

## **Application Note 7**

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# **FM Spectroscopy With Tunable Diode Lasers**

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# FM Spectroscopy With Tunable Diode Lasers

## Introduction

Tunable diode lasers are ideal for optical spectroscopy because of their narrow linewidths, large tuning ranges and stable outputs. Because they are more compact and rugged than traditional spectroscopic optical sources, like Ti:Sapphire lasers, dye lasers, color-center lasers, hollow-cathode lamps, and nonlinear systems (e.g. optical parametric oscillators), they have enabled spectroscopic methods to be used not only in laboratory environments but also in the real world. Applications of diode-laser spectroscopy include remote sensing, LIDAR, laser cooling and trapping of atoms,<sup>1</sup> frequency standards,<sup>2</sup> length standards,<sup>3</sup> trace gas detection,<sup>4</sup> and process monitoring.<sup>5</sup> They can be used to monitor environmentally important species, such as methane, carbon dioxide, and water. In semiconductor manufacturing, they can be used for closed-loop control of deposition processes, including electron-beam, sputtering, molecular-beam epitaxy (MBE), and thermal evaporation, resulting in a significant increase in the yield of existing devices and making possible new and improved ones. Indeed the field of tunable-diode-laser absorption spectroscopy has grown so large that a review of the research journals reveals that it has developed its own acronym, TDLAS, which stands for Tunable Diode Laser Absorption Spectroscopy.

There is a wide variety of laser-spectroscopic techniques available to researchers.<sup>6</sup> In this application note, we will focus on one particular technique—frequency-modulation spectroscopy, or FMS. FMS is a powerful technique that can achieve a high signal-to-noise ratio with a relatively simple experimental setup. In a typical FMS experiment, the wavelength of a continuous-wave laser is modulated at a particular frequency. As the center wavelength is scanned across the atomic transition, the wavelength modulation is converted into amplitude modulation, giving rise to a modulation in the optical absorption of a sample at the same frequency. (See figure 1 for a typical absorption line.)

Narrow-band demodulation techniques, such as phase-sensitive detection using a lock-in amplifier, then allow the absorption information to be realized at DC.

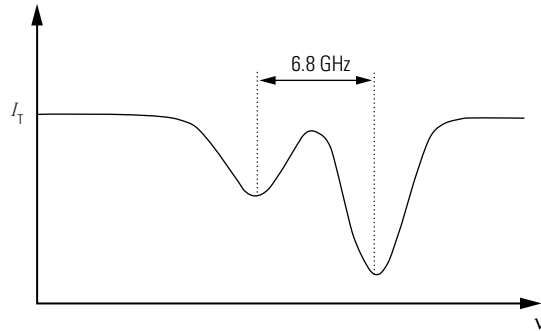


Figure 1: A typical absorption line for rubidium, showing transmitted intensity ( $I_T$ ) as a function of the laser frequency ( $\nu$ ).

Because the signal has been moved to a high frequency via modulation, FMS avoids the typical limitations of absorption measurements such as laser-intensity fluctuations, which peak at DC and fall off roughly as  $1/f$ , hence the name  $1/f$  noise. Using this technique, absorption sensitivities can reach the part per million (ppm) level. For example,  $H_2S$  has been detected at the ppm level in air;<sup>7</sup> absorption of Y has been measured at the ppm level,<sup>5</sup> and methane has been detected at the level of 12 ppb  $m/\sqrt{Hz}$ .<sup>8</sup>

As we will discuss later in this application note, further enhancements of the signal can be achieved by taking advantage of experimental geometries that cancel particular noise sources or undesirable aspects of the signal. Finally, if a reasonable level of attention is given to the optics and electronics, systematic errors can be suppressed and high accuracy can be achieved with FMS.<sup>9</sup> This is important, for instance, when trying to determine the exact center of an absorption line. For many applications the center must be found to better than 0.1% of the linewidth.

FMS is particularly well suited to diode lasers such as the New Focus Vortex™ Series and Velocity® Series external-cavity diode lasers because of their simple, yet powerful, modulation capabilities. The main methods for achieving wavelength modulation (shown with the maximum modulation rates for the Vortex) include mechanically “dithering” the mirror (3.5 kHz), modulating the injection current into the diode (100 MHz), and using external phase modulators (10 GHz).

