### Laser Frequency Stabilization by Means of Saturation Dispersion

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A method for frequency stabilization of 3.39  $\mu m$  He-Ne lasers, which makes use of the saturation dispersion of the  $v_3$ , P(7) methane transition is described. It is shown that with lasers which are stabilized by saturation effects, the absorptive and dispersive properties of the saturation resonance are not precisely distinguishable. This leads to a slightly asymmetric resonance and a shift of the apparent line-center. When using the dispersion for stabilization, the asymmetry can be detected and eliminated. Frequency stabilization to the true line center with an accuracy of  $5\times 10^{-12}$  was attained.

#### 1. Introduction

Saturated absorption provides a means of stabilizing the output frequency (or wavelength) of a gas laser, and thereby renders possible frequency standards in the visible and infrared parts of the spectrum.

For frequency standards the main objective is not necessarily to produce the narrowest resonance lines possible, as it would be in high resolution spectroscopy, but rather to find and to tune to the center of the resonance with minimum uncertainty. As long as the observed resonance is symmetric, this is only a question of signal-to-noise ratio and precise electronics. With the methane-stabilized He-Ne lasers as first described by Barger and Hall <sup>1</sup>, the signal-to-noise ratio is so high, however, that the random fluctuations are small compared to systematic effects, mainly caused by asymmetric distortions of the line, already at averaging times of less than a millisecond, at a level of uncertainty of about  $10^{-10}$  to  $10^{-11}$ .

Some of the asymmetric distortions are caused by the molecules themselves, like the unresolved hyperfine structure<sup>2</sup>, and possibly the second order Doppler and the photon recoil effect.

In this paper we deal with those asymmetries which are introduced by the particular method of observation. We analyze the performance of conventional servo systems, and describe a dispersion based stabilization scheme that overcomes most of the difficulties of saturated absorption line distortions.

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# 2. Saturated Absorption and Dispersion in Methane

The power output of a 3.39 µm wavelength He-Ne laser with an intracavity absorption cell filled with 1 Pa of methane shows a narrow (250 kHz HWHM)<sup>3</sup> peak at the center of the  $\nu_3$ , P(7) methane line. This peak, also called an inverted Lamb-dip, is caused by the increased saturation occurring in a standing-wave field. Fig. 1 shows how the power output (lower trace) and the frequency of such a laser vary, as its resonator length is scanned by means of a piezoelectric mirror translator. The zero lines of both curves are suppressed: The height of the saturation peak is only about 2% of the total power. The frequency trace was produced by generating a beat signal of approximately 5 MHz with a second laser and passing it through a frequency-to-voltage converter.

The curves show the two aspects of the saturation resonance: absorption and dispersion. As absorption and dispersion have different symmetries, incomplete separation of the two leads to asymmetrically distorted line-shapes.

When using either the absorption or the dispersion for frequency stabilization of a laser, these asymmetries are observed as a modulation-width dependent shift of the stabilized frequency.

### 3. Gas Lens Effect

As the saturation of the absorber depends on the strength of the saturating field, the Gaussian intensity distribution of the field within the laser causes the absorbing gas to form a distributed lens of frequency dependent focal length. This results in a frequency dependent mode geometry in the laser and,



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as a consequence, in a frequency dependence of the power output, reflecting the dispersion line shape.

The apparent saturated absorption line profile will therefore be a superposition of an absorption line and a weak dispersion line.

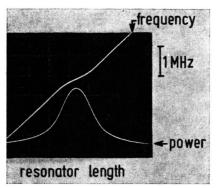


Fig. 1. Frequency and output power of a 3.39  $\mu m$  He-Ne laser with an internal methane absorption cell as a function of the resonator length.

From Fig. 1 one can find the maximum frequency deviation introduced by the saturation resonance to be  $\Delta f = \pm 150 \,\mathrm{kHz}$ . With a cavity length  $L = 60 \,\mathrm{cm}$ and  $f_0 = 88 \text{ THz}$  this means, that the optical length of the absorption cell changes by 2 nm as the frequency is tuned from one side of the resonance to the other. Although due to the geometry of the laser mode this is an average value, it is sufficient for a first estimate to assume that the change is 2 nm on the axis and zero at one beam radius from the axis. Concentrated in front of a mirror of radius of curvature R = 5 m, where the mode radius is 1 mm, this radial dependence of optical path length would be equivalent to a change of the mirror radius from 4.95 m to 5.05 m. With the geometry of our lasers  $(R_1 = 5 \text{ m}, R_2 = 2.5 \text{ m}, L = 0.6 \text{ m})$  this would result in a change of the mode radius by 0.2% at the far end mirror.

