



HAL
open science

PROGRESS TOWARD PHASE-STABLE OPTICAL FREQUENCY STANDARDS

Janet Hall, L. Hollberg, Ma Long-Sheng, T. Baer, H. Robinson

► **To cite this version:**

Janet Hall, L. Hollberg, Ma Long-Sheng, T. Baer, H. Robinson. PROGRESS TOWARD PHASE-STABLE OPTICAL FREQUENCY STANDARDS. Journal de Physique Colloques, 1981, 42 (C8), pp.C8-59-C8-71. 10.1051/jphyscol:1981808 . jpa-00221703

HAL Id: jpa-00221703

<https://hal.science/jpa-00221703v1>

Submitted on 4 Feb 2008

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

PROGRESS TOWARD PHASE-STABLE OPTICAL FREQUENCY STANDARDS

J.L. Hall,* L. Hollberg, Ma Long-sheng,† T. Baer‡ and H.G. Robinson°

Joint Institute for Laboratory Astrophysics, National Bureau of Standards and University of Colorado, Boulder, Colorado 80309, U.S.A.

Abstract.— The sensitivity of laser spectroscopy is usually limited by laser amplitude noise of a technical nature. We describe a new and very sensitive detection technique which basically eliminates this source of noise by encoding the resonance information into RF frequencies using FM modulation followed by optical heterodyne and RF phase-sensitive detection. The method is applied to laser stabilization and sub-Doppler spectroscopy. Calculation shows the method provides an available sensitivity within 3.4 dB of the theoretical limit for ideal absorption spectroscopy.

The design of useful and optimized optical frequency references tends to be an evolutionary kind of procedure. Usually each progressive step results from direct interaction between theory and experiment. After a time there tends to be a convergence toward a rather definite design and level of performance. For example, stabilized dye laser linewidths had converged to ~ 1 -2 kHz (1) and 633 nm He-Ne/I₂ lasers had an apparent reproducibility (2,3) $\sim 10^{11}$. Then recently, substantially through interaction with people outside the usual metrological community, some rather amazing new understandings have developed. For example, in the case of the 633 nm HeNe laser stabilized on I₂, the international intercomparisons (4-6) have been useful in demonstrating unmistakably that the actual frequency reproducibility attained can be about one decade worse than the apparent repeatability: both laser power density (5) and I₂ cell preparation (6) have been shown to be of serious concern even though the laser device appears to be operating normally. The former design equilibrium has been further undermined by the introduction (7) and widespread development (8,9) of laser devices based on the 612 nm He-Ne/I₂ coincidence which has a number of important advantages over the conventional 633 nm transition. External-cell experiments (10,11) with this system seem particularly promising, with repeatability in the sub-10⁻¹² domain already reported (10). As for laser stabilization, a suggestion by Drever (12) arising from his interest in gravity wave detection has been synthesized with local interest in dye laser stabilization and has led to a quantum advance in this field (discussed below). Extension of these techniques to precision high-sensitivity molecular spectroscopy was natural and has recently been reported (13). The whole RF sideband technique for sensitive spectroscopy was developed independently by Bjorklund (14) in connection with optical information storage and readout.

These RF electro-optic modulation techniques are extremely powerful for both laser spectroscopy and laser stabilization and form the unifying theme of this report. We first discuss the use of the FM sideband techniques for laser locking. We then illustrate application of the method to sub-Doppler spectroscopy with some precise iodine saturated absorption resonance profiles. Consideration of the absorption process in terms of "darkness radiation" emitted by the molecules allows us

*Staff Member, Quantum Physics Division, National Bureau of Standards.

†Permanent address : East China Normal University, Shanghai

‡Present address : Spectra Physics, Mountain View, California.

°1980-81 JILA Visiting Fellow, on leave from Department of Physics, Duke University, Durham, North Carolina.

readily to discuss nonequilibrium processes and to calculate the signal/noise ratio for several cases of interest. The report concludes with a brief look at a number of interesting new directions suggested by this work.

A fundamental problem in laser spectroscopy arises from the high level of amplitude noise which most lasers display, basically due to causes of a technical nature. A large number of ingenious techniques, such as polarization spectroscopy (15), interference spectroscopy (16) and Polinex (17) have been developed over the years to retain most of the signal while avoiding much of the laser's amplitude noise. Snyder, Raj, Bloch and Ducloy (18) recognized that the laser noise could be minimized by using higher modulation frequencies. However, the upper useful frequency was limited by the population relaxation process implicit in their use of four-wave mixing to obtain sub-Doppler resonances. It is instructive to look at the frequency distribution of the laser amplitude noise. For example in figure 1 we show the amplitude noise spectral density of our relatively quiet ring dye laser. It may be seen that the laser amplitude is shot-noise limited only for (Fourier) frequencies above approximately 2 MHz. It may also be seen that the noise density is ~ 80 dB higher at a more conventional modulation frequency such as 1 kHz. Although, as mentioned above, a plethora of laser spectroscopic techniques have been developed which aim to minimize direct feedthrough of this noise, for sensitive detection it is clearly attractive to encode our optical resonance information into an intermediate frequency signal above ~ 2 MHz.