FMS can be broken down into two regimes: wavelength modulation (WM) and frequency modulation (FM). In the case of WM, the modulation depth is very large, generating a large number of sidebands, but the modulation frequency is low ( $<1$  MHz); in the case of FM, the modulation depth is small but the frequency is very high ( $>100$  MHz). We will discuss FM in this application note, but the physics of WM and FM are nearly identical.<sup>10</sup> The methods discussed here have been used for atomic spectroscopy of Cs, Rb, K, Li, Ne, Pb, Tl, Y, Sr, and Ca. In addition, there are also numerous examples of diode lasers being used in molecular spectroscopy utilizing  $O_2$ ,  $I_2$ ,  $CO_2$ ,  $NO_2$ ,  $H_2O$ ,  $H_2S$ ,  $CH_3$ ,  $CH_4$ ,  $C_2H_2$ ,  $CH_3CH_2OH$ , and others. Many applications, such as interferometry and frequency stabilization of lasers,<sup>11</sup> benefit from virtually identical techniques. The narrow band transmission of a Fabry-Perot interferometer is quite similar to a narrow absorption line<sup>12</sup> (figure 2), so much of the discussion in this applications note is directly relevant to such a setup.

This application note is broken into two main parts. The first part explains essential elements of the physics behind FMS and certain details about the signals that are obtained. Then a simple FMS setup utilizing intensity noise cancellation is described and analyzed. The second part concerns itself with FM “saturation” spectroscopy. Saturation spectroscopy uses a setup similar to the first part, but gives much narrower signals that are free from Doppler broadening (typically the dominant source of line broadening). A short discussion of the physics of saturation is included. This note concludes with a reality check—a brief discussion of the most common experimental difficulties encountered with FMS.

## The Physics of FM Spectroscopy

Figure 2 shows a laser beam transmitted through a gas cell containing a resonantly absorbing atomic or molecular vapor and a laser beam transmitted through a Fabry-Perot interferometer. For the interferometer, the transmitted intensity (measured with a photodiode) has a narrow, peaked response centered around some frequency (or wavelength), while the absorption will display a narrow dip in the response around the central absorption frequency. Either situation could be analyzed here, but for the sake of clarity let’s look in detail at the absorption.

The central aspect of FMS is the modulation of the laser frequency and its effect on the intensity of light transmitted by the gas cell. Consider the situation where the frequency at which the laser is modulated is fairly low and the amplitude of that modulation is small. In this case the central laser frequency can be thought of as periodically increasing and decreasing by a small amount. Such a situation is shown in figure 3a where the laser linewidth is assumed to be much smaller than the width of the absorption. If the laser frequency is in the vicinity of an absorption line then the frequency modulation causes the absorption to modulate synchronously. In this way, the laser frequency modulation is mapped onto the laser’s transmitted intensity. Another way to state this is that the frequency modulation on the laser has been converted into an amplitude modulation by the absorption: FM has become AM. This change allows the photodiode to detect the modulation, since the photodiode cannot detect frequency changes. Figure 3a also demonstrates the phase relationship between the laser’s FM and the absorption’s AM. A final point concerning figure 3a is

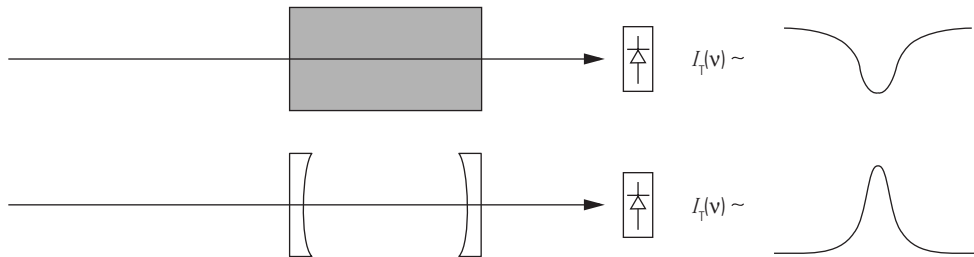


Figure 2: The intensity versus frequency of a laser beam transmitted through a gas cell containing an atomic or molecular vapor (top) and of a laser beam transmitted through a Fabry-Perot interferometer (bottom).

that the conversion of FM to AM at a particular frequency depends on the slope (or derivative) of the absorption at that frequency. This relationship between FMS and the slope will be formalized below.

