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LASER FREQUENCY STABILIZATION BY SATURATED ABSORPTION

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Abstract.- This paper summarizes the metrological results available about saturated absorption stabilized lasers. After recalling their fundamental limitations, we describe the present state of the art of the most important lasers: CO₂ at 10.6 μm, He-Ne at 3.39 μm, and visible iodine stabilized systems. We show that there is a real need for improved performances, and that we now have all the tools necessary to fulfill this need.

1. Introduction.- During the last decade, hundreds of papers (speculative, theoretical and experimental) have been published concerning saturated absorption and optical frequency standards, and all the metrology laboratories have built and studied their own He-Ne or CO₂ or argon stabilized lasers. About 20 more papers presented at this Symposium relate the latest theoretical and experimental developments in this field. We thought it could be useful to brush a coarse picture of the present state of the art in order to help the potential user in understanding what can be really expected from a saturated absorption stabilized laser. We will also try to compare the achievements with the present and future needs, hoping to stimulate some more efforts from the laser people. The performances of the various standards will be described by the usual quantities: accuracy, reproducibility and stability, defined by the Allan variance. We will specially emphasize the distinction between the reproducibility, which can be measured by comparing standards from different laboratories, and repeatability, which concerns one standard, or a series of similar standards originating from the same laboratory. This happens to be necessary since the intercomparisons of different optical standards show that the "reproducibility" estimates given by all the authors were usually too optimistic by an order of magnitude.

After a rapid description of the more or less fundamental limitations which are common to all saturated absorption frequency stabilized lasers, we will consider the experimental realizations, with special emphasis on the I₂ stabilized systems in the visible, the CH₄ stabilized systems at 3.39 μm and the CO₂ systems at 10 μm. We will finally compare their performances with the present and future needs.

2. Line shifts and asymmetries in saturated absorption.- In flat space, the conservation of the energy-momentum quadrivector leads to the following formula for the resonant emission and absorption frequencies of a two-level atom of velocity v, mass m and frequency splitting ω₀, with respect to an electromagnetic wave propagating along the z direction:

\[ \omega = \omega_0(t) \left[ 1 + \frac{\nu}{c} - \frac{\nu^2}{2c^2} \left( \frac{\hbar \omega_0}{2mc^2} \right)^2 \right] \]  

(1)

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This formula contains all the problems of the saturated absorption technique, if we neglect lightshifts, Zeeman or Stark shifts and the gravitational redshift.

A. Influence of collisions.- The time dependence of $\omega$, has been introduced here to symbolize the perturbation of the atomic energy levels by collisions with other atoms. This is not really a fundamental effect, but it is important since it is known that collisional shift and broadening have a non linear pressure dependence in saturated absorption \(^{(1,2)}\) which forbids the extrapolations to zero-pressure and limits the accuracy although this problem is now well understood theoretically. On the other hand, in the low pressure, high resolution case, the pressure shifts get very small, because the main effect becomes the velocity change of the molecule, which results in broadening but no shift. In methane at 3.39 \(\mu\)m, the pressure shift is only \(5 \times 10^{-14}/\text{mTorr} \) \(^{(3)}\), and very low values were also observed in iodine in the visible \(\text{\'4,5} \) .

So, the direct influence of collisions can be made negligible as concerns the reproducibility, and also the accuracy, for gas pressures in the sub-millitorr range.

B. Geometrical effects.- In the ideal case of infinite plane waves propagating along the x direction, the saturated absorption technique completely suppresses the first-order Doppler shift \((-2\Omega\omega)\). In the practical case of finite Gaussian waves one gets a broadening, and, eventually, a shift \(\delta\), if the 2 counterpropagating waves have unequal intensities and if the absorbing medium is not symmetrical relative to the waist. This "wavefront curvature shift" is probably negligible in all internal cell systems although it has never been fully evaluated in that case. It is also negligible in all the high resolution devices using expanded beams.

