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Recent developments in high resolution saturation spectroscopy obtained by means of acousto-optic modulators (*)

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Résumé. — Nous présentons trois applications des modulateurs acousto-optiques à la spectroscopie de saturation à très haute résolution.
On utilise d'abord un modulateur A/O pour stabiliser à la fois la fréquence et l'amplitude du faisceau fourni par un laser à Ar^+.
On décrit ensuite une nouvelle méthode de détection des signaux d'absorption saturée dans laquelle le faisceau saturant est à la fois décalé et modulé en fréquence. La modulation induite est détectée sur le faisceau sonde. Cela permet d'améliorer sensiblement le rapport signal/bruit et d'éliminer très efficacement tous les fonds parasites.
Enfin, l'utilisation d'un modulateur A/O supplémentaire permet d'étudier les formes de raies au moyen d'un seul laser.

Abstract. — We present three applications of acousto-optic modulators to high resolution saturation spectroscopy.
— First, an A/O modulator is used to improve both frequency and amplitude stabilization of Ar^+ lasers outside the laser resonator.
— Second, a new technique to detect saturation resonances is demonstrated. In this method the saturation beam is both frequency shifted and modulated and the induced modulation of the probe beam is detected resulting in a highly improved signal to noise ratio and a high degree of background cancellation.
— Third, an auxiliary A/O modulator is used for line shape studies with a single laser.

Introduction. — The improvement of the sensitivity of the experimental techniques used in saturation spectroscopy has been aimed at two main goals :

(i) The increase of the signal/noise ratio.

(ii) The cancellation of the various backgrounds.

Noticeable advances in these two directions have been made possible by the availability of efficient and broadband acoustooptic (A/O) and electrooptic (E/O) modulators. Indeed the use of these powerful tools has allowed the application in the optical domain of several methods already used in the electronics and microwave domains [1]. Thus it has been possible recently to take full advantage of the following concepts :

— The use of high modulation frequencies in order to transpose the useful information at frequencies where the source noise is very small [2].

— The heterodyne techniques which are able to provide huge amplification factors and enable one to reach the shot noise level.

— The judicious use of optical sidebands which opens the possibility of modulating at much higher frequencies than those usually allowed by the different relaxation rates [3, 4].

In this article, our aim is to describe some recent developments in high resolution saturation spectroscopy obtained by means of A/O modulators. Our experiments have been performed with Ar^+ lasers on the $^{127}I_2$ molecule [5]. The frequency and intensity stabilities of the commercial Ar^+ lasers are not good enough for these high resolution applications. Therefore we have improved these characteristics and describe, in the first part, the laser control methods used for this purpose. In the second part we describe a new technique used in saturation spectroscopy in which the pump beam alone is both frequency shifted and modulated and the induced modulation of the probe is detected. Then we describe an experimental set-up specially designed for accurate lineshape studies using only one laser.

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1. **Frequency and intensity stabilization of a commercial Ar^+ laser.** — 1.1 PRINCIPLE. — The jitter of our commercial laser is of about 20 MHz p.p. (depending on the water flow used to cool the tube). To be able to use this laser in high resolution spectroscopy it is necessary to prestabilize its frequency on the transmission fringe of an external passive confocal resonator. In the schematic diagram of this servo-system given in figure 1 we again find the well-known « double loop », with the slow PZT (C_L) and the fast one (C_R). Its principle has been given in reference [6] and it has already been described several times in detail [7, 8].

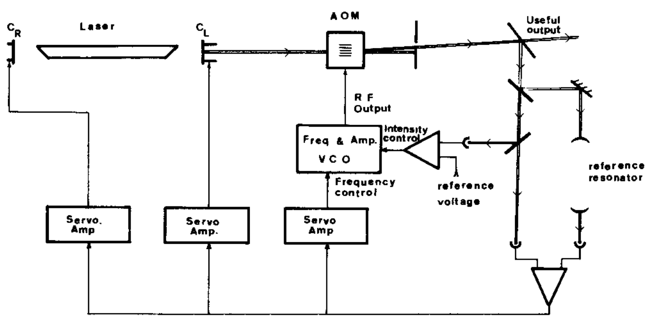

Fig. 1. — Scheme of principle of the external control of both the intensity and the frequency of the laser beam deflected by means of A/O M.