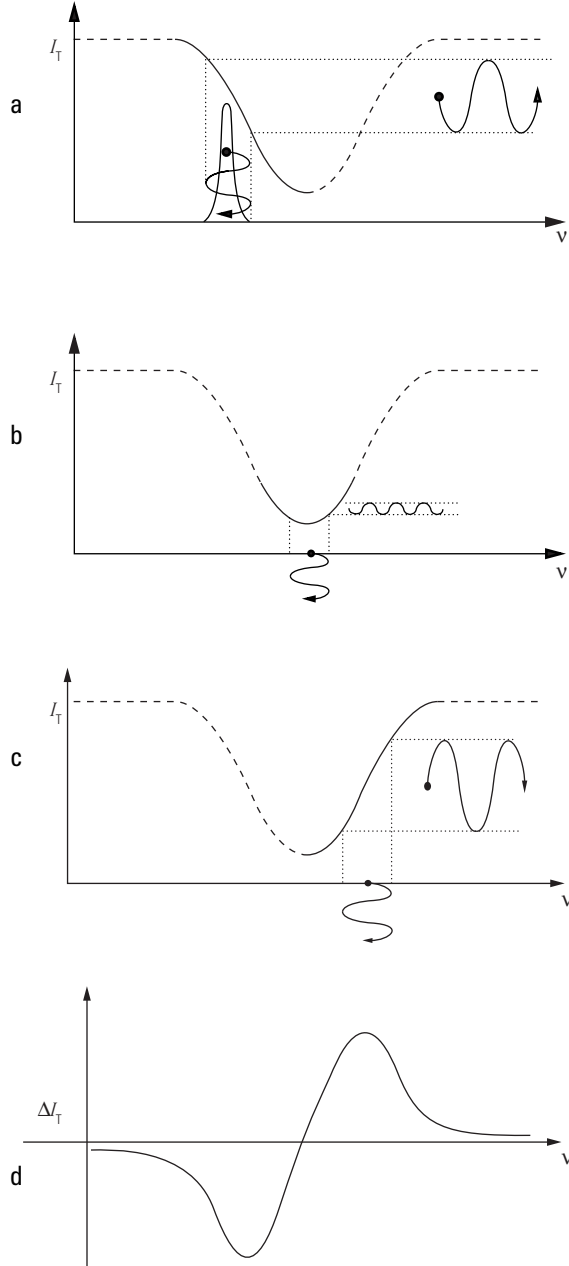


Figure 3: In frequency modulation spectroscopy, as the wavelength is scanned across the atomic transition, the wavelength modulation is converted into amplitude modulation, giving rise to a modulation in the optical absorption of a sample at the same frequency. As you continue to scan across the absorption profile, as is done in figures 3b and 3c, you can see that the amount of FM to AM conversion varies. Figure 3d is a plot of the ratio of AM to FM versus the laser frequency, taking phases into account.

Qualitatively, it is useful to continue the scan across the absorption profile, as shown in figures 3b and 3c. Near the point of maximum absorption, the conversion of FM to AM is very small, and in fact goes to zero at line center. On the other side of the absorption peak it can be seen that the FM to AM conversion is large again, but the phase relationship between the FM and AM has reversed. Finally, if the scan is continued away from the absorption, the AM goes away because of the lack of absorption and near-zero slope. In this way, one could plot the ratio of AM to FM versus the laser frequency and obtain the curve shown in figure 3d, where the change in phase has been represented by a sign change. This curve looks suspiciously like the derivative of the absorption, and this is indeed the case.

If the laser frequency  $\nu$  is modulated at the frequency  $\Omega$  (sometimes called “dithering”), with modulation amplitude  $m$ , then the transmitted intensity  $I_T$  through the vapor cell can be written

$$I_T(\nu) = I_T(\nu + m \sin \Omega t)$$

Here we take  $m, \Omega < \Gamma =$  linewidth of the absorption. Now we can expand  $I_T$  as a Taylor series

$$I_T(\nu + m \sin \Omega t) = I_T(\nu) + (m \sin \Omega t) \frac{dI_T}{d\nu} + \left( \frac{m^2 \sin^2 \Omega t}{2!} \right) \frac{d^2 I_T}{d\nu^2} + \left( \frac{m^3 \sin^3 \Omega t}{3!} \right) \frac{d^3 I_T}{d\nu^3} + \dots$$

and combine terms:

$$I_T(\nu + m \sin \Omega t) = \left[ I_T(\nu) + \frac{m^2}{4} \frac{d^2 I_T}{d\nu^2} + \dots \right] + \sin \Omega t \left[ m \frac{dI_T}{d\nu} + \frac{m^3}{8} \frac{d^3 I_T}{d\nu^3} + \dots \right] + \cos 2\Omega t \left[ -\frac{m^2}{4} \frac{d^2 I_T}{d\nu^2} + \dots \right] + \dots$$

