Precision Spectroscopy and Laser Frequency Control Using FM Sideband Optical Heterodyne Techniques

J.L. Hall*, T. Baer, L. Hollberg, and H.G. Robinson
Joint Institute for Laboratory Astrophysics
University of Colorado and National Bureau of Standards, 325 S.Broadway
Boulder, CO 80302, USA

In fundamental physical experiments using laser servolocking techniques,1 in passive ring laser gyro experiments, and in precision atomic/molecular spectroscopic measurements as well, the two overriding experimental concerns are maximizing the signal-to-noise ratio and obtaining highly symmetrical resonance profiles to facilitate precise line splitting. In this paper we discuss the technique of FM sideband optical heterodyne spectroscopy,2,3 which appears to be the experimentally optimum method for obtaining such high precision resonance profiles of maximal signal/noise ratio. We discuss the process in simple physical terms relative to stabilization of a laser to a resonant optical cavity, before turning to sub-Doppler resonance spectroscopy obtained by applying the sideband techniques to cw dye lasers and color center lasers. The final topic concerns our study of optical transients resulting from laser phase changes, studied with the optical heterodyne technique.

We begin with the observation that resonance absorption measurements in whatever frequency domain — rf, microwave or laser — are almost always limited in practice by residual noise of a technical nature present on the output of the coherent source. In the laser case this problem has led to the invention of polarization spectroscopy4 and interference spectroscopy5 as techniques to reduce the direct feedthrough of laser amplitude noise while preserving most of the signal. Recent progress has involved the use of high modulation frequencies6 and four-wave mixing7 to further improve the sensitivity. Although the technique of FM sideband optical spectroscopy recently has been invented independently at least twice,5,8 and although its realization and application in the laser domain as optical heterodyne saturation spectroscopy will undoubtedly provide major advances in sensitive and precise spectroscopy, in fact the technique was first discussed in connection with NMR spectroscopy by SMALLER9 and ACRIVOS.10 For historical interest and perspective about the optical work, we briefly describe the several component ideas developed for rf and microwave spectroscopy.

To stabilize a microwave klystron oscillator to a stable microwave cavity, POUND11 introduced the idea of a reflective modulator driven by an rf source, say at frequency ω, to produce sidebands on the carrier. A high Q reference cavity is used in a reflection-mode experimental setup. When the modulation frequency is high enough, the resulting FM sidebands lie well outside the passband of the resonant cavity and are fully reflected. However, the carrier, being in near resonance with the cavity, is reflected with a tuning-dependent phase and amplitude. When the reflected sidebands

*Staff Member, Quantum Physics Division, National Bureau of Standards.
and phase-shifted carrier are heterodyned in the detector, one has the
cavity's resonance information converted to amplitude and phase changes of
the output signal at the rf frequency \( \omega \). After rf amplification, sub-
sequent phase-sensitive detection of this signal relative to the modulator's
drive phase at \( \omega \) leads to the desired "discriminator-type" response func-
tion. This description is totally appropriate also for the optical case.\(^8\)

POUND recognized the advantages of processing signal information at an rf
frequency, thereby avoiding much of the intrinsic source noise and the de-
tector's low frequency excess noise. TREL and FAIRBANK,\(^12\) in building
superconducting cavity-stabilized oscillators, further improved the long-
term stability with an experimental topology in which the FM sidebands and
carrier leaving a phase modulator travel together along the same path to
the resonant cavity and -- after reflection -- on to the detector. Me-
chanical perturbations thus develop phase-shifts proportional only to the
difference in the k vectors.

STEIN and TURNEAU\(^3\) further improved the locking precision by meas-
uring in a separate detector, before interaction with the cavity, the re-
sidual AM component of the modulation sidebands. This AM information was
then used to servo-control the dc bias on the modulator crystals to produce
pure frequency modulation, free of any spurious AM. A locking stability of
\( \tau = 100 \) sec, \( 6\beta = 10 \text{ Hz} \) \( = 6 \times 10^{-12} \) was obtained with a microwave cavity
\( Q = 10^{11} \) (optimum stability = \( 10^{-8} \) cavity line widths).

