# Optical heterodyne spectroscopy with frequency- and amplitude-modulated semiconductor lasers

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Optical heterodyne spectroscopy has been performed with GaAlAs diode lasers by modulating the injection current at frequencies up to 2.6 GHz. This extremely convenient modulation scheme results in simultaneous frequency and amplitude modulation of the laser output. When the modulated laser output probes a narrow absorption line, a characteristic heterodyne beat signal occurs. We show that the observed spectra are similar to those obtained in pure frequency-modulation spectroscopy. The technique offers high sensitivity and fast detection speed and is applied to the detection of water-vapor absorption lines.

Optical heterodyne spectroscopy with frequency-modulated laser light is a powerful and versatile detection scheme in high-resolution laser spectroscopy. In early examples of this measurement technique, relatively low modulation frequencies were used and the observed spectra had a derivative-type line shape. Bjorklund[1] recently employed high modulation frequencies that exceeded the spectral width of a probed absorption line. This technique has opened up new applications for optical heterodyne spectroscopy.[2–6] The use of widely separated frequency-modulation (FM) side bands results in a large differential absorption for the FM light and consequently in a strong heterodyne beat signal that can be detected rapidly with high sensitivity.[2] Previously, narrow-band dye lasers were used for high-FM spectroscopy, and the requisite FM was produced by an external electro-optical phase modulator.

In this Letter we introduce a modified FM measurement technique in which the FM light spectrum is created by directly modulating the injection current of a single-mode diode laser at frequencies up to several gigahertz. This modulation method[7] is straightforward and convenient, but the desired FM is accompanied by an additional modulation of the laser output power. However, the results presented in this Letter demonstrate that frequency- and amplitude-modulated semiconductor lasers can be useful light sources for heterodyne spectroscopy at high modulation frequencies. The technique was applied to study Fabry–Perot resonances and water-vapor absorption lines. The observed spectra are analyzed with a line-shape theory that we have derived for the general case of heterodyne measurements with frequency- and amplitude-modulated laser light.

It should be pointed out that derivative spectroscopy with current-modulated semiconductor lasers has been performed in the past.[8,9] The use of modulation frequencies in the gigahertz regime generally offers better sensitivity and much higher detection speed. Single-mode GaAlAs diode lasers have little frequency and amplitude noise at radio frequencies, and the ultimate limit to the achievable detection speed is set by the radio frequency since the formation of the heterodyne beat signal requires several rf cycles to occur.

The experimental setup used for the heterodyne measurements is schematically illustrated in Fig. 1. A single-mode GaAlAs laser diode (Mitsubishi ML-5308) was used as the source; the laser diode was mounted onto a metal heat sink, which was put into a Plexiglas housing to reduce frequency and amplitude fluctuations resulting from temperature variations of the environment. Slow temperature drifts did not affect the experiments since all spectra were recorded on a millisecond time scale. The diode drive current consisted of three parts. A dc bias current from a stabilized power supply was combined with a current ramp that was controlled by the sawtooth output of an oscilloscope. This permitted continuous tuning of the laser frequency over more than 30 GHz without a mode hop. A small rf current provided by a rf synthesizer was capacitively coupled into the laser diode. A beam splitter was used to direct part of the laser power to a scanning Fabry–Perot interferometer or a spectrometer

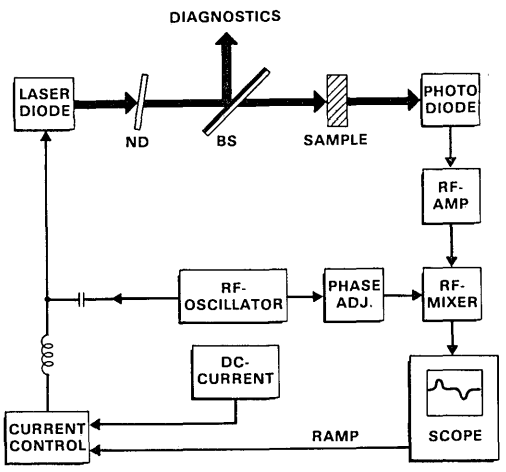


Fig. 1. Experimental arrangement for heterodyne spectroscopy with diode lasers.

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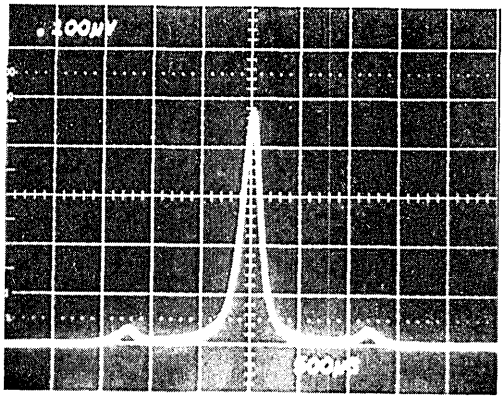


Fig. 2. Power spectrum of diode-laser output with the injection current modulated at 2.6 GHz.

for control purposes. The laser was typically operated at an output power of 5 mW, and the linewidth was 60 MHz.