In addition we use a more sophisticated method proposed in reference [9, 10] of « an acoustooptic frequency and intensity control system ». We shall only report here the guide-line ideas of this method and outline its advantages and disadvantages. The error signal provided by the reference resonator is used to drive both the « double loop » and a third one including an A/O modulator which is used to correct the fast frequencies. The A/O modulator operates here like a single sideband modulator. The output beam is frequency shifted by an ajustable value $\Delta(t)$. This R.F. is supplied by a power oscillator, the frequency of which is controlled by the high-frequency components of the error signal. We can write the instantaneous frequency of the shifted beam as :

$$\omega_i = \omega_L + \delta\omega_L(t) + \Delta(t)$$

where $\omega_L + \delta\omega_L(t)$ is the instantaneous frequency of the laser, $\delta\omega_L(t)$ represents the residual jitter. We can also write $\Delta(t) = \Delta + \delta\Delta(t)$. The idea is then to cancel the residual jitter by driving $\delta\Delta(t)$ with the error signal, so that ω_i remains as constant as possible. In fact each of the three transducers C_L, C_R and A/O M of this « Hi-Fi » servo-system is specialized in a frequency domain : C_L for « bass », C_R for « medium » and A/O M for « treble ».

Moreover, the intensity of the A/O deflected beam can be controlled by varying the power of the R.F. ($\Delta(t)$). Therefore this method provides the means of stabilizing both the frequency and the intensity of the deflected beam.

1.2 IMPLEMENTATION AND RESULTS. — Because of the use of frequency domain specialized transducers we can limit the study of these servo-systems to the linear approach and the implementation of these various loops is not very difficult. For the « double loops » we have used the elements already described in reference [5] or in more details in reference [8]. In the high speed loop we have used an A/O M type IM50 (SORO) in which the frequency shift is $\Delta = 250$ MHz and whose transmission is $\simeq 98$ % into the first-order shifted sideband when the R.F. power is about 1.5 watt. The preamplifier which is the common stage of the three loops is of course a high frequency and low noise operational amplifier. We have tried several types. A good choice seems to be the 50 K (Analog Devices) or the OP37 (P.M.I.). We have found it essential to use a high slew-rate amplifier to drive the V.C.O. (the 50 K for instance). So the bandwidth of the overall response is not « electronically » limited. The limitation comes mainly from the acoustic wave propagation delay, $\lesssim 1$ μs, but compared with our usual fast PZT the advantages in terms of bandwidth are still considerable. Another limitation of this technique could come from the angular variations $\delta\theta(t)$ corresponding to any frequency change $\delta\Delta(t)$. In our case it is easy to see that this limitation cannot be really important for the following reason : the main components of the jitter of our Ar^+ laser are between 10 Hz and 10 kHz. Above these values, the spectrum amplitude decreases rapidely with the frequency and the A/O M is only operated in the high frequency domain where the needed correction is very small ($\delta\Delta \lesssim 10$ kHz p.p. i.e. $\Delta\theta \lesssim 10^{-6}$ rd). Nevertheless, it is well-known that to increase the efficiency of a linear servo-system (of given open loop gain shape) in the low frequency domain, it is necessary also to increase its bandwidth in order to maintain the stability. The effect of the A/O transducer is thus essential. It could be compared to a very effective phase corrector placed inside the conventional double loop. Indeed the main corrections are still operated by the piezoelectric transducers.

In conclusion it is essential to adjust properly the respective transfer functions of these three loops using the following steps :

(i) Choice of our ideal open loop overall response $G_i(j\omega)$.

(ii) Estimate of the response of each transducer in the frequency domain where this response is a constant (α_L, α_R, α_{AO}).

(iii) Synthesis of the correctors necessary to obtain a good approximation of the ideal response ($G_L(j\omega)$, $G_R(j\omega)$, $G_{AO}(j\omega)$, these correctors are included in the servoamplifiers of figure 1). The Bode diagram of figure 2 gives an idea of this principle.

