# HETERODYNE SATURATION SPECTROSCOPY THROUGH FREQUENCY MODULATION OF THE SATURATING BEAM ☆

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Received 12 January 1982

We present a new spectroscopic method to detect saturated absorption signals. Its principle lies in a high frequency modulation of the saturation beam and a detection of the induced modulation of the probe combined with a frequency offset of the saturation beam. This method, which directly provides a dispersion-like lineshape, is very effective to increase the sensitivity close to the shot-noise limit and to cancel the various backgrounds.

In saturation spectroscopy, various methods have been recently proposed to improve the detection sensitivity. At the origin of these methods there are two major guiding ideas:

(i) The first one relies on an adequate choice of the local oscillator amplitude to reach the shot noise limit (saturated polarization [1], saturated interferometry [2]).

(ii) The second one consists in increasing the frequency of the detected signal in order to reach a frequency domain where the amplitude noise of the laser is negligible (high-frequency optically heterodyned spectroscopy [3], heterodyne saturation spectroscopy by means of phase modulation of the probe beam [4,5]).

All these methods are not generally able to completely cancel the background coming from either the linear absorption or the spurious interferometric fringes induced by imperfect optical isolation. Several additional methods can do it: for instance, the saturation chopper method [6] or the phase modulation of the pump beam by means of a vibrating mirror [7] eliminate the linear absorption. The pump-probe frequency shift [8,9] with the help of an Acousto-Optic (A/O) Modulator improves the optical isolation from the source.

We propose here a new method combining the advantages of high frequency detection with a very effective cancellation of all types of backgrounds. The saturation beam is both frequency-shifted by $\Delta$, and frequency-modulated at frequency $\Omega$ with the help of a A/O modulator and the induced amplitude modulation of the probe beam is detected (see fig. 1).

The saturating field is written as:

$$E_S = \tfrac{1}{2}\,\mathcal{E}_S \exp\{i([\omega + \Delta)t + (\delta/\Omega)\sin \Omega t + kz + \varphi]\} + \text{c.c.} \tag{1}$$

This field can be expanded into its Fourier components:

$$E_S = \tfrac{1}{2}\,\mathcal{E}_S \sum_{n=-\infty}^{+\infty} J_n(\delta/\Omega) \exp\{i[(\omega + \Delta + n\Omega)t + kz + \varphi)]\} + \text{c.c.} \tag{2}$$

where $J_n$ is the Bessel function of integer order $n$ and where $\delta/\Omega$ is the modulation index.

The counter-propagating probe field is:

$$E_P = \tfrac{1}{2}\,\mathcal{E}_P \exp[i(\omega t - kz)] + \text{c.c.} \tag{3}$$

☆ Work supported in part by D.R.E.T. and by B.N.M.
* Laboratoire associé au C.N.R.S., L.A. no. 282.

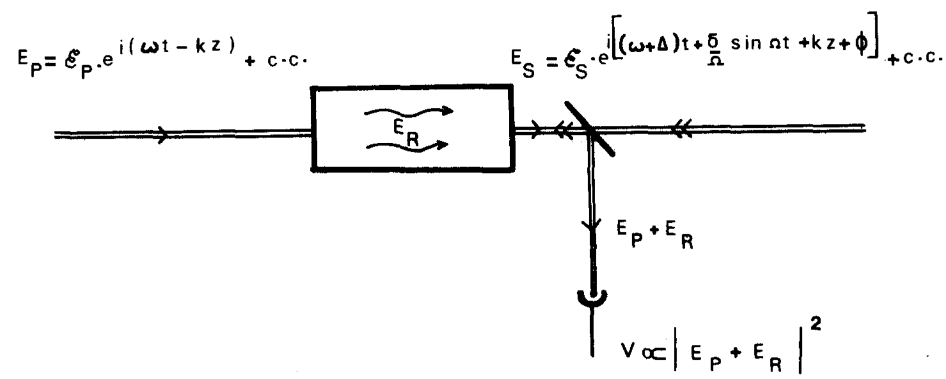


Fig. 1. Illustration of heterodyne saturation spectroscopy through frequency modulation of the saturating beam.

