



INTERNATIONAL ATOMIC ENERGY AGENCY
UNITED NATIONS EDUCATIONAL, SCIENTIFIC AND CULTURAL ORGANIZATION



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COLLEGE ON ATOMIC AND MOLECULAR PHYSICS:
PHOTON ASSISTED COLLISIONS IN ATOMS AND MOLECULES

(30 January - 24 February 1989)

Lecture 1 - "GRAMMAR" OF RADIATION-MATTER INTERACTION

Lecture 2 - TECHNIQUES FOR HIGH RES. (SUBDOPPLER)
SPECTROSCOPY

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MASSIMO INGUSCIO - HIGH RESOLUTION SPECTROSCOPY

LECTURE n. 1

"GRAMMAR" OF RADIATION-MATTER INTERACTION

a) EINSTEIN COEFF. ; LINEAR - NONLINEAR SPECTR.

b) LINESHAPES ; HOMOGENEOUS BROADENING (NATURAL, COLLISIONAL, SATURATION) ; INHOMOGENEOUS BROAD. (DOPPLER EFFECT) VOIGT CONVOLUTION

c) SPECTRAL REGIONS WHERE DOPPLER BROAD. NEGLECTIBLE : EXPERIMENTAL COLLISIONALLY BROADENED AND SHIFTED LINESHAPES.

a) INTRODUCTORY "GRAMMAR"

With spectroscopy we introduce radiation and its interaction with matter.

It is at the basis of modern quantum theory (Understanding of discrete spectra)

Well known from university courses the spectrum of the atom of hydrogen (Fig 1.1) for the interpretation of which it was necessary to assume discrete energy levels :

$$E_n = \frac{2\pi^2 e^4 m}{h^2} \frac{Z^2}{n^2} = R \frac{Z^2}{n^2}$$

$$e \text{ (electron charge)} = 1.60217739(49) \cdot 10^{-19} \text{ Coul.}$$

$$m \text{ (" mass)} = 9.1093897(54) \cdot 10^{-31} \text{ Kg}$$

$$h \text{ (Planck constant)} = 6.6260755(40) \cdot 10^{-34} \text{ J s}$$

$$R \text{ (Rydberg constant)} = 10973731.534 \text{ m}^{-1}$$

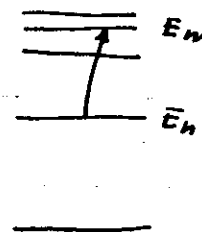
Line frequencies :

$$\nu_{n,m} = \frac{E_m - E_n}{h}$$

$n=1$ Lyman UV

$n=2$ Balmer visible

$n=3$ Paschen infrared



RYDBERG BOHR

$$\frac{1}{\lambda} = R \left[\frac{1}{m^2} - \frac{1}{n^2} \right]$$

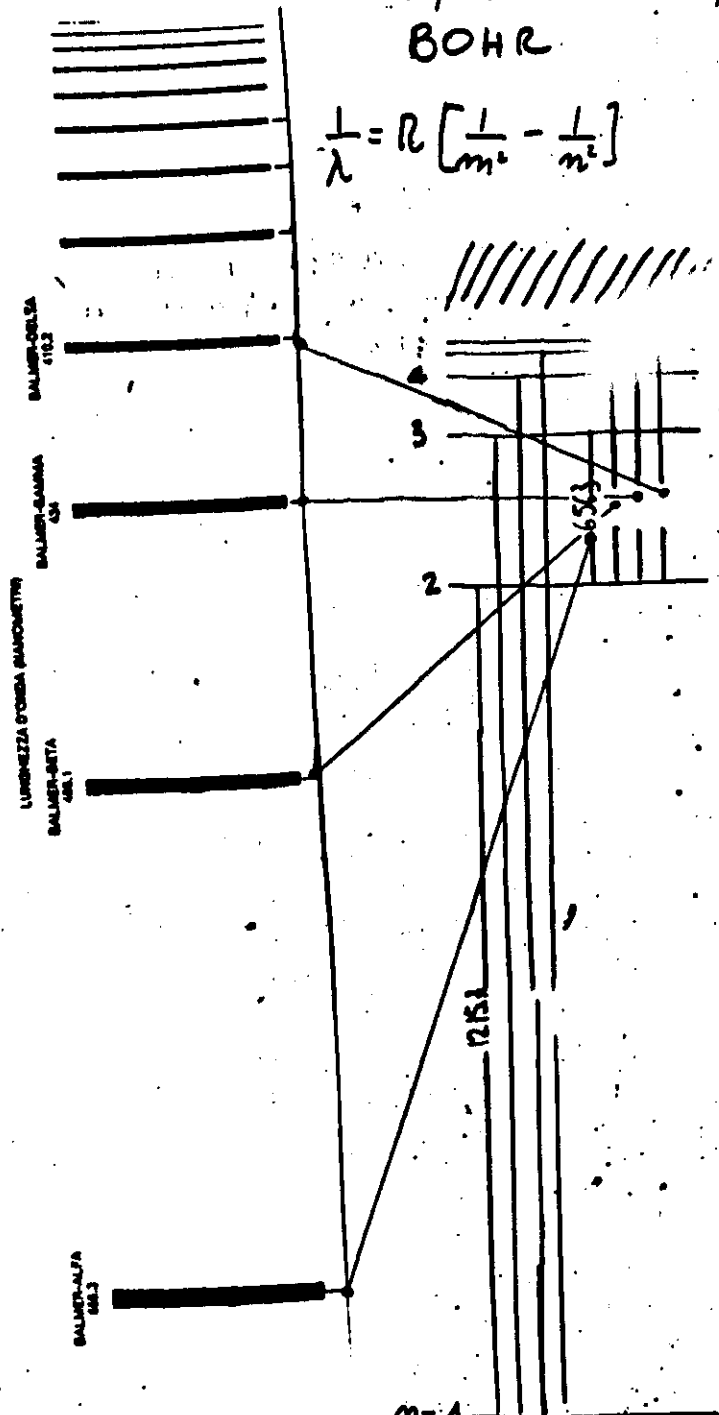
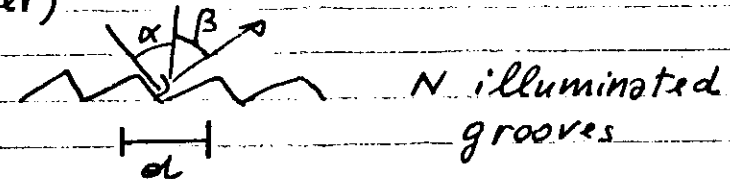


Fig. 1.1

Emission spectrum observed from a discharge by means of a grating spectrometer (The collimated parallel light is sent to a reflection grating consisting of many straight grooves "ruled" onto an optically smooth substrate - plus highly reflecting layer)



Constructive interference for

$$d (\sin \alpha \pm \sin \beta) = m \lambda$$

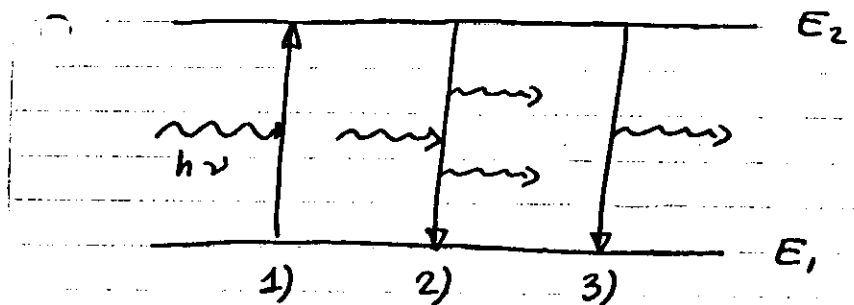
(dispersion at different m orders) $\Rightarrow$

$$\frac{\lambda}{\Delta \lambda} = \frac{\nu}{\Delta \nu} = \frac{N d (\sin \alpha \pm \sin \beta)}{\lambda} = \text{Res. Power} = m N$$

Typical resolving power $10^4 \sim 10^5 \Rightarrow$

in the visible $\Delta \lambda \approx 10^{-1} \text{ \AA}$; $\Delta \nu \approx 10 \text{ GHz}$

Interaction radiation-matter not only described by spontaneous emission but, more completely, by: 1) Absorption 2) Induced emission 3) Spontaneous emission.



3

The probabilities per second of the three events:

1) $\frac{d}{dt} P_{12} = B_{12} \rho(\nu)$ radiation en. density $\rho(\nu)$

2) $\frac{d}{dt} P_{21} = B_{21} \rho(\nu)$ "

3) $\frac{d}{dt} P_{21}^{\text{spont}} = A_{21}$ no radiation

$\Downarrow$

$$B_{12} = \frac{g_2}{g_1} B_{21}; \quad A_{21} = \frac{8\pi h \nu^3}{c^3} B_{21}$$

dipole approximation : $A \propto \nu^3 d^2$

el. dip. $(d_e)^2 \approx (e a_0)^2$

magn. dip. $(d_m)^2 \approx (\mu_0)^2$

$$(d_e)^2 \sim 10^5 (d_m)^2$$

- In the visible, for an allowed el. dip. transition
 $A \sim 10^8 \text{ sec}^{-1} \rightarrow \tau = \frac{1}{A} \sim 10^{-8} \text{ sec.}$

4

Dependence on $\nu^3 \rightarrow$ much longer lifetimes in the microwaves or radio frequency

For instance H ground state

$F=1$ magnetic dipole, gigahertz region
 21 cm
 $F=0$ $\tau = \frac{1}{A} \sim 10^7 \text{ years}$

(Emission observable only from very low density interstellar H clouds).

Stimulated transitions earlier in the microwaves; in the visible only after the advent of lasers (enough $\rho(\nu)$)

LINEAR AND NONLINEAR ABSORPTION

Optically thin layer the infinitesimal absorption dI/dz is proportional to the incident intensity I

$$\frac{dI}{dz} = -\alpha I$$

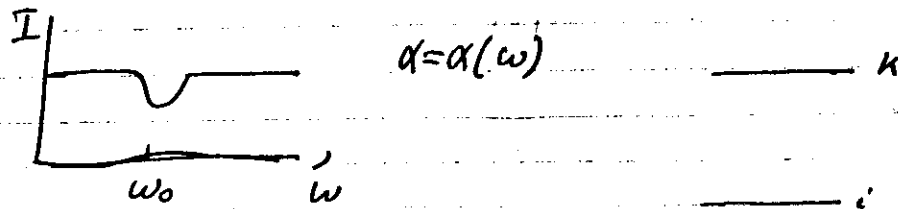
$\Downarrow$

$$I(z) = I_0 e^{-\alpha z}$$

α absorption coefficient (proportional to the imaginary part of the complex

index of refraction)

5



Total absorption $\int \alpha_{ik}(\omega) d\omega = \left(\frac{\hbar \omega_{ik}}{c} \right) (N_i - N_k \frac{g_i}{g_k}) B_{ik}$

definition of absorption cross section σ_{ik}

$$\alpha_{ik} = \sigma_{ik} (N_i - N_k \frac{g_i}{g_k});$$

$$\int \sigma_{ik} d\nu = \left(\frac{h \nu_{ik}}{c} \right) B_{ik} = \left(\lambda^2 / 8\pi \right) A_{ik}$$

We have

$$I(z) = I_0 e^{-\alpha z} = I_0 e^{-\sigma_{ik} [N_i - (\frac{g_i}{g_k}) N_k] z}$$

LINEAR ABSORPTION (As long as the population densities N_i and N_k of the two levels E_i and E_k are not noticeably altered by the interaction with the radiation field (weak signal approximation) one can regard them as constant.

