united nations educational, scientific and cultural organization

international atomic energy agency

the
abdus salam
international centre for theoretical physics

SMR 1302 - 16

WINTER SCHOOL ON LASER SPECTROSCOPY AND APPLICATIONS

19 February - 2 March 2001

Elementary Introduction to Subdoppler Laser Spectroscopy

Part I and II

M. INGUSCIO
L.E.N.S. - Lab. Europeo di Spettroscopie Non Lineari
Largo Enrico Fermi, 2 - Firenze, Italy

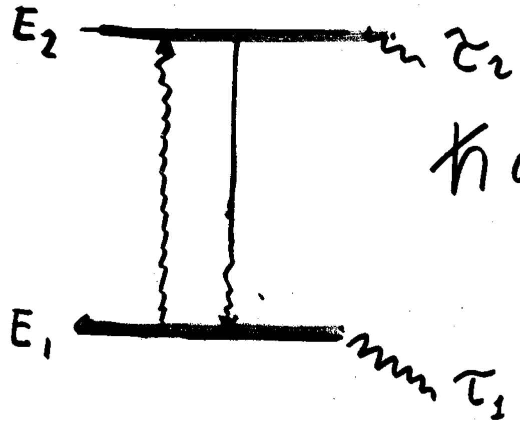
These are preliminary lecture notes, intended only for distribution to participants.

Elementary introduction to Subdoppler laser Spectroscopy

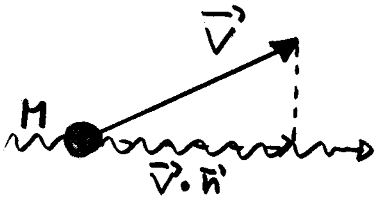
M. Inguscio # 1, 2

ALLARGAMENTO DOPPLER DI RIGHE SPETTRALI (1° ordine)

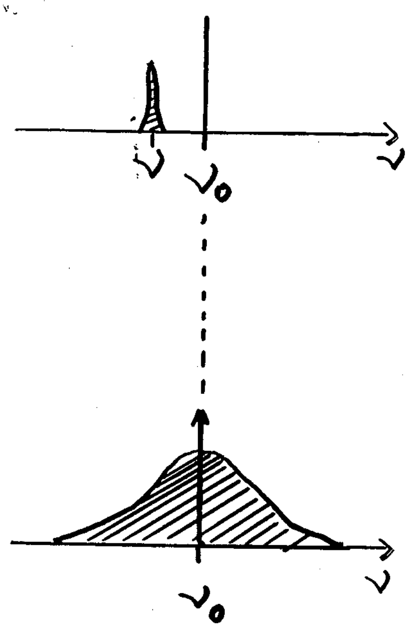
DOPPLER BROAD. (1st ORDER)



$$\hbar \omega_0 = E_2 - E_1$$



$$\nu = \nu_0 \left(1 + \frac{\vec{V}\cdot\vec{n}}{c}\right)$$



$$I = I_0 \, \mathrm{EXP}\left[-\frac{Mc^2}{2KT}\left(\frac{\nu-\nu_0}{\nu_0}\right)^2\right]$$

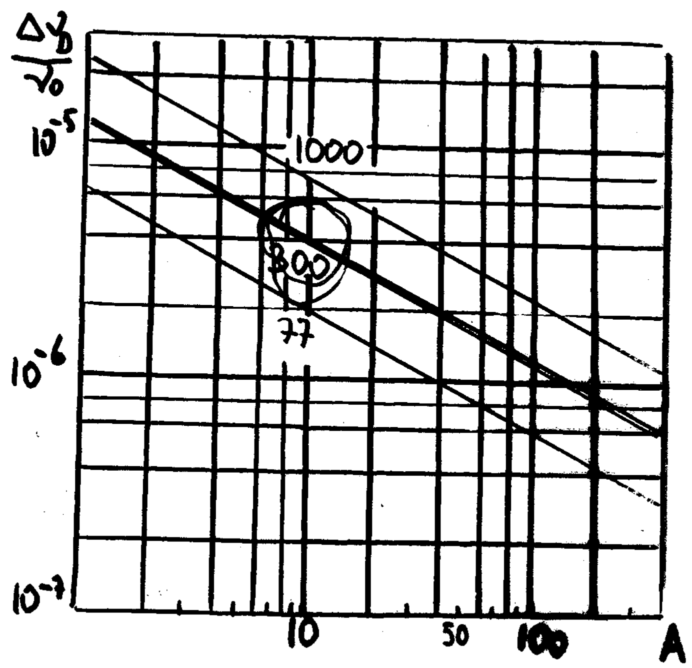
$$\boxed{\Delta\nu_D = 7.163\times10^{-7}\,\nu_0\sqrt{\frac{T}{A}}}$$

$10^8 - 10^{10}$ Hz

$\Delta\nu_D \gtrsim 10^2 \, \Delta\nu_{NAT}$ in the VIS.

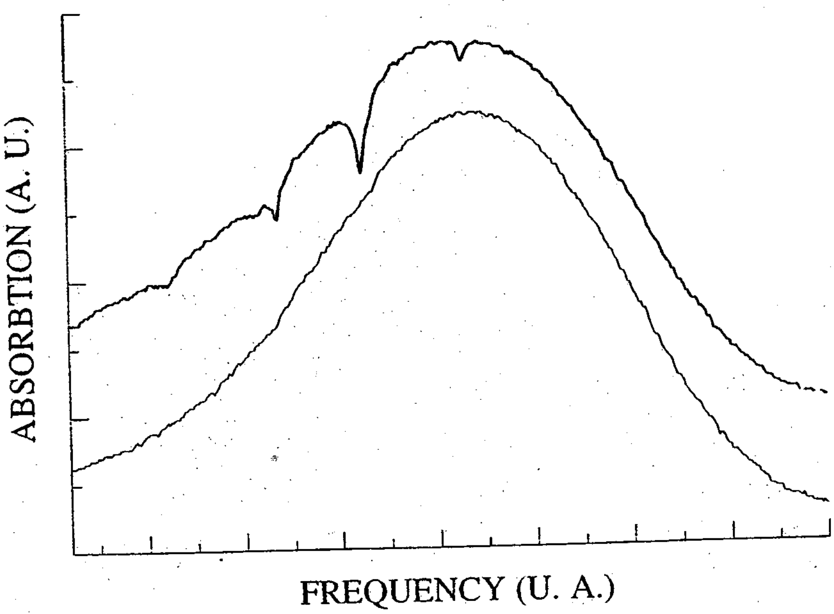
NOTE! $\Delta\nu_D$ independent from ν

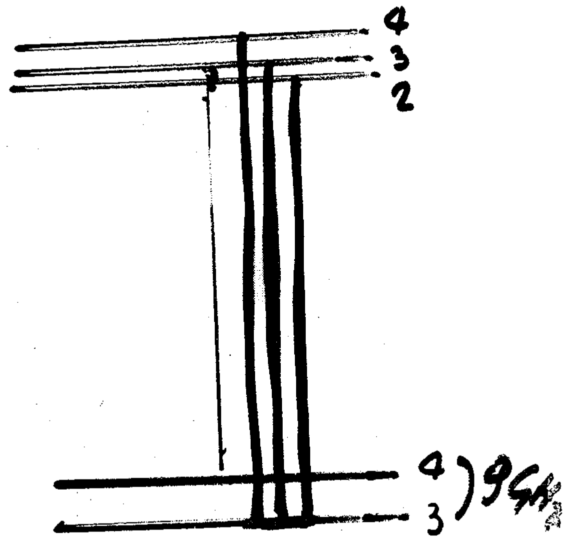
At room temp $\frac{\Delta\nu}{\nu} \sim 2\cdot10^{-6}$



SATURATION SPECTROSCOPY

Cs

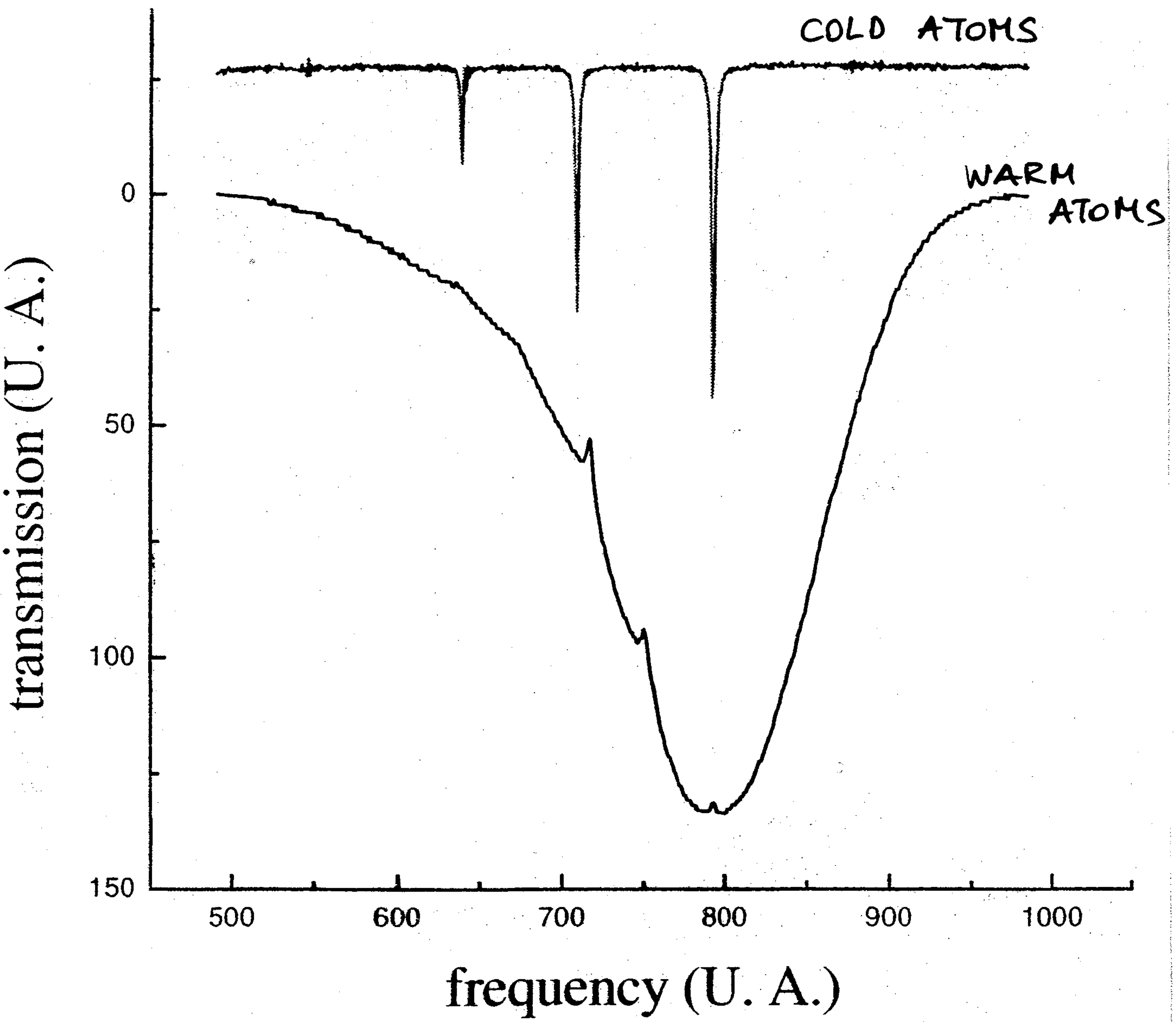




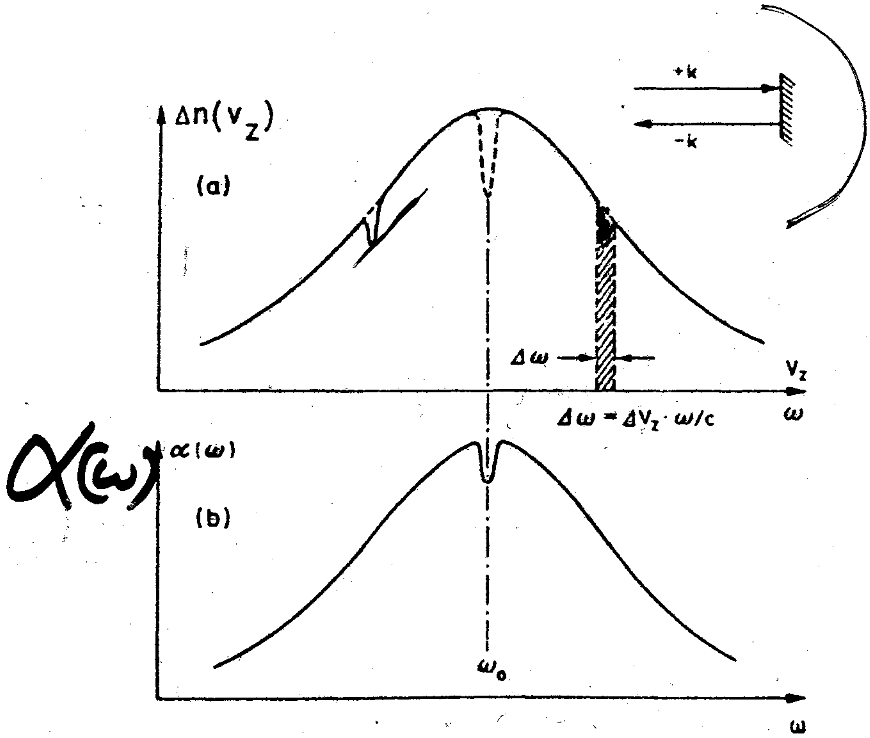
Different transitions masked by Doppler width.

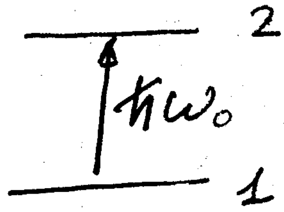
. . . as if atoms, at least in one direction, were not moving

NOW THEY ARE AT "REST,,



SUBDOPPLER SPECTROSCOPY





EFFETTO DOPPLER:

ATOMO CON VELOCITA' $\vec{v}$ SI MUOVE IN UN CAMPO LASER DI FREQUENZA ω_L E VETTORE D'ONDA $\vec{K}$

NEL SISTEMA DI RIFERIMENTO DELL'ATOMO ω_L E' VISTA COME $\sim \omega' = \omega_L - \vec{K} \cdot \vec{v}$

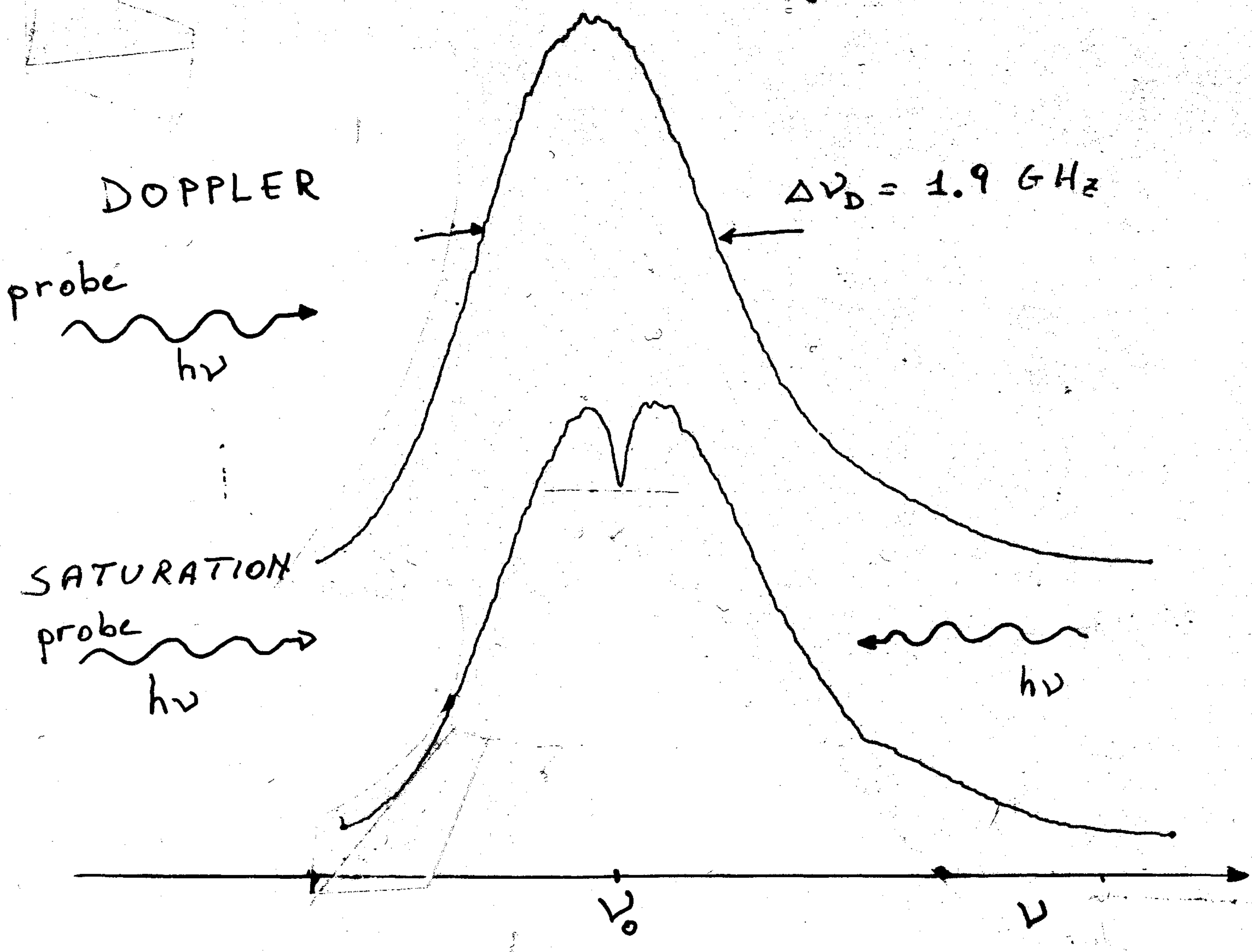
CONDIZIONE DI RISONANZA:

$$\omega_L = \vec{K} \cdot \vec{v} + \omega_0$$

OPTOGALVANIC SATURATION SPECTROSCOPY

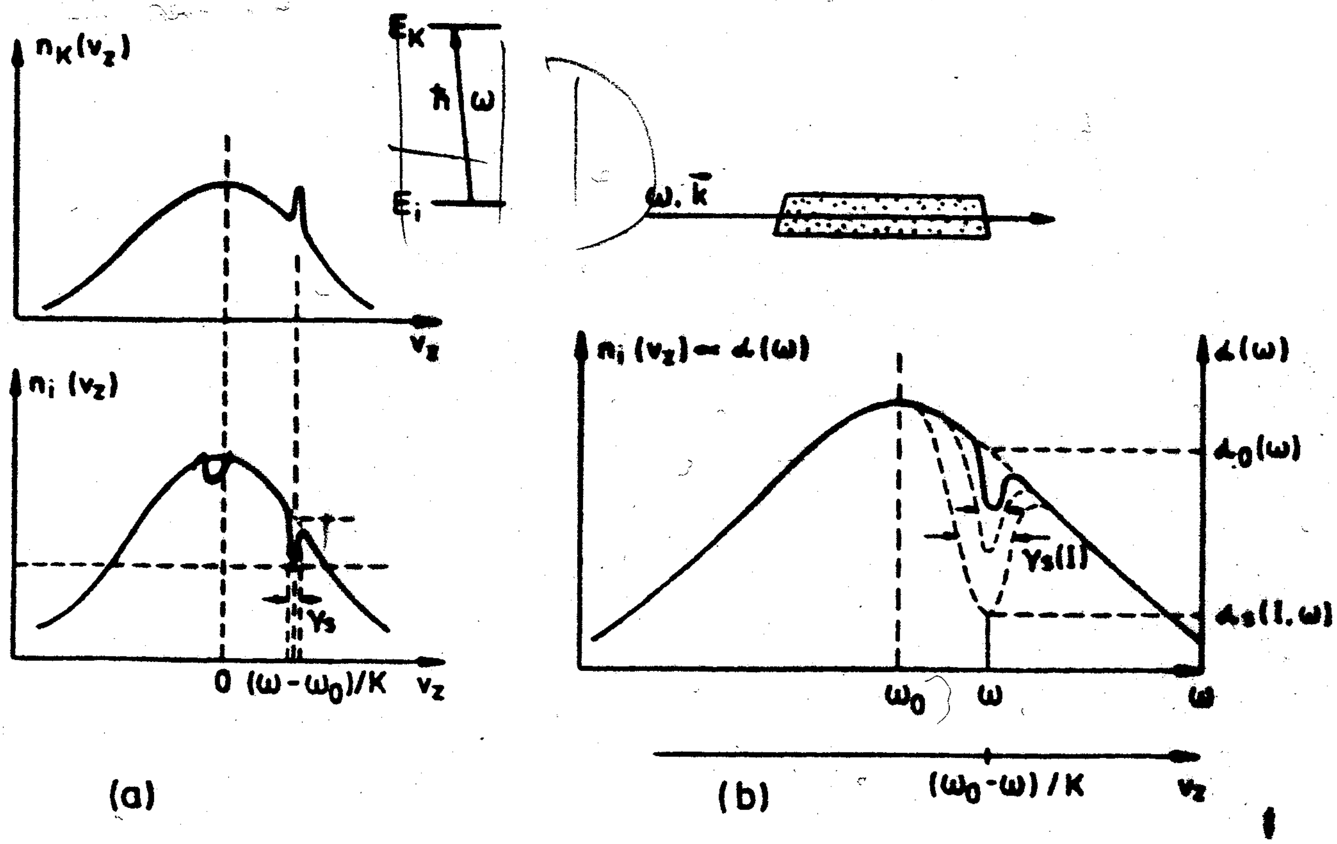
an example : Neon transition

FROM METASTABLE $1s_4 \rightarrow 2p_3$
$\lambda = 6074$ Å



HOW REMOVE THE DOPPLER BACKGROUND ?

SATURATION SPECTROSCOPY

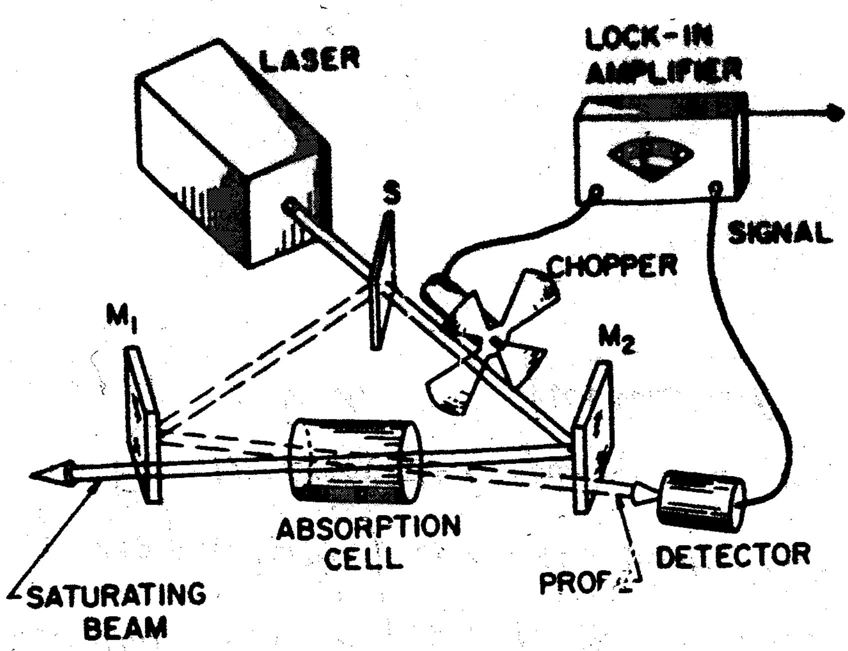


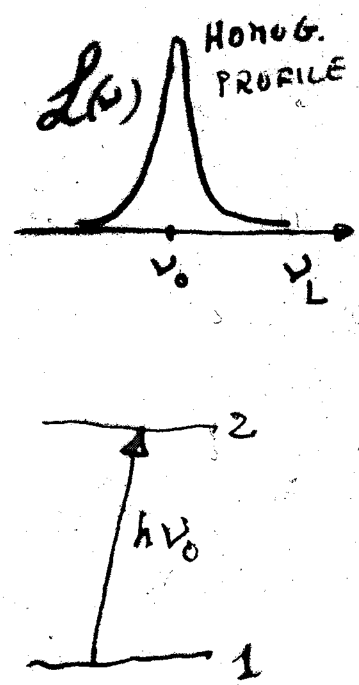
(a) (b)

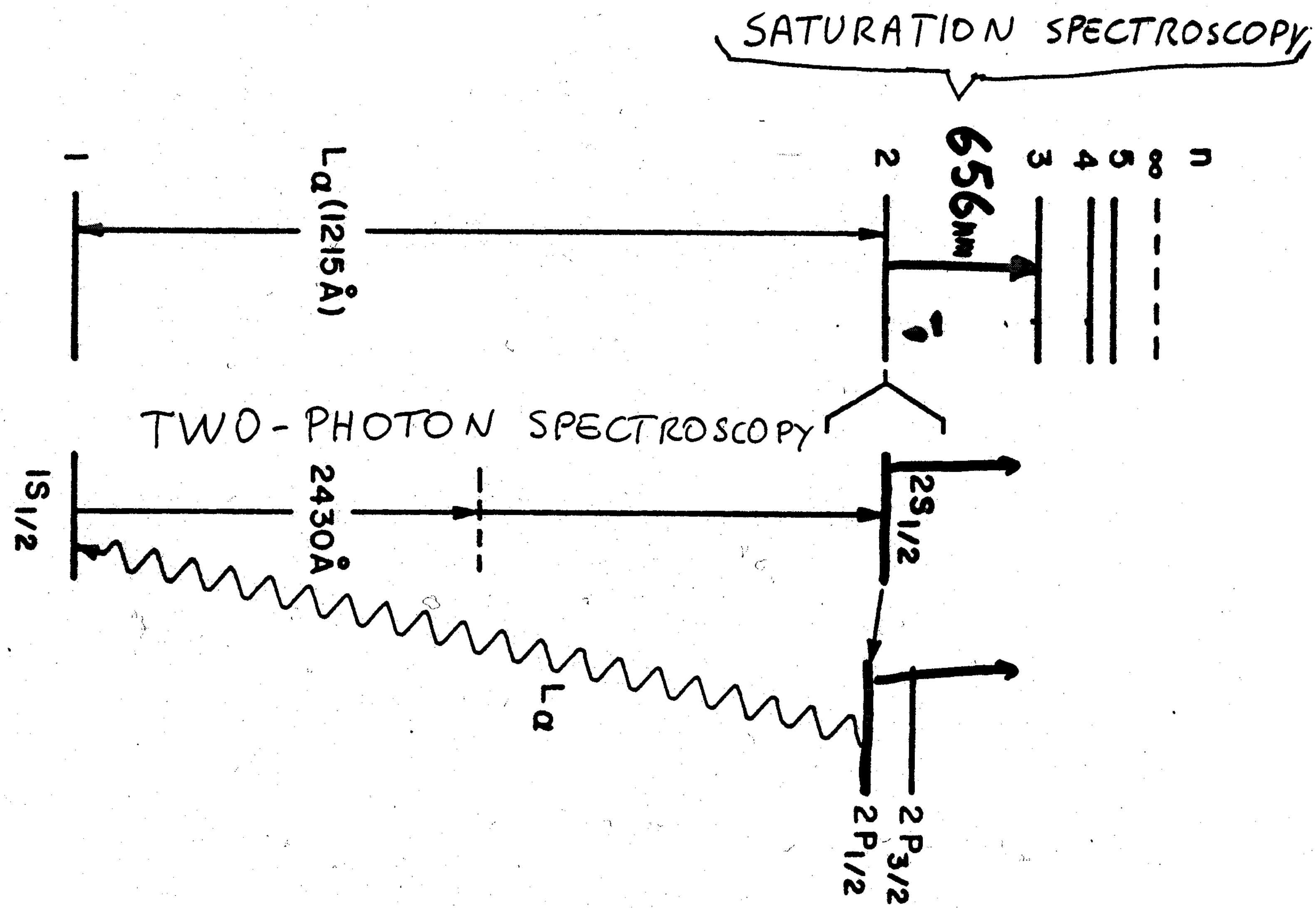
Hänsch

SCHEME

SATURATION SPECTROMETER






SATURATION SPECTROSCOPY
TWO-PHOTON SPECTROSCOPY
1
2
3
4
5
∞
n
Lα (1215Å)
656nm
1S1/2
2430Å
2S1/2
Lα
2P1/2
2P3/2

2.4.

