FREQUENCY MODULATION (FM) SPECTROSCOPY

Diploma paper
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ABSTRACT

This diploma paper describes and investigates applications of a recently developed method for measurements of weak absorption, named frequency modulation (FM) spectroscopy. Using an electro-optic modulator, sidebands are generated on a laser beam. These sidebands, equal in magnitude and opposite in phase, beat against the laser carrier and if not otherwise influenced, they cancel perfectly. When the laser frequency is scanned across a spectral absorption feature differential absorption of the sidebands occurs, which gives rise to a beat signal, detectable with standard radio frequency techniques. The detection shot noise limit, in general about 10^{-7} in relative absorption, can be reached. This limit has been reported by some other groups.

A sensitivity of 10^{-4} with 50 MHz modulation and 10^{-3} with 8.4 GHz modulation is presented.

This diploma paper does not extend the current knowledge concerning FM spectroscopy. It gives a short description of the theory and describes how it would be experimentally possible to reach the shot noise limit. It may thus be a useful starting point for further research on FM spectroscopy.

THEORETICAL PART

1. INTRODUCTION

Frequency modulation (FM) in connection with lasers is used in two main applications; for laser frequency control and as a method of optical heterodyne spectroscopy. In this presentation only the latter, frequency modulation spectroscopy, here abbreviated FMS, will be discussed.

FMS was first demonstrated by Bjorklund in 1980,[1] using a single-mode cw dye laser. It constitutes a high-resolution, high-sensitivity method of optical spectroscopy. It has since been used for high-resolution absorption spectroscopy with quantum-limited detection sensitivity. (See, e.g., Refs. 2 and 3). Using an electro-optic phase modulator (EOM) two frequency sidebands are generated on a laser beam. The sidebands are separated from the carrier by a frequency distance equal to the modulating radio frequency (RF) and are equal in magnitude and opposite in phase. This offers the possibility of zero background phase-sensitive detection. The zero background can be viewed as originating from a radio frequency signal arising from the beating of the upper sideband against the carrier, perfectly canceling the radio frequency signal arising from the lower sideband beating against the carrier. In practice, however, this perfect balance is disturbed by birefringence variations of the phase modulator and étalon effects due to the crystal and reflective surfaces along the propagation path after the modulator, resulting in a spurious residual amplitude modulation (RAM). RAM sets a sensitivity limit on FMS, but using various methods it is possible to totally suppress the RAM and achieve the quantum limit.[2,3]

When the frequency-modulated laser beam is obstructed by a narrow-band spectral feature, the sidebands will be differentially absorbed, giving rise to an amplitude modulation, beating at the RF modulation frequency. Using a photodiode, reacting fast enough for the radio frequency, the beat notes can be detected and through conventional radio frequency techniques, be demodulated to a signal directly proportional to the differential absorption.

An important feature of FMS is that the detection frequency can be moved to a region outside the laser linewidth where the laser RF noise is negligible. FMS will be quite insensitive to intensity fluctuations of the laser.

FMS should be distinguished from wavelength modulation spectroscopy (WMS), demonstrated by Tang and co-workers.[4] In WMS higher orders of sidebands are generated and the modulation frequency is considerably smaller. This means that an absorbing feature is probed by more then one sideband at one time. In FMS, practically only an upper and a lower sideband are generated and the modulation frequency is at least of the order of the width of the spectral feature of interest. This can be referred to as the FM condition. The higher modulation frequency is also a condition for achieving the highest sensitivity or the quantum noise limit.

Additional explanation of the experimental performance and the components used is given in the Appendix at the end of this paper.

2. FREQUENCY MODULATION SPECTROSCOPY (FMS)

2.1 Basic theory of FMS

The theory of FMS was first presented by Bjorklund.[1] We will follow his presentation when describing the basic theory of frequency modulation spectroscopy.

A single-frequency laser provides radiation at the optical carrier frequency ω_c. Using complex notation the electric field is described by

$$E_1(t) = E_0 e^{i\omega_c t} \tag{2.1}$$

After passing through the electro-optic modulator (EOM) (Fig. 1), which is driven by a sinusodially varying radio frequency (RF) field at frequency ω_m, the laser frequency spectrum is described by

$$\begin{aligned} E_2(t) &= E_0 e^{i(\omega_c t + M\sin\omega_m t)} \\ &= E_0 \sum_{n=-\infty}^{n=\infty} J_n(M) e^{i(\omega_c + n\omega_m)t}, \end{aligned} \tag{2.2}$$

where M is the modulation index and J_n are Bessel functions of order n. This is the case for a pure FM spectrum. It will be assumed that the modulation index is small compared with unity, then neglecting sidebands of second order and higher. The FM spectrum will have a strong carrier ω_c with a weaker sideband on each side at frequencies $\omega_c \pm \omega_m$, and it is described by

$$E_2(t) = E_0\left(\frac{M}{2}e^{i(\omega_c + \omega_m)t} + e^{i\omega_c t} - \frac{M}{2}e^{i(\omega_c - \omega_m)t}\right). \tag{2.3}$$

The beam is now passed through a sample (Fig. 1) of length L, characterized by an intensity absorption coefficient α and a refractive index η, both functions of the optical frequency.

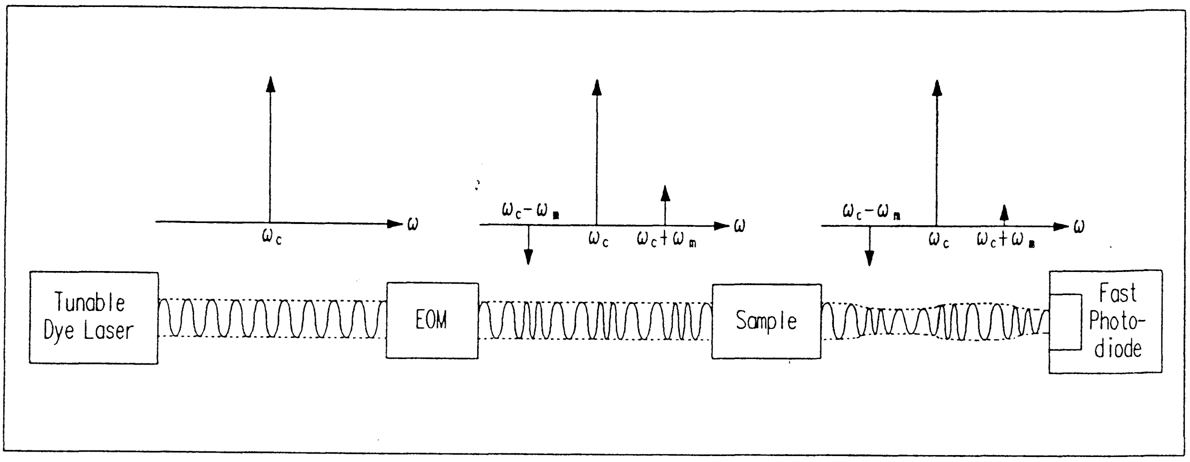


Fig. 1
Using an electro-optic modulator (EOM) sidebands are generated on a single mode laser beam with carrier frequency ω_c. The sidebands are equal in magnitude and opposite in phase and cancel perfectly. The FM beam is passed through an absorbing sample, which affects the perfect balance and gives rise to a sensitively detectable beat signal.

Bjorklund defines the complex quantities

$$T_n = \bar{T}e^{-\delta_n - i\phi_n}, \tag{2.4}$$

where

$$\delta_n = \alpha_n \frac{L}{2}, \qquad \phi_n = \eta_n L\frac{(\omega_c + n\omega_m)}{c}. \tag{2.5}$$

The subscript n = (-1,0,1) specifies the values at $\omega_c - \omega_m$, ω_c and $\omega_c + \omega_m$, respectively. Thus, the amplitude attenuation and the optical phase shift experienced by each frequency component are denoted by δ_n and ϕ_n, respectively. In Eq. (2.4) a possible broadband background is also taken into account

$$\bar{T} = e^{-\bar{\delta}_n - i\bar{\phi}_n}, \tag{2.6}$$

where the background amplitude attenuation $\bar{\delta}$ and the background optical phase shift $\bar{\phi}$ do not change on the scale of the FM spectrum in Eq. (2.3).[2]

It is now possible to describe the transmitted field

$$E_3(t) = E_0\left(T_0 e^{i\omega_c t} + T_1\frac{M}{2}e^{i(\omega_c+\omega_m)t} - T_{-1}\frac{M}{2}e^{i(\omega_c-\omega_m)t}\right). \quad (2.7)$$

The intensity $I_3(t)$ impinging on the photodiode is given by

$$\begin{aligned} I_3(t) &= \frac{c|E_3(t)|^2}{8\pi} \\ &= \frac{cE_0^2}{8\pi} e^{-2\delta_0}\Big[1 + \Big(e^{\delta_0-\delta_1}\cos(\phi_1-\phi_0) \\ &\quad - e^{\delta_0-\delta_{-1}}\cos(\phi_0-\phi_{-1})\Big)M\cos\omega_m t \quad (2.8) \\ &\quad + \Big(e^{\delta_0-\delta_1}\sin(\phi_1-\phi_0) \\ &\quad - e^{\delta_0-\delta_{-1}}\sin(\phi_0-\phi_{-1})\Big)M\sin\omega_m t\Big], \end{aligned}$$

where terms of the order M^2 have been dropped. The electrical signal of the photodiode will thus contain a beat signal with a modulation frequency ω_m if all the δ_n are not equal or all the ϕ_n are not equal. If $|\delta_0 - \delta_1|$, $|\delta_0 - \delta_{-1}|$, $|\phi_0 - \phi_1|$ and $|\phi_0 - \phi_{-1}|$ are small compared with unity, Eq. (2.8) simplifies to

$$\begin{aligned} I_3(t) = \frac{cE_0^2}{8\pi} e^{-2\delta_0}\Big(1 &+ (\delta_{-1} - \delta_1)M\cos\omega_m t \\ &+ (\phi_1 + \phi_{-1} - 2\phi_0)M\sin\omega_m t\Big). \end{aligned} \quad (2.9)$$

The quadrature component ($\cos\omega_m t$) of the beat signal is proportional to the difference in loss experienced by the upper and lower sidebands. If ω_m is small compared with the width of the spectral feature of interest (Fig. 4), the quadrature term is proportional to the derivative of the absorption. On the other hand, if ω_m is large and only one sideband probes the spectral feature at a time, we have the

FM spectroscopy condition and the absorption of the spectral feature can be directly measured. The in-phase component ($\sin\omega_m t$) of the beat signal is proportional to the difference between the phase shift experienced by the carrier and the average of the phase shift experienced by the sidebands. The quadrature component is proportional to the second derivative of the dispersion.

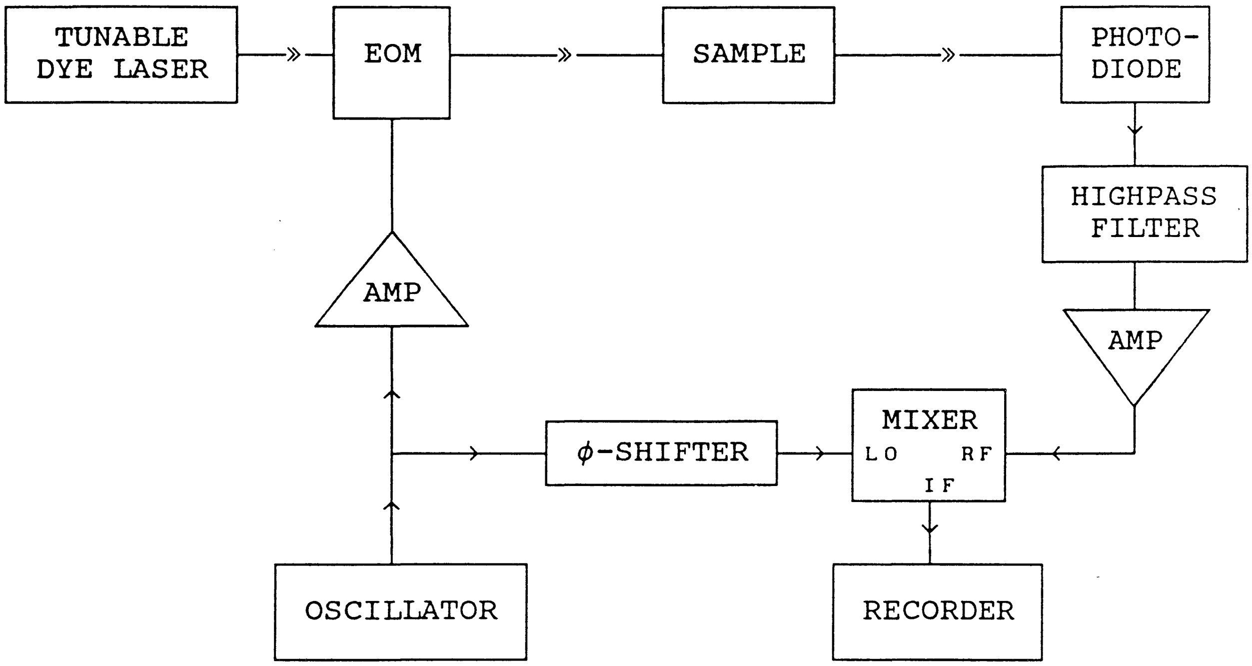


Fig. 2.
The basic set-up for FMS.

When ω_m is large compared with the width of the absorbing feature (Fig. 3) and if the laser ω_c is scanned and only the upper side band is probing, the losses and phase shifts experienced by the carrier and the lower sideband remain essentially constant. We can then write $\delta_{-1} = \delta_0 = \bar{\delta}$ and $\phi_{-1} = \phi_0 = \bar{\phi}$, where $\bar{\delta}$ and $\bar{\phi}$ are the constant background loss and phase shift, respectively. Bjorklund introduces the quantities $\Delta\delta = \delta_1 - \bar{\delta}$ and $\Delta\phi = \phi_1 - \bar{\phi}$ as the deviation from the background values, caused by the spectral feature.