At larger intensities I , N_i decreases, N_k increases
i.e. $N_i = N_i(I)$; $N_k = N_k(I)$

dI is no longer proportional to I
(NONLINEAR ABSORPTION)

Simple two non degenerate levels

6

$$\begin{array}{l} \xrightarrow{E_2} \\ \xrightarrow{E_1} \end{array} \quad g_i = g_1 = 1 \quad N = N_1 + N_2 = \text{constant} \\ B_{12} = B_{21} = B \quad \Delta N = N_1 - N_2$$

We also allow additional collisional induced transitions (non radiative) wff: C_{12} and C_{21}

Evolution of populations

$$\frac{dN_2}{dt} = B \rho(\omega) \underbrace{[N_2 - N_1]}_{R_2} + \underbrace{(A_{21} + C_{21}) N_2}_{R_1} - \underbrace{C_{12} N_1}_{R_1}$$

under stationary conditions

$$\frac{dN_1}{dt} = - \frac{dN_2}{dt} = 0 \quad \text{hence:}$$

$$\Delta N = \frac{\Delta N_0}{1 + 2 B \rho(\omega) / (R_1 + R_2)} = \frac{\Delta N_0}{1 + S}$$

where $\Delta N_0 = N_0 (R_2 - R_1) / (R_2 + R_1)$ is the population difference for $\rho = 0$

The saturation parameter

$$S = \frac{2 B \rho(\omega)}{R_1 + R_2} = \frac{B \rho(\omega)}{\bar{R}}$$

- represents the ratio of the induced transition probability to the mean relaxation probability
 $\bar{R} = (R_1 + R_2)/2$

If the only relaxation process is spontaneous emission ($R_1 = 0, R_2 = A$), then the saturation parameter S yields the ratio of induced to spontaneous transition rates

- $S=1$ means ΔN drops to $1/2$ of its unsaturated value ΔN_0

With $I(\omega) = c \cdot \rho(\omega)$

$$\Delta N = \frac{\Delta N_0}{1 + \frac{B I(\omega)}{c \cdot \bar{R}}} = \frac{\Delta N_0}{1 + I/I_s}$$

Saturation intensity $I_s = c \cdot \frac{\bar{R}}{B}$ stands

for that incident intensity which decreases

ΔN to $\Delta N_0/2$

Absorption

$$\alpha = \frac{\alpha_0}{1+S} = \frac{\alpha_0}{1+I/I_s}$$

- i.e. in a pure two-level system $\alpha \rightarrow 0$ for $I \rightarrow \infty$ - Easier to saturate transitions

- with high B and low R values. 8

Note that this is just a very elementary introduction, for instance the system should be generalized by allowing additional relaxation paths.

b) LINESHAPES

$$\begin{array}{c} \text{-----} E_2 \\ \Delta E = E_2 - E_1 = h\nu_0 \\ \text{-----} E_1 \end{array}$$

But the energy of the states is not exactly defined (because of external perturbations or of indeterminism principle...), hence absorbed or emitted radiation is distributed around ν_0

$I = \tilde{I}(\nu)$ lineshape.

NATURAL BROADENING

Spontaneous decay of excited levels. Coefficient A is independent from time, hence we have an exponential decay of the emitted intensity with a time constant

$$\tau = \frac{1}{A}$$

- The frequency distribution can be computed by means of the Fourier transform of an exponential

decaying wave

$$I(\nu) = \frac{\Gamma/2\pi}{(\nu - \nu_0)^2 + (\frac{\Gamma}{2})^2} \quad \text{LORENTZIAN}$$

with a width $\Gamma = \Gamma_{\text{NAT}} = 1/2\pi\tau = \frac{A}{2\pi} = 2\gamma$

FROM INDETERMINATION PRINCIPLE $\Delta E \cdot \Delta t = h/2\pi$
 $\Delta\nu = \frac{\Delta E}{h} = 1/2\pi\tau$

(All this for an excited level decaying only and directly to the ground state)

In general $2\pi\Gamma = \frac{1}{\tau_1} + \frac{1}{\tau_2}$

LORENTZIAN properties:
 very smooth decaying to zero $\propto (\nu - \nu_0)^{-2}$,
 not negligible absorption even at large
 distances from the line center.

Known the lineshape it is possible to compute
 the cross section of absorption, and
 in particular its value at center

$$\sigma(\nu_0) = \frac{\lambda^2}{4\pi^2} g_2 \frac{A}{\Gamma} \sim \frac{\lambda^2}{2\pi} g_2$$

i.e. absorption or emission at the center of
 a naturally broadened line is independent
 of τ (hence from the dipole moment):

9

stronger lines are only larger (note that
 this is rigorously true only for resonance
 transitions for which $\tau_1^{-1} = 0$ and
 $\tau_2^{-1} = A$).

10

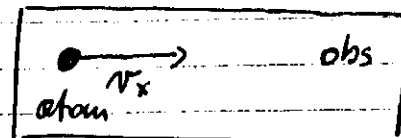
NATURAL ONE is an example of homogeneous
broadening (broadening identical for each
 atom of the system and the interaction
 with a monochromatic radiation beam is
 the same for all the atoms).

Inhomogeneous lineshape = typical: Doppler
 effect: each atom has its own lineshape
 which however is frequency shifted
 by Doppler effect.

DOPPLER BROADENING

Gas at room temperature $\frac{v}{c} \sim 10^{-6}$ i.e.

we can expand in series the relativistic
 formula and take the first two terms



$$\nu = \nu_0 \left[1 + \frac{v_x}{c} - \frac{v_x^2}{2c^2} \right] \quad (1)$$

$\sim 10^{-12}$ negligible
 with respect to the
 natural $\frac{\nu}{\Delta\nu}$ ($10^9 \sim 10^8$)

Substituting v_x from (1) into
 the Maxwell velocity distribution, we

obtain

$$I(\nu) = \frac{2}{\Delta \sqrt{\pi}} \exp \left[-4 \frac{(\nu - \nu_0)^2}{\Delta^2} \right]$$

When Δ is the width of the Doppler line measured at $\frac{1}{2}$ the maximum.

Width at half maximum (FWHM = Full Width Half Maximum)

$$\begin{aligned} \text{FWHM} = \Delta \nu_D &= \Delta \sqrt{\ln 2} = \frac{2\nu_0}{c} \sqrt{2 \ln 2 \frac{kT}{M}} = \\ &= \boxed{7.16 \cdot 10^{-7} \nu_0 \sqrt{\frac{T}{M}}} \end{aligned}$$

T in Kelvin, M atomic or molecular mass.

Note that $\Delta \nu / \nu_0$ is independent from the transition frequency and at room temperature is $\sim 2 \cdot 10^{-6}$

Comparison with natural width in the visible:

$$\Delta \nu_D \approx 10^9 \text{ Hz} \approx 10^2 \Delta \nu_{\text{NAT}}$$

(MASKING EFFECT).

VOIGT PROFILE

12

Real situation: natural and Doppler both present as independent processes, hence the lineshape must be computed by a convolution; Voigt integral

$$I(\nu - \nu_0; \frac{\Gamma}{\Delta}) = \frac{\Gamma}{4\pi\sqrt{\pi}} \int_0^\infty \frac{\exp[-4(u - \nu_0)^2/\Delta^2]}{(\nu - u)^2 + \frac{\Gamma^2}{4}} du$$

— only numerical solution.

Comparison Lorentzian - Gaussian in FIG. 2

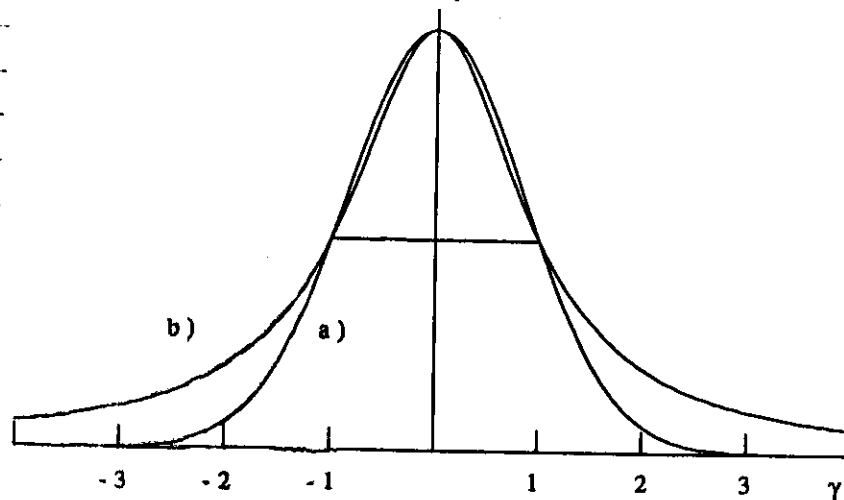


FIG. 2

a) Gaussian; b) Lorentzian same width and height.

Far from the center Lorentzian is predominant; near the center Gaussian is predominant; intermediate region Voigt profile is necessary.

COLLISIONAL (PRESSURE) BROADENING

Time of collision practically instantaneous with respect to radiative time.

$$\tau_c \sim \left(\frac{\sigma}{v}\right)^{1/2} / N$$

N relative velocity; σ cross section $\leq 10^{-13} \text{ cm}^2$

At room temperature $N \sim 10^5 \text{ cm/sec}$
and $\tau_c \sim 10^{-12} \text{ sec}$.

Atom then unperturbed until next collision.

Frequency of collisions: $f_c = \bar{v}_r N$

N perturber density; $\bar{v}_r$ relative mean velocity.

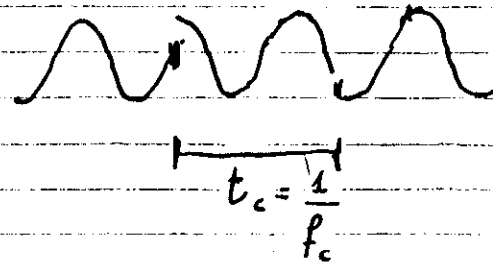
$$t_c = \frac{1}{f_c} \quad \text{time interval between two consecutive collisions.}$$

For $N \leq 10^{20} / \text{cm}^3$ $t_c > \tau_c$ hence collisions are well separated in time; we can neglect the contribution of multiple simultaneous collisions and all the perturbation effects are proportional (in intensity) to the density of the perturber.

