High Sensitivity and High Resolution Laser Spectroscopy of C_2H_2 Molecules with Optical Waveguides in the 1.5 μ m Wavelength Region

林泰生 (Tyson Lin)

逢甲大學光電學系/物理教學研究中心

Department of Photonics and Physics Teaching Center

Feng Chia University, Taichung 407

2011.3.14 於清華大學物理系





Outline

- 1. Introduction
- 2. Advantages to laser spectroscopy with optical waveguides
- 3. Hollow fiber absorption spectroscopy
- 4. Fiber evanescent wave spectroscopy with tapered fiber



主要研究方向:

→高解析雷射光譜與氣體偵測
 →原子分子光譜及其頻率精確量測
 →雷射穩頻及光頻率標準
 →雷射及其相關研究



最近從事研究及有興趣主題:

- 1. HD分子1.5µm波段泛音譜帶雷射光譜
- 2. CO₂及C₂H₂分子1.5µm波段雷射光譜
- 3. 半導體雷射穩頻
- 4. Fabry-Perot共振腔增强吸收法之研究
- 5. 中空光纖吸收室研究
- 6. 光纖消散波光譜法之研究
- 7. 测量HD、CO2等分子在1.5µm波段之精確譜線頻率
- 8. 自組外腔室可調頻二極體雷射之研究



Applications of Laser Spectroscopy in the 1.5 µm Region

- Fundamental Physics
- Optical Communications
- Environmental Gas Tracing and Detection

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- Astrophysics
- Frequency Standards and Metrology



Molecular Spectroscopy in the 1.5 μm Region

Absorption of molecules in the 1.5 µm region : Overtone or combination band transitions : HD, C₂H₂, CO, NH₃, HCN Hot Band Transitions: CO₂, H₂O

- \Rightarrow Small electric dipole moment
- \rightarrow Weak transition or even very weak transition
- \rightarrow Difficult to obtain high resolution spectroscopy



Example of Weak Absorption Spectrum in the 1.5 μ m region: HD & C₂H₂



Obtain Weak Absorption Spectrum in the 1.5 µm Region

Basics:

➢ Increase number density of molecules

 → i.e. increase pressure of sample
 ⇒ pressure broadening

 ➢ Increase intensity of light source

 → high intensity tunable power usually not available

 ➢ High sensitivity spectroscopic techniques

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Methods for Weak Absorption Spectroscopy in the 1.5 μm region

Traditional way :

For the example of HD:

- \rightarrow very long absorption cell
 - or multipass absorption cell \sim path up to 3 km

 \rightarrow high pressure : 10² Torr to above 1 atm

→grating spectrograph



Laser Spectroscopy of Weak Absorption in the 1.5 µm Region

Since the invention of laser, provides:

- ⇒Low NOISE tunable laser source, and high sensitivity PD
- ⇒High sensitivity spectroscopic techniques to enhance the SNR (訊噪比):
 - (1) Frequency Modulation
 - →high modulated frequency
 - (2) Difference Spectroscopy Technique
 - →reduce noise and background
 - (3) FM Sideband Techniques
 - \rightarrow combine FM and Difference spectroscopy
 - (4) Intracavity absorption
 - (5) Cavity enhanced absorption



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Frequency Modulation :

- ⇒對雷射頻率做某一頻率f的調制 因此
- →只有雷射造成的訊號才被取出
- → 只取出調制頻率的雜訊
- → 雜訊降低, 增強SNR
- → 取微分訊號, 訊號背景也被降低
- →f 越高, SNR 越大