The final simplified equation is

$$I_3(t) = \frac{cE_0^2}{8\pi} e^{-2\bar{\delta}}(1 - \Delta\delta M\cos\omega_m t + \Delta\phi M\sin\omega_m t). \qquad (2.10)$$

When an absorbing feature causes the differences $\Delta\delta$ and $\Delta\phi$ to be nonzero, the sideband amplitudes are unbalanced and the beam emerging from the sample will be amplitude modulated. Note, while the EOM FM modulates the laser, the signal is AM detected. Note too, that an absorbing feature can be probed

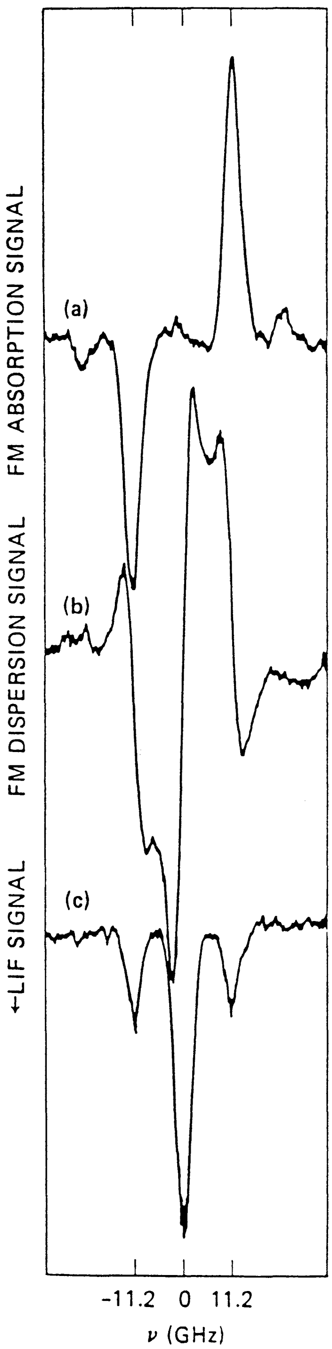


Fig. 3
a) FM absorption, b) FM dispersion, and c) D-line laser-induced fluorecence signals produced by the Na D_2 resonance line line at at 5890Å. [From Ref. 5]

by one sideband either by scanning the laser frequency ω_c or by tuning the radio frequency ω_m. However, in the case of high-frequency modulation of a resonant cavity modulator (see Sect. 7.1), it is not advisable to tune the radio frequency.

Some favorable qualities of FMS now come to light. When no absorbing sample is present the mixer output will be zero. This zero background signal can be viewed as being due to a perfect cancellation of the two RF signals arising from the upper and lower sidebands beating against the carrier. As will be seen later, the optical equipment along the laser beam path, will disturb this perfect cancellation and give rise to an amplitude modulation, even when no sample is present. This disturbance is called residual amplitude modulation (RAM) and it can be suppressed by various methods. Another advantage with FMS is that the modulation frequency ω_m, can be chosen such that the detection signal frequency is well beyond the maximum of the laser amplitude noise[6] (f^{-1}) and the detection shot noise limit can be reached.

With phase-sensitive detection electronics (Fig. 2) it is possible to choose the signal, absorption or dispersion, to be detected. The electrical signal from the high-speed photodetector will contain beat notes at the modulation frequency, where the modulation depth is proportional to the absorption and/or the dispersion. By demodulating the beat signal through standard radio frequency techniques, the absorptive or dispersive FM spectrum is obtained at the mixer intermediate-frequency (IF) output. In this way, the phase relation between the LO and RF port of the mixer is adjusted. This is easily done by varying the cable lengths, moving the photodetector or by a special device, a phase-shifter, which in the microwave region (in our case) is a simple line-stretcher. However, in practice the phase is adjusted to yield an intermediate of both in order to achieve maximum detectable signal strength. It may be instructive to compare this performance with the commonly used lock-in technique.

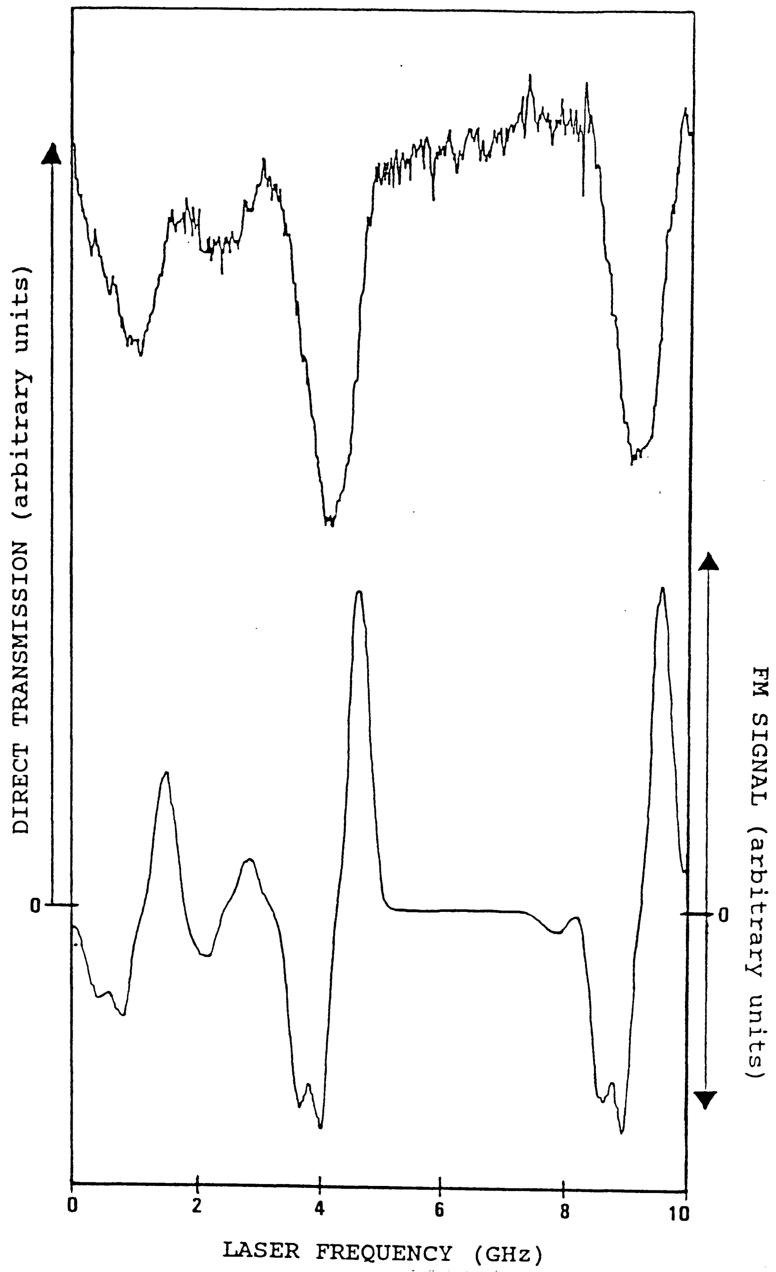


Fig. 4
Comparison of direct and FM absorption signals of I_2, recorded with a single mode cw dye laser and 50 MHz frequency modulation. The zero level is indicated.

The mixer output will be proportional to

$$\hat{F}(\lambda) = I_0 e^{-2\bar{\delta}} \times \begin{cases} -\Delta\delta M & \text{(quadrature)} \\ \Delta\phi M & \text{(in phase)} \end{cases} \qquad (2.11)$$

where the hat on $\hat{F}(\lambda)$ denotes either the quadrature or in-phase Fourier component of $I_3(t)$ in Eq. (2.10) and the wavelength $\lambda = \lambda_c = 2\pi c/\omega_c$ of the carrier is used instead of the frequency ω_c. The abbreviation F is introduced, as will be seen later, to distinguish between the attractive pure FM case, where the signal arises from a zero background and a case with a time dependent background variation, caused by RAM.

Fig. 3 illustrates the different lineshapes of the absorption and dispersion signals. For a detailed lineshape analysis see Ref. 7. Note, that the modulation frequency is large compared with the width of the absorbing profile; only one sideband is probing at the time.

2.2 Noise characterization of FM spectroscopy

In Ref. 7 Bjorklund *et al.* give a detailed signal-to-noise analysis of FMS. With the simplifications of a pure absorptive spectral feature, probed with an isolated sideband and no background absorption present ($\Delta\Phi = \bar{\delta} = 0$), they derive the minimum detectable absorption $\Delta\delta_{min}$, with unity signal-to-noise ratio (SNR).

The optical power $P_3(t)$ incident on a photo-conductor of area A is given by Eq. (2.10)

$$P_3(t) = P_0(1 - \Delta\delta M\cos\omega_m t), \qquad (2.12)$$

here $P_3(t)=AI_3(t)$ and $P_0 = \frac{AcE_0^2}{8\pi}$ is the total laser power. The two dominant sources of noise in FMS are thermal noise $\bar{i}^2_{TN}$ (rms noise power) and shot noise $\bar{i}^2_{SN}$ (rms noise power), generated at the photo-detector. The rms power of the beat signal is $\bar{i}^2_s(t)$. The signal-to-noise (S/N) ratio will be

$$\frac{S}{N} = \frac{\overline{i_S^2(t)}}{\overline{i_{SN}^2} + \overline{i_{TH}^2}} = \frac{\frac{1}{2}g^2e^2\eta^2\left(\frac{P_0}{\hbar\omega_c}\right)^2\Delta\delta^2M^2}{2g^2e^2\eta\left(\frac{P_0}{\hbar\omega_c}\right)\Delta f + \left(\frac{4kT}{R}\right)\Delta f}, \qquad (2.13)$$

where η is the quantum efficiency and g is the gain of the photo-detector, Δf is the bandwidth of the detection electronics, k is Boltzman's constant, T is the temperature in K, e is the electronic charge and R is the detection electronic's input impedance.

When $P_0 \geq P_{0min} = \frac{2kT\hbar\omega_c}{\eta g^2 e^2 R}$, shot noise dominates over thermal noise, and we have the shot-noise-limited condition

$$\frac{S}{N} = \frac{\eta\left(\frac{P_0}{\hbar\omega_c}\right)\Delta\delta^2M^2}{4\Delta f}, \qquad (2.14)$$

and minimum detection absorption $\Delta\delta_{min}$, with unity S/N will be given by

$$\Delta\delta_{min} = \frac{2}{M}\left(\frac{\Delta f}{\eta\left(\frac{P_0}{\hbar\omega_c}\right)}\right)^{1/2} \qquad (2.15)$$

Thus, it is advantageous to increase the total laser power P_0, to keep the bandwidth of the detection electronics low, to increase the modulation index M and have a quantum efficiency near unity. The main problems in actually achieving quantum-limited performance in FMS will be discussed in following sections.

2.3 Residual amplitude modulation (RAM), limitation of FMS

The sensitivity limit of FMS (as it has been described so far) is set by residual amplitude modulation (RAM), caused by imperfect cancellation of the sidebands even when no sample is present. RAM noise appears in laser scans as fringes (multipass-effect, Fig. 5 and 18) and/or as a sloping baseline (single-pass effect, Fig. 6). It limits the sensitivity to $\Delta\delta_{min} \sim 10^{-5} - 10^{-2}$.[2] Using various methods, it is possible to subtract these disturbances and reach the shot noise limit.

The multi-pass effect, due to etalon fringes, is effectively reduced by placing every parallel optical surfaces (including the EOM) at an angle to the direction of beam propagation. The single-pass effect has not yet, to the author's knowledge, been fully understood. It makes it appearance as a sloping baseline and is not easily suppressed.

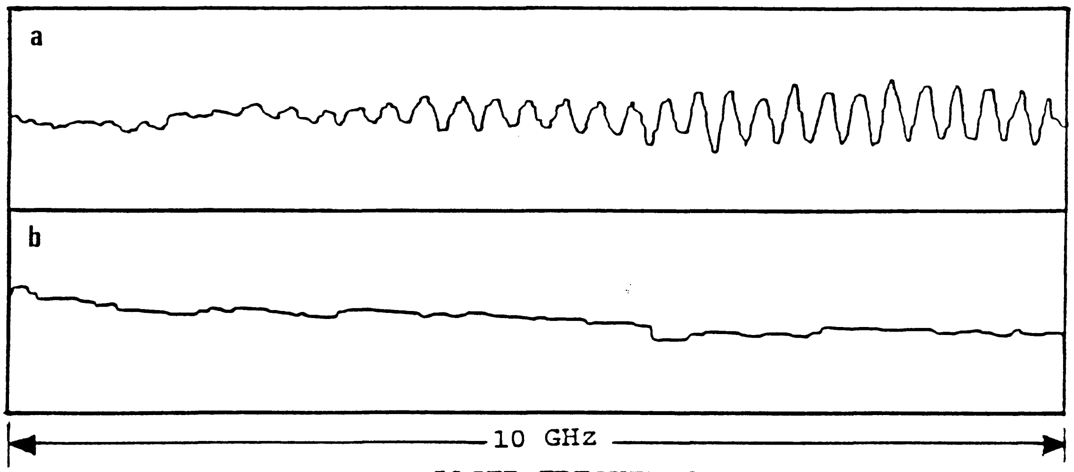


Fig. 5 a and b

Recordings of the FM signal without absorption cell.

a) *Fringes originating from etalon effects in the electro-optic modulator.*

b) *The result of tilting the modulator by a few degrees.*

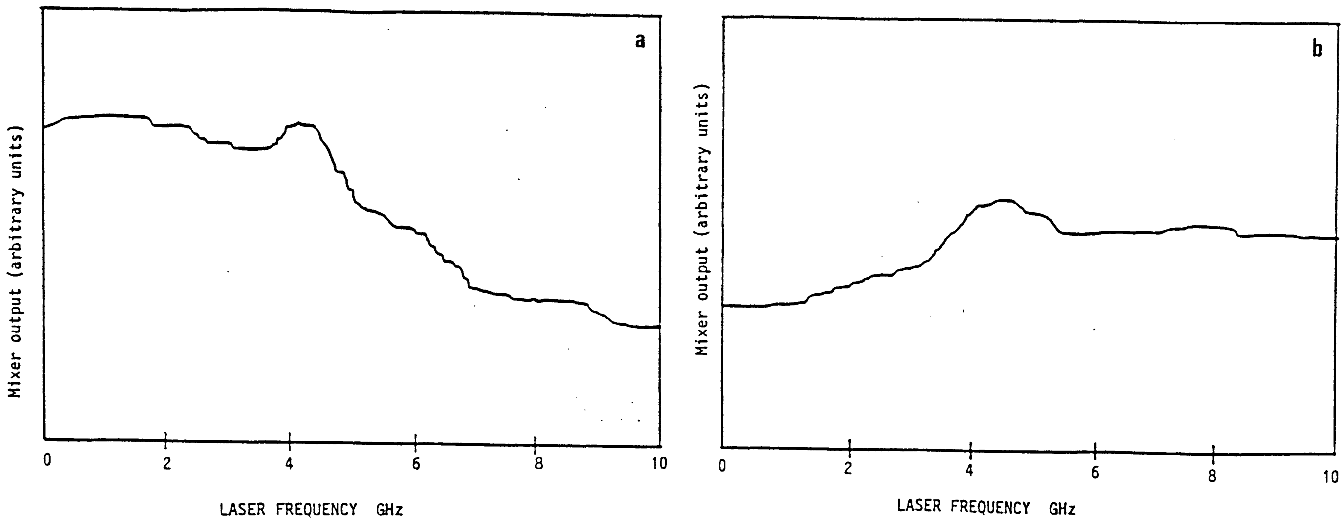


Fig. 6 a and b
FM signal of the Na D_2-line demonstrating the sensitivity limitation due to single-pass RAM (sloping baseline), at two different temperatures: a) 39 °C and b) 37 °C. See also Fig. 19. The recordings were made with a single-mode cw dye laser and 50 MHz frequency modulation.