(Under this approximation phenomena are described by impact theories)

(Treatment of a single collision and then average over velocity distribution

and impact parameters.



Long distance interaction between two atoms or molecules described by means of a potential (attractive):

$$\Delta V_n(r) = C_n^k / r^n$$

constants C_n^k depends on the k state of the perturbed atom and on the perturber while n depends on the type of interaction:

$n=2$ linear Stark effect and charged perturbers (electrons and ions in a plasma).

$n=3$ resonant dipole interaction.

$n=4$ quadratic Stark effect.

$n=6$ dipole — induced dipole.

Short distance (atomic radius) repulsive forces (we neglect)

- k During a collision the instantaneous transition frequency between two states k and i , as a function of z , is given by:

$$h\nu_{ki}(z) = E_k - E_i + \Delta V_k(z) - \Delta V_i(z)$$

i.e.

change of phase in the emitted radiation (can be computed if you know the trajectory of the collision).

Roughly: Lineshape given by the Fourier transform of an exponentially decaying wave, losing phase coherence with a frequency Γ_c .

Again Lorentzian

$$\Delta\nu_c = \Gamma_N + \frac{\bar{\sigma} \bar{v} N}{\pi} = \Gamma_N + \Gamma_c$$

More sophisticated theories evaluate the effect of collisions with high impact parameter, producing only small phase displacements, which summing produce a shift in the center frequency.

D Real part of cross section $\sigma_r \rightarrow$ line broadening
Imaginary part (σ_i) $\rightarrow$ " shift.

Collisional lineshape, again a Lorentzian

$$\Gamma_N + \Gamma_c$$

$$\frac{(\nu - \nu_0 \pm \Delta\nu_s)^2 + (\Gamma_N + \Gamma_c)^2}{}$$

$$\uparrow$$

$$\Delta\nu_s = \sigma_i \bar{v} N / 2\pi$$

for neutral perturbors always $\sigma_i < \sigma_r$

(Pressure shift smaller than pressure broad)

IF WE MOVE FROM VIS. TO

LOWER FREQ, WE HAVE DECREASE

OF NATURAL WIDTH, DECREASE

OF DOPPLER WIDTH, HENCE

WE CAN OBSERVE COLLISIONAL

LINESHAPES EVEN AT LOW PRESSURES.

WE GIVE EXAMPLES IN THE

FAR INFRARED ($\sim 10^{12}$ Hz)

FOR PRESSURE BROADENING AND

SHIFT OF POLAR MOLECULES

(DIPOLE-DIPOLE INTERACTION

DURING THE COLLISION).

FAR INFRARED: $30 \mu\text{m} \div 1 \text{ mm}$

ELECTROMAGNETIC SPECTRUM

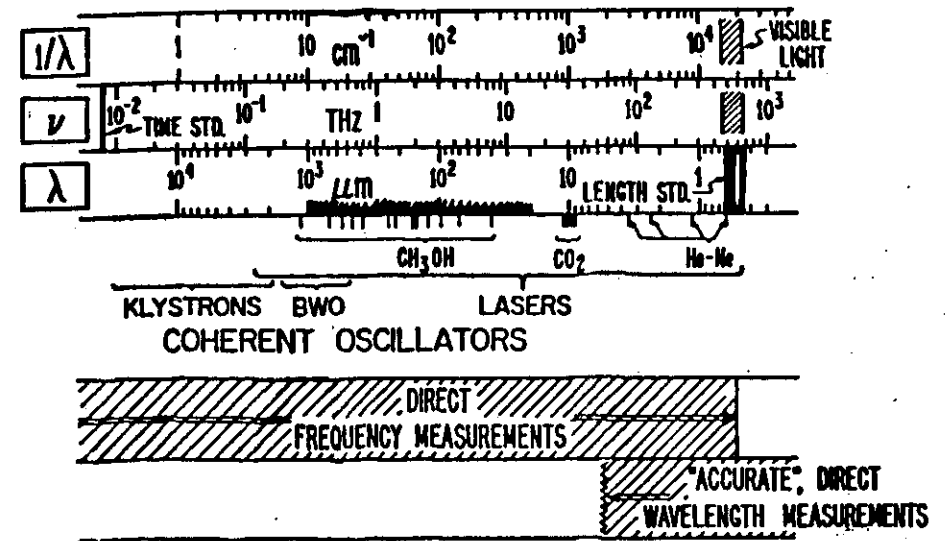


Figure 1. The electromagnetic spectrum showing oscillators, frequency and wavelength standards, and measurement regions. The extension of frequency measurements into the infrared has produced an overlap of the accurate ($< 4 \times 10^{-9}$) wavelength measurement region and the frequency measurement region for the first time. The frequency standard is the zero field hyperfine structure separation in the ground state of ^{133}Cs which is defined to be 9 192 631 770 Hz. The wavelength standard is the transition between the $2p_{10}$ and $5s_3$ levels of ^{86}Kr with the vacuum wavelength of this radiation defined to be $1/1\,650\,763.73 \text{ m}$.

3rd ORDER GENERATION

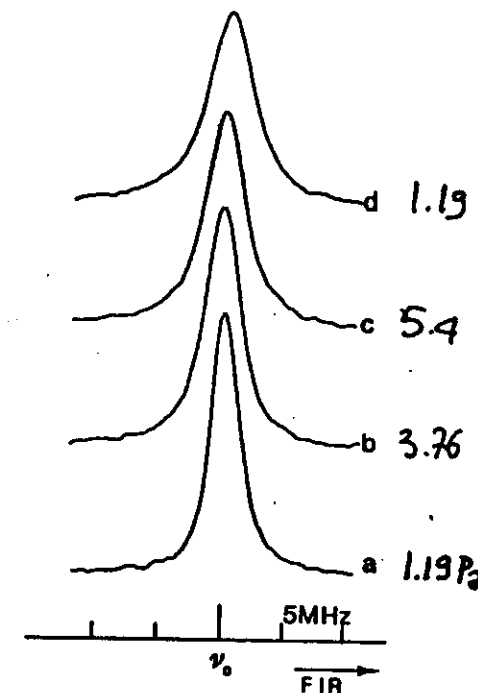
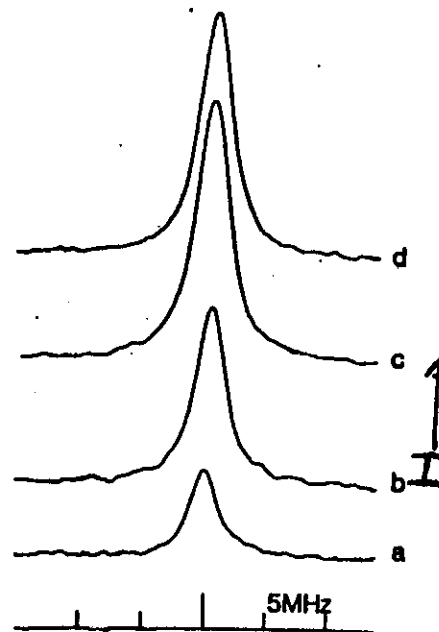
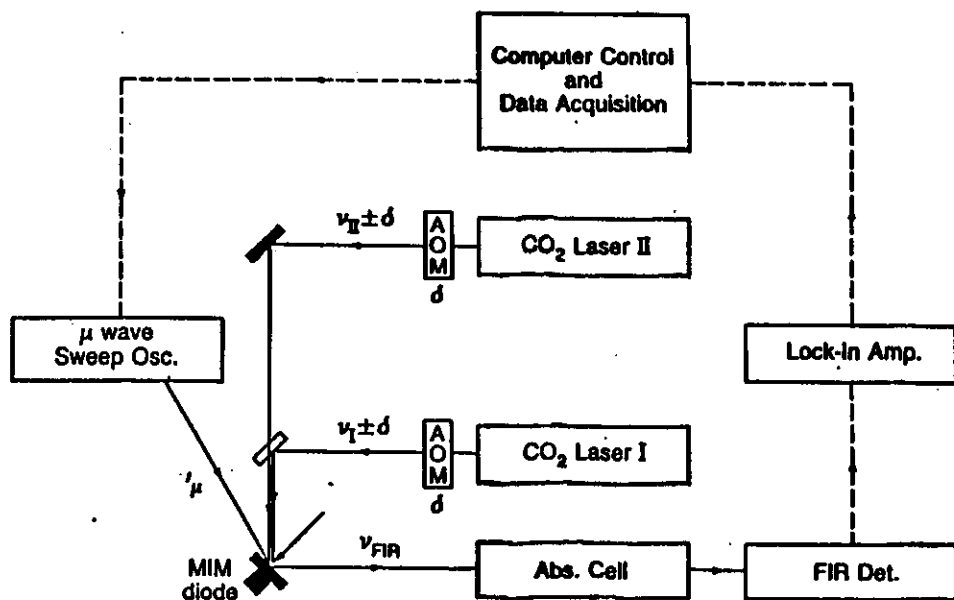
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DOPPLER FREE SIGNAL

20

PUMP INTENSITY

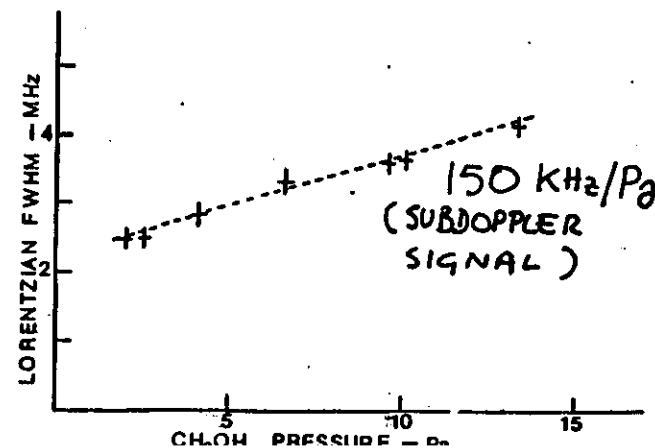
PRESSURE BROADENING



μ WAVE - ± 20 GHz

SPECTRAL COVERAGE
COMPLETE BY MEANS
OF ONLY 150 PAIRS

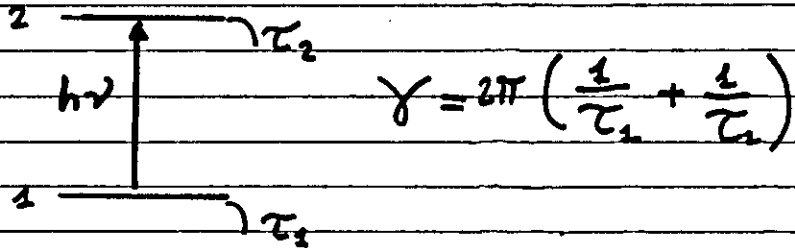
(DOPPLER
BROADENED:
226)