A very powerful and appropriate optical modulation technique has been suggested recently by Bjorklund (14) and by Drever (12) and demonstrated by Drever *et al.* (19). This method consists of putting FM sidebands on a probe signal and looking for a differential phase change or a sideband amplitude unbalance due to interaction with the resonant medium. In fact this is the optical equivalent of a method first discussed with much the same motivation by Smaller (20) and by Acrivos (21) in the context of NMR. In the optical domain we may conveniently modulate the laser's phase with an external electro-optic phase modulator driven by an RF signal. Alternatively we may frequency modulate the laser with an intracavity electro-optic crystal (or possibly even with a very fast piezo-driven mirror). The modulated laser

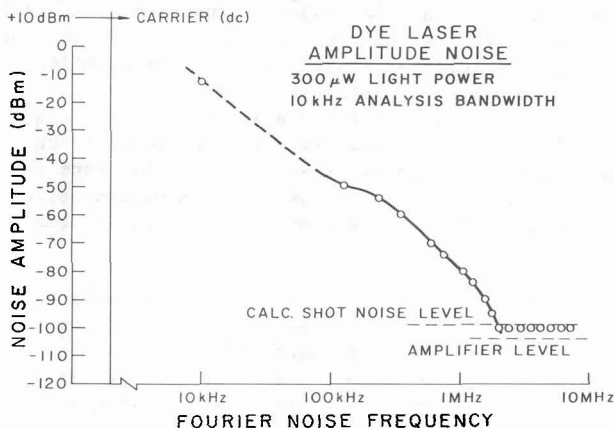


Fig. 1.: Amplitude spectral noise density of our ring dye laser. The shot-noise floor at -148 dB/√Hz relative to the carrier is reached for Fourier frequencies above 2 MHz.

field is of the form

$$\begin{aligned}\bar{E}(t) &= \bar{E}_0 \sin(\Omega t + \beta \sin \omega t) \\ &= \bar{E}_0 \left\{ \sum_{n=0}^{\infty} J_n(\beta) \sin(\Omega + n\omega)t + \sum_{n=1}^{\infty} (-1)^n J_n(\beta) \sin(\Omega - n\omega)t \right\}\end{aligned}\quad (1)$$

where $J_n(\beta)$ is the Bessel function of order n and β is the modulation index. (In phase modulation, β is the peak phase retardation. In FM, $\beta = \Delta\omega/\omega$ where $\Delta\omega$ is the peak frequency deviation, and ω is the RF modulation frequency.) This type of modulation process is very attractive for the intended spectroscopic heterodyne purpose as there is ideally no background intensity modulation at the IF frequency ω for any value of β . In practice, the most interesting range is $\beta \lesssim 1.1$, for which the modulated signal reduces essentially to the carrier and its first two sidebands

$$\bar{E}_{in} \approx \bar{E}_0 [J_0(\beta) \sin \Omega t + J_1(\beta) (\sin(\Omega + \omega)t - \sin(\Omega - \omega)t)]$$

The spatial dependence corresponds to running waves in the (+) z direction. This frequency/phase modulated field will be the input to our absorption cell or resonant cavity for laser stabilization.

As emphasized by Bjorklund (14), a photodetector receiving this pure signal will display no photocurrent at the modulation frequency ω : the two beat photocurrents between the carrier and either sideband are equal and opposite, leading ideally to perfect cancellation. The sideband structure displayed in equation (1) has full odd symmetry around the carrier; thus the resonances observed at frequency ω with this signal will also display full odd symmetry around their spectral center. Experimental verification is discussed below. A second advantage of the RF sideband method is that without loss of signal we are free to choose any convenient value for the modulation frequency, for example, one which is high enough to avoid the laser noise and to resolve sub-Doppler resonances of interest. On the other hand it is convenient to use an RF modulation frequency low enough to minimize pickup of the underlying Doppler profile and to avoid interaction with other hfs components. Lenth *et al.* (22) have drawn attention to the high data rate possible with these techniques arising from a combination of high intrinsic signal/noise ratio with a high Nyquist sampling frequency. A fourth important advantage of these RF sideband techniques for metrology is that the laser output per se is unmodulated. Finally, Drever (12) has pointed out that, even on a time scale much shorter than the resonance inverse linewidth, there is information available about changes of the laser's phase. Before considering such transient response in detail, it is convenient first to discuss stabilization of frequency with these techniques.

Figure 2 shows an improved version of the Pound/Drever laser frequency/phase stabilizer (19). For simplicity, we initially will ignore the portion of the diagram labelled "AM control loop," and describe first the operation of the basic system. A laser to be phase/frequency stabilized is externally phase-modulated (or internally frequency modulated). The appropriate RF frequency is chosen by reference to noise spectral data like those in figure 1. We typically use 7-15 MHz for dye laser locking to have an adequately-high Nyquist frequency for the wideband dye-laser servo, although lower frequencies — down to ~2 MHz — would be adequate for low noise. After the modulator the laser beam has two first-order FM sidebands as described previously.