The frequency- and amplitude-modulated laser output was passed through the sample and impinged onto an avalanche photodiode of at least 3-GHz bandwidth. A variable-length air line permitted changing the phase of the local oscillator signal relative to the photodiode signal. The dc output of the mixer was directly displayed with an oscilloscope that was synchronized with the laser-frequency scan.

Figure 2(a) shows a Fabry–Perot scan of the laser output when the drive current was modulated at $\nu_m$ = 2.6 GHz, the frequency limit of the rf synthesizer. The observed power spectrum consisted primarily of a strong carrier at $\omega_0$ and two sidebands at $\omega_0 \pm \omega_m$ that have equal amplitude. In case of simultaneous frequency and amplitude modulation the optical field is given by

$$E(t) = E_0[1 + M\sin(\omega_m t + \psi)] \times \exp[i(\omega_0 t + \beta \sin \omega_m t)], \quad (1)$$

where $\omega_0$ is the carrier and $\omega_m$ is the modulation frequency. $M$ is the amplitude-modulation (AM) index, $\beta$ is the FM index, and $\psi$ defines the phase difference between AM and FM. The instantaneous optical frequency is $\omega(t) = \omega_0 + \beta\omega_m \cos \omega_m t$. For small modulation index the radiation field is described by a strong carrier at $\omega_0$ and two weak sidebands at $\omega_0 \pm \omega_m$. By expanding Eq. (1) the intensities at these frequencies can be calculated:

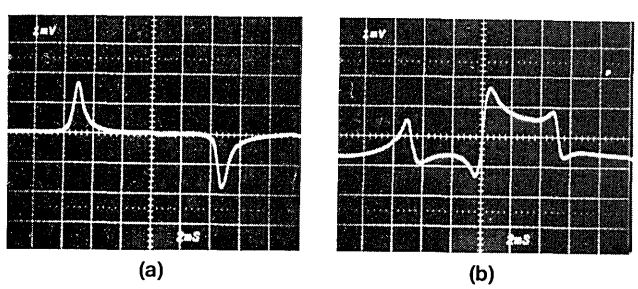
$$E^2(\omega_0) = J_0^2(\beta) + M^2 J_1^2(\beta)\cos^2\psi,$$

$$E^2(\omega_0 \pm \omega_m) = J_1^2(\beta) + \frac{M^2}{4}\{[J_0^2(\beta) + J_2^2(\beta)]\} - \frac{M^2}{2}J_0(\beta)J_2(\beta)\cos 2\psi \pm M[J_0(\beta) + J_2(\beta)]\sin\psi. \quad (2)$$

Here $J_n(\beta)$ are the $n$th-order Bessel functions. The observed symmetrical power spectrum [Fig. 2] means that the phase difference between FM and AM is either $\psi = 0$ or $\psi = \pi$. The phase difference $\psi$ depends critically on the laser's operating conditions and was found to be $\psi \approx \pi/2$ for modulation frequencies below 1.5 GHz.[10] In contrast to laser radiation, which is frequency modulated by means of an external electro-optic phase modulator, the intensity of the drive-current-modulated diode laser is modulated at $\omega_m$, i.e., $E^2(t) = E_0^2(1 + 2M \sin \omega_m t)$.

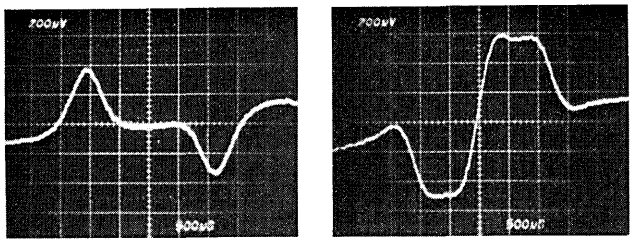
FM spectroscopy is based on the heterodyne detection of the change in the transmitted radiation power spectrum that occurs when the frequency-modulated light probes an absorption line. A beat-frequency signal at the modulation frequency $\omega_m$ occurs if there is a difference in the absorption of the two sidebands at $\omega_0 \pm \omega_m$ if the index of refraction for the carrier changes relative to the average of that for the sidebands.[1] In the ideal case of purely frequency-modulated light ($M = 0$), the effect of the probed absorption line converts FM into AM and is thus essential for any rf signal to occur. If the laser light used is simultaneously frequency and amplitude modulated, a rf signal can be detected even when no sample is present.

To test the possibility of performing heterodyne spectroscopy by modulating the drive current in a GaAlAs laser at high frequency, we used a Fabry–Perot resonator in the reflection mode to simulate an absorption line. The Fabry–Perot resonator had a free spectral range of 21 GHz, and its resonance line had a FWHM of 480 MHz. We modulated the laser-diode drive current at $\nu_m$ = 2.6 GHz and measured the heterodyne beat signal that arises when the carrier frequency is tuned through the vicinity of the resonance. Adjusting the phase of the phase-sensitive detection electronics (see Fig. 1) permitted recording either the in-phase or the quadrature component of the beat signal (Fig. 3). The spectra resemble those in Refs. 1–4, in which an absorption line is probed with a pure FM light spectrum produced with an external phase modulator; however, a few differences are noteworthy. The in-phase component of the beat signal consists of two peaks, which occur when the upper and lower sidebands sweep through the resonance; the two spectral structures have a slightly asymmetrical, non-Lorentzian line shape with a tail on the right-hand side. The quadrature component of the signal has a nonvanishing background, and all the positive-going peaks are stronger than the corresponding negative-going peaks. In order to understand this line-shape behavior we have made a complete line-shape analysis for heterodyne spectroscopy with frequency- and amplitude-modulated light. The general theory together with further experimental data will be presented elsewhere, and we



(a) (b)

Fig. 3. Heterodyne-spectroscopy signals of Fabry–Perot resonance: (a) in-phase component, (b) quadrature component.



