couplings**
- **Phase Modulation of the Source**

A Typical Setup



NICE-OHMS EMF



Phase Modulations in NICE–OHMS

Weak Absorption in a Resonant Cavity

Equivalent Absorption Length:

$$L_{eq} = \frac{2\,\mathscr{F}\,L_{cav}}{\pi}$$

Cavity "Bandwidth":

$$\Delta_\omega = \frac{FSR}{\mathscr{F}}$$

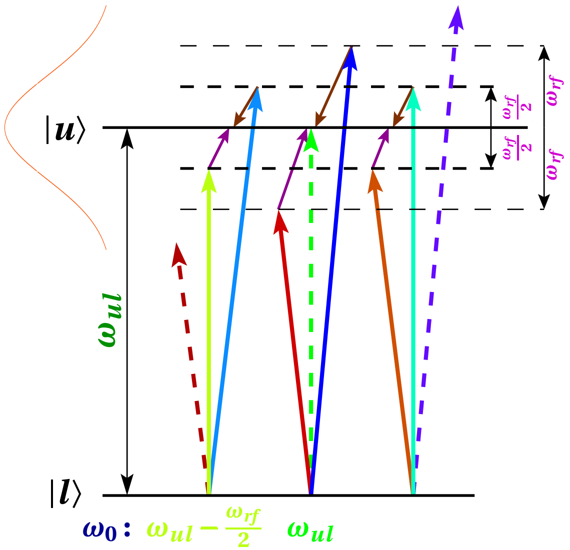
EM Field Enhancement at resonance ($\mathscr{R}_{cav} \sim 0$):

$$\mathscr{I}_{cav} = \frac{\mathscr{F}}{\pi}\mathscr{I}_{in}$$

Photon noise:

$$\sqrt{\frac{2\,e\,\Delta_\nu}{\eta\,\langle P\rangle}} = \left(\alpha L_{eq}\right)_{min}$$

Interaction with a TriChromatic EMF in Cavity



Background

- **"2-level System"**
  - Analytical Solution of the Liouville Equation in the Frequency Domain
  - NonLinear Polarization in the Frequency Domain
  - 2-step Iteration (Moderate Saturation)
  - Multichromatic EMF (forth and back), Dirac or Lorentzian Profile
  - Numerical Integration over the Doppler Shift (Boltzmann Distribution)
  - Lamb Dips and Cross-Sideband Resonances
  - Zeeman Structure and EMF Polarization (σ or π)
  - Zeeman Sublevels with a unique Relaxation Time
  - Transit Time Approximation
  - Direct Absorption, "Absorption-like", "Dispersion-like"
  - Comparison with O. Axner Group Data
- **Approximated Model**
  - Zeeman Sub-transitions (Averaged "Saturation Coefficient")
- **Not in the Model**
  - Strong Saturation
  - N-level System
- **Recovering Dipole Moment and Number Density**

Solutions

$$\Delta\mathscr{P}^{(DC)}_{m_u m_l}(\omega_0,\delta) = \mathscr{P}_0\, l_{eq} \begin{pmatrix} F_u & 1 & F_l \\ -m_u & \Delta m & m_l \end{pmatrix}^2 \times$$

$$\frac{J_0^2(\xi)\,\mathrm{d}\alpha^{(0)}_{ul}(\omega_0,\delta) + J_1^2(\xi)\left[\mathrm{d}\alpha^{(0)}_{ul}\left(\omega_0+\omega_{rf},\delta\right) + \mathrm{d}\alpha^{(0)}_{ul}\left(\omega_0-\omega_{rf},\delta\right)\right]}{1+3\begin{pmatrix} F_u & 1 & F_l \\ -m_u & \Delta m & m_l \end{pmatrix}^2 S_{ul}(\omega_0,\delta)}$$

$$\mathrm{d}\mathscr{P}^{(\cos)}_{m_u m_l}(\omega_0,\delta) = J_0(\xi)\,J_1(\xi)\,\mathscr{P}_0\, l_{eq} \times \begin{pmatrix} F_u & 1 & F_l \\ -m_u & \Delta m & m_l \end{pmatrix}^2 \times$$