To observe narrow resonances with FM spectroscopy from a laser frequency control resonator, it is convenient to use a reflection-mode spectrometer. The

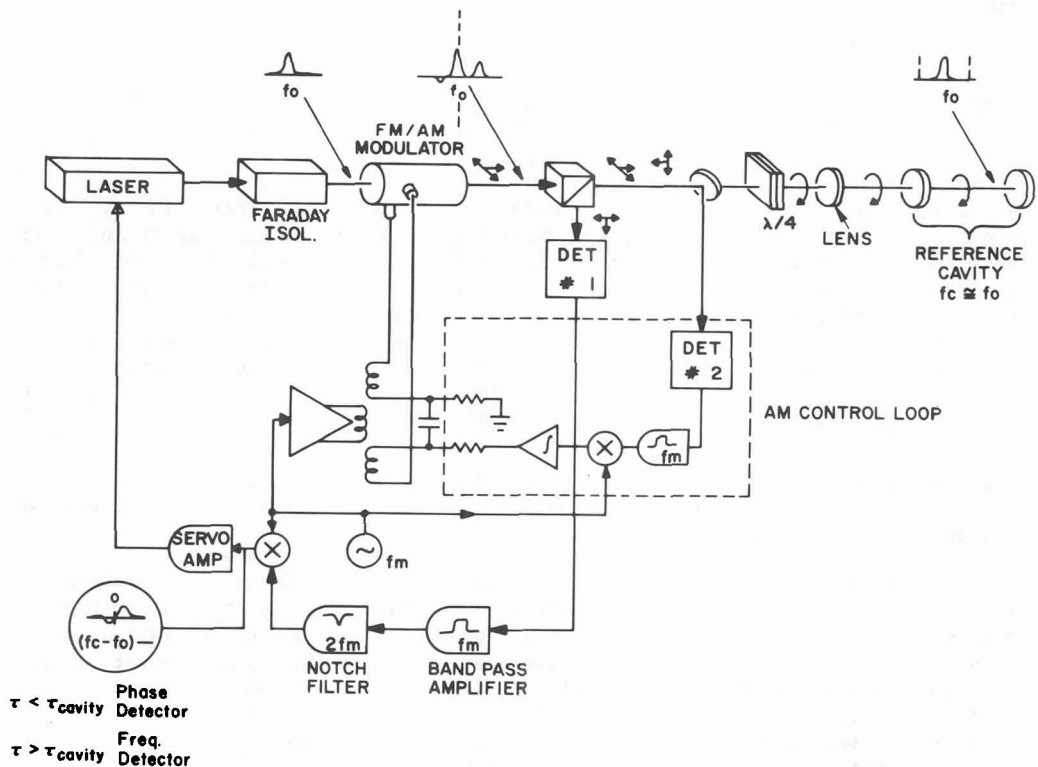


Fig. 2.: Accuracy control of the Pound/Drever Stabilizer via additional AM control loop. See text for a detailed explanation of the basic Pound/Drever stabilizer, without the AM control loop. Here we add detector #2 which measures the spurious amplitude modulation on the beam presented to the cavity. After phase-sensitive detection and integration, the resulting DC bias voltage is applied to the modulator crystal to drive the net AM on the modulator's output to zero.

"three-port circulator" may be conveniently formed with a polarizing prism and $\lambda/4$ plate (or Faraday rotator of 45° rotation). The input laser field with its sidebands passes through the polarizing prism and wave plate to the cavity where the sidebands are effectively totally reflected, as they lie completely outside the resonant linewidth of the cavity. The carrier however, is in near resonance with the cavity and leads to a build-up wave inside the resonator with a relative phase and amplitude that depend on the laser/cavity detuning. The leakage wave back through the coupling mirror adds vectorially to the directly-reflected laser carrier frequency component. The result (in the adiabatic limit) is an effective cavity reflection coefficient which exhibits a narrow dip at resonance, with the expected Lorentzian shape and associated rapid phase variation. The net reflected electric field of the carrier with quadrature-phase is the one useful for producing a discriminator-type resonance. This field, combined with the two reflected sideband fields, is steered to the detector (output) port of the polarizing prism by virtue of the orthogonal polarization resulting from two passages through the wave plate (or Faraday rotator). These three fields, incident on a fast photodetector, will generally give rise to a photobeat current at the modulation frequency unless the two beat currents exactly cancel. Such perfect cancellation occurs when the cavity

reflection phase shift is the same for all three optical frequencies: this condition produces an AM-free signal, although the sideband/carrier amplitude ratio may be larger than that obtainable by frequency/phase modulation due to resonant absorption of the carrier. Any relative cavity/laser detuning will yield a net RF photocurrent which, after amplification, is phase-sensitively detected to recover the desired dispersion-shaped resonance (for laser frequency/phase control).

Figure 3 shows the intensity transmitted by the cavity (top trace) as the cavity frequency is scanned through the optical carrier and its several sidebands. The lower curve shows the phase detector output. One can note that we really are detecting the electric field (not intensity) with the heterodyne method by comparing the strength of the $J_2(\beta)$ second sideband with the $J_1(\beta)$ first sideband signal in its intensity measure (top trace) and field measure (lower trace). Further, we note the window of width 2ω over which the error signal has the correct sign. If the laser happens to briefly fall out of lock, it will be recaptured from anywhere within this window. This property is of inestimable value in providing a lock which is extremely tight yet still robust and tolerant of dye laser bubbles, etc.