The intrinsic resonant symmetry associated with pure FM sideband spec-
troscopy is a fundamental issue for high precision lineshape studies and
precision servo locking. It appears that our optical studies may be the
first to investigate this point. See below.

To choose the rf modulation frequency intelligently we consider the fre-
quency distribution of the laser's amplitude noise, presented in Fig. 1 for
our cw ring dye laser. Several observations may be made from the data.

1. For sufficiently high Fourier frequencies, \( \omega/2\pi > 2 \text{ MHz} \), the dye
laser output is indeed quiet enough to closely approach the fundamental
photoelectron shot noise limit associated with the dc light level. (Not
shown on this graph are some extremely high rates around 17 and 85 MHz
that arise from argon laser mode-beating. It is impressive how strongly
these frequencies propagate through the dye laser!)\(^8\)

2. At the usual kHz modulation frequencies the laser noise spec-
tral density is increased by \(-80 \) dB (\(-10^8 \) power ratio) over the fundamental
shot noise level. Under such conditions it is not surprising to find that
over the years laser spectroscopists have developed a very large body of experi-
mental techniques which aim to exclude laser noise from the measurement
channel.

3. For this particular photodiode preamplifier, a light level of 300 \( \mu \text{W} \)
is marginally sufficient for the shot noise level to exceed the noise floor set
by the amplifier. A larger feedback resistor, \( R_F \), could be used to re-
duce the amplifier noise and useful rf bandwidth proportionally (present
\( R_F = 3k \) gives \( f_{\text{dB}} = 17 \text{ MHz} \)). A further necessary criterion is
\( R^2 R_F > 0.025 \) V to mask thermal noise from the feedback resistor.

The use of optical FM sideband techniques has been suggested in relation
to interferometric gravity-wave antennas.\(^14\) From a physical discussion of
the transient ringing behavior of the carrier field stored in the proposed
high finesse resonant optical cavity, Drever\(^8\) was led to suggest use of the
cavity "leakage" field as a phase reference for phase locking a laser to
the reference cavity. This surprising idea of phase locking a laser onto
itself via a reference cavity does not appear to have been previously sug-
gested although the stabilized microwave oscillator of Stein was in fact
operating in this transient regime in view of the high Q of the supercon-
ducting cavity. POUND has shown\(^15\) that this system exhibits a smooth
crossover into his previously described cavity frequency discriminator
mode for times longer than the cavity ringing time.

It may be interesting to present some information about our experimental
laser stabilization results\(^16\) with these methods. To establish the reality
of tight, phase-locking of lasers to a cavity, we used the Pound/Drever

Small portions of each beam were mixed together via a separate beam
splitter and avalanche diode receiver. The resulting 100 MHz beat was down
converted in a balanced mixer by heterodyne with a stable frequency synthe-
sizer. A surprisingly stable beat frequency was produced by tuning the
synthesizer about 600 Hz away from the 100 MHz inter-laser beat. We could
easily see that the rf heterodyne line width -- and hence the independent
laser line widths -- were well below 100 Hz. A complete description will
be published elsewhere,\(^16\) and new experiments are being prepared.

We now turn to the use of FM sideband techniques for sub-Doppler spec-
troscopy. The two most striking features of this method are the high
signal-to-noise ratio and the excellent symmetry of the observed resonances. The physical origin of narrow resonances within the Doppler profile is most easily understood by representing the original pure phase-modulated wave in terms of an optical carrier \( \omega \) plus FM sidebands spaced by the modulation frequency \( \omega_m \). We have

\[
E = E_0 \sin(\omega t + \beta \sin \omega t) = E_0 \left( \sum_{n=0}^{\infty} J_n(\beta) \sin(\omega_n t) + \sum_{n=1}^{\infty} (-1)^n J_n(\beta) \sin(\omega_n t) \right)
\]