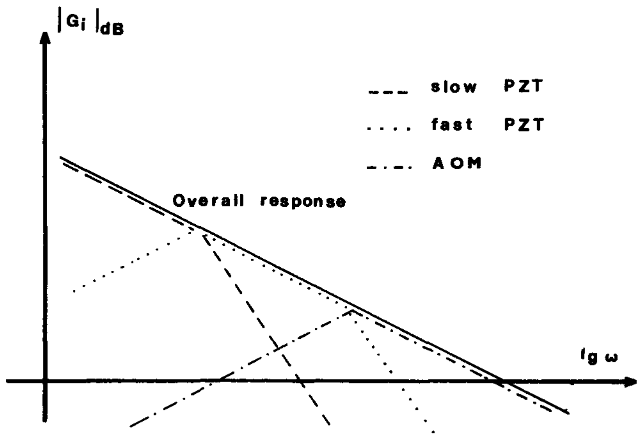

Fig. 2. — Bode diagram of the three loops used in the frequency stabilization servo-system.

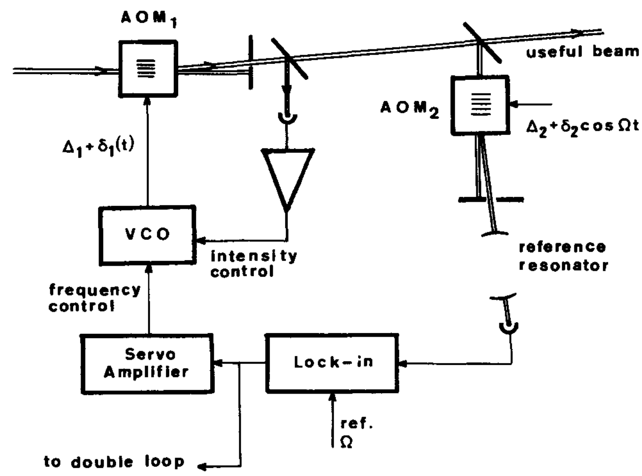

Fig. 3. — Laser frequency locking to the transmission peak of a reference Fabry-Perot cavity by means of frequency modulation with an A/O M.

When these three loops operate properly the overall bandwidth is about 300 kHz and we usually obtain a residual frequency jitter $\lesssim$ 3 kHz rms.

The implementation of the intensity servo-loop did not present any difficulty. The bandwidth was also of about 300 kHz (limitation arising also of course from the time delay imposed by the acoustic wave). We have not tested in detail the efficiency of the intensity stabilization. We can only say that in our experiments the efficiency was sufficient because as we will see later we were able to reach the shot noise level.

1.3 Alternative methods and improvements. — The A/O M can also provide other advantages in the laser frequency stabilization domain. It is well-known that the difficulty of the precedent method is that an accidental frequency jump can unlock the servo-system if this jump steers the laser frequency on the wrong side of the resonator fringe. New methods (which mostly originate in the early microwave domain [1]) have been recently proposed in order to avoid that problem :

— The polarization spectroscopy of a reflecting reference cavity [11].

— The R.F. phase modulation technique [3, 4, 10].

We offer a challenge to these methods by applying the use of the early idea [12] which consists of modulating the frequency of the laser and of detecting in phase the amplitude modulation of the light transmitted by the reference resonator. One obtains a dispersion-like lineshape which corresponds to the derivative of the Airy function when the modulation index is small. The use of an auxiliary A/O M to modulate the laser beam frequency gives a new interest to this method (see Fig. 3). Indeed we now find the following advantages :

(i) The frequency shift Δ_2 provides a good spatial isolation [9, 13] and thus allows the use of a very high finesse non confocal reference resonator.

(ii) The frequency modulation Ω can now be high ènough (3 MHz for instance) to reach a region where the source noise is very small (lower than the shot noise or the electronic noise). Then we do not need any difference or ratio techniques which are ordinarily used in order to cancel the effect of the amplitude noise of the laser.

(iii) The servo bandwidth is not limited by the carrier frequency.

(iv) The method is simple to implement.