2.4 Theory of residual amplitude modulation

In Ref. 2 a detailed theoretical description of RAM by Gehrtz *et al.* can be found. We will give a short summary following their notation. As has already been noted, RAM makes its appearance, experimentally, as an intensity modulation of the laser beam, even when no sample is present. The intensity after the EOM, in the case of pure FMS, is

$$I_2^F(t) = \frac{cE_0^2}{8\pi} = I_0, \tag{2.16}$$

where the superscript F is introduced to denote the pure case. If a RAM disturbance is introduced (in the EOM), the intensity will be time dependent

$$I_2(t) = I_0[1 + 2R\sin(\omega_m t + \Psi)], \quad (2.17)$$

where R denotes the RAM strength and Ψ the RAM phase shift (Cf. Eq. (2.2)). Both R and Ψ are functions of the laser wavelength λ and the modulation frequency ω_m, in general[2]. Modifying Eq. (2.2) to describe the effects of RAM on the FM spectra, thelaser frequencyspectrum will be

$$E_2(t) = E_2^F(t)\left(1 + R\sin(\omega_m t + \Psi)\right). \quad (2.18)$$

Similar to the description in the case of pure FMS, the same simplifying assumptions are made; FM index M and RAM index R are assumed to be small compared with unity and only terms depending on the fundamental of the modulation frequency ω_m are retained.

The RAM-contaminated intensity emerging from the sample is

$$I(t) = F(t) + e^{-2\bar{\delta}}B(t), \quad (2.19)$$

where F(t) is the pure FM component, Eq. (2.10), and B(t) is the background signal, arising even when no sample is present:

$$\begin{aligned} B(t) &= I_0(2R\sin\Psi\cos\omega_m t + 2R\cos\Psi\sin\omega_m t) \\ &= I_2 2R\sin(\omega_m t + \Psi). \end{aligned} \quad (2.20)$$

When no sample (ns) is present, the intensity is

$$I_{ns}(t) = I_0 + B(t). \quad (2.21)$$

If the laser carrier frequency is scanned, there will be a signal at the mixer output, which is proportional to

$$M(\lambda) = \hat{F}(\lambda) + e^{-2\bar{\delta}}\hat{B}(\lambda) \tag{2.22}$$

and

$$M_{ns}(\lambda) = \hat{B}(\lambda) \neq 0. \tag{2.23}$$

$\hat{F}(\lambda)$ denotes the pure FM signal that would be recorded if no RAM was present, Eq. (2.11). The hat on the background signal $\hat{B}(\lambda)$ denotes the Fourier component of the corresponding time-dependent signal B(t), Eq. (2.20). It is dependent on the LO phase selection

$$\hat{B}(\lambda) = I_0 \times \begin{cases} 2R\sin\Psi & \text{(quadrature)} \\ 2R\cos\Psi & \text{(in phase).} \end{cases} \tag{2.24}$$

The wavelength dependence of the background signal $B(\lambda)$ can originate in the RAM index $R(\lambda)$ as well as in the RAM phase shift $\Psi(\lambda)$.[2]

2.5 Suppressing residual amplitude modulation

Different methods have been used to achieve shot-noise-limited sensitivity in FMS. The first report, to the author's knowledge, was published by Levenson *et al.*,[8] who used FMS to achieve a shot-noise-limited sensitivity, when detecting the small gain induced by the stimulated Raman effect in deuterium. Further, Gehrtz *et al.*[2] presented a double-beam, single-detector technique and Wong and Hall[3] demonstrated a servo control technique. Other methods have been demonstrated, which reduce the RAM to near the quantum limit. Two methods will shortly be presented here.

2.5.1 Double-beam technique

A double-beam, single-detector technique was used in Ref. 2 to efficiently suppress the RAM and perform quantum-limited FMS (Fig. 7). Using two beam splitters (BS), the frequency-modulated laser beam is divided into a sample and a reference beam and subsequently recombined at the photodiode. A chopper (Ch) is used to chop both beams synchronously but 180° out of phase. It is important, in order for the RAM nulling to work at arbitrary modulation frequencies, that the RF phases in the two beams are equal. This is accomplished by keeping the beam path lengths equal.

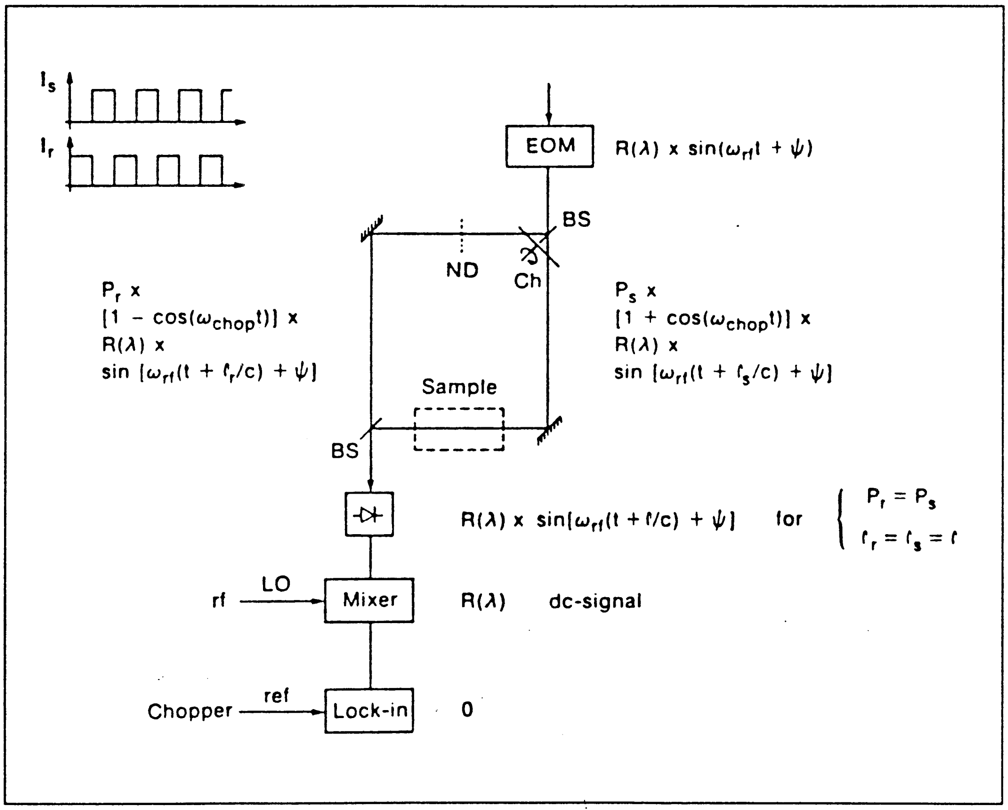


Fig. 7
Schematic illustrating the double-beam RAM-nulling technique. [From Ref. 2]

The reference beam will carry the no-sample background signal (Eq. 2.21) and the sample beam carries the complete signal (Eq. 2.19). The mixer output will be proportional to

$$M_{tot}(\lambda) = P_s\hat{F}(\lambda)(1 + \cos\omega_{chop}t) + 2P_r\hat{B}(\lambda), \qquad (2.25)$$

where P_s and P_r denote the relative light power in the two beams; the FM signal $\hat{F}(\lambda)$ and the background signal $\hat{B}(\lambda)$ have been introduced in Sect. 2.4. Demodulating the mixer output with a lock-in amplifier the total signal will be proportional to $P_s\hat{F}(\lambda)$ and in the absence of any absorbing sample the attractive zero background is obtained.

2.5.2 Servo control of amplitude modulation

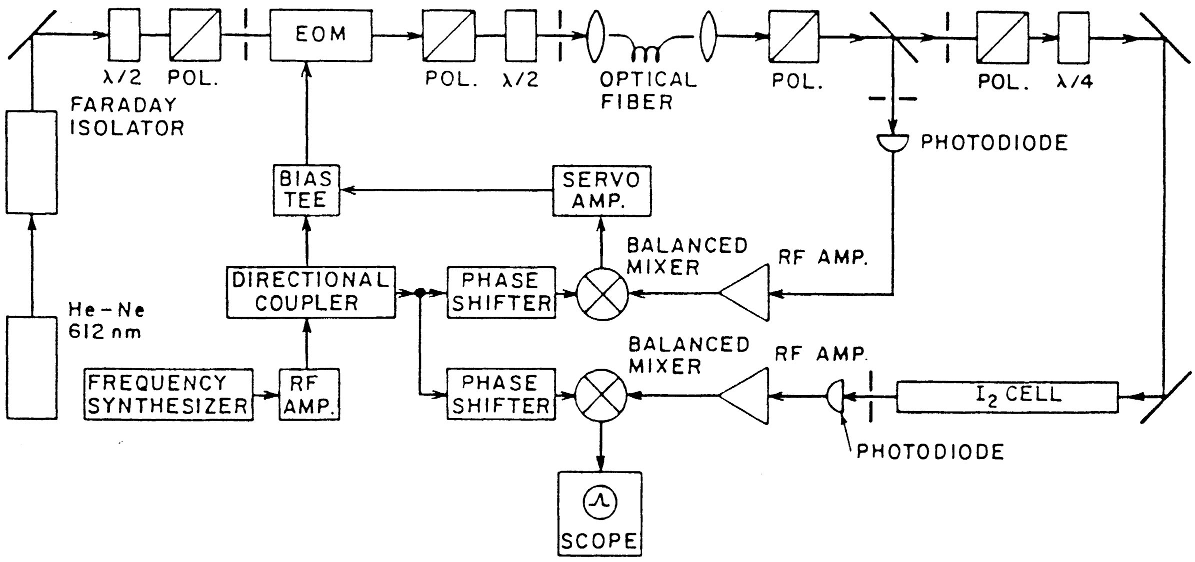


Fig. 8
Schematic of an experimental set-up for RAM reduction. The laser AM reduction is achieved by monitoring the in-phase signal and feeding the error signal into a servo amplifier, which corrects the birefringence of the crystal. [From Ref. 3]

3. TWO-TONE FREQUENCY MODULATION SPECTROSCOPY

Two-tone frequency modulation spectroscopy (TTFMS) is a method of performing high-frequency modulation FMS while using low-frequency detection. With the use of broadband multimode lasers, requirements of higher modulation frequency are raised, as the increased laser bandwidth introduces more laser noise into the FM signal. Additionally, since the FM signal is proportional to the differential absorption of the upper and the lower sidebands, FM detection of (atmospheric) pressure-broadened spectral lines, which have widths of the order of 10 GHz, requires a modulation frequency of the same order. Since, in ordinary FM spectroscopy, the signal is detected at the modulation frequency ω_m, a photo-detector with a bandwidth response higher than ω_m is necessary. High-speed photodiodes have the disadvantage of being manufactured by only a few companies, and have a small active area which is easily damaged, apart from being very expensive. The 10 GHz PIN diode we purchased cost 22000 SEK and has an active area of 0.0036 mm^2. The way of overcoming these problems is to use two-tone modulation. With this technique, the laser is simultaneously modulated by two closely separated frequencies which generate sideband pairs on the laser beam (Figs. 9 and 10). This is accomplished by using two oscillators and an additional mixer to provide the sum and difference of the two oscillator frequencies, one high (GHz) and one low (MHz). Using the heterodyne technique, an FM absorption signal can be detected at the difference frequency (sideband pair separation), which will be twice the low-oscillator frequency, and the advantages of high-frequency modulation are still retained.

3.1 Theory of two-tone FMS

The theoretical description of two-tone FMS does not differ much from that of single-tone FMS. We will give a short presentation. For a detailed analysis see Ref 9.

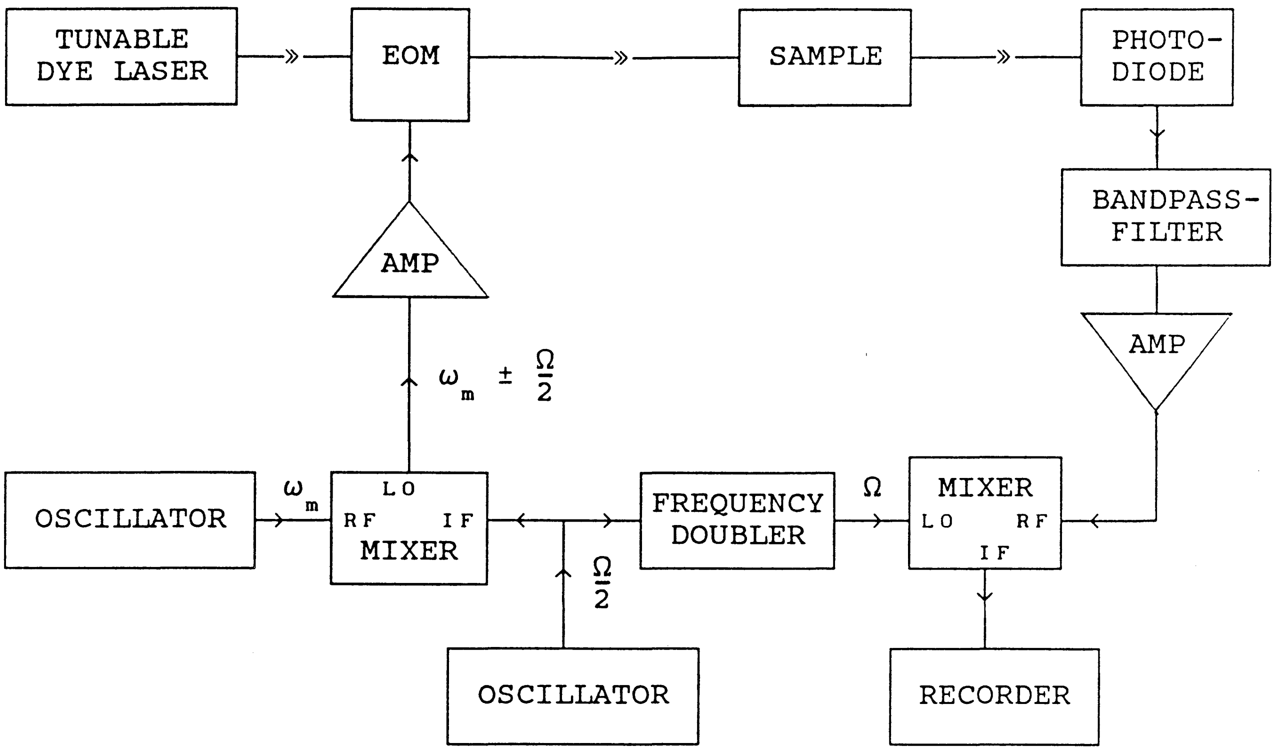


Fig. 9
The basic set-up for two-tone FMS.