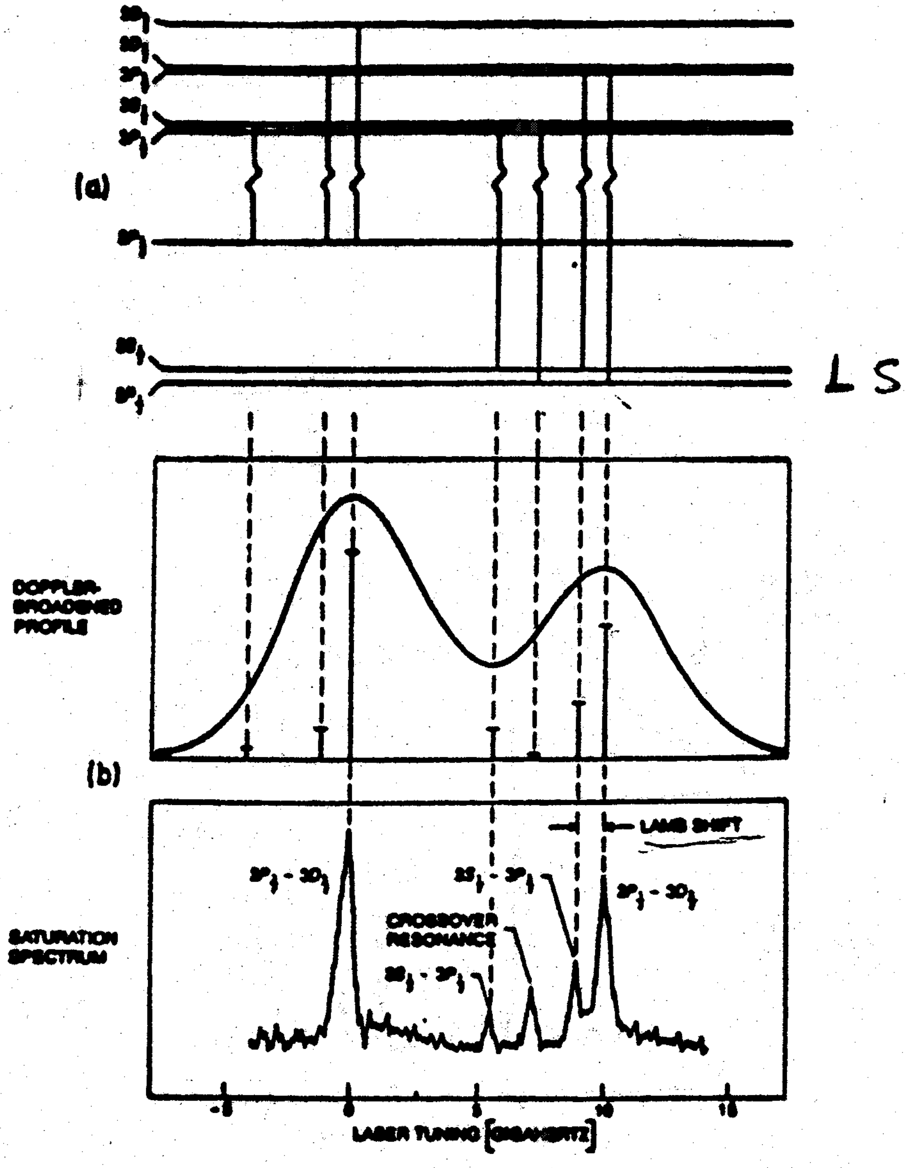
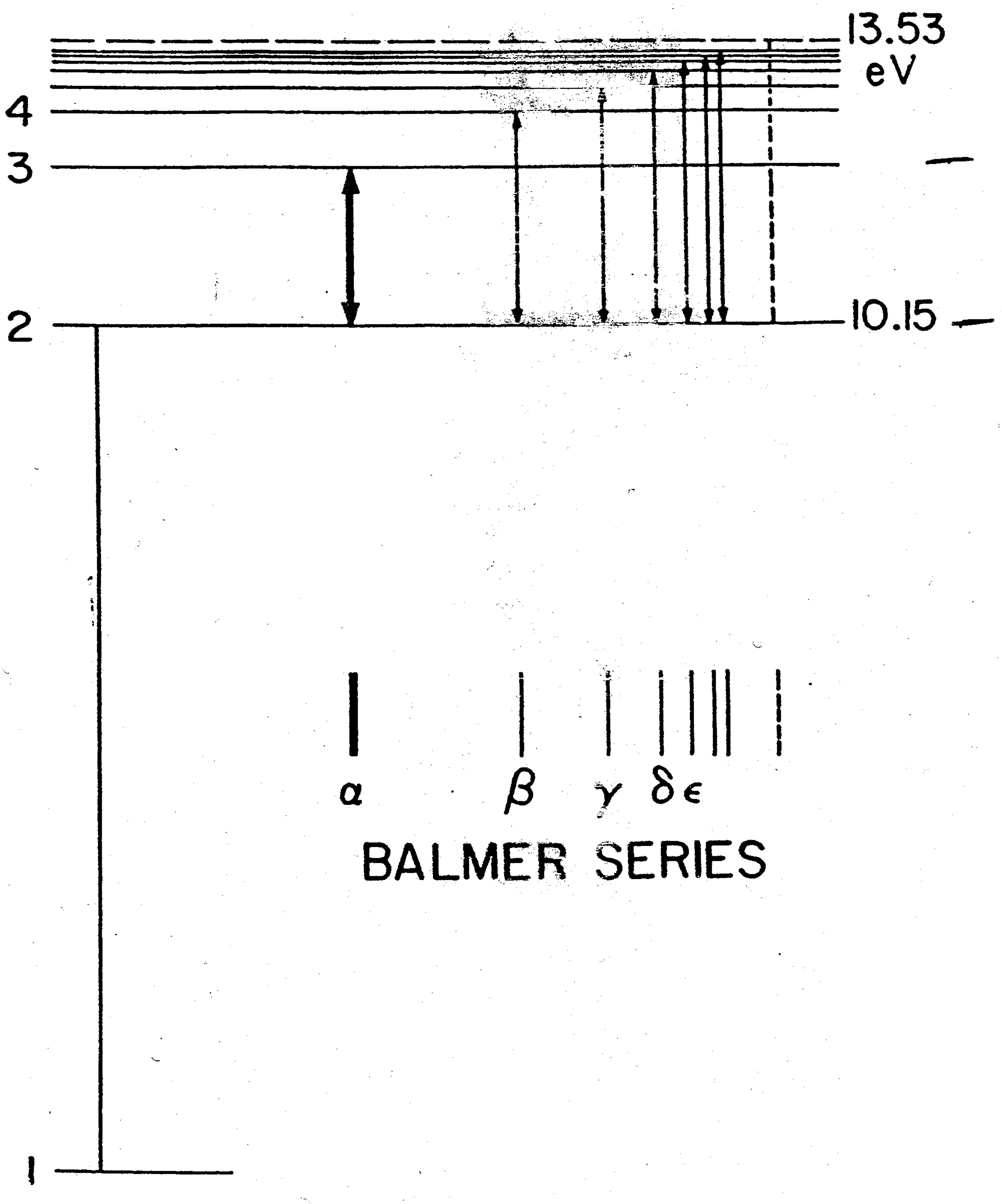


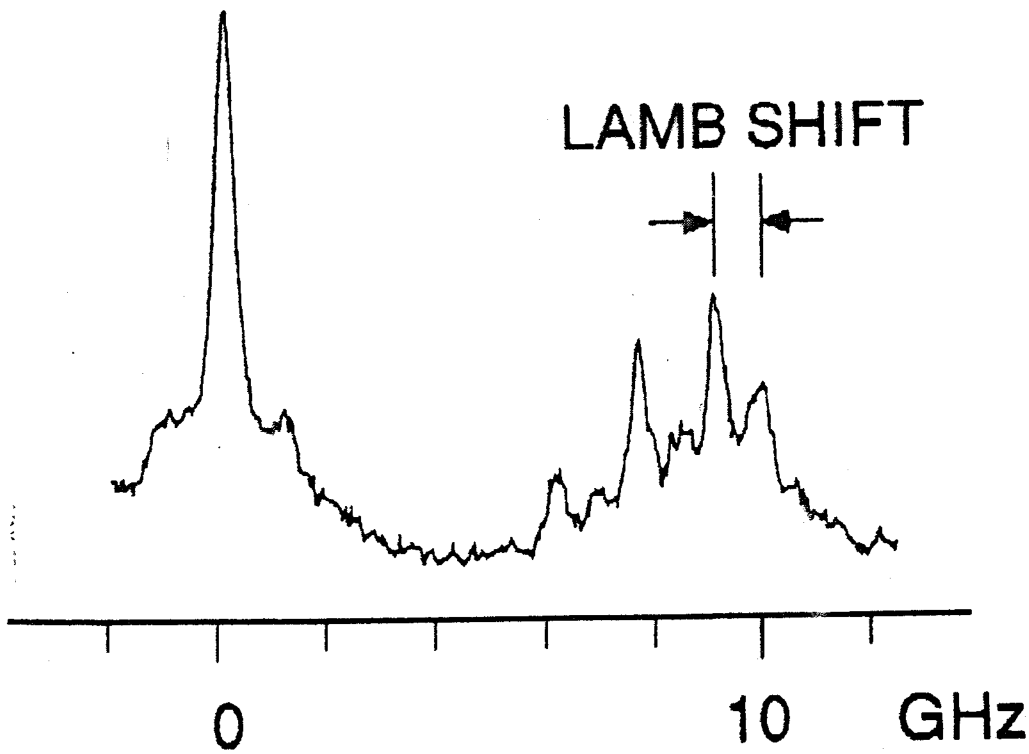
Fig.10.21a,b. Measurement of the Rydberg constant by saturation spectroscopy of the Hydrogen Balmer α transition. (a) Level scheme, (b) Doppler profiles and saturation spectrum of the Balmer α line in a hydrogen discharge [10.31a]

SUBDOPPLER RESOLUTION OF HYDROGEN BALMER α, WITH OBSERVATION OF FINE STRUCTURE AND LAMB SHIFT.

HYDROGEN TERMS

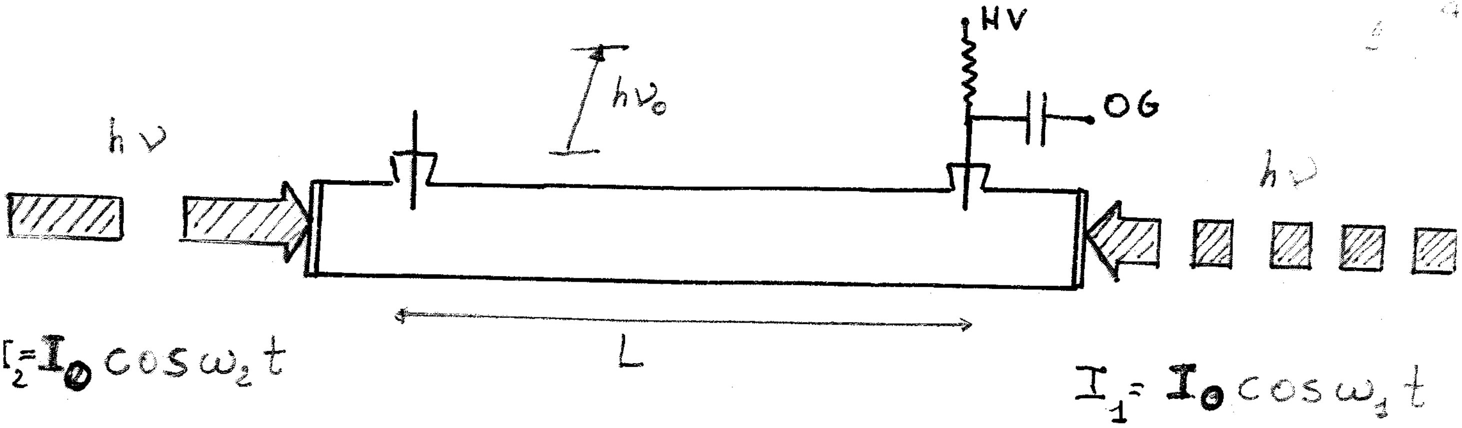


EVEN WITH A SMALL DIODE LASER



SOREM & SCHAWLOW Opt. Comm. 5, 148 (1972)

INTERMODULATED OPTOGALVANIC SPECTR.



THE OG SIGNAL INDUCED BY TWO COUNTERPROP. BEAMS, MODULATED AT FREQUENCIES

$$\omega_1 = 2\pi f_1 \quad ; \quad \omega_2 = 2\pi f_2$$

$$I = I_1 + I_2$$

is given by

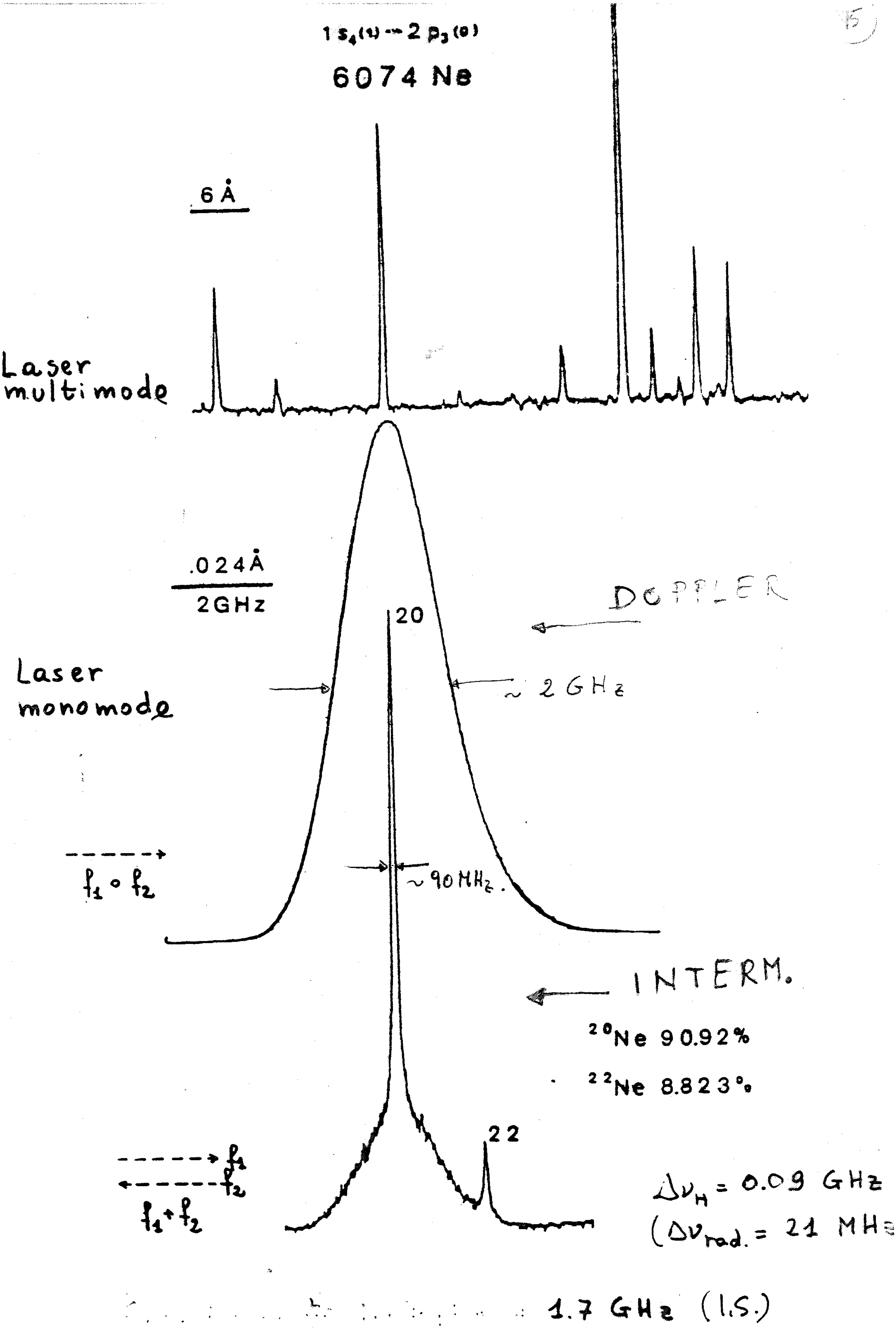
$$S_{OG} = K I \left[1 - e^{-\alpha(\omega) L} \right] \qquad k = \text{const.}$$

where

$$\alpha(\omega) = \alpha_0(\omega) \left[1 - \frac{S_0}{2} \frac{(\gamma_s/2)^2}{(\omega - \omega_0)^2 + (\gamma_s/2)^2} \right]$$

If $\alpha L << 1$ ("optically" thin sample)

$$S_{OG} = K L \alpha_0(\omega) \left[I - \frac{I^2}{2 I_{SAT}} \frac{(\gamma_s/2)^2}{(\omega - \omega_0)^2 + (\gamma_s/2)^2} \right]$$


1 s4(1) → 2 p3(0)
6074 Ne
6 Å
Laser multimode
.024 Å
2GHz
DOPPLER
20
Laser monomode
~ 2 GHz
f1 ∘ f2
~90 MHz
INTERM.
20Ne 90.92%
22Ne 8.823%
22
f1
f2
f1+f2
Δν_H = 0.09 GHz
(Δν_rad. = 21 MHz
1.7 GHz (I.S.)

Being :

$$I = I_1 + I_2 = I_0 \sin \omega_1 t + I_0 \sin \omega_2 t$$

$$= I_0 \left(\sin \omega_1 t + \sin \omega_2 t \right)$$

$$S_{OG} = K L \, \alpha_0(\omega) \Big|_{\omega_1} + \quad \Leftarrow \text{DOPPLER}$$

$$K L \, \alpha_0(\omega) \Big|_{\omega_2} + \quad \Leftarrow \text{DOPPLER}$$

$$K L \, \alpha_0(\omega) \Big|_{(\omega_1+\omega_2) \text{ or } (\omega_1-\omega_2)} \times \frac{(\gamma_s/2)^2}{(\omega-\omega_0)^2 + (\gamma_s/2)^2} \quad \Leftarrow \text{SUB-DOPPLER}$$

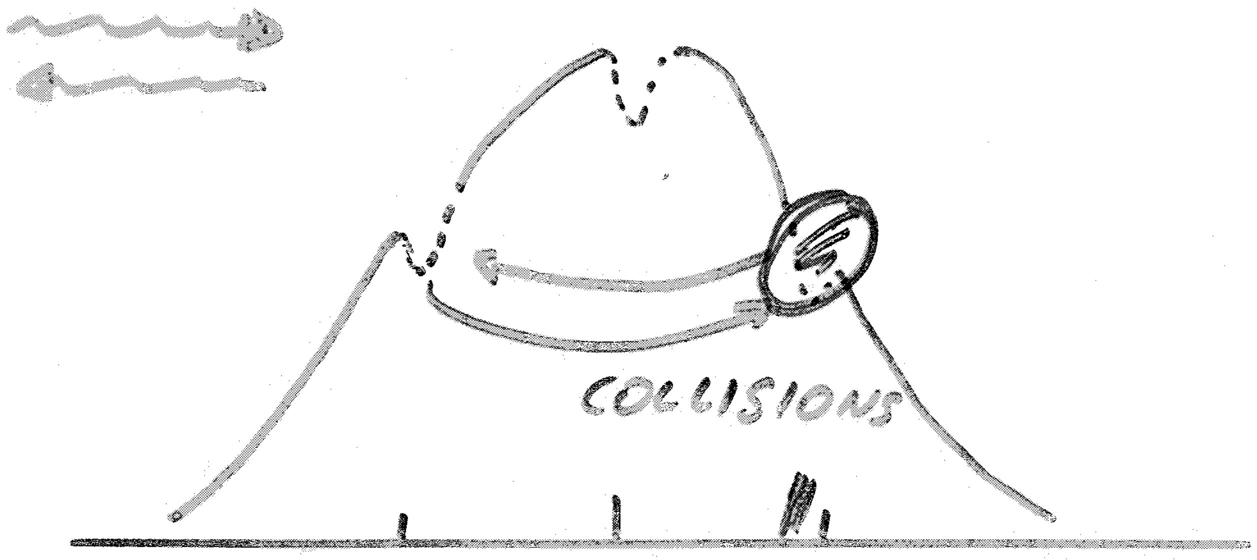
FINALLY

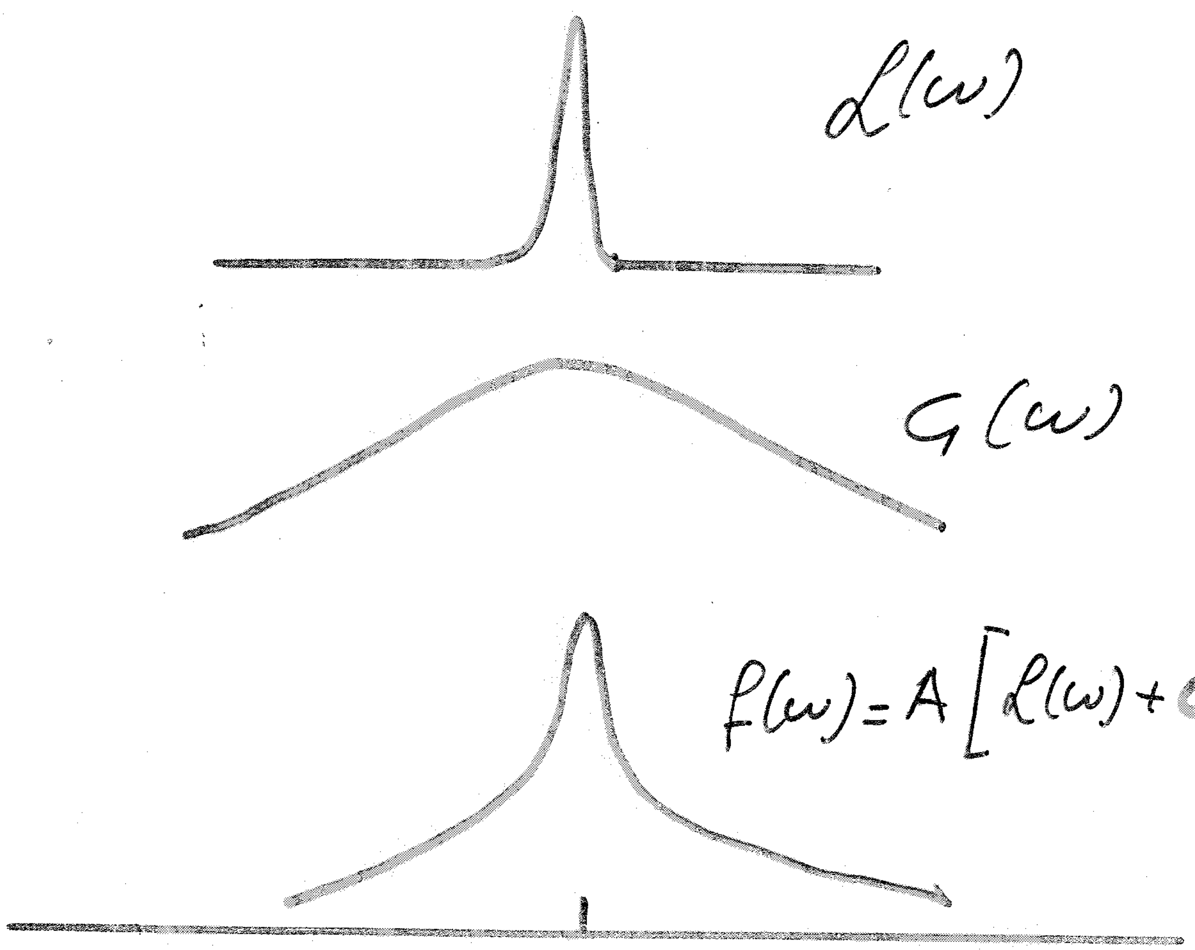
$$S_{OG}^{(\omega_1+\omega_2)} = K \cdot e^{\left[\frac{(\omega-\omega_0)}{\Delta\omega_D}\right]^2} \cdot \frac{(\gamma_s/2)^2}{(\omega-\omega_0)^2 + (\gamma_s/2)^2}$$

