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WINTER COLLEGE ON LASERS, ATOMIC AND MOLECULAR PHYSICS

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Molecular Spectroscopy with IR and FIR Lasers

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Introduction

The development of lasers has made possible a detailed study of excited molecular and atomic states. Their spectral purity makes it possible to record absorption spectra with a resolution depending on the state of the system under investigation rather than on the instrumentation. Furthermore the power available from many lasers is sufficient to significantly change the population of levels involved in a transition, leading to an entirely new class of spectroscopic experiments.

Under proper experimental conditions, population inversion can also be obtained. This is the basis for the development of optically pumped lasers. Among these, molecular lasers pumped by CO_2 or N_2O lasers and emitting in the Far Infrared (FIR) play an outstanding role. The dramatic importance of these sources for molecular and atomic spectroscopy is mainly due to the fact that before their discovery (Chang and Bridges, 1970) the FIR region of the electromagnetic spectrum ($\lambda = 30 \mu\text{m}$ to 1 mm) was only scarcely covered by suitable photon sources. Furthermore these lasers have been demonstrated to be by themselves powerful sources of information on the infrared (vibrational) and far infrared (rotational) spectra of the molecules used as active medium.

The purpose of these lectures is to describe:

- 1) the physical mechanisms involved in the laser action;
- 2) the new nonlinear spectroscopic techniques developed thanks to these lasers;
- 3) the application of these techniques to some particular case.

Optically pumped three-level lasers.

In optically pumped FIR lasers the active medium is a polar molecular gas which absorbs (Fig. 1) the radiation from a strong IR laser (usually a $10 \mu\text{m}$ CO_2 laser). The absorption of pump radiation leads to inversion between rotational levels in an excited vibrational state. The population inversion is possible because the energy of the pump radiation is larger than kT and the other levels of the excited vibrational state are much less populated. The relaxation

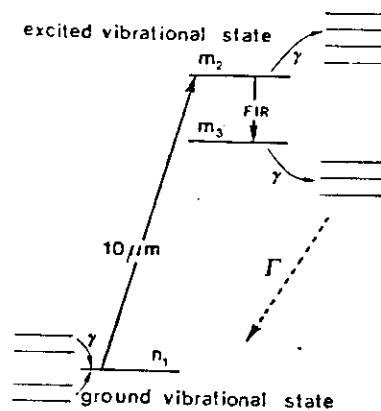


FIG. 1

between the levels is caused by collisions. The rotational relaxation within a given vibrational state is fast ($\tau^{-1} = \gamma = 10^8 \text{ Torr}^{-1} \text{ sec}^{-1}$) and tends to thermalize the population among the levels. The vibrational relaxation Γ is slow ($10^3 \text{ Torr}^{-1} \text{ sec}^{-1}$) and hence also the collisions with the walls, proportional to $p^{-1} d^{-2}$ (p gas pressure, d diameter of the laser resonator) are important to remove population from the lower laser level. That suggested the use of small cross section waveguide resonators to improve the efficiency of the lasers. A recent complete rate-equation analysis of the threshold conditions for CW operation can be found in Strumia, 1982. In the most favourable cases

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pump thresholds of only a few hundred mWatts are enough for cw operation. Output levels up to several tens of milliwatts have been obtained in cw operation from several molecules and on many different laser lines.

The active molecules can be considered as a coupled three-level system interacting with applied radiation fields. A peculiarity of the system in this case is that one of the coupled transitions generates itself a laser emission. Different processes manifest themselves depending on whether one or both of the coupled transitions are homogeneously or Doppler broadened.

The homogeneous width of the levels is determined by collisions and saturation:

$$\Delta\nu_{\text{homogeneous}} = \Delta\nu_{\text{collisions}} \left(1 + I/I_{\text{sat}}\right)^{1/2} \quad (1)$$

For the polar molecules active in the FIR it is $\Delta\nu_{\text{coll}} \approx 10-40 \text{ MHz/Torr}$, as a consequence at the typical cw operation pressures (5-500 mTorr) the homogeneous width results of the order of a few MHz, weakly affected by the actual pressure. In fact at lower pressures the saturation intensity I_{sat} decreases and the saturation broadening compensates the decrease of $\Delta\nu_{\text{coll}}$.

As for the inhomogeneous Doppler width, it is of the order of several ten MHz in the IR pump transition, while in the FIR emission it can range from less than 1 MHz to more than 10 MHz, depending on the wavelength and on the molecular mass.

As a consequence the three coupled level system can be considered Doppler-Doppler or Doppler-Homogeneous broadened, depending on the FIR wavelength, on the pressure and on the

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pump intensity.