These two beams interact inside the sample cell to create a non-linear macroscopic polarization which reradiates an electromagnetic field, $E_R$, in the direction of the probe. This reemitted field exhibits sidebands at all harmonic frequencies, $\omega + p\Omega$:

$$E_R \propto \tfrac{1}{2} \sum_p \mathcal{E}_R(p) \exp\{i[(\omega + p\Omega)t - kz]\} + \text{c.c.} \tag{4}$$

where the amplitude $\mathcal{E}_R(p)$ may be complex.

As an example, in the framework of third-order perturbation theory, four-wave mixing processes induced by two arbitrary Fourier components of the pump beam ($\omega + \Delta + n\Omega$ and $\omega + \Delta + n'\Omega$) and the probe field yield two new re-radiated fields at frequencies $\omega \pm (n - n')\Omega$ [3]. In this simple approach, one easily understands that the two signal fields interfere with the probe on the detector to yield a heterodyne beat at the frequency $|n - n'|\Omega$. As it is now well-known [3,10–11], the re-emitted fields are not resonantly enhanced for the same laser frequency and thus the beat signal exhibits a doublet structure split by $|n - n'|3\Omega/2$. In the case of two-level systems ($E_a < E_b$), the intensity of the beat signal (i.e. the optical power per unit surface modulated at the frequency $(n - n')\Omega$) is given by [ref. [11], eqs. (31)–(32) and Appendix A]:

$$I(n,n') = \frac{\hbar\omega L}{16}\frac{\sqrt{\pi}}{ku}(N_a - N_b)\frac{|\mu^4 \mathcal{E}_S^2 \mathcal{E}_P^2|}{\hbar^4} J_n(\delta/\Omega) J_{n'}(\delta/\Omega) S(n,n') \exp[i(n-n')\Omega t] + \text{c.c.}, \tag{5}$$

where $ku$ = Doppler width, $N_\alpha$ = population density of level $\alpha$, $\mu$ = electric dipole matrix element, $L$ = interaction length, with the complex lineshape given by:

$$S(n,n') = \sum_{\alpha=a,b} \frac{C_\alpha}{\gamma_\alpha + i(n-n')\Omega}\left[\frac{1}{\gamma + i[\nu + (n - n'/2)\Omega]} + \frac{1}{\gamma - i[\nu + (n' - n/2)\Omega]}\right], \tag{6}$$

with $\nu = \omega - \omega_0 + \Delta/2$.

$\gamma_\alpha$ is the relaxation rate of level $\alpha$, $\gamma$ the optical dipole relaxation rate, $\omega_0$ the transition frequency and $C_\alpha$ is a geometrical coefficient depending on angular momenta and incident beam polarizations. Relation (6) is valid in the Doppler limit approximation. In $S(n, n')$, the first factor describes the population response to the incident modulation at frequency $(n - n')\Omega$, and the second one exhibits the doublet structure. A general expression of the signal modulated at frequency $p\Omega$ may therefore be written as:

$$S(p\Omega) = \sum_n J_n(\delta/\Omega) J_{n-p}(\delta/\Omega) S(n, n-p). \tag{7}$$

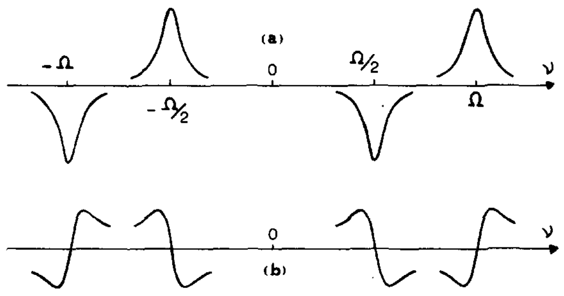


Fig. 2. Predicted lineshape for $\Omega \gg \gamma$, (a) absorption, (b) dispersion.