So, the transmitted intensity contains a DC term, a term oscillating at  $\Omega$ , a term oscillating at  $2\Omega$ , and so on. If phase-sensitive detection is performed at  $\Omega$ , for instance using a lock-in amplifier, the coefficient of the  $\sin \Omega t$  term can be extracted. In particular, since we have assumed that  $m$  is small, the coefficient of the  $\sin \Omega t$  term is essentially  $m$  multiplied by the first

derivative of the transmitted intensity (or absorption). In an analogous way, detection at  $2\Omega$  reveals the second derivative,  $3\Omega$  the third derivative, and so on. For this reason, FMS is sometimes called “derivative” spectroscopy.

### A Simple FM Spectroscopy Experiment

It is easy to improve the basic FMS setup described above by reducing the noise that arises from laser-intensity fluctuations—typically the largest source of noise. Basically, the experiment above is repeated, but a second laser beam is derived from the same laser which is *not* passed through the vapor cell (figure 4). The beam that passes through the cell performs the FM to AM conversion, and possesses intensity fluctuations. The second beam also has laser-intensity fluctuations but no induced AM. If the absolute intensities of the two beams after the vapor cell are balanced so they have the same power (and this procedure should be performed away from the absorption feature), and one measures the *difference* between the two photocurrents, then the laser intensity fluctuations will exactly cancel.

Such a differential measurement is easily accomplished using the New Focus Model 20X7 Nirvana auto-balanced photoreceiver. The Nirvana’s patented auto-balancing circuit uses a low-frequency feedback loop to automatically maintain DC balance between the signal and reference arms. In effect the circuit behaves as a variable-gain beamsplitter, so you won’t have to manually balance the power in the two beams. This circuit, in conjunction with the subtraction node, cancels common-mode laser noise with greater than 50-dB rejection at frequencies less than 125 kHz. Combining this auto-balancing detector with phase-sensitive detection (a lock-in amplifier) allows you to obtain the derivative of the absorption signal free from intensity noise and with other noise sources suppressed due to the modulation.

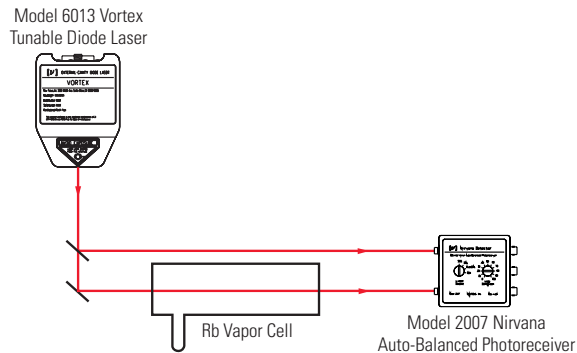


Figure 4: Setup of a typical FM spectroscopy experiment. In the setup above, a second laser beam that does not pass through the vapor cell is used to eliminate laser-intensity fluctuations. A dual balanced detector, such as the Model 20X7 Nirvana, is used to make a differential measurement.

You can perform this experiment very easily with the New Focus Vortex laser. This laser’s external-cavity design creates a narrow-linewidth output which can be scanned across by many tens of GHz across an absorption feature. A BNC input to a piezo-electric transducer allows you to modulate the laser frequency by up to 3.5 kHz. Vortex lasers are also guaranteed to be mode-hop free over their entire tuning ranges and can be built to almost any desired wavelength range covered by existing semiconductor diode lasers.

### The Physics of Saturation Spectroscopy

As in typical FM spectroscopy, FM saturation spectroscopy measures the intensity of a laser beam passed through a gas cell. The important difference is that, in saturation spectroscopy, a strong laser beam (typically at the same frequency) is sent through the vapor cell *counter-propagating* (or with a very small angle) to and overlapped with the original beam passing through the cell (figure 5). This results in significantly narrowing the widths of absorption lines, as will be shown.

