# LASERS AS STABLE FREQUENCY SOURCES

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## Abstract

The development of low noise gas lasers and the advent of Doppler free spectroscopy techniques has opened up the possibility of building frequency and length standards in the optical domain. The problems encountered, the present state of the art and the application field of stabilized lasers are briefly reviewed.

## I. Generalities

The oscillation frequency $\nu$ of a single mode free running gas laser is primarily determined by the length L of its resonant cavity, according to the relation $\nu = \frac{C}{2L}$ , when n is an integer, and C is the speed of light. Such a laser when placed in good environmental conditions can exhibit a very high short term frequency stability [1]. However, the oscillation frequency can lie anywhere within the amplification bandwidth of the active medium (Fig.1) and

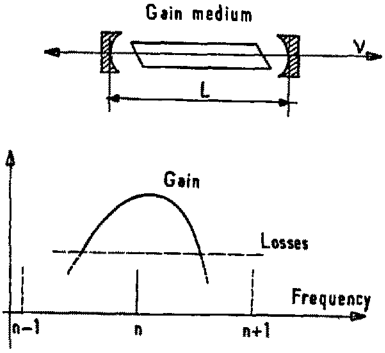

Fig.1 Scheme of a single mode free running laser

no significant accuracy can be defined for this device. To obtain a frequency standard, it is therefore necessary to lock the output frequency $\nu$ to some absolute frequency reference $\nu_{ref}$, the best of which is the center of a suitable atomic or molecular absorption line. It is customary to characterize a stabilized oscillator by its :

- stability (the degree to which it will produce the same frequency over a period of time in continuous operation)
- reproducibility (the degree to which it will produce the same frequency from unit to unit, of from one run to another)
- accuracy (the degree to which the difference between the delivered frequency and the frequency of the corresponding unperturbed reference line can be evaluated)

Stability and reproducibility call for the narrowest possible linewidth $\Delta\nu$ of the reference line compatible with other constraints such as signal to noise ratio.

The absorption linewidth in low pressure molecular gas of interest here is essentially determined by the Doppler effect due to molecular thermal motion. In the optical domain, $\frac{\Delta\nu}{\nu_{ref}} \simeq 10^{-6}$ which is too large to obtain any interesting figures for the metrological qualities listed above.

It is thus necessary to have a means of observing absorption lines without any Doppler broadening. Three techniques have been investigated up to now for this purpose :

- saturated absorption (S.A.) [2]
- use of molecular beams excited by a light beam perpendicular to it [3]
- two photons absorption [4].

All three techniques lead to absorption lines with a relative linewidth $\sim 10^{-9}$ suitable for frequency stabilization. However, saturated absorption has been the most studied and we will mainly discuss this case.

## 2. The Saturated Absorption Technique

We assume that a Doppler-broadened transition between two levels a, b ($E_a - E_b = h\,\nu_{ref}$) is submitted to a monochromatic standing light wave (Fig.2).

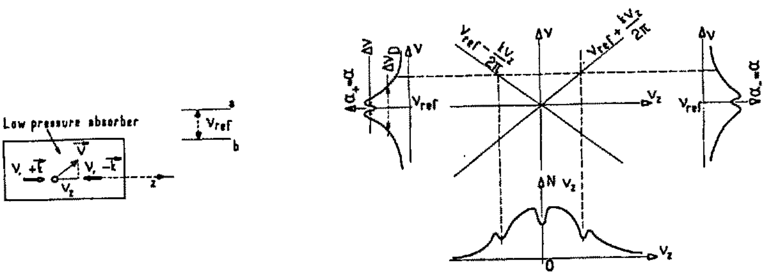

Fig.2 The saturated absorption technique in a strong monochromatic standing light wave, and the shape of the absorption coefficient of the medium for light waves propagating along the + z and - z directions.

The field interacts, resonantly with molecules whose velocity satisfy the condition

$$\nu = \nu_{ref} \pm \frac{k\ v_z}{2\pi} \qquad (1)$$

where $\nu$ is the frequency of the laser field, $\pm\ v_z$ the projection of the velocity of the molecule on the direction of propagation of each of the travelling waves forming the standing wave. It the amplitude of the field is sufficient to change the level population (to saturate), two holes are then burned in the population difference distribution $N(v_z)$. When the field frequency is tuned, the holes overlap. Both travelling waves change the population difference with $kv_z = 0$. The degree of saturation is twice as large in this case. The dip which appears in the center of the Doppler absorption contour $\alpha_+ = \alpha_- = \alpha$ is called the saturated absorption dip . The ratio of its width compared to the Doppler width is typically $10^{-2}$ or $10^{-3}$.