C. Quadratic Doppler effect.- The quadratic Doppler effect is a redshift \((-\frac{v^2}{2c^2})\). Its order of magnitude for the most probable velocity of a Boltzmann distribution at \(T \text{\,K}\) is $\omega_0 \frac{KT}{2c^2}$, that is: $-1.72 \times 10^{-12}$ for \(CH_4\) at 3.39 \(\mu\)m, $-1.08 \times 10^{-13}$ for \(OsO_4\) at 10.6 \(\mu\)m, and $-1.08 \times 10^{-13}$ for \(I_2\) at 300\(^\circ\)K. Its temperature dependence is $-5.7 \times 10^{-15} K^{-1}$ for \(CH_4\) \(^{(3)}\), and less for the heavier molecules, so it does not seriously limit the reproducibility, but it is necessary to evaluate it precisely in order to reach an accuracy of $10^{-14}$. In the low saturation limit, Ch.J. Bordé \(^{(7)}\) could give an analytical expression for the lineshape in saturated absorption, including the quadratic Doppler effect and the beam geometry. The important result is that the quadratic Doppler effect causes power dependent, as well as resolution dependent asymmetry and shift of the resonance line. This effect has been demonstrated in a spectacular way by Barger \(^{(8)}\), in a calcium beam. This seems to forbidden the use of light molecules in high accuracy work, unless one can cool them or operate a velocity selection.

D. Recoil effect.- The importance of the fourth term in eq.\((1)\), the recoil effect has been pointed out by Kol'tchenko et al \(^{(9)}\), as soon as in 1968. In saturated absorption lines, it produces a splitting $\delta = 1.33 \times 10^{-9} \frac{\lambda^2}{M}$ where $\lambda$ is the wavelength in microns and \(M\) is the molecular weight. The value of this splitting is $2.45 \times 10^{-11}$ in \(CH_4\), $4 \times 10^{-13}$ in \(OsO_4\) and $8.3 \times 10^{-12}$ in \(I_2\). In most cases, this splitting is not resolved, and it results an inaccuracy from the fact of the two components of the doublet have unequal intensities if the relaxation rates of the two atomic levels are different. More detailed theoretical studies show that the splitting becomes smaller than $\delta$ when power broadening occurs \(^{(10,11,12)}\). Their results are confirmed by experimental data \(^{(8,13)}\). In the case of pure transit broadening, the effective relaxation rates of the two level are given by the inverse of the transit time : they are equal and there is no shift due to the recoil. Another interesting case happens at higher pressure, where the ratio of the amplitudes of the two components becomes pressure independent. High reproducibility can be obtained in that case, but the accuracy is limited.
Considering the above problems, it seems that ultrahigh resolution is not compatible with high accuracy, unless the atoms are cooled. On the other hand, the artificial broadening sources may induce shifts (collisions, transit time). A very good compromise may be the case of the $\text{I}_2$ molecule in a low pressure cell or a beam. For well chosen transitions, the linewidth is lifetime-limited to about 50 kHz ($Q = 10^{10}$), so that the recoil effect can be described accurately; the quadratic Doppler effect is weak enough, compared to the linewidth, to produce only a shift and no asymmetry. The light beam waist size needs only be a few millimeters to avoid any geometrical effect.

After this paragraph, whose main goal was to remember orders of magnitude and to give the references of some important theoretical contributions, let us try to understand the experimentalist's problems.

3. Experimental results.- The metrological data available today concern mainly the following systems:

- stabilization to $\text{I}_2$ hyperfine components in the visible. The best documented and most widely developed stabilized laser is the He-Ne with an internal $\text{I}_2$ cell at 633 nm. The most performant ones are the $\Lambda$ laser and He-Ne laser stabilized on external cells at 515 and 612 nm.
- He-Ne-CH$_4$ systems at 3.39 $\mu$m.
- CO$_2$ lasers locked by saturated absorption in OsO$_4$ and SF$_6$ and by saturated fluorescence in CO$_2$.

Excellent stabilities have been obtained with the He-Xe laser at 3.51 $\mu$m, locked on H$_2$CO (14) but this system is not much documented yet.

