Frequency-Stabilized Lasers—a Driving Force for New Spectroscopies.

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1. - Introduction and overview.

It is particularly appropriate to consider the evolution of stabilized lasers within the context of a Fermi School concerned with the Frontiers of Laser Spectroscopy, as the stabilization of lasers has clearly provided one of the stimulating influences for the development of new spectroscopic methods. It is hoped that this lecture will serve as a useful introduction to this art of stabilized lasers, especially for those who may be joining the sport from other fields. The remainder of this overview section may be regarded as an «executive summary» of the main text.

The first laser stabilization experiments with laser/absorber systems such as HeNe/CH$_4$ and HeNe/I$_2$ led to unprecedented laser stability performance, based on intracavity saturated absorption. The use of HeNe/CH$_4$ and CO$_2$/OsO$_4$, for example, yielded stability performance in the $10^{-13}$ domain. Later the added flexibility of experiments with an external cell led to further significant stability improvements. The natural extension of this work is to employ tunable lasers. This approach has dominated the past decade-and-a-half, with frequency stabilization performance ultimately limited by the same systematic shifts that trouble the gas laser work. However, in using broadly tunable sources—such as dye lasers, Ti:sapphire lasers and diode lasers—the first challenge concerns narrowing the laser spectrum and guiding the center frequency to explore interesting resonances. It has become clear that prestabiliza-

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tion of the laser on the nonsaturable resonance of a stable cavity is a good strategy: with adequate feedback system design, one can effectively replace the intrinsic noise of the laser with the measurement noise of the stabilizer system. By now sub-hertz frequency control and optical phase locking have been demonstrated with most of these tunable sources. The ready access to modulation of the diode laser can lead to a very simple but impressive source, while the external stabilizer approach [1] is attractive for dye and optically pumped solid-state sources. Current work on amplitude stabilization of the laser pump may lead to reduction of the «intrinsic» solid-state laser noise as well.

With the current explosion of interest in atom trapping techniques we can look forward to major progress in the narrow-line laser/super-sharp absorber high-resolution spectroscopy business. Applications range from atomic clocks to cold-atom collision physics to tests of special relativity. The combination of ultra-stable lasers with cold-atom interferometry will be especially powerful in offering new tests of atomic-charge neutrality and of time-reversal invariance via new limits on atomic electric-dipole moments. Remarkably, a practical instrument for oil and gas prospecting might be based on a laser-diode/atom-interferometric measurement [2] of local \( g \).

2. – The gas laser epoch.

21. Lamb-dip stabilization. – One of the important approaches to laser stabilization came up early in the history of gas laser development, when the technical advances in reducing optical losses allowed saturation effects to play a larger role. This provided researchers with the sub-Doppler intracavity saturated-emission resonance which came to be known as Lamb's central tuning dip, in recognition of Lamb's fundamental contributions to the theory [3] of the effect. A clear physical picture was provided [4] by Bennett Jr., who discussed the saturation effect in terms of «holes» «burned» into the Gaussian velocity distribution by the strong fields. The saturation of the amplifying medium would be maximized at the line center tuning, as there both cavity running waves would appear to have the same frequency in the rest frame of a single-velocity group. These zero-axial-velocity atoms would then interact with increased strength, but still would yield less stimulated emission than in the detuned case where two separate velocity groups could contribute to the output power.

The width of the power dip at the central tuning was fixed in part by collisions \( (\approx 10 \text{ MHz/mbar}) \), and in part by natural decay, augmented by the broadening effect of the intracavity laser transition rates themselves. Servo techniques enabled one to lock the laser frequency to the center of the power dip with an attainable frequency stability \( \approx 10^{-10} \) or better, limited mainly by vi-
Fig. 1. – Lamb's (central tuning) dip in pure $^{20}\text{Ne}$ laser. Laser operated by r.f. discharge in 0.12 Torr (0.16 mbar) of pure $^{20}\text{Ne}$. Frequency increases to right. Cavity $\text{fsr} = 465 \text{MHz}$. Powers increase from (a) 1.4 $\mu\text{W}$, (b) 4.2 $\mu\text{W}$, (c) 5.6 $\mu\text{W}$. Weak asymmetry to high-frequency side probably due to small amount of $^{22}\text{Ne}$ impurity. Lamb dip widths are about (a) 30 MHz, (b) 36 MHz, (c) 39 MHz.

brations. The long-term stability was compromised by the gradual pressure decrease of the HeNe gain cell, which reduced the pressure-induced blue shift associated with Ne-He collisions.

An important advance in this field was the development by BENNETT and KNUTSON [5] of a new, single-gas operating environment for the neon laser at 1.15 $\mu\text{m}$. The total pressure was reduced 15-fold to ~0.2 Torr which in turn provided a saturation feature of much higher contrast and sharpness (see fig. 1). The pressure shift of this line was reduced by the low-pressure operation, and removed as an important contributor to long-term drift by our easy ability to measure changes in the pressure of the one-component active gas. Interestingly, atomic physics also came to our advantage to make the shift intrinsically small, ($\sim 1.4 \pm 0.8$) MHz/mbar, as was eventually measured by MAGYAR in some careful experiments [6].

Certainly a stability and reproducibility in the $< 10^{-12}$ range would have obtained if this option had been strongly pursued [7]. CHEBOTAYEV [8] pointed out that adding hydrogen improved the power of the spectral doublet at 1.15 $\mu\text{m}$, and discussed measurement of the speed of light via interferometry and optical heterodyne detection of the 51 GHz beat frequency [9].