To give an impression of the performance of such a stabilizer we present in figure 4 a record of the beat waveform between a pair of such stabilized lasers. For this test, we stabilized both to a single reference cavity to almost eliminate problems due to cavity drift. The isolation of the two lasers (one HeNe and one jet-stream dye laser, both operating at 633 nm) was assured by use of orthogonal polarizations, different electro-optic modulation crystals and RF frequencies, and 1 interference order difference in the control cavity. The ~ 100 MHz beat frequency, obtained in a separate avalanche photodiode, was then heterodyned into the audio domain using a phase-stable frequency synthesizer. The beat frequency, chosen to be ~ 600 Hz, is displayed in figure 4. The signal has been low pass filtered (10 kHz fc) and sampled (at $50 \mu\text{s}/\text{channel}$). Inspection of figure 4 shows there is remarkably little fast phase noise. The laser beat line width, estimated from a histogram of

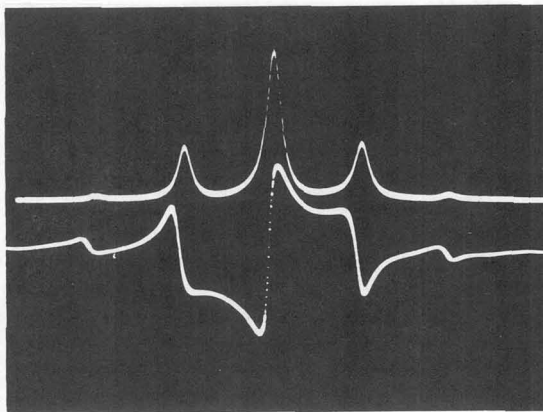
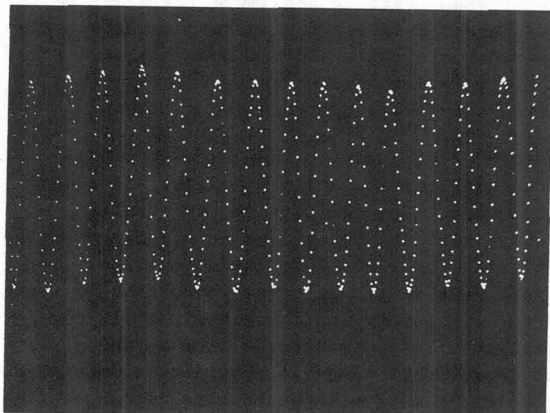


Fig. 3.: Signals in the Pound/Drever Stabilizer. Upper curve, transmitted laser intensity as cavity is tuned over sideband structure of phase-modulated laser beam. First and second sidebands visible symmetrically on both sides of the carrier (prominent central line). Lower curve, output of RF phase-detector with phase reference adjusted for dispersion. Lock point is the zero in the central high slope region. Note that the error signal sign is correct for servo purposes over the full spectral interval 2ω .



LASER BEAT WAVEFORM

Sampling time 50 μ s/channel
 beat frequency \sim 600 Hz
 display bandwidth \approx 10 kHz

Estimated frequency noise \ll 100 Hz!

Fig. 4.: Laser Beat Waveform. Approximately 600 Hz "sinewave" beat between two separate lasers independently-stabilized to an optical cavity. Display bandwidth 10 kHz. Sampling time 50 μ s/channel. Estimated frequency noise below 100 Hz. See text for details.

the number of sampling dots per half cycle, is \sim 60 Hz (19). The basic limiting phenomena are associated with drifts of the dc levels due in part to ground loops and other purely electronic problems. But the main drift problems arose from incidental amplitude modulation produced in the nominally-pure FM modulation process. As noted below, it is straightforward to implement an AM control loop which drives the time-averaged amplitude modulation to zero. It is interesting to note that the same technique has been used earlier by Stein and Turneaure (23) in locking a microwave oscillator to a microwave cavity.

Implementation of the optical AM control loop ("accuracy servo") is facilitated by using as phase modulator a multi-crystal modulator of the type designed originally for amplitude modulation. We place the laser input polarization just a few degrees away from the axis that exhibits electro-optic modulation (rather than at 0° which would give only phase modulation or at 45° which would give only amplitude modulation). Thus at our chosen orientation we have the desired phase modulation plus a small amplitude modulation component of sign and magnitude which are controllable via the dc bias imposed on the crystal. An integrator and HV amplifier can form the closed loop "accuracy servo." In practice, we find it is very important to use flat response photodiodes of reasonably large area (\approx 3 mm ϕ). Apparently on a very sensitive level there are spatial variations in the sideband phases or amplitudes, perhaps arising from spurious reflections within the modulator unit and/or from stria in the modulator crystal itself. With modest care a background AM signal level 60 dB below the maximum signal is readily obtained. This value would correspond to a possible offset in the cavity lock frequency of about 1/1000 of the finesse line width. A detailed investigation of these accuracy-limiting issues is in progress.

We turn now to the application of these RF sideband methods to spectroscopy, especially sub-Doppler spectroscopy using saturated absorption techniques. Figure 5 shows our "optical heterodyne saturated absorption laser spectrometer." We have in the gas cell in addition to the phase-modulated probe beam a counter-running, unmodulated wave at the laser frequency. Saturation by this beam produces sharp