A. Stabilization by saturated absorption in iodine.— The iodine molecule $^{127}\text{I}_2$ has a very rich absorption spectrum in the visible, which contains about 50 000 strong lines between 15 000 and 20 000 cm$^{-1}$. Each of these lines has a hyperfine structure 900 MHz wide featuring 15 or 21 main components. The weakly radioactive $^{129}\text{I}_2$ has a similarly dense spectrum, with different hyperfine structures because of the different nuclear spin (9/2 instead 5/2). The lifetimes of the excited levels corresponding to these transitions vary between 0.3 $\mu$s for highly predissociated levels and ten of microseconds for the low Franck-Condon levels which lie near the dissociation limit, so the Q factor corresponding to the natural linewidth varies between $10^9$ and $10^{11}$. It results that any laser emitting between 5000 and 6500 Å, with a tunability of 1 GHz or more, will probably coincide in frequency with some of these hyperfine components. This fact has been used to frequency stabilize argon and krypton lasers at different wavelengths (15,16) and He-Ne lasers at 633 (17) and 612 nm (18).

Let us first describe the achievements and the limitations of the best known system, the He-Ne laser with an internal iodine cell at 633 nm.

1) Internal cell systems.

Since the first observation of saturated absorption of the $R(127)6-3$ line of $^{127}\text{I}_2$ by Hanes and Dahlstrom (17) with a He-Ne laser at 633 nm, all the wavelength metrology labs in the world have built similar systems and they use it, now, instead of the krypton lamp, for most applications. A kind of "standard" set-up has established by himself: the typical system is a 40 cm long cavity, containing a 10 cm iodine cell whose pressure is fixed around 100 mTorr with a cold finger at 15°C and a 20 cm long He-Ne gain cell. Single frequency operation results from the short length and from the intracavity absorption losses. Transverse modes are eliminated by diffraction losses due to the small diameter of the He-Ne tube (1.1 mm typically). The saturated absorption signals have a contrast of $2 \times 10^{-3}$ and a width of 5 MHz (FWHM). Because of the slanted background due to the He-Ne gain curve, it is necessary to use a 3rd harmonic detection (19), the optimum modulation amplitude being 7 MHz p. to p. Such systems have now been built in about 15 different countries (20 to 32) and probably more than 50 laboratories use it now as a reference for wavelength metrology or high resolution spectroscopy.

During the first years (1970-76), the main effort was put on the improvement of the servo-loop electronics: the weak contrast of the saturated absorption signals
ask for low noise and low offset servo loops. Furthermore, the use of the 3rd harmonic detection puts stringent limits on the tolerable harmonic distortions of the frequency modulation (33).

By 1975, most of the laboratories agreed upon a stability of about $2 \times 10^{-13}$ for $\tau > 100$ s, probably limited by fluctuations of the iodine pressure ($-1$ MHz/Torr), and a repeatability of 1 to 2 parts in $10^{-11}$. Everybody was unhappy with the weak contrast, and with the broad lines, which should have been 10 times narrower, according to the lifetime measurements (34) of the excited level, but, strangely enough, nobody had tried to modelize the physical system. The theoretical basis was available (35), but the difficulty was to evaluate all the relevant parameters. The quantitative model we made in 1976 (36) allowed us to get a good description of the contrast and width of the signals and of their variation with the iodine pressure. It resulted from this model that the contrast would be improved by a factor of ten when heating the I$_2$ cell to 200°C, at constant pressure, which was rapidly verified experimentally (37,38), allowing one to improve the short-term stability of the laser (39). But the elaboration of this model also taught us about the limitations of internal cell devices: the main one is that in simple He-Ne-I$_2$ systems, it is not possible to reach a narrow linewidth, because, when decreasing the iodine pressure, one increases the power broadening. This could be avoided only by expanding the beam in the iodine cell, which is difficult with such a low gain laser, or by running it very close to threshold, which would be very noisy, and anyway very uneasy to control and to use. Although it describes correctly the contrast and width of the lines, this model does not include the causes of asymmetries and shifts, which govern the reproducibility of the lasers.