The saturated absorption signal can be observed directly when the absorber is outside the laser source (Fig.3).

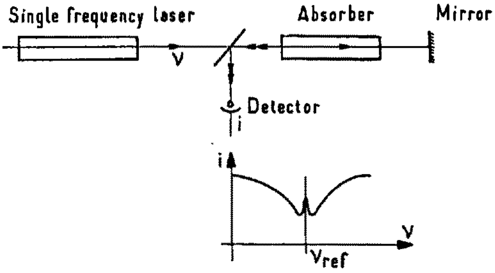

Fig.3 External cell saturated absorption set-up

The observation of narrow resonances of saturated absorption can also be made when the absorber is placed inside the laser resonator. When such a situation occurs, it is obvious that the amplifying medium must satisfy the following conditions :

1) the absorption line must coincide with the gain line of the active medium or lie inside the gain profile.

2) the laser emission intensity must be sufficient to saturate the absorber.

Figure 4 shows that the dip in the absorption line leads to a maximum of transmission. The "inverted Lambdip" which appears on the laser output power profile at $\nu = \nu_{ref}$ is suitable for frequency stabilization.

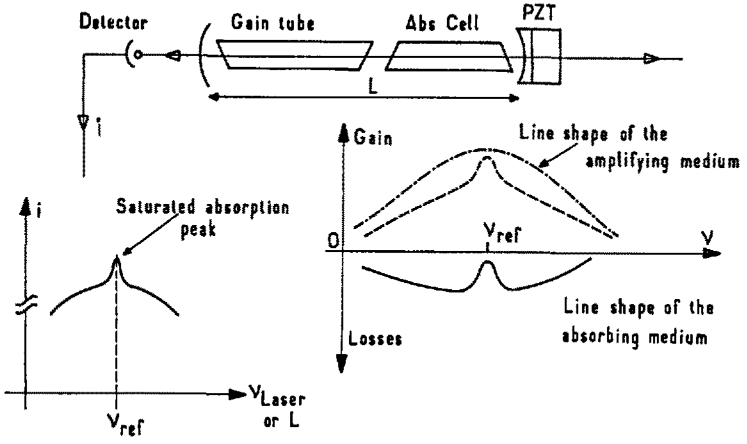

Fig.4 Laser with a non linearly absorbing medium inside the resonator

## 3. Principle of the Servo System Used to Stabilize the Laser Frequency

The experimental set-up enabling such a stabilization is depicted on Fig.5. A small frequency modulation is applied to the laser. This modulation

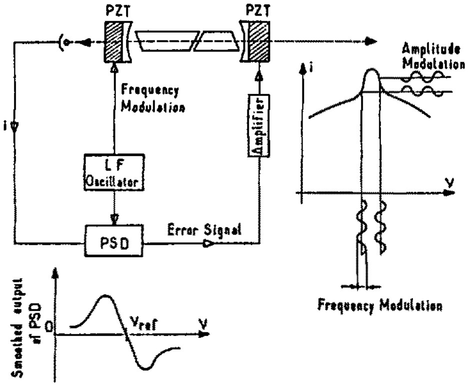

Fig.5 Principle of the servo system used to stabilize the laser frequency.

results in an intensity modulation of the laser output power. The intensity modulation is fed in a phase sensitive detector and suitably smoothed. A DC error signal is obtained. A servo system controls the laser length so as to maintain zero PSD voltage. The laser frequency is continually referred to the centre frequency of the saturated peak and perturbations of the optical frequency will automatically be compensated for.

## 4. Performances

Several practical optical frequency standards based on this scheme have been studied. Up to now, they use low noise single mode gas lasers and non linear absorber molecules whose absorption frequency coincides accidentally with the laser gain line. They are :

- the infrared He-Ne laser stabilized to a saturated absorption line of $CH_4$ at 3.39 µm [5],
- the visible He-Ne laser stabilized to S.A.L. of iodine at 0.612 µm and 0.633 µm [6],
- the visible $Ar^+$ laser stabilized to S.A.L. of iodine at 0.5145 µm [7],
- the infrared $CO_2$ laser stabilized to S.A.L. of $CO_2$, $SF_6$, $OsO_4$ at 10.6 µm [8].