The laser frequency spectrum after modulation by the two frequencies ω_1 and ω_2 is

$$\begin{aligned} E_2(t) &= E_0 e^{i\omega_c t} \sum_{n=-\infty}^{n=\infty} J_n(M_1) e^{in\omega_1 t} \sum_{m=-\infty}^{m=\infty} J_m(M_2) e^{im\omega_2 t} \\ &= E_0 e^{i\omega_c t} \sum_{n,m} J_n(M_1) J_m(M_2) e^{i(n\omega_1 + m\omega_2)t}. \end{aligned} \tag{3.1}$$

The following simplifying approximations will be made: $M_1 = M_2 = M$ and $M < 1$, only the zeroth- and first-order side-bands are considered to be significant (Fig. 10) and the two modulation frequencies are expressed as

$$\begin{aligned} \omega_1 &= \omega_m + \frac{\Omega}{2} \\ \omega_2 &= \omega_m - \frac{\Omega}{2}, \end{aligned} \tag{3.2}$$

where $\Omega = \omega_1 - \omega_2$ will be the detection frequency, which is twice the low-oscillator modulation frequency.

The modulated laser field after interaction with a sample is given by

$$E_3(t) = E_0\Big[T_0\Big(e^{i\omega_c t} - \frac{M^2}{4}e^{i(\omega_c+\Omega)t} - \frac{M^2}{4}e^{i(\omega_c-\Omega)t}\Big) + T_{+1}\Big(\frac{M}{2}e^{i(\omega_c+\omega_m+\frac{\Omega}{2})t} + \frac{M}{2}e^{i(\omega_c+\omega_m-\frac{\Omega}{2})t}\Big) + T_{-1}\Big(-\frac{M}{2}e^{i(\omega_c-\omega_m-\frac{\Omega}{2})t} - \frac{M}{2}e^{i(\omega_c-\omega_m+\frac{\Omega}{2})t}\Big)\Big]. \quad (3.2)$$

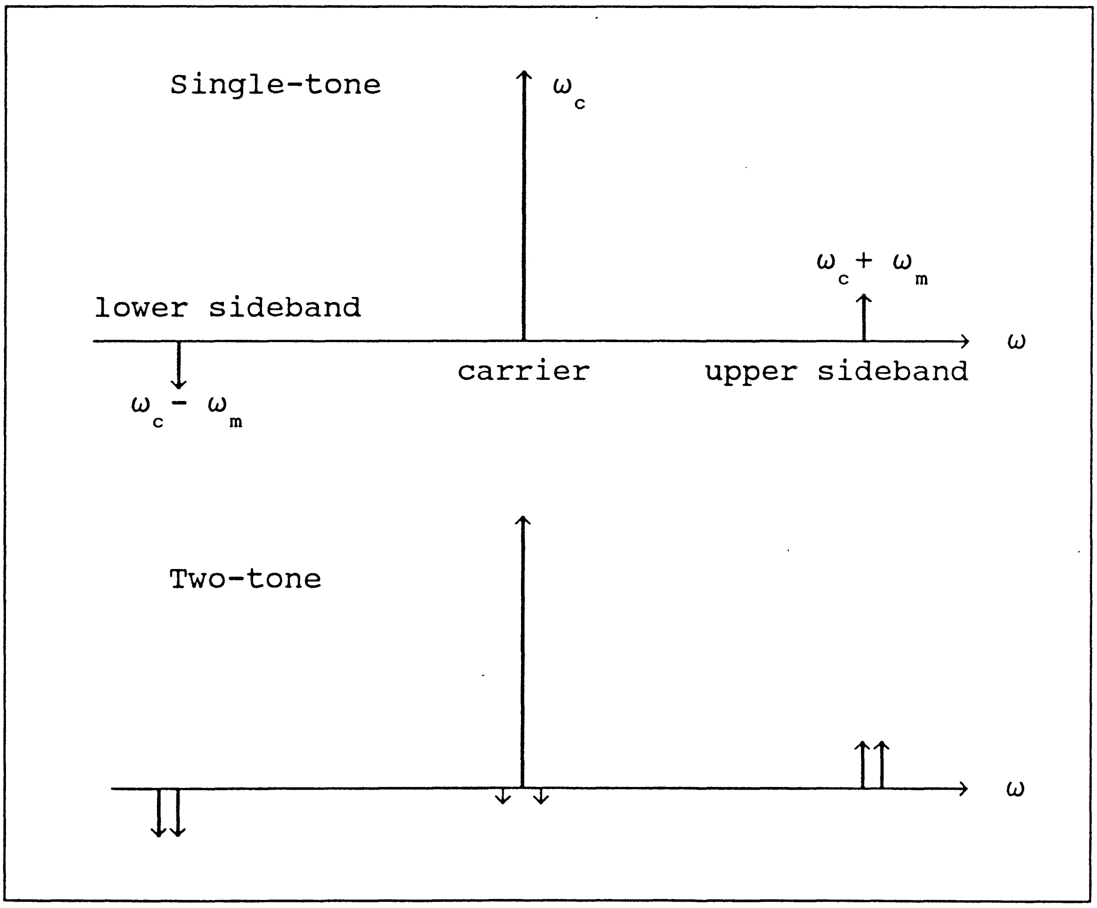


Fig. 10
Spectral components of the laser after single-tone and two-tone modulation, respectively. Note that no consideration has been taken of proportionality.

The intensity impinging on the photodiode is given by $I_3(t) = \frac{c|E_3(t)|^2}{8\pi}$. With heterodyne detection electronics, a mixer with the LO-port reference signal from the low-frequency oscillator frequency-doubled, the interesting beat-signal at Ω is extracted from the photodiode output current (Fig. 9). There are other frequencies, but these are then rejected. Thus the intensity (at Ω) can be written

$$I_3(\text{at } \Omega) = \frac{cE_0^2M^2}{16\pi} (e^{-2\delta_1} + e^{-2\delta_{-1}} - 2e^{-2\delta_o})\cos\Omega t \qquad (3.4)$$

which for small absorptions simplifies to

$$I_3(\text{at } \Omega) = \frac{cE_0^2M^2}{8\pi} (2\delta_o - \delta_1 - \delta_{-1})\cos\Omega t. \qquad (3.5)$$

There is no dispersion signal and the absorption signal arises from the difference between the absorption of the carrier and the sum of the absorption of the sidebands. This could be thought of as a second derivative. Note that the signal is proportional to M^2 instead of, as earlier, M. The attractive feature of conventional FM detection is preserved, that is, the signal arises from a zero background.

Fig. 11 illustrates the lineshape of two-tone signals. Consider Eq. (3.5) again. When the laser is tuned across an absorbing line, first the upper sideband pair will be absorbed, causing δ_1 to become nonzero, resulting in a net negative signal. Scanning the laser further will cause the central group of sidebands to be absorbed and $2\delta_0$ to become nonzero, in turn causing a net positive signal, twice as high as the foregoing and the subsequent net negative signal, arising when the lower sideband pair in turn is absorbed and δ_{-1} becomes nonzero.

The earlier derived expression for the minimum differential absorption that can be detected (Eq. 2.15) also holds for two-tone FMS detection, except that M is replaced by M^2 and $\Delta\delta_{min}$ signifying $\delta_0 - \delta_1$ instead of $\delta_1 - \delta_{-1}$. The RAM still makes its appearance and the shot-noise limit difficult to reach.[9]

When a multimode dye laser beam is two-tone modulated, a detection frequency of a few MHz will be well within the envelope of the laser. Care has to be taken when choosing detection frequency. If the frequency is not coincident with the laser mode spacing or integral multiples of it and is outside the width of a single mode, there should not be problems with laser noise in the signal.[9]

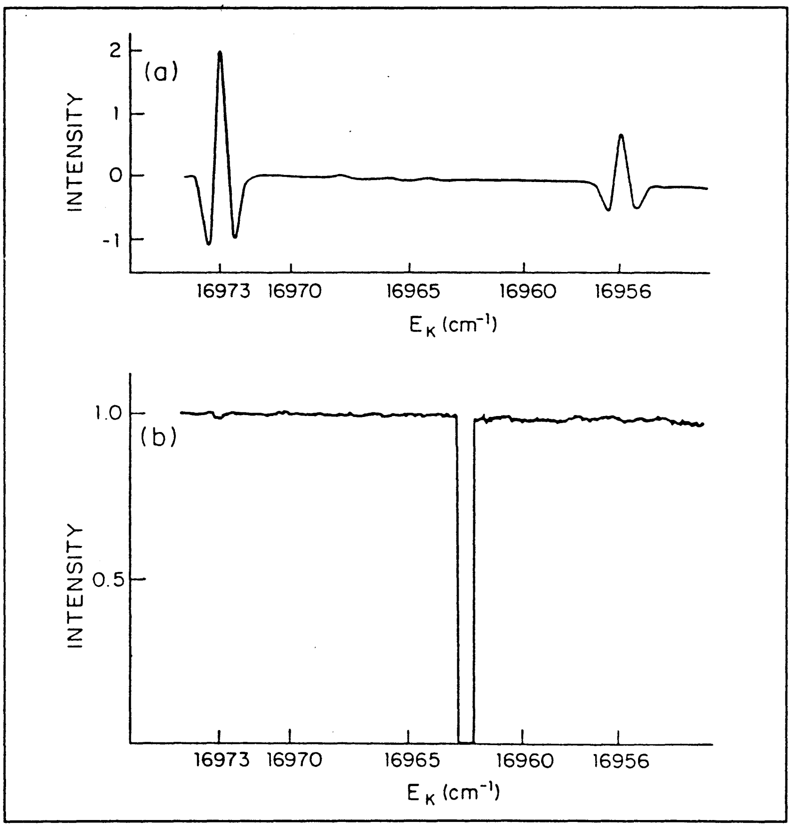


Fig. 11

Comparison of direct and two-tone FM absorption signals of the Na D lines, with 2% absorption in the D_2 line. (a) FM signal with M = 0.25 at 17 GHz frequency modulation. (b) Direct absorption signal. The D_2 line is barely visible and the D_1 line cannot be seen. [From Ref. 9]

4. FMS WITH SECOND HARMONIC DETECTION

It may be instructive to take a short glance at FMS with second harmonic detection. Diagnostics on broadband spectral features, together with the use of pulsed multimode lasers, increase the requirements on increased modulation frequency relative to the spectral widths of interest and the laser linewidth. This is accomplished with second harmonic demodulation, demonstrated by Janik *et al.*.[10] If the modulator drive power, and thus the modulation index, is increased, the second order sideband pair will gain in significance (Fig. 12). Second harmonic demodulation will depend on the second sidebands as well as the first, allowing the modulation frequency to be effectively doubled. It will also yield a symmetric spectral feature proportional to M^2. Fig. 13 illustrates two recordings, absorptive and dispersive, of the Na D_2 line, made by a pulsed dye laser with a linewidth of 4 GHz and 8.4 GHz modulation. The RF split off from the oscillator to drive the mixer LO port is frequency doubled to 16.8 GHz. Thus, the photodetector current at 16.8 GHz will be demodulated.

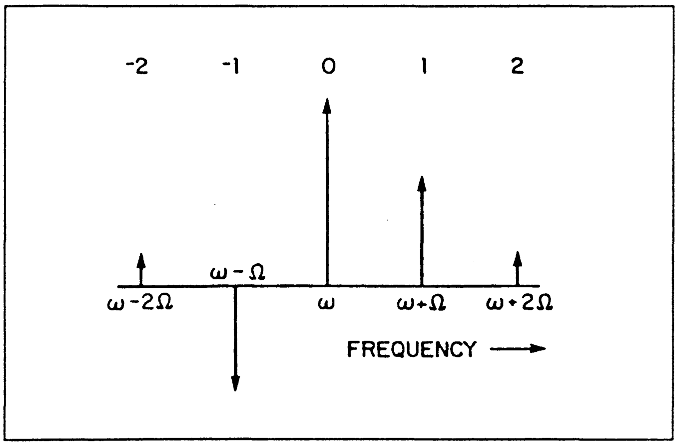


Fig. 12
Amplitude spectrum of a laser at frequency ω modulated at frequency Ω. The directions of the arrows indicate the phases of the sidebands and the lengths of the arrows of their amplitudes. The numbers labeling the sidebands correspond to the number of microwave photons added to the optical photon. [From Ref. 10]

Janik *et al.* report a higher SNR than for those signals obtained by demodulating the FM signal at the modulating frequency of 8.4 GHz. A real disadvantage of this method (presently), is the need for a high-speed photodetector, fast enough to follow the doubled frequency.