Saturation spectroscopy essentially eliminates the Doppler broadening of absorption lines, which is usual-

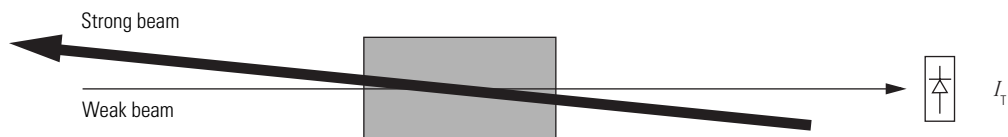


Figure 5: In an FM saturation spectroscopy experiment, a strong laser beam (typically at the same frequency) is sent through the vapor cell counter-propagating to (or with a very small angle) and overlapped with the original, weak beam passing through the cell.

ly the largest source of line broadening. For many applications, it is very important to realize a narrow absorption width near the “natural” (or quantum-limited) width so that line center can be determined with high accuracy. Once the Doppler broadening is removed, other interesting effects—such as the hyperfine structure—become resolved. It also allows the study of much smaller sources of line broadening, which will reveal more subtle physics (collisional shifts, time-of-flight broadening, magnetic-field effects, photon-recoil effects, and pressure broadening are just a few).

Doppler broadening arises because atoms or molecules are in motion. An atom with velocity  $\vec{v}$  whose absolute angular absorption frequency is  $\omega_0$  absorbs light from a laser with angular frequency  $\omega=2\pi\nu$  only if the following Doppler condition is satisfied:

$$\omega = \omega_0 + \vec{v} \cdot \vec{k} \quad \text{or} \quad v = v_0 \left(1 + \frac{V_{\parallel}}{c}\right)$$

Here the wave vector of the laser beam is

$$|\vec{k}| = \frac{2\pi}{\lambda},$$

$\lambda$  is the laser wavelength,  $V_{\parallel}$  is the component of velocity along  $\vec{k}$ , and  $c$  is the speed of light. The laser frequency does not have to be in exact resonance with the *absolute* absorption frequency in order for *moving* atoms to absorb light. Hence, a wide distribution of velocities (typically Maxwell-Boltzmann) leads to a distribution of resonance frequencies and line broadening.

So, if the laser frequency is scanned across the absorption profile, the full Doppler-broadened line-shape will be obtained. At any given laser frequency, however, the light will be absorbed by only a *subset* of the available atoms—those whose velocity along the laser beam is such that the Doppler condition is fulfilled. Such a subset of the atoms is called a “velocity group.” Figure 6a demonstrates the idea of velocity groups further by showing the number of atoms in the