2.2. Intracavity saturated absorption—atoms and molecules. – In 1967 an important new idea was tried: LEE and SKOLNICK [10] and LISITSYN and CHEBOTAYEV [11] showed that it was attractive to use two separate cells in the laser cavity. One cell could contain a HeNe mix to give good gain and output power. The other would contain an absorber gas under conditions more favorable to serving as a sharp, stable reference transition. In these two experiments, pure neon was the absorbing reference gas. This choice ensured the needed wavelength coincidence, but required also a weak discharge to produce population in the neon absorbing level. Almost instantly many workers realized that molecules would be interesting as absorbers because their rich spectra helped
ensure the necessary spectral overlap with the laser transition. Furthermore, molecules can offer sharp absorptions beginning from the ground state. The attractive system of HeNe oscillating at 3.39 μm, saturating the P(7) line in methane (CH₄) was introduced at basically the same time by Barger and Hall [12] in the U.S. and by Chebotayev and his associates in Novosibirsk [13]. Frequency reproducibility of 10⁻¹¹ was obtained along with stabilities in the 10⁻¹³ range in the very early experiments! This HeNe-plus-methane system represents, even now, one of the best-performing optical frequency standards known, based on the advanced techniques to be discussed momentarily.

However, in keeping with our spectroscopic-frontiers theme, and noting that methane spectroscopy has served as a testing ground for spectroscopic ideas since long before the laser epoch, it seems a brief historical note would be appropriate here. In fig. 2 we show the progress over some 35 early years, beginning with the rotational resolution by Nielsen and Nielsen [14] in 1956 of the ν₂ fundamental band at 3.39 μm of CH₄. (The modern laser physics student, armed with sophisticated data acquisition hardware/software in his personal computer, may well notice the ordinate axis label in fig. 2a) («deflection») and reflect upon the efforts invested by earlier generations of students.) The line from J″ = 7 in the ground state to J′ = 6 in the vibrationally excited state, P(7) in usual notation, looks in fig. 2a) quite sharp and reasonable at this resolution level of ~ 1500. However, by the ’60’s people had become sensitized to the issue of symmetry in spectroscopy, perhaps from the ready observation of symmetry-dependent NMR and ESR spectra of solids. The CH₄ molecule, with its four equivalent hydrogen atoms, might initially be expected to exhibit perfect tetrahedral symmetry. But the nuclear spins, each of I = 1/2, can be combined in several ways, leading to a total I of 2, 1 and 0. Of course, an overall symmetry is required for the nuclear part of the product-type Born-Oppenheimer wave function, and so we recognize not all states should be available. Also, one might first suppose the energy splittings between these states should be only associated with hyperfine interactions, but Hecht [15] showed in 1960 that a Coriolis-type interaction could lower the symmetry of the excited state in a subtle way, resolving it into 6 components (see fig. 2c)). Basically, the ν₂ = 1 triply degenerate vibration can give rise to a superposition in which a circulation is evident, designated by L = 1. Coriolis-type forces couple L = 1 and ν₂ = 1, giving six levels within the excited state, as shown clearly in fig. 2b) by the high-resolution grating spectra (nearing the Doppler limit) by Henry et al. [16]. As indicated, the Coriolis component which interacts with the unshifted HeNe laser is designated F⁰₂, and corresponds to one of the I = 1 nuclear configurations. Nearby levels E (I = 0) and A₁¹¹ (I = 2) can be accessed with a Zeeman-shifted HeNe laser. When tunable lasers became available, PINE used infrared difference frequency generation in a nonlinear crystal (LiNbO₃) to make beautiful systematic measurements [17] of this spectrum.

After this digression, we return to fig. 2d) which shows the sub-Doppler
Fig. 2. ~ 35 years' progress in methane spectroscopy: a) shows the CH₄ spectrum obtained in 1935 by Nielsen and Nielsen (ref. [14]). The lines appear reasonable at this resolution (~ 1500) and give no hint of internal structure. b) shows the small splitting appearing between the different symmetry species, induced by a Coriolis interaction in the vibrationally excited state. From ref. [16]. c) shows the symmetry splitting predicted by Hecht (ref. [15]), although the designations follow the convention of Herzberg. d) Saturated-absorption peak in CH₄. HeNe laser at 3.39 μm is excited by r.f. discharge. CH₄ cell at 12 mTorr (16 mbar) is located inside laser cavity. HeNe pressure has been adjusted to bring laser frequency into near coincidence with the saturated-absorption feature. Power output is ~ 200 μW and peak contrast is ~ 12%. Peak width is ~ 270 kHz. Cavity free spectral range is 250 MHz. Note cross-over resonances in two-mode region near cusps (ref. [12] (1969)).

saturated-absorption peak obtained almost simultaneously in Boulder and Novosibirsk in 1968. In this figure, the peak shows a contrast of some 12% in the 200 μW output, which rises to 15% at maximum output power (~ 0.8 mW). (Unfortunate hysteresis in the displacement of the PZT gives a doubled trace.) The peak width of ~ 270 kHz is controlled by power and pressure broadening of
a basic linewidth fixed by the limited transit time of crossing the light beam.