Apart from the studies made in each lab., the problem of reproducibility has been particularly studied by the B.I.P.M. group, through the numerous intercomparison they made with other laboratories (23,40). This work pointed out two problems: the first one is trivial and concerns the purity of the iodine: any non-condensable vapor, present in the iodine cell with a pressure of 0.05 Torr or more, can produce a shift as large as 100 kHz ($2 \times 10^{-10}$), without affecting in a noticeable way the linewidth or contrast of the signal in an internal cell system. So, it is necessary to be able to check the absence of a foreign vapor either by using the Hanle effect (41) or by a direct fluorescence lifetime measurement from a long lived state. The second problem is an asymmetry of the lines, which results in a modulation shift of $-5$ kHz/MHz. Among the possible origins of this effect, one can imagine the following:

a) Asymetric "Fano lineshape" resulting from the I$_2$ predissociation: according to a theoretical evaluation (42) this effect should be too small.

b) Harmonic distortions coming from the saturated dispersion effect (frequency pulling) (43) would also be too small.

c) Frequency dependent diffraction losses due to the gas-lens effect (44 to 46) because of the radial intensity dependence of the gaussian beam, the iodine cell may slightly focus or defocus the beam according to the sign of the frequency detuning relative to the nearest hyperfine component. If the diffraction losses are significant, this effect will superimpose a dispersion shaped power variation upon the saturated absorption signal, producing a line asymmetry. In He-Ne lasers, a diffraction loss of 2 to 5 % results from the small diameter of the gain cell, and this effect may be relatively important. Unfortunately, it is difficult to evaluate it, since its amplitude and even its sign may change with the cavity geometry. A recent computation of this effect made by one of us (P.C.) on the basis of Le Floch's model (46) shows that, in a typical He-Ne I$_2$ system, it can produce modulation shifts as large as a few kHz/MHz, but with a sign opposite to what has been generally observed. It is possible that this diffraction loss effect may be compensated by the fact that the number of active Ne atoms is also changed by the beam geometry modification.

d) the asymmetry due to wavefront curvature. Once again, a coarse evaluation (47) predicts a negligible effect, but the effect has never been carefully evaluated in an internal cell system.

e) Modulation and electronic distortions: the presence of 2nd harmonic distortion in the frequency modulation of the laser with a rate of 0.5 to 1 % would be sufficient to explain the modulation shifts (48). But since their sign is the same.
for all lasers, the phase of this second harmonic distortion should be the same. This is unlikely, unless the distortion comes from an hysteresis effect common to all P.Z.T. The measurements which have been made of the PZT distortions \((45, 47)\) seem to rule out this possibility.

In these internal cell systems, all the parameters of the gain and the absorption cells, the cavity geometry and losses, and the modulation amplitude have to be taken into account simultaneously in order to make a realistic evaluation of the above effects. Furthermore, it is extremely difficult to make any significant experimental test isolating one of these effects, since all the parameters are coupled. For instance, if one tries to isolate the "diffraction losses - gas-lens" effect by adding a pinhole inside the laser cavity, he will simultaneously modify the light intensity, and then the power broadening, and then the ratio of the modulation amplitude to the homogeneous width. A frequency shift will result whatever the effect \(a, b, c, d\) or \(e\) is responsible for the asymmetry, and no usable information can be obtained.

We think it is wise to conclude that these laser are useful and reliable standards at the precision level of 1 part in \(10^{10}\), provided the operating conditions are close to the "standard" device described above. Even if the asymmetry was explained, it would be illusory to hope for 1 part in \(10^{11}\), because it would require the specification of too many operating parameters with too high a precision.

The above considerations are also valid for internal cell \(A^\ast\) stabilized lasers and He-Ne lasers at 612 nm, with only slight differences. The obvious way to improve the situation is to use an external cell system. This is unfortunately difficult at 633 nm, because the \(R(127)11-6\) line is a weak one, but it has been done at 515 and 612 nm \((4,49)\).

2) External cell systems.

A) Argon lasers at 515 nm. - This system has been studied by Camy \((4)\) and Spiweck \((50)\). The iodine lines at 515 nm are interesting because they are strong and potentially narrow. Using expanded laser beams in a low pressure \(I_2\) cell, Camy obtained a linewidth of 70 kHz, close the natural linewidth. He reported a stability of \(5 \times 10^{-14}\) for \(\tau = 100\) s, and a repeatability of \(1.5 \times 10^{-12}\), limited by the laser beams geometry and alignment. He also demonstrated the possibility of using Doppler generated level crossings, which have the advantage of being free of recoil splitting, but the drawbacks of being weaker than the main lines and sensitive to light-shifts.

