Frequency-modulation spectroscopy: a new method for measuring weak absorptions and dispersions

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Received September 17, 1979

A new type of wavelength-modulation laser spectroscopy is accomplished by utilizing an external phase modulator driven at radio frequencies large compared to the width of the spectral feature of interest. The spectral feature is probed by a single frequency-modulated (FM) sideband, and the associated absorption and dispersion are measured by monitoring the resulting radio-frequency beat signal. Experimental results are presented for the measurement of Fabry–Perot resonances, I_2 vapor absorption lines, and saturation holes in Na vapor.

Recently, Tang and co-workers demonstrated the increased spectral resolution and experimental convenience that can be achieved by using rapidly tunable laser sources for wavelength-modulation spectroscopy.[1,2] In this Letter, a new and different method for wavelength-modulation laser spectroscopy is proposed and demonstrated. This method utilizes an external phase modulator to produce the wavelength modulation and is capable of detecting weak absorption or dispersion features with the full spectral resolution characteristic of cw dye lasers. The modulator converts the single-axial-mode dye-laser input into a pure frequency-modulated (FM) optical spectrum with a low modulation index. The key concept is that the modulator is driven at radio frequencies that are large compared to the width of the spectral feature of interest. As a result, the FM sidebands are widely separated and the spectral feature of interest can be probed by a single isolated sideband. Both the absorption and the dispersion associated with the spectral feature can be separately measured by monitoring the phase and amplitude of the rf heterodyne beat signal that occurs when the FM spectrum is distored by the effects of the spectral feature on the probing sideband. Since single-mode dye lasers have little noise at radio frequencies, these beat signals can be detected with a high degree of sensitivity. Furthermore, the entire line shape of the spectral feature can be scanned either by tuning the dye-laser input with the rf constant or by tuning the rf with the dye laser set at a fixed wavelength.

It should be noted that Harris *et al.* have previously recognized that rf beats provide a sensitive probe of FM distortion and utilized this effect to stabilize the center frequency of an FM laser with respect to the center of the laser gain profile.[3] In their experiments, the separation of the FM sidebands was small compared with the width of the gain profile, and the gain and dispersion could not be separately measured. Pulse-shaping effects have previously been observed when the outputs of frequency-chirped pulsed lasers[4] or FM cw lasers[5] were passed through optical elements exhibiting group velocity dispersion. It should also be noted that Brewer and others have utilized heterodyne techniques to detect optical free-induction decay signals in coherent optical transient spectroscopy.[6] Eesley *et al.*[7] and later Owyoung[8] have utilized heterodyne detection to measure separately the real and imaginary parts of the third-order nonlinear susceptibility. However, to the author's knowledge, the FM spectroscopy technique described in this Letter is the first to make possible heterodyne detection of the real and imaginary parts of the first-order linear susceptibility.

Figure 1 is a schematic of a typical experimental arrangement for FM spectroscopy. A single-frequency laser provides radiation at the optical carrier frequency, ω_c, and the electric field, $E_1(t)$, is given by $E_1(t) = \frac{1}{2}\tilde{E}_1(t) + \text{c.c.}$, where $\tilde{E}_1(t) = E_0 \exp(i\omega_c t)$. The beam is passed through a phase modulator driven by a sinusoidally varying rf field at frequency ω_m. The modulated beam has a pure FM spectrum described by

$$E_2(t) = \frac{E_0}{2} \sum_{n=-\infty}^{\infty} J_n(M) \exp[i(\omega_c + n\omega_m)t] + \text{c.c.}, \tag{1}$$

where M is the modulation index and the J_n are Bessel functions of order n. Throughout this Letter, it will be assumed that $M \ll 1$ so $J_0(m) \cong 1$, $J_{\pm 1}M = \pm M/2$, and all other terms vanish. Thus the FM beam is described by a strong carrier at frequency ω_c with two weak sidebands at frequencies $\omega_c \pm \omega_m$.