If modulate the Laser Frequency $\omega_L(t) = \omega_0 + a \sin \Omega t$



Fig.6.2. Absorption spectroscopy with a frequency modulated single-mode laser



Then the Intensity of Laser Becomes:

$$\begin{split} \mathbf{I}_{\mathrm{T}}(\boldsymbol{\omega}_{L}) &= \mathbf{I}_{\mathrm{T}}(\boldsymbol{\omega}_{0}) + \Sigma(a^{\mathrm{n}}/\mathrm{n}!) \sin^{\mathrm{n}}\Omega \mathbf{t} \times [\mathrm{d}^{\mathrm{n}} \mathbf{I}_{\mathrm{T}}/\mathrm{d}\boldsymbol{\omega}^{\mathrm{n}}]_{\boldsymbol{\omega}_{0}} \\ &= \mathbf{I}_{\mathrm{T}}(\boldsymbol{\omega}_{0}) - a\mathrm{L}\mathbf{I}_{0}\{ [a/4 (\mathrm{d}^{2}\alpha/\mathrm{d}\boldsymbol{\omega}^{2})_{\boldsymbol{\omega}_{0}}] \\ &+ [(\mathrm{d}\alpha/\mathrm{d}\boldsymbol{\omega})_{\boldsymbol{\omega}_{0}} + \cdots] \sin\Omega \mathbf{t} \\ &+ [-a/4 (\mathrm{d}^{2}\alpha/\mathrm{d}\boldsymbol{\omega}^{2})_{\boldsymbol{\omega}_{0}} + \cdots] \cos 2\Omega \mathbf{t} \\ &+ [-a^{2}/24 (\mathrm{d}^{3}\alpha/\mathrm{d}\boldsymbol{\omega}^{3})_{\boldsymbol{\omega}_{0}} + \cdots] \sin 3\Omega \mathbf{t} \\ &+ \cdots \} \end{split}$$

, where $I_T(\omega_L) = I_0 \exp(-\alpha(\omega)L)$ and the weak absorption condition $\alpha(\omega)L <<1$ are used. 逢甲大學光電學系 Dept. of Photonics, FCU, *www.photonics.fcu.edu.tw*



Signals Demodulated with the Lock-in Amp: Derivative Spectroscopy



Fig.6.3. Lorentzian line profile $\alpha(\omega)$ of halfwidth γ (FWHM) (a) with first (c) and third (d) derivatives



Methods of Frequency Modulation :

調制通常對: (1) PZT → 約至數kHz (2) Thermal → 無法很快 (3) Current → 可至MHz或更快,但伴隨 RAM

(4) EOM → 可至MHz或更快,但伴隨 RAM

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Difference Spectroscopy





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FM Sideband (Heterodyne) Spectroscopy

Concept :



Fig. 1. Schematic diagram illustrating the principle of FM spectroscopy and the basic setup used in the experiments.

 $E_{i}(t) = E_{o} \exp(i\omega_{c}t)$ $E_{2}(t) = E_{o} \exp[i(\omega_{c}t + Msin\omega_{rf}t)] = E_{o} \sum_{n=0}^{n=0} J_{n}(M)e^{i(\omega_{c}+n\omega_{rf}t)}$ $= E_{o} \sum_{n=0}^{M} e^{i(\omega_{c}+\omega_{rf}t)t} + e^{i\omega_{c}t} - M e^{i(\omega_{c}-\omega_{rf}t)t} \sum_{r=0}^{n=0} E_{s}(t) = E_{o} \sum_{n=0}^{n=0} T_{n}e^{i(\omega_{c}+\omega_{rf}t)t} - T_{-1} \sum_{n=0}^{M} e^{i(\omega_{c}-\omega_{rf}t)t} \sum_{r=0}^{n=0} T_{s}(t) = I_{o}e^{-2\delta}(1 - \Delta\delta M \cos\omega_{rf}t + \Delta^{2}\phi M \sin\omega_{rf}t)$

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FM Sideband (Heterodyne) Spectroscopy: Signal





High Resolution: Removing Broadening

對氣體分子的譜線寬:

在溫度T下,由於熱運動 ⇒ Doppler Broadening

在氣壓P下,由於分子碰撞 ⇒ Pressure Broadening

→掩蓋了分子躍遷真正的譜線寬,進而無法精確決定中 心譜線值

→利用高解析光譜法得其natural linewidth:

在低氣壓做SubDoppler光譜,

如: Saturation Absorption Spectroscopy(飽和吸收光

譜),

Doppler-free Two Photon Spectroscopy,



Concept of Saturated Absorption



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Saturated Spectroscopy Using Cavity

對於弱吸收分子 → high saturation intensity 因此須

- → 增強入射pumping beam光強度
- →增加有效吸收長度
- → 增加probe 與pumping beam之重合度
- ⇒Cavity:可同時提供probe beam與足夠強pumping beam,及完美的重合度
- (1) Intracavity Spectroscopy

→雷射輸出功率受到影響

- (2) Fabry-Perot Cavity Enhanced Absorption
 - →External Cavity

→Stable Cavity





Intracavity Absorption Spectroscopy



Fig.6.4. Intracavity absorption detected either by monitoring the laser output $P(\omega_L)$ with detector 2 or the laser-induced fluorescence $I_{Fl}(\omega_L)$ with detector 1



F-P Cavity Enhanced Absorption :

- 高反射低損耗之反射鏡,可做high Finesse的共振 腔
 - → Example : 7 ≥10000, FSR = 1GHz (L=15 cm)
 - \Rightarrow Cavity linewidth ~ 100 kHz , i.e. cavity photon storage time ~ 10 μs
 - ⇒相當於等效長度為 3×10⁸×10 µs = 3 km!
 - \Rightarrow At resonance : $v = v_0$
 - $P_t \propto 1 2\delta/(1 r_1 r_2)$ (δ:absorption per pass

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即相當於:吸收由δ增強至 $2\delta/(1 - r_1r_2)$

Pound-Drever-Hall Scheme



 $\tau > \tau_{cavity}$ Freq. Detector

1

phase-modulation sidebands offset by $\pm f_m$ from carrier frequency f_c Sidebands and some carrier, reflected from reference cavity, are steered by quarter-wave plate and polarizer to detector. Phase-sensitive detection against modulation source t gives bipolar error signal proportional to frequency offset $f_0 - f_1$ in the adiabatic regime. In transient regime, the system functions as an optical phase detector (see text)





de Labachelerie, Nakagawa, Awaji, and Ohtsu's

Setup



Fig. 1. Experimental setup showing the ECSL structure, the FP cavity, and the arrangement for long-term stabilization. ISO, 60-dB isolator; PBS, polarizing beam splitter; $\lambda/4$, quarter-wave plate; DBM, double-balanced mixer.



de Labachelerie, Nakagawa, Awaji, and Ohtsu's Result



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Fig. 3. (a) Derivative of the line that was used for frequency stability measurements. (b) Longest wavelength sub-Doppler line recorded with $^{13}\mathrm{C}_{2}\mathrm{H}_{2}.$

NICE-OHMS:Noise-immune cavity enhanced

optical heterodyne molecular spectroscopy

Jun Ye, L.-S. Ma, J.L. Hall (JILA & NIST)

- ⇒利用Pound-Drever-Hall Scheme將雷射頻率鎖於F-P Cavity
- :降低雷射之linewidth及frequency noise
 - ⇒ Phase Modulator 所產生的sideband與F-P cavity 的mode spacing 相同
 - ⇒利用FM sideband光譜技巧看分子之飽和吸收光譜

好處:

- 1. Sideband 與center frequency間之Heterodyne可消除來自laser noise
- 2. 可做高頻調制,提高SNR.
- \rightarrow Sensitivity ~ 10⁻¹³;
- →利用此法已觀察到C₂H₂在1.064 µm(dipole moment ~3×10⁻⁶ D)之飽和吸收!



NICE-OHMS: Experimental Scheme

Ye et al.



Fig. 2. General experimental schematic for the NICE-OHMS spectrometer.

- A:高频调制,看分子之FM sideband 光馨用
- δ: 較低頻調制,便需射利用Pound - Drever 法额频 F-P cavity.