Nevertheless too high a carrier frequency introduces a significant angular sweep of the deflected beam and then a broadening of the reference resonator lineshape. To avoid this problem one can use a double-pass acousto-optic system, the principle of which is described in chapter 3. This A/O frequency modulation method has been successfully operated and the preliminary results are promising.

2. **Heterodyne saturation spectroscopy through frequency modulation of the saturation beam.** — 2.1 Experimental set-up. — The schematic diagram of the new method is given in figure 4. The basic saturated absorption ring used in our previous experiments [2, 5, 13] has been slightly modified.

The two basic ideas are :

1) To modulate the frequency of the saturation beam and to detect the induced modulation of the probe beam like in the method proposed in [17].

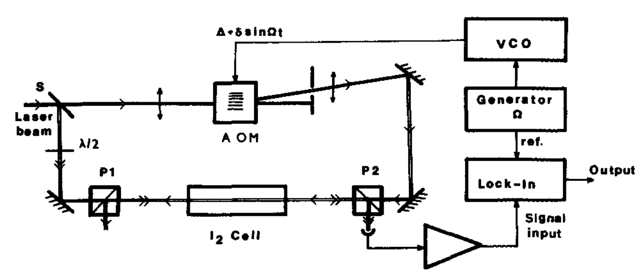

Fig. 4. — Principle of heterodyne saturation spectroscopy by means of frequency modulation of the pump beam.

2) To shift the frequency of the saturation beam to provide high optical isolation.

The laser beam is divided by the beam splitter into two counter-propagating beams. The pump beam transmitted through S is frequency shifted by Δ and frequency modulated at frequency Ω with a p.p. amplitude $2\,\delta$ by means of an A/O modulator. The R.F. $\Delta \simeq 70$ MHz is given by a power oscillator (~ 1.5 W) whose frequency is controlled by the output signal of a low frequency generator ($\delta \sin \Omega t$). Two Glan prisms P_1 and P_2 yield two orthogonal polarizations inside the cell. They are located close to the cell windows in order to reduce the effect of depolarization of the beams in the ring and then to prevent the feedback towards the laser with the best efficiency. After their interaction inside the sample cell, the pump and the probe beams are respectively deflected by P_1 and P_2. The probe beam whose intensity can be adjusted by the $\lambda/2$ plate, is then detected by a fast photodiode (FND 100), amplified by a low noise preamplifier (type TL71 or better type OP27) and demodulated by the usual lock-in amplifier technique.

2.2 PRINCIPLE OF THE METHOD. — In a simple approach [8] the signal detected in such a saturated absorption experiment is given by :

$$S \propto I_P + K I_P \exp\left[-\left(\frac{\omega_P - \omega_0}{\omega_D}\right)^2\right] + K' I_P^2 \exp\left[-\left(\frac{\omega_P - \omega_0}{\omega_D}\right)^2\right] + \beta \frac{\gamma I_S I_P}{\gamma + i\left(\frac{\omega_S + \omega_P}{2} - \omega_0\right)} \quad (1)$$

I_P, ω_P and I_S, ω_S are the powers and the frequencies of the probe and of the saturating beams respectively. ω_D is the Doppler width, ω_0 the resonance frequency of the transition, γ the optical dipole relaxation rate, K, K' and β are proportionality coefficients depending on the medium properties. This relation is, of course, only valid in the Doppler limit approximation. The first term represents the amplitude of the continuous background, the second one describes the linear absorption, the third-one corresponds to self-saturation of the probe and the last one describes the useful saturation signal. When the electric field E_S of the pump beam alone is modulated :

$$E_S = \frac{1}{2} \mathcal{E}_S \exp\left(i\left[(\omega_L + \Delta)\, t + \frac{\delta}{\Omega} \sin \Omega t + kz\right]\right) + \text{c.c.} \quad (2)$$

and when δ and Ω are small compared with γ (adiabatic approximation), the relation (1) is still valid and it is noticeable that only the last term is modulated.

$$S(\Omega) \propto \frac{\gamma I_P I_S}{\gamma + i\left(\omega_L + \frac{\Delta}{2} + \frac{\delta}{2} \cos \Omega t - \omega_0\right)} \quad (3)$$