21

d) 8 CH₃OH GROUND STATE^③

SATURATION BROADENING



$$\Delta \nu_{\text{hom}} = \gamma \sqrt{1 + I/I_s}$$

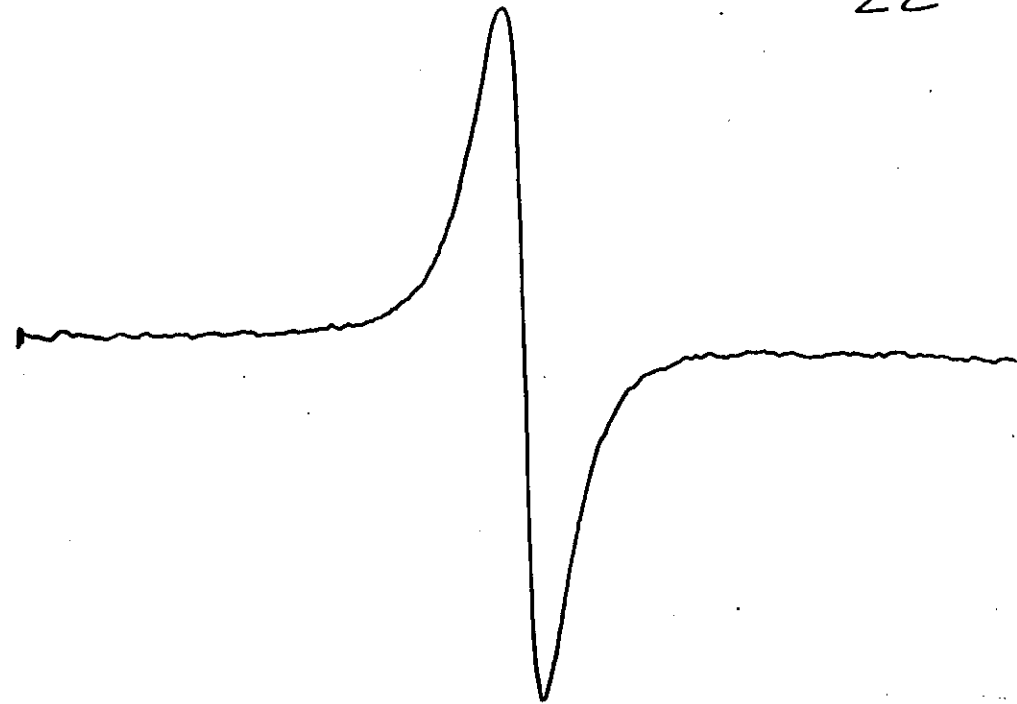
$$I_s = \frac{\epsilon_0 c \hbar^2 \gamma A}{2 |\mu_{12}|^2 \tau}$$

SAT.
INTENSITY

$$S = I/I_s$$

SATURATION
PARAMETER

22



- 155 m Torr
- 400 ms T.C.
- $\Delta C O_2 = 10 R(16) - 10 R(38) (^{13}C)$
 $= 999\ 465.831\ \text{MHz}$
- $\nu = 999\ 416.100 (50)\ \text{MHz}$

23. CH3OH ground state

23

RESOLUTION

CAN BE MASKED BY COLLISIONS

24

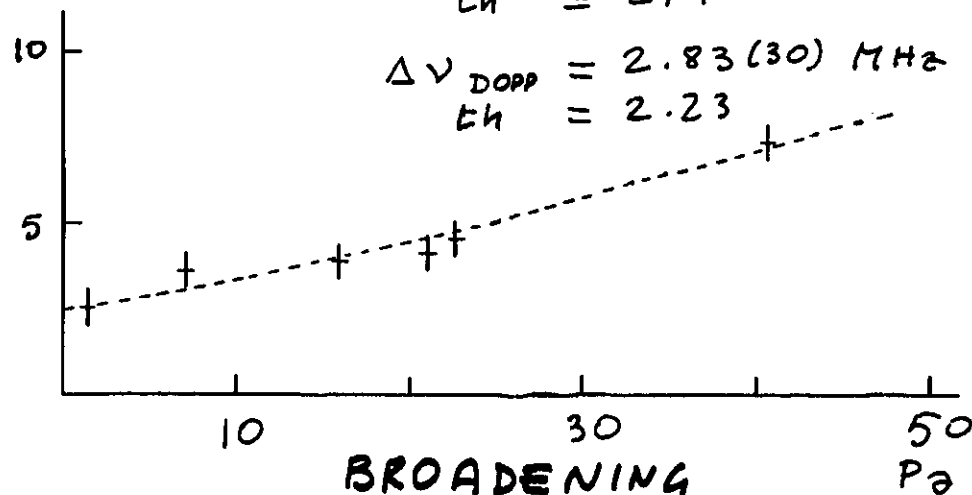
$\Delta\nu$ (MHz)

$$\Delta\nu_{\text{coll}} = 268(25) \text{ KHz/Pa}$$

$$\epsilon_h = 271$$

$$\Delta\nu_{\text{Dopp}} = 2.83(30) \text{ MHz}$$

$$\epsilon_h = 2.23$$



BROADENING

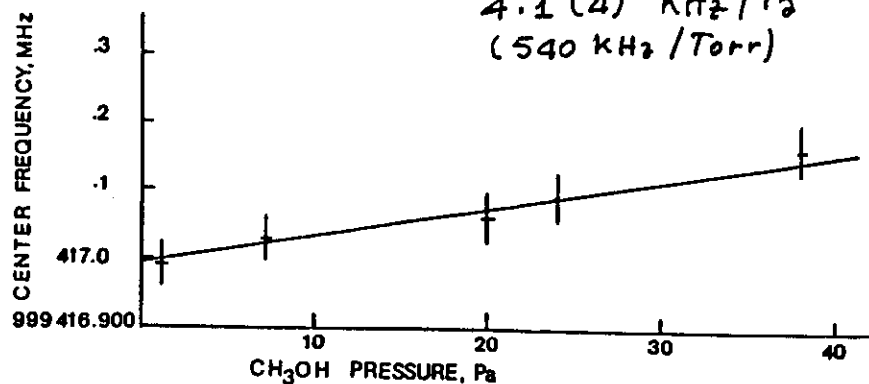
PRESSURE

SHIFT

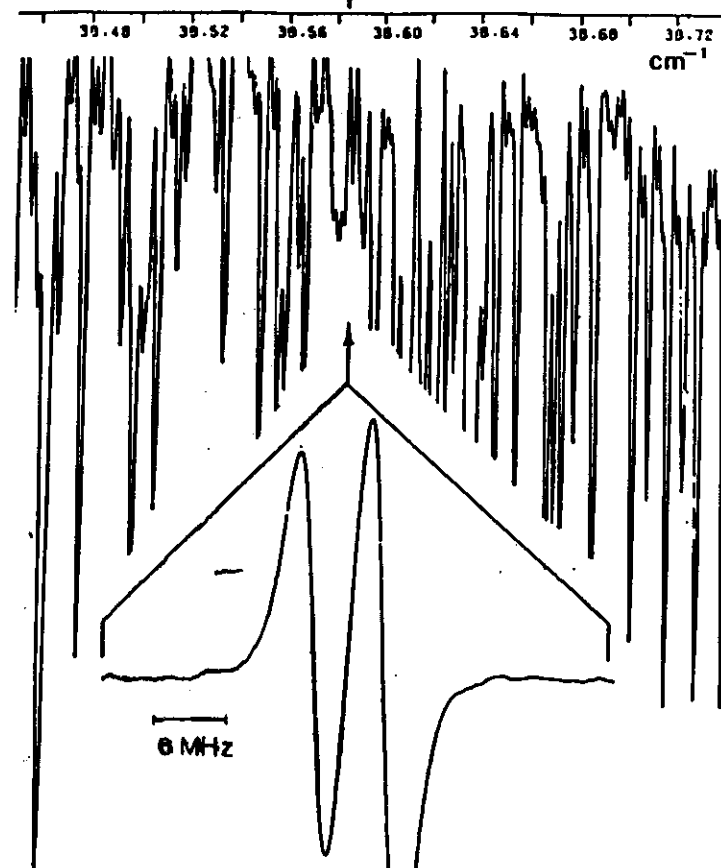
CENTER FREQUENCY, MHz

$$4.1(4) \text{ KHz/Pa}$$

$$(540 \text{ KHz/Torr})$$



259 μm

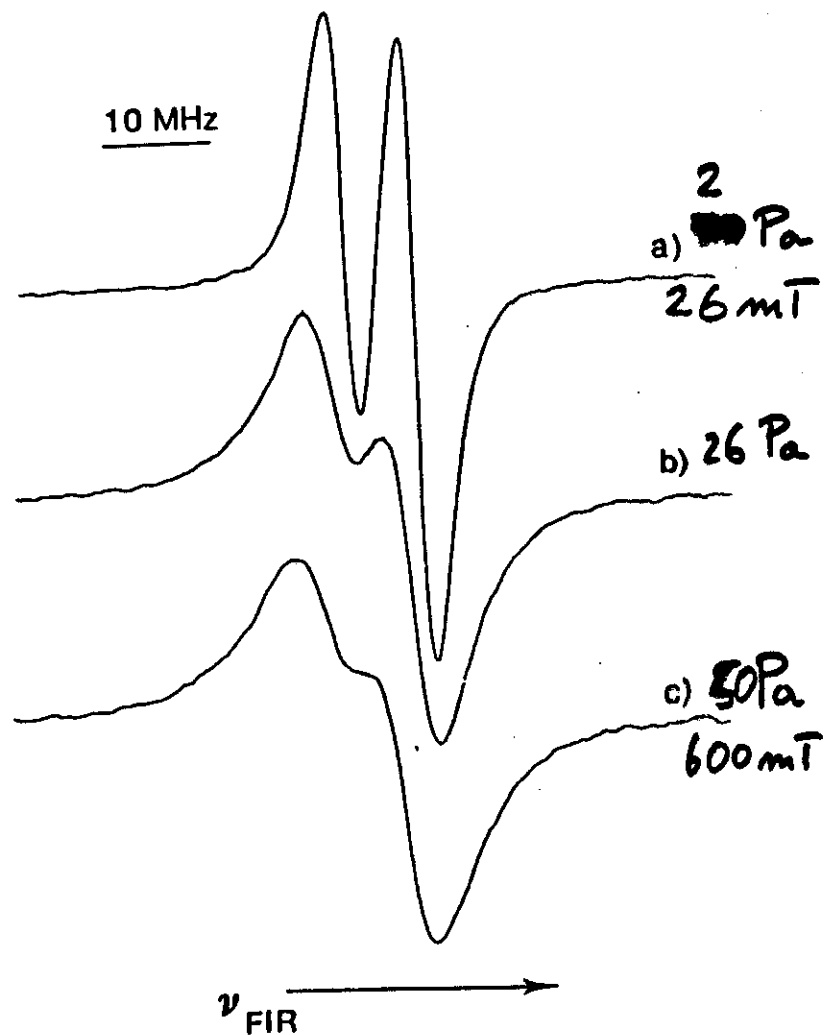


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J. MOL. SPECTROSC. (1972)