Types of Optical Waveguides for Laser Spectroscopy

- Hollow metal tube or silicate tube with dielectric and silver coatings :1996 Kozodoy *et al.*, observation of CO_2 spectrum; 2002 Fetzer *et al.*, observation of CO_2 and NH₃ spectrum with coiled hollow waveguide in 1.5 µm.
- Hollow photonic bandgap fiber:, Henningsen *et al.* (2005), R. Thapa *et al.*(2006), saturated absorption of C₂H₂
- Tapered optical fiber: Takiguchi *et al.*, saturated absorption of C_2H_2

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Advantages of Optical Waveguides for Laser Spectroscopy

- Providing high laser intensity
- Highly overlapping for saturated absorption
- Interaction with less amount of gases sample
- Precisely detection of wanted area and gases
- Avoiding absorption of unwanted gases on delivery of laser beam
- Available for remote sensing





Laser Spectroscopy of C_2H_2 with the Hollow Fiber Absorption Cell in the 1.5 μ m

- The hollow fiber has been used on the conventional absorption spectroscopy, but not saturated absorption, of gas and liquid for several years
- Example of demonstrating the saturated absorption of R(9) line , $v_1 + v_3$ band of C₂H₂ molecule at 1520 nm



Hollow Waveguide

• Y. Matsuura *et al.* (Prof. Miyagi group, *Tohoku University, Japan*),

"Hollow-fiber delivery of high-power pulsed Nd:YAG laser light"

- Reduced the propagation losses of the fibers in the near-infrared region by producing a silver film very smoothly on the hollow silicate tube.
- Use of an ultrasonic wave for mixing of the silver and the reducer solutions in the silver-plating process.

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*Opt. Lett. 23, 1858 (1998)

Hollow Waveguide:



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Hollow Fiber from Tohuko Univ

FWHM=10.7° 1999.3.8 25 • The performance of two hollow 20Attenuation (dB) fibers given by 15 Prof. Miyagi (宮 10 城光信教授): 1-m-long polymer-coated 1.5 0.5 1.0Wavelength (µm) fibers for the 700-mm bore size.

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Hollow Fiber as Absorption Cell



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Coupling Laser Beam into Hollow Fiber

- 對有鍍銀的中空砂光纖,其傳播最低模態為TE₀₁模態, 為圓偏振。(Marcatili & Schmeltzer)
- 但在自由空間傳播的TEM₀₀模無法耦合成TE₀₁,所以只能以TEM₀₀的雷射耦合入光纖,以HE₁₁混合模傳播。因此由Matsuura et al.的研究,要選擇耦合光束束腰小於 0.64a(a為中空光纖的內半徑),且耦合鏡焦距須符合:



其中f=耦合鏡的焦距, D=高斯光束強度在 $1/e^2$ 的直徑, λ =中空光纖內傳播的光波長。

E. A. J. Marcatili and R. A. Schmeltzer ,"*Hollow metallic and dielectric waveguides for long distance optical transmission and lasers*", J. Bell Syst. Tech., **43**, pp.1783-1864 (1964)

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Y. Matsuura, T. Abel, and James. A. Harrington, "Optical Properties of Small-Bore Kollow Glass Waveguides", Appl. Opt., vol.34, pp.6842-6847, (1995)

Laser Spectroscopy of C_2H_2 with the Hollow Fiber Absorption Cell in the 1.5 μ m



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Saturated Absorption of C₂H₂with the Hollow Fiber Absorption Cell



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Laser Spectroscopy of C₂H₂with the Hollow Fiber Absorption Cell: (a) Only Pump beam modulated



DFB : Distributed Feedback laser diode ; ECTDL: External Cavity Tunable Diode Laser; EDFA : Erbium Doped Fiber Amplifier ; PC: Polarizer Controller ; PBS : Polarization Beam Splitter ; $\lambda/2$: Halfwave plate ; P.D. : Photo Detector



Spectroscopy with Modulation Transfer Technique

1980, J.J Snyder *et. al.*: modulation transfer between two beams interacting with the same group of atoms or molecules



J. J. Snyder, R. K. Raj, D. Bloch, and M. Ducloy, "*High-sensitivity nonlinear spectroscopy using a frequency-offset pump*", Opt. Lett., 5, pp.163-165(1980)



Laser Spectroscopy of C₂H₂with the Hollow Fiber Absorption Cell in the 1.5 µm

The linewidth of spectrum is 8 MHz at 43 mTorr.