If $\gamma_s \ll \Delta\omega_D$

$$S_{OG}^{(\omega_1+\omega_2)} = K \frac{(\gamma_s/2)^2}{(\omega-\omega_0)^2 + (\gamma_s/2)^2} \quad \text{HOMOGENEOUS LINESHAPE}$$

VELOCITY CHANGING COLLISIONS

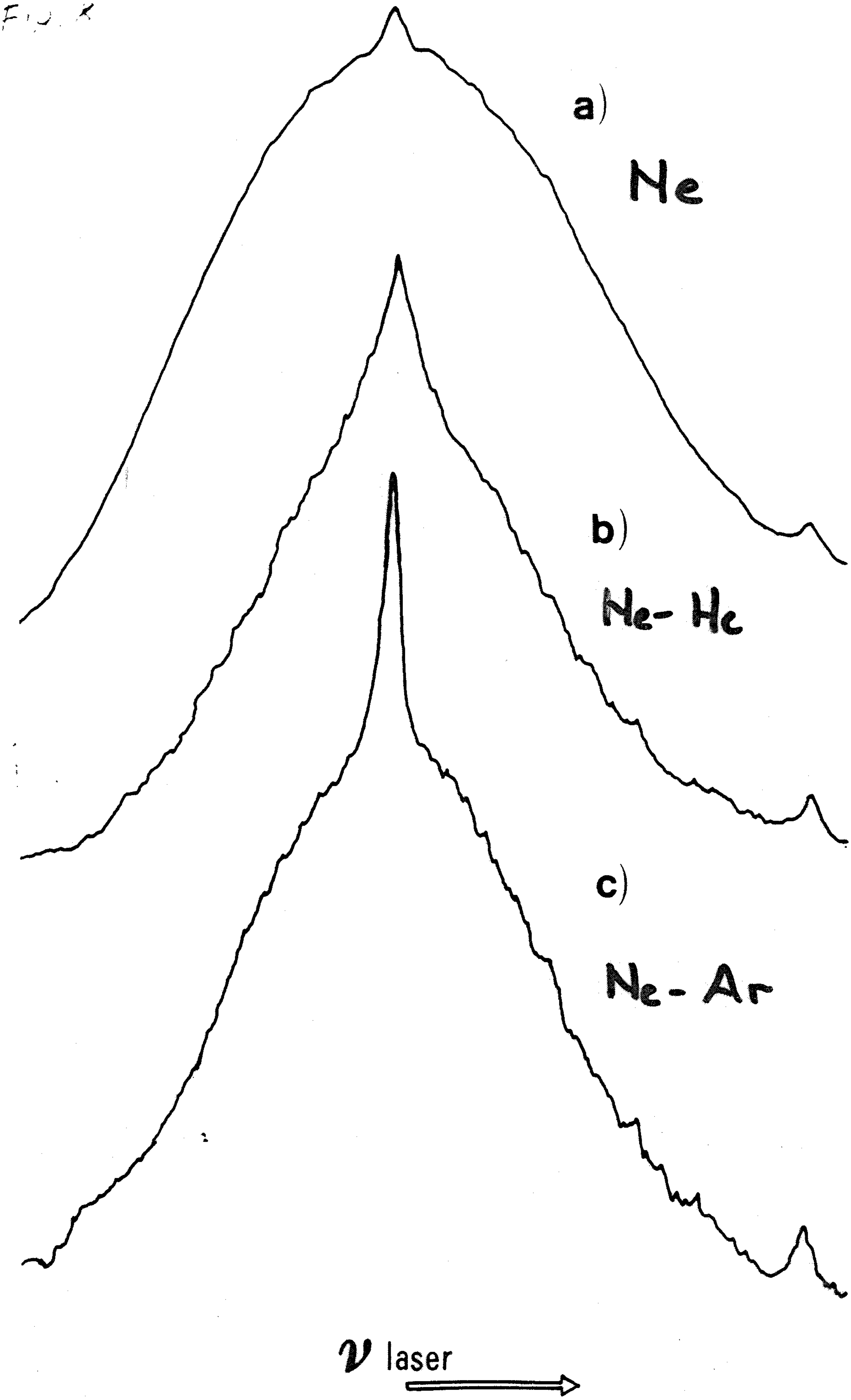




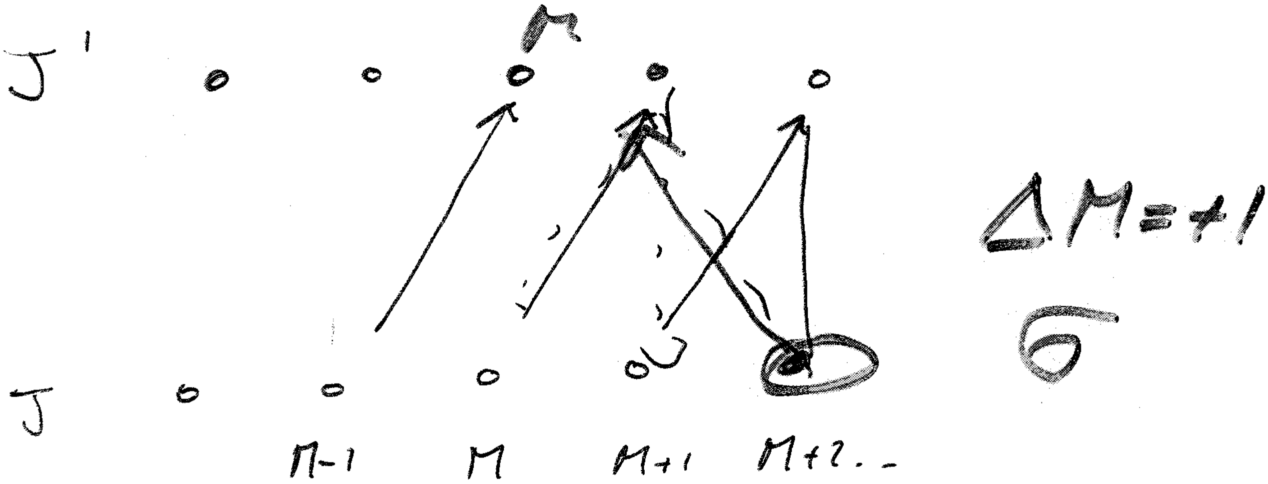
$$f(\omega) = A\left[L(\omega) + C\,G(\omega)\right]$$

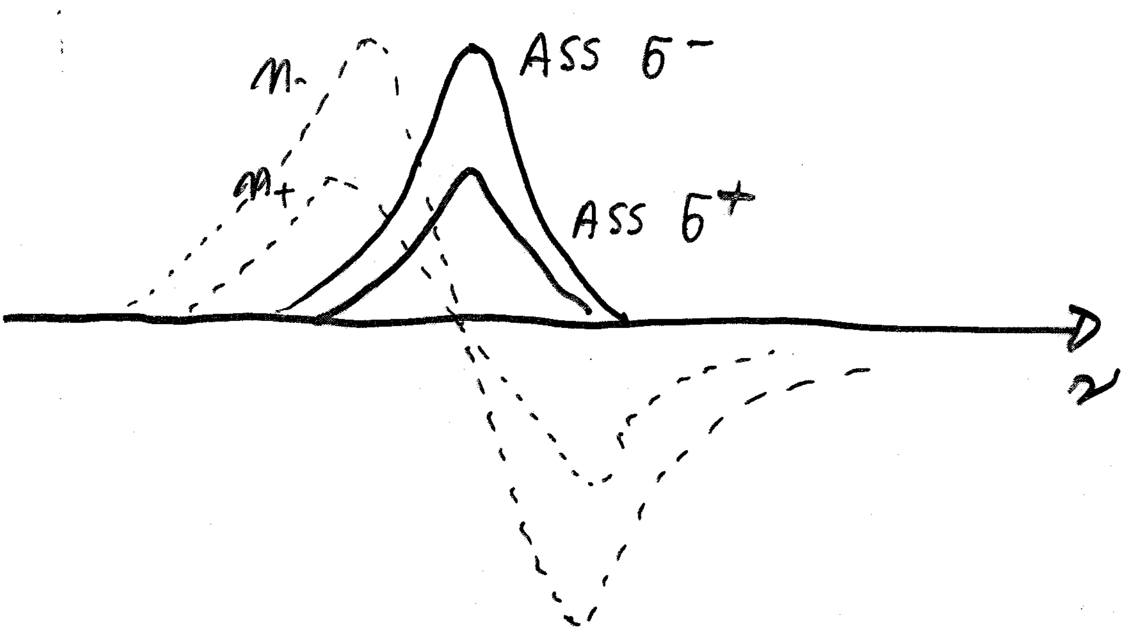
$$C \sim 2\sqrt{\pi}\,\lg 2\,\frac{\Gamma_1 \gamma}{\gamma_2\,\Delta\gamma_D}$$

Fig. 8

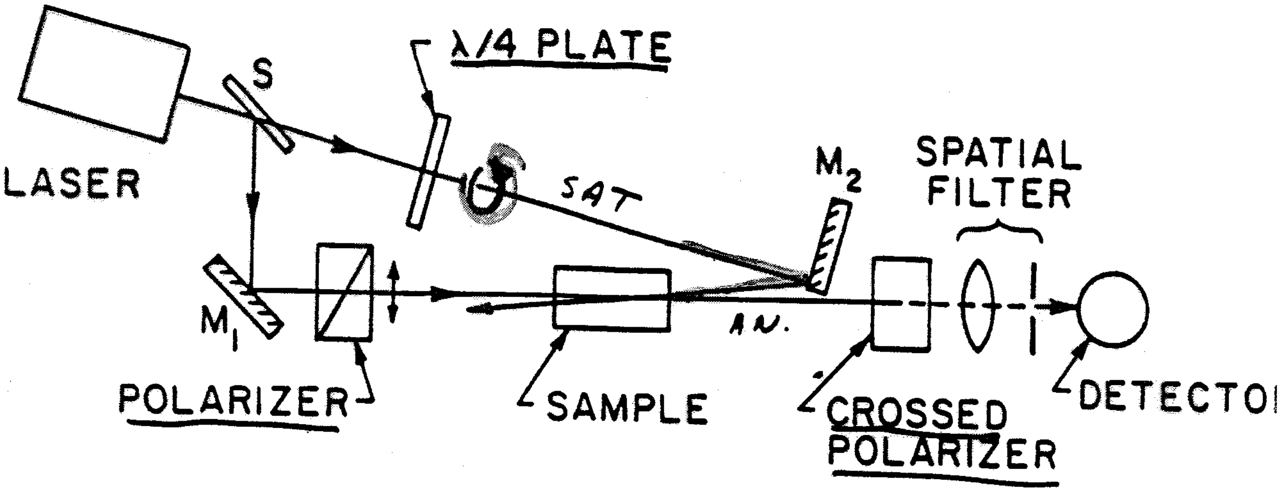


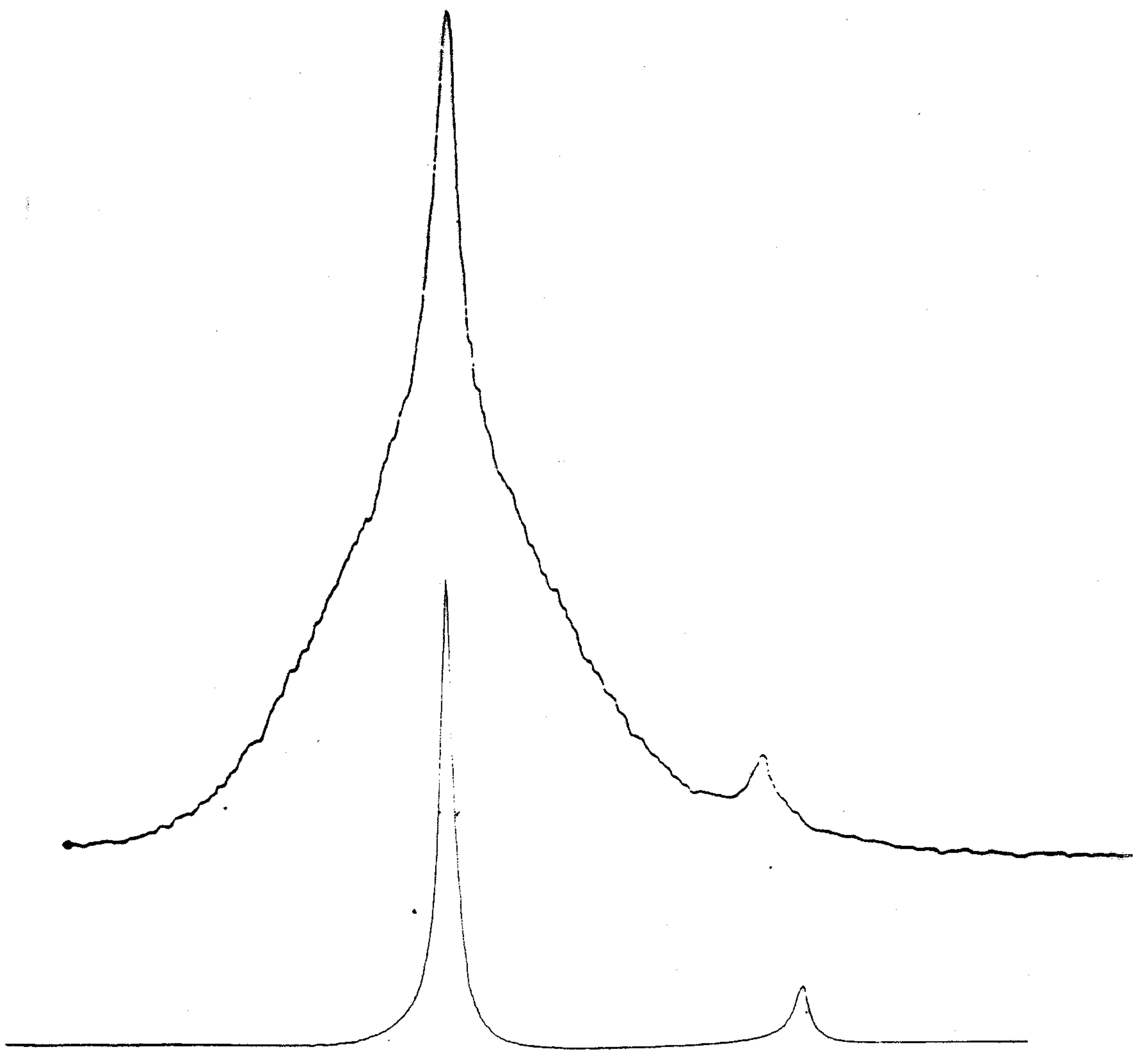
CON LUCE POL. CIRCOLARE
PROVOCO UN'ORIENTAZIONE
(SAT. SELETTIVA IN M)



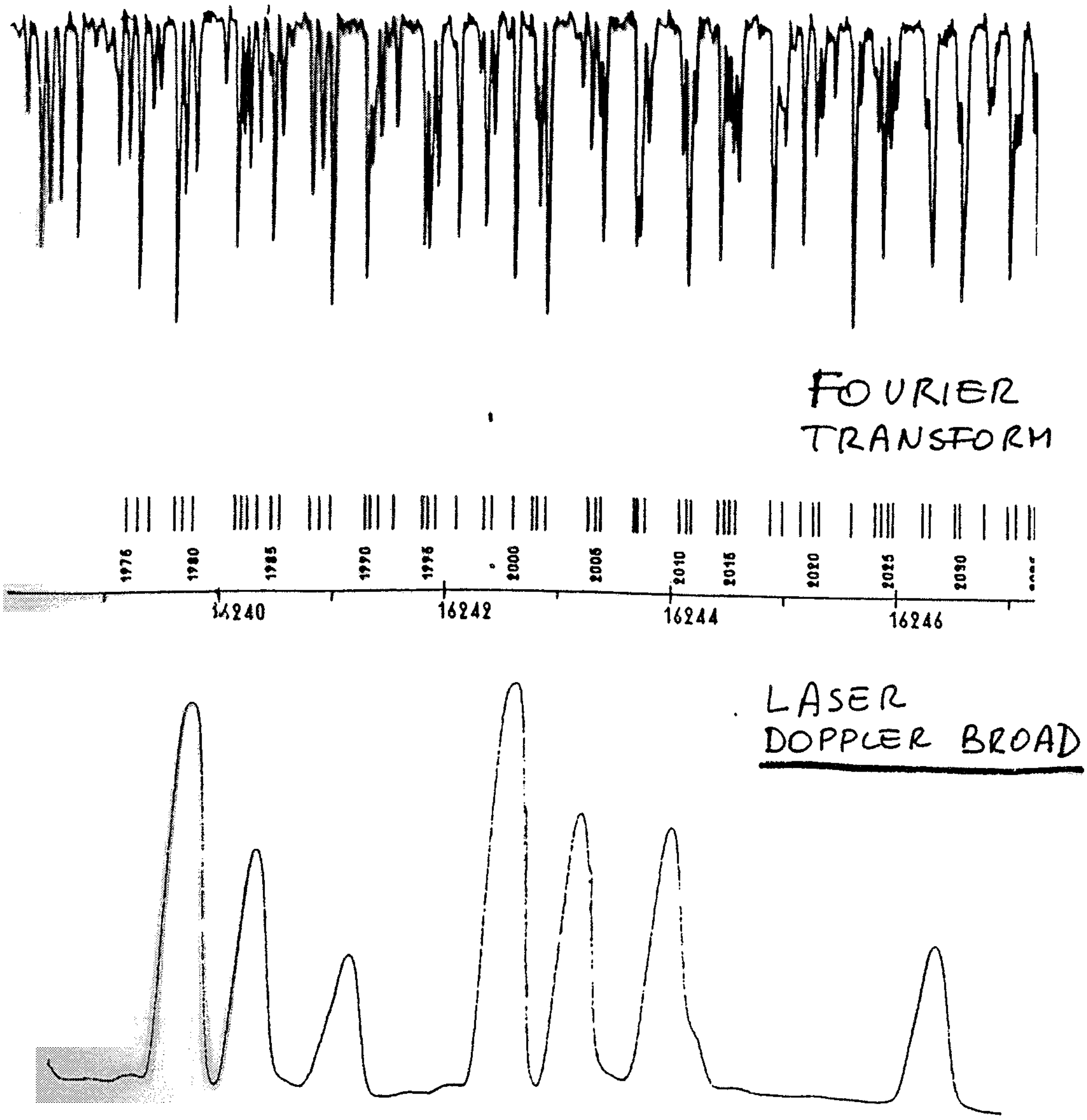


CIRCULARLY POLARIZED LIGHT
CREATES AN ORIENTATION
(SATURATION SELECTIVE IN M)


λ/4 PLATE
S
LASER
SAT
M2
SPATIAL
FILTER
M1
AN.
POLARIZER
SAMPLE
CROSSED
POLARIZER
DETECTO



I_2 SPECTRUM (GOOD FOR λ CALIBRATION) 9



1939	16232.7893	2.1	24	1971	16238.4971	1.1	32	2003	16242.8937	.7	51	2035	16247.2425	.7	42
1940	16232.9659	.8	32	1972	16238.5940	.6	31	2004	16243.2738	.6	23	2036	16247.4690	1.7	17
1941	16233.0872	.7	33	1973	16238.7526	.5	38	2005	16243.3428	.9	17	2037	16247.6540	.5	44
1942	16233.1332	.4	41	1974	16239.1054	.6	26	2006	16243.3862	.5	38	2038	16247.8451	6.6	17
1943	16233.2791	2.4	15	1975	16239.1964	.6	25	2007	16243.6739	.7	39	2039	16247.8973	.7	38
1944	16233.6513	1.0	20	1976	16239.2974	.5	47	2008	16243.7098	.8	38	2040	16248.2979	.6	43
1945	16233.7256	2.3	45	1977	16239.4006	1.8	16	2009	16243.7774	1.0	21	2041	16248.4641	.6	27
1946	16233.7472-	.9	40	1978	16239.6225	.4	53	2010	16244.0828	1.0	24	2042	16248.5285	.5	46
1947	16233.8414	.7	25	1979	16239.6932	.7	31	2011	16244.1465	1.3	46	2043	16248.6747	.8	21
1948	16234.1210	.5	48	1980	16239.7828	.7	27	2012	16244.1902	.7	18	2044	16248.9210	.6	37
1949	16234.4643	.8	31	1981	16240.1529	.5	40	2013	16244.4252	.6	42	2045	16249.1452	.5	33
1950	16234.5782	.6	30	1982	16240.2074	.8	21	2014	16244.4804	1.1	18	2046	16249.3674	.9	26

$\pm .002\ cm^{-1}$

$\pm 60\ MHz$

Fig. 15 Registrazione in fluorescenza stimolata di una regione dello spettro della molecola di I_2 e confronto con le righe tabulate .