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A. GOZZINI, PISA SNS 1987
25



M. INGUSCIO - LECTURE N. 2

TECHNIQUES FOR HIGH RES. (SUBDOPPLER)
SPECTROSCOPY

- a) ATOMIC BEAMS
- b) SATURATION SPECTROSCOPY
- c) POLARIZATION SPECTR.
- d) TWO-PHOTON
- e) APPLICATIONS

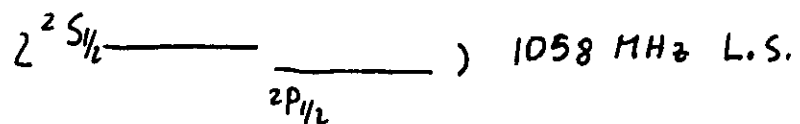
TR

HIGH RESOLUTION (SUBDOPPLER)

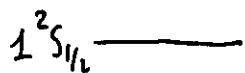
SPECTROSCOPY NECESSARY FOR:

- 1) OBSERVATION OF HOMOGENEOUS LINEWIDTHS (LIFETIMES, ATOMIC INTERACTIONS ...)
- 2) OBSERVATION OF STRUCTURES (SPLITTINGS) OF THE LEVELS (FINE, HYPERFINE, ISOTOPE ...)

A FUNDAMENTAL EXAMPLE (QUANTUM ELECTRODYNAMICS): LAMB SHIFT IN ATOMIC HYDROGEN



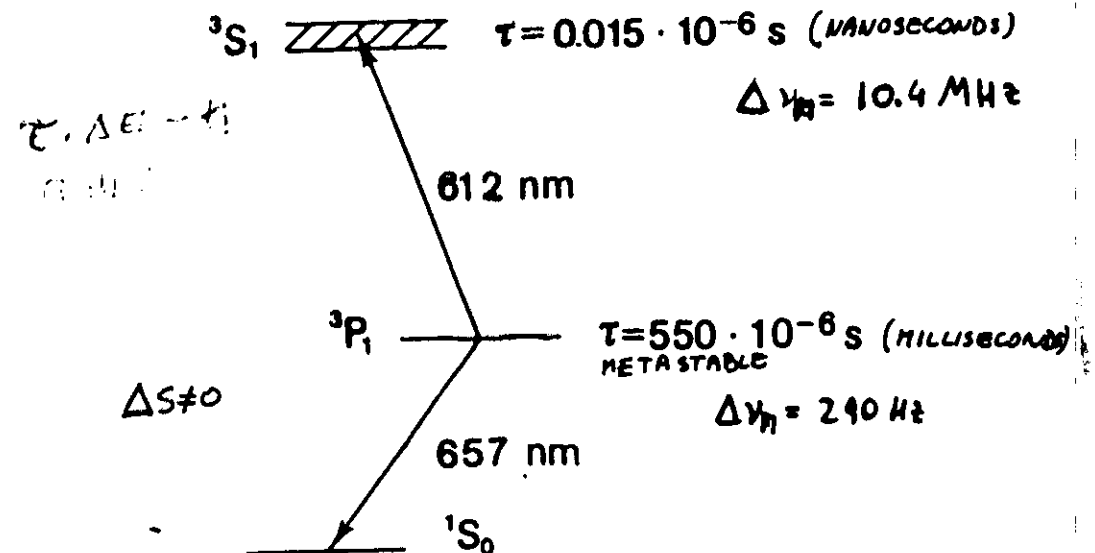
DOPPLER WIDTH OF BALMER α $\Delta\nu \sim 6000$ MHz



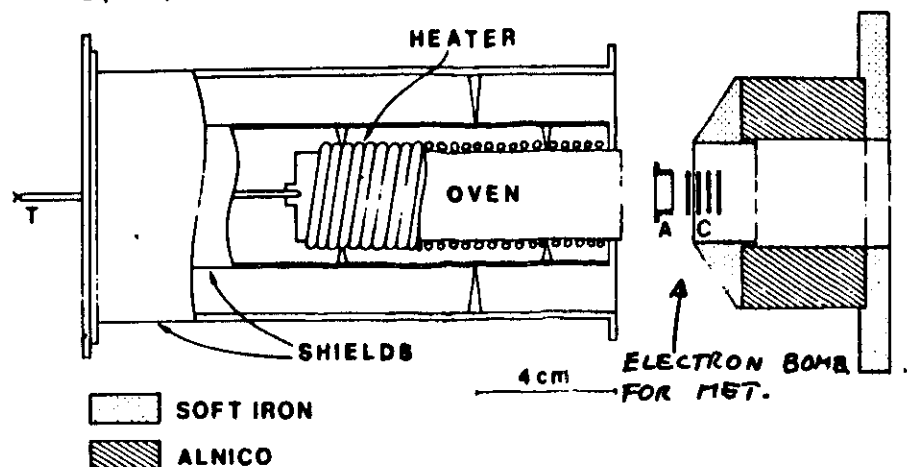
EARLIER APPROACHES

- LOWERING T (NOT VERY EFF. ($\sqrt{T}$) AND LIMITED TO SPECIES WITH ENOUGH DENSITY AT LOW TEMP.)
- ATOMIC COLLIMATED BEAMS + ORTHOGONAL OBSERVATION.

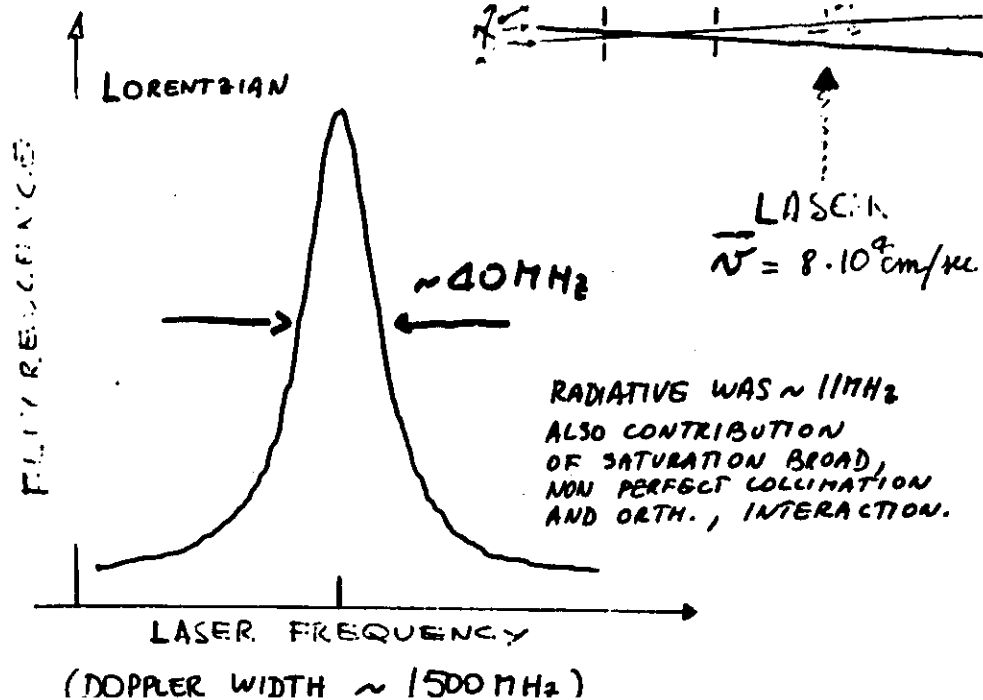
EXAMPLE: OBSERVATION OF THE TRANSITION AT $612 \text{ \AA}$ IN AN ATOMIC BEAM OF METASTABLE CALCIUM



OVEN FOR THE PRODUCTION
OF ATOMS (600°C)

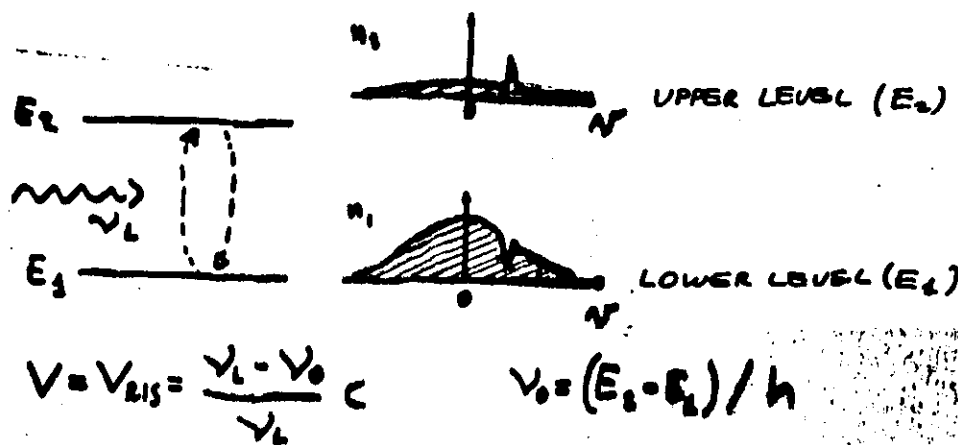


ATOMIC BEAM



LASER SOURCE { MONOCROMATIC ($\Delta \nu_{\text{LAS}} < \Delta \nu_{\text{DOPPLER}}$)
DIRECTIONAL
INTENSE (ENOUGH TO SATURATE)

SATURATION SPECTROSCOPY
(WE CHANGE THE EQUILIBRIUM ENERGY DISTR.
BETWEEN THE LEVELS, SELECTIVE IN VELOCITY)



COUNTER PROPAGATING BEAMS : AT RESONANCE
AT LINE CENTER WE HAVE A "DIP", WITH

2.3.