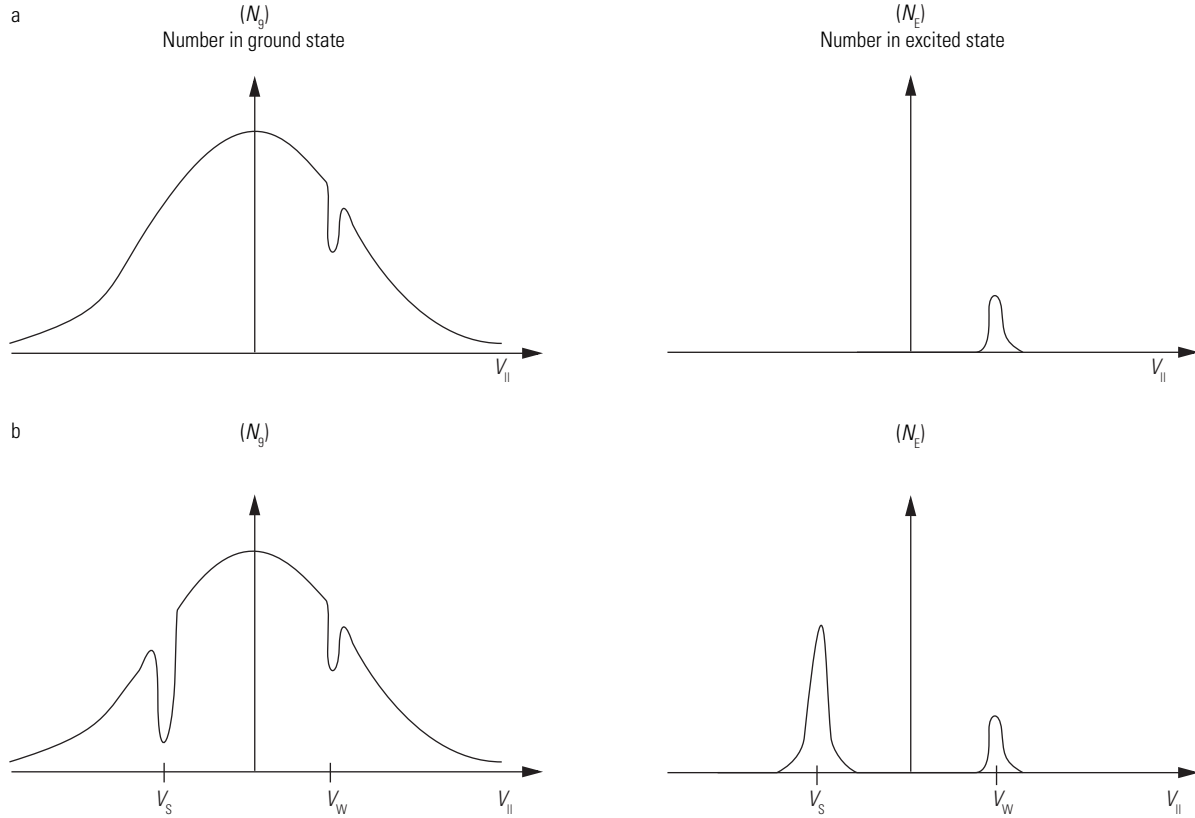


Fig. 6: a) Number of atoms in the ground state versus  $V_{\parallel}$ . For a given laser frequency, only a single, narrow velocity group interacts with the laser, which drives atoms in that velocity group to the excited state. b) Number of atoms in the ground state versus  $V_{\parallel}$  when the effect of the strong, counter-propagating beam is taken into account.

ground state versus  $V_{\parallel}$ . For a given laser frequency, only a single, narrow velocity group interacts with the laser, which drives atoms in that velocity group to the excited state. Equivalently, this causes a depletion of the number of atoms in the ground state for that velocity group (the dip in figure 6a).

If we assume the above analysis applies to the original, weak beam passing through the cell, and now consider the effect of the strong, counter-propagating beam, we get the situation shown in figure 6b. The counter-propagating beam (of the same frequency) interacts with a velocity group  $V_S$  whose Doppler shift must be opposite that of the original velocity group,  $V_W$ . At first glance, the presence of the strong beam appears to have no effect—the two laser beams interact with completely independent groups of atoms. If only the transmission of the weak beam is monitored (as in figure 5), then the signal ought to be the same as when the strong beam is not present.

This is not always true, however. Consider the situation when the laser frequency  $\nu$  is very near the exact atomic resonance frequency  $\nu_0$ . In that case, the velocity groups interacting with the laser beams have  $V_{\parallel} \approx -V_{\parallel} \approx 0$ , and both beams begin to interact with the *same* group (same atoms). The two beams will compete for the atoms, but it is not a fair fight. The strong beam quickly depletes the number of atoms in the ground state with  $V_{\parallel} \approx 0$ , leaving very few to interact with the weak beam. To the weak beam, it appears that there are no ground state atoms to interact with because the strong beam has driven them all to the excited state. Hence, when  $\nu \approx \nu_0$ ,  $V_{\parallel} \approx 0$ , then the weak beam travels through the vapor cell with *very little absorption*, even though it is at resonance. Though not a rigorous definition, this effect is known as “saturation.”

### A Typical FM Saturation Spectroscopy Setup

For the above example, the transmitted intensity for the weak beam will look something like figure 7. The absorption looks normal (as if the strong beam were absent) except near exact resonance, where the absorption is very small due to saturation from the strong beam. In the middle of the Doppler-broadened lineshape, there is now a very narrow feature around

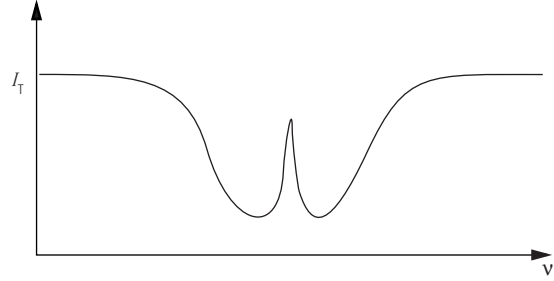


Figure 7: Transmission of the weak beam from the setup in figure 5. When the laser frequency is very near the exact atomic resonance frequency  $\nu_0$ , so that  $V_{\parallel} \approx -V_{\parallel} \approx 0$ , the absorption looks normal except near exact resonance, where the absorption is very small due to saturation from the strong beam. In the middle of the Doppler-broadened lineshape, there is now a very narrow feature around exact line center.

exact line center. At this point, FMS could be performed exactly as above, and the derivative of the narrow absorption feature could be obtained. The larger Doppler-broadened shape is still present, however, contributing its own derivative to the signal. A better scheme for a saturation-spectroscopy setup, shown in figure 8, allows the Doppler-broadened contribution to the lineshape to be completely subtracted.