Of course, other laser/molecular absorbing systems could be imagined. Hartman and Dahlstrom [18] were quick to introduce the HeNe/127I2 system in the red laser at 633 nm. Knox and Pao [19] and Desslattes, Sweitzer and their colleagues [20] pointed out the contrast advantages of a similar transition in the He35Ne/129I2 system. In later times a relatively large number of other systems have been investigated, including HeNe/127I2 at 612 nm, 594 nm, 543 nm and 640 nm [21]. These systems work with the HeNe laser constrained to oscillate on one of several possible neon transitions, through the use of intracavity wavelength selectors such as prisms or special ‘edge’ coatings. Another old favorite [22] is the Ar+ ion laser operating with 127I2. Because of the greater complexity of this laser, and especially its vastly higher power capability, it is typical for this combination to be operated with the absorber contained in a cell external to the laser cavity [23]. Other lasers and molecular absorbers have been found to give attractive performance, the most general system being CO2 lasers operating with an intracavity cell containing CO2 at low pressure [13]. The needed spectral coincidence is clearly ensured for every line. The associated 4.3 μm saturated fluorescence signals are widely used for stabilization [24]. Another excellent system uses the CO2 laser with an external cell, employing molecules such as SF6, SiF4 and OsO4 [25, 26]. The 3.39 μm HeNe laser was used to explore saturated-absorption spectroscopy and laser locking with a variety of methyl halide gases [27].

23. Saturated-absorption wavelength/frequency standards—redefinition of c. – The use of these various laser systems as reproducible standards of frequency/wavelength has been extensively studied: many variations and intercomparisons have been performed [28], mainly by the several national standards laboratories and a few other institutes. Progress has been monitored by an international committee of experts (Comité Consultatif pour la Définition du Mètre, or CCDM) which has recommended operating conditions within which frequency reproducibility is enhanced [29]. The good general agreement has made it possible also to offer recommended values for the frequency/wavelength under such conditions. In 1983 there was an international redefinition of the meter in terms of the frequency of a stable laser and an adopted value for the speed of light of 299,792,458 m/s, exactly [29]. This idea has worked out well, with the 1992 meeting of the CCDM being able to propose almost 10-fold more precise values for the absolute optical frequencies of these recommended transitions [30]. A number of laboratories now have specialized frequency synthesis chains operating to connect different of these optical frequency references to the primary standard of time/frequency, the cesium clock, which operates at 9,192,631,770 GHz, by definition. As an infrared standard, the CO2/OsO4 system in particular has served well, with many lines now being known in abso-
hute frequency at the 50 Hz level [25]. A particularly good approach employs the OsO₄ reference molecules in an external resonator [31]. Also the HeNe/CH₄ system has been measured by many laboratories and an uncertainty in the ~200 Hz region seems realistic: \( v = 88.376 \pm 1.606 \) kHz [30]. Clairon and his colleagues have recently given a much improved frequency [32] for the red 633 nm iodine-stabilized laser: \( 473.61 \pm 12 \) kHz.

A significant difference exists between the iodine-based and other molecular systems with respect to the presence of internal structure within the saturated-absorption line. It happens that the main hyperfine energy arises in I₂ from an interaction of the nuclear electric-quadrupole moment with the electric-field gradient of the molecule. These splittings of tens of MHz are readily resolved in the optical sub-Doppler spectra, in contrast to other molecular cases where the absence of quadrupolar splitting leaves only magnetic energies in the tens of kHz range. For a time in the early 1970's there was a productive industry of laser-based measurements of these small hyperfine splittings, stimulating an improvement in the attainable optical resolution. Since the main broadening turned out to be associated with «time of flight» of the molecules as they crossed the interrogating laser beam, it was not surprising that absorption cells of larger transverse aperture were soon introduced. The JILA group pursued this direction, along with Chebotayev, Bagaev and their colleagues in Siberia, and Bödde and his associates in Paris. It proved possible to resolve the methane hyperfine spectrum [33] optically and soon to observe the consequences of the recoil [34] when a photon was either absorbed or emitted. Figure 3a) represents a continuation of fig. 2d), where that single transit-limited peak has been explored with an absorption cell of 30 cm aperture. Note the frequency axis scale has been usefully magnified by 10⁴. The magnetic hyperfine structure is clearly resolved and the pure of heart can appreciate the incipient spectral doubling associated with recoil in both absorption and emission. After this epoch, the Boulder group turned toward frequency stabilization of tunable lasers operating in the visible. However, Chebotayev and his associates improved their methane setup (see fig. 3b)) and pursued another extremely promising avenue.

2.4. Optical selection of the slowest atoms. – This idea [35] is based on enhancing the detection sensitivity to such a degree that one could afford to select molecules which have basically zero velocity along the axis (this is the usual group involved in saturation spectroscopy) but also have near-zero velocity transverse to the axis. Their absorption contributions can be relatively enhanced—perhaps it would be better to say they are not so disastrously decreased in amplitude—when the interrogating laser power is strongly reduced. Then only the slowest particles will experience the resonant drive for a time long enough to develop appreciable excitation. Maintaining a collision-free interaction will require severe reduction of the pressure, which also cuts the signal size. However, this velocity selection approach has been used successfully
in Siberia (one wonders if their reduced ambient temperatures imply substantially larger fractions of slow molecules!) with the added advantage that the extended interaction time leads to surprisingly narrow resonances for a given size laser beam. A 1988 preprint by Bagaev and Chebotayev presented a 1 kHz linewidth which they had obtained—at 88 THz!—about 20-fold smaller than the scaling studies [36] would predict for a 5 mm diameter beam. This 20-fold re-
duced velocity means an effective temperature below 1 K, and a relativistic
time dilation shift below 1 Hz. So the stability, reproducibility and accuracy ca-
pability of this laser system can be in the ~ 10^{-14} region! This work has been
further improved [35], leading to the data shown in fig. 3c): a 50 Hz optical reso-
nance linewidth at 88 THz! The aperture here was increased to 30 cm, so the
cooling effect is more than a factor of 20. This has been absolutely inspiring
work—a clever mixture of brute-force scaling, careful engineering and auda-
cious new physics ideas.