In particular, the modulation induced on the probe at frequency $\Omega$ is given by

$$S(\Omega) = \sum_{n,\alpha} J_n(\delta/\Omega) J_{n-1}(\delta/\Omega) \frac{C_\alpha}{\gamma_\alpha + \mathrm{i}\Omega} \left[ \frac{1}{\gamma + \mathrm{i}[\nu + (n+1)\Omega/2]} + \frac{1}{\gamma - \mathrm{i}[\nu + (n-2)\Omega/2]} \right]. \tag{8}$$

A noteworthy property is that the signal always has an odd symmetry with respect to the line center ($\nu = 0$). In the hypothesis where the modulation index is smaller than 1/2, the only important contributions come from $J_0$ and $J_{\pm 1}$ (The $J_1 J_2$ terms are $\lesssim 3\%$ of the $J_0 J_1$ contributions). Thus the signal lineshape is of the form

$$S(\Omega) \approx J_0(\delta/\Omega) J_1(\delta/\Omega) \sum_{\alpha=\mathrm{a,b}} \frac{C_\alpha}{\gamma_\alpha + \mathrm{i}\Omega} \left[ \frac{1}{\gamma + \mathrm{i}(\nu+\Omega)} + \frac{1}{\gamma - \mathrm{i}(\nu - \Omega/2)} - \frac{1}{\gamma + \mathrm{i}(\nu + \Omega/2)} - \frac{1}{\gamma - \mathrm{i}(\nu - \Omega)} \right]. \tag{9}$$

The predicted quadruplet structure appears either in absorption (fig. 2a) or in dispersion (fig. 2b), depending on the detection phase. When $\Omega$ and $\delta$ are small compared with the linewidth $\gamma$, $S$ reduces to the usual derivative of the saturated absorption signal:

$$S(\Omega) \approx -J_0 J_1 \sum_\alpha \frac{C_\alpha}{\gamma_\alpha + \mathrm{i}\Omega} \frac{2\Omega\nu\gamma}{(\gamma^2 + \nu^2)^2}. \tag{10}$$

Corresponding experiments have been performed on the 43–0 P(13) transition of the $^{127}I_2$ molecule with a frequency-stabilized single-mode $Ar^+$ laser [16]. A detailed description of the experimental set-up is given in ref. [12]. Fig. 3 gives the first (lowest frequency) main components of the spectrum of P(13). In this experiment, the overall frequency shift was $\Delta = 70$ MHz, the modulation frequency $\Omega = 50$ kHz and its amplitude $\delta = 500$ kHz. The beat signal was demodulated with a lock-in detection working at the same reference frequency $\Omega$. The $I_2$ cell was operating at a rather low pressure $P = 10$ mT for which the total linewidth is about 440 kHz (FWHM).

Typical powers were 1 mW for the pump and 0.5 mW for the probe. The beam diameter inside the cell was $2w$

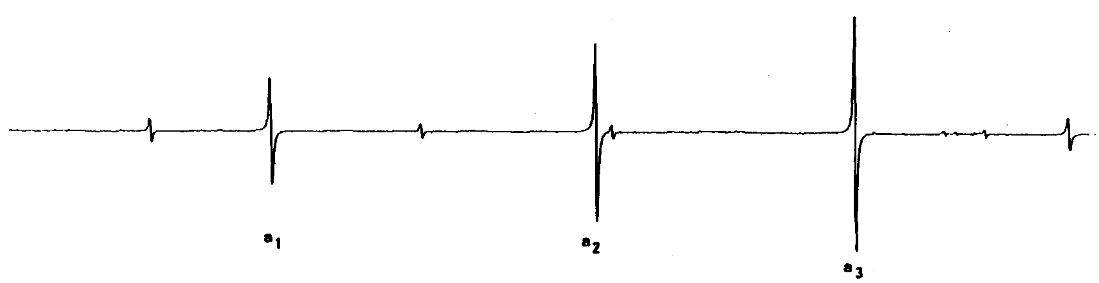


Fig. 3. Low-frequency region of the spectrum of $^{127}I_2$ corresponding to the transition 43–0 P(13) for $\Omega = 50$ kHz and $\delta = 500$ kHz, $P = 10$ mT which corresponds to a width equal to 440 kHz (FWHM). Frequency increases from left to right.