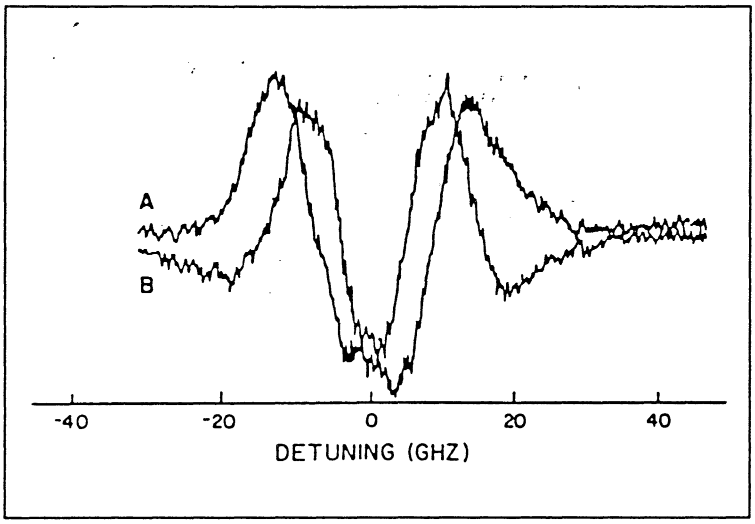


Fig. 13
Observed signal from the 5890 Na line: a) the absorption signal; b) the dispersion signal. [From Ref. 10]

5. APPLICATIONS

5.1 Pulsed lasers

In many applications, such as combustion diagnostics and remote sensing, a pulsed laser is preferred. However, when performing FMS with a pulsed tunable dye laser, the large laser bandwidth, FWHM in the GHz region, has to be considered. Random phase modulations occur within the envelope of the laser (i.e. beat signals at ω_m arise from different frequency components of the carrier that are ω_m apart) and contribute noise to the mixer output signal. To achieve an acceptable signal-to-noise ratio the modulation frequency has to be increased as far as possible outside the envelope. Fig. 14 illustrates the effects of raising the modulating frequency with respect to the laser bandwidth; two typical recordings of an FM signal produced by absorption from Fabry-Perot etalon are shown. It appears, that the signal-to-noise ratio is markedly improved by increasing the modulation frequency. In Fig. 15, a plot of the inverse of the adjusted signal-to-noise ratio, $(N/S)[0.1/(M^2/4)]^{-1/2}$, vs the modulation frequency, denoted ν, is shown.

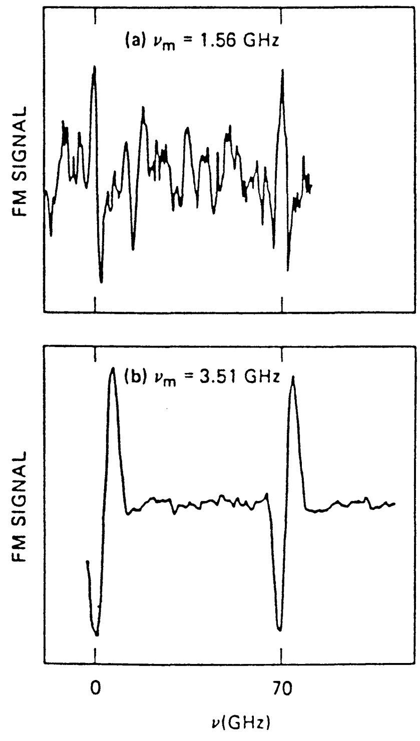
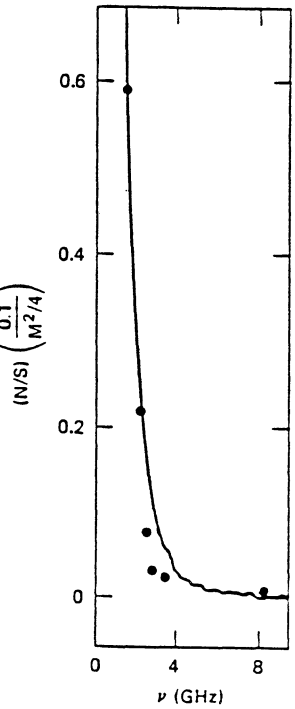


Fig. 14
FM signal produced by a 2 GHz wide, 12% absorption from a tunable Fabry-Perot etalon with a) ν_m = 1.56 GHz, and b) ν_m = 3.51 GHz. Two cycles of absorption from the tunable etalon are shown. The dye laser linewidth is $\Delta\nu$ = 1.0 GHz (FWHM) in both a) and b). [From Ref. 5]

The quantity $M^2/4$ is the amount of energy in each sideband relative to that of the carrier, and the adjusted signal-to-noise ratio, $(S/N)[0.1/(M^2/4)]^{1/2}$, is the signal-to-noise ratio, which would be obtained for 10% of the carrier energy in each sideband. From Fig. 15, it is evident that the noise in pulsed FMS falls off as the wing of the laser line.

The best sensitivity in pulsed performance, to the author's knowledge, is reported by Tran *et al.* in Ref. 5 and is 10^{-3}.

Fig. 15
Noise-to-signal as a function of modulation frequency. The dye laser spectrum, which has a FWHM of $\Delta\nu$ = 1.0 GHz, is also shown [From Ref. 5]



5.2 Diode lasers

FM spectroscopy can conveniently be carried out with diode lasers. The frequency modulation is internally achieved by directly modulating the injection current, which only requires a few milliwatts of radio frequency power, thus, eliminating the need for an expensive high power amplifier and, of course, an electro-optic modulator. Together with two-tone frequency modulation it provides a cheap and easy way to perform FMS, in the IR region. A disadvantage is that diode lasers are sensitive to RAM noise. Near-quantum-limit sensitivity has been demonstrated,[11,12] where laser excess noise was the limitation and recently quantum noise limited sensitivity was demonstrated by Carlisle *et al.* (Ref. 13).

EXPERIMENTAL PART

The experiments were carried out with two different phase modulators; one with a modulation bandwidth of 0-50 MHz and another, a resonant cavity modulator, working at resonances around 8 GHz. The reasons for using the low-frequency modulator were that it was available in our laboratory together with all RF equipment (in that frequency region) and that it provided an excellent opportunity to learn the basics of FMS. It also made it possible to perform Doppler-free frequency-modulated saturation spectroscopy.

If high/highest sensitivity is to be reached, the modulation frequency ought to be at least of the same order as the width of the spectral feature of interest; see Sect. 2.1. However, at a modulation of 50 MHz, a sensitivity (theoretical estimate) of 10^{-4}, where the sloping-baseline RAM made its appearance, was easily reached. No actual effort was made to increase this sensitivity. In view of the greater difficulties in the X-band (8 - 12.4 GHz), in the microwave region, it is tempting to suggest not to use a higher frequency than necessary.

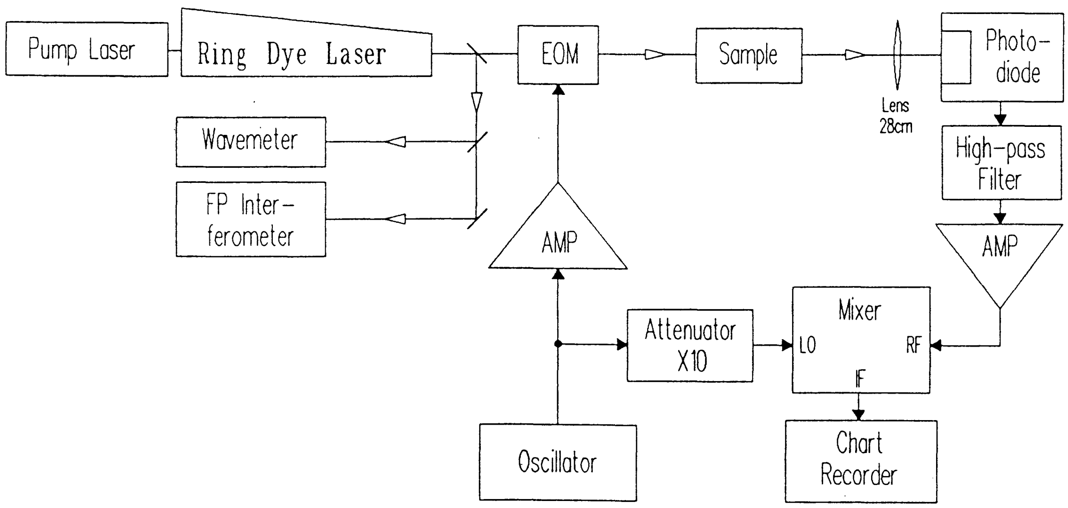


Fig. 16
Experimental arrangement for FM spectroscopy, using a single-mode cw dye laser and 50 MHz frequency modulation.

6. FMS WITH 50 MHZ FREQUENCY MODULATION

6.1 Measurements on I_2

The equipment employed in the experiment using 50 MHz frequency modulation was as follows (Fig. 16): the tunable dye laser was a Coherent 699-21 ring laser (linewidth 1 MHz) pumped by a Spectra-Physics argon ion laser. The EOM was a Gsänger light modulator LM0202P, which has four 45° y-cut ADP crystals and a frequency response of 0-50 MHz. The modulator was driven close to 50 MHz by General Radio Co. Unit oscillator Type 1211-c, amplified by a Boonton Radio Co. power amplifier, Type 230A, to give a maximum RF power of approximately 4 Watt. Power was split off from the oscillator output to drive (with 5 dBm) the LO port of a Minicircuits Laboratories ZFM-2 double balanced mixer. The photodiode was a Hammamatsu, its output was filtered by a high-pass filter (a capacitor with 100 ns time constant) and then amplified 410 times by an EG&G ORTEC AN302/N quad amplifier. A mixer will give a better noise characteristic if small signals are amplified before the RF port. Fig. 17 shows an oscilloscope picture, which is a double exposure of the photodiode output signal when the sidebands are set off-resonance, zero signal, and when they are set on-resonance, at maximum slope for an I_2 transition, for maximum beat signal.

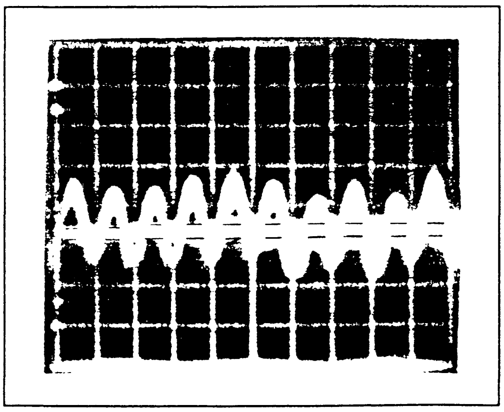


Fig. 17
Double exposure of photodiode signal, where the sidebands are set off-absorption resonance, zero signal, and on-resonance for maximum beat signal.

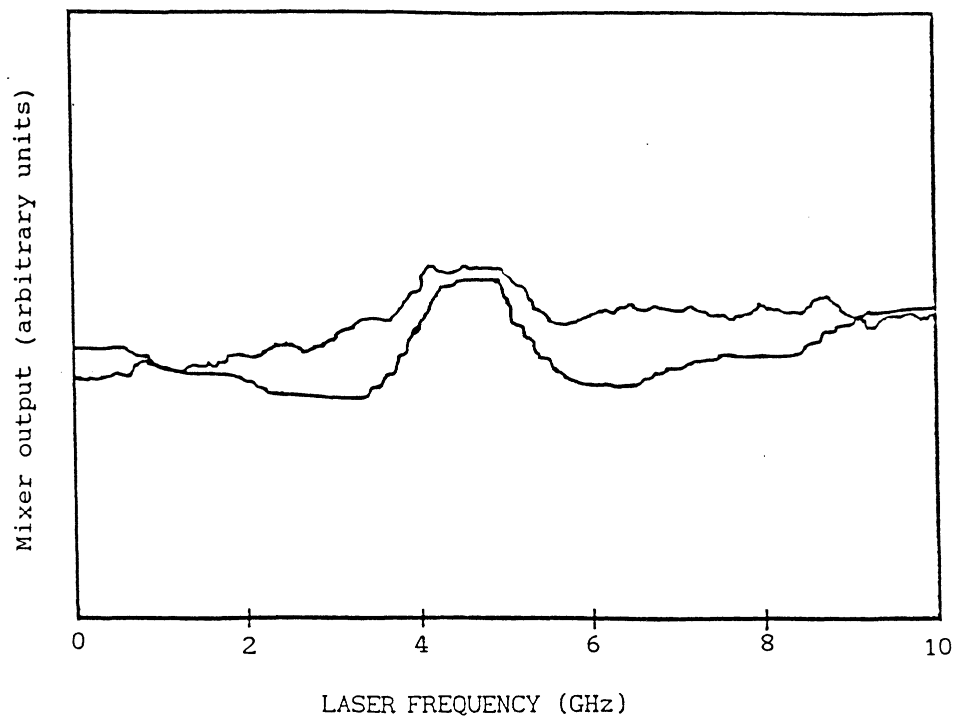


Fig. 19

FM signal of Na D_2 line at two different temperatures; the upper trace at 37 °C and the lower trace at 43 °C. The temperature of 37 °C corresponds theoretically to an absorption of 2.7×10^{-4}. The recordings were made with a single mode cw dye laser and 50 MHz frequency modulation.

while the laser was repeatedly scanned over 10 GHz, the temperature was recorded while tuning through the maximum of the absorption signal. The measurements proceeded until the signal disappeared, that is, the temperature and thus the vapor pressure were so low that the number of sodium atoms was not sufficient to give rise to a signal which was detectable with the equipment employed in the experiment. In Fig. 19 (and Fig. 6) some of the recorded absorption profiles are given for various temperatures. At a temperature of about 35°C, the signal was hardly recognizable.

In Ref. 14, Fairbank *et al.* present a thermodynamically derived plot of sodium vapor density against temperature. They also present an expression for the peak absorption cross section dependence of T

$$\sigma(5890Å) = (10^{-12}cm^2)(8.037 - 1.31\times10^{-2}T + 2.30\times10^{-5}T^2), \qquad (6.1)$$

$$\sigma(5896Å) = (10^{-12}cm^2)(3.968 - 6.40\times10^{-2}T + 1.11\times10^{-5}T^2), \qquad (6.2)$$

where T is in °C. For a temperature of 37 °C, the cross section of the D_2 line is calculated to be 7.58×10^{-12} cm^2 and, using Fig. 20 from Ref. 14, the sodium vapor density N is determined to be 5.0×10^6 cm^{-3}. The absorption is given by $\delta = \sigma NL$, where L is the length of the absorption cell. Thus, the absorption is theoretically estimated to be 2.7×10^{-4}. This result should be compared with the predicted sensitivity limit in the previous section. From the SNR ~5 shown in fig. 19 (upper trace) for an absorption of 2.7×10^{-4}, absorptions as small as 5.5×10^{-5} should be detectable with SNR ~1.

The measurements were sensitive to RF disturbances; when measuring with the highest sensitivity a small hand-waving in the laboratory was enough to totally drown the FM signal.