where \( \beta \) is the modulation index and \( J_n(\beta) \) is the Bessel function of order \( n \). To simplify the discussion we may assume a small modulation index \( \beta \), so the full FM signal collapses to the carrier and its first-order modulation sidebands. After passing through our absorption cell these three frequencies will be incident on the photodetector where they will produce, in addition to the three terms at dc, two alternating currents at the rf modulation frequency \( \omega_m \). An essential point of the FM spectroscopy method, as emphasized by BJORKLUND, is that these two rf currents are equal but of opposite signs. This cancellation, characteristic of pure phase modulation, can be affected by the optical medium by phase shift of any of the spectral components or by differential attenuation of one of the sidebands. BJORKLUND emphasized the latter mode, especially where the modulation frequency was so high that only one of the sidebands interacted appreciably with the absorbing medium. For precise spectroscopy, on the other hand, we prefer to use sidebands spaced close together with respect to the Doppler width and/or possible hyperfine structure. Of course the modulation frequency must be high enough to avoid the regime of laser technical noise. See Fig. 1.

Figure 2 shows a schematic diagram of the apparatus employed in our spectroscopy experiments. The output beam of a cw ring dye laser beam passes through a beam splitter, an external phase modulator, and a 50 cm \( l_2 \) absorption cell. This 1 mW probe beam of 3 mm diameter causes little saturation. After passing an additional beam splitter, this information-bearing probe beam is detected by a photoreceiver broadly tuned to the rf frequency (\( \omega/2\pi = 15 \) MHz) driving the phase modulator. The resulting rf signal at \( \omega \) is coherently detected in a doubly-balanced mixer whose phase reference comes from the rf source at \( \omega \) via a step-selected delay cable. The strong unmodulated laser beam from the first beam splitter passes through an acousto-optic modulator whose Bragg-deflected output beam is introduced via the second beam splitter to become the counter-running saturating beam (\( \omega \) = 20 mW).

The probe and saturating beams are coaxial, collimated, and precisely anti-parallel. As noted by SNYDER et al., the frequency shift (\( \Delta \omega = 80 \) MHz) associated with the acousto-optic Bragg-scattering prevents interferometric noise problems due to optical scattering of the saturating beam back into the probe channel direction: heterodyne terms with this radiation do not lie at the information-bearing frequency \( \omega \). The sub-Doppler resonances are efficiently isolated from any drifts or background by using phase sensitive detection of the rf mixer output. See Fig. 2.

This apparatus responds to differences in the medium's refractive index across the spectral interval \( 2\omega \). Since our modulation frequency, \( \omega_m \), is much smaller than the Doppler line width, the probe beam alone picks up only a very weak signal due to the overall Doppler profile. However, when we supply a counter-running, saturating beam, we can affect the absorption/displacement on a velocity-resolved basis. For example, suppose the saturating beam modifies the population of the velocity group detuned by one homogeneous line width on either side of the group resonant with the carrier frequency in the probe beam: these tuning conditions will produce the maximum dispersive effect. (Tuning the saturating beam to resonate with the same molecules as the probe carrier leads to zero dispersive effect.) These small changes in the medium's index of refraction integrate over the cell length to a resonant phase shift for the carrier which unbalances the equality of the rf detector currents at the modulation frequency \( \omega_m \). When the saturating beam influences the molecules nearly resonant with either sideband field, an analogous dispersion-shaped feature will result, so that two auxiliary resonances are obtained with \( 2(\omega/2) \) separation. Under suitable experimental conditions the entire signal will be purely odd symmetric around the central dispersion resonance.

To illustrate these ideas experimentally we have used the 127I \( \text{I}_2 \) line at 589.2141 nm. Figure 3c shows a spectrum taken with the rf phase shift set to recover the saturated dispersion signal. The signal-to-random-noise ratio for the display is \( \approx 200 \) in a 10 kHz bandwidth. This signal-to-noise ratio should permit laser stabilization to \( < \text{\text{10}}^{-6} \) of a typical 1 MHz \( l_2 \) line width in a 1 s averaging time.