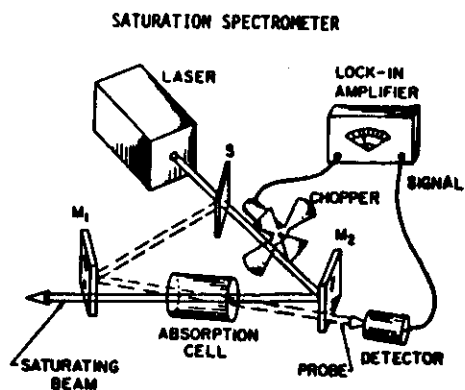
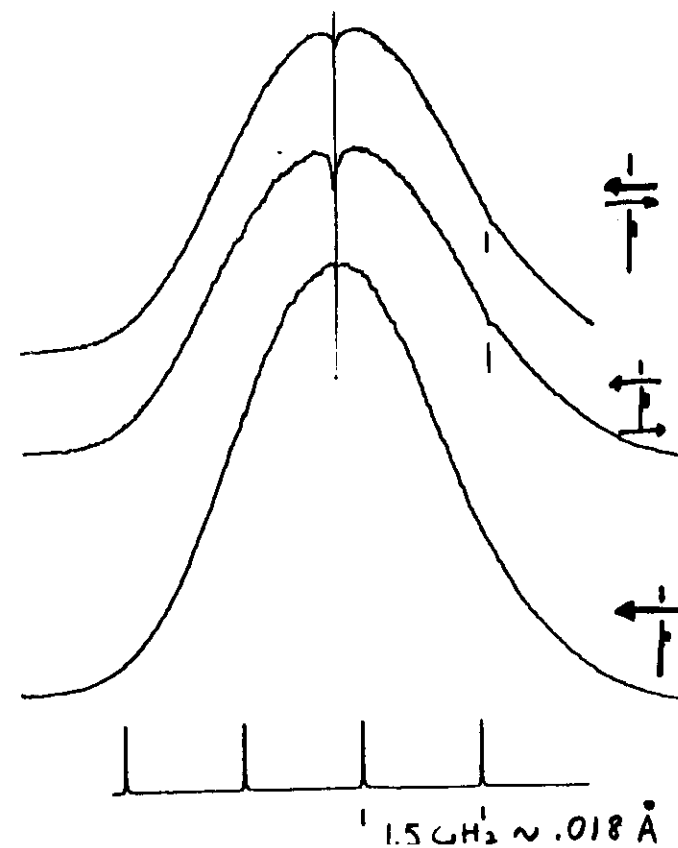


Fig. 1. Apparatus for Doppler-free saturated absorption spectroscopy of a gas sample.

EXPERIMENTAL APPARATUS

NeI $1s_5 \rightarrow 1p_1$ 5944.5 Å
 950 mTorr, 50 mA, $I_L = 50 \text{ mW}$



2.4.

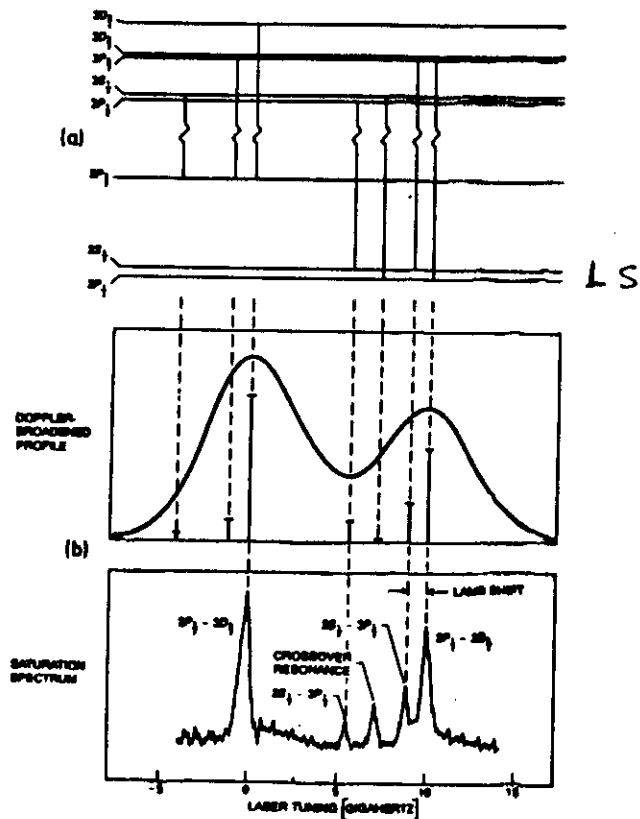


Fig.10.21a,b. Measurement of the Rydberg constant by saturation spectroscopy of the Hydrogen Balmer α transition. (a) Level scheme, (b) Doppler profiles and saturation spectrum of the Balmer α line in a hydrogen discharge [10.31a]

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

SOMETIME THE SENSITIVITY
OF SATURATION SPECTR. NOT
ENOUGH (DIP CONTRAST)

NEW TECHNIQUES

- POLARIZATION SPECTROSCOPY
(ILLUSTRATED FOR THE HYPERFINE
STRUCTURE OF MOLECULAR IODINE)
- INTERMODULATED SPECTR.
(NEXT LECTURE COMBINED WITH
OPTOGALVANIC DETECTION).

I₂ SPECTRUM (GOOD FOR λ CALIBRATION)

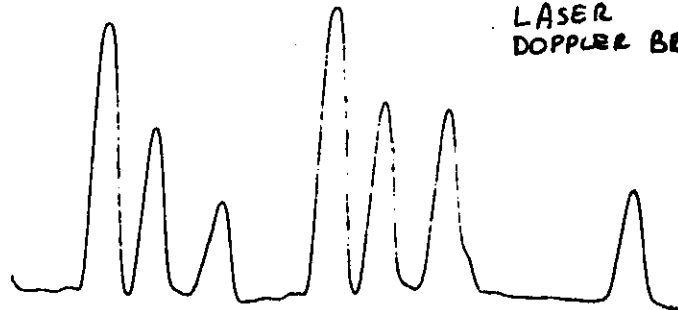
9



FOURIER
TRANSFORM



LASER
DOPPLER BROAD.



1939	16232.7093	2.1	34	1971	16236.4971	1.1	32	2003	16242.8937	.7	31	2030	16247.0429	.7	42
1940	16232.9059	.8	32	1972	16236.5948	.8	31	2004	16243.2738	.8	23	2030	16247.4480	1.7	17
1941	16233.0872	.7	33	1973	16236.7526	.8	30	2005	16243.3620	.9	17	2037	16247.6548	.8	46
1942	16233.1232	.4	31	1974	16236.1054	.8	29	2006	16243.3662	.8	30	2038	16247.8451	0.6	17
1943	16233.2791	2.4	15	1975	16236.1964	.8	25	2007	16243.4739	.7	30	2039	16247.8973	.7	30
1944	16233.6513	1.6	20	1976	16236.2974	.8	47	2008	16243.7890	.8	30	2040	16248.2979	.6	63
1945	16233.7256	2.3	45	1977	16236.4986	1.6	10	2009	16243.7776	1.8	21	2041	16248.4661	.8	27
1946	16233.7472	.9	40	1978	16236.6125	.4	63	2010	16244.0020	1.0	24	2042	16248.5808	.9	46
1947	16233.8410	.7	25	1979	16236.6930	.7	31	2011	16244.1065	1.3	46	2043	16248.6767	.8	21
1948	16234.1210	.8	48	1980	16236.7828	.7	27	2012	16244.1982	.7	10	2044	16248.8010	.6	37
1949	16234.4043	.6	31	1981	16236.1895	.8	48	2013	16244.4252	.6	42	2045	16249.1482	.5	33
1950	16234.5702	.6	30	1982	16240.2074	.8	21	2014	16244.4804	1.1	10	2046	16249.3676	.9	26

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

$\pm 60 \text{ MHz}$

Fig. 15 Registrazione in fluorescenza stimolata di una regione dello spettro della molecola di I₂ e confronto con le righe tabulate.

DIMERI + HYPERFINE

10

15 comp. J. value
21 comp. K. value

HYPERFINE INTERACTIONS
FROM SPECTRA

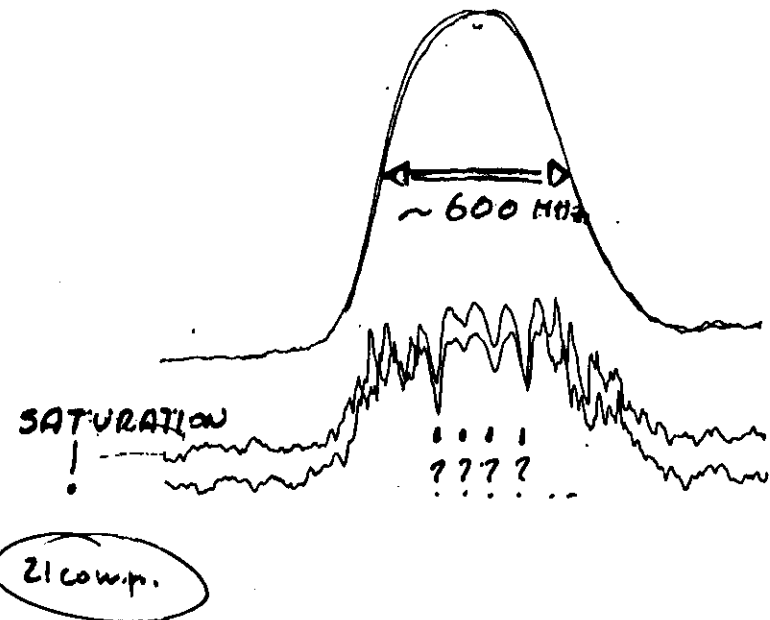


Fig. 22 Struttura iperfina della molecola di Iodio. Profilo di assorbimento con fascio opposto saturante.

POLARIZATION SPECTR

11

(SATURATION COMBINED WITH OPTICAL PUMPING).

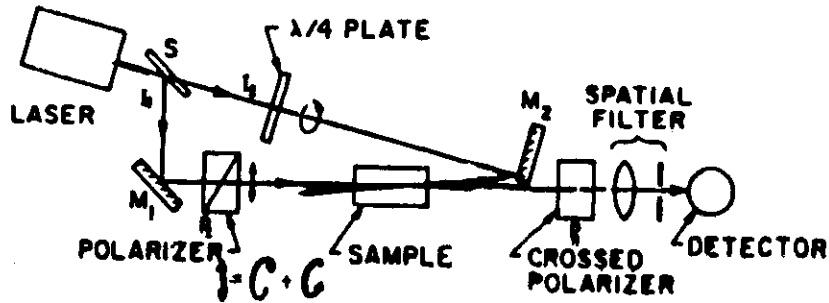
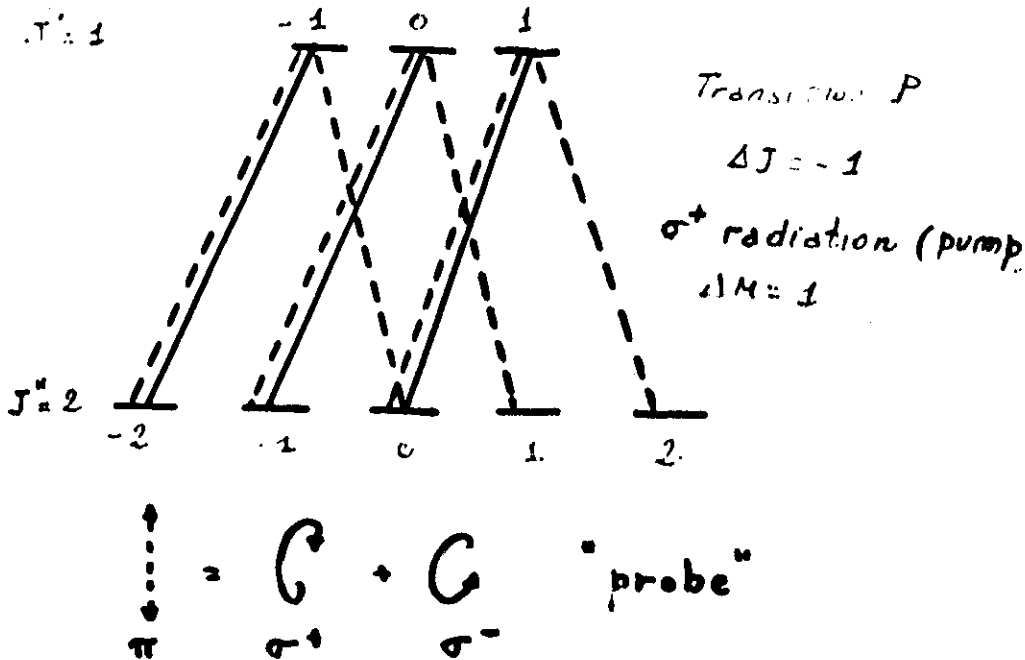