Doppler-Doppler broadened system: it is possible to observe tuning of the FIR emission by tuning the pump radiation across the absorption Doppler profile. If a detuning Δp is introduced

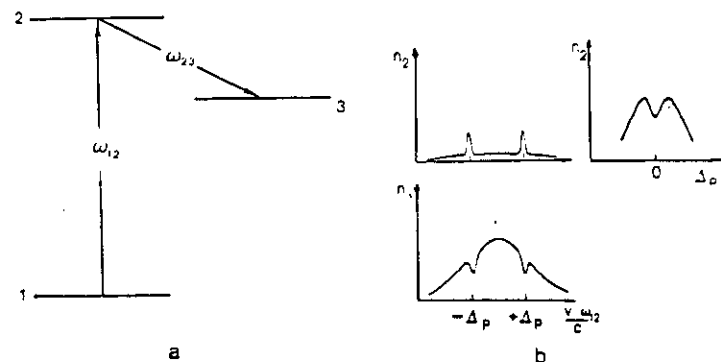


FIG. 2

between the pump frequency and the center of the absorption (Fig 2), only the molecules with the proper velocity component along the laser direction, $v = c \Delta p / \omega_{12}$, can be excited. If the homogeneous broadening is negligible also on the rotational transition, the FIR emission is Doppler shifted and the gain curve consists of the sum of two Lorentzian curves centered at frequencies:

$$\Omega_{\pm} = \omega_{23} \pm \omega_{23} \Delta p / \omega_{12} \quad (2)$$

corresponding to FIR co- and counter propagating with the pump beam. (If Raman-type processes are considered, an anisotropy in intensity and width of the two curves is also introduced).

As a consequence of Eq. 2, by scanning the FIR resonator length when a detuning Δp is present, two different peaks can be observed in the FIR emission if the splitting:

$$\Delta V_{FIR} = 2\omega_{23} \Delta p / \omega_{12} = 2\Delta p \lambda_{IR} / \lambda_{FIR} \quad (4)$$

exceeds the homogeneous width. A typical experimental recording is shown in FIG 3. Here a waveguide CO₂ laser tuned on the 10 R(34) line at 10.158 μ m is used to pump CH₃OH and lead to FIR emission at 62.98 μ m. The emission profile is recorded for different

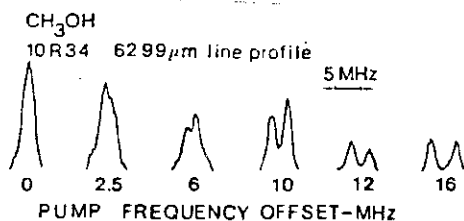


FIG. 3

offsets of the pump frequency. No FIR splitting is detected for zero pump detuning, while a FIR splitting increasing with an increasing pump detuning is observed, in agreement with the theoretical slope $\lambda_{IR}/\lambda_{FIR} = 0.16$. It should be noted that the effect can also be observed by scanning the pump frequency while the FIR resonator is tuned off the exact resonance position: the FIR signal will be at a local minimum when the pump offset sweeps the FIR frequency through resonance (Henningen et al 1982).

A more general effect, which is observable also in the Doppler-homogeneous case of broadening is the IR-FIR Transferred Lamb Dip (TLD) (Inguscio et al. 1979).

Since the pump is effected in a resonator configuration, the system interacts with forward and backward propagating IR beams

and with standing wave FIR radiation. A saturation is generated in the IR Doppler profile, causing a decrease of the absorption and of the number n_2 of excited molecules (FIG 2 b).

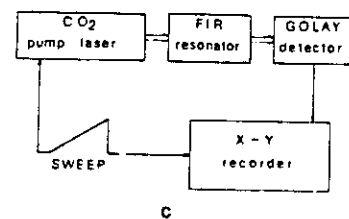
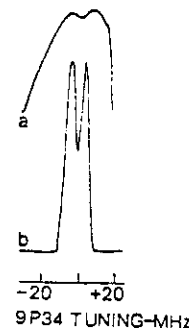


FIG. 4

as shown in FIG. 4 a. The overall FIR laser intensity depends on the number of excited molecules, therefore zero pump detuning causes also a dip in the FIR laser emission, as recorded in fig. 4b. The 2→3 transition generates a laser emission and hence is easy to investigate by means of the experimental apparatus schematically shown in FIG. 4c.

An important feature is that the IR saturation effect is "amplified" due to the nonlinear detection via the FIR laser oscillation (compare FIG 4 a and b): Dip contrasts up to 100% can be obtained by a proper choice of the laser threshold. TLD is a general technique which has been already applied in a wide wavelength range (370 to 1200 μ m) and to several laser molecules (CH₃F, CH₃Br, CH₃OH, C₂F₂, NH₃...).