We have taken $\omega_P = \omega_L$ and $\omega_S = \omega_L + \Delta + \delta \cos \Omega t$. In its present configuration our experimental set-up is only sensitive to the saturated absorption signal. The signal detected by the photodiode is thus proportional to Re $S(\Omega)$. When the frequency reference of the lock-in is Ω one obtains a dispersion-like lineshape ready to use for frequency laser stabilization. In fact, the output signal of the lock-in reduces to the first derivative of Re $S(\Omega)$

$$V_S(\Omega) \propto \frac{-2\gamma^2 I_P I_S(\omega_L + \Delta/2 - \omega_0)}{\left[\gamma^2 + \left(\omega_L + \frac{\Delta}{2} - \omega_0\right)^2\right]^2}. \quad (4)$$

The preceding analysis shows the main advantages of this new method :

— high frequency modulation (but with the limitation $\Omega < \gamma$);

— cancellation of all types of backgrounds.

In spite of its simplicity this approach gives an idea of the expected lineshape when the experiment is operated in order to obtain a signal of metrological interest (i.e. a useful error signal for frequency stabilization of the laser).

However a more sophisticated approach is necessary if this method is to be used for accurate measurements and in any case where δ or Ω are $\gtrsim \gamma$ [19]. Then one has to take into account the different Fourier components of the pump beam and one can use an analysis in terms of degenerate four-wave mixing [14]. In this approach (see reference [15]) the signal field E_R is the sum of the various electromagnetic fields reemitted by the medium. Since the photodetector current is proportional to the intensity of the total impinging beam (probe, E_P + reemitted fields, E_R), the heterodyne beat signal results from the interference term $E_R \cdot E_P$.

Another major advantage of this method is the simplicity of the experimental set-up. However it is necessary to take some care in its implementation and to use proper optics in order to eliminate any wave front deformation. A more serious difficulty could some from any amplitude modulation of the pump at frequency Ω (this could come for instance from a non linearity in the response of the V.C.O.). This point is easy to verify by monitoring the pump beam amplitude. In our experiment we have not seen any spurious modulation of this kind. Anyway we have seen in part 1 that such a problem is easy to solve by stabilizing the intensity of the pump beam by means of the acoustooptic modulator itself.

2.3 EXPERIMENTAL RESULTS AND PROSPECTS. — 2.3.1 *Results.* — Our experiments have been performed on the 43-0 P(13) transition of the $^{127}I_2$ molecule