The entire laser spectroscopy community was saddened recently to learn of
the sudden death of our colleague Veniamin P. Chebotayev on September 1,
1992, just shortly after our Fermi School meeting. He was 53 years of age. His
contributions are many, varied and important, ranging from many of these
methane spectroscopy tricks, to suggestions for measuring the speed of
light [8], to Doppler-free two-photon spectroscopy [37], to optical Ramsey
fringes [38], to optical clocks [39], frequency synthesis [40], and many, many
other now widely employed techniques. We will all miss his cheerful style and
his many good ideas in physics.

3. The tunable laser arrives.

3’1. Frequency stabilization is a severe problem for tunable lasers. — Many
spectroscopists welcomed the arrival in the early 1970's of tunable dye lasers
and the rapid development of various additional methods of frequency exten-
sion, including new dyes, harmonic generation, frequency mixing, etc. Now one
could tune to the real transition of physical interest! Of course, a laser that is
free to be tuned to another color will itself freely tune to a wrong color: the pres-
sure was on to learn how to control these potentially wonderful new sources.
Single-mode operation became routine with the inclusion of a servo-controlled
intracavity etalon to increase the losses for all but a selected mode. Commercial
systems appeared with the capability of a 30 GHz tuning range. A stabilization
system provided useful but relatively rough stabilization to a tunable cavity
(= MHz). Even now, it remains an interesting challenge to provide stabiliza-
tion at the kHz level (or better) of a laser which can tune 50 THz in 30 GHz
windows!

3’2. Cavity stabilization of tunable lasers. — Stabilization of a tunable laser
to an optical resonator [41] has a number of advantages compared with the pre-
vious nearly universal scheme of stabilization of nearly untunable lasers to
quantum absorptions. For one thing, a cavity has the interesting property that
it has many uniformly spaced sharp resonances throughout a spectral region vs.
just one, or at best a few, lines for a molecule. Having many potential lock
points should be a useful advantage for a widely tunable laser. Another impor-
tant advantage is the totally generic aspect of cavity resonances: they can be
designed to occur at whichever wavelength we find useful. A more subtle, but crucial, difference arises from the fact that the cavity resonant system is basically linear. For our locking purpose, if the obtained signal/noise performance is insufficient, it can be increased by allocating a larger power for the task. With quantum absorbers this extra power would broaden the resonance and so degrade the useful frequency discriminant.

While a fringe side locking method[42] was in vogue first, as accuracy dreams became more demanding it was necessary to use the cavity resonance information in a better way. One method currently in use employs r.f. phase modulation of the laser source to produce optical sidebands on the laser signal[43]. The perfect antisymmetry of the sidebands produced by pure FM/phase modulation leads to a cavity frequency discriminator curve which is ideal for laser locking. DREVER et al.[44] have discussed an interesting aspect of this discriminator when used in reflection: the stored field in the resonator can serve as a phase reference against which the present optical phase can be compared. This leads to the capability of an optical phase lock at short times, changing to a frequency lock for times long compared with the cavity storage time. The detection S/N ratio, of course, depends on the employed bandwidth. The interesting effect that occurs here is that we can broaden the servo bandwidth to large values ($\sim 10$ MHz) with the extra bandwidth-induced noise leading to a slight reduction of the optical-phase measurement quality. However, from FM theory we know that, if the optical phase tracks the sinewave ideal to within 0.1 rad r.m.s., we will have only 1% of the power in the accompanying phase modulation sidebands[45]. So we paid only a little in noise performance—and that noise appears far from the carrier—in exchange for starting our servo locking curve at a much higher unity gain frequency. With the gain function rising toward lower frequency at the rate of $\approx 10$ dB/octave, now when we reach the ugly range $\sim 200$ kHz for Ar$^+$-pumped lasers, we just may have enough gain to reduce the parasitic frequency/phase noise to our desired low value ($\ll 1$ rad). Eventually, below the cavity linewidth of perhaps tens of kHz, the cavity no longer stores a reference phase, so we begin to have a frequency measurement output. The measurement noise in the bandwidth from this frequency downward will be compared by the servo with the frequency discriminator output slope, and so we can see that the servo will impose this FM equivalent onto the laser as the servo drives toward a zero apparent error signal[46].

The bottom line of this story is that, even for dye lasers, one can expect to be able to suppress the intrinsic noise of the laser more-or-less totally with feedback, replacing it with measurement noise. The value of this noise is worth noting for its smallness: For an invested 1/4 mW one can readily engineer a sub-hertz bandwidth for the tunable laser, be it dye or Ti:sapphire! Recently Ohtsu’s group has increased the speed of analog locking[47] of a simple single-frequency diode laser to achieve $\sim 10$ Hz linewidth and phase locking. External cavity operation of diode lasers allows us to enter this domain also[48] with
Fig. 4. – Wave form of optical heterodyne signal. Two lasers are independently locked onto adjacent axial orders of a stable interferometer leading to a beat frequency of \( -500 \) MHz. This r.f. beat is heterodyned to near 1/2 Hz using a frequency synthesizer. The resulting wave is low-pass filtered \( (f < 10 \text{ kHz}) \) and digitized at 60 samples/s. From least-squares fits to several such data sets, the apparent linewidth is found to be below 50 mHz per laser.

these attractive sources. It should be emphasized that this degree of locking is expressed relative to the resonator’s frequency. If that frequency is being affected by vibrations or temperature changes, then surely our narrow-linewidth tunable laser will faithfully track these changes. So the problem has two parts: laser locking and cavity stabilization. Let us consider the laser locking part first.