Since the effect is originated in the absorption and only "transferred" to the emission, the center frequency does not depend on the FIR

recording of the infrared bands dip on a vibrational transition of CH₃OH is obtained using the optoacoustic technique at pressures typical for consequent FIR action (~100 mTorr),

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resonator tuning - interesting measurements are shown in FIG. 5 for the CH_3OH emission at $133.12 \mu\text{m}$, pumped by the CO_2 9 P 24 laser line. The TLD on the top of the FIR profile is recorded at different pressures of CH_3OH being gas. Pressure affects the emitted intensity, the tuning of the FIR resonator (via the refractive index), hence the symmetry of the FIR line, but not the frequency position of the TLD. Also the width of the TLD is not strongly affected by pressure and this is consistent

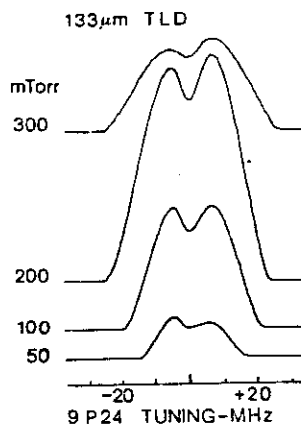


FIG. 5

with previous considerations on Ep. 1.

These features allow experimental investigation of the IR transition via the coupled FIR emission. The simultaneous recording of TLD and CO_2 tuning curve allows the determination of pump offsets: the detection in the coupled FIR emission avoids the ambiguity that exists in two-level spectroscopy when several absorptions are located within the CO_2 laser tuning range.

A typical situation is illustrated in Fig. 6

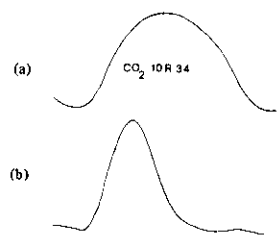


FIG. 6.

This broad absorption is centered by at least

for the CH_3OH absorption around the CO_2 10 R 34 laser line. The mode profile (total tuning 240 MHz) is recorded in a). The optoelectronic spectrum (b) from CH_3OH at 380 mTorr reveals a broad absorption at the low frequency side.

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Three vibrational transitions overlapping within the Doppler profile - A total of ~~nine~~ different

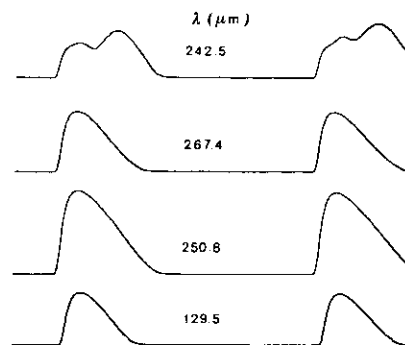


FIG. 7

Dip is recorded only on the line whose pump transition center is accessible - By using all the larger tuning range of FIG. 6 the TLD can be observed also on the other three lines and other two new laser lines, at a further different offset value, can be discovered. In FIG. 8 it is recorded as an example the TLD on the $250.8 \mu\text{m}$ line, inasmuch in FIG. 7 and one of even larger offset

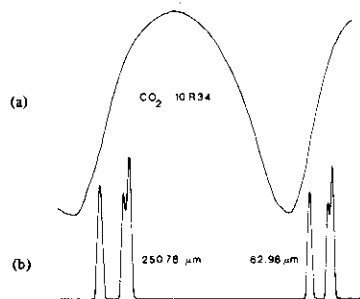


FIG. 8

lines. It is worth stressing that the power of the TLD technique in determining whether FIR emissions share a common pump transition is of fundamental importance in the assignment of the quantum numbers to the pump-emission cycle.

TLD and Stark Effect

Optically pumped FIR lasers can also be operated in presence of a static electric field. Since the active molecules are polar, they display a significant Stark effect. The energy splitting of the levels for a symmetric top molecule is given by:

$$W = -\mu E \frac{KM}{J(J+1)} \quad (5)$$

where μ is the electric dipole moment in Debye ($1D = 5.0344 \text{ MHz/V cm}^{-1}$), J the angular momentum, K and M its components along the molecular symmetry axis and the external electric field respectively. The Stark effect is more complex in the case of asymmetric rotors since K is not a good quantum number. (In general a perturbative treatment can be done and the first term is quadratic in the field). Linear Stark effects can be observed also on asymmetric rotor molecules when the K splitting is very small or when the K degeneracy is restored by some additional effect. This is for instance the case of the most prolific FIR laser molecule: CH_3OH . Here an internal rotation restores the K degeneracy for most of the levels. In this case the Stark energy splitting is given again by Eq. 5, but the effective value of μ may now change from one state to another.