B) He-Ne laser at 612 nm. - The \(R(47)9-2\) and \(P(48)11-3\) \(127I_2\) lines at 612 nm are strong enough to give good signals in an external cell, but the He-Ne laser at 612 nm is rather weak (300 \(\mu\)W typically). So, we chose to enhance the saturated absorption signals by putting the \(I_2\) cell inside a high finesse (100) Fabry Perot resonator, whose resonant frequency is servo locked to the laser frequency \((51)\). The multipass effect in the Fabry Perot greatly increases the contrast, so that it is possible to use low iodine pressures (3 \(m\)torr) and to obtain narrow linewidths (200 kHz) and large S/N ratio. The Fabry Perot cavity, including a telescope, defines a wide beam, of high geometrical quality, the two counterpropagating waves being self-matched. These conditions are highly suitable for metrological applications. A first comparison of two similar systems allowed us to demonstrate long term stability \((\tau > 100\) s) of \(2 \times 10^{-13}\) and a repeatability of \(6 \times 10^{-13}\) (300 Hz), unfortunately limited by electronic offsets.

We could verify the high symmetry of the lines from the absence of modulation shift, and we were not able to measure any power shift within the resolution of \(6 \times 10^{-13}\), so we can still expect some improvements. We think this technique could be useful in conjunction with other lasers (argon, dye or optically pumped \(Na, Li\) lasers), providing a reference grid in the visible with a reproducibility of \(10^{-12}\) or better.

C) The He-Ne \(CH_4\) system at 3.39 \(\mu\)m. - The He-Ne laser at 3.39 \(\mu\)m stabilized on the \(F'(2)\) component of the \(P(7)\) line of the \(v_3\) branch of \(CH_4\) is one of the most important systems for historical reasons, because it was used by Barger and Hall \((52)\), who first demonstrated the feasibility of high performance optical frequency stan-
dards, and for practical reasons, because it lies in this narrow part of the spectrum where both wavelength and frequency can presently be measured with high precision. This is why it was used for the determination of the velocity of light (53).

The work of Barger and Hall was followed by many studies in different countries. Most of the metrological studies (evaluation of stability and repeatability) were made with simple internal cell systems (54,55,56,57), while more fundamental studies were undertaken, mainly at JILA (Hall and co-workers) and Novosibirsk (Chebotayev and co-workers), in order to increase the resolution and to reach the fundamental limits. Other groups from USSR studied different ways of improving the contrast of the CH_4 resonance via mode competition in linear or ring laser cavities (58,59). Contrasts of up to 80% have been obtained, in that way, with a linewidth of 100 kHz or less, which leads to a potential stability of 10^{-15}/\sqrt{t}. The frequency pulling of the laser, due to saturated dispersion has also investigated and used for locking the lasers (59,43).

The most significant results from the viewpoint of metrology are the following: the simple internal cell systems typically show a stability of 10^{-13}/\sqrt{t} and a repeatability of 1-2 10^{-11}. They exhibit a power shift and a line asymmetry at high intensity: both can be explained by the presence of hyperfine structure of the F_2(2) line (60), which is not resolved in these systems, but has been clearly resolved by Hall and Bordé (61), and later by other groups (59,62). This hyperfine spectrum contains 3 main components with a spacing of \approx 11 kHz plus 3 weaker ones and some Doppler generated level crossings. Even at the highest resolution (1 kHz) obtained by Hall and Bordé, all the components are not completely resolved. Every stabilized laser using the F_2(2) line is locked "somewhere" in the middle of this hyperfine structure, and the locking point varies with the resolution and the light intensity, because the relative intensities of the hyperfine components and the D.G.L.C. have different saturation parameters. These effects can be computed in order to evaluate the power shift and line asymmetry of a given experimental set-up. Bagaev and Chebotayev (3) could find some specific conditions in which the frequency of their lasers is not too much dependent on power and modulation parameters. Using two identical lasers functioning in these well defined conditions, they could reach a stability of 5 10^{-15} for \tau = 100 s, and a repeatability of 3 10^{-14}. It is possible that a similar laser built independently in another laboratory would confirm a reproducibility of \approx 10^{-13}, but the necessity of specifying all the operating conditions in order to get a good reproducibility is not a very satisfactory situation, although there is already a precedent, with the krypton lamp.