The beam is next passed through a sample of length L whose intensity-absorption coefficient, α, and index

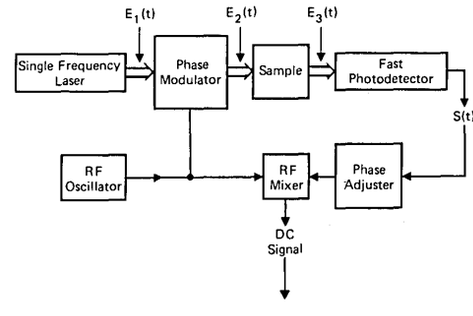

Fig. 1. Typical experimental arrangement for FM spectroscopy.

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of refraction, η, are functions of optical frequency. It is convenient to define the quantities $T_n = \exp(-\delta_n - i\phi_n)$, $\delta_n = \alpha_n L/2$, and $\phi_n = \eta_n L(\omega_c + n\omega_m)/c$, where $n = -1,0,1$ denotes the values at $\omega_c - \omega_m$, ω_c, and $\omega_c + \omega_m$, respectively. Thus δ_n describes the amplitude attenuation and ϕ_n describes the optical phase shift experienced by each frequency component. The transmitted field is $E_3(t) = \frac{1}{2}\tilde{E}_3(t) + \text{c.c.}$, with

$$\tilde{E}_3(t) = E_0\left[T_0 e^{i\omega_c t} + T_1\frac{M}{2}e^{i(\omega_c+\omega_m)t} - T_{-1}\frac{M}{2}e^{i(\omega_c-\omega_m)t}\right]. \quad (2)$$

The slowly varying intensity envelope, $I_3(t)$, of the beam impinging upon the photodetector is given by $I_3(t) = c|\tilde{E}_3(t)|^2/8\pi$. Dropping terms of the order of M^2, the general result, valid for all values of δ_n and ϕ_n, is

$$I_3(t) = \frac{cE_0^2}{8\pi}e^{-2\delta_0}\{1 + [e^{\delta_0-\delta_1}\cos(\phi_1-\phi_0) - e^{\delta_0-\delta_{-1}}\cos(\phi_0-\phi_{-1})]M\cos\omega_m t + [e^{\delta_0-\delta_1}\sin(\phi_1-\phi_0) - e^{\delta_0-\delta_{-1}}\sin(\phi_0-\phi_{-1})]M\sin\omega_m t\}. \quad (3)$$

The photodetector electrical signal $S(t)$ is proportional to $I_3(t)$ and thus will contain a beat signal at the rf modulation frequency if all the ϕ_n are not equal or if all the δ_n are not equal.

If $|\delta_0 - \delta_1|$, $|\delta_0 - \delta_{-1}|$, $|\phi_0 - \phi_1|$, and $|\phi_0 - \phi_{-1}|$ are all small compared with 1, Eq. (3) simplifies to

$$I_3(t) = \frac{cE_0^2}{8\pi}e^{-2\delta_0}[1 + (\delta_{-1}-\delta_1)M\cos\omega_m t + (\phi_1+\phi_{-1}-2\phi_0)M\sin\omega_m t], \quad (4)$$

and the in-phase ($\cos\omega_m t$) component of the beat signal is proportional to the difference in loss experienced by the upper and lower sidebands, whereas the quadrature ($\sin\omega_m t$) component is proportional to the difference between the phase shift experienced by the carrier and the average of the phase shifts experienced by the sidebands. If it is further assumed that ω_m is small compared with the width of the spectral feature of interest, the in-phase component becomes proportional to the derivative of the absorption, whereas the quadrature component becomes very small and is proportional to the second derivative of the dispersion. These conditions correspond to those of conventional wavelength-modulation spectroscopy.[9]

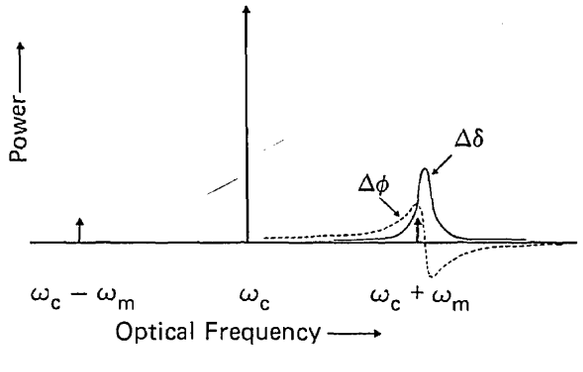

Fig. 2. Frequency-domain illustration of FM spectroscopy.

