CHAPTER THREE

High-Accuracy Spectroscopy with Semiconductor Lasers: Application to Laser-frequency Stabilization

LEO HOLLBERG, V. L. VELICHANSKY,* C. S. WEIMER,† and R. W. FOX

United States Department of Commerce, National Institute of Standards and Technology, Boulder, Colorado

3.1 INTRODUCTION

In this chapter, we focus on the performance of diode lasers used for high-precision spectroscopy, with an eye toward optical frequency and wavelength references in the visible and near-infrared spectral region. A brief summary is given of the existing optical references and the major physical processes that limit the performance of optical

*Currently, Lebedev Institute of Physics, Moscow, Russia.
†Currently, Ophir Corp., Littleton, Colorado.

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frequency-wavelength standards. Present state-of-the-art high-stability diode laser systems and laser technology are outlined. Particular attention is given to our present results in high-resolution diode-laser spectroscopy of calcium and the prospects for the future.

For many years there has been the promise of future optical frequency standards that could augment or even replace the now traditional microwave frequency standards. In terms of reproducibility, the best proven standard is still the microwave frequency standard based on the cesium ground state hyperfine structure. The present performance of the best cesium standards is a frequency uncertainty of $(\delta\nu)/\nu \approx 1 \times 10^{-14}$. None of the proposed optical standards in the visible region have been tested at a level that is comparable to this. Nevertheless, projected performance is quite impressive, and significant progress has been made in the past few years by using cold trapped ions and neutral atoms.

Prior to the present era of laser cooling and trapping, the highest resolution that had been achieved in the visible was the work of Barger and Bergquist, who in the late 1970s demonstrated optical Ramsey-fringe widths of approximately 2 kHz on the 657-nm calcium transition [1, 2]. Now with laser cooling, Bergquist and collaborators have demonstrated an approximately 50-Hz-wide optical transition at 563 nm in a single, trapped mercury ion [3]; Hansch's group has demonstrated approximately 7-kHz-wide lines at 486 nm on the very important hydrogen 1S to 2S transition [4]; Ertmer's group has detected approximately 2-kHz-wide lines on the 457 nm transition in magnesium in a magnetooptical trap [5]; and Riehle and collaborators have also shown approximately 2-kHz-wide resonances on the 657 nm line in calcium in a magnetooptical trap [6].

In the visible, the most common wavelength-frequency reference is the I_2-stabilized helium–neon laser at 633 nm. This system provides very good performance, has been carefully studied, and is well understood at the approximately 3-kHz level of precision. Achieving much higher performance with the 633 nm helium–neon seems improbable, given the width of the transition ($\sim$5 MHz), the cell-dependent systematic shifts, and the amount of time that has gone into optimizing the system. However, it is an excellent stabilized laser and its widespread use is not only because of its high accuracy, but also because of practical considerations: it is compact, relatively low cost, easy to use, and it lases in the visible. The excellent performance is due in a large part to the many years of developmental work that have been invested throughout the world.

Relative newcomers to the field of high-performance optical references are diode lasers stabilized to the rubidium 2-photon transition at 778 nm [7], and the I_2-stabilized frequency-doubled Nd:YAG-laser at 532 nm [8]. Both of these systems look very promising and are already competitive with existing standards, but neither has been developed, nor tested to the degree that prior systems have, so the ultimate performance is yet to be determined.

In the infrared spectral region there are two other well-established standards for quality frequency references, the CH_4-stabilized helium–neon laser at 3.39 μm, and the OsO_4-stabilized CO_2 lasers at 10 μm. In the best cases, frequency stabilities have been reported at the level of a few hertz. In terms of fractional frequency uncertainty, the CH_4 system is now known by optical frequency measurements to $(\delta\nu)/\nu \approx 3 \times 10^{-12}$, and the OsO_4 system is known to 50 Hz, $(\delta\nu)/\nu \approx 2 \times 10^{-12}$ [9].

For a new optical reference to be useful, it needs to provide something that cannot be achieved with existing standards. For example, it could feature better stability, higher accuracy, access to different spectral regions, lower cost, portability, or better reliability, or it might be more easily realized in the lab or transferred to other locations. For a combination of these reasons, diode lasers are beginning to play an important role in high-precision spectroscopy. They are obviously lower cost and more compact than other tunable laser sources, but importantly they give us access to different spectral regions. Until recently, the best stabilized lasers have either been dye lasers or gas lasers. Because of their transportability the gas lasers have served the role of wavelength and frequency-transfer standards. But stabilized gas lasers have only limited tunability, so we have had to rely on a few fortuitous overlaps of narrow molecular lines with common gas-laser wavelengths. With some limitations, tunable diode lasers now give us the opportunity to choose the best atomic or molecular transitions and, at the same time, have the option of a transportable instrument. Some of the interesting possibilities that are now being pursued with diode lasers are the alkaline earth atoms (calcium, strontium, and barium) and ions (Ca^+ and Sr^+).