$$\frac{\mathrm{d}\alpha^{(0)}_{ul}\left(\omega_0+\omega_{rf},\delta\right) - \mathrm{d}\alpha^{(0)}_{ul}\left(\omega_0-\omega_{rf},\delta\right)}{1+3\begin{pmatrix} F_u & 1 & F_l \\ -m_u & \Delta m & m_l \end{pmatrix}^2 S_{ul}(\omega_0,\delta)}$$

$$\mathrm{d}\mathscr{P}^{(\sin)}_{m_u m_l}(\omega_0,\delta) = J_0(\xi)\,J_1(\xi)\,\mathscr{P}_0\, l_{eq} \times \begin{pmatrix} F_u & 1 & F_l \\ -m_u & \Delta m & m_l \end{pmatrix}^2 \times$$

$$\frac{2\mathrm{d}\phi^{(0)}_{ul}(\omega_0,\delta) - \mathrm{d}\phi^{(0)}_{ul}\left(\omega_0+\omega_{rf},\delta\right) - \mathrm{d}\phi^{(0)}_{ul}\left(\omega_0-\omega_{rf},\delta\right)}{1+3\begin{pmatrix} F_u & 1 & F_l \\ -m_u & \Delta m & m_l \end{pmatrix}^2 S_{ul}(\omega_0,\delta)}$$

And (Numerical Sums and Integrals):

$$\Delta\mathscr{P}_{ul}^{(DC)}(\omega_0) = 3\sum_{m_u}\sum_{m_l}\int \Delta\mathscr{P}_{m_u m_l}^{(DC)}(\omega_0,\delta)$$

$$\mathscr{P}_{ul}^{(\cos)}(\omega_0) = 3\sum_{m_u}\sum_{m_l}\int \mathrm{d}\mathscr{P}_{m_u m_l}^{(\cos)}(\omega_0,\delta)$$

$$\mathscr{P}_{ul}^{(\sin)}(\omega_0) = 3\sum_{m_u}\sum_{m_l}\int \mathrm{d}\mathscr{P}_{m_u m_l}^{(\sin)}(\omega_0,\delta)$$

$$\Omega_{ul}^2 = \frac{\mathscr{F}}{\pi}\left|\frac{E_0\mu_{ul}}{\hbar}\right|^2$$

$$\mathrm{d}\mathscr{N}_0(\delta) = \mathscr{N}_0\sqrt{\frac{\ln 2}{\pi}}\frac{1}{\Delta_D}\mathrm{e}^{-\ln 2\left(\frac{\delta}{\Delta_D}\right)^2}\mathrm{d}\delta$$

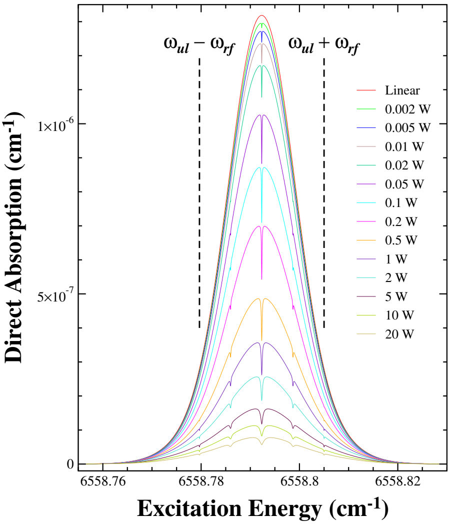
Approximations

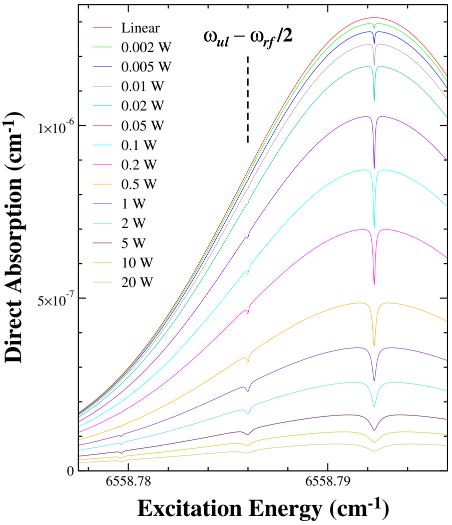
$$3\begin{pmatrix} F_u & 1 & F_l \\ -m_u & \Delta m & m_l \end{pmatrix}^2 S_{ul}(\omega_0,\delta) =$$

$$2\Omega_{ul}^2 \frac{\gamma_{ul} T_1}{d g_{rot_l}} \sum_{\kappa} \left[\frac{1}{\left(\omega_{ul} - \omega_0 + \kappa\omega_{rf} + \delta\right)^2 + \gamma_{ul}^2} + \frac{1}{\left(\omega_{ul} - \omega_0 + \omega_{rf} - \delta\right)^2 + \gamma_{ul}^2} \right]$$

Simulation C_2H_2: R_0 ($I = 0$): Direct Absorption (σ Polar.)