The idea behind the setup in figure 8 is as follows. First, a strong, “saturating” beam has been added that is counter-propagating to and overlapped with one of the weak beams. Second, the vapor cell has been shifted so that *both* weak beams pass through the cell. The effect of these changes is summarized in figure 9. The difference between the two photocurrents has completely subtracted the Doppler-broadened lineshape,

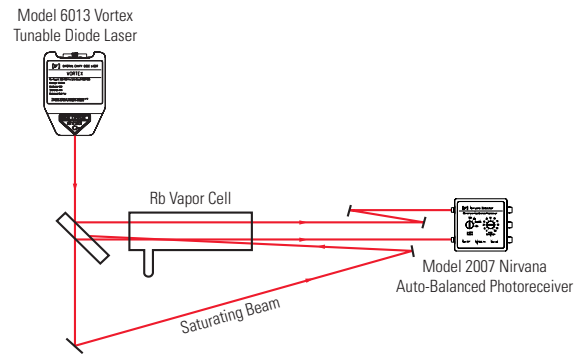


Figure 8: Typical FM saturation spectroscopy setup where a strong “saturating” beam has been added that is counter-propagating to and overlapped with one of the weak beams. The vapor cell has been shifted so that both weak beams pass through the cell. The difference between the two photocurrents has completely subtracted the Doppler-broadened lineshape and intensity noise, leaving only the narrow saturation feature.

leaving only the narrow saturation feature. In addition, if the intensities of the two weak beams are balanced, then the intensity noise of the laser will be cancelled. Such a setup is often used to conveniently resolve the hyperfine structure of atomic spectra that is normally buried beneath the Doppler broadening (figure 10).

### Difficulties with FMS

It is fairly common for the sensitivity of FMS to approach a fundamental noise limit, and in many cases (particularly for higher modulation frequencies) this noise limit is achieved. The fundamental noise source here is photon shot noise (or counting noise) on the amplitude of the laser beam used to study the absorption.<sup>13</sup> For the relatively low modulation frequencies suggested in this application note, shot noise may be the dominant noise source, but not necessarily the *only* significant noise source. In particular, diode lasers tend to have intensity fluctuations (and especially frequency fluctuations<sup>14, 15</sup>) that extend to very high frequencies, making the intensity noise cancellation technique very important. Other difficulties with FMS may hinder the sensitivity well before the fundamental shot noise limit is reached, however.

One of the most common problems with FMS is spurious AM on the laser beams,<sup>16</sup> at the same frequency as the intended FM, but *not* associated with the absorption. Such AM, when demodulated, leads to offsets on the derivative signal obtained from FMS. This can be a problem when it is desirable to have the derivative signal cross zero at exact line center. Often, however, this offset is not of great concern, and can be nulled electronically. Using a geometry with dual photodetectors will minimize this effect, but not necessarily eliminate it. A common source of unwanted AM comes from residual étalon effects, for instance from the win-

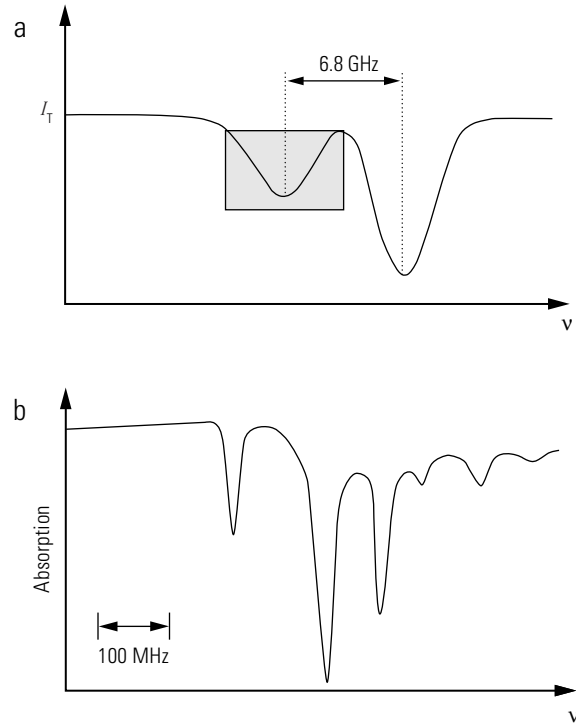


Figure 10a demonstrates the absorption measurement of Rb in a non-saturated setup. Figure 10b shows the hyperfine structure of one of the features from the Rb-absorption line, as measured using the saturated-spectroscopy setup of figure 8.