DOPPLER + HYPERFINE

15 comp J even

21 comp J odd

~ EQUAL INTENSITY FOR J > 30

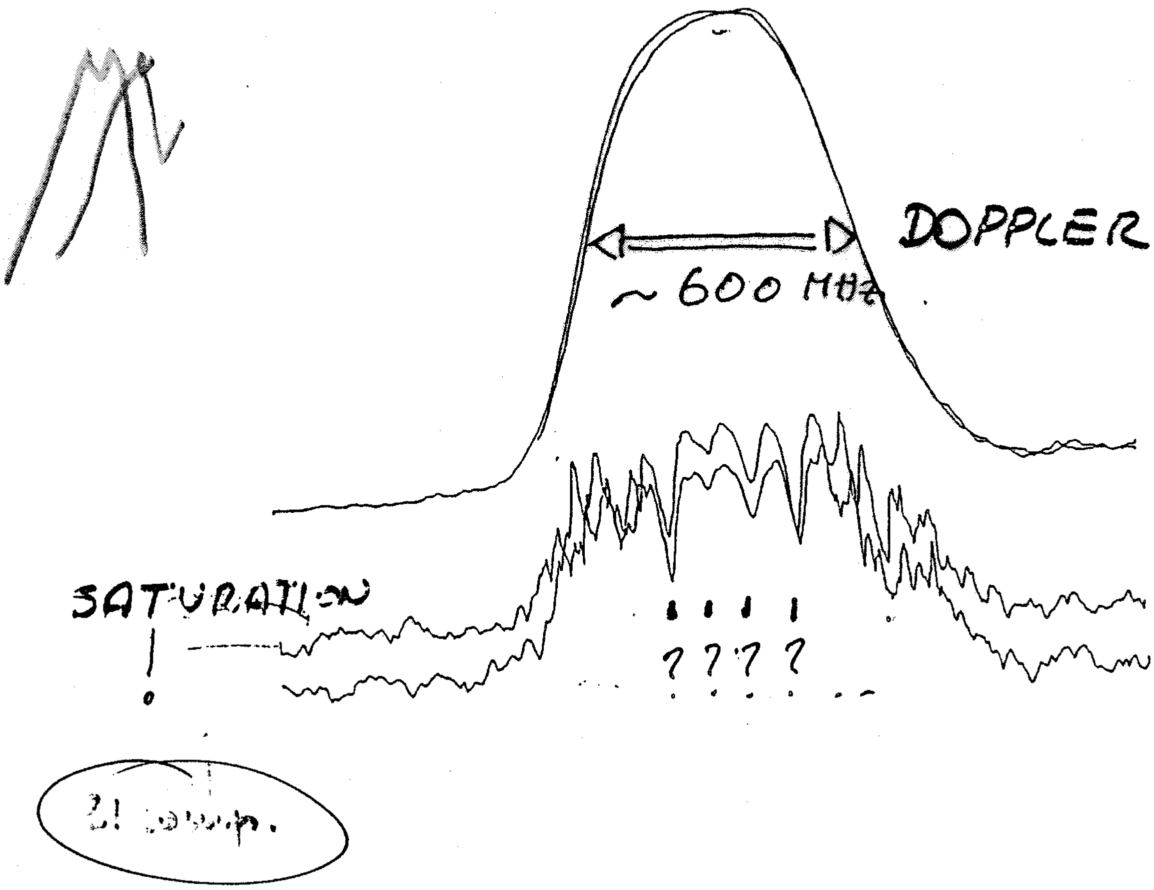
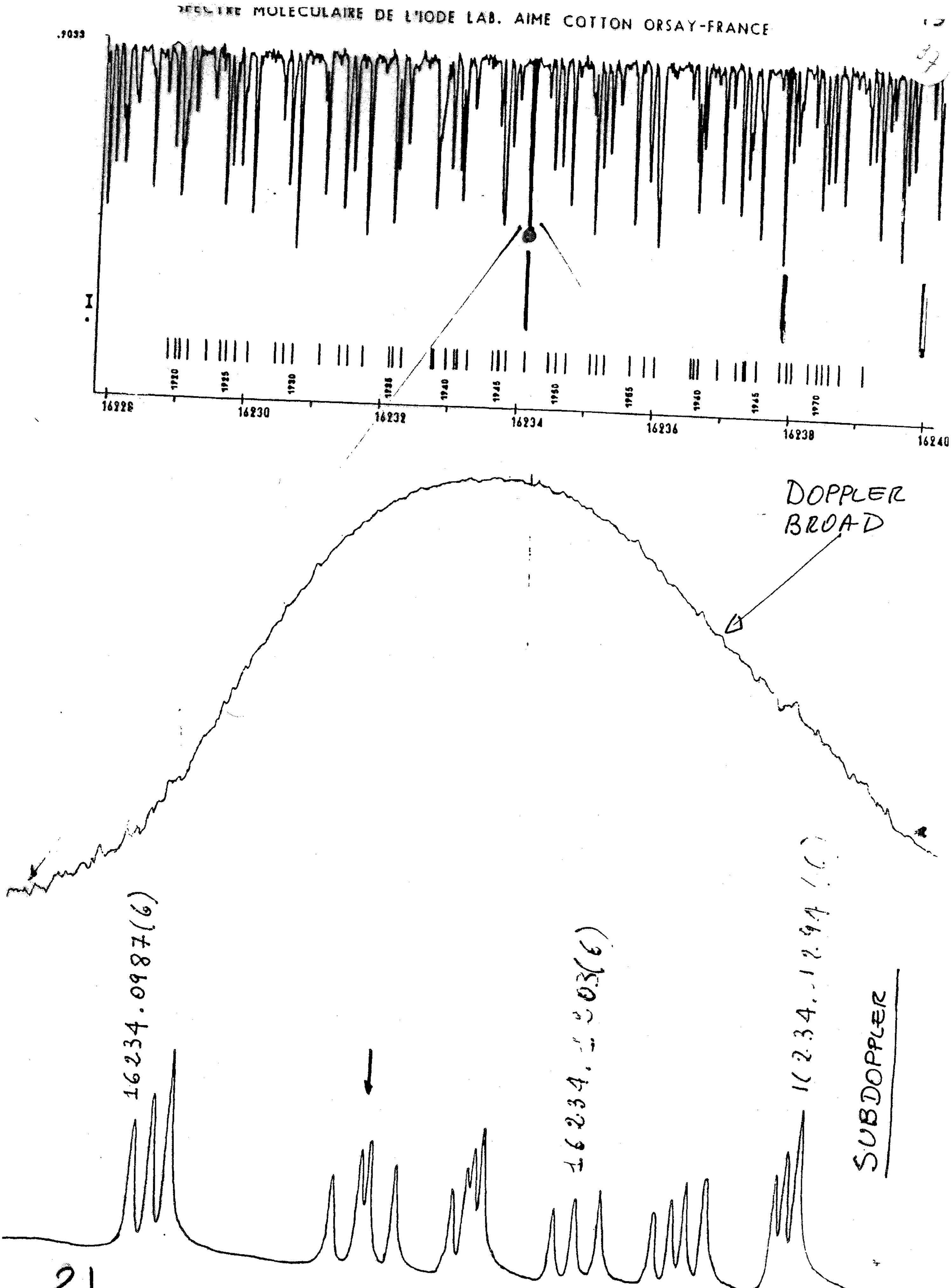


Fig. 22 Struttura iperfina della molecola di Iodio .
Profilo di assorbimento con fascio opposto saturante .

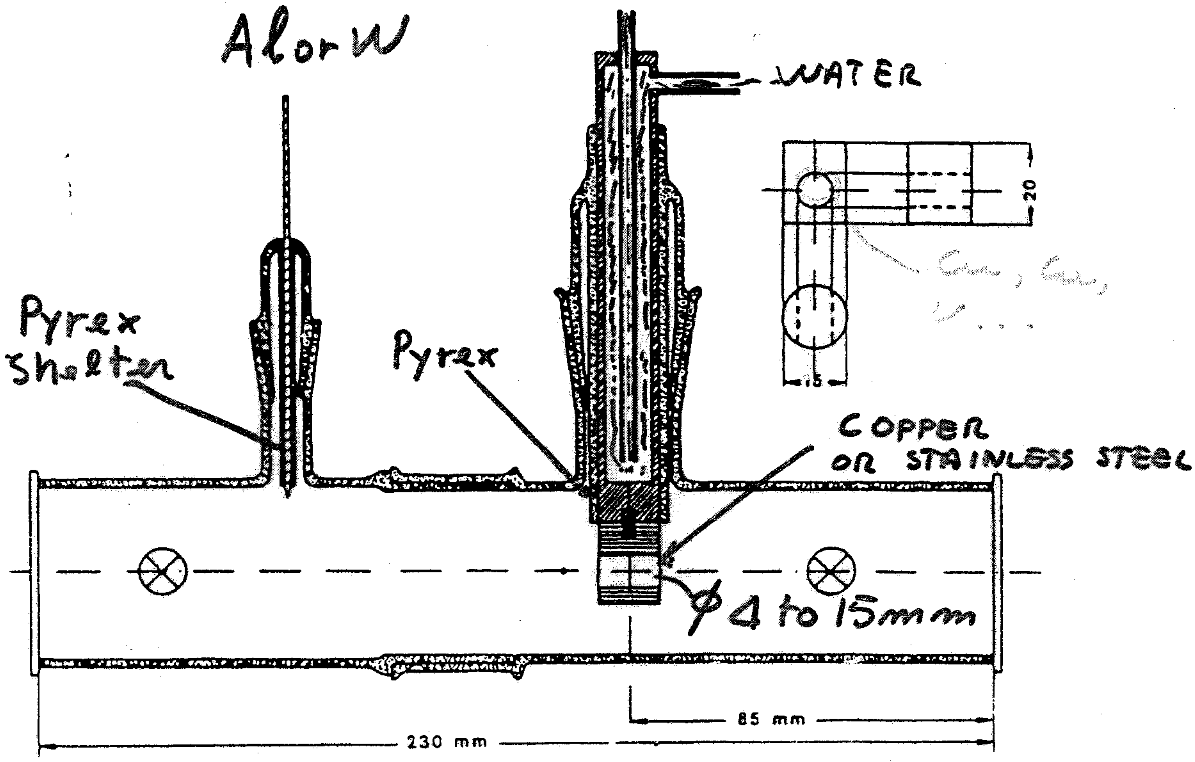


21 COMPON. Polarization spectra of hpf componen

OG INVESTIGATION OF REFRACTORY ELEMENTS

$I = 2 \div 50$ mA

p (Ar or Ne) ≈ 0.2 Torr
(sealed off)
OR
FLOWING INTO THE LASER

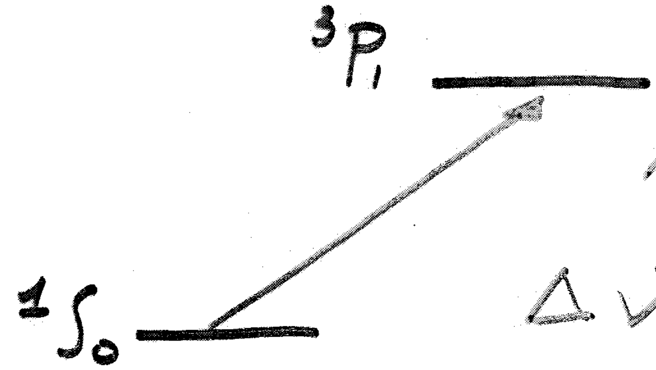


M. Inguscio. "High Resolution Intermodulated and Double Resonance Atomic Spectroscopy in a Hollow Cathode" J. Phys. (Paris) 44 C (1983)

Hollow Cathode Discharge

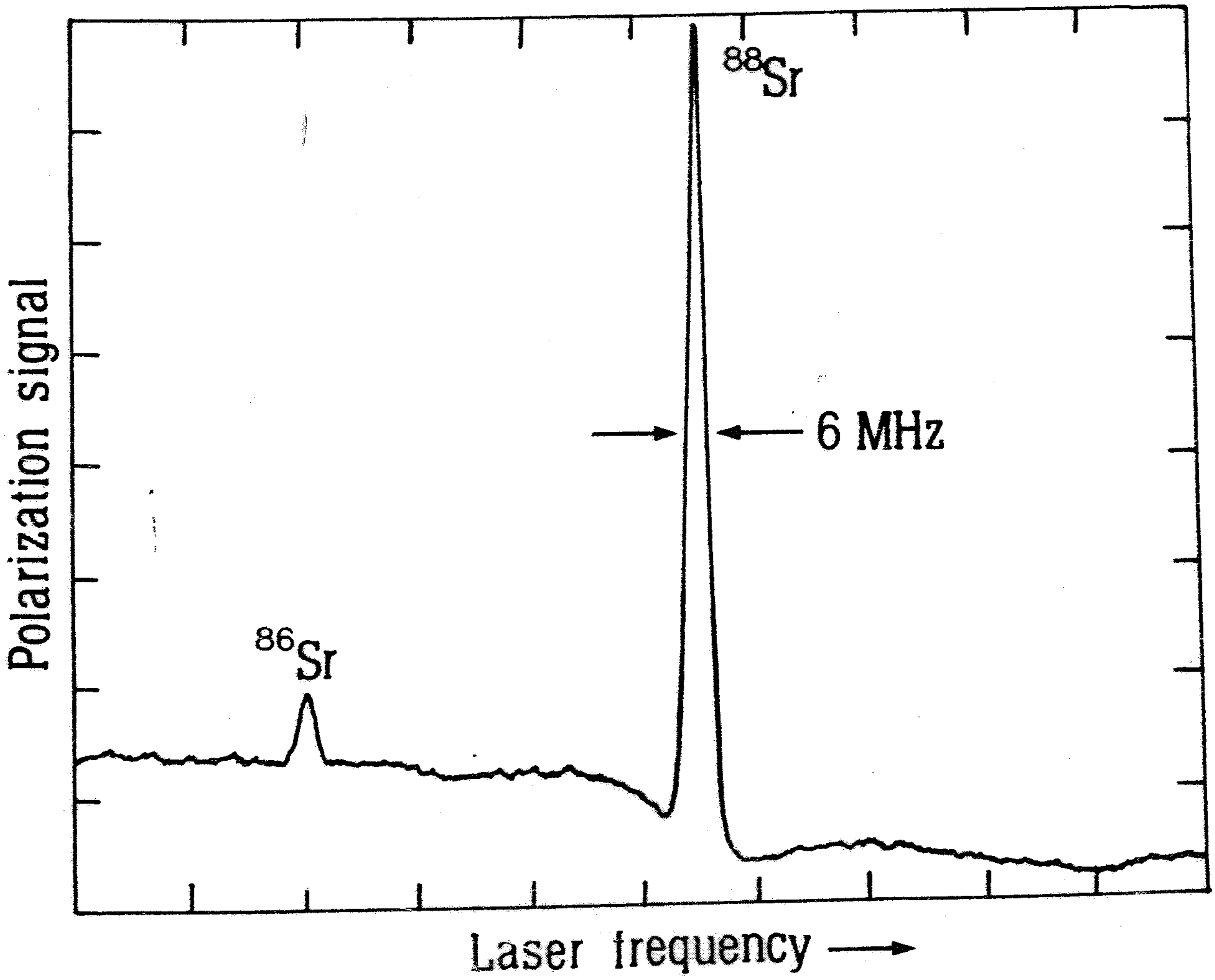
Sr intercombination line

83



$\lambda = 689\ nm$

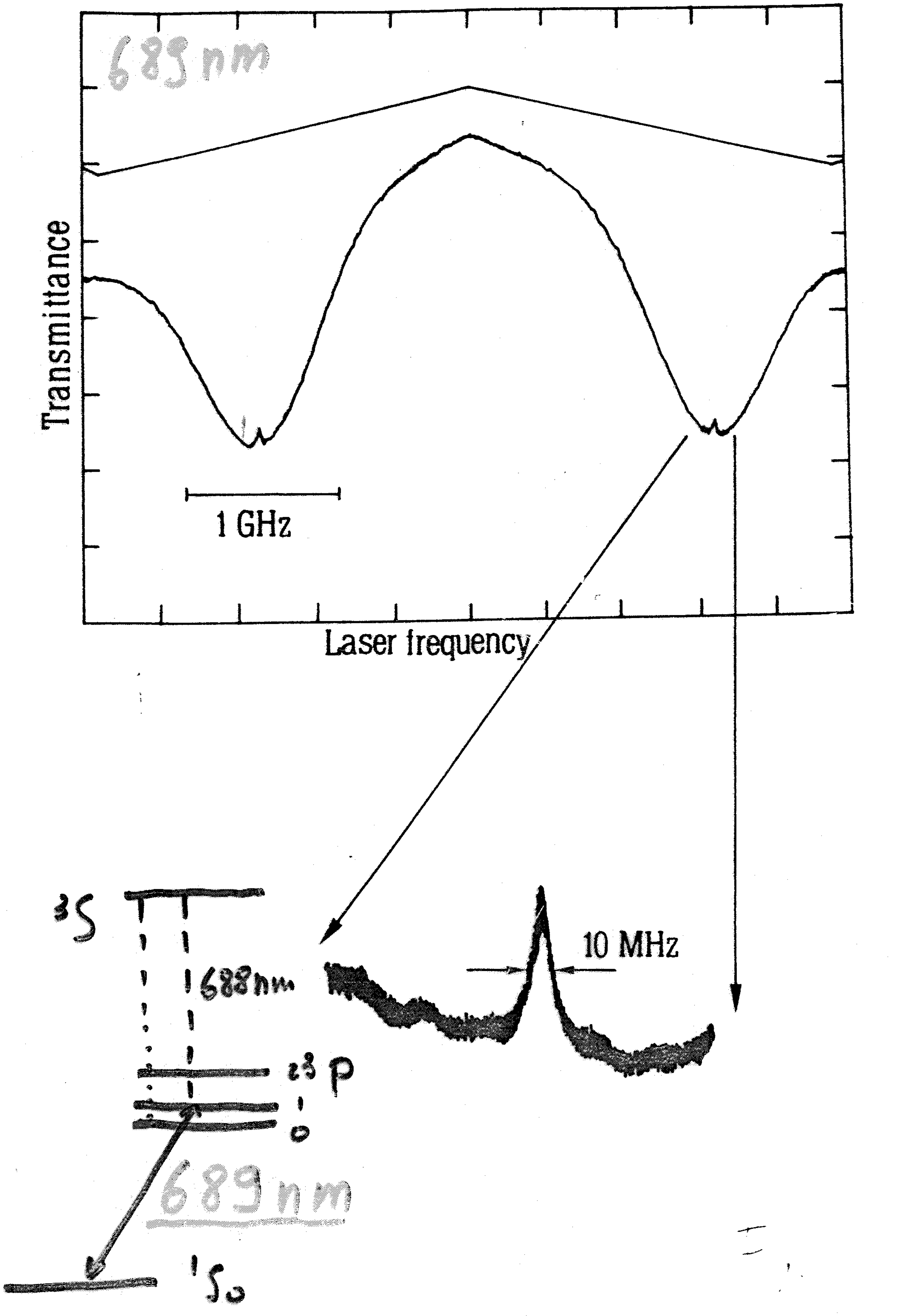
$\Delta \nu_{RAD} \sim 60\ KHz$



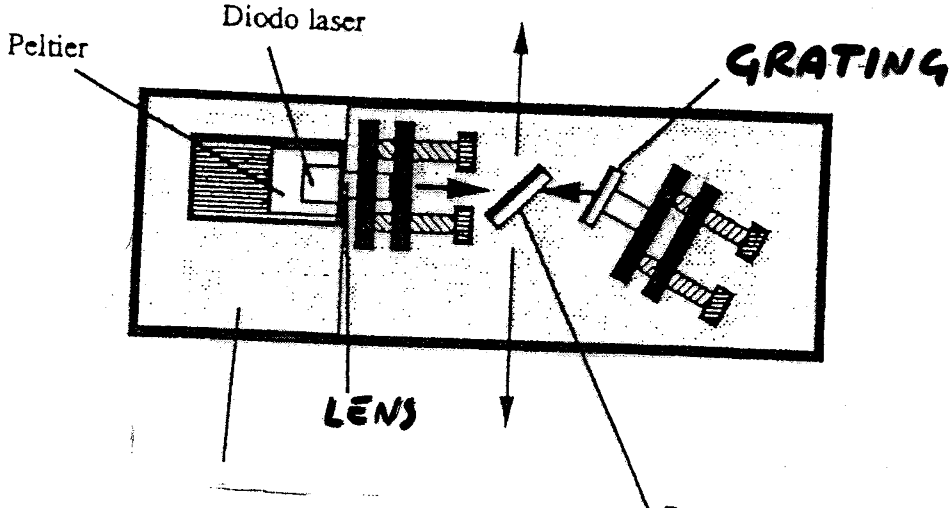
POLARIZATION SPECTROSCOPY

(HOLLOW CATHODE DISCHARGE)

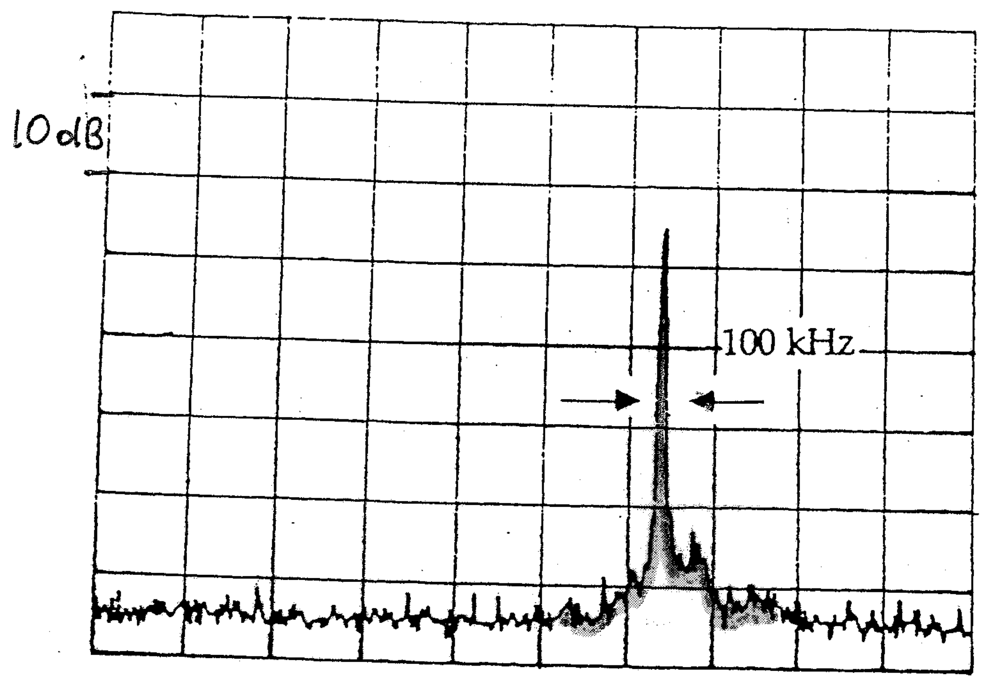
ABSORPTION FROM Sr IN A HOLLOW CATHODE



GRATING FEEDBACK NARROW BAND SEMICONDUCTOR DIODE LASERS



$\frac{\Delta\nu}{\nu} \lesssim 10^{-11}$ (fast)

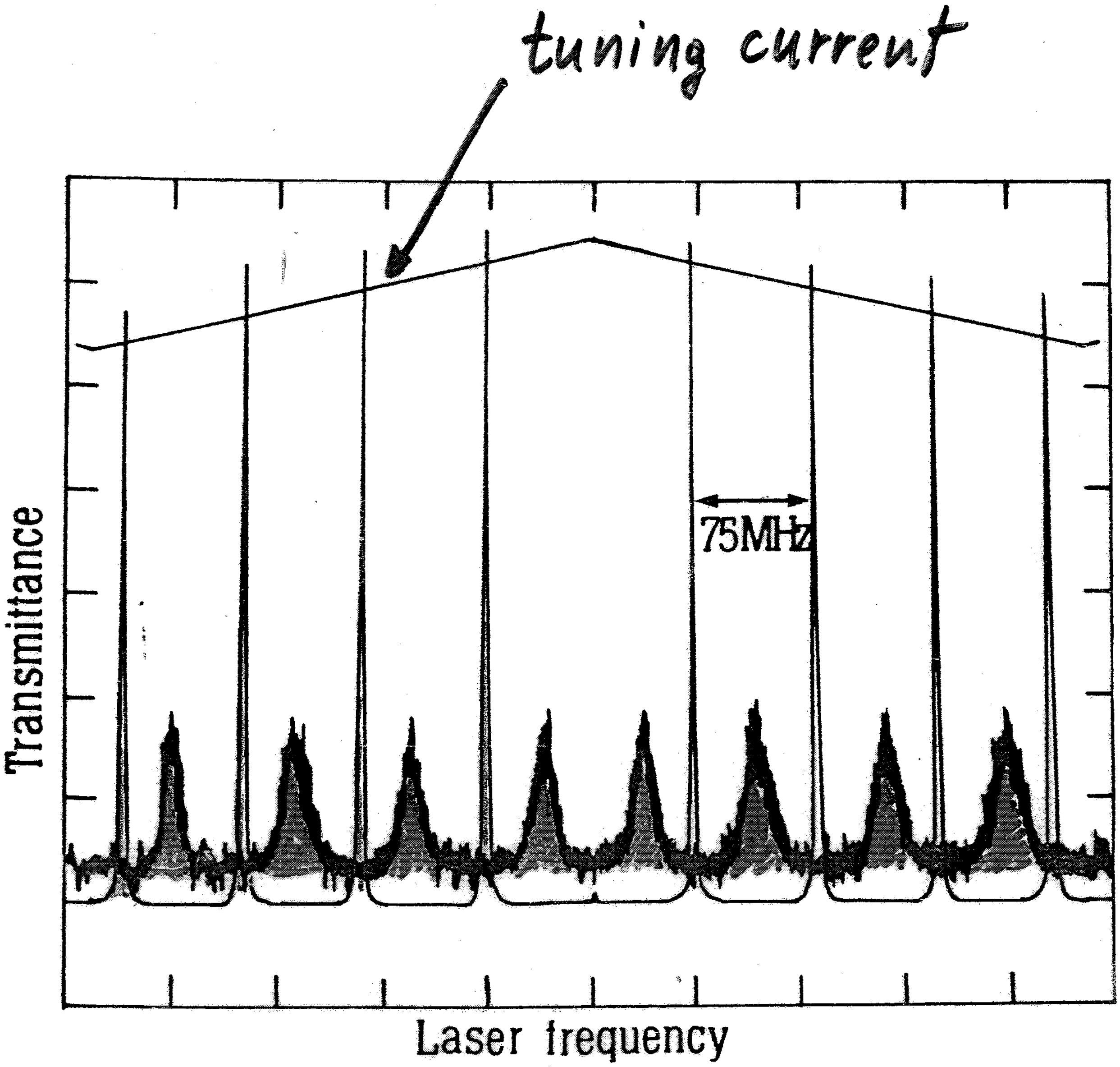


BEATING 2 INDEPENDENT SDL

2 GHz apart 50 msec SWEEP

InGaP laser at 690 nm

LASER → (   )