Fig.10.34. Schematic experimental arrangement for polarization spectro

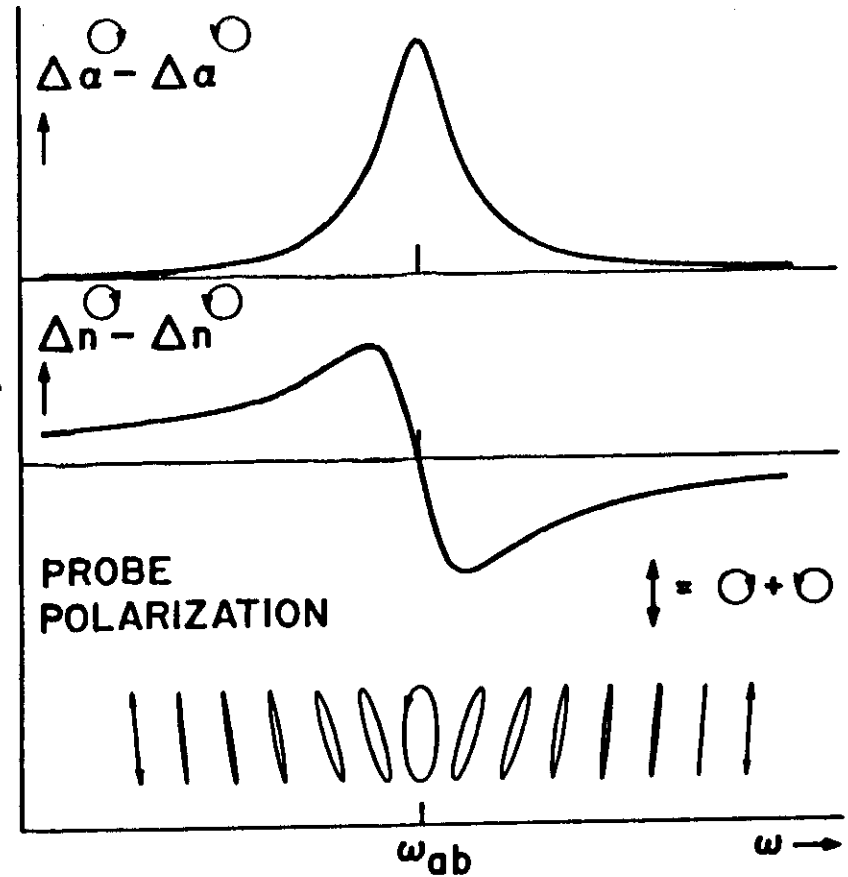


INDUCED BIREFRINGENCY

12

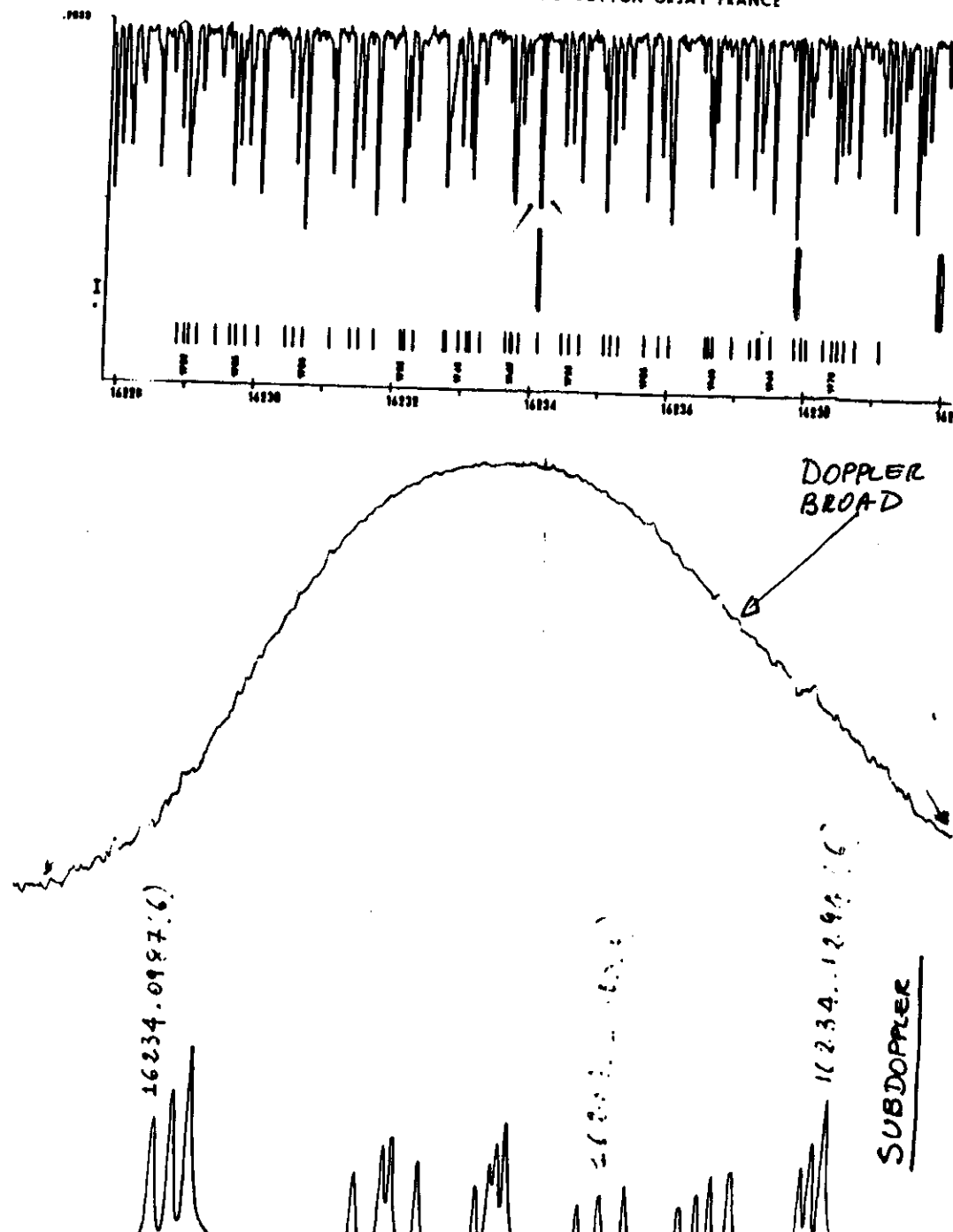
21

DOPPLER FREE LASER POLARIZATION SPECTROSCOPY

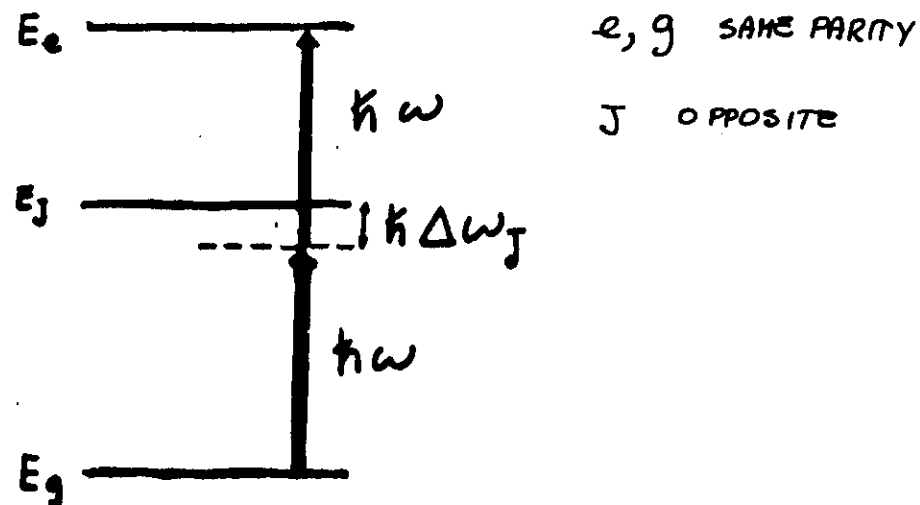


LASER FREQUENCY

$$I_T = I_0 \left[\frac{1}{2} (1 + \cos 2\alpha) + \frac{1}{2} (\sin 2\alpha)^2 \right]$$



2 PHOTON TRANSITIONS



ONLY REQUIREMENT: ENERGY CONSERVATION
AT THE END OF THE PROCESS.

$$E_e - E_g = 2 \hbar \omega$$

(POSSIBLE IMPORTANCE OF A NEARBY RES.
INTERMEDIATE LEVEL).

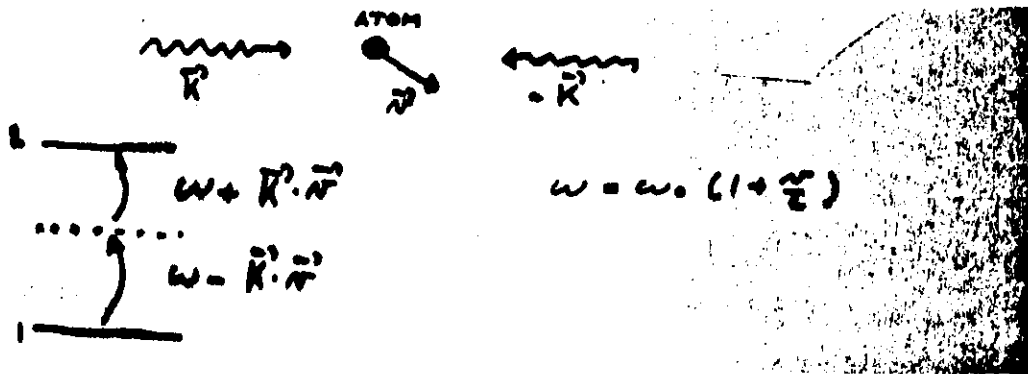
FIRST OBS. MICROWAVES (KASTLER)

INTEREST IN ATOMIC AND MOLECULAR SPECTROSCOPY

1) DIFFERENT SELECTION RULES FROM SINGLE PHOTON (PARITY)

2) TWO PHOTON EXCITATION ALLOWS TO USE VISIBLE OR NEAR UV RAD. FOR ENERGY JUMPS CORRESPONDING TO SHORTER WAVELENGTHS

3) ELIMINATION OF 1st ORDER DOPPLER WHEN PHOTONS ARE ABSORBED FROM COUNTER-PROP. BEAMS.