An important example of resolved Stark pattern is shown in FIG. 9 for the CH_3F laser emission at $496 \mu\text{m}$ ($J, k: 12, 2-11, 2$). The observed Stark intensity pattern

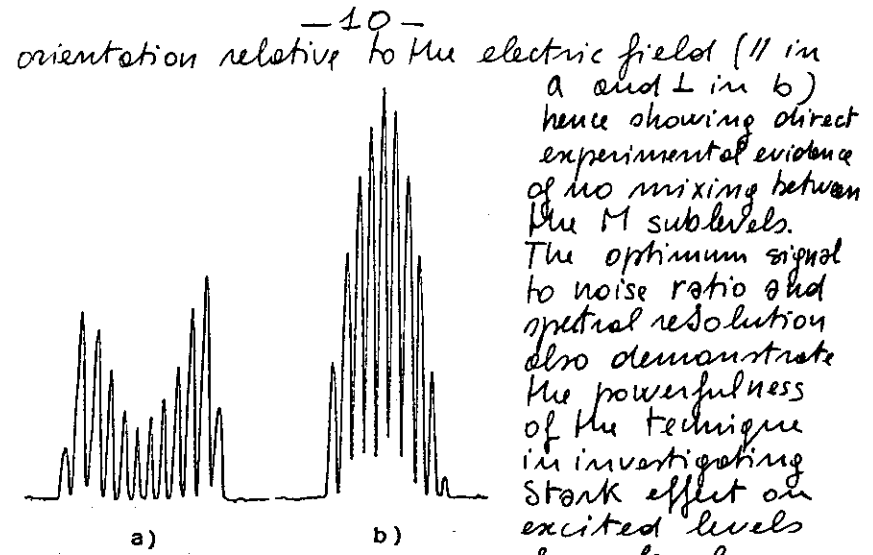


FIG. 9

in case of more complex molecules, different Stark lineshapes can be obtained, depending on the selection rules and quantum numbers. The different behaviours can be found in Strauss and Inguscio, 1982 and can significantly help the assignment of transitions.

Combining the Stark effect with the TLD technique it is also possible to investigate the effect of the electric field in the pump transition by monitoring the FIR emission. An example is illustrated in FIG 10, which refers to the optical pumping of a CH_3OH line at $205.6 \mu\text{m}$ whose pump transition is close to the high frequency end of the tuning range of a CO_2 laser oscillating on the $9P(34)$ line (tuning range $\pm 100 \text{ MHz}$ in the recording). The experiment is a sort of Laser Stark spectroscopy measurement

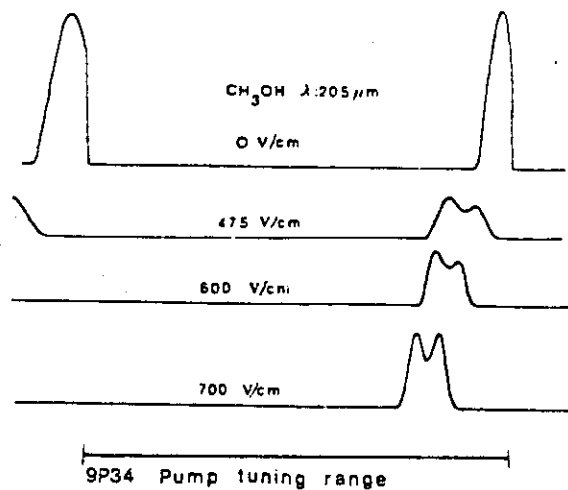


FIG. 10

laser tuning range, the different offsets of the TLD allow the investigation of the effect.

The techniques can also be applied to cases for which the Stark effect is not linear -

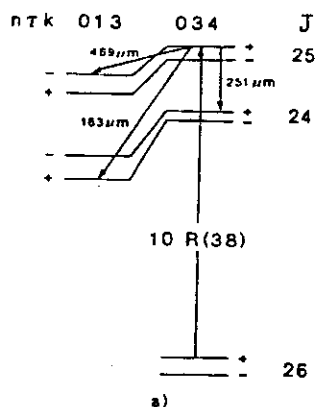


FIG. 11

upper and lower level (see "intracavity triple

but the pumped FIR emission is detected instead of the absorbed (or transmitted) radiation. At zero electric field the line center is not accessible and no TLD is detected. When the pump transition is Stark tuned across the CO_2

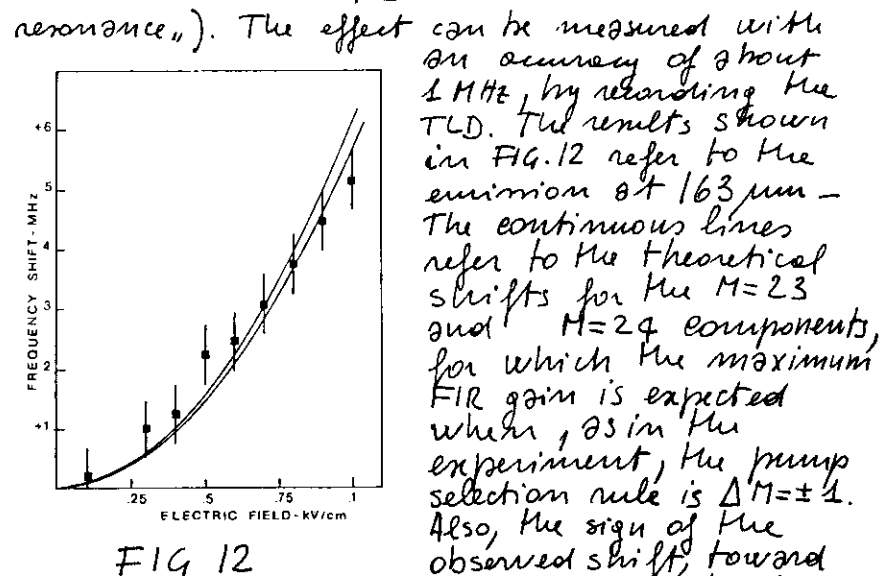


FIG. 12

high frequencies, is in agreement with the assignment as an $A^+ \rightarrow A^+$ transition.