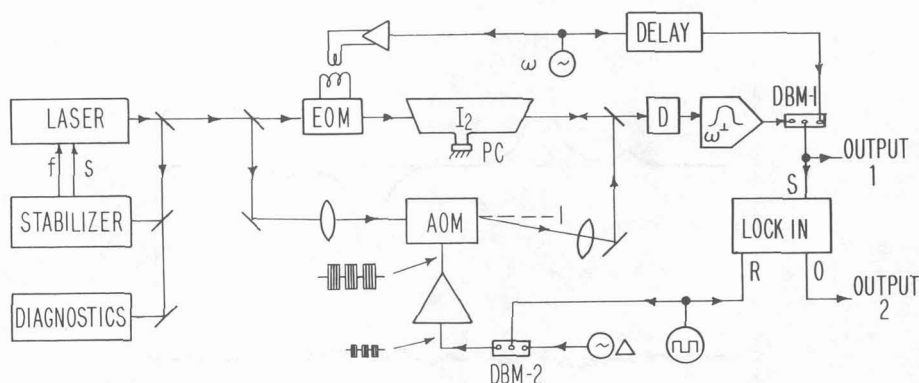


Fig. 5.: Optical Heterodyne Saturation Spectrometer. The frequency-stabilized single frequency laser output is divided into probe and saturating beams by a beam splitter. The probe beam is phase modulated by an Electro-Optic Modulator and passed through the Iodine cell whose pressure is controlled by Peltier Cooler. The saturating beam, frequency offset and chopped by the Acousto-Optic Modulator, is collimated and aligned coaxial and antiparallel to the probe beam in the I₂ cell. The signal-bearing probe beam is detected by a fast photodiode (D) whose output is filtered for the RF component at frequency ω and applied to the signal port of an RF Doubly-Balanced Mixer. The RF reference signal is phase-shifted by an adjustable delay line. The dc output of DBM-1 may be further processed by a lockin amplifier to recover the signal (output 2) synchronous with the saturation chopping.

Bennett holes in the population distribution which give rise to sharply-tuned dispersive/absorptive features as the probe beam and its modulation sidebands tune over them. The sub-Doppler signals arise when the absorbing medium produces a resonant phase shift of the carrier, or when it produces an attenuation or phase shift of either sideband. A summary of the theory of the resulting line shapes was given in Ref. 13 where we also showed that one can obtain resonances of exceptional signal/noise ratio. However, for metrological purposes the symmetry of resonances is perhaps even more interesting than their signal/noise ratio. A remarkable property of the FM sideband technique is that modest apparatus misadjustment (e.g. RF phase shifter setting) does not compromise the intrinsic line symmetry. We have recently published an experimental investigation (13) of this point in which we showed that residual line asymmetry was certainly below 1/500. This interesting, but not yet overwhelming asymmetry limit was controlled by small residual errors in the profile fits. In figure 6 we show the resonances in absorption (curve a) and dispersion (curve c) phase. The five-fold magnified residuals are plotted in curves b and d, respectively. In our fitting functions the saturation broadening was not treated in adequate detail; subsequent work shows that inclusion of the spatially-inhomogeneous saturation effects associated with the gaussian mode shape gives a nice improvement in the fit quality. When still higher S/N records are available, we will want to include different saturation factors for each of the probe's frequency components, even though the probe-generated saturation broadening is rather small.

To explain the principles involved and to clarify the origin of the noise inherent in previous spectroscopic practice, it is useful to consider an absorbing/gas sample interacting with an interrogating radiation field. In figure 7 we show such a gas cell being illuminated by a monochromatic optical input field

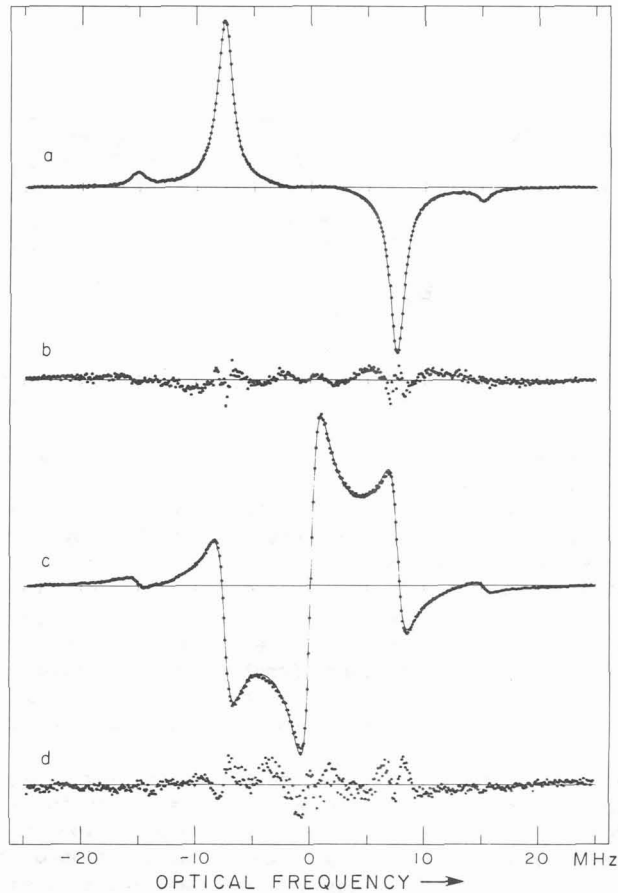


Fig. 6.: Modulated saturation optical heterodyne spectrum of central hfs component of I_2 line at 589.2140 nm, along with best-fitting theoretical representations. (a) Experimental absorption-phase profiles (\bullet) and best-fitting theoretical shape (solid line). Frequency scale 100 kHz per channel. (b) Residuals of absorption-phase fit (5 \times expanded vertical scale). (c) Dispersion-phase profiles and best-fitting theoretical shape. (d) Residuals ($\times 5$) of dispersion fit. Note that the residuals have essentially pure odd symmetry.