### 4 .1. Frequency Stability

Frequency stability as a function of the observation time has been determined for these standards in different laboratories using a beat frequency method between two stabilized lasers operating under similar conditions. Fig.6 is a plot of the results in the form of Allan variance curves. The long term

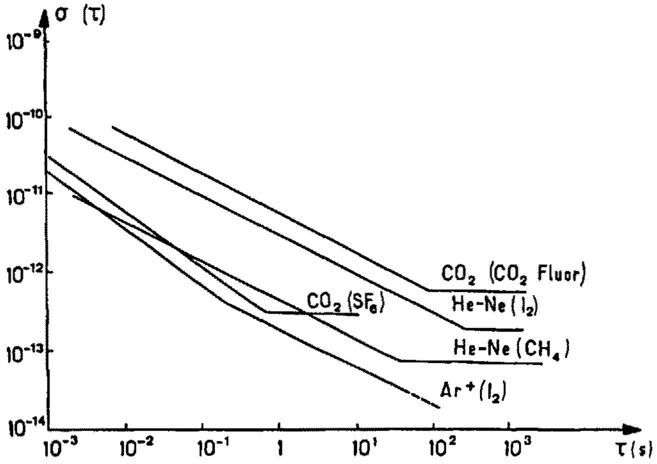

Fig.6 Relative frequency stability curves as a function of the observation time τ.

frequency stability is reaching parts in $10^{13}$ for $SF_6$ [9] and Iodine [10], and even parts in $10^{14}$ for $CH_4$ [11].

### 4.2. Reproducibility

The main effects which limit the reproducibility (and also accuracy) of frequency stabilized lasers are the following :

. Some are clearly of technological origin, such as electronic offsets in the servo system or modulation distorsions [12].

. Others have a physical origin depending on the system under considerations such as frequency shifts due to molecular collisions (Iodine) or to an unresolved hyperfine structure ($CH_4$) [13].

The present reproducibility of these optical frequency standards is $\simeq 10^{-11}$ Let us mention that it does not reach the fundamental limitation ($< 10^{-12}$) which are linked to the molecular photon interaction. With the saturated absorption technique, these fundamental limitations are [14] :

- the residual first order Doppler effect not completely eliminated by the saturated absorption technique [15],
- the second order Doppler effect [16],
- the recoil effect which gives rise to a splitting of the saturated absorption lines into two close peaks unresolved in conventional devices [17].

### 4.3. Accuracy

At the present time, the accuracy of small Iodine conventional saturated absorption devices may be estimated at about a few $10^{-11}$. In the case of small $CH_4$ devices, it is even unsuitable to give a significant figure because the emitted frequency does not correspond to a given transition between two $CH_4$ energy levels.

## 5. Applications

### 5.1. Metrology

The stabilized He-Ne lasers are already applied as standards of wavelength. Their reproducibility is far superior to that of the primary length standard, the Krypton lamp.

The absolute vacuum wavelength of the Methane and Iodine stabilized He-Ne lasers have been measured by direct interferometric comparison with the $^{86}$Kr primary standard line [18]. A by product of these precise measurements has reveal an uncertainty of ± $4 \times 10^{-9}$ in the 1960 definition of the meter.

By measuring both the frequency and the wavelength of the stabilized He-Ne laser at 3.39 µm [19], one obtains an experimental value for the speed of light recommended to be $C = \lambda f = 299\ 792\ 458$ m/s in 1973 by CCDM.

The main source of error is due to the uncertainty in the definition of the meter which, when reported on the speed of light, gives an error of ± 1.2 m/s. This value is 100 fold smaller than the former determination of C. One can predict that the speed of light and stabilized lasers will play a central role in length metrology. May be they will contribute to a new definition for the meter.

## 5.2. Geophysics

Geophysical research uses stabilized lasers as light sources for strain meters where the movement of rocks over long periods are studied [20]. In this experiment, a slave He-Ne laser is frequency locked to the transmission maxima of a long Fabry Perot interferometer. The frequency of the laser is therefore related to the length of the interferometer by $f = n \frac{C}{2L}$ where n is an integer, C is the velocity of light and L is the length of the interferometer. Then $\frac{\Delta f}{f} = - \frac{\Delta L}{L}$. This laser is compared with a reference stabilized He-Ne laser and the resulting beat frequency variations are a direct measure of the earth strain variations. A beat frequency shift of 10 MHz between two visible He-Ne lasers in an interferometer 30 m long correspond to a strain of 2. $10^{-8}$. This apparatus has been used to detect earthquakes [21].