Sub-Homogeneous Spectroscopic Resolution

Saturation and in general nonlinear spectroscopy of the transitions between two energy levels usually allows the resolution of structures larger than the homogeneous width. TLD is a saturation technique applied in three-level configuration yielding a resolution higher than the corresponding one in conventional high resolution techniques applied to two-level schemes. As an example we shall illustrate a typical case where structures are resolved which should be masked within the homogeneous linewidth in two-level spectroscopy. In FIG. 13a it is schematically shown a case in which the energy splittings are larger than the homogeneous width in

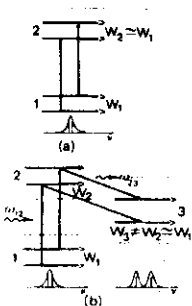


FIG 13

both the levels, but they are nearly equal. Therefore the photons associated to the $1 \rightarrow 2$ transitions have nearly the same energy and cannot be resolved in the homogeneous linewidth. In b) it is shown how, at least in principle, the three level configuration can be successful in overcoming the difficulties present in the two-level scheme. In fact the photons associated to the $1 \rightarrow 2$ transitions, unresolved within the homogeneous linewidth, are coupled to the photons of the $2 \rightarrow 3$ transitions, where the line structures are resolved. The $2 \rightarrow 3$ photons can be used to "label" $1 \rightarrow 2$ corresponding photons if a suitable technique is provided. TLD can do that, transferring and resolving in the far infrared the infrared saturation spectrum.

Experimental examples of the scheme illustrated in FIG 13a can be found in Stark spectroscopy of polar molecules: 1 and 2 can be regarded as two different vibrorotation levels, W_1 and W_2 as the Stark splittings. Let the energy separation between contiguous Stark sublevels be given by Eq. 5.

Depending on the radiation polarization, transitions are induced with $\Delta M = 0$ or ± 1 selection rules. The $1 \rightarrow 2$ line consists of various components equally spaced by a quantity $|W_2 - W_1| \cdot E$. In case of a Q branch transition ($\Delta J = 0, \Delta K = 0$), the difference between W_1 and W_2 is due only on the M values in the two vibrational states, differing only

of a few parts in 10^2 . Hence the line Stark structure is unresolved in spite of an energy sublevels splitting larger than the levels width both in 1 and 2 states. Higher resolution, overcoming the homogeneous linewidth limitation, is obtained in molecular lasers optically pumped in presence of an electric field (FIG. 14). The infrared $1 \rightarrow 2$ transition Stark