Clearly these changes in mode size can effect the power output of the laser in several ways. An increase of the mode radius may increase the losses at beam limiting apertures, but it may as well increase the available power through a better filling of the plasma volume.

As the saturation peak is only 2% of the total power, with a power output proportional to the mode volume, the gas lens effect would shift the top of the line by as much as 10% of the HWHM.

Analysis of the expected superposition of absorption and dispersion lineshapes is complicated by the asymmetries introduced by the baseline tilt.

#### 4. Tilted Baseline and Dispersion

For taking the picture shown in Fig. 1, the laser and the detector diode were adjusted in such a way as to produce an apparently symmetric lineshape. In general, however, the saturation peak is observed on top of a more or less tilted baseline. This tilt may have at least three different origins:

- 1) The saturation peak does not coincide with the center of the gain curve.
- 2) Reflections from the detector diode or other surfaces give rise to interference fringes.
- The piezoelectric mirror translator slightly tilts the mirror.

With a servo-system using sinewave frequency modulation and first harmonic synchronous detection, the baseline tilt leads to a frequency shift, which also depends on the modulation width. As a number of parameters influence the magnitude of the baseline tilt, and as some of them are not stable with time, the baseline is also responsible for the drifts and fluctuations limiting the medium term frequency stability of these lasers. It has been proposed and reported by several authors 4 that thirdharmonic detection be used to suppress the influence of the baseline. This would work perfectly if one could produce a pure sinewave frequency modulation. If one simply applies a sinewave voltage to the piezoelectric mirror translators, however, the frequency modulation is strongly distorted by the dispersion of the saturation line. Although this distortion leaves the resonance itself symmetrical, the baseline, now as a function of resonator length instead of frequency, is no longer a straight line, which would be ignored by the third harmonic lock, but takes the shape of the upper trace in Figure 1.

Figure 2 shows the calculated frequency shift produced by a baseline tilt for sinewave resonator length modulation and first and third harmonic synchronous detection. Lorentzian lineshapes <sup>5</sup> were assumed for both absorption and dispersion with magnitudes as observed with our lasers.

The result shows that in this case third harmonic detection gives only a slight improvement and does not suppress the effect of a tilted baseline.

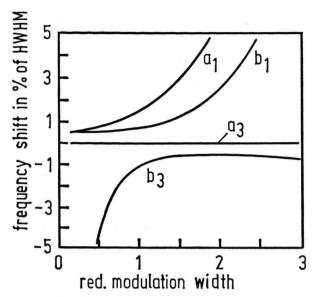


Fig. 2. Calculated combined effects of dispersion and tilted baseline for sinewave resonator length modulation and first and third harmonic sinewave detection. The reduced modulation width equals 1, if the peak length deviation produces a frequency deviation equal to the HWHM in an empty resonator. — Subscripts 1 and 3 denote first and third harmonic detection. — The base-line slope was chosen to be one per cent of the line height per HWHM. With no dispersion, (curves a<sub>1</sub> and a<sub>3</sub>) the third harmonic lock completely eliminates the effect of the base-line tilt. If the dispersion is as strong, however, as in our lasers the frequency shift (curves b<sub>1</sub> and b<sub>3</sub>) has about the same magnitude for the third harmonic lock as for the first harmonic.

A controlled-frequency modulation using a second laser as a reference is difficult to implement because of the relatively slow response of the piezoelectric tuning elements.

## 5. Frequency Stabilization Using Saturation Dispersion

The difficulties encountered in the attempt to observe the pure saturated absorption line-shape make it seem advantageous to use the dispersion rather than the absorption for finding the true center of the resonance. A quantitative picture of the saturation dispersion is given in Figure 3. It shows the measurement of the first derivative  $\mathrm{d}f/\mathrm{d}l$  of the laser frequency f to the resonator length l as a function of laser frequency. This was measured by modulating the resonator length and using an up-down-counter for synchronous detection, while a controlled frequency sweep was performed against a stabilized reference laser. It can be seen that  $\mathrm{d}f/\mathrm{d}l$ 

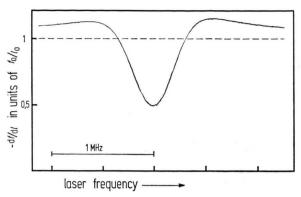


Fig. 3. Measured first derivative df/dl of laser frequency to resonator length as a function of laser frequency.

at line center is only half the value of  $\mathrm{d}f/\mathrm{d}l$  far away from the center.