3.3 Precision of locking to a cavity resonance. – In fig. 4 we show the beat between two HeNe lasers locked onto adjacent orders of a single Fabry-Perot resonator [49]. The optical frequencies differed by one unit of the free spectral range \( (\sim 500 \text{ MHz}) \), so the 500 MHz r.f. beat signal from the photodetector was heterodyned with a stable 500 MHz synthesized frequency to produce the difference frequency which was digitized in time to produce the wave form shown. The absence of fast phase noise shows that both of these independent optical-frequency lock circuits work extremely well, since any errors would show up in the difference. This phase-stable picture was obtained only after we used a special quartz oscillator for the frequency synthesizer, since, even though the radiofrequency is one million times lower than the locked optical frequency, the performance domain we are entering places almost unattainable demands on stability and spectral purity—of the r.f. oscillator! Obvious interest attaches to increasing the beat frequency to perhaps 10-fold higher values and selling this beat frequency wave form for r.f. synthesis tasks.

In fig. 5 we present data for the system of fig. 4 using the Allan variance presentation. Please remember that these curves relate to the quality of locking
lasers to a common cavity, which substantially reduces sensitivity to cavity instabilities: Here we are testing the locking.

As to the accuracy of locking to the optical resonances, we have shown in an earlier experiment that with some care one can reach the 1 Hz domain. Some new experimental strategies to separate the cavity reflection from spurious reflection information are being explored at present. Experience in the r.f. domain has shown that with enormous and persistent effort one can hope to find a reproducible «center» of a resonance to within about $10^{-6}$ linewidths, assuming the line shape is free of asymmetry and other problems. For the current cavities, this criterion would be about $1/10$ Hz! Clearly there is room for major resourcefulness here because the shot-noise-defined linewidth is conveniently expressed in milli- or micro-hertz. It is worth re-emphasizing that basically any c.w. laser can in principle be stabilized at these levels: all it takes is lots of servo gain and the requisite short time delay to make it applicable. We turn now to the question of the cavity's intrinsic stability.

4. Stability of the cavity resonance itself.

4.1. Estimating environmental requirements. A useful calculation relative to stable cavities is to estimate the required isolation from vibration and changing temperature changes. Contemporary low-expansion materials have expansion coefficients in the neighborhood of $2 \cdot 10^{-8}$, which would correspond to about 10 MHz per degree. So to have stability in the kHz domain implies temperature control in the 0.1 mK range. Careful temperature servo design can approach this range, given favorable environmental stability. Still, considering the nice linewidth data presented above, why not try...
for 1 Hz? The associated temperature drift over the duration of the experiment must then be 0.1 µK. Double-shell dual-temperature regulators can bring one into the sub-mK range, but it is easy to see that soon we will want to return to locking the system to quantum absorption lines.

As for vibrations, it is also easy to estimate the scale of the sensitivity using the published values of elastic moduli, density and Poisson’s ratio. One finds a compressional distortion of about +10 MHz, assuming the cavity were to be supported vertically from the bottom. Holding it suspended from the top end would lead to a stretching, giving about −10 MHz frequency shift. One could hope by symmetry of mounting to achieve something like 100 kHz/g as the vibration sensitivity. A typical laboratory has 1 milli-g vibration levels at the ~30 Hz frequency associated with mechanical imbalance of rotating electrical equipment. One should reach the sub-kHz level on an ordinary vibration-isolated optical table, while another 50 or 60 dB reduction would be necessary to reduce the phase modulation to sub-radian levels. This proves to be possible!

4’2. Active vibration isolation. – In fig. 6 we show some data on the performance of a 4’ × 8’ optical table which is servo-controlled vertically. The table employed the customary air legs, the vertical vibrations were sensed with a sensitive vertical accelerometer, and the vertical actuator was a low-cost “woofer” from a local stereo store. A time-proportioning air feed/exhaust of the air legs provided the long-range vertical control in preparation for future refinement, i.e. including a tilt servo as well. The seismometer has a simple resonance at 0.8 Hz associated with its suspended mass, and the first of its spurious resonances is at 200 Hz. With the aid of a simulation software package, it was feasible to design an effective 11th-order servo control loop for this system, as indicated in fig. 6.

Fig. 6. – Influence of air legs with tilt and vertical servos. Vertical (velocity) noise on 4’ × 8’ optical table. Vertical sensor is a low-noise seismograph of moving-coil design, 0.8 Hz resonance. Servo ‘actuator’ is electromagnetic type. Servo design compensates excess phase, is of eleventh-order design, but not highly optimized. The peak at 60 Hz is an artifact. Electronic ‘bubble’ tiltmeter feeds time-proportioning pneumatic tilt servo to give 0.5 µrad r.m.s. residual tilt noise.
However, the problem with such an active reduction system is that the directional stability is heavily compromised by the air-legs' integrating character, and these tilts lead to unacceptable low-frequency length changes of the cavity frequency. Of course, we can plan to stabilize the laser table tilts as well. The present cavity mounting is horizontal and shows a tilt-induced cavity frequency shift of 10 Hz/μrad (several times larger than expected). Our preliminary E/W tilt servo shows a noise residual of about 0.5 μrad r.m.s., which leads to a frequency excursion of the beat between our two independent systems of about (5 + 10) Hz as expected. An additional and faster tilt sensor is being considered to increase the tilt servo bandwidth well beyond the present ~1 Hz. The tilt motion of the table at present shows a spectrum peaked up around 6 s/cycle, the known center of the microseismic band associated with ocean wave loading on the sea floor.