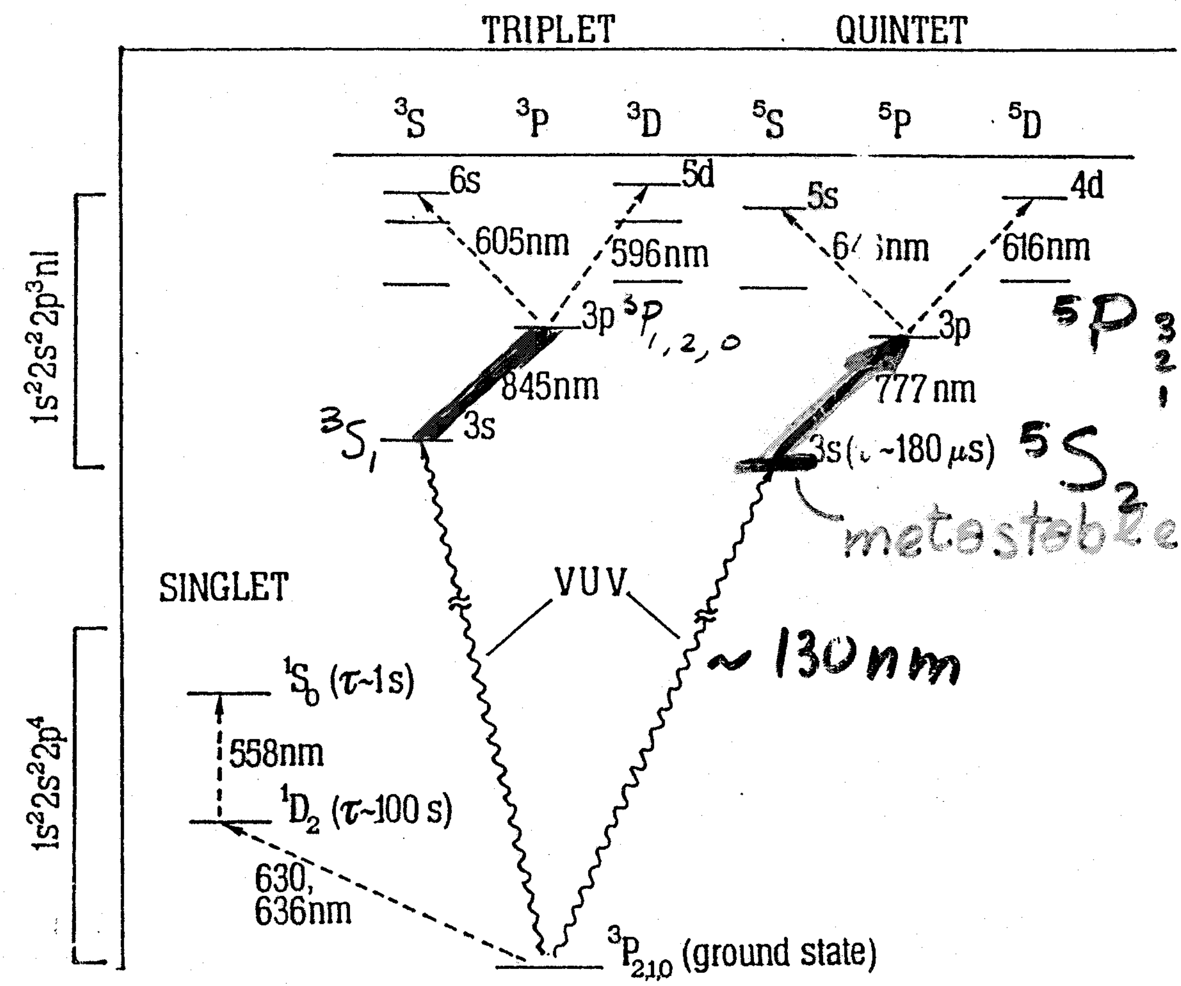


G. Tino, M. de Angelis, L. Gianfrani, M. Barsanti, M. I.

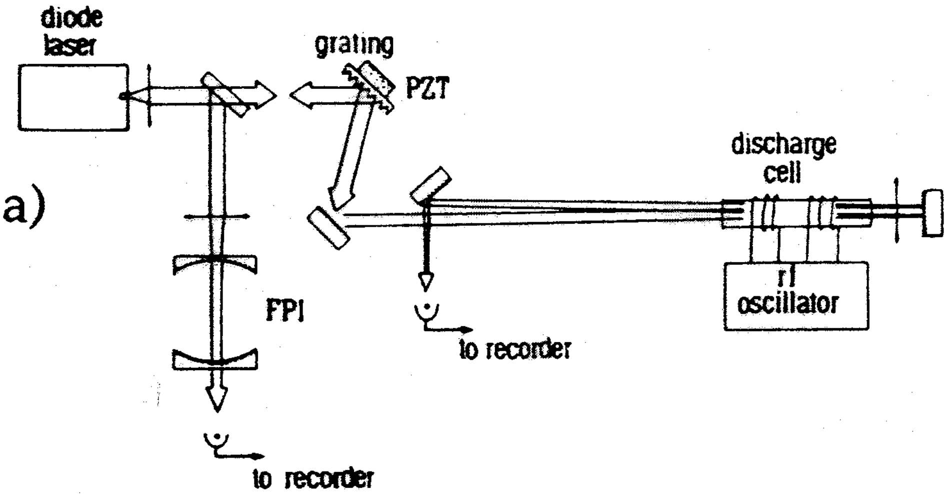
Appl. Phys. B (1992) (NAPOLI + FIRENZE)

high resolution spectroscopy
and OXYGEN-ATOM

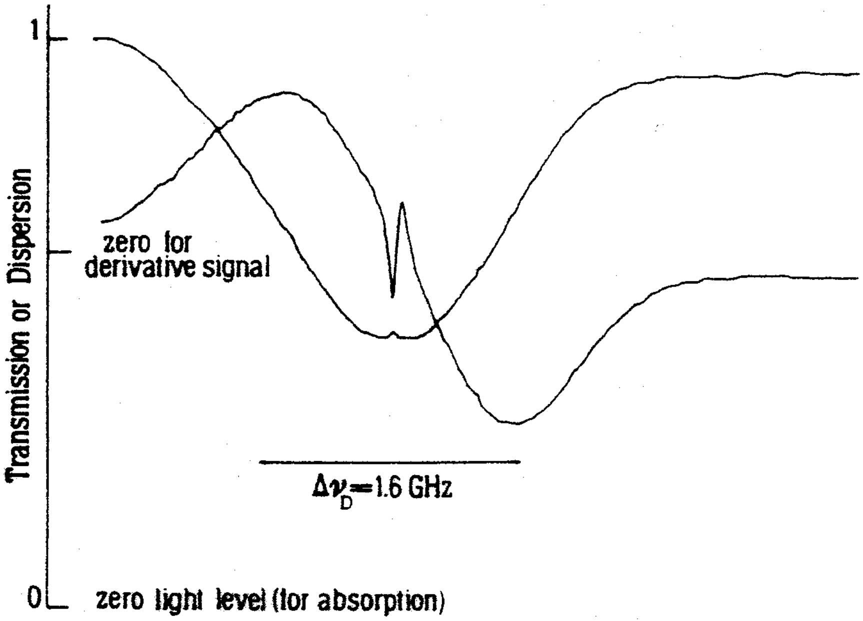
- Isotope and hyperfine structures
- Cooling



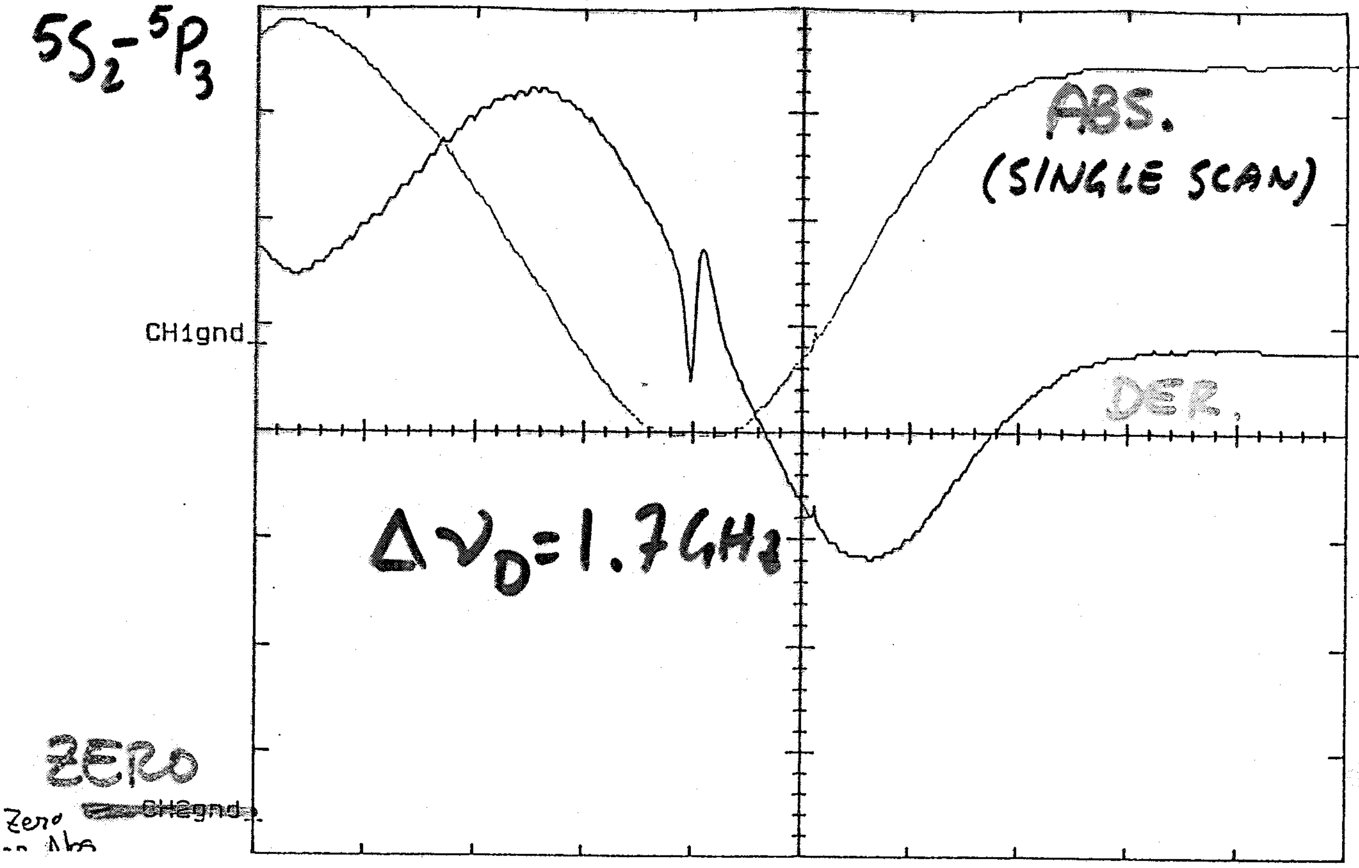
main difficulty is the level scheme of the atom itself a)


diode laser
grating
PZT
discharge cell
FPI
rf oscillator
to recorder
to recorder

b)


1
Transmission or Dispersion
zero for derivative signal
Δν_D=1.6 GHz
0
zero light level (for absorption)

Abs vs

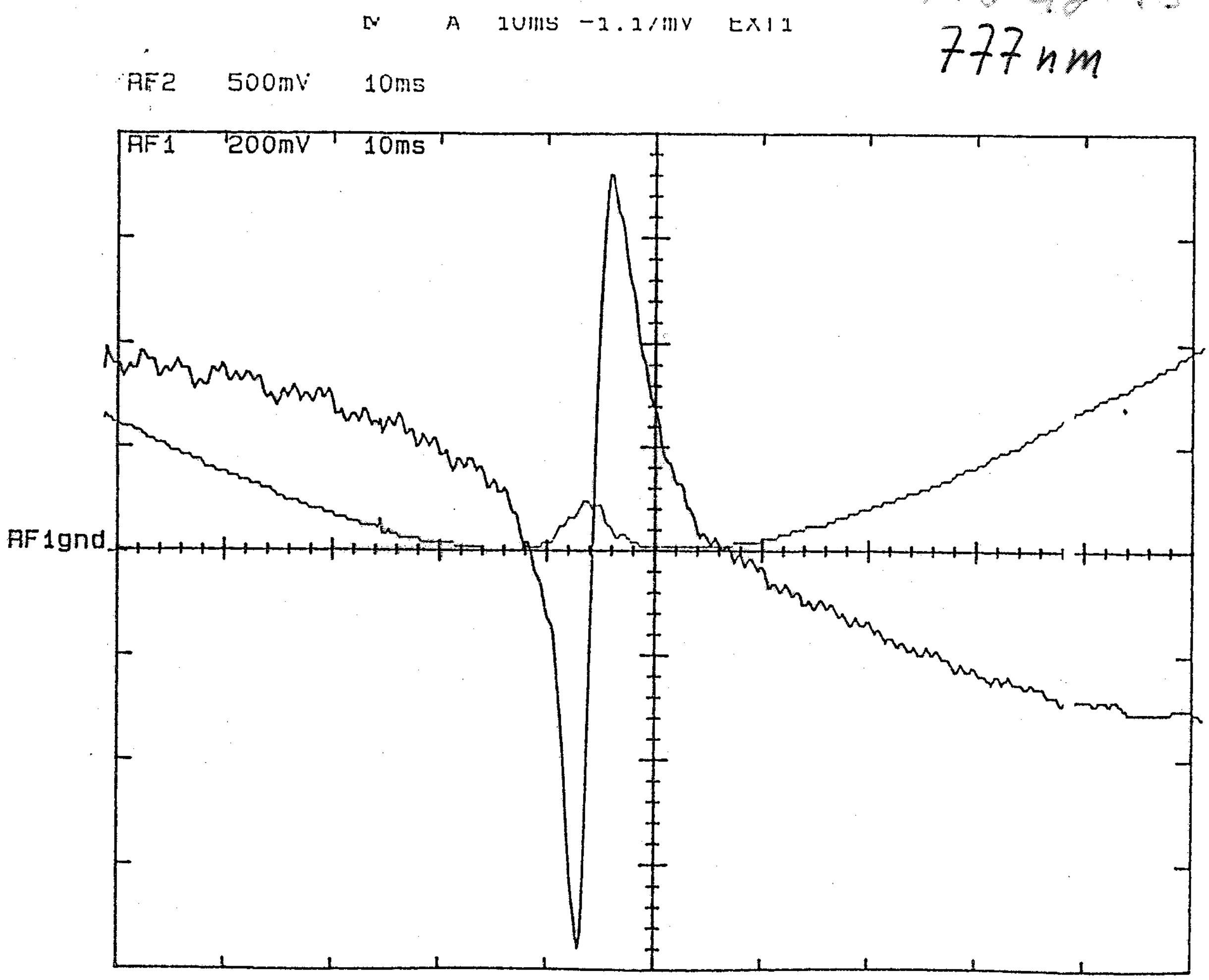


note the low noise level!

SATURATION SPECTROSCOPY

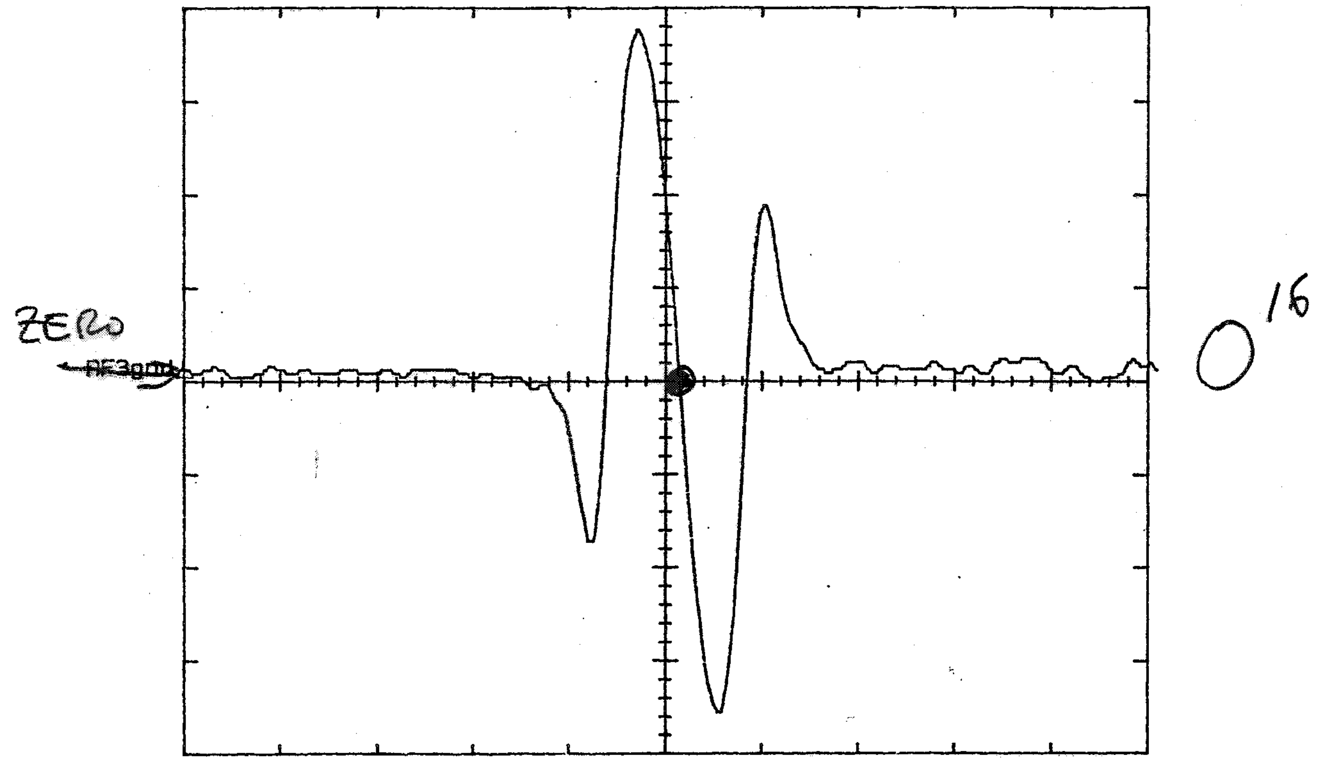
AlGaAs
777 nm

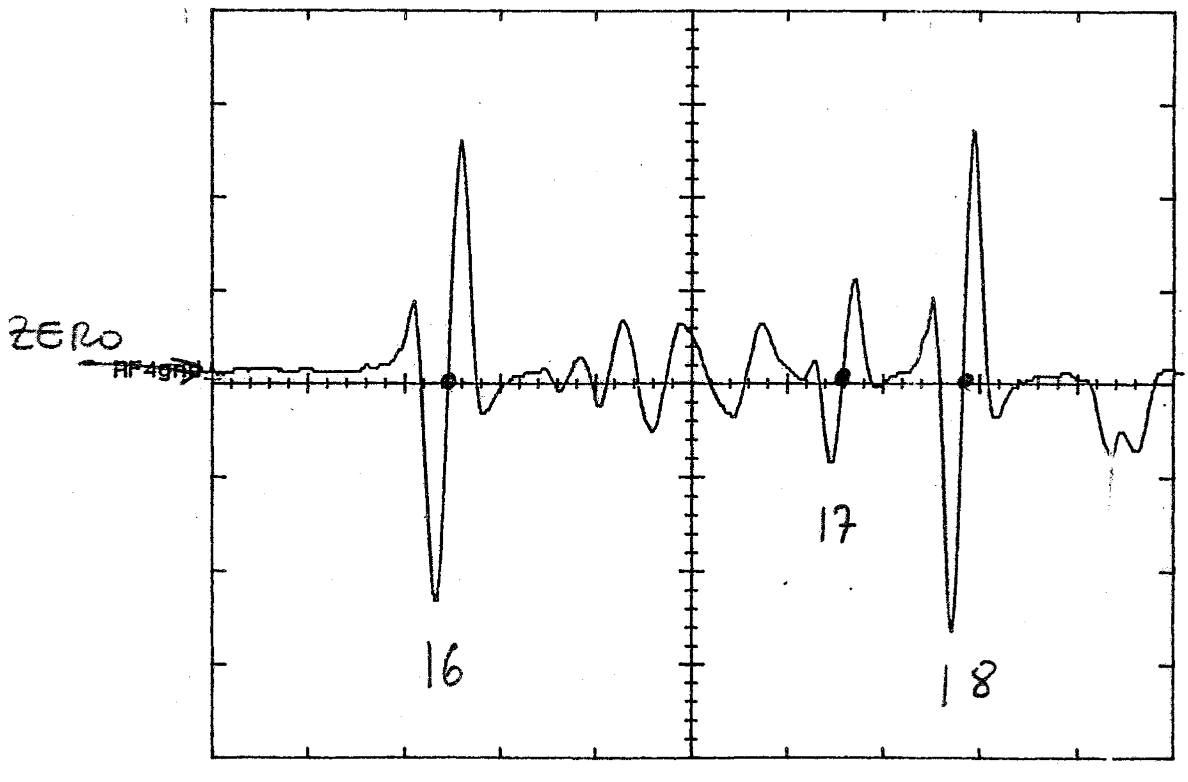
9/2



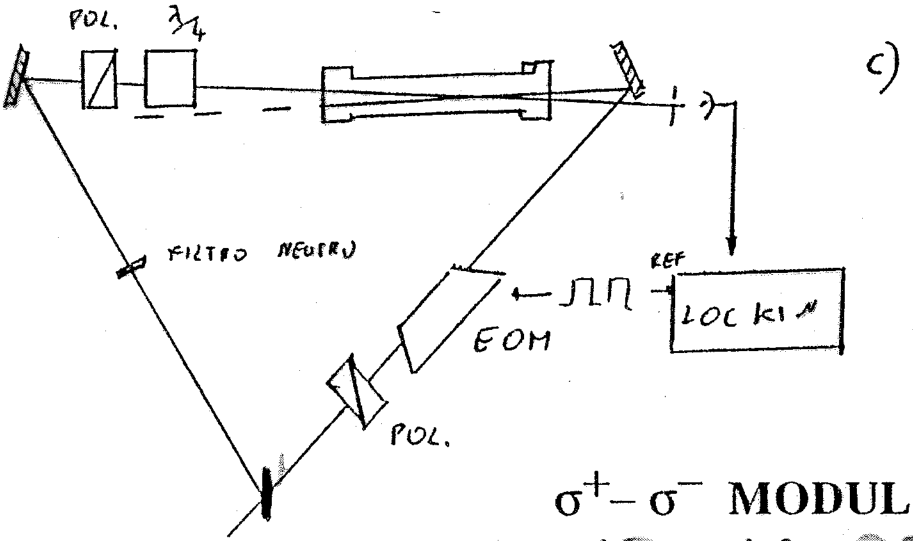
P.R. LETTERS 64 2999 (1990) G.M. Tino et al.