ENERGY CONSERVATION

$$E_2 - E_1 = \hbar(\omega + \vec{k} \cdot \vec{n}) + \hbar(\omega - \vec{k} \cdot \vec{n}) = 2\hbar\omega$$

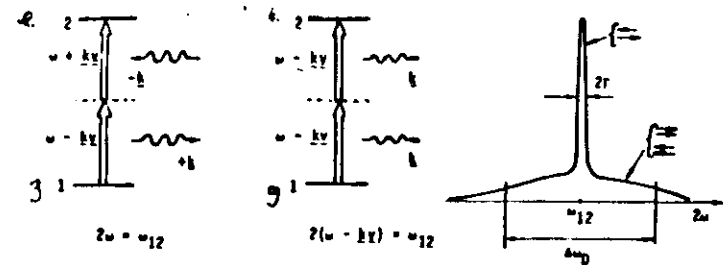
At resonance satisfied by all atoms independently

LINE SHAPE

$$P_{12}(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt e^{i(\omega - \omega_0)t} \langle e^{i\vec{k} \cdot \vec{r}(t)} \rangle$$

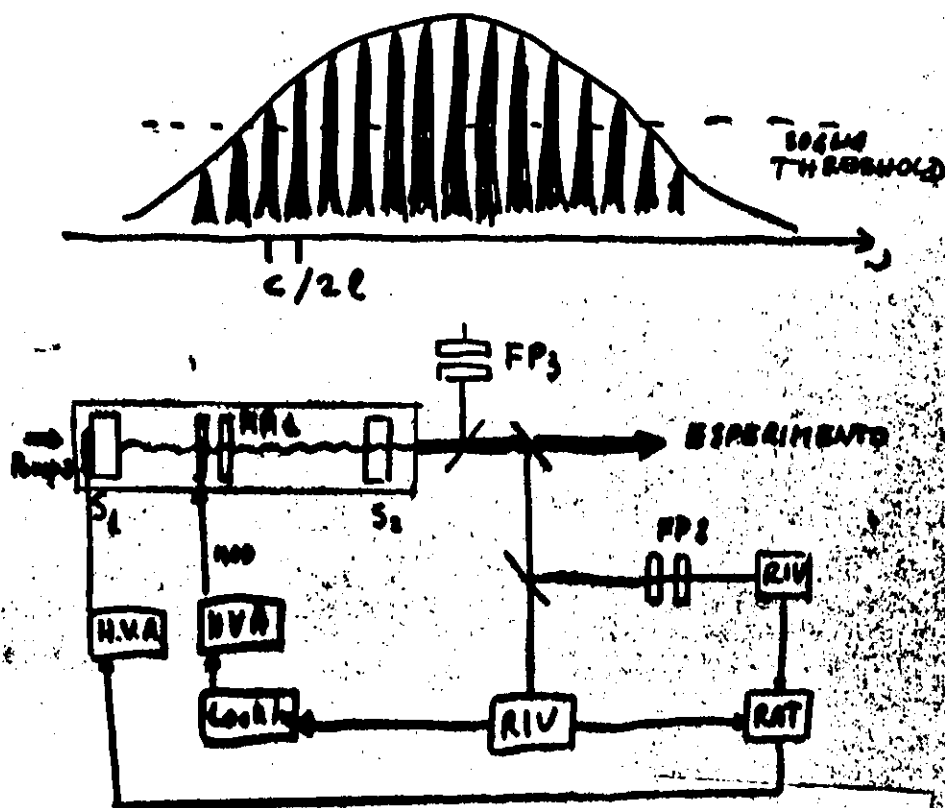
$$P_{12}(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt e^{i(\omega - \omega_0)t} \langle e^{i\vec{k} \cdot \vec{r}(t)} \rangle = \frac{\Gamma}{1 + (\omega - \omega_0)^2 \frac{\Gamma^2}{4}}$$

(i = 4, 2)



LORENTZIAN
OVER A

APPARATO SPERIMENTALE: IL LASER
 APPARATUS EXPERIMENTAL (BYE LASER)



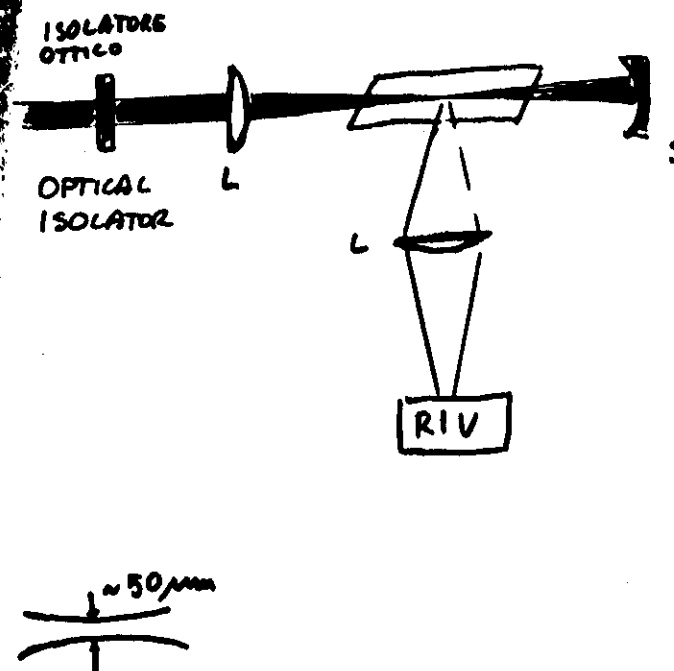
FP1 MODE SELECTION

FP2 CONTROL OF FREQUENCY

FP3 CALIBRATION OF FREQ SCALE

$\Delta \nu_{\text{vis}} \sim 1 \text{ MHz} \sim 10^{-5} \text{ \AA}$ in the visible

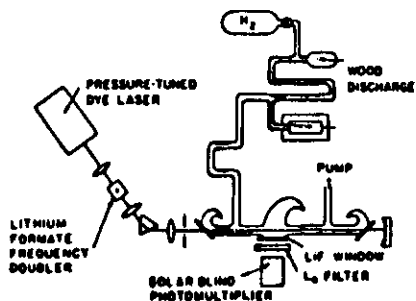
HIGH ENERGY DENSITY
 (STRONG FOCUSING)



HYDROGEN

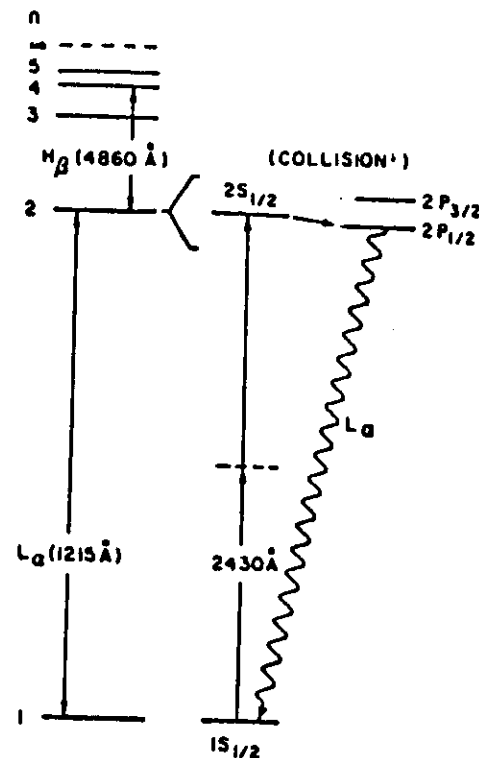
218

N_2 1 MW
 Dye
 4860 50 kW
 2430 LiF
 < 1 kW
 10 nsec
 $\Delta \nu \sim 100 \text{ MHz}$



Hänsch et al 1975

7.3.

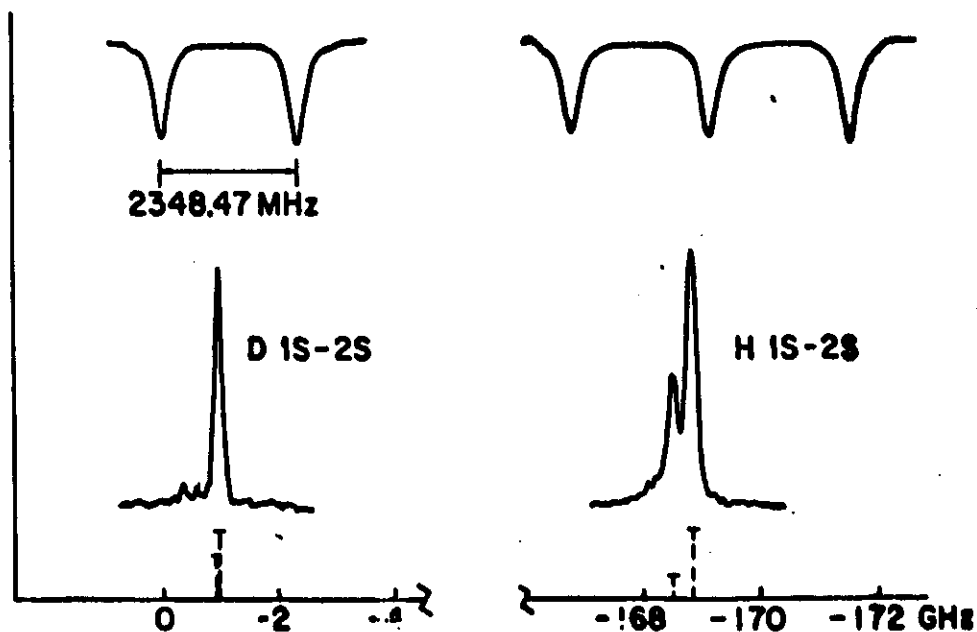


$$1215 \times 2 = 2430 \times 2 = 4860$$

ISOTOPE SHIFT OF HYDROGEN 1S-2S

RELATIVE ORDER

$n=0$ 71 72 73



DYE LASER FREQUENCY DETUNING ($-\nu$)

$\Delta \nu < 2\%$ $\Delta \nu_{D,H} = 0$

7.4.