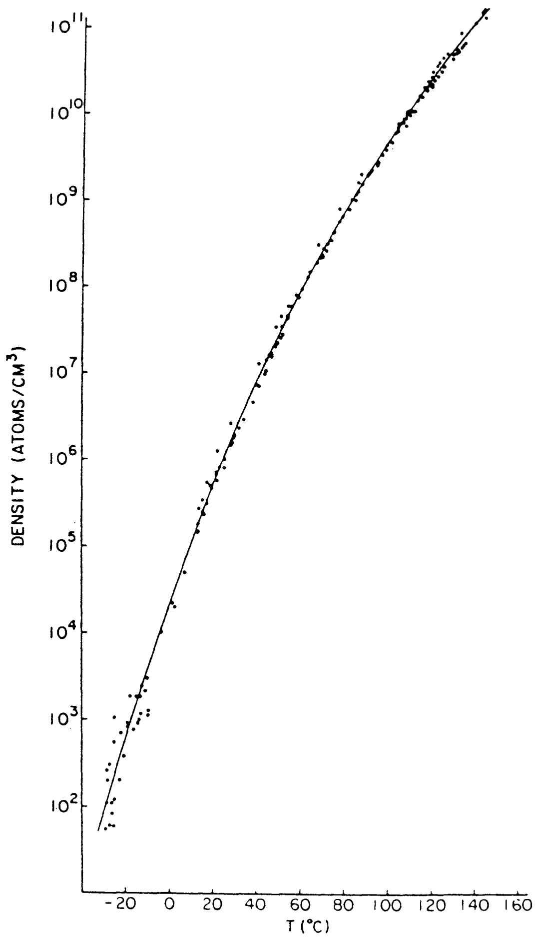


Fig. 20
Sodium vapor density measurements from Ref. 14. The solid line is a thermodynamically derived curve, using ΔH_0° = *25600 cal/mole. The curve was used in this work in order to estimate absorptions.*

6.3 Doppler-free FM saturation spectroscopy

During the work with FMS, experiments with optical bistability, using a saturation spectroscopy arrangement, were carried out in the same laboratory. We used this opportunity to perform Doppler-free FM saturation spectroscopy. The experimental set-up is shown in Fig. 21.

The equipment of the FM part of the experiment was the same as previously described. The "saturation equipment" employed was as follows: the tunable dye laser was this time a Coherent 599-01 single-mode laser, pumped by an argon-ion laser. The laser beam was chopped by a Soro electro-optics (type MAR-50 unit) acousto-optic modulator (AOM). It was driven, at a frequency of ω_{ch} = 30.5 kHz, by a Hewlett-Packard pulse generator, model 8003A. The mixer IF output

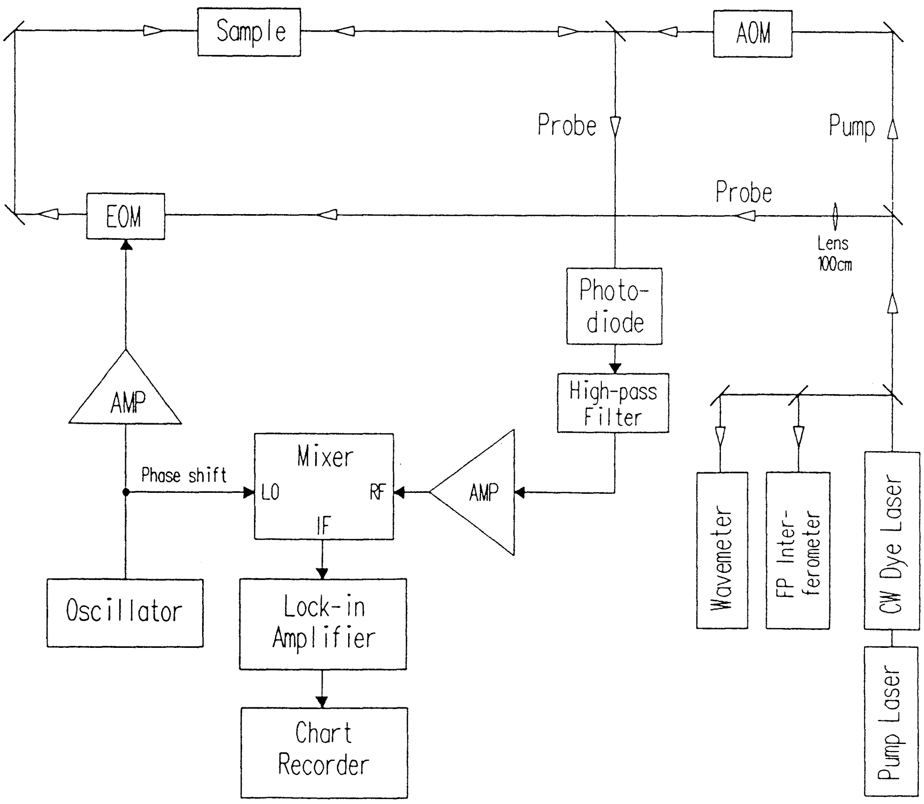


Fig. 21
Experimental arrangement used for Doppler-free FM saturation spectroscopy.

signal, containing the demodulated RF beat (due to absorption or dispersion in the cell) was sent to a Stanford Research Systems SR510 lock-in amplifier, referenced to the AOM. The FM signal was recorded on a strip chart recorder.

Let us again take a look at Fig. 21. The laser beam is split into a probe and a pump beam. The probe beam passes through the electro-optic modulator and the Na cell while the pump beam passes through the acousto-optic modulator and counter propagates the probe beam in the Na cell. Thus, when the laser is scanned across a spectral profile, the probe beam emerging from the cell will contain the sample-induced

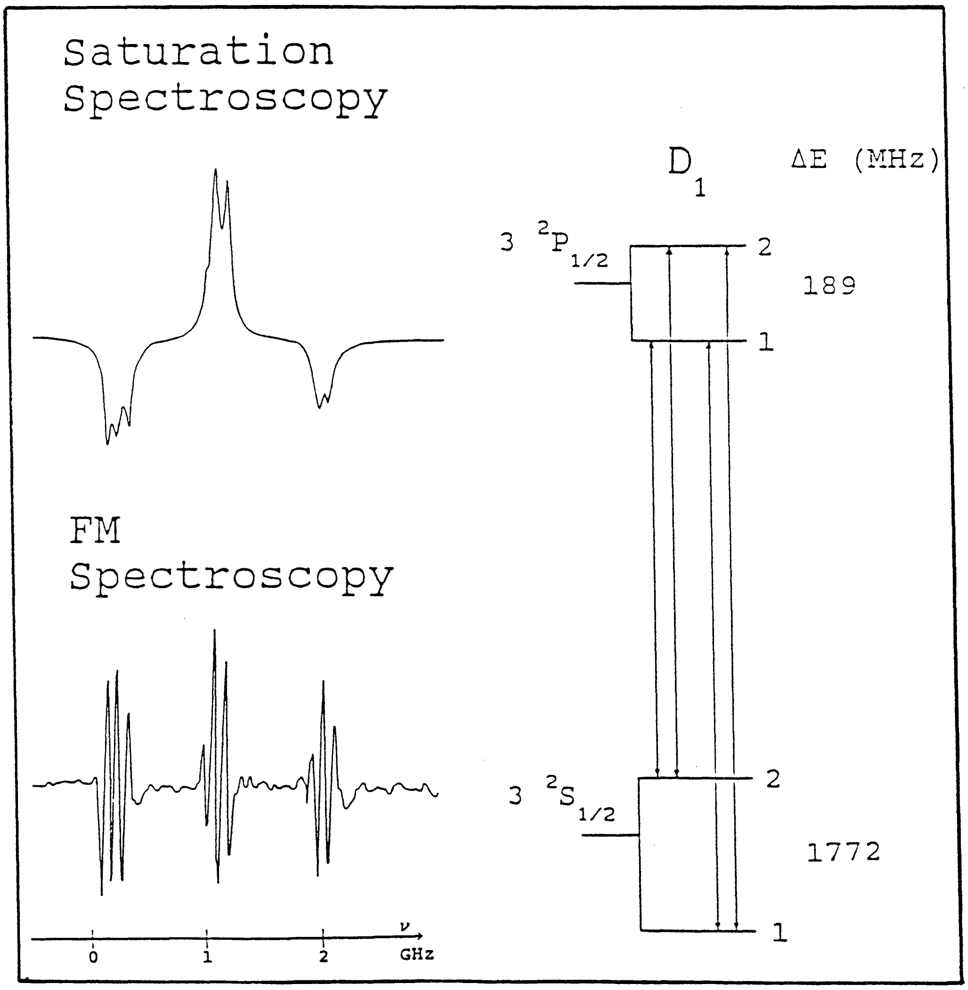


Fig. 22
Doppler-free saturation spectra and FM spectra of the sodium D_1 line. The FM recording was made with 50 MHz frequency modulation.

absorption beat signal, chopped at a frequency ω_{ch}. The probe beam is then split off and detected with the previously mentioned RF technique.

In Fig. 22 two recordings of the Na D_1 line are shown; one recorded with pure saturation spectroscopy, i.e. without the EOM and with the detector signal directed directly to the lock-in amplifier, the other with the FM equipment in place. The Na cell, same as in previous section, was heated to a temperature of roughly 100°C. It appears, that FMS clearly resolves the hyperfine structure, as well as the cross-over signals. It should be mentioned that no effort was made, (such as slow-speed laser scans with a high lock-in time constant), to record beautiful, well-resolved doppler-free saturation spectra. Both recordings were made under the same conditions. The measurements were disturbed by a strong RF interference. Probably it originated from interference between the AOM and the EOM drivers.

Saturation spectroscopy together with FM may be a useful aid when improving the performance and convenience of Doppler-free measurements on low-density media.

7. FMS WITH 8.4 GHZ FREQUENCY MODULATION

One of the purposes of this work, was to produce a working FMS arrangement, useful for diagnostics of broadband spectral features. Pressure-broadened lines in the atmosphere or in combustion flames, have a width in the GHz region. To achieve high (highest) sensitivity in FMS it is important that the modulation frequency is of the same order as the width of the absorbing feature. (See Eq. (2.9)). The use of pulsed tunable dye lasers also requires the modulation frequency to be raised. The signal-to-noise ratio obtained in FMS becomes larger as the modulation frequency is increased relative to the laser linewidth.[5] These considerations, make an electro-optic modulator, working around 8.4 GHz, attractive.

7.1 Description of the high frequency electro-optic modulator

Our electro-optic modulator (EOM) is a resonant cavity optical modulator constructed by Thomas F. Gallagher and his group. It is described in detail in Ref. 15 and Ref. 16. It uses the dispersive properties of a waveguide to achieve phase (velocity) matching of the optical and RF waves. The electro-optic material is lithium tantalate ($LiTaO_3$), in which the bulk velocity of the modulating RF wave and an optical beam differ by a factor of 3. The optical wave is the faster. The $LiTaO_3$ crystal forms a dielectric in a rectangular waveguide through a copper block (Fig. 23).

A modulator constructed in this way provides efficient use of the microwave power used to impart the modulation. This gives a high modulation index at the expense of the bandwidth of the modulation frequency. The cavity of our modulator has several modes between 8.0 and 8.6 GHz (the region where we have been working) where it resonates, more or less efficiently (Figs. 25 and 26). With the aid of a Teflon screw in the bottom of the copper block it is possible to tune and

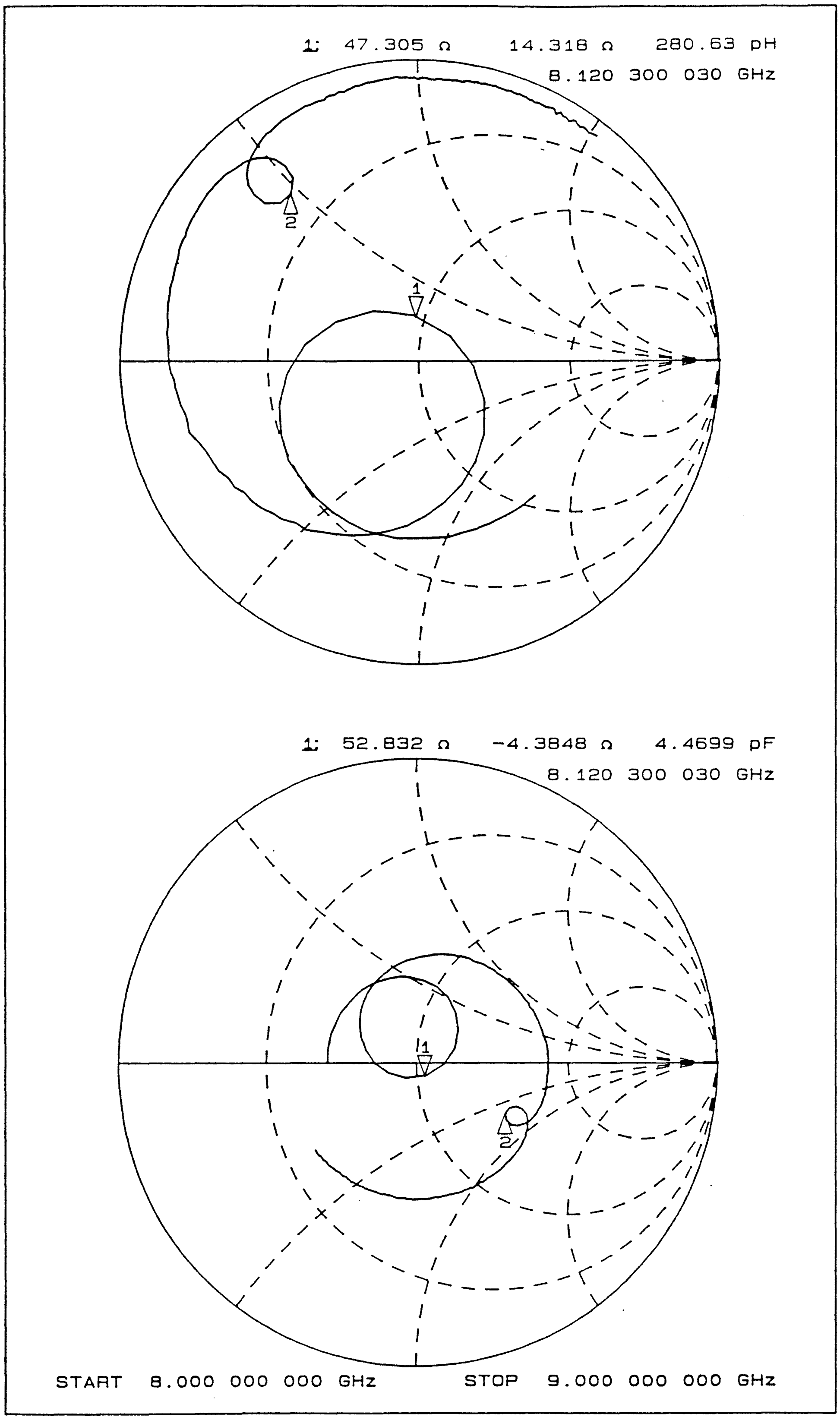


Fig. 26

Smith chart of the electro-optic modulator, used to impart about 8.4 GHz phase modulation to a laser beam. In the lower recording a 3 dB attenuator was inserted between the analyzer and the modulator. The recordings were made with an HP-8720A network analyzer.