3rd DERIVATIVE (NO BASELINE)





25/9/90 F. MARIN, P. DENATALE, M. I., M. PREVEDELLI, L. R. ZINK and G. TINO OPTICS LETT. 15 Jan. 1992



c)

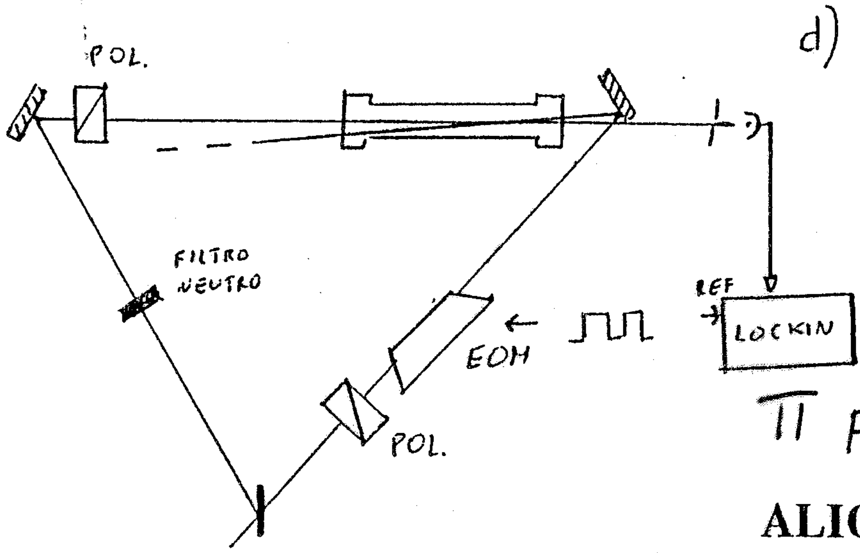
σ pump polarization

σ⁺– σ⁻ MODULATION

ATOMIC ORIENTATION

$$OSS = \sum_{-J}^{+J} n_{m_J} m_J$$

d)

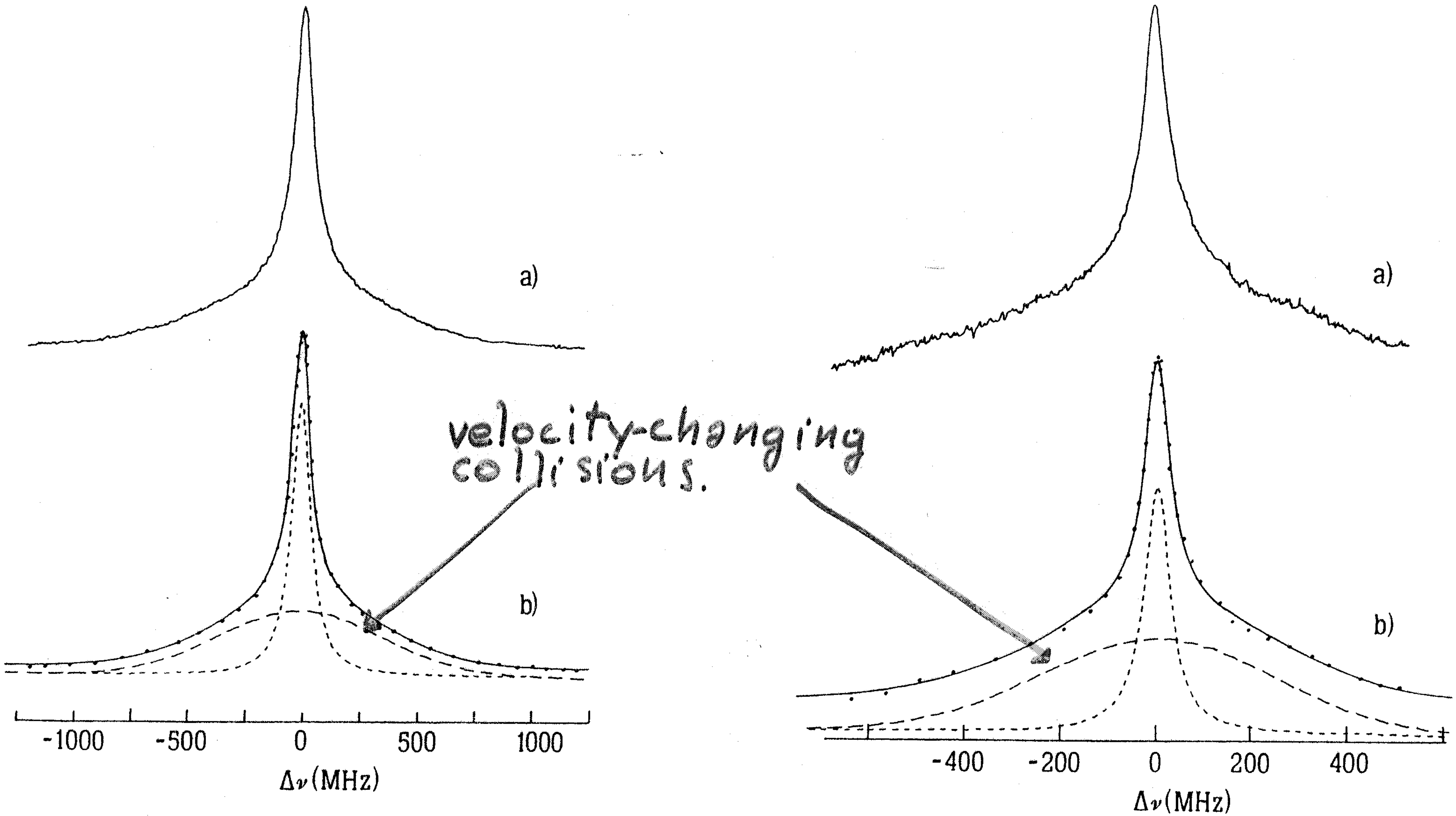


π pump polarization

ALIGNMENT

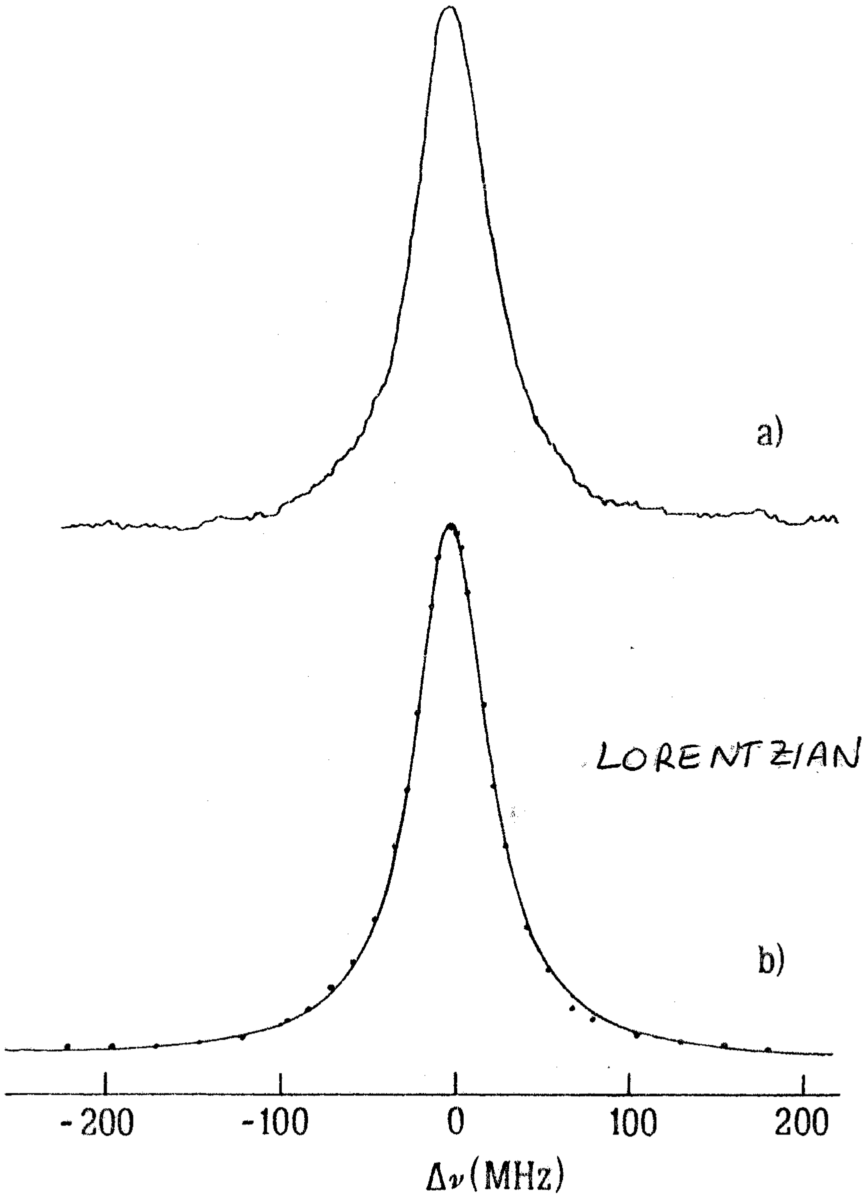
ATOMIC ALIGNMENT

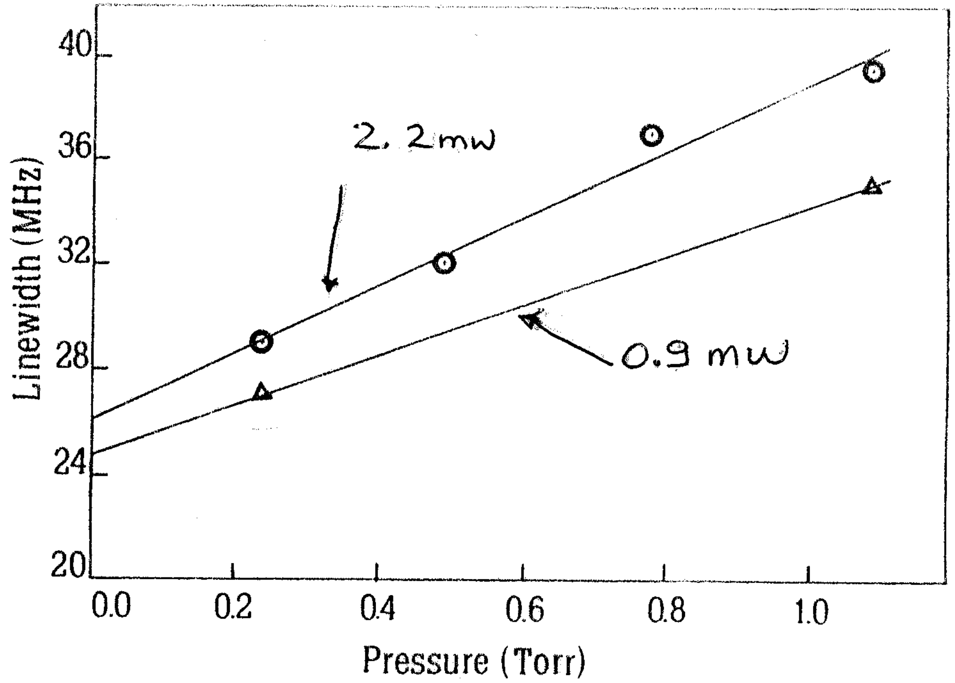
$$OSS = \sum_{-J}^{+J} n_{m_J} m_J^2$$



SATURATION (POPULATION)

σ^+ / σ^- (ORIENTATION)





$\tau(^5P_3) = 7\,\text{nsec}$

π

RADIATIVE LINEWIDTH LIMITED RESOLUTION

M. de Angelis et al. Phys. Rev. A 44 5811 (91)

High sensitivity molecular spectroscopy with semiconductor diode lasers (an introduction)

M. Inguscio # 3

MOLECULAR SPECTRA

ELECTRONIC – UV

VIBRATIONAL – IR

ROTATIONAL – MW → FIR

~ no VISIBLE – NEAR IR

Sources

Detectors

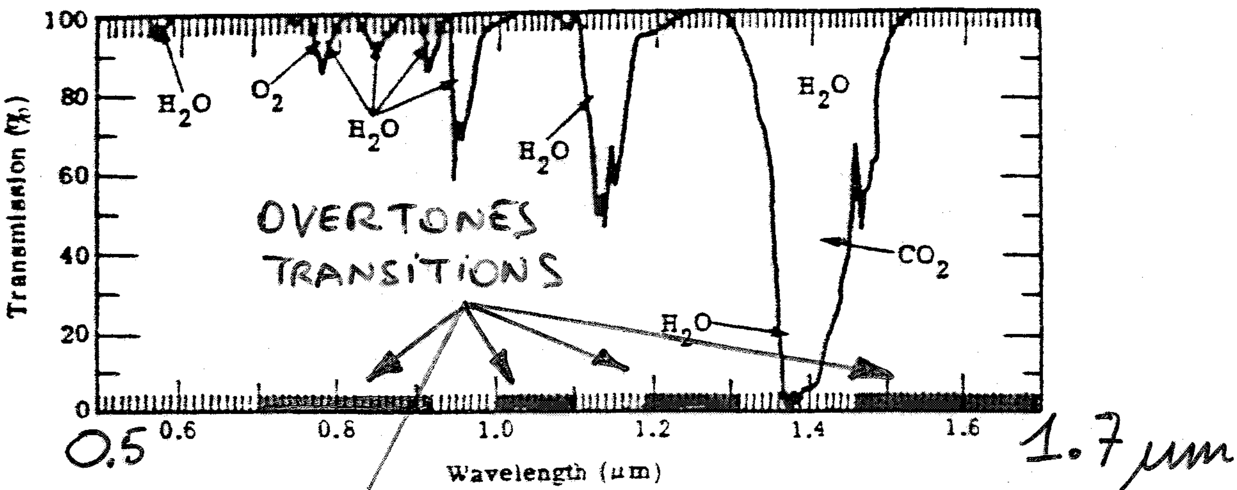
Transmission (Atmospheric or environment windows)

TRACE GAS SENSING

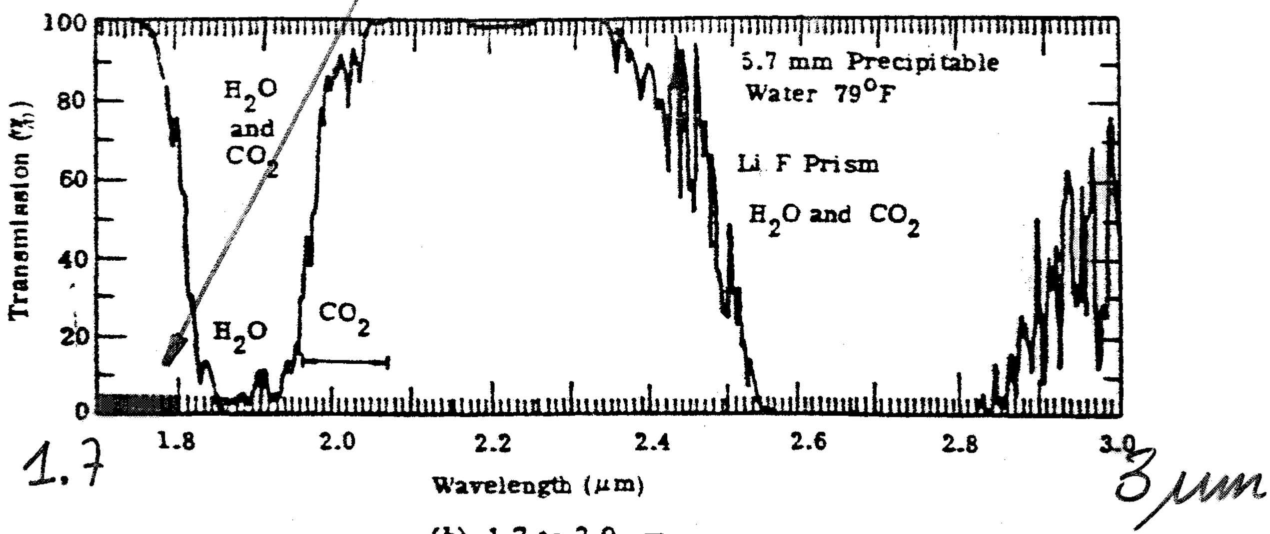
SEA LEVEL
100 meter path length

③

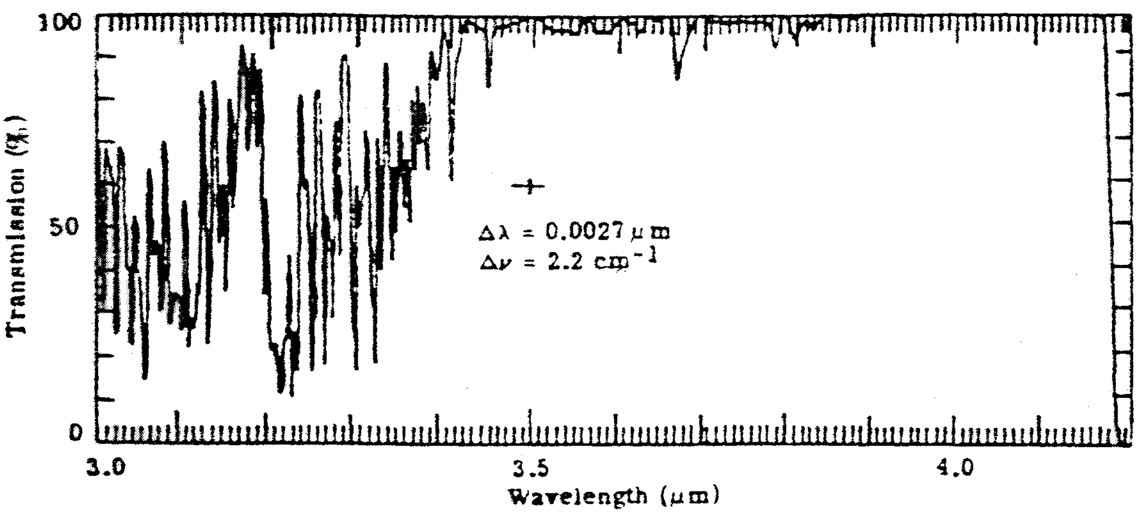
ATMOSPHERIC ABSORPTION



(*a*) 0.5 to 1.7 μm.



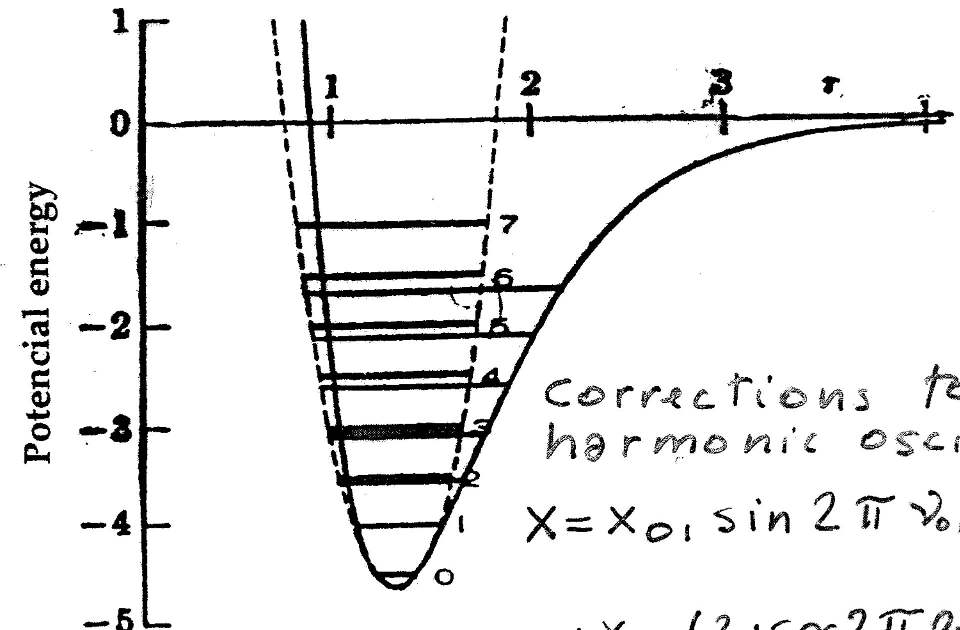
(*b*) 1.7 to 3.0 μm.



(*c*) 3.0 to 4.2 μm.

THE OVERTONE TRANSITIONS

(Intensity depends on the anharmonicity)

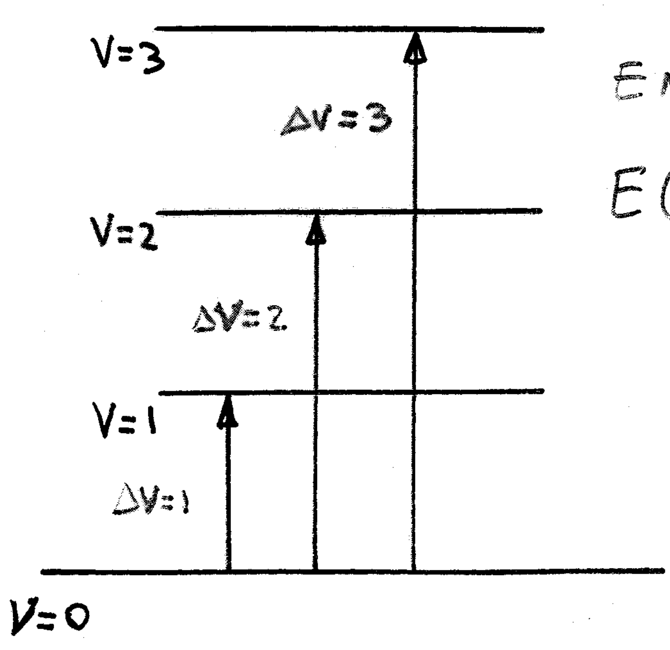


Corrections to the harmonic oscillation

$$X = X_{01} \sin 2\pi \nu_0 t \; +$$

$$+ X_{02} (3 + \cos 2\pi\, 2\nu_0 t) +$$

$$+ X_{03} \sin 2\pi\, 3\nu_0 t + \dots$$



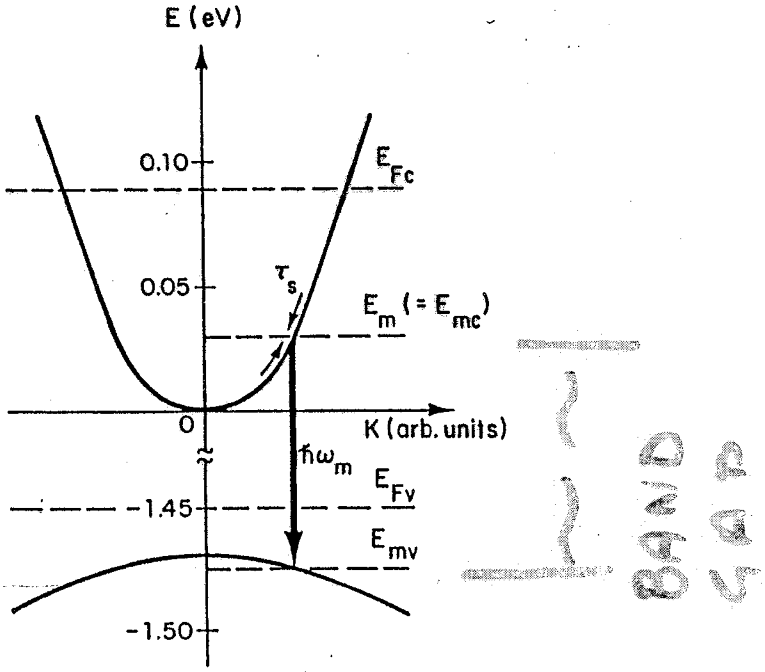
ENERGIES

$$E(v) = h\nu_0 \left(v+\tfrac{1}{2}\right) - h\nu_0 X_e \left(v+\tfrac{1}{2}\right)^2$$

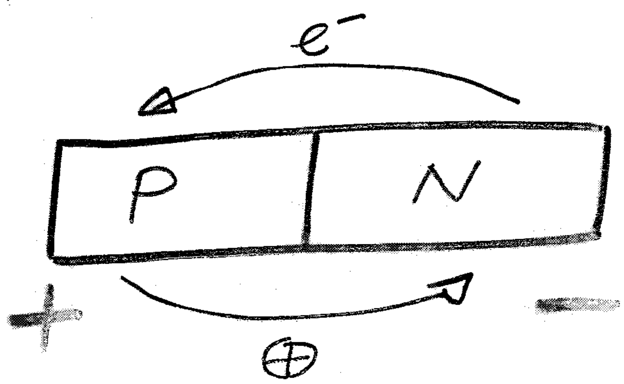
$$+ h\nu_0 y_0 \left(v+\tfrac{1}{2}\right)^3 \dots$$

DIRECT INTERBAND LASER TRANSITION

(Ga Al) As

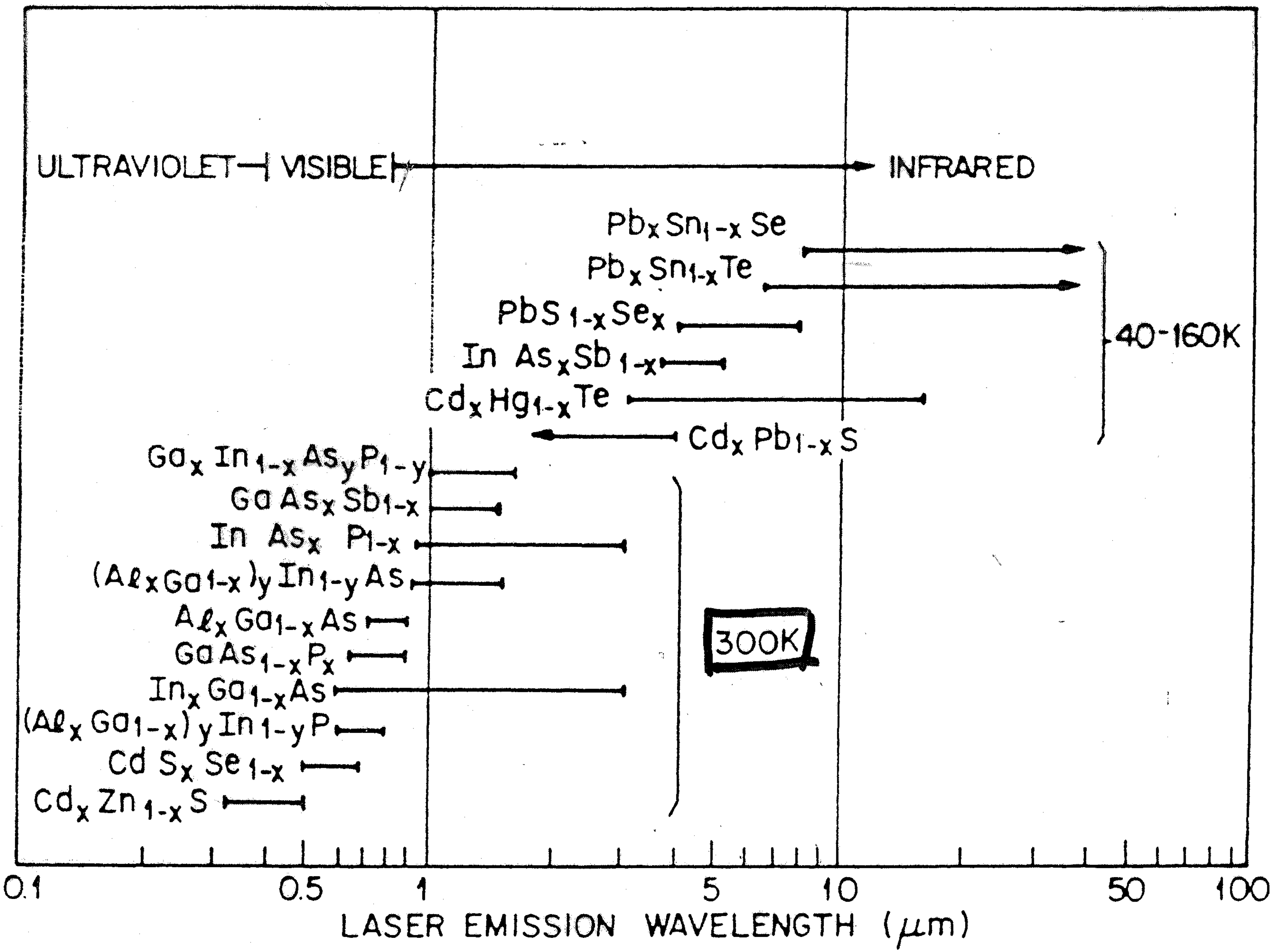


ig. 8 *Schematic diagram of the simplified band structure of (GaAl)As showing the direct interband laser transition.*



Also In Ga P

In Ga AsP



Emission wavelenghts of direct semiconductors

SHOT NOISE $\propto \sqrt{N_{phot}}$ (fluctuations in the photon distrib.)