The obvious way to overcome the H.F.S. problem is to use the E-line of CH_4, which lies 3 GHz to the red of the F_2(2), and has no structure. In 1975, we showed that it was not difficult to tune the CH_4 gain curve with a magnetic field of 18 Tesla in order to observe the E-line. The gain on the \sigma^+ and \pi components is easily quenched with a CH_4Br absorption cell (63). More recently, Koreshlyavskii et al (64) made a deeper study of the metrological properties of E-line stabilized He-Ne lasers. As expected, the power shift and line asymmetry are much weaker than with the F_2(2) line. They could observe an asymmetry related to diffraction losses. This problem was solved by increasing the gain cell diameter, and they obtained a repeatability of 5 10^{-13} with a simple system (65). Their lineshape is very symmetrical, but there remains a weak power shift, which is probably due to the quadratic Doppler effect. It seems that this fundamental effect will limit the reproducibility and accuracy of CH_4 stabilized lasers to about 1 part in 10^{13}, at least when one uses a cell. Another cause of power shift in a cell is the coupling of power broadening with nonlinear collisional shift and broadening (66).

It is difficult to understand why the E-line is not yet of general use in CH_4 stabilized lasers. The main reason is probably that the repeatability of 2 10^{-11} obtained with the F_2(2) systems was considered as being "good enough" for their applications in frequency synthesis chains and in wavelength measurements. Unfortunately, intercomparisons have been made in 1976 between a few CH_4 stabilized lasers from NPL, BIPM and PTB (57), whose conclusions are that, when used in similar conditions, the lasers under test where capable of a reproducibility of \pm 2 10^{-11}, but that shifts as large as 1.2 10^{-10} could be observed, especially when one of the lasers was locked by the saturated dispersion technique instead of saturated absorp-
Furthermore, there is now a real need to improve the reproducibility and accuracy of 3.39 \( \mu \)m stabilized lasers, since the VNIIPTRI group was recently able to measure the frequency of an He-Ne CH\(_4\) laser by comparison with the Cs clock frequency, the whole frequency synthesis chain being phase-locked\(^{67}\). The measurement precision was then limited only by the Cs clock accuracy, and the value they got for the \( F^2(2) \) line frequency is:

\[
\nu_{F^2(2)} = 88 376 161 603.4 \text{ kHz}
\]

with an error bar of \( \pm 1.4 \text{ kHz} (1.5 \times 10^{-11}) \) corresponding to the CH\(_4\) stabilized laser repeatability.

The lack of reproducibility of the "He-Ne-F\(^2(2)\) line" system is probably responsible for the 10 kHz disagreement between this measurement and previous ones\(^{68}\).

D) CO\(_2\) stabilized lasers. - The CO\(_2\) stabilized lasers at 10.6 \( \mu \)m, like the 3.39 \( \mu \)m He-Ne lasers, played an important role in the measurements of the velocity of light\(^{69,70}\). They are an essential part of the frequency synthesis chains. The experimentalists working on CO\(_2\) stabilized lasers have taken 2 main directions:

- a) the first one is the stabilization by saturated fluorescence in CO\(_2\)\(^{71}\). This technique has been extremely useful in the frequency synthesis chains and for calibration of the infrared spectrum. It provides a convenient reference for each CO\(_2\) or N\(_2\) laser line. Unfortunately, one has to find a difficulty compromise between pressure broadening and S/N ratio, which limited the stability to \( 10^{-12} \) about, and the reproducibility to 5 kHz (1.5 \( 10^{-10} \)). This technique is then a little bit obsolete now, in regard to the measurement precision of the phase locked frequency synthesis chains.

- b) much more promising were the saturated absorption experiments in SF\(_6\) and OsO\(_4\) reported for the first time in\(^{72}\) and\(^{73}\). Most of the work realized on these molecules up to now has been spectroscopical more than metrological (74,75), but excellent stabilities (5 \( 10^{-14} \) for 10 s) have already been obtained\(^{76}\). The SF\(_6\) molecular lines have narrow hyperfine structures, which are usually not resolved, and which limit the reproducibility to a few kHz (10\(^{-10}\)), just like in the CH\(_4\)-\( F^2(2) \) line case. The 192\(^{192}\)OsO\(_4\) molecule is far more interesting for metrology, since it has no hyperfine structure and, also, because it is very heavy, so the quadratic Doppler and recoil effects are weak.