The FM spectroscopy condition is achieved when ω_m is large compared with the spectral feature of interest and only one sideband probes the spectral feature. Figure 2 shows the case of a Lorentzian spectral feature probed by the upper FM sideband. This sideband can be scanned through the spectral feature either by tuning the input laser frequency ω_c or by tuning the radio frequency ω_m. In either case, the losses and phase shifts experienced by the carrier and lower sideband remain essentially constant. Thus $\delta_{-1} = \delta_0 = \bar{\delta}$ and $\phi_{-1} = \phi_0 = \bar{\phi}$, where $\bar{\delta}$ and $\bar{\phi}$ are the constant background loss and phase shift, respectively. If the quantities $\Delta\delta = \delta_1 - \bar{\delta}$ and $\Delta\phi = \phi_1 - \bar{\phi}$ are defined to express the deviations from the background values caused by the spectral feature, then Eq. (4) further simplifies to

$$I_3(t) = \frac{cE_0^2}{8\pi}e^{-2\bar{\delta}}[1 - \Delta\delta M\cos\omega_m t + \Delta\phi M\sin\omega_m t]. \quad (5)$$

Thus the in-phase component of the beat signal is now directly proportional to the absorption induced by the spectral feature, whereas the quadrature component is directly proportional to the dispersion induced by the spectral feature. Furthermore, if M is known, the absolute values of $\Delta\delta$ and $\Delta\phi$ can be determined by comparison of the ac and dc portions of $I_3(t)$.

The rf beat signal arises from a heterodyning of the FM sidebands with the carrier frequency, and thus the signal strength is proportional to the geometrical mean of the intensity of each sideband and the carrier. The null signal that results with pure FM light can be thought of as arising from a perfect cancellation of the rf signal arising from the upper sideband beating against the carrier with the rf signal from the lower sideband beating against the carrier. The high sensitivity to the phase or amplitude changes experienced by one of the sidebands results from the disturbance of this perfect cancellation.

Experiments using the setup shown in Fig. 1 have been conducted to demonstrate FM spectroscopy for a variety of samples. A Spectra-Physics 380 ring dye laser provided the tunable single-frequency-input radiation at wavelengths between 5700 and 6300 Å. The modulator was a single-crystal $LiTaO_3$ Lasermetrics Model 1097 phase modulator terminated with 50 Ω and driven by approximately 20 mW of rf power at frequencies between 50 and 1500 MHz from a Hewlett Packard Model 8690B sweep oscillator. An MRD 510 photodiode, reverse biased by 60 V, was utilized as the photodetector. The rf beat electrical signals were detected either directly with a Hewlett Packard 851 Spectrum Analyzer or by homodyne detection using a Mini-Circuits ZFM-4 double-balanced mixer. The spectrum analyzer was not phase sensitive and thus detected the square root of the sum of the squares of the in-phase and quadrature rf signals. Phase-sensitive detection was accomplished by using the double-balanced mixer, which produced a dc signal proportional to the amplitude of that portion of the rf signal that was in phase with the local-oscillator rf input. The phase

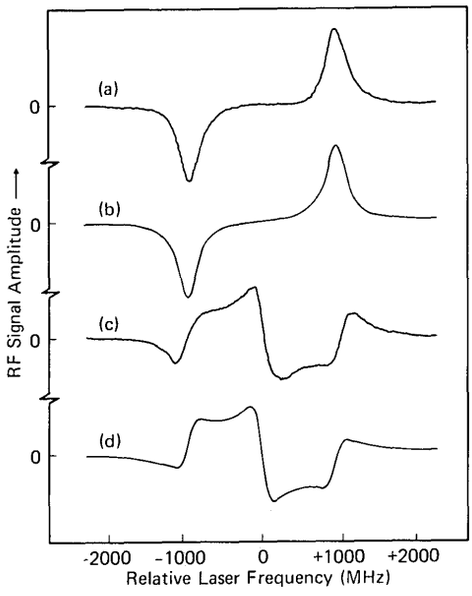

Fig. 3. Experimental and theoretical line shapes for FM spectroscopy of a Fabry–Perot resonance: (a) experimental in-phase signal, (b) theoretical in-phase signal, (c) experimental quadrature signal, (d) theoretical quadrature signal.