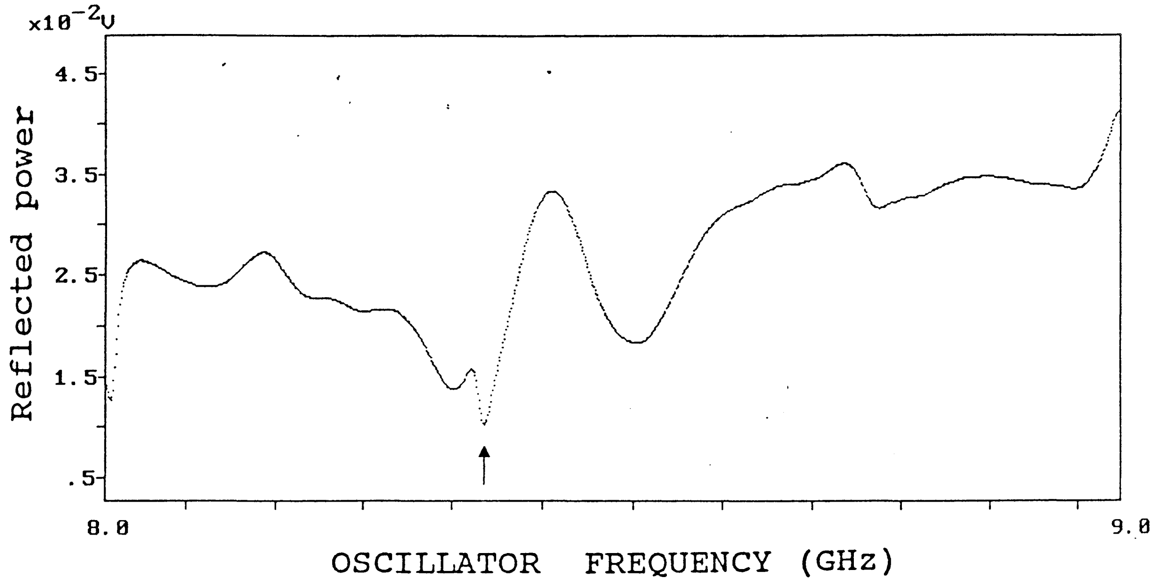


Fig. 27
Reflected power from the resonant cavity electro-optic modulator, showing one resonance.

7.2 Measurements on Na with a pulsed laser

One of the main goals was to investigate the capability of FMS in conjunction with a pulsed dye laser. However, after only a brief application with a single mode, continuous-wave dye laser modulated at 8.4 GHz, the step to pulsed performance turned out to be too large. It was difficult (for the author) to fully estimate and understand the importance of different parameters in order to improve sensitivity. It was necessary to return and acquire a deeper understanding of the easier cw application. Then, it immediately became apparent, that RF disturbances were limiting the sensitivity. Fig. 29 illustrates a recording of the D_2 line at a temperature of 121 °C and an absorption length of 7.2 cm, measured with a time constant τ = 5s and a modulation frequency of 8.43 GHz.

Experimental setup: The equipment employed in this experiment was as follows (Fig. 28): The pulsed dye laser was a Quantel TDL50, it was pumped by a YG580 actively Q-switched Nd:YAG

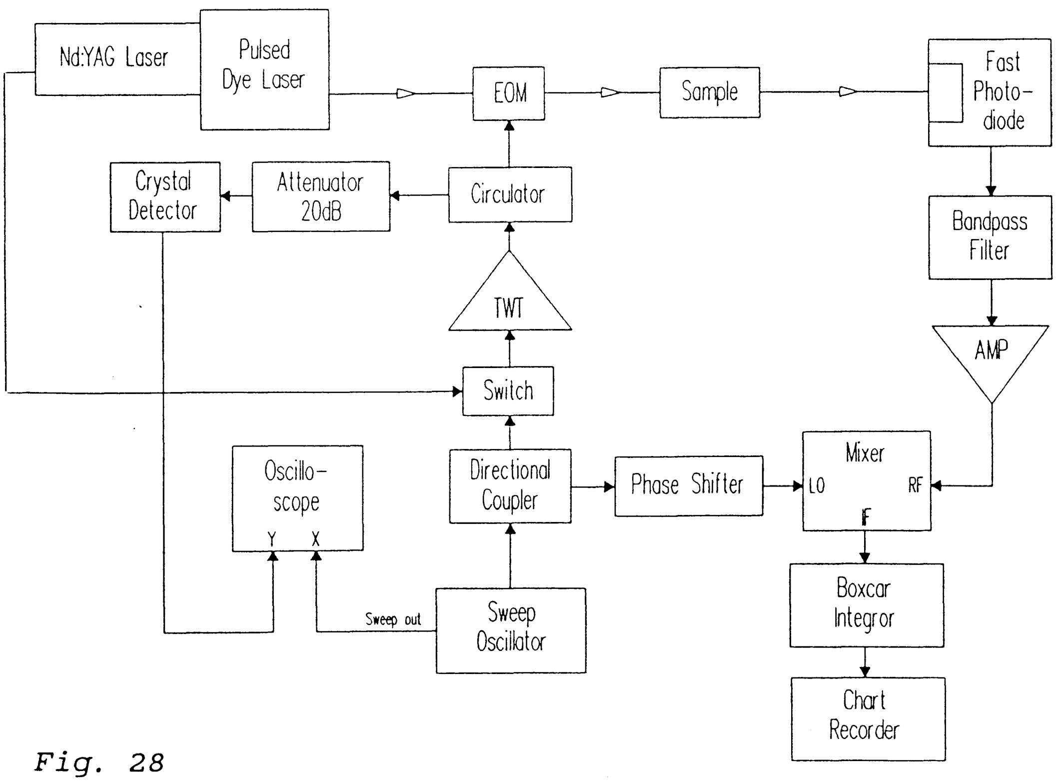


Fig. 28
Experimental arrangement for FM spectroscopy, using a pulsed dye laser and 8.4 GHz frequency modulation.

laser operating at 10 pulses/sec. The dye laser output had typical shot-to-shot intensity fluctuations of ~10%. The EOM, described in Sect. 7.1, was driven at 8.43 GHz by a Philips PM7022 sweep oscillator amplified to about 7 W peak power with a Hughes 1177H traveling wave tube (TWT) amplifier. The FM beat signal was detected by an Antel ultra high speed photodetector AR-S2. The photodetector output was filtered by a Sierra Microwave Technology SMF8012 bandpass filter with frequency range 8.0 to 12.0 GHz, amplified with an Avantek AM-8404M GaAs-FET amplifier and mixed in an Avantek DBX158L double-balanced mixer with the LO port driven by the sweep oscillator. The mixer output was directed to an EG&G model 162 boxcar integrator and the FM signal was recorded on a strip chart recorder. The absorbing Na cell was arranged in the same way as described in Sect. 6.2. A microwave switch was utilized, synchronously with the laser pulses, to chop the microwave drive to the modulator, with a duty cycle of ~10%, to avoid overheating of the $LiTaO_3$ crystal. For an explanation of the use of the circulator and the crystal detector, see Sect. 7.1 and the Appendix. The Appendix also contains a more detailed description of some of the various components used.

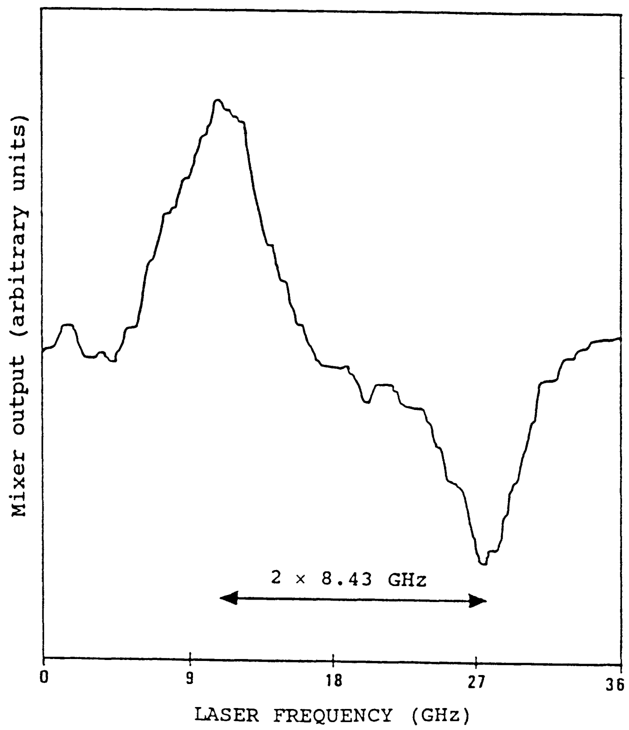


Fig. 29
FM spectra of Na D_2 line recorded with a pulsed dye laser and 8.43 GHz frequency modulation.

7.3 Measurements on I_2/Na with single-mode cw laser

Experimental setup: The equipment employed in this experiment (Fig. 30) was almost the same as that described in the previous section. The tunable dye laser was a Coherent 699-21 ring laser pumped by a Spectra Physics argon-ion laser. The mixer output signal was amplified by a Stanford Research Systems SR510 lock-in amplifier, referenced to the chopping frequency of the microwave switch. The FM signal was recorded on a strip chart recorder. The polarization rotator was used to rotate the laser beam polarization along the optical axis (y-axis, Fig. 23) of the uniaxial $LiTaO_3$ crystal, in order to obtain the largest possible electro-optical effect.

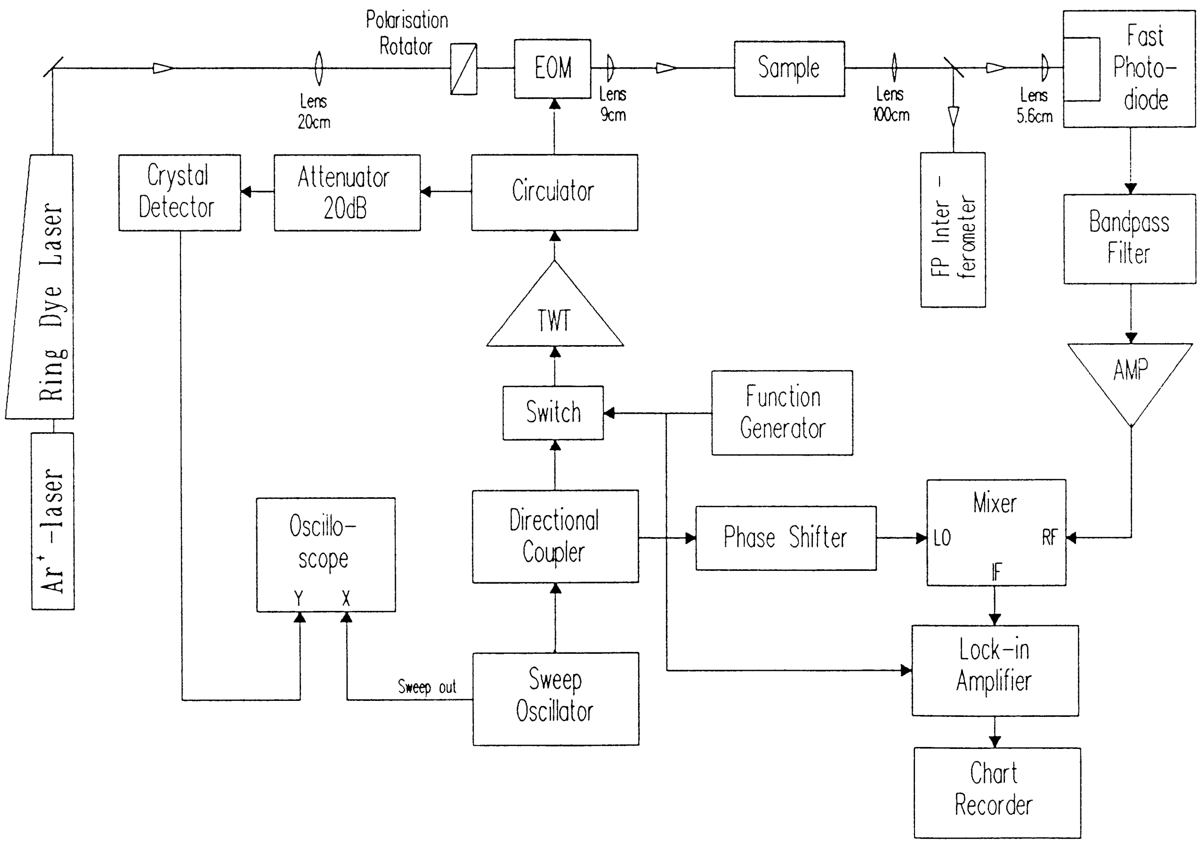


Fig. 30

Experimental arrangement for FM spectroscopy, using a single-mode cw dye laser and 8.4 GHz frequency modulation.

Fig. 31 illustrates a spectrum of I_2, recorded in a 29 GHz laser scan, using two methods: laser induced fluorescence and FMS.

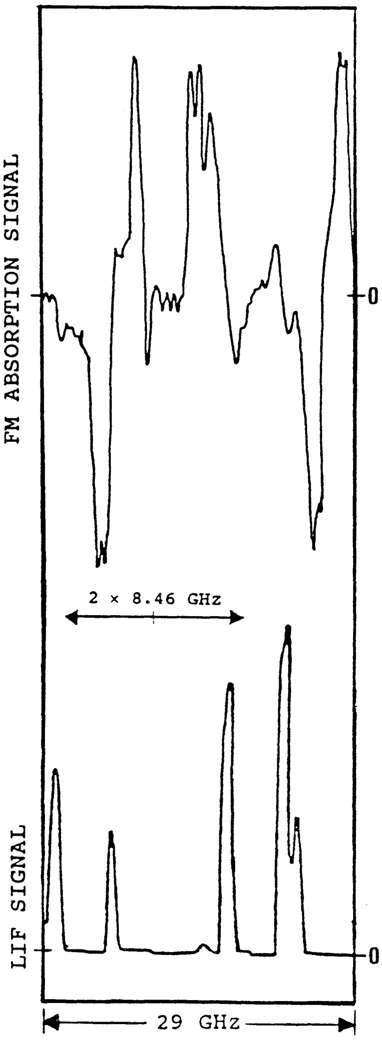


Fig. 31
FM signals and laser induced fluorescence (LIF) signals of I_2 at room temperature, recorded with a single-mode cw dye laser and 8.46 GHz frequency modulation.