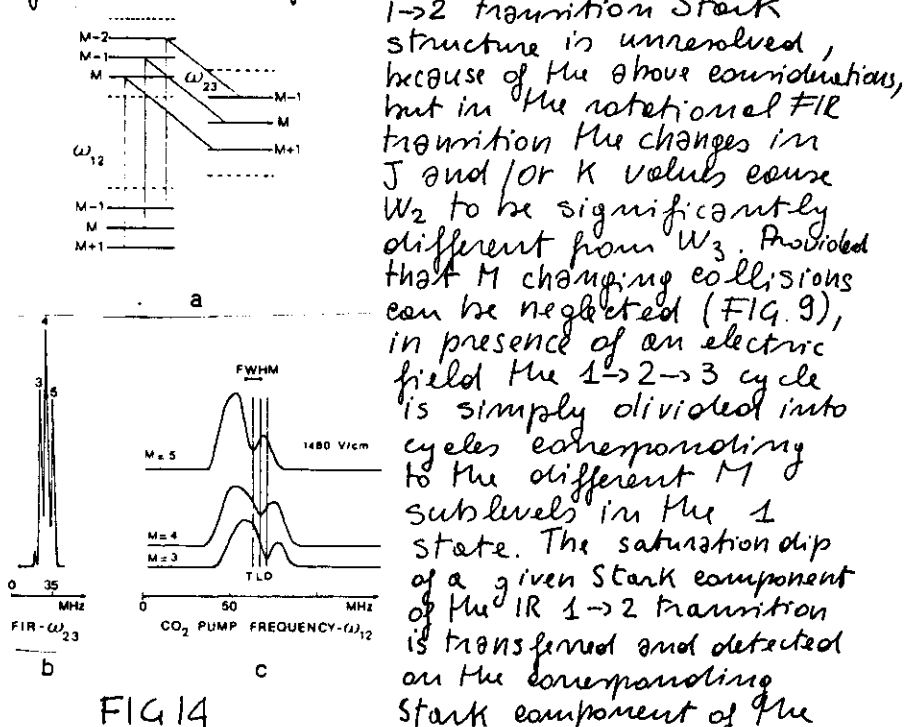


FIG 14

structure is unresolved, because of the above considerations, but in the rotational FIR transition the changes in J and/or K values cause W_2 to be significantly different from W_3 . Provided that M changing collisions can be neglected (FIG. 9), in presence of an electric field the $1 \rightarrow 2 \rightarrow 3$ cycle is simply divided into cycles corresponding to the different M sublevels in the 1 state. The saturation dip of a given Stark component of the IR $1 \rightarrow 2$ transition is transferred and detected on the corresponding Stark component of the coupled FIR emission. The technique is demonstrated in FIG 14 b, c for the FIR emission at $205 \mu m$ from CH_3OH pumped with the 9-P(34) CO_2 laser line. The three level quantum numbers are: $(J, K): (6, 5) \rightarrow (6, 5) \rightarrow (5, 4)$ and the Stark splittings

in the three levels are: $W_1 = 53.3$, $W_2 = 54.5$, and $W_3 = 61.0 \text{ MHz/KV cm}^{-1}$. The Stark splittings in the $1 \rightarrow 2$ infrared transition are given by $|W_2 - W_1| = 1.2 \text{ MHz/KV cm}^{-1}$, while in the coupled $2 \rightarrow 3$ FIR transition they are: $|W_3 - W_2| = 6.5 \text{ MHz/KV cm}^{-1}$. The levels width γ is of the order of a few megahertz. As a consequence for electric fields of the order of 1 KV/cm , the Stark structure of the levels is resolved but cannot be directly observed in the $1 \rightarrow 2$ transition line since $|W_2 - W_1| < \gamma$. On the contrary with electric fields of the order of 1 KV/cm $|W_3 - W_2| > \gamma$ and the Stark structure of the $2 \rightarrow 3$ transition can be observed (FIG 14b: FIR laser oscillation can be obtained on individual Stark components). By setting the FIR cavity length for oscillation on a given Stark component, and by scanning the CO_2 frequency (FIG 4c) it is detected the Doppler-free transferred Lamb-dip corresponding to the saturation of the component of the IR pump transition to which the observed FIR component is coupled. That is typically shown in FIG. 14c (Inguscio et al 1980). The transferred Lamb dips corresponding to different Stark FIR components are clearly observed at different positions of the CO_2 frequency. The electric field intensity was 1480 V/cm , corresponding to a dip frequency separation $|W_2 - W_1| \approx 1.8 \text{ MHz}$. These dips should be unresolved in conventional sub-Doppler two-level spectroscopy (the homogeneous linewidth of the two level transition can be estimated from the dips FWHM to be $\gamma \approx 5 \text{ MHz}$). A discussion of this superresolving technique in terms of information capacity of atomic and molecular spectroscopy can be found in Inguscio 1982.

Intracavity triple resonance

In laser intracavity double resonance experiments, the absorption of radio-frequency or microwave radiation by the medium under study is detected as a change in the laser output power. This technique has been extensively applied in the IR region with the medium as an intracavity absorber. The sensitivity of the technique is based on the pumping action of the laser and on the nonlinear response of the laser.

In optically pumped FIR lasers the active medium itself can be used to probe the rotational structure of the lasing molecule. Since the radiofrequency (or microwave) resonance can be detected on the effect on the FIR output amplitude, the experiment can be regarded as a triple-resonance one (IR-MW-FIR).

The experiment described in this lecture is based on the levels scheme illustrated in FIG 11 of pg. 11. The $10.1 \mu\text{m}$ CO_2 R(38) line pumps the $P_2(26)$ A+ line, where the subscript denotes the value of K , which is the axial component of the rotational angular momentum for the transition, and A+ indicates the torsional symmetry. The pumping of methanol molecules into the excited state leads to FIR laser emission at $163, 251$ and $469 \mu\text{m}$. The Δ_{JK} splitting between the A+ and A- levels, which is a K-doubling splitting akin to splitting in a rigid asymmetric rotor, is represented by:

$$\Delta(J, K) = \frac{(J+K)!}{(J-K)!} [S(K) + J(J+1)T(K)] \quad (6)$$

where the $S(K)$ and $T(K)$ parameters depend on

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the vibrational and torsional states. This parameters can be estimated from a simultaneous least-squares fit of the microwave data and IR laser data (Arimondo et al. 1980), at least for the ground state, hence obtaining $\Delta(26,4) = 577.5$ MHz. For the excited state, before the experiment here described, the values could only be estimated: $\Delta(25,4)$ and $\Delta(24,4)$ in the 300-400 MHz range, whereas $\Delta(25,3)$ and $\Delta(24,3)$ in the 5-6 GHz range. In the experiment the 250 mW radio frequency power was coupled into the FIR waveguide using a coaxial guide (Fig 15a)