$\vec{E}_{in}(t) = \vec{E}_0 \cos \Omega t$. The absorbing gas in general attenuates and phase shifts the input signal, so the field exiting after the cell length L may be written in the form $\vec{E}_{out}(t) = \vec{E}_0 e^{-\alpha L/2} \cos(\Omega t - \phi)$. In this expression α and ϕ are due just to the resonant absorption process. It is highly instructive to focus our attention on change field, $\vec{E}_D(t) = \vec{E}_{in}(t) - \vec{E}_{out}(t)$. In a very real sense this field $\vec{E}_D \approx \vec{E}_0(\alpha L/2 \cos \Omega t + \phi \sin \Omega t)$ represents a "darkness wave" radiated by the medium in equilibrium to provide the "absorption" we associate with Beer's exponential attenuation law. Microscopically, the atomic dipoles — coherently phased by the applied driving electric field — radiate in the forward direction with a sign and phase associated with a driven dispersion oscillator. In terms of the complex susceptibility $\chi = \chi' - i\chi''$, the "darkness" field takes the form $\vec{E}_D = \vec{E}_0 [kL\chi''/2 \cos \Omega t +$

"DARKNESS WAVES"

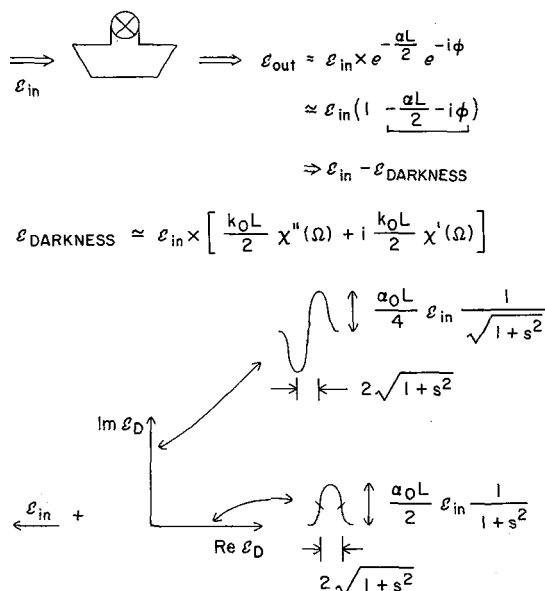


Fig. 7.: Looking at Absorption as a Radiative Process. The absorption occurring within the gas cell may be usefully considered as arising from forward "darkness waves" radiated by dipoles coherently driven by the applied input field. The lower part of the figure shows in the rotating frame how the intrinsic subtraction occurs. The particular functions shown would occur in a Doppler-free (atomic beam) case, but closely-related results are also obtained in the Doppler limit. See Ref. 13 for definition of the effective saturation parameters, S , and more general formulae.

$kL\chi'/2 \sin\Omega t$]. The physical reality of the "darkness wave" is readily demonstrated by experiments in which the input laser frequency (or phase) is abruptly switched to a new value: the "darkness radiation" appropriate for the original condition persists for a time of the order of the transverse, dipolar relaxation time. Such coherent optical transients ("free induction decay transient") following laser frequency switching have been widely utilized for gas phase relaxation spectroscopy (24,25). Phase switching transients have also been studied (26). To make closer connection with the previous discussion, we note here the strong physical resemblance of this matter-generated "darkness wave" to the wave leaking out of a high-finesse Fabry-Perot resonator. The latter field has a damping time associated with the lifetime of photons in the optical cavity or, ultimately, with the related spectral linewidth of the cavity's resonance function. Drever suggested use of this radiation as a "flywheel" or "phase memory" to allow measurement of the instantaneous laser phase (12). By now we have successfully used both "cavity storage" and "darkness wave" radiations for laser phase-locking.

We have suggested that the influence of laser noise can be vastly reduced if we can encode our resonance information to an appropriately-high intermediate frequency

(IF) for processing using heterodyne techniques. Alternatively we could consider signal detection at zero IF frequency (homodyne detection) using an ideal laser having only the inescapable quantum fluctuations (pure shot noise). It is interesting to compare the sensitivity of the two approaches. Let $\eta = c\epsilon_0 \cdot A \cdot e / \hbar \Omega \cdot \eta_0$ where A is the effective area of the laser field distribution and η_0 is the diode's quantum efficiency. The quantity η is effectively the diode sensitivity in amps/(volt/m)². Homodyne detection corresponds to the usual straightforward absorption signal: there will be but one frequency, the role of the "local oscillator" being played by \vec{E}_{in} and the signal by \vec{E}_{Dark} . The "beating" of these two fields in the diode will produce a diode current

$$i_{diode} = \eta \langle (\vec{E}_{in} - \vec{E}_{dark})^2 \rangle \approx \eta \langle \vec{E}_{in}^2 \rangle - 2\eta \langle \vec{E}_{in} \cdot \vec{E}_{dark} \rangle = i_{DC} + i_{homodyne}$$

The quadratic term in \vec{E}_{dark} is neglected because of its small relative size. In $i_{homodyne}$, the quadrature phase component of \vec{E}_{dark} averages to zero in the "phase-sensitive homodyne detection process." We can see that a noise modulation on the laser output present in i_{DC} could add directly to the homodyne signal and thus tend to obscure the absorption. Finally,

$$|i_{homodyne}| = 2\eta \langle E_{in} \cdot (E_{in} \frac{\alpha L}{2}) \rangle = \eta E_{in}^2 (\alpha L) = \eta \frac{E_0^2}{2} \alpha L = i_{DC} \cdot \alpha L$$