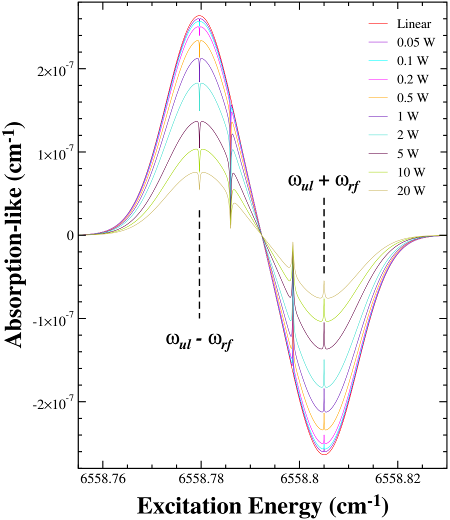
$1_0^1 3_0^1$ ($\Sigma_u^+ \leftarrow \Sigma_g^+$), $\mu = 10.7$ mD, $\omega_{rf} = 380$ MHz, $\xi = 0.4$ (1 mTorr, $w_0 = 0.45$ mm, $\Gamma_{rel.} = 1$ MHz)

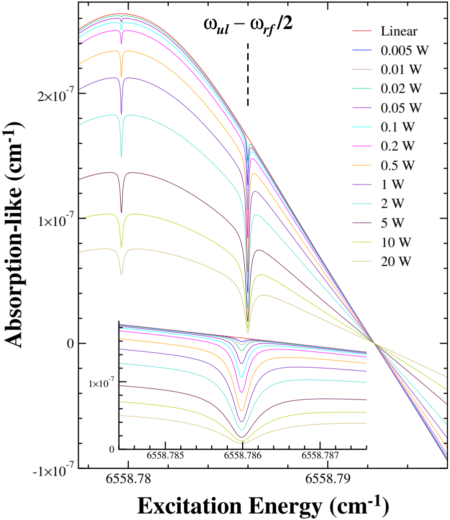




Simulation C_2H_2: R_0 ($I = 0$): Absorption-like (σ Pol.)

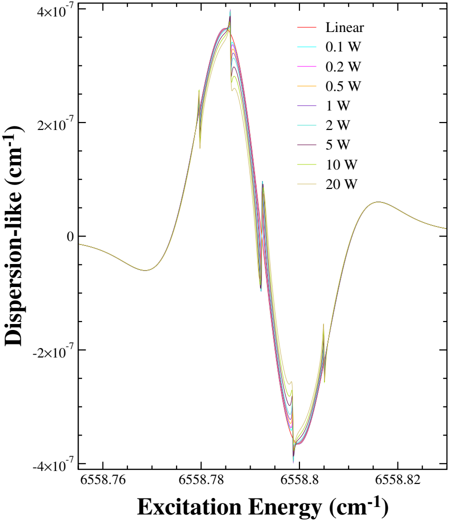
$1_0^1 3_0^1$ ($\Sigma_u^+ \leftarrow \Sigma_g^+$), $\mu = 10.7\,\mathrm{mD}$, $\omega_{rf} = 380\,\mathrm{MHz}$, $\xi = 0.4$ (1 mTorr, $w_0 = 0.45\,\mathrm{mm}$, $\Gamma_{rel.} = 1\,\mathrm{MHz}$)

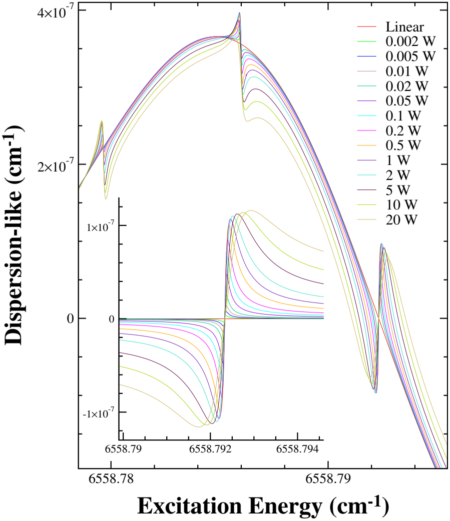




Simulation C_2H_2: R_0 ($I = 0$): Dispersion-like (σ Pol.)