Another type of signal may be obtained when the radio frequency phase reference has been adjusted 90° from this present quadrature location. In
the in-phase position we will be sensitive mainly to changes in the amplitude of either of the two sideband signals. This detection mode has recently been used by BJORKLUND and LEVISON\(^1\) for sub-Doppler spectroscopic experiments, with such a high FM frequency \(\omega\) that only one sideband appreciably interacts with the Doppler distribution. In contrast, our probe frequency separations are much smaller than the Doppler width; in the absence of the saturating beam our linear absorption signal at \(\omega\) approximates the derivative of the Doppler profile. With the saturating beam present, there are two resonant tuning conditions in which preferential bleaching occurs for one or the other of the two phase shift sidebands. Considering that the beat terms of these sidebands with the carrier have opposite signs, we again find a saturation resonance profile with odd symmetry around the center frequency. Figure 3b shows the in-phase saturation resonance signal at the rf balanced mixer output in which the large frequency scan clearly shows the Doppler (derivative absorption) background. For comparison, note that the rf dispersion signal (Fig. 3c) even without saturation chopping is almost perfectly free of additive Doppler background problems. The corresponding fluorescence signal is displayed in Fig. 3a. The optical frequency scale can be estimated from the observed spacing between the central and lower frequency singlets: (580±10) MHz.

We have made a simple rate equation theory of this optical heterodyne saturation spectroscopy, extending the results of BJORKLUND\(^2\) to include the effects of finite absorption and the nonlinear absorption/dispersion response of the resonant medium. We calculate the path-integrated phase and amplitude changes of the several spectral components of the probe beam due to the (nonlinear) medium's (complex) index of refraction. We follow the cross-terms of interest to recover rf currents at \(\omega\), phase-shifted and attenuated by the medium. The medium is modeled by an infinitely wide distribution of narrow, saturable velocity packets. Molecular transit time effects are lumped into the effective relaxation rate. After the Doppler convolution it is found that the sharp nonlinear resonances have pure odd symmetry\(^3\) for all values of the rf phase shift, modulation index, and absorption coefficient. This absence of underlying opposite symmetry terms is fundamentally important in using these signals for laser frequency locking. See below. To facilitate presentation of our results, we introduce the notation

\[
L = \frac{\Gamma\omega^2}{\Gamma + (\omega)^2}, \quad D = \frac{\Gamma^2\omega^2}{\Gamma + (\omega)^2},
\]

with \(\omega = \Omega - \pi\Omega/2(2\Omega/2), \quad \Omega = \pi\Omega/2(2\Omega/2)\), and \(\omega = \Omega - \pi\Omega/2(2\Omega/2)\). The natural center frequency, \(\Omega_0\), is shifted to \(\Omega = \Omega_0 + \Delta/2\), depending on which acousto-optic sideband was chosen. \(\Gamma\) is the homogeneous width of a velocity packet, power-broadened by the average of the sideband and saturating intensities. The path-integrated absorption measure is \(A = \alpha L\), where \(\alpha L\) is the saturated absorption coefficient and \(L\) the cell length. For the in-phase case, in the limit of modest modulation index \(\beta + 1\) and small absorption \(A << 1\), we have \(S_{\text{sat}}^0 = B + A\gamma(L - L')\gamma(L - L')\gamma(L - L')\cos\omega t\), which is clearly interpretable as the beating by the saturating beam of the opacity resonant with the four sidebands. The term at \(\omega\) vanishes because of equal but opposite contributions from each pair of sidebands.

The rf quadrature term in the same low absorption, modest modulation index limit is \(S_{\text{sat}}^0 = -CA_1[(\omega_0 + \Omega_2)(0^+ + 2^+ + 4^+ + 6^+) + 1^+ + 3^+ + 5^+]\sin\omega t\). At low saturation, the model gives \(B = C\), which is well confirmed experimentally\(^2\). The most conspicuous effect of higher order terms in the absorption \(A\) is a small step near zero detuning in the in-phase signal.