The dependence of laser frequency on resonator length can be used to derive the error signal for a loop that stabilizes the laser frequency to the minimum of the curve in Fig. 3, which, except for the asymmetric distortions represents the same frequency as the maximum of the saturated absorption line.

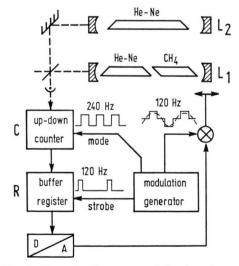


Fig. 4. Dispersion frequency stabilization scheme.

The error signal is derived in the following way: The frequency of the laser is sampled at three different resonator lengths:  $l_0$ ;  $l_1 = l_0 + \Delta l$  and  $l_2 = l_0 - \Delta l$ . The frequencies corresponding to the three lengths are  $f_0$ ,  $f_1$  and  $f_2$ . If  $f_0$  coincides with the frequency of the central turning-point of the frequency vs. resonator length curve (Fig. 1), then  $f_1$  and  $f_2$  will be equally spaced about  $f_0$ , i.e.  $f_1 + f_2$ 

 $=2f_0$ . If  $f_0$  is not the turning-point frequency, then  $F=f_1+f_2-2f_0$  can be used as the error signal.

The set-up for frequency-stabilizing a laser is shown in Figure 4. The resonator length of laser  $L_1$  is periodically set to the three values by means of a piezoelectric mirror translator. Its frequency is measured as the beat frequency with the free-running laser  $L_2$  using an up-down-counter. The counting direction of this counter is controlled at twice the modulation frequency, so that after one modulation period its content is proportional to  $F = f_1 + f_2 - 2 f_0$ .

Although we have used this 3-point interrogation scheme, which gives better short term stability, most of the measurements were carried out with sinewave modulation in order to avoid distortions. With f(l) representing the frequency vs. resonator length curve of the laser, and  $l(t) = l_0 + l_{\rm m} \sin \omega_{\rm mod} t$  describing the modulation, the contents of the counter in this case is:

$$N = \int [f_1(l_0 + l_m \cdot \sin \omega_{\text{mod }} t) - f_2] \cdot \operatorname{sgn}(\cos 2 \omega_{\text{mod }} t) \cdot dt.$$

As the counter is not reset to zero, it also acts as the integrator in the servo loop. Its content is peridodically transferred to the buffer register R, thereby suppressing the modulation frequency and its haramonics, and converted to an analog signal, which — after some filtering — controls the resonator length.

All measurements were performed using the setup shown in Figure 5. Lasers  $L_1$  and  $L_3$  contain an absorption cell. The output of both lasers is mixed

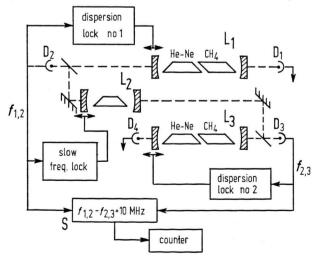


Fig. 5. Set-up for measuring the frequency stability and frequency shifts of dispersion-locked lasers.

with the output of a common local oscillator  $L_2$  by means of Photodiodes  $D_2$  and  $D_3$ .  $L_1$  and  $L_3$  are stabilized to the turning point of the dispersion curve as described above.  $L_2$  is frequency controlled at about 3 MHz from the frequency of  $L_1$  by a slow frequency control loop that ignores the modulation of  $L_1$ . The slow control loop prevents the beat frequencies from exceeding the frequency range of the up-down-counters. The beat frequencies  $f_{1,2}$ ,  $f_{2,3}$  generated by  $D_2$  and  $D_3$  are subtracted by a digital frequency subtractor S. 10 MHz are added to the frequency difference so that the sign of the latter can be recognized. The output of S is processed by a computing counter,  $D_1$  and  $D_4$  serve to monitor the saturated absorption line-shapes.

To measure the frequency stability of the dispersion locked lasers, the square root of the two-sample Allan variance  ${}^6\sigma_y(\tau)$  was recorded.

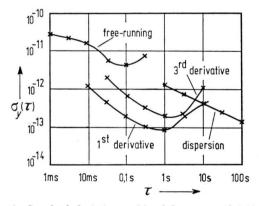


Fig. 6. Standard deviation  $\sigma_y(\tau)$  of frequency of 3.39  $\mu m$  He-Ne lasers, free-running and stabilized by three different methods as a function of averaging time  $\tau$ .