Ultrahigh resolution experiments are in progress at LPL (Villetaneuse) which should result soon in a linewidth much smaller than 1 kHz (3 \( 10^{-11} \))\(^{77}\). It would be surprising if the resulting reproducibility was worse than \( 10^{-12} \), but up to now, no result better than \( 3 \times 10^{-11} \) has been reported at 10.6 \( \mu \)m.

4. Conclusions. - Optical frequency standards were considered as extremely promising in 1970, on the simple basis that much higher line \( Q's \) could be obtained in the optical range than in the microwave range, leading to much better stabilities. This part of the contract has been partially realized, since the best CH\(_4\) or SF\(_6\) or I\(_2\) stabilized lasers already have a better short-term stability than all the microwave standards. But the goal of improving reproducibility and accuracy has not been achieved. We see many reasons for that:

- a) non linear spectroscopy brings the high-resolution capability, but also a lot of new problems, which were not realized in 1970. They are now well understood theoretically, but they are difficult to overcome experimentally at all level better than \( 10^{-13} - 10^{-14} \).

- b) more trivial problems, like unresolved hyperfine structures result from a wrong choice of the reference line. The use of 192\(^{192}\)OsO\(_4\) molecules and of the E-line of CH\(_4\) should straighten this situation.

- c) the repeatabilities of \( \pm 2 \times 10^{-11} \) achieved at 3.39 \( \mu \)m and 633 nm as soon as 1973, were considered as "good enough" for most applications. This may have slowed down the enthusiasm of many for working towards better metrological performances: in most laboratories, frequency stabilized lasers were not studied for themselves, but they were used as tools for well defined applications, such as length measurements, frequency synthesis, high resolution spectroscopy, measurement of the Rydberg...
constant, geophysics, relativity, all subjects which often seem more rewarding than pure metrology.

Hopefully, "good enough" in not a long-lived statement in physics: the successfull operation of a phase-locked frequency synthesis chain asks for CO, and 3.39 \mu m He-Ne lasers having a reproducibility of 10^{-14}, which could be used as secondary standards allowing the extension of the synthesis up to the visible with this high accuracy, without the need of operating the whole chain simultaneously.

There are other reasons for improving the visible standards: the measurements of the Rydberg constant now approach the limit were their accuracy is limited by the He-Ne-\text{I}_2 laser reproducibility (78). The new high resolution frequency meter proposed by J. Snyder could provide the "lambdameters" with a resolution of 10^{-12}(79) which should result in an improvement of their accuracy to the 10^{-11} level.

Since we now have the needs, and the adequate tools, we can hope it will not take ten more years before we can see some true optical frequency standards.

References

(1) ALEKSEYEV V.A., ANDREEYVA T.L., SOBEL'MAN I.I., Sov. Phys. JETP 64 (1973) 813
For more references and compilation of further works, see :
(2) BAGAEV S.N., BAKLANOV E.V., CHEBOTAYEV V.P., JEPT Lett. 16 (1972) 344
(3) BAGAEV S.N., CHEBOTAYEV V.P., Applied Physics 7 (1975) 71
(4) BORDE Ch. J., CAMY G., DECOMPS B., DESCUBES J.P. and J. VIGUE, To be published in Journal de Physique (Oct. 81)
(6) HALL J.L. and BORDE Ch. J., Appl. Phys. Lett. 29 (1976) 788
(7) BORDE Ch. J. in Laser Spectroscopy III (Springer-Verlag, N.Y. 1977) p. 120
(16) GILL P. and BENNETT S.J., Metrologia 15 (1979) 117
(18) BENNETT S.J. and CEREZ P., Optics Com. 25 (1978) 343
(19) WILSON G.V.H., J. Appl. Phys. 34 (1963) 3276
(24) HELMACE J., BAYER-HELMS F., Metrologia 10 (1974) 69
(26) TURMA W. and VAN DER HOEVEN C.J., Appl. Optics. 14 (1975) 1896
(31) TANAKA K., SAKURAI T. and KUROSAWA T., Jap. J. Appl. Phys. 16 (1977) 2071
(77) BORDE Ch. J., This issue