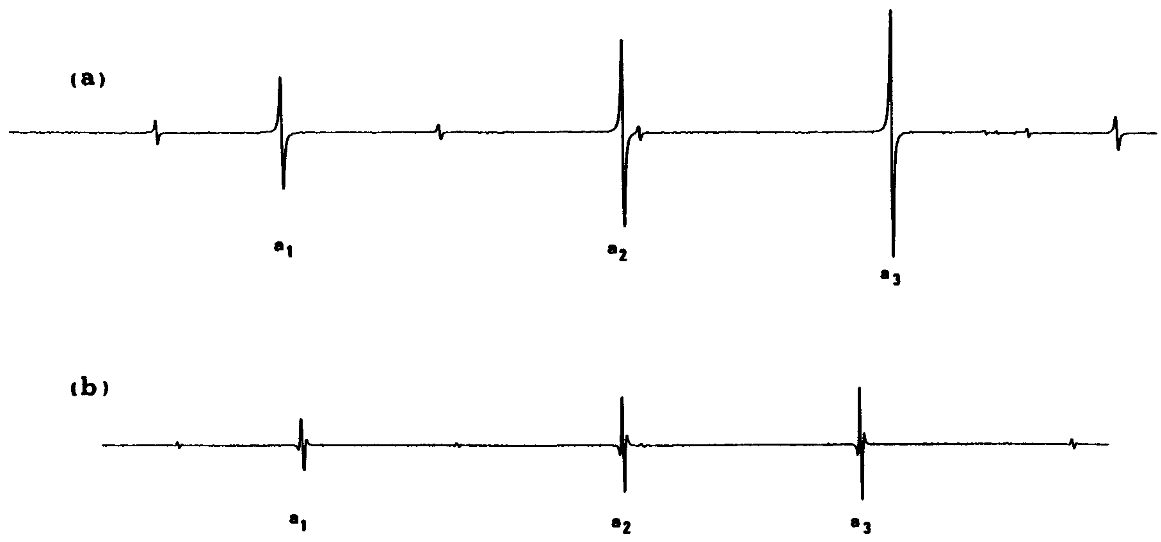

Fig. 5. — Low frequency region of the $^{127}I_2$ spectrum corresponding to transition $v = 0 \rightarrow 43$, P(13), $\lambda = 514.5$ nm ($p = 10$ mT, laser intensity $\simeq 1$ mW). $\nu_{a_2} - \nu_{a_1} = 71\,865.2$ kHz (see ref. [5]). (*a*) Frequency modulation of the pump beam at $\Omega = 50$ kHz ($\delta = 500$ kHz). Detection of the first harmonic. (*b*) Third harmonic of the saturated absorption signal in conventional low frequency modulation (at 1 kHz) of the laser beam.

with our prestabilized Ar^+ laser. Figure 5*a* shows the first main components of this spectrum obtained with the new method. These results are to be compared to those obtained with the conventional spectrometer of reference [8] (see Fig. 5*b*). The experimental conditions (I_2 pressure $\simeq 10$ mT, laser intensity $\simeq 1$ mW, beam diameter $2\,w \simeq 6$ mm) were the same except the time-constant of the lock-in which was 20 times larger in figure 5*b* than in figure 5*a*. One can see that the sensitivity differs by a factor $\simeq 5$ at the advantage of the new method. The residual noise is of the order of magnitude of the calculated shot noise. If the frequency modulation is increased up to 1.5 MHz, each line is split into four resonances (Fig. 6*a*, *b*). For a frequency modulation of 2.4 MHz these four resonances are completely resolved. Their shape (absorption (Fig. 6*a*, *c*)

Fig. 6. — Lineshape splitting for high modulation frequencies.

a) absorption, *b*) dispersion: $\Omega = 1.5$ MHz

c) absorption, *d*) dispersion: $\Omega = 2.4$ MHz.

or dispersion figure (6*b*, *d*) depends on the phase of the lock-in reference. The positions of the resonances centers are $\nu_0 \pm \Omega$ and $\nu_0 \pm \Omega/2$ where ν_0 is the ordinary line center. It is noticeable that each component of this quadruplet is not modulation broadened. The theoretical interpretation of these results is given in reference [15].

2.3.2 *Conclusion and prospects.* — This method, very simple to achieve, increases the detection sensitivity in saturation spectroscopy close to the ultimate limit corresponding to the shot noise and provides a ready to use error signal which is very convenient for metrologic and spectroscopic applications. The very effective cancellation of all various backgrounds makes this method particularly attractive in the domain of metrology.

However the above method may present two limitations in some extreme cases :

(i) The detection frequency is limited by the relaxation rates.

(ii) The probe acts like the local oscillator to create the heterodyne beat in the photodetector current and thus there is no independent control of local oscillator and reemitted signal field [13-15]. The heterodyne amplification effect is then limited by the probe intensity (saturation effects).

The phase modulation [3, 4] (or frequency modulation by means of an A/O M) takes care of the first difficulty but cannot completely solve the second one. An alternative technique could be based on the combination of pump frequency modulation with saturation interferometry [16, 17] which allows one to separate the probe and the local oscillator. In figure 7 we present a possible scheme of this method where the detection frequency can be increased to very high values.

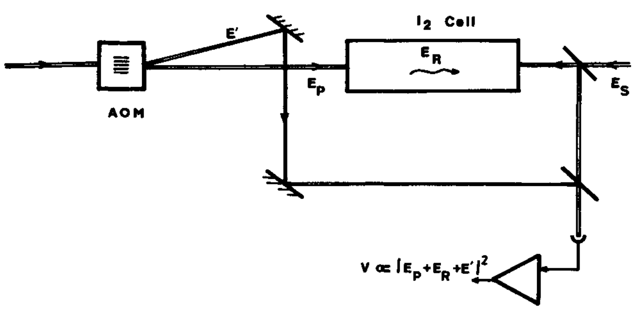

Fig. 7. — Possible interferometric scheme for a sensitive heterodyne detection of saturated absorption.