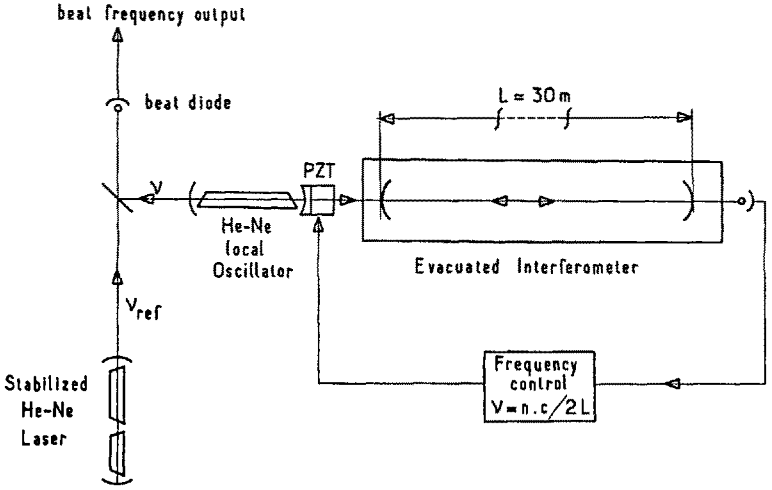

Fig.7 A Strain Meter Design

Figure 8 illustrates the sensitivity of the method. We can see the initial arrival of a burst of seismic energy and subsequent arrivals after the pulse has travelled around the surface of the earth. Pulses up to the fifth transit are visible on the record.

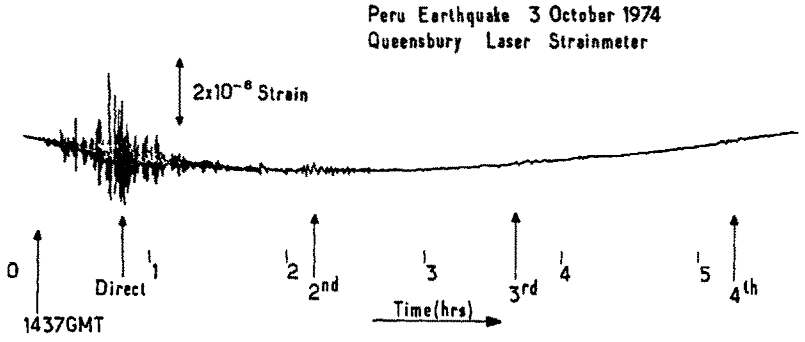

Fig.8 Record of the earth strain variations during an earthquake. From [21].

## 5.3. Gravity measurements

Exact knowledge of gravitational acceleration is important for many measurements (barometry, geological and mining prospection, determination of daily and secular g variations.

The principle of the absolute measurement of g is the following : we use the fundamental law of dynamics $e = \frac{1}{2} g t^2$ which applies for a body falling in vacuum. e represents the distance, while t is the duration of the fall. The distance measured by a Michelson interferometer is expressed by the number of interference fringes of a laser beam reflecting upon an optical corner which constitutes the moving body. The time is measured by mean of a high stability clock (Rb clock). The accuracy of g measurement is $10^{-9}$ [22].

## 5.4. Relativity tests

New versions of the fundamental tests of relativity based on the Michelson-Morley [23] and Kennedy Thorndike [24] experiments can be realized using frequency metrology with stabilized lasers instead of interferometry. A recent version of the Michelson-Morley experiment demonstrates a gain in sensitivity by a factor of 12 000 [25].

## 5.5. Frequency stabilized CW dye lasers for spectroscopic applications

The tunable CW dye laser has become an increasingly important laboratory source of light for spectroscopic and even metrological purposes. Sources suitable for sub-Doppler atomic spectroscopy where one is dealing with linewidths of ∿ 10 MHz are commercially availables. But the much narrower and stable dye source needed for molecular high resolution spectroscopy requires many man hours of technology [26]. The spectrum of the frequency fluctuations of a dye laser turns out to be essentially conditionned by the perturbations occuring in the jet stream region. The low frequency fluctuations (0 - 1 KHz) can be ascribed to thermal drifts, drafts, jet stream thickness fluctuations due to vibrations in the dye circulator. They result in a frequency jitter of a few MHz and a drift of ∿ 1 MHz/minute, provided elementary precautions are taken for vibrations and acoustical isolation. In an intermediate frequency range (tens of KHz) the mechanical resonances of the nozzle can induce a line broadening of 150 KHz. We have also high frequency noise which we think to be due to surface waves in the dye stream.