dows of the photodetector, the windows of the vapor cell, or some combination of partially reflective surfaces. AM arising from étalons tends to give sloping backgrounds (the derivative of the étalon transmission) which have a different frequency dependence than the desired absorption. The poor mechanical and thermal stability of such étalon effects leads to noisy, drifting background signals. These effects are easily searched for and eliminated.

Another source of AM is a residual étalon within the laser cavity itself. Up to this point, we have only been discussing low-frequency modulation of diode lasers, performed using a PZT that changes the length

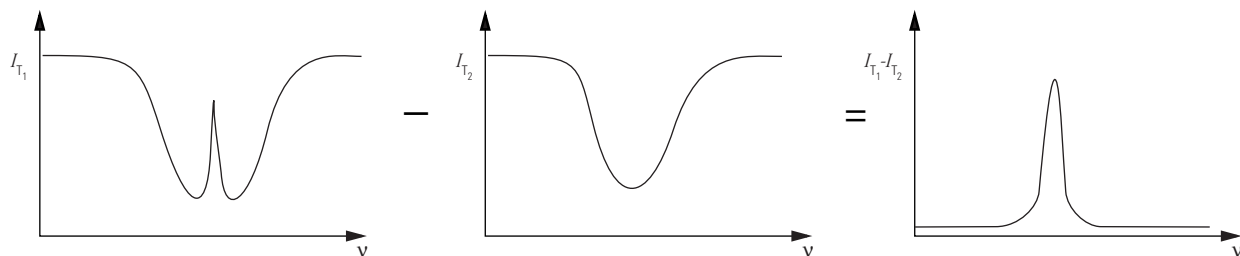


Figure 9: Transmitted intensity versus frequency of the setup shown in figure 8.

of the diode's external cavity. Theoretically this results in pure FM. However, residual étalon effects occur within the laser cavity itself between the anti-reflection coated front facet of the laser diode and the back facet of the laser diode. Residual reflection from the coated facet can result in 10% modulation in external-cavity diode lasers.

Another possible source of AM should also be mentioned here. Whereas we've only discussed low-frequency modulation using the PZT, another widely used technique is to modulate the diode-laser injection current, which also causes modulation of the laser frequency. However, this method produces AM as well as FM<sup>17,18</sup> (the ratio depends on the modulation frequency and the particular diode), leading to problems such as those mentioned above. The benefit arises because the injection current can be modulated at *very* high frequencies, up to several hundred MHz (or even GHz). In the case of the 6000 Series Vortex lasers, you can modulate the current with a bandwidth of up to 1 MHz through the current-modulation input on the driver or up to 100 MHz by applying the signal to the laser diode via a connector on the laser head. As mentioned, this AM is tolerable for many experiments. It was also recently shown that the residual AM can be reduced 50 dB by injection locking one diode laser to another.<sup>19</sup>

A final common source of problems with diode lasers *in general* is due to optical feedback from reflective surfaces throughout the optical setup. Any residual reflection that leads back into the laser will tend to form a second external cavity, and perturb the laser frequency. Like étalon effects, the poor mechanical and thermal stability of such reflections tends to add a great deal of noise to the laser frequency. By using a good isolator, you can avoid common feedback problems.

## Summary

Frequency-modulation spectroscopy is a powerful tool that can achieve high sensitivities with a relatively simple experimental setup. Tunable diode lasers, in particular, can make the setup even simpler because they provide a narrow, tunable output that can be easily modulated. As new diode laser wavelengths become available the field of TDLAS (tunable diode laser absorption spectroscopy) will continue to grow rapidly,

particularly when combined with nonlinear optical techniques that allow conversion of available wavelengths to the blue,<sup>20</sup> UV,<sup>21,22</sup> or far infrared.<sup>8</sup>

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