(a) (b)

Fig. 4. Heterodyne spectra of water-vapor absorption line at $\lambda = 816.7$ nm: (a) in-phase component, (b) quadrature component.

discuss here only the final result obtained under simplifying assumptions that are justified for the experimental conditions of Fig. 3. Starting with Eq. (1) and assuming small signal modulation ($M, \beta \ll 1$), the light intensity impinging upon the photodetector is

$$I(t) \sim e^{2\delta_0}\{1 + [\beta(\delta_{-1} - \delta_1) + M(\phi_{-1} - \phi_1)] \cos \omega_m t + [\beta(-2\phi_0 + \phi_{-1} + \phi_1) + M(2 + 2\delta_0 - \delta_{-1} - \delta_1)]\sin \omega_m t\}, \quad (3)$$

where $\delta_0$ and $\delta_{\pm 1}$ are the amplitude-absorption losses at the frequencies $\omega_0$ and $\omega_{\pm 1} = \omega_0 \pm \omega_m$, respectively, and $\phi_{0,\pm 1}$ are the corresponding phase shifts induced by the sample. A phase difference of $\psi = 0$ between the FM and AM of the laser has been assumed, and the absorption losses and the phase shifts experienced by the two sidebands and the carrier are assumed to be small, i.e., $|\delta_{-1} - \delta_1| \ll 1$ and $|\phi_0 - \phi_{\pm 1}| \ll 1$. For $M = 0$, expression (3) simplifies to Eq. (4) in Ref. 1, which describes the case of pure FM. Expression (3) completely explains the observed line shapes of the spectra in Fig. 3. The observed deviations form pure FM-spectroscopy spectra result from contributions of absorption and dispersion effects to both the in-phase and the quadrature part of the heterodyne beat signal. The line-shape theory predicts correctly a zero background of the in-phase term of the signal, whereas the quadrature term exhibits a nonvanishing background. If $M$ and $\beta$ are known, an analysis of the measured line shapes and a calibration of the signal strength using a spectral structure of known absorption and dispersion permits the absolute determination of $\delta(\omega)$ and $\phi(\omega)$ of the sample under investigation. The overall effect of the AM on the observed spectra is rather small since for the current-modulated diode laser used we found that $\beta/M \approx 20$.

Optical heterodyne measurements with current-modulated diode lasers can be applied to high-resolution laser spectroscopy. We have successfully used the technique to measure water-vapor absorption lines between 816 and 818 nm. An absorption cell of 1-m length containing pure water vapor (18 Torr) was used at room temperature in a triple-pass configuration, resulting in approximately 5% absorption of a water-vapor line in the 820-nm region. Figure 4 shows oscilloscope traces of the in-phase and the quadrature component of the heterodyne beat signal with the diode-laser drive current modulated at $\nu_m = 2.6$ GHz. Temperature tuning with a Peltier cooler was used to tune the diode-laser wavelength to the vicinity of the water-vapor absorption line. The dc bias current was ramped (see Fig. 1) in order to sweep through the 1.5-GHz-wide resonance. The background slope of the signal in Fig. 4 results from a periodic wavelength dependence of the laser intensity impinging upon the photodetector and is caused by étalon effects in the parallel-absorption cell windows, which were neither wedged nor antireflection coated. Note that the spectra shown are direct displays of the output of the double-balanced mixer and that they are recorded on a millisecond time scale with no additional signal averaging. The quadrature component of the beat signal [Fig. 4(b)] can be used to lock the carrier frequency to the center of a water-vapor absorption line by using a technique that has been employed to frequency stabilize ring dye lasers with accuracies of better than 1 kHz.[4]

In conclusion, we have performed heterodyne absorption spectroscopy with GaAlAs diode lasers that, through current modulation, were simultaneously frequency and amplitude modulated at frequencies up to 2.6 GHz. The technique has been tested by studying Fabry–Perot resonances and water-vapor absorption lines. The line shapes of the measured spectra deviate characteristically from those observed in pure FM spectroscopy but can be completely explained with a straightforward line-shape theory. The method can be used with any single-frequency semiconductor laser with the modulation-frequency limit set by the laser-diode characteristics. The simple and convenient modulation and tuning methods of diode lasers combined with the high sensitivity and fast detection speed of heterodyne detection makes this measurement technique interesting for applications such as laser remote probing of gaseous species, readout of frequency-domain optical memories,[5] frequency locking of semiconductor lasers, and high-resolution laser spectroscopy.

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