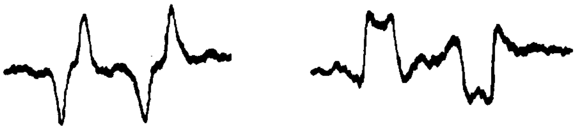


Fig. 4. Experimental lineshape of line $a_3$ when $\Omega$ = 2.4 MHz and $\gamma \approx$ 200 kHz ($P \approx$ 6 mT). (a) absorption, (b) dispersion.

$\approx$ 6 mm. In these conditions the signal looks like a single line whose shape is close to the derivative of a saturated absorption resonance (eq. (10)).

When the modulation frequency is increased to 2.4 MHz, the quadruplet is resolved and can be observed either in absorption (fig. 4(a)), or dispersion when the lock-in reference phase is changed by $\pi/2$ (fig. 4(b)). In this experiment the modulation index is about 0.2, for which value, eq. (9) closely describes the experimental results. We have verified that the resonance centers are located at the predicted values $\pm\Omega$, $\pm\Omega/2$.

For a careful analysis of the lineshape which is needed for metrological applications one should take into account a number of other physical effects like: the recoil effect, the second order Doppler effect, the beam geometry, collisions effects, etc. This development is outlined in Appendix A.

This new method is very simple to implement and only needs an A/O Modulator to yield a single dispersion-like line shape directly. This is obtained for moderate modulation frequencies ($\lesssim$50 kHz for $I_2$). At higher frequencies the medium cannot follow the modulation and the signal intensity decreases. This could be a limitation for dye lasers, but not for gas lasers for which it is possible to reach the shot noise limit at the mW power level for much lower modulation frequencies. A dispersion-like lineshape, which is very important in metrology and spectroscopy is also achieved with either ordinary frequency modulation of the laser or phase modulation of the probe beam only. However in the latter methods, the cancellation of the backgrounds (induced by probe linear absorption and optical feedback) is not complete and requires complementary tricks (pump modulation etc.).

The power of our method lies in its high capability to eliminate the various backgrounds and its insensitivity to any feedback of a residual amplitude modulation of the pump.

## Appendix A

One can start with expression (105) of ref. [13] which gives the gradient of the third-order change in the absorbed power for the probe beam as:

$$\frac{\mathrm{d}\overline{W}^{(3)}}{\mathrm{d}z} = -\hbar\omega \frac{\sqrt{\pi}}{ku}(N_\mathrm{a} - N_\mathrm{b}) \frac{\mu^4 \mathcal{E}_\mathrm{S}^2 \mathcal{E}_\mathrm{P}^2}{16\hbar^4} \left[\iint \mathrm{d}x\, \mathrm{d}y\, U_\mathrm{S} U_\mathrm{S}^* U_\mathrm{P} U_\mathrm{P}^*\right]$$

$$\times \Bigg\{ 4 \sum_{m,m',n,n' \atop -\infty}^{+\infty} \sum_{\alpha=\mathrm{a,b}} C_\alpha J_m(\delta_\mathrm{P}/\Omega_\mathrm{P}) J_{m'}(\delta_\mathrm{P}/\Omega_\mathrm{P}) J_n(\delta_\mathrm{S}/\Omega_\mathrm{S}) J_{n'}(\delta_\mathrm{S}/\Omega_\mathrm{S})$$

$$\times \mathrm{Re}\Bigg( \exp\{\mathrm{i}[(m-m')\Omega_\mathrm{P} + (n-n')\Omega_\mathrm{S}]t\} \int_0^{+\infty} \mathrm{d}v_\mathrm{r}\, F(v_\mathrm{r}) \int_0^{+\infty} \mathrm{d}\tau \int_0^{+\infty} \mathrm{d}\tau'$$