To test the full theoretical function from which the above limiting forms were derived, we obtained high precision profiles of an isolated hfs component near the Doppler center of the \(1^2\) line at 589.2141 nm. See Figs. 3 and 4. The dye laser was locked to a highly stable high finesse cavity which was itself locked to a He-Ne local oscillator which was in turn scanned relative to a \(125^1\) stabilized He-Ne laser via a computer-driven frequency synthesizer. Linear laser width was \(< 10\) kHz. The data are displayed as points in Fig. 4a (in-phase case) and Fig. 4c (dispersion phase). The full theory, separately least-squares fitted to the two sets of data, is as plotted as solid lines in Fig. 4a. Residuals of the fits, magnified five-fold, are shown in Figs. 4b and 4d. The agreement with the experimental data is seen to be truly remarkable! For example, the fitted displacement of the absorption phase components is \((7.56±0.01)\) MHz versus \(15.114\) MHz expected. The dispersion phase gives \((7.51±0.05)\) MHz displacement. The absorption phase gives a line width \((HWHM) = (0.77±0.01)\) MHz; the dispersion phase also gives \((0.77±0.01)\) MHz. Consideration of the spatial averaging of saturation further improves the fits. The \(I_2\) pressure was \(< 1\) mbar.

The use of the dispersion-phase signal for laser locking is extremely attractive as no slight error (e.g., in setting the rf phase-shift) can spoil the symmetry of the resonance (see Fig. 4). For example, our several tests on the dispersion-phase residuals, Fig. 4d, show no spurious term of even symmetry to within the noise level (<1/500 of the pp dispersion signal). Thus we can immediately see the prospect of laser stabilization to basically any of the iodine hfs lines with kilohertz accuracy.

Another interesting aspect of these resonances concerns their excellent signal-to-noise ratio. If the laser is shot-noise-limited at the chosen modulation frequency, theory offers promise of an ultimate sensitivity limited only by quantum noise in the signal\(^2\). A third interesting aspect of these resonances concerns their behavior under very rapid changes in the laser phase if the probe beam is intense enough to cause saturation. The carrier frequency component of the probe
beam will find its own velocity group with which a strong resonance is established, leading to a nonlinear macroscopic polarization at this frequency. This polarization cannot follow the rapid laser phase change and continues to radiate at the original optical phase. However, the sidebands, produced by modulation of the laser, carry its rapid phase shift. Heterodyne terms between the field radiated by the macroscopic polarization at the carrier frequency (with the original phase) and the now phase-shifted sidebands lead to a transient signal in the optical detector. Following a step laser phase jump of \( \theta \) at \( t = 0 \), a simple analysis leads to a transient signal of \( E_0 J_1(\alpha t/2)(\beta t S) \) exp \( -i \omega t \sin \theta \) prior to rf demodulation. \( S \) is the saturation parameter. The sign of the transient signal is determined by the sign of the laser phase shift. Thus we have a "phase memory" provided by the molecules! This physical effect is closely related to the Stark switching \(^{23} \) and frequency switching \(^{24} \) coherent optical transients. These signals due to laser phase slewing are dramatically visible as a \( \sim 30 \) fold "degradation" of the quadrature phase signal-to-noise ratio relative to the in-phase case, if the laser stabilization is deactivated and if the probe beam is strong enough to cause appreciable saturation.\(^{25} \)

The transient signals from the balanced mixer can be used for an extremely high speed phase-control loop of the dye laser anywhere within the Doppler profile. Experiments with both dye and color center lasers confirm this expectation. Interesting new effects at longer times are observed.

As a final remark we note that the necessary pure FM sidebands can be easily produced by a weakly-driven phase modulator crystal located inside the laser resonator. We used LiNbO\(_3\) inside our color center laser both as the FM modulator and also as the fast phase/frequency feedback element in a frequency control loop. It also seems feasible to detect the residual AM on the laser output for use in centering the intracavity etalon.