The minimum noise condition will arise if the laser has no amplitude fluctuations whatsoever of a technical nature. We then will have a pure shot-noise limited detection process, resulting in an rms noise current of $i_n = \sqrt{2ei_{DC}B}$, where B is the effective detection bandwidth. The minimum detectable absorption signal therefore corresponds to $i_n = i_{homodyne}$, giving

$$(\alpha L)_{min, Homodyne} = \frac{(2ei_{DC}B)^{1/2}}{i_{DC}} = \left[\frac{2eB}{(\eta E_0^2)/2} \right]^{1/2}$$

In heterodyne detection, to obtain the frequency-offset "local oscillator" needed for a nonzero "IF frequency," we may convert a portion of the original laser beam into sidebands by the phase modulation process previously described. Suppose we produce two sidebands of amplitudes $E^+ = J_1 E_0$, $E^- = J_1 E_0$ and frequencies $\Omega^+ = \Omega + \omega$ and $\Omega^- = \Omega - \omega$, respectively. The carrier frequency is Ω and its amplitude will be $E^c = J_0 E_0$. The main heterodyne signal current of dispersion phase will arise from the carrier-related darkness wave beating with each sideband. Within the total current $i_{DC} = \eta \langle E_{total}^2 \rangle$, we will have two heterodyne terms at ω :

$$i_{het} = 2\eta \left[\langle E^+ E^c \rangle \frac{\alpha L}{2} + \langle E^- E^c \rangle \frac{\alpha L}{2} \right] \sin \phi$$

$$= \eta \cdot \alpha L \cdot \frac{E_0^2}{2} \cdot 2J_1(\beta)J_0(\beta) \cos \omega t \sin \phi$$

The modulation-dependent function $2J_1(\beta)J_0(\beta)$ has its maximum value of 0.677 at $\beta \approx 1.1$. Thus we see that if the total available laser power is to be used (i.e. we have to "spend" some of our carrier power to produce sidebands for the heterodyne process), then the signal cost is a factor 0.67 (≈ -3.4 dB). If we had a phase modulation process that produced no higher order sidebands, for a sideband relative amplitude f, the remaining carrier would be $(1-2f^2)^{1/2}$. The function $2(1-2f^2)^{1/2}$ has a maximum of $1/\sqrt{2}$ (≈ -3 dB) at $f=1/2$. The total power falling on a diode, and

hence the shot-noise generated background noise is unchanged by the modulation. Thus we conclude that for a signal cost of <3.4 dB, the use of FM sideband techniques enables us to encode the resonance information to a frequency high enough that we can totally escape the laser technical noise. Further, we see that simple external phase modulation (or laser frequency modulation) is a completely satisfactory way to produce the necessary phase-coherent local oscillator sidebands.

It is also worth noting that often the laser power output is larger than the necessary or optimum level for our spectroscopic task. For example, in saturated absorption spectroscopy, excess power leads to wider spectral lines. In this limit, optical heterodyne spectroscopy even more closely approaches the optimum signal recovery possible using shot-noise-limited homodyne detection.

Usually, in practice the experimental absorption sensitivity is far worse than the theoretical limiting value. It is interesting to estimate the magnitudes involved. For example, a 1 mW probe beam offers an intrinsic $S/N = 4.3 \times 10^7$ for 1 Hz detection bandwidth [$i_{DC} = 600 \mu A$, $i_{noise} = 1.4 \times 10^{-11} A$]. A two level atom has an absorption cross section $\sim \lambda^2 \sim 10^{-10} \text{ cm}^2$ for its resonance line. For net $S/N = 10$ in a 10 cm absorption length we would need 230 atoms/cm³, which corresponds to about 10^{-14} torr. Taking the Doppler broadening into account (~ 1 GHz vs natural linewidth ~ 10 MHz) increases the necessary pressure 100 fold to 10^{-12} torr. It is clear that very weak absorption (or dispersion) can be studied with this high sensitivity.

With this kind of remarkable sensitivity a number of interesting possibilities present themselves: For example, two photon absorption should be visible in absorption when the carrier and either sideband sum to the two-photon allowed transition energy (27). Related two photon processes in four-wave mixing have been studied with heterodyne techniques (28). Several types of two photon dispersion signals may also be expected. Alternatively, unprecedented values of signal/noise ratio may be expected for more conventional absorption samples. We believe that it will be particularly attractive to study molecular absorbers within a "build-up" resonator external to the laser source. An obvious candidate is the 612 nm line in I_2 using a HeNe laser. Two distinct modulation frequencies may be used as in passive hydrogen devices (29) to control cavity tuning and laser frequency separately. The high available sensitivity may be important in ameliorating the light intensity shifts noted in the introduction.

Finally, some quite interesting four-component patterns arise when the detector views the originally-unmodulated "saturating" beam. Modulation is transferred from the counter-running modulated wave to the unmodulated one via a multiplicity of Doppler-free four wave resonant mixing processes. J. Shirley has given a theory of these signals and these results will be reported in detail elsewhere (30). The signals arise basically from pulsating population gratings and so carry interesting information about relaxation rates as discussed previously by Raj et al. (31).