3. **Line shape studies by means of A/O M.** — 3.1 PRINCIPLE OF THE EXPERIMENTAL SET-UP : SEE FIGURE 8. — The idea is to lock the prestabilized laser to a reference line with a first saturation set-up using the previous method (of Fig. 4) and to use a part of the beam to monitor the shape of the same line by sweeping the deflected beam frequency in a second similar set-up (see Fig. 8) [18].

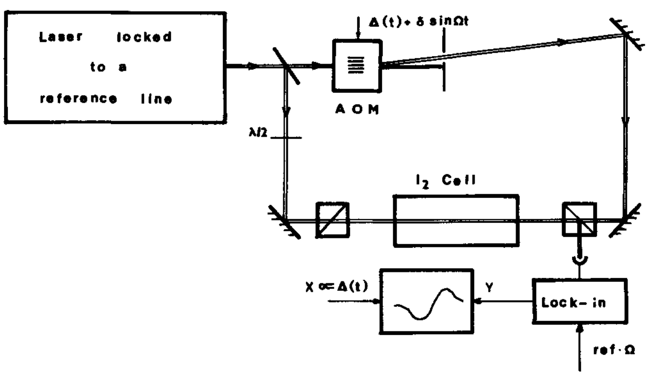

Fig. 8. — New technique for accurate lineshape studies with a single laser by means of two A/O M.

The main problem is to obtain a relative stability of about 10^{-6} between the generator which gives Δ in the first set-up (i.e. the reference one) and the one which gives a swept frequency $\Delta(t)$. The easiest solution is to use two separate synthesizers. A cheaper solution consists of using two ordinary V.C.O.'s and to lock the frequency of the one which provides $\Delta(t)$ on the one which gives Δ, with an adjustable offset around the value $\Delta(t) = \Delta$. We have successfully used this second solution. One can record either the first or second harmonic of the saturated absorption signal by using Ω or $2\,\Omega$ as frequency references for the lock-in. However if we want to record the lineshape for high pressure values it is necessary to use a frequency excursion of about 5 MHz in which case the associated angular variation of the deflected beam is not negligible.

3.2 THE DOUBLE-PASSED A/O SYSTEM. — In order to cancel the angular variations related to the frequency scanning we used a double-pass A/O M system [9] the principle of which is given in figure 9. The input field $E_i = \mathcal{E}_i\, e^{i(\omega t - kz)}$ is focused by O_1 in the A/O M where it is deflected by the travelling acoustic wave $A = \mathcal{A}\, e^{i(\Delta t - Kx)}$. The frequency of the deflected beam is $\omega + \Delta$ and because of the linear momentum conservation its wave vector is $\mathbf{k} + \mathbf{K}$. By means of a reflector composed of the optical system O_2 and the mirror M, the deflected beam is reflected and refocused in the A/O M, where it interacts again with the acoustic travelling wave. This retroreflected field, which wavevector $-(\mathbf{K} + \mathbf{k})$, is thus deflected again by the acousto-optic wave. The output electric field is then

$$E_S = \mathcal{E}_S\, e^{i[(\omega + 2\Delta)t + kz]}$$

The useful beam is extracted by means of the Glan prism P and the $\lambda/4$ plate. Thanks to this system the direction of this beam is not modified by any change of the frequency Δ.