(b) Both Pump Beam and Probe Beam Modulated: Modulation Transfer Spectroscopy Technique



DFB : Distributed Feedback laser diode ; ECTDL: External Cavity Tunable Diode Laser; EDFA : Erbium Doped Fiber Amplifier ; PC: Polarizer Controller; PBS : Polarization Beam Splitter ; $\lambda/2$: Halfwave plate ; P.D. : Photo Detector \circ



(b) Both Pump Beam and Probe Beam Modulated: Modulation Transfer Spectroscopy Technique

The linewidth of spectrum is 6 MHz at 45 mTorr~47 mTorr.



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Evanescent Wave of Optical Fiber

 Evanescent wave from total internal reflection as incident angle θ₁ > critical angle θ_c, with



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Evanescent Wave of Optical Fiber

• For $n_1=1.5$, $n_2=1$, $\lambda=1.5\mu m$, $\theta_1=45^\circ$ ($\theta_c=41.8^\circ$), then $\delta=0.65\mu m$ and the evanescent wave propagates along the interface with its *E* field decaying rapidly by a factor 1/*e* within a very short range 0.65 μm (smaller than its wavelength).

The E field decays by a factor 1/e at a distance $\delta=1/\alpha$ from the interface,

$$\delta = \frac{\lambda_0}{2\pi n_1 \sqrt{\sin^2 \theta_1 - \sin^2 \theta_c}}$$



Evanescent Wave of Tapered Optical Fiber



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PHOTO

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Evanescent Wave of Tapered Optical Fiber

1992 J. Opt. Soc. Am. A/Vol. 16, No. 8/August 1999

J. Bures and R. Ghosh

Power density of the evanescent field in the vicinity of a tapered fiber

Jacques Bures and René Ghosh

Laboratoire des Fibres Optiques, Département de Génie Physique et de Génie des Matériaux, Ecole Polytechnique de Montréal, C.P. 6079, Succursale Centre-Ville, Montréal, Québec H3C 3A7, Canada

Received October 2, 1998; accepted March 22, 1999; revised manuscript received March 29, 1999

The power density in the vicinity of a tapered fiber is calculated, with the vectorial model of step-index circular waveguides. For the fundamental HE_{11} mode carrying a power of 1 Watt, we show that it is possible to obtain theoretical densities in the range of 10^8 W/cm² at the fiber surface. The promising use of such intense evanescent fields as "atomic mirrors" is considered, and the feasibility of these guides is investigated. © 1999 Optical Society of America [S0740-3232(99)00308-7]

OCIS codes: 060.2310, 060.2340, 060.2400.



Evanescent Wave of Tapered Optical Fiber

- J. Bures and R. Ghosh calculate the power density in the vicinity of a tapered fiber in the fundamental mode HE_{11} as a function of waveguide radius and wavelength.
- The model used is that of the exact vectorial theory developed by Snyder and Love* for circular step-index waveguides.
- *A. W. Snyder and J. D. Love, *Optical Waveguide Theory* (Chapman & Hall, London, 1983), Chap. 12.



Power Density of Evanescent Wave of Tapered Optical Fiber



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Power Density of Evanescent Wave of Tapered Optical Fiber



Fig. 4. Fraction of the power carried by the HE_{11} mode outside the fiber as a function of radius r, for a few common wavelengths. Also indicated are the loci of maximum density at the core–cladding interface and the limit of the single-mode regime.