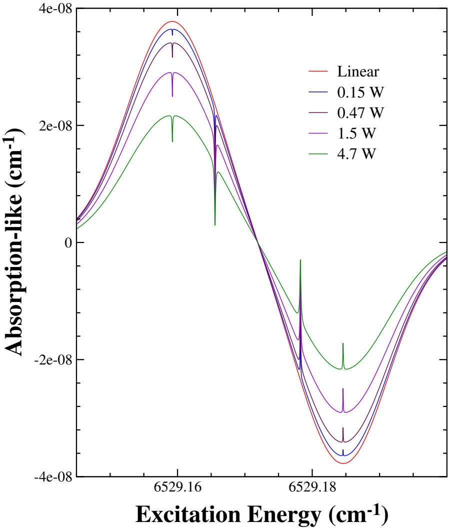
$1_0^1 3_0^1$ ($\Sigma_u^+ \leftarrow \Sigma_g^+$), $\mu = 10.7\,\mathrm{mD}$, $\omega_{rf} = 380\,\mathrm{MHz}$, $\xi = 0.4$ (1 mTorr, $w_0 = 0.45\,\mathrm{mm}$, $\Gamma_{rel.} = 1\,\mathrm{MHz}$)

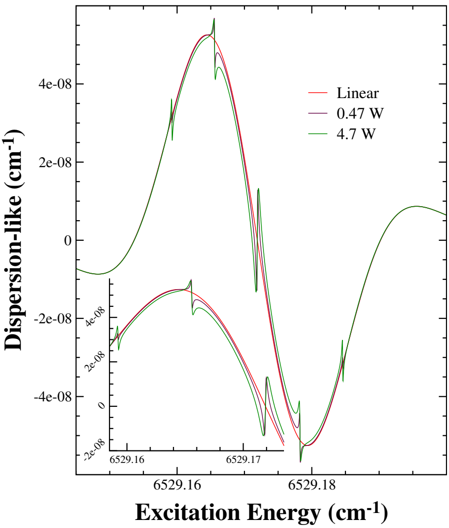




Simulation C_2H_2: $P_{11}(I=1)$, $\mathcal{N} = 3.2\times10^{11}\,\mathrm{cm}^{-3}$, σ Pol.

$1_0^1 3_0^1$ $(\Sigma_u^+ \leftarrow \Sigma_g^+)$, $\mu = 10.75\,\mathrm{mD}$, $\omega_{rf} = 380\,\mathrm{MHz}$, $\xi = 0.36$ ($w_0 = 0.54\,\mathrm{mm}$, $\Gamma_{rel.} = 0.5\,\mathrm{MHz}$)





Summary

- Analytical Solutions + Doppler Shift Integral (Numerical) + Sum over the Sub-Transitions (“saturation coefficients” < 100)
- The Formalism (involving 2 counter propagating beams) reproduces the Lamb-dips and the Cross-Sideband Resonances: Position, Intensity and Width
- Can be applied to any value of the Phase Modulation Index (ξ)
- Spatial Beam Profiles: Longitudinal and Transverse (through additional Numerical Integration)
- Can be applied to non-monochromatic Sources
- Benchmarked on 2 transitions of C_2H_2 (P_{10}, $I = 0$, and P_{11}, $I = 1$) in σ Polarization (very good agreement with O. Axner data)
- Weak sensitivity of the “Dispersion-like” signal with the Saturation (under the present experimental conditions)
- Optimum Sensitivity: Modulation Index, and Doppler vs. Sub-Doppler
- Approximation by mean “saturation coefficient” (excellent for “Dispersion” signal)
- Applicable to Saturated FMS (i.e., without cavity)

Perspectives

- Simultaneous determination of the Number Density and of the Transition Strength
- The Strong Saturation Regime?
- Extension to N-level System?
- Application to Multichromatic EMF (OFC)
- Metrology
- Line Shape Analysis
- Etc..

Thank for Your Attention

Ove Axner

Swedish French Institute

lpca Laboratoire de Physico-Chimie de l'Atmosphère

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