adjustment was accomplished by varying the cable lengths.

A Fabry–Perot resonator used in the reflection mode was employed to test the line-shape theory in the FM spectroscopy limit. The resonator had a 5-GHz free spectral range and a finesse of 16.8. The attenuation and phase shift experienced by the reflected beam when the laser was scanned through a resonance closely approximated the behavior of a Lorentzian absorption line with a full width at half-maximum of 298 MHz. Figures 3(a) and 3(c) show, respectively, the experimental in-phase and quadrature signals obtained when the sidebands were scanned through the resonance by tuning the input-laser carrier frequency, ω_c, with the rf modulation frequency, ω_m, kept at a constant value of 925 MHz. As the laser is scanned toward increasing frequency, the in-phase data clearly show a negative-going signal, which reproduces the absorption line shape probed by the upper FM sideband, followed at a spacing of $2\omega_m$ by a positive-going signal, which reproduces the same absorption line shape probed by the lower FM sideband. The quadrature data show three overlapping dispersion curves, which occur when the upper sideband, the carrier, and then the lower sideband sweep through the resonance. Figures 3(b) and 3(d) show, respectively, the theoretical line shapes for the in-phase and quadrature signals, calculated by using Eq. (5). The agreement is quite good. In addition, similar results were obtained for 30-MHz-wide saturation holes in Na vapor, in which, by utilizing the techniques of polarization-selective optical heterodyne detection,[10] the optical transmission versus laser wavelength could be made to mimic the profile of a weak, isolated absorption line with a line shape characteristic of the hole. A series of experiments in I_2 vapor was performed to determine the limit of sensitivity to small absorptions. The I_2 cell was 9.5 cm long and at room temperature provided a rich structure of absorption lines, the stronger of which had approximately 25% absorption and had widths in the 1–2-GHz range. The experiments were performed with ω_m in the 200–1000-MHz range, and thus conditions were intermediate between the wavelength modulation and FM spectroscopy limits. For each I_2 line, the laser was tuned to that relative position that produced the maximum rf beat signal. Typically, for lines with 25% peak absorption, and with ω_m = 440 MHz, an rf beat signal with rms voltage of 1.4×10^{-3} V was measured by using the spectrum analyzer. When the I_2 vapor was frozen out of the cell, the rf signal level was reduced by a factor of 400 to a residual level of 3.4×10^{-7} V. Thus an absorption line with 25% divided by 400 or 0.06% peak absorption would produce a signal as big as the residual background. However, since this background signal was essentially constant as the laser was scanned, absorptions as small as 0.005% could be detected. It should be possible to enhance this sensitivity further by utilizing a dual-beam method to compensate the signal for fluctuations in the background signal.

In conclusion, FM spectroscopy has been demonstrated to be capable of measuring both the absorption and dispersion associated with weak spectral features. Since the method utilizes an external phase modulator, and since modulation frequencies up to 30 GHz are within the state of the art,[11] it should be possible to achieve 2 cm^{-1} of spectral coverage with any fixed-frequency laser source. By trading off sensitivity and utilizing wideband electronics to detect the rf beat signals, FM spectroscopy should be capable of monitoring time-varying absorption changes with a temporal resolution in the nanosecond regime.

The author wishes to thank R. G. Brewer, R. G. Devoe, S. E. Harris, J. P. Heritage, A. C. Luntz, M. D. Levenson, and R. V. Pole for helpful discussions. The author wishes especially to thank M. D. Levenson for help in conducting several of the experiments.

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