43. Long-term cavity length/frequency stability. – Our earlier estimates have indicated that it will require some effort to obtain adequate long-term environmental conditions appropriate for hertz level stabilities, even assuming the cavity per se experiences no drift. Of course, all physical materials do show drift with time, so it is interesting to see what can be achieved experimentally. This information on drift and uniformity of drift will be useful for long-term experiments in space, and for relativity experiments which depend on a fixed length, as opposed to a fixed frequency. One can imagine the internal molecular vibrations as being of very high frequency (−10^{12}/s), continually exploring the shape and extent of the intermolecular potential wells. In some cases a structural defect gives rise to another nearby potential minimum, but one which is inaccessible energetically. There are permanent intermolecular forces of chemical and polarization origin (van der Waals) that bias the specimen with their attractive force. It seems quite reasonable that occasionally the system somehow manages to pool different vibrational energies to jump into the preferred minimum-energy configuration. Perhaps this biased fluctuation process is the physical picture underlying the perpetual slow shortening we call «creep». In ULE[51] this process leads to shortening rates near 10 kHz/day asymptotically, after the larger response due to mirror substrate/spacer body mutual roughness accommodations have slowed down (t ≥ 1 month). These Fabry-Perot reference cavities employ mirrors fabricated with particular care for low optical loss (L ≤ 10 p.p.m.) and are «optically contacted» onto the spacer. An initial shortening ≈ the residual surface roughness has been suggested by Jacobs et al.[52]. However, creep at the later times appears to be a homogeneous process which becomes very uniform in time. Some interesting photo-assisted aging effects can be seen also.

The low-expansion material «Zerodur» is a mixed-phase material in which the low expansion is assured by controlling the crystallite size growth by careful thermal annealing[51]. One could imagine that the creep rate might be asso-
Fig. 7. — Long-term drift of Zerodur cavity. Zerodur rod is hanging at 24°C constant temperature in vacuum: curve a) quadratic fit \( y = -35.6073 + 0.167x - 4.497 \times 10^{-5}x^2 \), curve b) standard line fit. Rapid initial drift after mirror contact not shown. Slow drift of length and deceleration are similar to that observed by BAYER-HELMs et al. Near day 600 we increased the stabilized temperature by 2.4°C, resulting in basically zero drift during the one-month experiment, before restoring the original setpoint.

associated with the gradual stress cracking of these crystalline zones under the internal compressive forces. Such a model would account naturally for the gradual deceleration of the creep as observed by BAYER-HELMs et al. Figure 7 shows our long-term data for one cylindrical spacer of 15 cm \( \varnothing \times 30 \text{ cm} \). The deceleration we observed is basically in accord with the value they observed for the gauge blocks.

We supposed that heating the Zerodur by several degrees would increase the creep rate by providing more available thermal energy. However, the data in the middle of fig. 7 show the physics to be vastly more complex. By increasing the storage temperature by 2.4°C from 24°C we found the creep rate changed more than 10-fold: the creep rate decreased to nearly zero at the higher temperature. An interesting and possibly related hysteresis of thermal expansion in the temperature range below \(-30°C\) has been observed by LINDIG[54] and by JACOBS[55]. A new formulation (Zerodur M) is now available and long-term tests are being prepared using this material[51].

5. – Tuning the laser relative to a fixed cavity.

5.1. Using an acousto-optic modulator for tuning. – The utility of a stable reference etalon in the optical domain is now quite clear, and we have indicated that sub-hertz linewidth and sub-hertz frequency predictability for perhaps one hour are now basically available from our stabilized laser. Unfortunately it is al-
most certain that our interesting atomic-physics resonances will not occur at
one of the cavity’s resonance frequencies. The required frequency offset can
usefully be made up by a modulator which produces a frequency-offset output
beam. Perhaps the most attractive such device is an acousto-optic modulator
(AOM), which produces a (single-direction) input/output frequency shift equal
to the r.f. drive frequency. So one can use a variable r.f. drive frequency, ob-
tained from a computer-programmable source, to obtain the required tuning. A
nice idea is to double-pass this AOM so that the angular offset intrinsic to the
Bragg scattering can be cancelled in a second pass, while the frequency offset is
doubled. The retroreflected beam can be produced with a curved mirror and
isolated with polarization optics[56], or with a collimating lens and 90° roof
prism. In the latter case the beam also emerges on the source side, but spatially
resolved from the input beam. Wide-band modulators allow one to scan a full
free spectral range of the reference cavity, and appropriate software allows the
laser to be scanned many cavity fsr’s by seamlessly joining sequential
scans[57]. Some kind of gain normalization can be introduced in software or by
a hardware intensity-leveling loop to overcome the change in Bragg-scattering
efficiency as the frequency is scanned.

52. Example 1: Line-narrowing and scanning a Ti:sapphire laser. – This
AOM tuning system has been implemented for our Ti:sapphire laser and is
very effective in controlling the laser relative to the stable cavity frequency.
For convenience we employ two AOM-based systems in cascade, the first using
a tunable reference cavity and fast-locking system to line-narrow the laser. This
error signal is derived with a Pound-Drever discriminator, and applied back to
the laser to correct its «instantaneous» frequency. A fast PZT is used, but to ob-
tain a more rapid control the high-frequency components are sent to the volt-
age-controlled oscillator which feeds the first AOM just outside the laser itself.
This AOM has very small frequency excursions to deal with and so, in the inter-
est of optical efficiency, is not doubly passed. This frequency-servo AOM
serves the additional roles of optical isolator and AO intensity controller, in ad-
dition to its role as the fast actuator in locking the laser to the reference cavi-
ety[58]. The intensity noise is reduced by > 30 dB at mid-kHz frequencies, and
a linewidth in the ~ 10 kHz range is obtained. This cavity uses an intracavity
Brewster plate for tuning in the usual way, but its galvanometer is fitted with
flex pivots and angular-position feedback to basically eliminate the troublesome
hysteresis and «stick-tion» often observed otherwise. This line-narrowed and
«civilized» tunable Ti:sapphire laser is then locked to the long-term reference
cavity and scanned as described above[59].