800 nm, 5mw $\sim 10^{-8}$

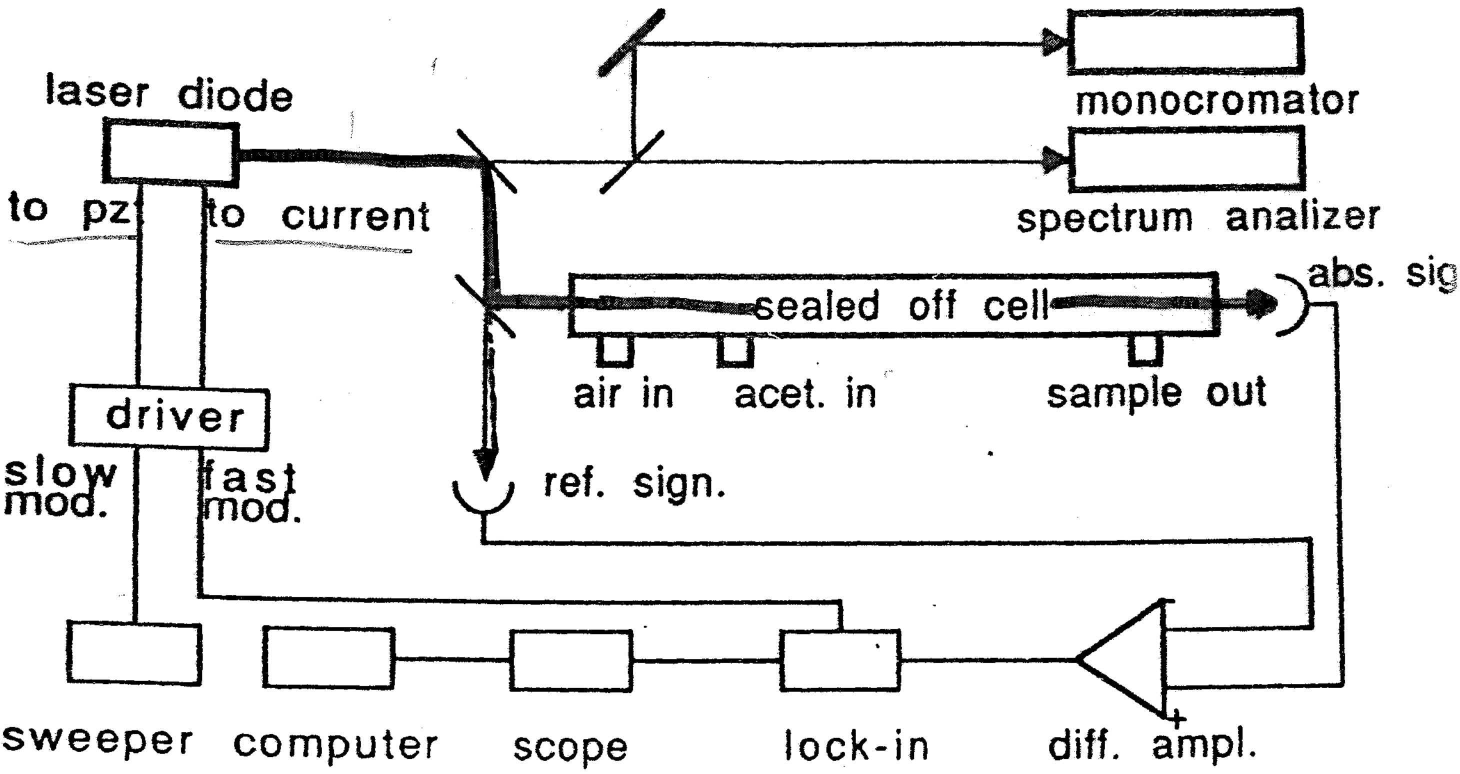
here we are a factor
10 ~ 20 db worse, so
we expect $\boxed{\frac{\Delta I}{I} \sim 10^{-6}}$

↓

HIGH SENSITIVITY IN <u>PURE ABSORPTION</u> EVEN FOR VERY WEAK TRANSITIONS

general ...

EXPERIMENTAL APPARATUS (SIMPLEST) FOR OVERTONE SPECTROSCOPY

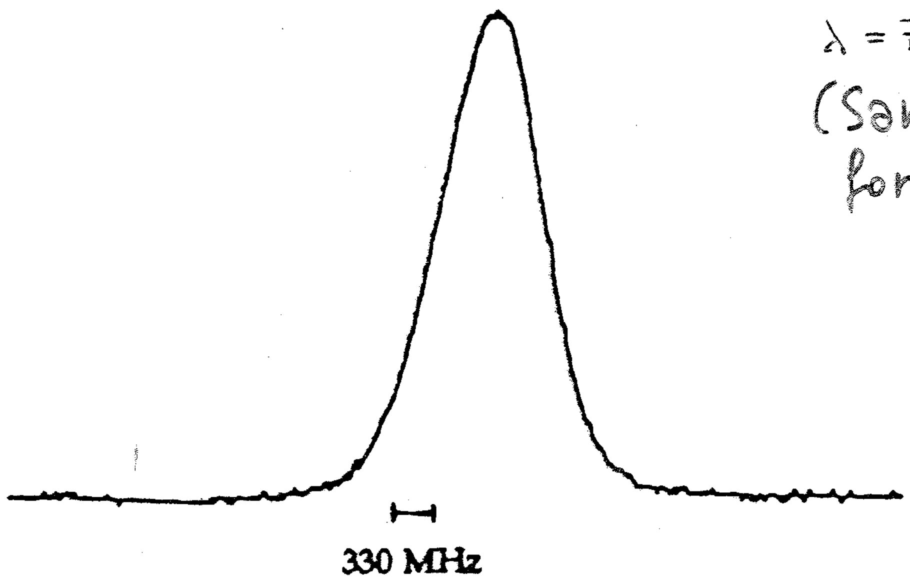


simple source, simple detector,
simple electronics ...

PURE ABSORPTION SIGNAL FOR THE R(9) COMPONENT (C_2H_2)

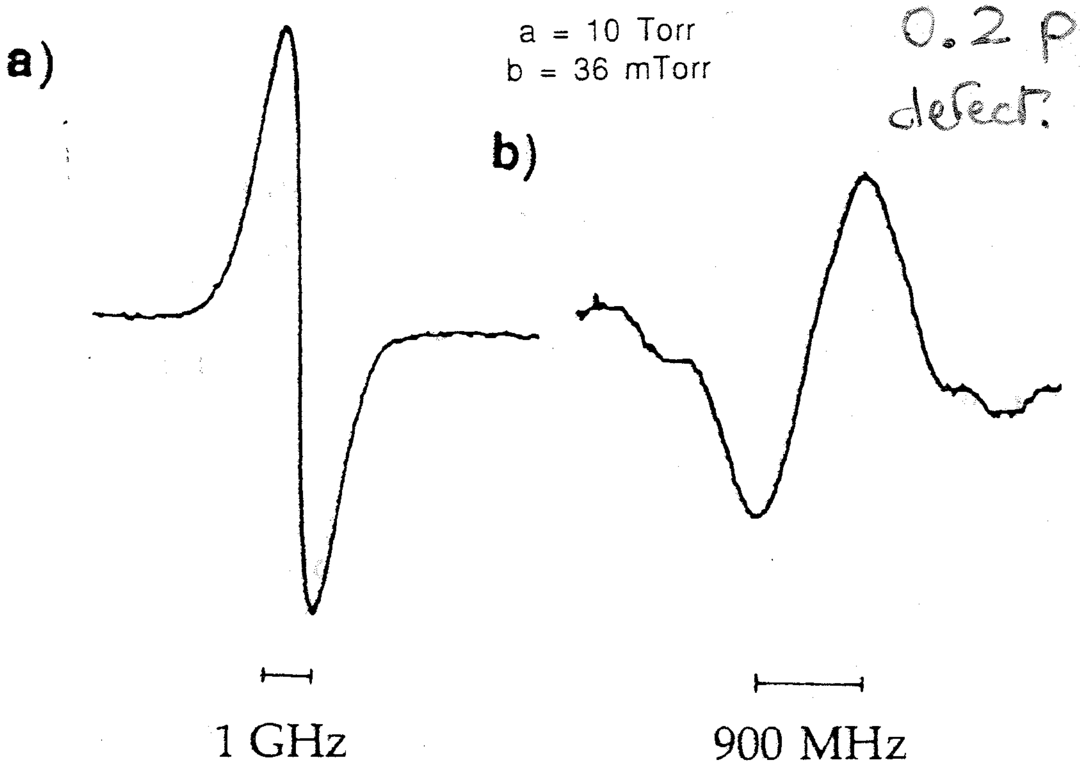
pressure = 10 Torr
pathlength = 1.5 meter
absorption = a few percent

3rd OVERTONE
λ = 788 nm
(Same laser for Rb)



DERIVATIVE SIGNAL

0.2 ppm×m
detect. limit



<u>Limit of detection in absorption : 10^{-6}, 1Hz detect. bandw.</u>

<u>Last limit using FM techniques : up to 10^{-8}</u>

<u>Air pressure detection limit : 0.2 ppm</u>

F.S. PAVONE ET AL., APPL. OPTICS 32, 255 (1993)

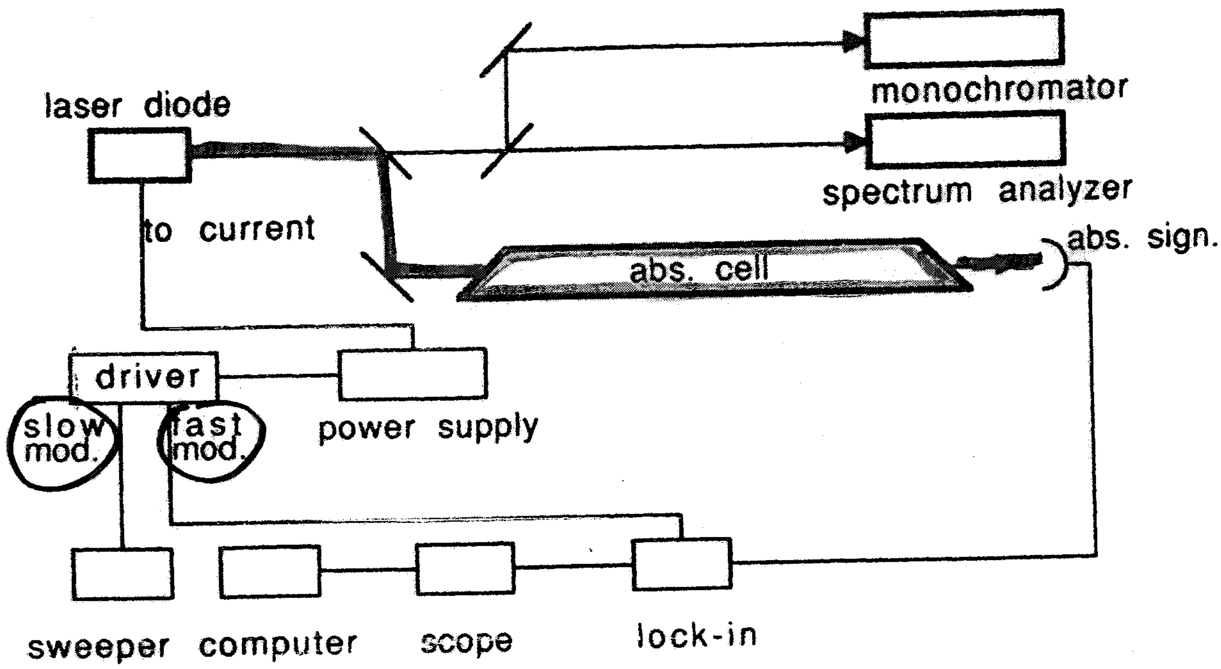
IMPROVING SENSITIVITY...

1st METHOD :

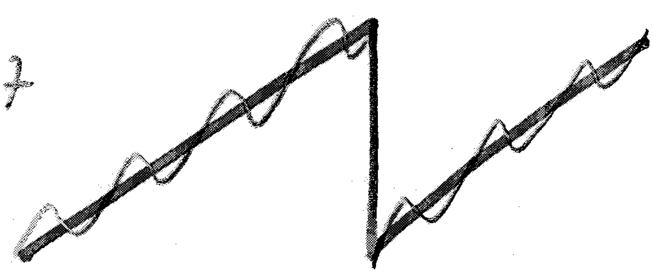
LOW WAVELENGTH MODULATION

MODULATION FREQUENCY 1 KHz

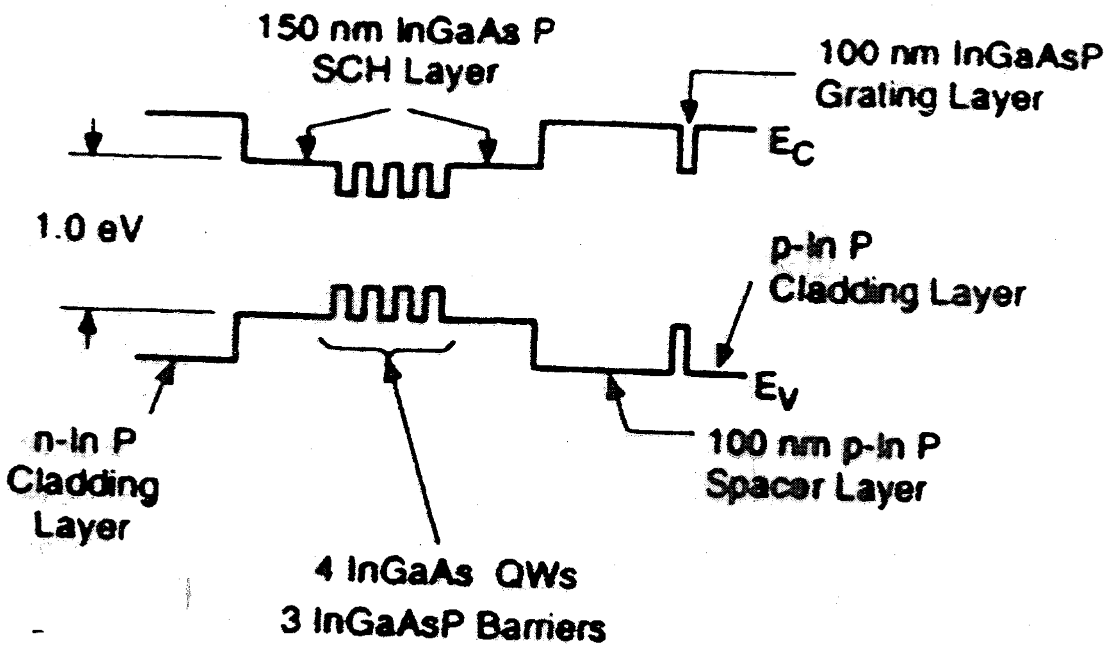
(SMALLER THAN MOLECULAR LINEWIDTH AND THE ABSORPTION IS PROBED BY MANY SIDE BANDS)

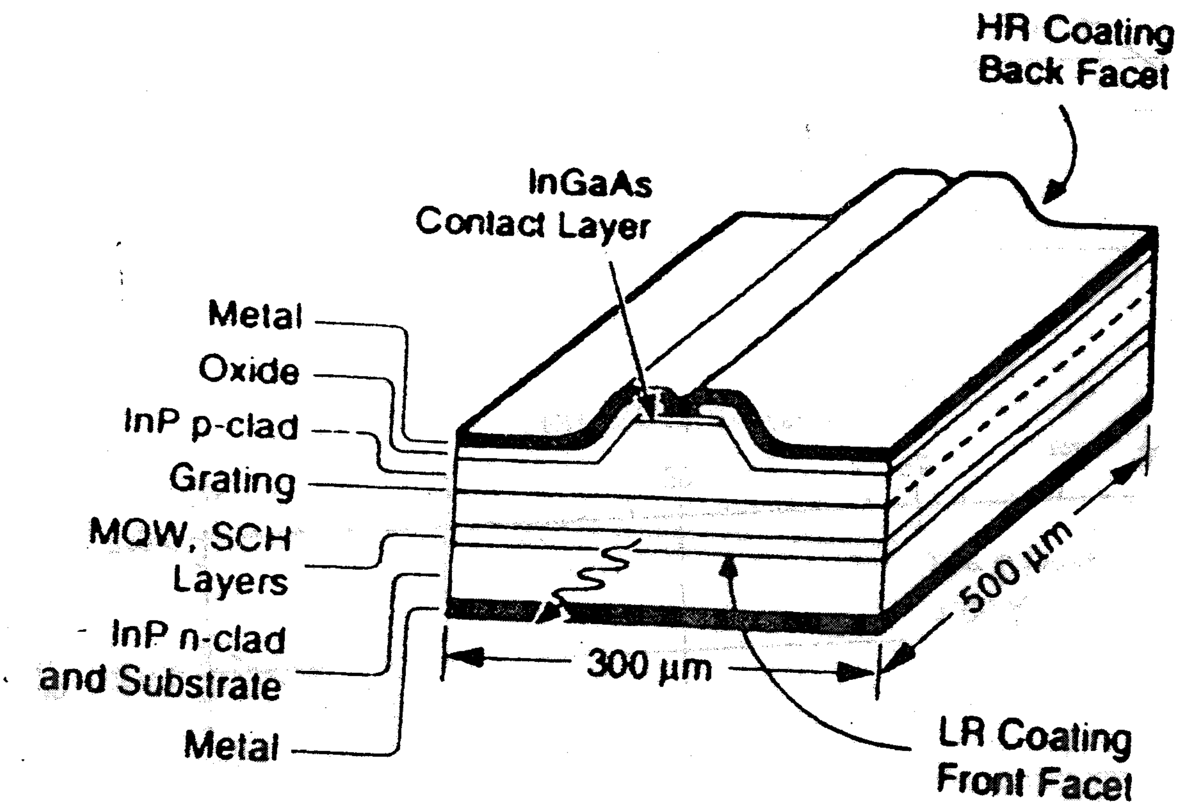


$\alpha_{MIN} = 4.5 \; 10^{-7}$



DFB LASER

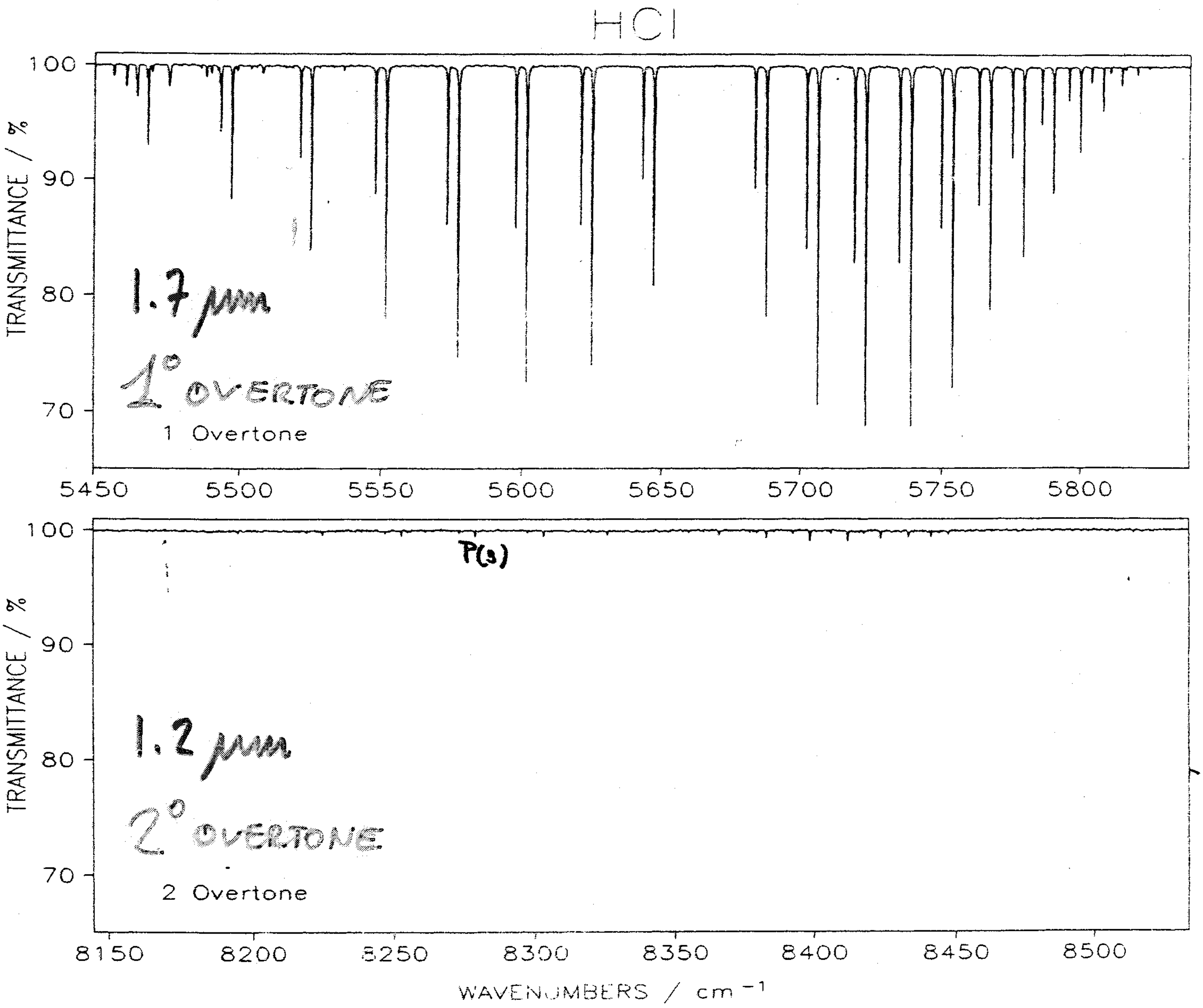




Physical structure and energy band diagram of near-infrared DFB lasers.