Figure 5 shows the resolved hfs of the P(2) line of HF near 2.6 \( \mu m \). The full line width at half height is about 50 kHz, still importantly limited by transit broadening (\( \omega_0 = 2.5 \) mm) and laser line width (6y = 25 kHz).

In this paper we have described a new technique, optical heterodyne saturation spectroscopy, which can produce narrow peaks with high signal-to-noise ratios, limited only by the fundamental fluctuation in the signal itself. The method provides a resonance of essentially ideal symmetry, uncontaminated with offsets or terms of the opposite symmetry. It thus should allow unprecedented accuracy in locking to a quantum absorber's resonance. The method also provides a fast dispersion-phase signal which may be directly used for reducing the spectral width and frequency slewing rate of the laser source.

Fig. 5. Optical heterodyne saturated absorption spectrum of P(2) line of HF. Taken with color center laser at \( \sim 2.6 \mu m \). Separation of two main hfs peaks \( \sim 200 \) kHz, line width \( \sim 50 \) kHz. Noise at left is artifact. Bold lines indicate fit limits.
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17. Polarizing beam dividers/combiners could be used with appropriate wave plates or Faraday devices to produce a more energy-efficient superposition of probe and saturating beams.
18. Note that only the probe beam has the sideband frequency offset but that both probe and saturation beam optical frequencies change with laser tuning: Thus the first-order sideband resonances are offset from $\omega^c$ by $\omega/2$. Line widths however are still given by $(\omega^2)^c/\Gamma = 1$.
20. Small symmetry departures can arise from multiplicative effects of the Doppler background.
21. We find $C/B = [1+|I_p/(I_0)|]^{1/2}/[1+(\Omega/J_0)]$ where $I_p$ is the probe beam intensity and $I_0$ is the saturation intensity.
25. The rf phase can be precisely set for the in-phase condition by tuning for a minimum of this noise from the balanced mixer. A calibrated phase change of 90° then produces accurate tuning for the dispersion-phase signal.

High Precision Laser Interferometry for Detection of Gravitational Radiation

K. Maischberger, A. Rüdiger, R. Schilling, L. Schnupp, W. Winkler, and H. Billinger
Max-Planck-Institut für Astrophysik, Karl-Schwarzschildstraße 1

1. Introduction

The "First Generation" gravitational wave experiments, carried out in the late 1970's, were not able to confirm definitely the existence of gravitational radiation. The strain sensitivity of these detectors was limited to $5x10^{-18}$ m/m. Later studies by astrophysicists suggest that a sensitivity about four orders of magnitude higher would be required for a realistic chance to detect gravitational waves [1].

To achieve this goal, the "Second Generation" experiments follow three key strategies: use of seismic isolators, reduction of background noise to the lowest possible level, and the construction of interferometers, which will be the focus of this paper.

Two properties of gravitational radiation are favourable for a laser interferometer:
(a) Gravitational radiation causes free test masses to exhibit displacements $\delta z$ which are proportional to their distance $z$. In an interferometer the distance $z$ between the mirrors, acting as test masses, can be made very large.
(b) Expected sources of gravitational radiation to be detectable on earth are catastrophic events in the universe (e.g. collapsing stars). The events are expected to have a typical duration of a few milliseconds. Thus, we need an interferometer to have low noise in the frequency range from, say, 1 Hz to a few kHz, only. This demand differs favourably from many other potential laser applications in which long term stability is of importance.

Interferometric techniques for the detection of gravitational radiation - all based on a Michelson configuration - are being investigated in a number of laboratories. In 1971 the first prototype detector was put in operating at Hughes Laboratories [2]. Pioneering work was also done at MIT [3]. Since then activities were started by groups in Munich [4, 5] and Glasgow [6], and recently at Caltech.

2. The Michelson Interferometer

2.1 Principle and Fundamental Limits

The principle of the Michelson interferometer is shown in Fig.1a: the input laser beam is split up into two beams which after reflection by the mirrors...