In this paper we have discussed the new and extremely powerful FM sideband technique which is attractive both for spectroscopy and for laser frequency stabilization. The method provides resonance profiles of exceptional signal/noise ratio in principle and in practice: the signal/noise ratio is only 3.4 db less than that associated with ideal absorption spectroscopy. The method provides resonances of basically ideal (odd) symmetry around the spectral center: experimentally we have demonstrated line profiles anti-symmetric at least to 1:500. A further advantage is that the necessary "discriminator curve" for the servo control of laser frequency is obtained without imposing frequency modulation on the laser per se. This in turn greatly enhances the utility of these devices for metrology. Finally, investigations

of metrologically-interesting resonances in external resonator gas absorption cells are in progress and laser line-narrowing into the sub-Hertz domain seems imminent.

The authors are indebted to G. C. Bjorklund and M. D. Levenson for pre-publication copies of their recent and closely-related work. We also thank M. Ducloy, D. Bloch and E. Giacobino for preprints and useful discussions. One of us (JLH) is happy to acknowledge many stimulating conversations with R. W. P. Drever. We thank J. Shirley for several theoretical contributions to this work. This work has been supported in part by the National Bureau of Standards under its program of precision measurement research for possible applications to basic standards and in part by the National Science Foundation and Office of Naval Research through grants to the University of Colorado.

References

1. HELMCKE J., LEE S. A. and HALL J. L., Appl. Opt. (submitted).
2. HANES G. R. and DAHLSTROM C. E., Appl. Phys. Lett. 14 (1969) 362.
3. CÉREZ P. and BRILLET A., Metrologia 13 (1977) 29.
4. CHARTIER J. M., HELMCKE J. and WALLARD A., IEEE Trans. Instrum. Meas. IM-25 (1976) 450.
5. MELNIKOV N. A., LAYER, H. P. and HALL J. L., unpublished.
6. ROWLEY W. R. C. and MELNIKOV N. A., private communication; ROWLEY W. R. C. and MARX B. P., Metrologia 17 (1981) 65.
7. CÉREZ P. and BENNETT S. J., Appl. Opt. 18 (1979) 1079.
8. CHARTIER J. M., private communication, and quoted in Ref. 10.
9. CIDDOR P. E. and BROWN N., Opt. Commun. 34 (1980) 53.
10. CÉREZ P., Brilliet A., MAN-PICHOT C. and FELDER R., IEEE Trans. Instrum. Meas. IM-29 (1980) 352.
11. DSCHAO K., GLASER M. and HELMCKE J., IEEE Trans. Instrum. Meas. IM-29 (1980) 354.
12. DREVER R. W. P., private communication.
13. HALL J. L., HOLLBERG L., BAER T. and ROBINSON H. G., Appl. Phys. Lett. 39 (1981) 680; also in Laser Spectroscopy V, eds. A. R. W. McKellar, T. Oka and B. P. Stoicheff (Springer-Verlag, Heidelberg, 1981) p. 15.
14. BJORKLUND G. C., Opt. Lett. 5 (1980) 15; BJORKLUND G. C. and LEVENSON M. D., Phys. Rev. A 24 (1981) 167.
15. WEIMAN C. and HANSCH T. W., Phys. Rev. Lett. 36 (1977) 1170.
16. KOWALSKI F. V., HILL W. R. and SCHAWLOW A. L., Opt. Lett. 2 (1978) 122.
17. DABKIEWICZ Ph., HANSCH T. W., LYONS D. R., SCHAWLOW A. L., SIEGEL A., WANG Z-Y. and YAN G-Y., in Laser Spectroscopy V, eds. A. R. W. McKellar, T. Oka and B. P. Stoicheff (Springer-Verlag, Heidelberg, 1981), p. 178.
18. SNYDER J. J., RAJ R. K., BLOCH D. and DUCLOY M., Opt. Lett. 5 (1980) 163.
19. DREVER R. W. P., HALL J. L., KOWALSKI F. V., HOUGH J., FORD G. M. and MUNLEY A. J., September 1979.
20. SMALLER B., Phys. Rev. 83 (1951) 812.
21. ACRIVOS J. V., J. Chem. Phys. 36 (1962) 1097.
22. LENTH W., ORTIZ, C. and BJORKLUND G. C., Opt. Lett. 6 (1981) 351.
23. STEIN, S. R., thesis, Stanford University, pp. 19-21, 1974 (under J. P. Turneaure).
24. HALL, J. L. in Atomic Physics III, eds. S. J. Smith and G. K. Walters (Plenum, New York, 1973), pp. 615-646.

25. SHOEMAKER R. L. and BREWER R. G., Phys. Rev. Lett. 27 (1971) 631; BREWER R. G. and GENACK A. Z., Phys. Rev. Lett. 36 (1976) 959.
26. GENACK A. Z., WEITZ D. A., MACFARLANE R. M., SHELBY R. M. and SCHENZLE A., Phys. Rev. Lett. 45 (1980) 438; errata p. 1044.
27. One expects a two-component Doppler-free resonance with splitting ω , centered on and antisymmetric around one half the natural frequency.
28. BLOCH D., DUCLOY M. and GIACOBINO E., quoted in DUCLOY M. and BLOCH D., J. Physique, to appear Jan. 1982.
29. WALLS F. L., US Patent 4-122408, issued Oct. 24, 1978.
30. HOLLBERG L. and SHIRLEY J., private communication.
31. RAJ R. K., BLOCH D., SNYDER J. J., CAMY G. and DUCLOY M., Phys. Rev. Lett. 44 (1980) 1251.