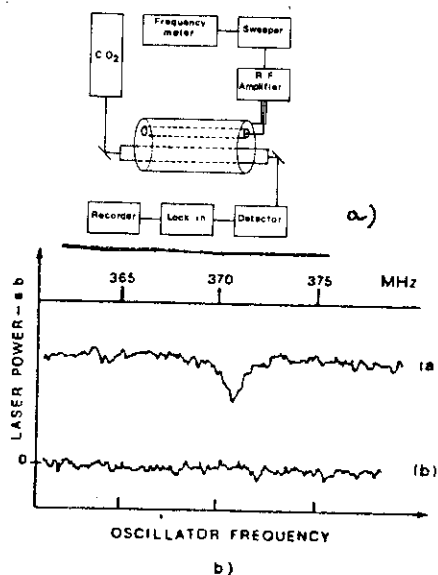


FIG. 15

states involved in the resonance is created

The radiofrequency field was applied between the internal and external conductors whose diameter was chosen in order to create a 50 Ω coaxial guide. A resonance signal at 370.8(5) MHz was observed when the FIR cavity was tuned for laser action on any of the 163-, 251-, and 489- μ m lines (Fig 15b, lower trace) was obtained in absence of CO₂ pumping to show that the population in the

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by IR pumping - This experiment is another example of determination of otherwise inaccessible structures using optically pumped molecular lasers (in this case the asymmetry splitting in a vibrational excited state). Furthermore the assignment of the three FIR laser lines is confirmed, proving that all of them originate from the same level.

The described experimental scheme can also be the basis for a study of the laser mechanism. The power P emitted by a laser operating at frequency ν with cross-sectional area A , length l , internal loss factor L , and mirror transmittance T may be written:

$$P = \frac{\hbar^2 \nu A \epsilon_0 \gamma_u \gamma_l}{2 \mu^2 g} \left(\frac{G}{L+T} - 1 \right) T \quad (7)$$

where the gain G on the lasing transition may be written as $G = 2 R \mu^2 g l / \hbar^2 \epsilon_0 \gamma_u (\gamma_u + \gamma_l)$.

γ_u and γ_l are the relaxation rates of the upper and lower levels, respectively, of the lasing transition (2 and 3 in Fig. 2a); μ is the dipole moment of the transition; g is the degeneracy of the lower level; ϵ_0 the vacuum dielectric permeability. R denotes the pumping rate for the upper level (CO₂ pumping from a ground vibrational level). The radio-frequency resonance produces an additional relaxation γ_r for the levels irradiated by radio frequency, dependent

on the relaxation parameters of the radiofrequency transition. Thus double resonance in the lower level of the pumping transition modifies the pumping rate to $R(1 + \gamma_u/\gamma_g)$, with γ_g the relaxation rate in the ground level (1), because the double resonance increases the number of molecules involved in the pumping action. Double resonance in the upper or lower levels of the laser transitions is described by a change in the relaxation rate: $\gamma_u + \gamma_r$ or $\gamma_l + \gamma_r$ for double resonance in the upper or lower level respectively. Supposing γ_r small compared with other rates, for the change in power produced by the double resonance in the ground (g) or upper (u) and lower (l) levels of the laser transition, it can be written:

$$\Delta P_g = \frac{\hbar^2 \nu A \epsilon_0 \gamma_u \gamma_l}{2 \mu^2 g} \frac{G l T}{L + T} \frac{\gamma_r}{\gamma_g} \quad (8)$$

$$\Delta P_u = \frac{\hbar^2 \nu A \epsilon_0 \gamma_l T}{2 \mu^2 g} \left(\frac{G l}{L + T} \frac{\gamma_u}{\gamma_u + \gamma_l} + 1 \right) \gamma_r \quad (9)$$

$$\Delta P_l = \frac{\hbar^2 \nu A \epsilon_0 \gamma_u T}{2 \mu^2 g} \left(\frac{G l}{L + T} \frac{\gamma_u}{\gamma_u + \gamma_l} - 1 \right) \gamma_r \quad (10)$$

Because the collisional broadenings γ_u and γ_l have the same magnitude, at large gain G the double resonance signals ΔP_u and ΔP_l

are equal and opposite. At low gain G , the double resonance signal in the upper level is larger than that in the lower one. In effect, the laser power is weak, and the population of the lower level is small, so that the double resonance in the lower level produces a small change in the laser output power. If the relaxation rate γ_r is comparable with γ_u and γ_l , the double resonance signal ΔP_g has the same intensity as ΔP_u , which is the double resonance signal in the upper level.