Power Density of Evanescent Wave of Tapered Optical Fiber



Fig. 8. Relative transmitted power at λ =633 nm by HE₁₁ at the end of the stretching process. Inside the figure is an enlargement of the beats between HE₁₁ and the TE₀₁, TM₀₁, and HE₂₁ group. The regime becomes single mode for ρ <0.23mm, and the leaky modes are responsible for the observed loss for an elongation > 7.3 mm. (J. Bures and R. Ghosh)

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Measured Loss for Single-mode Optical Wave Guiding



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Eric Mazur : the Balkanski Professor of Physics and Applied Physics at Harvard University

L. Tong, R. R. Gattass, J. B. Ashcom, S. He, J. Lou, M. Shen, I. Maxwell, and E. Maxwell, "Subwavelength diameter silica wires for low-loss optical wave guiding," Nature 42, 819 (2003).

Silica Microfibers

Fractional power inside the core



L. Tong et al., Opt. Express 12,1025 (2004)

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- 童利民(Limin Tong)教授:浙江大學光电信息工程学系
- L. Tong, J. Lou, and E. Mazur, "Single-mode guiding properties of subwavelength-dia silica and silicon wire waveguides," Opt. Express 12, 1025-1035 (2004),

Nano- Fiber : Univ of Tokyo

Fabrication of Nano-fiber Resonators for Cavity-QED Experiments with Neutral Atoms

Sept. 2008 CREST Quantum Information Okinawa Summer School

Institute of Physics, University of Tokyo <u>Masato Takiguchi</u>, Yutaka Yoshikawa, Yoshio Torii, Takahiro Kuga







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Nano- Fiber : Univ of Tokyo group



Road Map



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Nano- Fiber for Saturated absorption



Nano- Fiber for Saturated Absorption



The mode-field diameter increases as d decreases.





Nano- Fiber for Saturated Absorption

Saturated absorption spectra of the P(9) transition of ¹²C₂H₂



Linewidth vs fiber diameter



Nano- Fiber for Saturated Absorption

Linewidth vs fiber diameter



Fiber diameter [nm]	Measured linewidth (FWHM) [MHz]	Transit-time broadening [MHz]
400	51 ±2	17 ±2
600	73 ±3	38 ±3
780	132 ± 7	97 ± 7

(Pressure broadening: 35 MHz)

Masato Takiguchi, Yutaka Yoshikawa, and Takahiro Kuga, "SATURATED ABSORPTION SPECTROSCOPY OF ACETYLENE MOLECULESWITH AN OPTICAL NANO-FIBER", ICOLS 2009, Hokkaido, Japan



FCU's Tapered Fiber



*2011 物理年會壁報

Yung-Hsiang Lai (賴永祥), Tyson Lin (林泰生), Che-Chung Chou(周 哲仲), Cheng-Wen Wu (吳正文), Po-Neng Chang (張博能) **Department of Photonics, Feng Chia University**



Fiber-optic Evanescent Wave Spectroscopy Experimental Setup



DFB Diode laser (NEL. NKL1556STB) Wavelength meter(EXFO, WA-1550) FG: function Generator (1) modulation (2) scan: Detecter(EOT, ET-3010); TC: temperature Controller; CD: current Driver; LIA: Lock-in amplifier(SR850 DSP); Optical Fiber (Corning SMF-28e):core diameter 8.2 µm, cladding diameter $125 \pm 0.7 \mu m$; fiber taper: 5.2-µm-diameter, 0.4-cm-long





Lock-in Amp demodulation signal of optical power (Acetylene pressure: 20 Torr)





Lock-in Amp demodulation signal of optical power (Acetylene pressure: 60 Torr)



With Difference Detection



New Focus Balanced Photoreceiver Model 1617-AC



Results



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Future Works:

- **Development of frequency standards in the** 1.5 µm region using FEWS.
- Laser spectroscopy of Molecules : Fabry-Perot cavity enhanced absorption Hollow Fiber absorption Fiber evanescent wave
- Precise frequency measurement of molecular transition frequencies with frequency comb: HD, CO₂, CO...

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• Gas sensing of CO, CO₂, C₂H₂

Thank you for your attention!