3.3 PRELIMINARY RESULTS. — In figure 10 we record the first harmonic (Fig. 10*a*) and the second harmonic (Fig. 10*b*) of the a_6 line by means of this experimental set-up including the double-passed A/O system. These preliminary results are promising especially those of the second derivative. Indeed, we have found it inte-

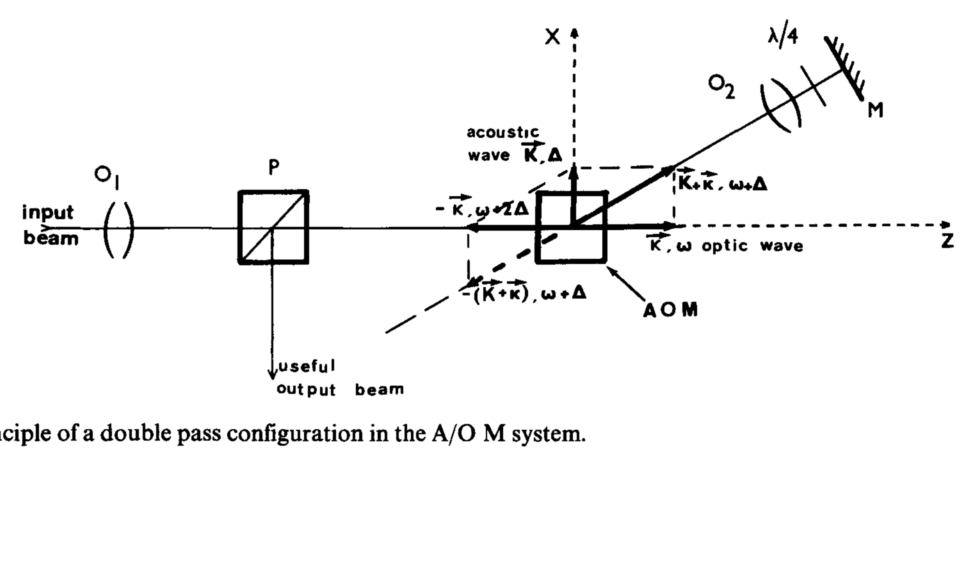

Fig. 9. — Principle of a double pass configuration in the A/O M system.

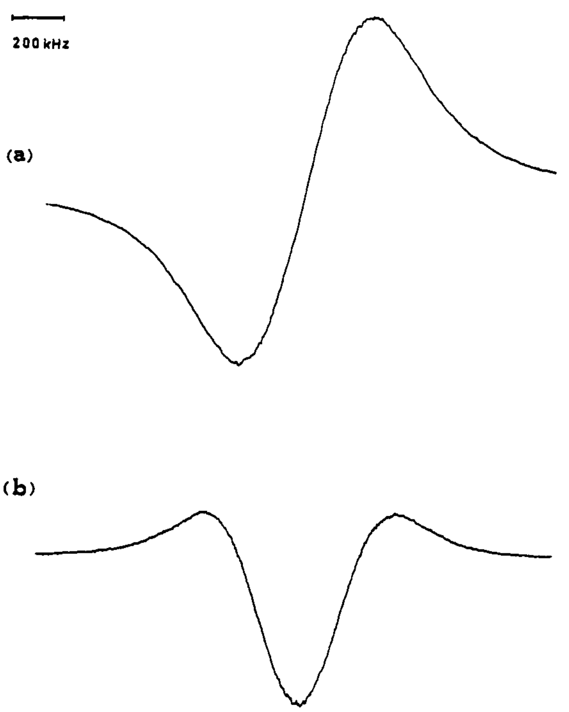

Fig. 10. — Lineshape of the a_6 resonance. (*a*) First harmonic (Ω = 50 kHz); (*b*) Second harmonic at $2\,\Omega$ = 100 kHz. Pump beam, 0.3 mW. Probe beam, 0.3 mW. Beam diameter, $2\,w$ = 2 mm.

resting to use this signal to lock successively $\Delta(t)$ on its two « zeros ». From the knowledge of these two frequencies and of the modulation amplitude δ, it is easy to calculate the experimental linewidth Γ. Our first experiments show that this method is very convenient for broadening measurements.

4. **Conclusion.** — We have shown experimentally that heterodyne spectroscopy through frequency modulation of the saturation beam is a very efficient method to increase the detection sensitivity in the domain of saturation spectroscopy. At the same time we have found that A/O M's are a very convenient tool for laser frequency and intensity control. The usual difficulties (wave front deformations and angular variations) can be easily solved by means of some complementary optical tricks. Lineshape studies which are of fundamental interest especially for metrological applications are greatly facilitated by these new methods.

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