The result is shown in Figure 6. For comparison, measurements for the same lasers controlled to the center of the saturated absorption line either by using the first or third derivative of the absorption line profile, are also shown.

Although the short term frequency stability of the dispersion locked lasers is inferior to that of the absorption locked lasers, the long term stability of the dispersion locked lasers is superior. We attribute this to the elimination of the influence of the "tilted baseline".

#### 6. Asymmetry of the Dispersion Line

The stabilized frequency of a dispersion locked laser depends on the modulation width, as can be seen from the measurement Figure 7 a. The possibility that this might be caused by a non-linearity of the piezotranslators was ruled out by measurements of the non-linearity with the up-down counter. The non-linear distortions of the system consisting of sine-wave generator, phase-shifter, high voltage amplifier and piezotranslator does not exceed 0.01% at a modulation width of 2 MHz. The dependence of stabilized frequency on modulation width must therefore be caused by an asymmetry of the dispersion line.

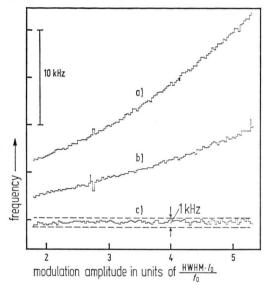


Fig. 7. Modulation width dependent frequency shifts of a dispersion stabilized laser (curve a). Curve b was taken with the output power regulated, curve c with optimum compensation.

As we expect the asymmetry to be introduced by refractive index changes with laser power in the gain cell, an attempt was made to eliminate the asymmetry by regulating the power to a value independent of laser frequency, using the output signal of D<sub>1</sub> to control the discharge current through the gain cell. The dependence of frequency on the modulation width is in this case indeed weaker (Fig. 7b), but the asymmetry is not removed completely, since changing the discharge current changes e.g. the electron density in the gain tube and consequently introduces new sources of refractive index changes.

We can on the other hand expect the optical path length changes in the gain tube to be proportional to the laser power for small power variations. It should therefore be possible to compensate for the refractive index change by an equivalent change in resonator length, i. e. by adding a certain amount of the absorption signal from D<sub>1</sub> and D<sub>4</sub> to the voltage applied to the piezotranslators. Curve c in Fig. 7 shows that with the right amount of absorption signal added, the stabilized frequency becomes independent of modulation width, indicating that the asymmetry has the expected absorption line shape.

The compensation of the asymmetry should therefore not only remove the dependence of stabilized frequency on modulation width but also result in a stabilization to the true line center, within the uncertainty of the compensation. With our set-up this uncertainty is about 600 Hz.

The measured frequency difference of two lasers, which were individually adjusted to emit modulation width independent frequencies, was indeed found to be smaller than 500 Hz. The two lasers have different resonators and they were operated at different power levels, so that an incidental equality of the stabilized frequencies seems unlikely.

In conclusion we note that: Absorptive and dispersive properties of the resonance to which a laser is to be stabilized may become mixed to some extent, leading to an asymmetric resonance. When dispersion is used for stabilization, the asymmetry can be detected and overcome by compensation. Stabilizing to the true line center is possible.

In view of the well-known hyperfine structure 2 of the  $v_3$ , P(7) methan transition used, which leads to a power dependent shift of the apparent line center of about 10<sup>-12</sup> per 10% power change, no further attempt was made to improve on the present accuracy of 5:10<sup>-12</sup> by improving the compensation technique.

Applied to an unsplit line, however, the frequency stabilization by means of saturation dispersion should be much better suited to finding the center of a spectral line, than are the stabilization schemes which wave been used so far.

<sup>&</sup>lt;sup>1</sup> R. L. Barger and J. L. Hall, Phys. Rev. Lett. 22, 4 [1969].

<sup>&</sup>lt;sup>2</sup> J. L. Hall and C. Bordé, Phys. Rev. Lett. 30, 1101

<sup>3</sup> Half width at half maximum.

<sup>&</sup>lt;sup>4</sup> See e.g. A. J. Wallard, J. Phys. E 5, 926 [1972].

<sup>&</sup>lt;sup>5</sup> Absorption line shape:  $L_{\rm A} \propto 1/(1+\Omega^2)$ , dispersion line

shape:  $L_D \propto \Omega/(1+\Omega^2)$ . J. A. Barnes et al., IEEE Transact. Inst. Meas. IM-20, 105 [1971].