REF: D.E. COOPER, R.U. MARTINELLI,
LASER FOCUS WORLD, NOVEMBER 1992

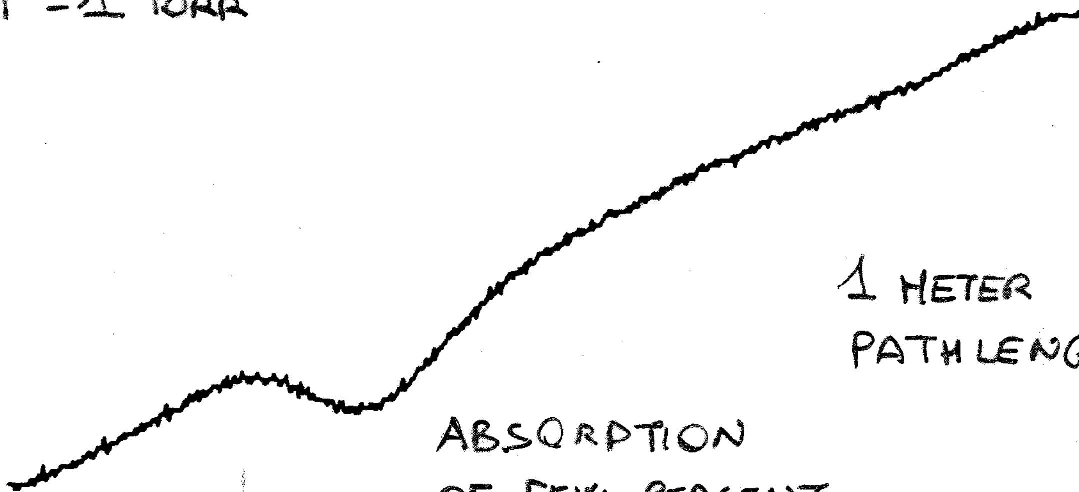
HCl fourier transform spectra



P(3) COMPONENT

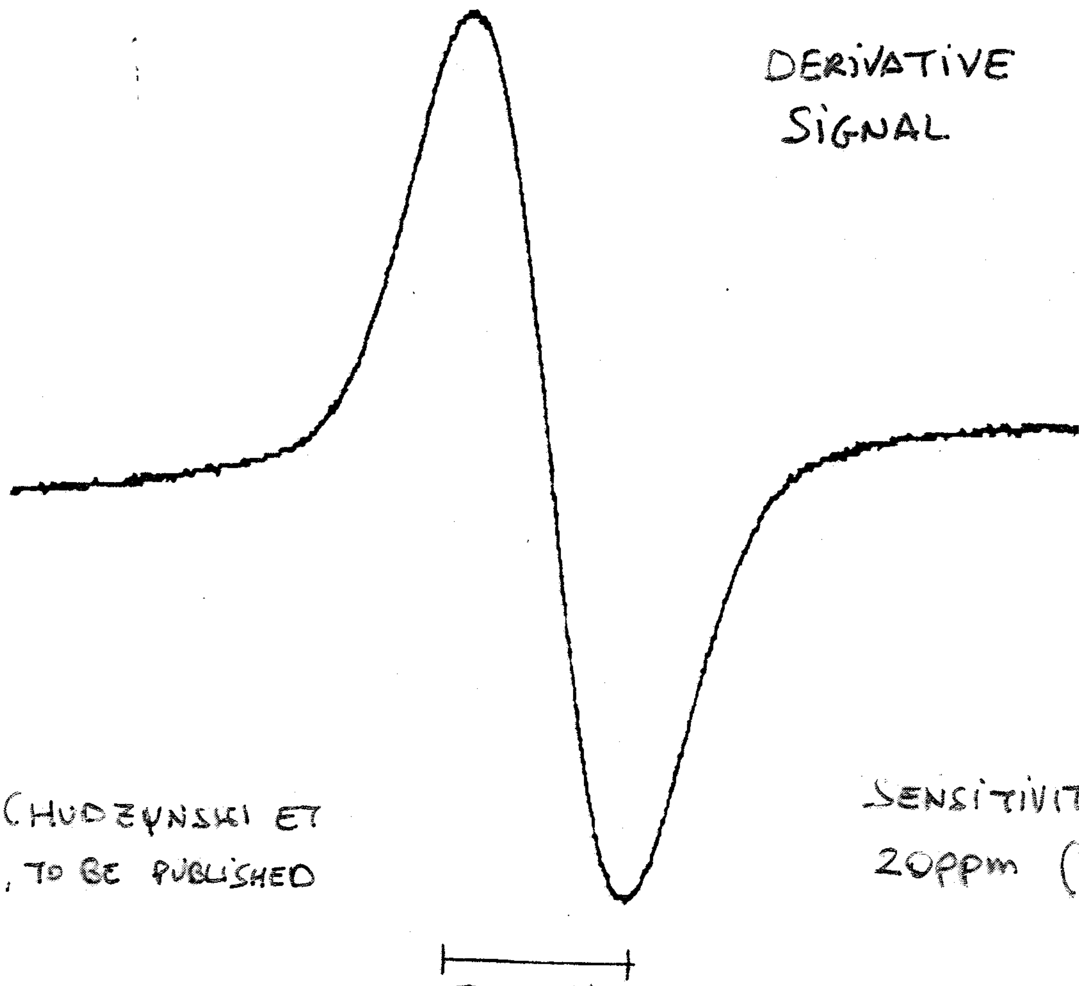
HCl 2^{nd} OVERTONE AT 1.2 µm

P = 1 TORR



1 METER PATHLENGTH

ABSORPTION OF FEW PERCENT



DERIVATIVE SIGNAL

J. CHUDZYNSKI ET AL., TO BE PUBLISHED

SENSITIVITY REACHED: 20ppm (PURE HCl)

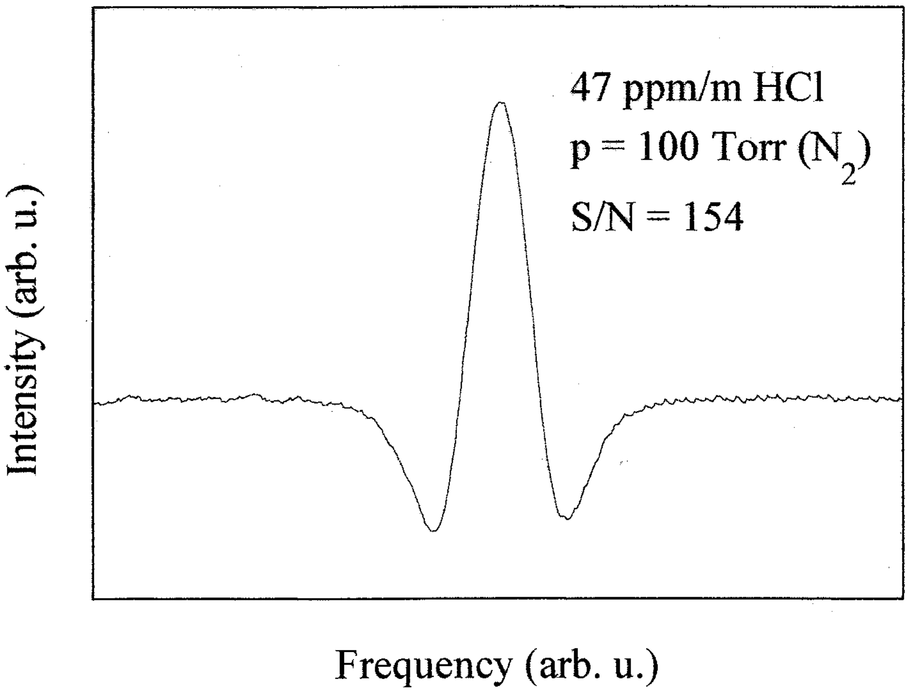
500 MHz

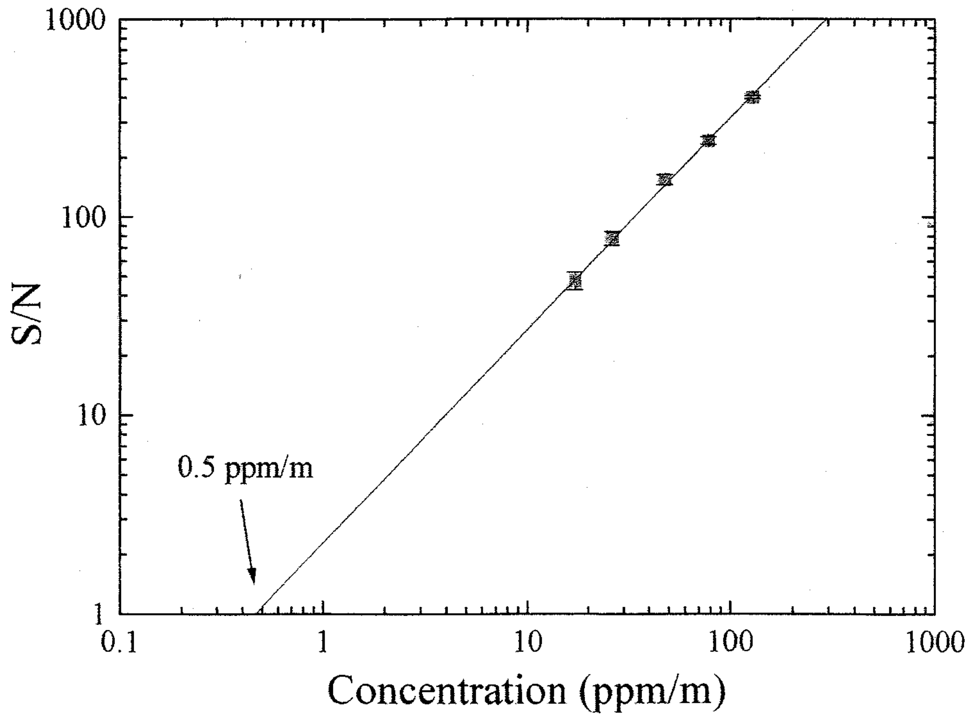
Detection of HCl on the first and second overtones at 1.7 μm and 1.2 μm using semiconductor diode lasers

C. Corsi[1], S. Czhudzynsky[2], F. D'Amato[3], M. De Rosa[4], K. Ernst[2]

LENS-INFM

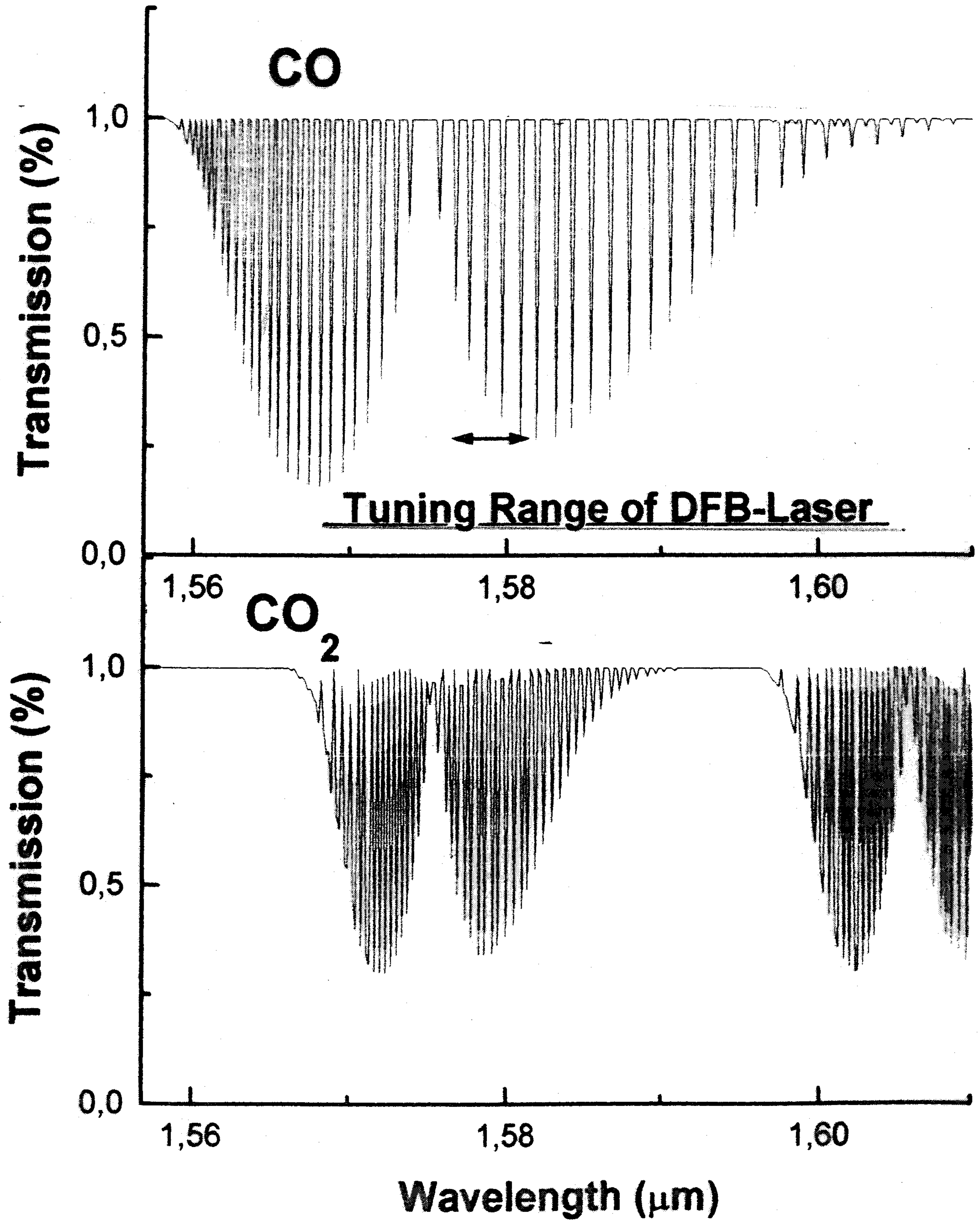
[1] Dip. di Neuroscienze, Univ. Firenze; [2] Dip. di Fisica, Univ. Warsaw;
[3] ENEA, Frascati; [4] SIT, Firenze



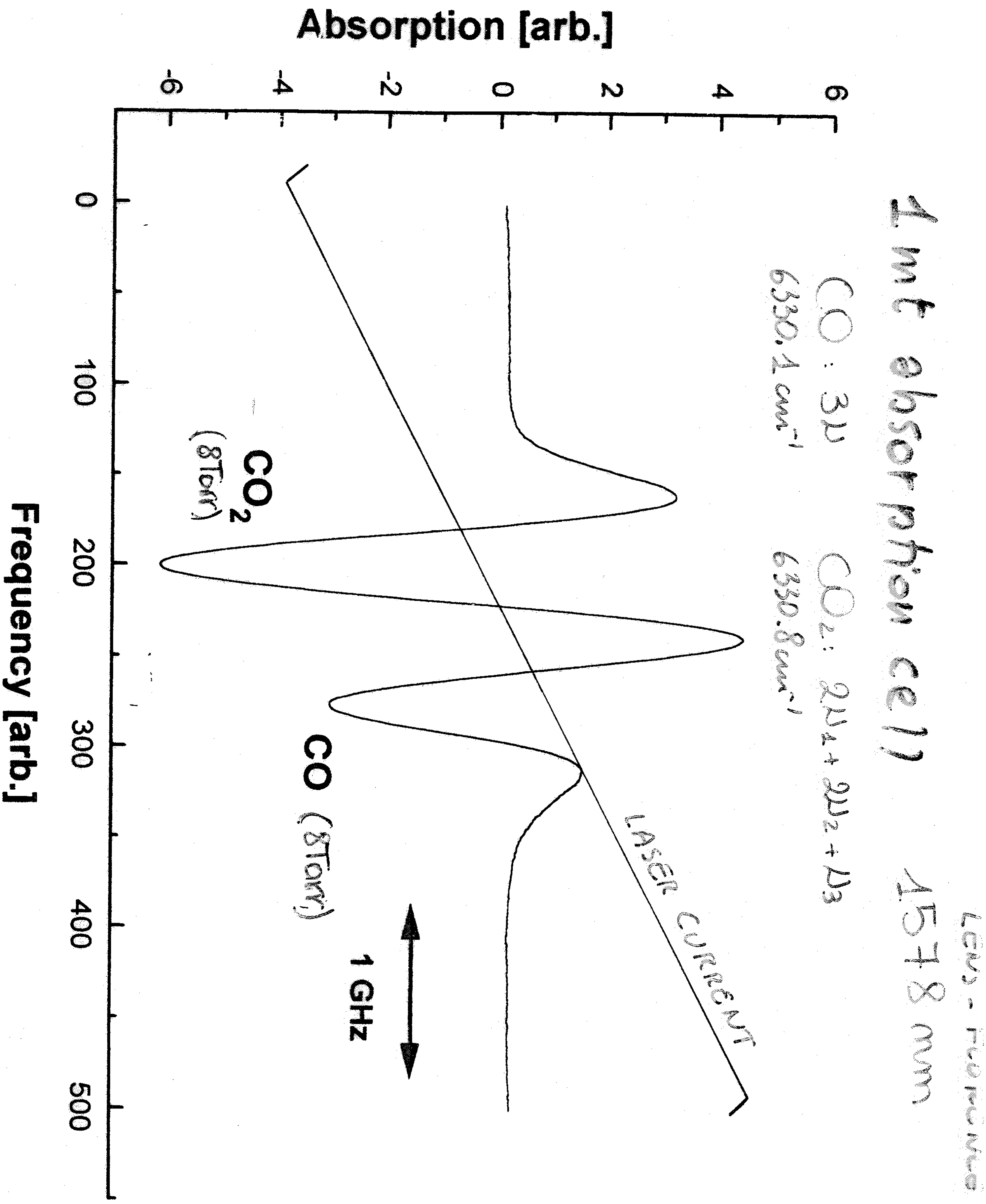


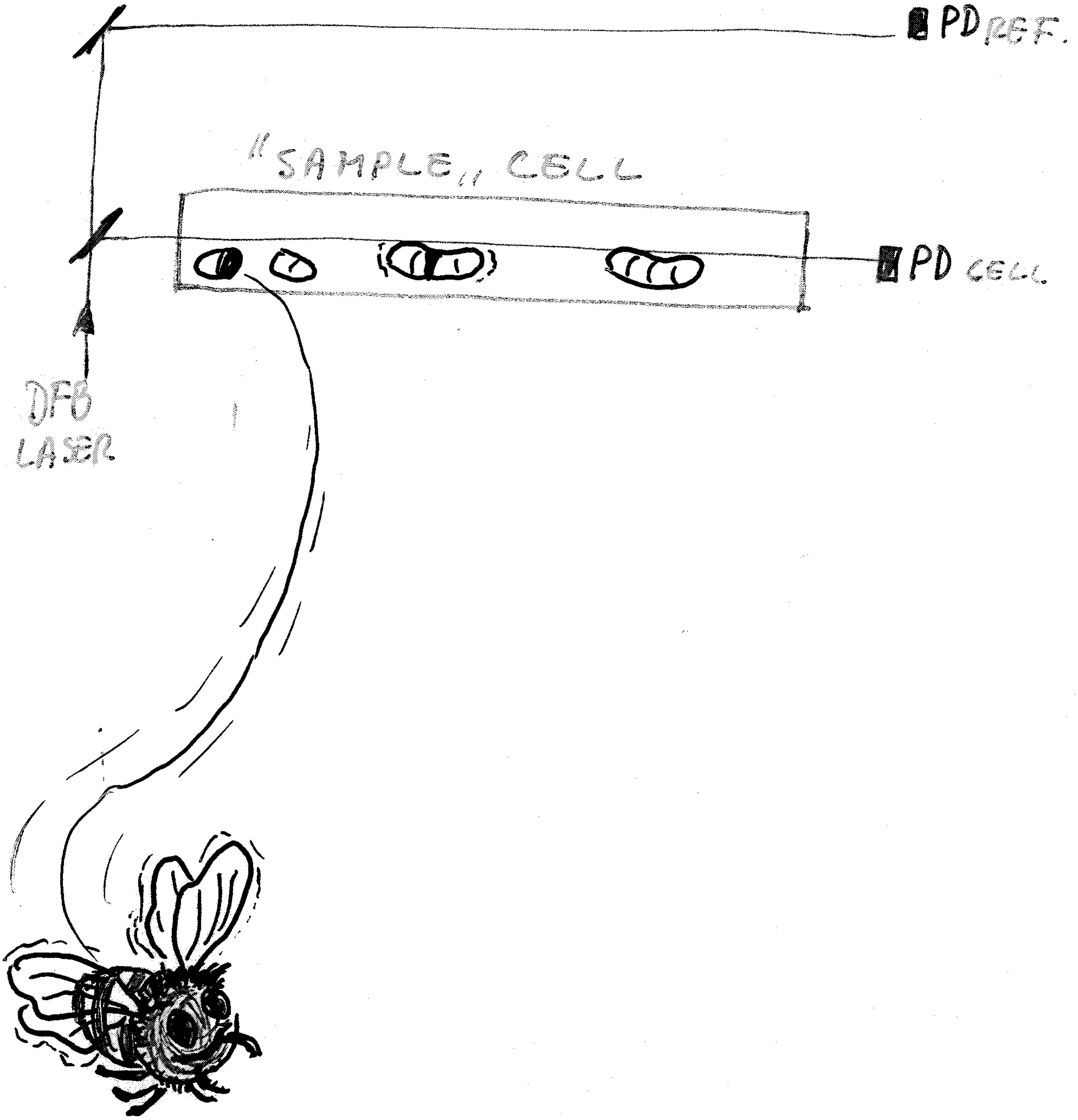
FOURIER TRANSFORM

Overtone Spectra of CO and CO_2 in the 1.5µm region



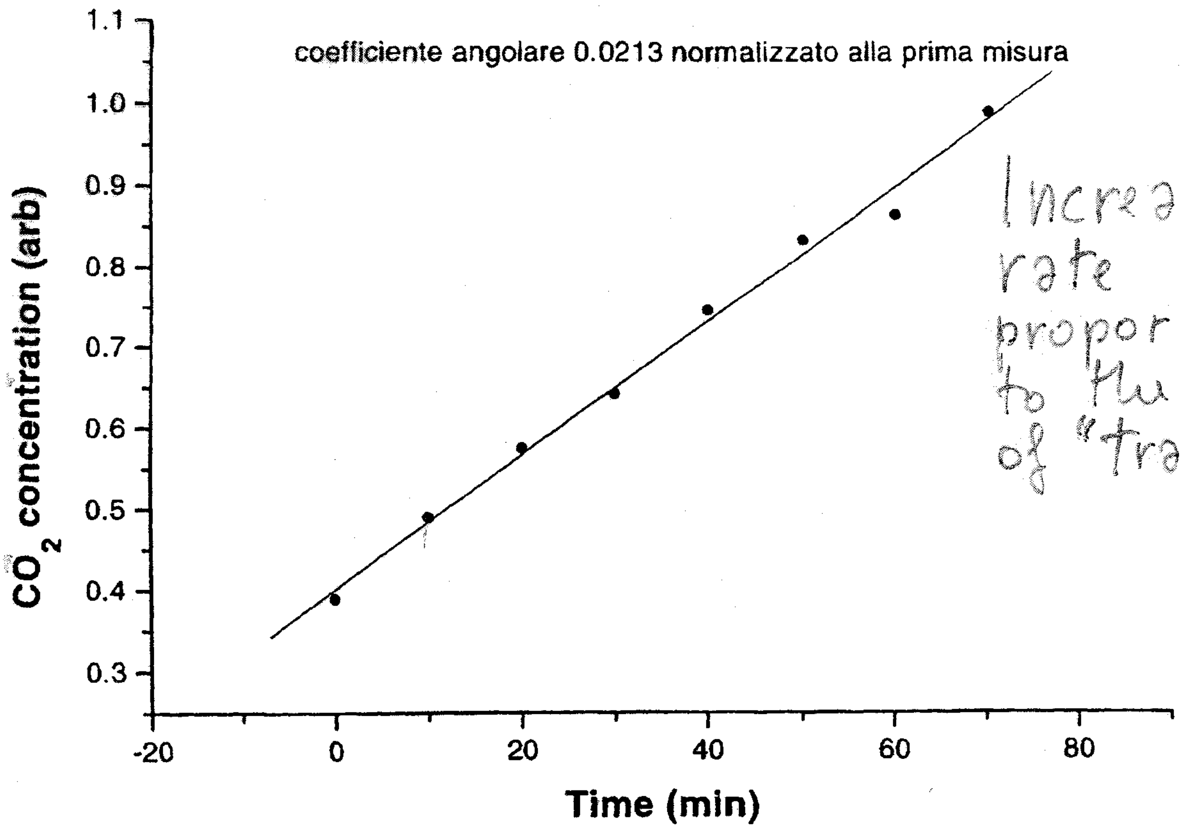
SIMULTANEOUS DETECTION BY ONE SINGLE SCAN





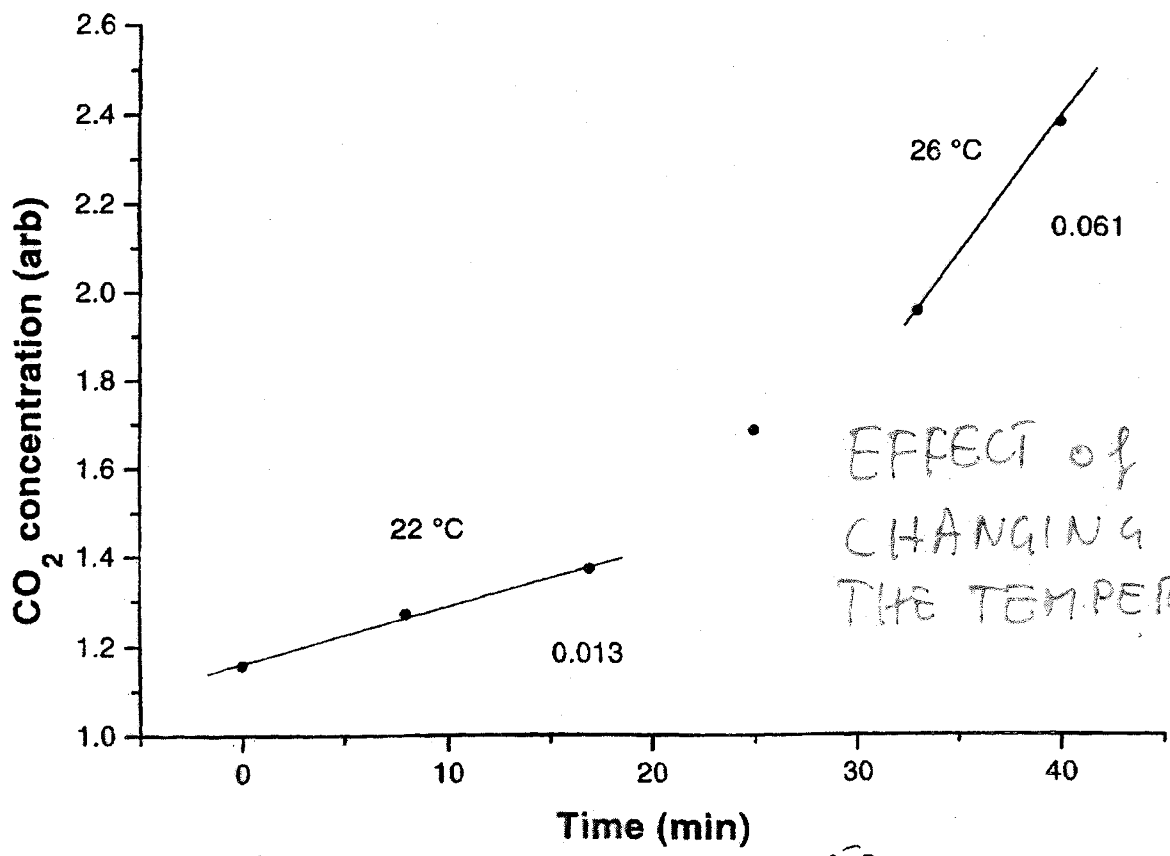
TWH

(4)



Increase rate proportional to the number of "trapped bees"

(5)



EFFECT of CHANGING THE TEMPERATURE

- 52 -

Why using diode lasers for monitoring gases?

- fast response time (ms)

  → DETECTION OF HAZARDOUS GASES

- selectivity

- non-intrusiveness

  → HOSTILE ENVIRONMENTAL CONDITIONS

- sensitivity

  → DETECTION AT PPM LEVEL

- low energy consumption
- low cost
- small-size
- remote sensing

  → FIBER OPTICS

} IDEAL FOR "IN SITU" MEASUREMENTS

Applications

Industrial Applications

-Control of combustion processes

O_2, CO, HCl

-Detection of hazardous gases

CO, H_2S, CH_4

-Monitoring of exhaust gases

CO, CO_2

-Monitoring of emmission gases in power plants

NH_3

Medical Applications

-Non invasive diagnostic of human breath

$^{12}CO_2$, $^{13}CO_2$, NH_3, C_2H_6

Geophysical Applications

-Monitoring of volcanic gases

H_2S, H_2O , CO_2