$$\times \exp[-v_\mathrm{r}^2(A\tau'^2 + 2B\tau\tau' + C\tau^2)]\, \exp[-(\gamma_\alpha + \mathrm{i}(n-n')\,\Omega_\mathrm{S})\tau']$$

$$\times \exp[-2(\gamma + \mathrm{i}(\omega_0 - \omega) + \mathrm{i}(\tfrac{1}{2}n - n')\,\Omega_\mathrm{S} - \mathrm{i}\,\tfrac{1}{2}m'\Omega_\mathrm{P})\tau] \Bigg)\Bigg\},$$

where we have used the notations and conventions of the present paper. We have introduced two distinct modulation frequencies $\Omega_S$, $\Omega_P$ and two modulation amplitudes $\delta_S$, $\delta_P$ for the saturation beam and for the probe beam; $F(v_r)$ is the transverse velocity distribution and the coefficients $C_\alpha$ take into account the laser polarization and the level degeneracy [14].

The functions $U_S$ and $U_P$ describe the transverse gaussian dependence of the laser beams. In the plane wave limit ($U_S \equiv U_P \equiv 1, A = B = C = 0$) the previous integrals are easily evaluated:

$$\left\{\quad\right\} = 4 \sum_{\substack{m,m',n,n' \\ \alpha=a,b}} J_m(\delta_P/\Omega_P) J_{m'}(\delta_P/\Omega_P) J_n(\delta_S/\Omega_S) J_{n'}(\delta_S/\Omega_S)$$

$$\times \mathrm{Re}\Big(\exp[\mathrm{i}\{(m-m')\Omega_P + (n-n')\Omega_S\}t]$$

$$\times \frac{C_\alpha}{\gamma_\alpha + \mathrm{i}(n-n')\Omega_S} \, \frac{1}{2\gamma + 2\mathrm{i}(\omega_0 - \omega) - \mathrm{i}m'\Omega_P + \mathrm{i}(n-2n')\Omega_S}\Big).$$

We recover the results of the text by setting the probe wave modulation index equal to zero and by a $\Delta/2$ shift of $\omega$:

$$\left\{\quad\right\} = \sum_{n>n'} J_n(\delta/\Omega) J_{n'}(\delta/\Omega) \exp\{\mathrm{i}(n-n')\Omega t\} \frac{C_\alpha}{\gamma_\alpha + \mathrm{i}(n-n')\Omega}\left[\frac{1}{\gamma - \mathrm{i}\{\nu + (n' - n/2)\Omega\}}\right.$$

$$\left. + \frac{1}{\gamma + \mathrm{i}\{\nu + (n - n'/2)\Omega\}}\right] + \sum_n J_n^2(\delta/\Omega) \frac{C_\alpha}{\gamma_\alpha} \, \frac{1}{\gamma + \mathrm{i}\{\nu + n\Omega/2\}} + \text{c.c.}$$

Similarly one can also derive the lineshape for experiments where only the probe beam is frequency modulated by setting $\delta_S$ equal to zero. In the general case of gaussian laser beams the coefficients $A, B, C$ are obtained from their expressions in ref. [13] and integrations over $v_r$ and $\tau'$ are performed as in the absence of modulation.

The recoil structure can be easily taken into account by the simple following replacement rule [13,14].

$\omega_0 \to \omega_0\{1 + \hbar\omega_0/2Mc^2\}$ in the term involving the lower level population,

$\omega_0 \to \omega_0\{1 - \hbar\omega_0/2Mc^2\}$ in the term involving the upper level population.

The transverse Doppler effect is taken care of, by replacing $\omega$ with [13,14]

$\omega/(1 - v^2/c^2)^{1/2} \approx \omega(1 + v_r^2/2c^2)$.

Finally elastic collisions are also introduced by the simple modification of the relaxation constants described in ref. [15].

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