In order to obtain a high short and long term frequency stability together with some tunability of the laser, it is necessary to use 3 successives servo loop whose principle is depicted in Fig.9.

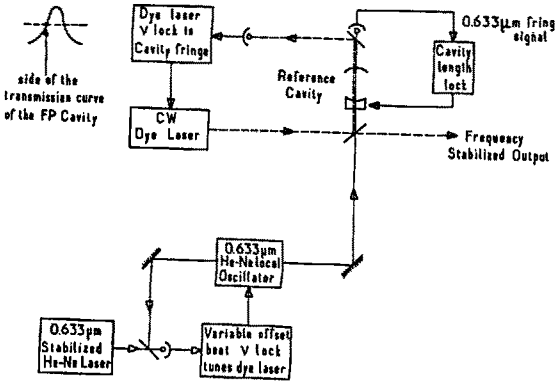

Fig.9 Experimental set-up used to stabilize the frequency of a CW dye laser

The first fast loop acting on the cavity output mirror PZT locks the dye laser frequency to the side of the transmission curve of an external reference cavity, thereby contributing to the short term frequency stability of the dye laser. To achieve good long term frequency stability of the laser, the length of the reference external cavity is locked to the emission line of a single frequency He-Ne transfer oscillator. The frequency of the local oscillator is stabilized by offset locking it to a iodine stabilized He-Ne laser using standard techniques.

Such CW dye lasers exhibit linewidths of a few tens of KHz whereas the long term frequency stability is reaching a few parts in $10^{11}$ for observation times longer than 100 s.

## 5.6. Lambdameter

The recent development of high resolution laser spectroscopy has created, in many laboratories, the need for accurate optical wavemeters. For instance, the study of heavy molecules or of atomic Rydberg states requires a fast and reliable way to measure the wavelength of the laser with a precision better than a tenth of a wavenumber (or one Doppler width). In two photon spectroscopy where very narrow ($\ll 10^{-3}$ cm$^{-1}$), but sometimes very weak signals are to be expected, one eventually wishes an accuracy of the order of a milliwavenumber, which is, by far, not achievable by any monochormator.

We have realized a highly performant and easy to operate lambdameter. The basic set-up is described in Fig.10 [27]. Fringes of an unknown and a reference wavelengths are simultaneously generated through the motion af a moving double corner cube reflector in a Michelson type interferometer. An electronic device compare the two wavelengths to within one hundredth of a fringes and numerically display the result.

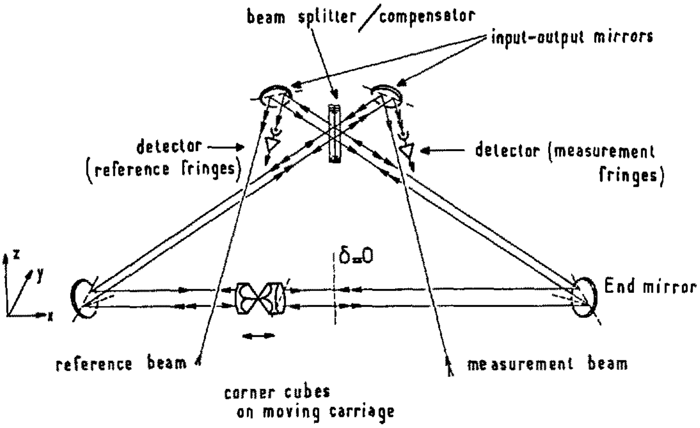

Fig.10 Design of an improved lambdameter

Most of the systematic errors are eliminated by the total superposition of the optical paths of the reference and the measured beams ensuring a high potential accuracy. As an illustration we have measured with this device the wavelength of the $\lambda = 612$ nm radiation generated by a He-Ne laser stabilized on a saturated absorption peak of iodine. The results exhibit a statistical dispersion of $\pm 6\ 10^{-9}$ about a mean value which agrees to within $2 \times 10^{-9}$ with a more exact result obtained at BIPM [28].

## Conclusion

The developments in stabilized laser physics over the past few years have led to great improvements in frequency stability and reproducibility. Important applications in metrology, geophysics, and super high resolution spectroscopy offer to physics new tools which hold the promise of exciting new observational techniques and measurement capabilities.

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