Of course, if the cavity thermal environment is inadequate, the system may
show excessive drift in frequency: we presently have about 1 MHz/h with a
simple system. One is then glad to have the nearly inertia-less tuning capabili-
ty, which allows a software subroutine to revisit a previous strong atomic/
molecular resonance to confirm/re-measure the zero-crossing central frequency, which is separately recorded whenever it is measured. At present we time-stamp all data so that, by making use of this drift data vector, we can make a posteriori correction of drifts in the scan axis. With a foreground/background multitasking approach we hope to calculate the necessary corrected axis frequencies as we proceed. Line fitting accuracy and reproducibility in the range \( \sim 10^{-3} \) linewidths is readily attainable with this approach[57].

5.3. Example 2: Phase-locking laser diodes. – Another approach to precise laser scanning is the tuning of one laser relative to a highly stabilized reference laser. With optical phase-locking techniques, one can transfer the full stability of the reference laser to the «tunable» laser, while enabling precise frequency scans via a tunable frequency offset in the locking loop. The most interesting applications of this stabilization technology may be for laser diode systems, as their reliability and relatively low cost facilitate the design of multilaser approaches to a physical experiment. Unfortunately, for most of the diode lasers now available, the injected charges we intended to correct the laser's frequency at time \( t \) are still present somewhere near the index-guided laser channel after an appreciable time. Taken with the large thermal tuning rate, the result is that the frequency change for an injected milliamperes correction signal has a drastic phase shift with frequency. We have measured a > 90° shift at 25 MHz for the common Sharp LT024780 nm laser[51]. Such large phase changes within a feedback loop are, of course, unacceptable for stability reasons, so it is useful to build multiple phase compensators to develop a more pleasant transfer function[48]. Previous work in this compensator area has been done by Prof. Ohtsu's group[47]. With a five-stage compensator we locked > 98% of the emitted diode laser power into the phase-locked carrier. With a more favorable diode operating at 852 nm (STC-LT50A-03U)[51], a single compensator allowed a > 20 MHz unity gain frequency and > 99.8 power concentration into the carrier[48]. Work on the locking/acquisition strategy continues.

5.4. Moving to higher/wider frequency scans. – Locking with higher and higher beat frequency offsets is another worthwhile and important research area. Applications include bringing a full hyperfine spectrum into a single precise scan, establishing precise frequency intervals between different reference lines, and simplifying various schemes of optical-frequency synthesis such as the all-optical «Divide and Conquer!» scheme being developed by HÄNSCH and his associates[60]. WONG et al. [61] suggest another promising approach to tunability, frequency-locking the outputs of a nearly degenerate optical parametric oscillator.

5.4.1. R.f. harmonic mixing in photodetector. The straightforward approach is to obtain a fast photodetector—bandwidths up to 60 GHz are com-
mercially available—and heterodyne its r.f. output in a microwave mixer to produce a beat in the comfortable range ≈ 100 MHz for the phase-locking loop. Unfortunately this approach requires an r.f. source output at the optical beat frequency. As the radiofrequency increases, such sources rapidly become extremely expensive! With a metal-insulator-metal diode as their mixer, DRUILLINGER et al. used a FIR laser at 119 μm as the «local oscillator» source to detect beats at 2.5 THz[62]. Probably some microwave harmonic-locking approach will be the way to obtain useful bandwidth increases. For example, it has been shown[9] that r.f. drive of a Schottky-barrier detector serves to generate relatively high harmonics internally and recently even 4 THz beats[63] have been detected as the 60th harmonic with this approach! With the development of fast nonlinear transmission lines, one has available for the first time electrical pulses in the picosecond domain. It will be interesting to try to build «sampling-type» optical detectors, for example by putting the optical-signal pair into the last photodetector/Schottky-barrier diode in the transmission line[64].

54.2. Broad-band modulation: modulator-in-a-cavity approach. Another promising approach is based on strong r.f. modulation: a microwave modulator is enclosed in a low-loss cavity. The sideband produced on the first transit is used as the source for a second sideband, and the second for a third and... KOUROGI, NAKAGAWA and OHTSU have reported[65] a spectral width of ~ ± 4 THz, made up of individual lines spaced by the 5.6 GHz modulation frequency. Enhancements of this scheme certainly can be imagined: A nice improvement of the efficiency could be accomplished by «re-cycling» the light reflected back toward the source from the entrance mirror. (Since the in-coupled light is spectrally scattered to many frequencies, there is a painful loss of in-coupling efficiency as the internal cavity leakage field at the input frequency rapidly becomes too feeble to cancel the directly reflected input beam and thus to accomplish the desirable «optical impedance matching».) This same recycling cavity, if short enough, could be resonance-free until one reaches the desired high-order sideband, perhaps several THz away. The frequency-shifted power in this line would be coupled back toward the source and could be separated with a Faraday isolator system. Such schemes may make it feasible to transfer the stability of one optical source in a phase-coherent manner to another source located an appreciable frequency interval away. Perhaps a shift of 1% of the optical frequency could be accomplished in a single step! This would reduce the number of «Divide and Conquer» stages[60] to six or seven.

6. – Applications of stable lasers.