So far the best sensitivity we have reached with a modulation frequency of 8.4 GHz is about 10^{-3}. The limit is restricted by etalon effects (RAM) and work is at the moment being done in our laboratory to improve the sensitivity. The measurements, at this current sensitivity, are highly sensitive to rf disturbances. The strong rf field in the laboratory seems to originate from the TWT amplifier.

Recently, the technique of using two modulation frequencies was investigated. A theoretical description of two-tone FMS can be found in Sect. 3. It was found that this method is insensitive to rf disturbances. As detection is carried out at a low frequency, corresponding to the separation between the sideband pairs, the strong rf disturbances from the (high-power) modulation frequencies can be filtered out. Fig. 32 shows the experimental setup. The components used are the same as those described in previous sections.

Fig.33 shows a two-tone FM recording of the Na D_2 line at a temperature of 68 °C, theoretically estimated to an absorption of 1%. The spectrum was recorded on a personal computer which was interfaced to the lock-in amplifier. The software used to record the data (and to plot it in an x-y format) was supplied by Stanford Research Systems together with its SR510 lock-in amplifier. The modulator was driven by approximately 10 watts of microwave power, which put approximately 20% of the carrier energy in each sideband (Fig. 24), and corresponds to a modulation index, M, close to unity ($0.2 \approx M^2/4$).

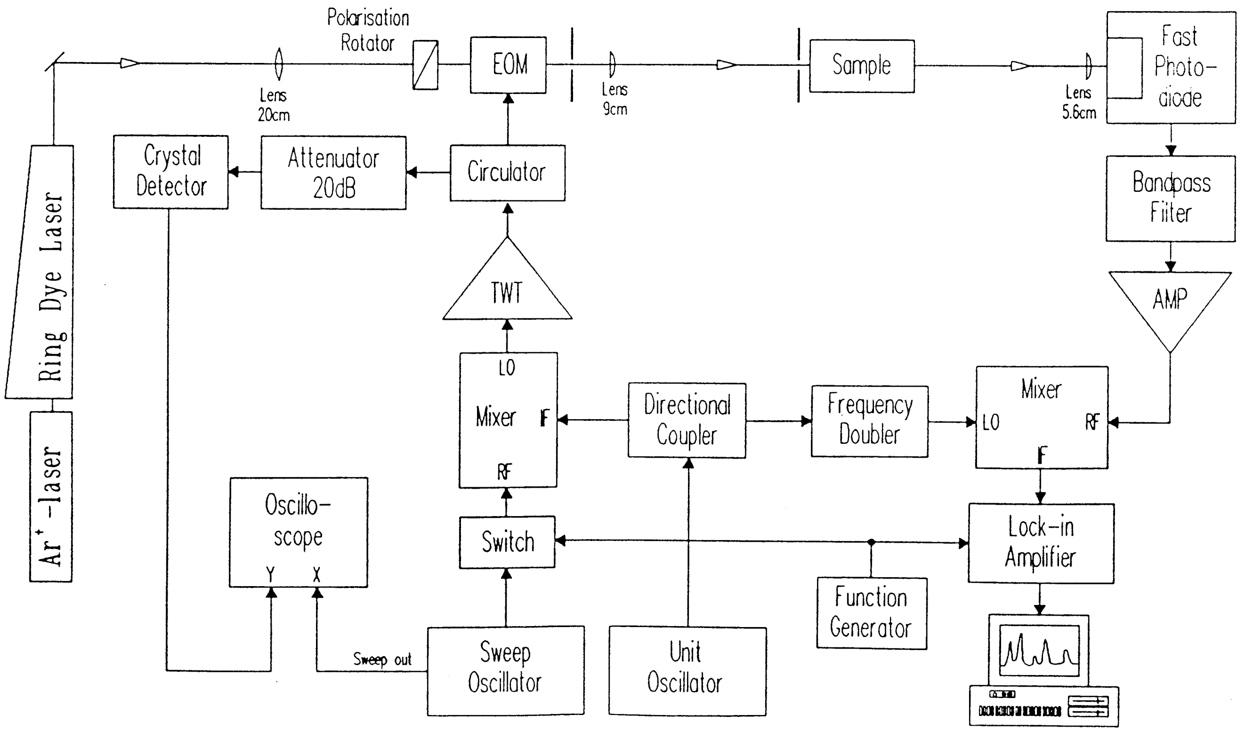


Fig. 32
Experimental arrangement for two-tone FM spectroscopy.

The bandwidth of the detection electronics, Δf, was 1 Hz. The Na cell arrangement was the same as that described in Sect. 6.2 and the absorption was calculated in the same way from Ref. 14. The sensitivity was limited by etalon fringes originating from the parallel end surfaces of the Na cell. The cell was tilted as much as possible (~7.5°) and the remaining etalon effect can be seen in Fig. 33, where the upper sideband is lifted. From the SNR ~30 shown in fig. 33 for an absorption of 1 %, we conclude that absorptions as small as 3×10^{-4} can be detected with SNR ~1. The minimum differential absorption $\Delta\delta_{min}$ that can be detected by using two-tone FM technique is given by Eq. 2.15, but with M replaced by M^2. For our system ($M^2 = 0.8$, $P_0 = 1$ mW, $\Delta f = 1$ Hz, $\eta = 0.6$), we find that $\Delta\delta_{min} \sim 6\times10^{-8}$.

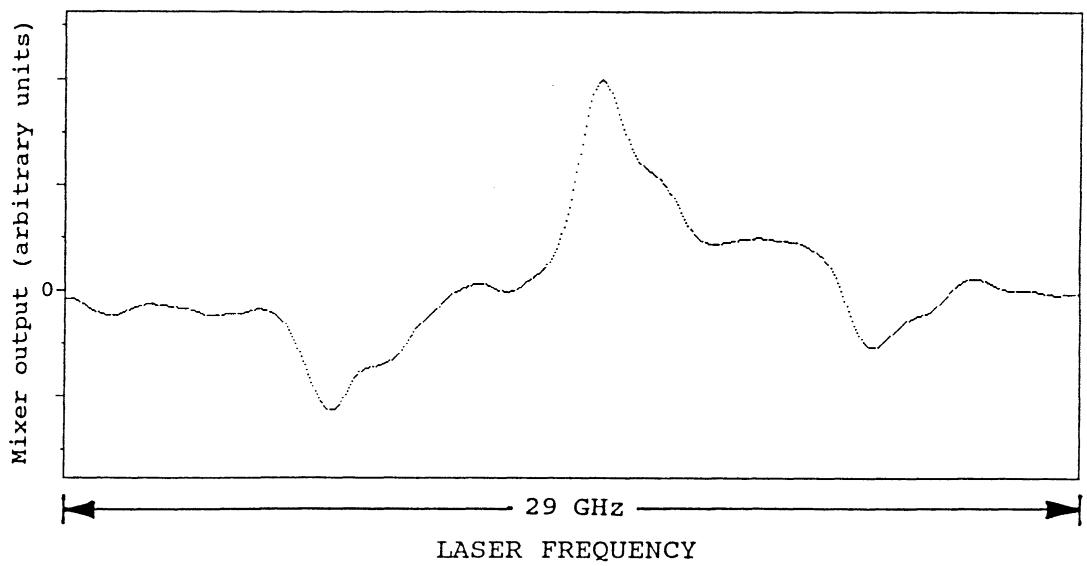


Fig. 33
Two-tone FM spectra of a 1 % absorption in the Na D_2 line, recorded with a single-mode cw dye laser, scanned over 29 GHz. The modulation frequencies were 8.44 GHz - 5.4 MHz and 8.44 GHz + 5.4 MHz.

ACKNOWLEDGMENTS

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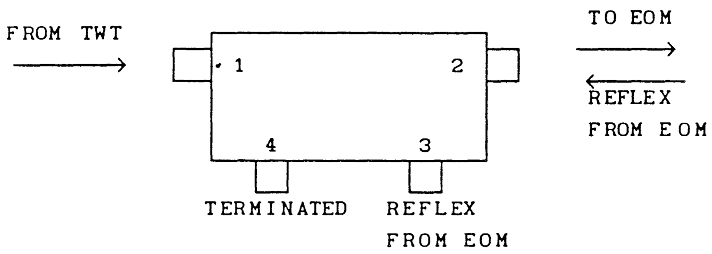
APPENDIX

This appendix has been written as a glossary, in an attempt to provide valuable help, for physicists, who do not have any insight into microwave technology, and to give a more detailed explanation of the components necessary to put frequency modulation spectroscopy into practice. It does not claim in any way to be complete.

Circulator, a device that acts as a commutator for power. See figure below.

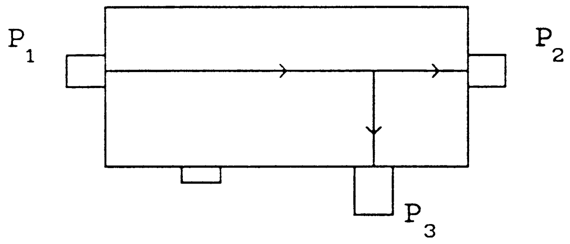


Power entering port 1 will be transmitted to port 2 only and power entering port 2 will be transmitted to port 3 only and so on. The circulator we used, a Sierra Microwave Technology SMC7012H, had four ports. It was used to find and keep control of a resonance in our electro-optic modulator. At resonance, the maximum power will be coupled into the modulator. Thus, when the oscillator is swept, it will leave a well defined dip in the reflected power, which is monitored with a crystal detector at the "next" port in the circulator. In the beginning of our measurements, a dual directional coupler was used to achieve the same performance.

Connectors/cables, in the microwave region, BNC connectors are no longer used. Care has to be taken regarding standing waves, attenuation, etc. We used SMA connectors together with a double-shielded RG400 cable. N connectors were also used. In the microwave region the power attenuation has to be considered. Even though the RG400 cable is suitable, it has an attenuation of 1.5 dB/m.

Diplexer, a device useful to separate a video signal from a radio-frequency. The electrical signal from the high-speed photodetector, will contain beat notes due to differential absorption influence, from a narrow-band spectral feature on the laser sidebands, as well as a direct, absolute absorption signal.

Directional coupler, a device that couples a distinct fraction of the incoming power into a second direction, as port 3 in the figure bellow.



Coupling is defined as follows:

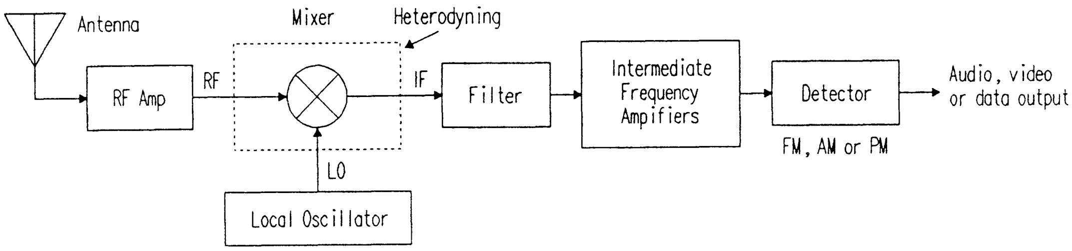
$$\text{coupling(dB)} = -10\log\frac{P_3}{P_1}.$$

We used a MA-COM FM-48117A with 6 dB coupling, to split off enough power from the oscillator output to drive the local oscillator (LO) port of a mixer with about 5 dBm.

dBm, the decibel represents a ratio, therefore a reference level must be stated if absolute values are required. The abbreviation indicates that the reference level is taken relative to 1 mW. Thus, dBm means "decibels relative to 1 milliwatt." Once familiar with it, it is a very convenient way of making circuit calculations.

Isolator, a device that permits power flow in only one direction. In the beginning of our experiments, when a dual directional coupler was used to achieve the properties of a circulator, care had to be taken regarding the high standing wave ratio (off resonance) of the modulator in order to protect the traveling wave tube amplifier from reflected power. A 3 dB attenuator was used in the beginning, between the dual directional coupler and the modulator, to lower the standing wave ratio below a safe level (Figs. 25 and 26), but this was a tremendous waste of "expensive" microwave power.

Mixer, in RF systems a mixer refers to a circuit with a nonlinear component that causes sum and difference frequencies of the input signal to be generated. This process is called heterodyning.



Superheterodyne receiver. The detector can be for FM (frequency modulation), AM (amplitude modulation) or PM (phase modulation), because virtually every modern receiver uses the heterodyning (mixing) effect.

Almost every high-frequency receiver uses a mixer to down-convert the received RF signal to an intermediate frequency (IF) signal. Transmitters use the sum frequency to up-convert the carrier to a higher frequency. When performing FM spectroscopy a double balanced mixer is to be used. In such a device good isolation between the RF and LO, RF and IF and LO and IF side is implemented in order to suppress unwanted high-order products and the only output will be sum and difference frequencies. In FM spectroscopy, the difference between two RF signals with the same frequency, of which one is amplitude-modulated, is of interest. It is important to choose a mixer, which provides a dc signal at the IF output. In two-tone FM spectroscopy, where a sum and a difference of a low (MHz) and a high (GHz) frequency are required, the same mixer can be used, but now with the low frequency connected into the IF port and the output through the LO port.

Oscillator, a Philips PM7022 sweep oscillator (YIG) was used to produce the modulation frequency. It is practical to sweep the frequency, when looking for a well defined resonance in the electro-optic modulator. However, consideration has to be given to the phase stability. If the phase or frequency of the oscillator fluctuates it will produce (phase) noise at the double balanced mixer, resulting in a noisy dc signal. This noise can be minimized if the propagation times for the optical signal (to the RF port) and the local oscillator (to the LO port) are equal. Thus, adding delay lines would provide an improvement. But, at our current measurements at a sensitivity of 10^{-3} with 8.4 GHz of modulation frequency, it is impossible to estimate the importance of the oscillator phase stability, in order to reach the detection shot noise limit. Thus, the oscillator should be as stable as possible.[6]

Phase shifter, in the GHz range were the wavelength is short, we used a simple line-stretcher (Omni-Spectra) to mechanically extend the path length between the oscillator and the LO port of the mixer. In this way, the phase relation between the LO and the RF side of the mixer is changed.

Switch, an Avantek AHS-1802 microwave switch (switching speed of maximum 25 ns), was used to chop the microwave power drive to the modulator by a duty cycle of ~10%, in order to protect the $LiTaO_3$ crystal from overheating. At the same time, components after the TWT amplifier such as attenuators with a maximum average drive power of 2 W were protected.

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