Direct frequency measurements

Gain curve of optically pumped FIR molecular lasers is in general homogeneously broadened. The linewidths of the resonator modes are larger than the gain curve and as a consequence the frequency of the emitted radiation is that of the molecular transition, only slightly perturbed by the cavity tuning via pulling effects. Frequency instabilities down to 2×10^{-12} have been demonstrated in apparatus especially designed for metrologic purposes.

Also for conventional laboratory apparatus in free running operation, the uncertainty in the frequency reproducibility is of the order of a few parts in 10^7 . For instance the measurements of the frequency of the $496 \mu\text{m}$ emission from CH_3F , performed in three different laboratories with three different experimental apparatus agree within 200 KHz. Hence frequency measurements can significantly improve the knowledge of the active

molecule structure (high rotational quantum numbers are involved, enhancing physical effects which in microwave spectroscopy are too small to be detected).

FIR frequency measurements were made possible since the development of nonlinear devices capable of harmonic generation and of sum- or difference- frequency generation from two or more incident radiations. Metal-insulator-metal (MIM) diodes, Schottky barrier diodes, and Josephson junctions (JJ) have been used.

A first possibility is the measurement by mixing the FIR laser radiation with a reference signal in the microwaves.

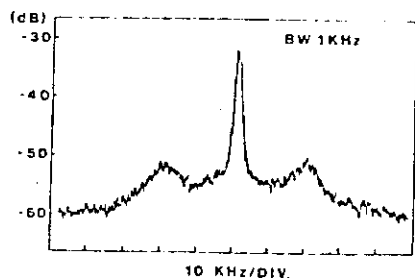


FIG 16

For instance in Fig 16 it is shown the beat note obtained by mixing a CH_3F FIR laser with the 8th harmonic of a 75 GHz Klystron in a point contact MIM diode. The full width of the signal at -30 dB is about 2 KHz. The Klystron is phase locked to a 5 MHz low noise crystal oscillator. The frequency of the $\text{CH}_3\text{F } \nu_3(J, K):12,2 \rightarrow 11,2$ transition was determined to be 604297.46 MHz (Bove et al 1977). Once included in a least-squares fit of all the microwave, laser Stark and infrared data related to the same band, accurate values for the rovibrational constants can be obtained. An example of the

improvement is in the table (from Arimondo and Inguscio, 1979). It is worth noting that CH_3F is a good reference molecule for microwave and infrared spectroscopy.

	Present Result	Previous Result	Source	Units
ν_0	31 436 560.7 (17)	31 436 559.7 (18)	3	MHz
$A''-A''-B''+B''$	43.96(52)	44.49(63)	3	MHz
$D_K''-D_K''$	-9F (16)	-145 (74)	3	KHz
H''	25 197.545(13)	25 197.57(3)	3	MHz
L''	57.41(26)	55.5(12)	3	KHz
$L''-L''$	516.5 (42)	575 (163)	3	KHz
$L''-B''$	-0.4 (3)	-	3	Hz
$L''-K''$	0	-	-	-
$L''-K''$	0	-	-	-
B''	25 536.1464(11)	25 536.1466(15*)	1	MHz
D_K''	60.20(11)	59.87(20*)	1	KHz
$D_K''K''$	439.90(34)	440.27(40*)	1	KHz
B''_J	-0.5 (8)	-10.7 (40*)	1	Hz
B''_JK''	8 (10)	20 (10*)	1	Hz
B''_KJ	33 (17)	32 (20*)	1	Hz
C_K''	0	0.318	20	-

Another, more general, technique to measure FIR frequencies is that of synthesizing an appropriate local oscillator signal from the difference frequency of two Lamb dip fluorescence stabilized CO_2 lasers. In this case radiations from two CO_2 lasers, oscillating on the proper lines, and from the FIR lasers are sent on the MIM diode. The beat note from the diode is detected on the spectrum analyzer. In some cases two CO_2 laser lines are not enough to synthesize a frequency close enough to the one of the FIR laser, and also power from a microwave source is sent to the diode. Measurements in Fig 17 report the signal to noise ratio (in dB) of the beat note as a function of the resistance

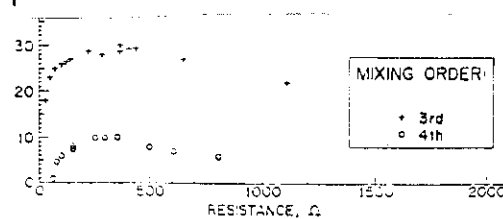


FIG 17

of the point contact diode. The 3rd order data refer to the measurement of the 170.6 μm line from a CH_3OH laser heterodyned with 10 P(36) and 10 R(40) CO_2 laser lines. In the 4th order experiment the frequency from a 127.03 μm $^{13}\text{CD}_3\text{OH}$ laser is measured by beating with the 10 R(22) and 9 P(10) CO_2 laser lines and a phase locked Klystron at 9491.170 MHz.

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