Of course, there are many applications of frequency-stabilized lasers in science and industry. One of the dreams is to build optical-frequency clocks with
dramatically improved characteristics. BERGQUIST and his associates have discussed [66] some of their results about locking a stable laser to a single trapped Hg⁺ ion. Trapped ions represent an excellent system since the ion is held basically without perturbation, for days if we like. Our JILA group is exploring an alternative idea, the so-called «atomic-fountain clock» [67, 68]. In this scheme we imagine bringing neutral atoms to near-zero velocity in a Zeeman-optical trap. One can capture easily $10^7$ atoms in a mm³. Now the magnetic fields are switched off, the molasses light further red-detuned and a few milliseconds long post-cooling interval begins. The product is ~ 3 million atoms within an 8 mm³ ball. Switching the vertical frequencies of the molasses has a profound effect by forming molasses in a moving frame—one could say «walking-wave» molasses—where the captive atomic ball is now moving upward at the speed $v = \lambda \delta \nu$. These preparation fields are now switched off and the supersharpest optical clock transition is irradiated for a few milliseconds. This excitation prepares a coherent admixture of ground and excited atomic states which forms the atomic-frequency coherence of interest. After the atoms reach apogee, fall and re-enter this clock excitation region, a second laser excitation pulse creates a second amplitude for being in the atomic excited state. Another laser can then interrogate the system to enjoy the Ramsey fringes between the two excitations, so as to judge the fraction of the atomic ball that underwent transitions into the excited state of the clock transition. Repeating this process with different detunings gives an appreciation of the location of the atomic center frequency.

CHEBOTAYEV and his associates have considered this approach in some detail.

Several groups are busy investigating microwave clocks built along such lines [69]. Using the «cycling» optical resonance line transition, alkali metals can be cooled efficiently to the Doppler limit, $kT - \hbar \nu$. One then uses another cooling mechanism such as polarization gradient or «Sisyphus» cooling to reach a residual velocity of ~ 3 recoil units. Unfortunately, these supercold alkali atoms do not appear to have an attractive optical clock transition. In fact, surprisingly, one of the problems for an optical standard is an apparent dearth of fully satisfactory atomic transitions, if we put on the additional restriction that the atom must also have a fast and basically closed two-level system for laser-cooling purposes.

The next idea is to use group II metals such as calcium. The initial cooling proceeds very efficiently as before to the Doppler limit. Now there is no hyperfine structure to employ for the second stage of cooling. One possibility is to switch to a weaker transition, such as the $^1S_0 - ^3P_1$ intercombination line. The long lifetime assures a narrow linewidth (~ 400 Hz), but this is uncomfortably weak for a second stage of Doppler cooling, and uncomfortably broad for an atomic-fountain clock, where we might prefer a natural width of (1 ~ 10) Hz. Still, laser-cooled calcium atoms would give an extremely good optical-frequency reference possibility. HOLLBERG has shown that diode lasers will be suitable for this system, so one can see the advantages of portability and low-cost.
potential, along with an inaccuracy of probably $\sim 10^{-15}$ due to light shift effects.

Another possibility in calcium would be to use the $^1S_0 \rightarrow ^3P_2$ magnetic-quadrupole transition as the clock. This line has a superlow natural linewidth [70] ($\Delta \sim 6 \cdot 10^{-5}$ /s), but even so we estimate a power of only some 10 mW would be sufficient to excite the Ramsey resonance using a build-up cavity around the atoms. This would be excited with a spectral comb to produce saturated-absorption signals from atoms with a number of distinct residual velocities. An alternative approach based on use of two-photon excitation is initially attractive since all atoms can contribute to the Doppler-free peak [37], but we are worried about problems with a.c. Stark shifts.

Frequency-doubled Nd laser output can give strong signals in the green on at least five different ro-vibronic transitions in the $I_2B \leftarrow X$ spectrum, although predissociation may limit the linewidths to $\sim 100$ kHz. These linewidths are ($10 \div 100$)-fold larger than one would like for a primary standard, but the signals are very strong and the laser intrinsic stability sufficient that a practical «good enough» standard can be envisioned, based on diode-laser-pumped Nd. Of particular value for this case of broad lines is the method of modulation-transfer spectroscopy [71], which provides good immunity to additive baseline/frequency shifts due to spurious AM produced by the phase modulator. Our experiments so far yield an Allan variance of $\sim 200$ Hz at one second, and a reproducibility from system to system of the same order. Both values represent more than two-orders-of-magnitude improvement over the standard 633 nm HeNe/$I_2$ laser system. So the future seems bright indeed!

7. - Conclusions.

Building on our pre-laser-era spectroscopic heritage, rather spectacular progress in laser stabilization has been made over the past 25 years. We saw first the development of methane-based stable lasers, whose accuracy capability was crippled by relativistic time dilation shifts of $\sim 10^{-12}$. These lasers are still very convenient and practical at this level. The development of external cavity resonances in OsO$_4$ are similar in principle but more attractive in the number of resonances available. What will be the ultimate system of preference in the visible remains to be seen. Ca with laser diode excitation at 657 nm will obviously be interesting. Maybe the magnetic fine-structure transitions between ground-state $^3P$ levels will be good. Diode-pumped, frequency-doubled Nd looks good for precision with simplicity. Maybe...

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[21] The journal Metrologia has a significant fraction of these papers. The biennial Conferences on Precision Electromagnetic Measurements are published in the IEEE Transactions on Instrumentation and Measurement.


[49] I would like to express my appreciation to D. Hils whose painstaking work has been the mainstay of the HeNe results.


[51] These trade names are used for technical communication purposes only, with no implied recommendation.