3.2 LIMITS TO SPECTROSCOPIC ACCURACY

In evaluating the merits of various optical references, it is easy to focus on the linewidth and signal-to-noise ratio that can be obtained with a given transition. These are clearly important parameters, but they do not necessarily guarantee high accuracy. More often than not, the accuracy, and even the stability, are limited by systematic effects, rather than the signal size, the fundamental noise processes, or the linewidth. The effects that limit the precision and accuracy in optical frequency references are discussed in many places [9]. An early article by Hall [10] provides a clear insight into the physical processes that limit the performance of high-resolution saturated-absorption spectroscopy. Precise theoretical treatments of many of the important effects were done by Bordé and collaborators [11–14].

In the highest performance neutral-atom systems, we are almost always limited by velocity-dependent shifts and broadening due to limited observation times. Even with the thermal velocity reduced by laser cooling and trapping, gravitational acceleration limits the practical observation times. Trapped ion systems in contrast have essentially unlimited observation times but are limited in signal size by a relatively small number of ions. With neutrals we can use large numbers of atoms and hence have large signals, but we do not have unperturbed atoms that can be observed for times much longer than approximately 1 s.

Some of the important physical effects that limit the resolution and accuracy in optical spectroscopy are listed in Table 3.1. For any specific system, one effect or another may be the dominant one. But because of optical phase uncertainties and second-order Doppler effects, it is clearly difficult to push beyond 10^{-12} fractional frequency uncertainly using fast atoms. Surprisingly, even at low pressures, collision with other atoms actually limit the accuracy that can be achieved in many frequency standards. For example, at the highest levels of accuracy, collision effects are significant even in cesium atomic clocks, some microwave trapped-ion systems, and I_2-stabilized lasers.

3.3 DIODE LASER TECHNOLOGY

3.3.1 Extended Cavity Diode Lasers

The sensitivity of diode lasers to optical feedback, can be an advantage for stabilizing the laser frequency. When used judiciously, optical feedback can easily narrow the linewidth of a solitary laser from tens of megahertz to tens of kilohertz (extended cavity diode lasers, ECDLs, are a good example of this).

In the case of ECDLs (as with optically locked lasers), the natural wavelength of the solitary laser chip is independent of the extended cavity; it is a function of temperature, injection current, and semiconductor properties. For stable ECDL operation at a specific wavelength we have to suppress the modes of the solitary chip with good antireflectance coatings, or we must rely on stable current and temperature control to keep the natural frequency of the laser chip within the control range of the grating. This control range depends critically on the dispersion, the feedback level, and the reflectance of the output facet. For a typical ECDL with a facet reflectance of a few percent, the control range of the grating might be approximately 10 GHz, whereas the solitary mode spacing might be approximately 120 GHz. Thus we are faced with the fact that the ECDL laser can still jump modes if the environment is not stable or as the laser ages. If, on the other hand, the facet reflectance is reduced with a broadband coating to a power reflectance of 10^{-4}, the grating can control the wavelength over the entire gain curve ($\sim$40 nm for AlGaAs lasers), and broad single-mode scans are possible.

3.3.2 Amplitude Noise

Even though the amplitude-modulated (AM) noise level on diode lasers is very low, it does increase at low Fourier frequencies. Because we are greedy for a low signal-to-noise ratio in our experiments, we

TABLE 3.1 Typical Uncertainties in Optical Frequency–Wavelength References

Effect	Dependence, or Frequency Uncertainty $\delta\omega/\omega_0$	Hot Atoms[b]	Cold Atoms[b]	Comments
Uncertainty in size of signal	$(N\gamma)^{-1/2}$			Amplitude uncertainty improves with number of atoms N, and linewidth γ
Natural linewidth	γ/ω_0	◇ 10^{-8} to 10^{-14}	◇ 10^{-8} to 10^{-14}	Frequency uncertainty improves with ω_0 and $1/\gamma$
First-order Doppler	$\mathbf{k}\cdot\mathbf{v}/\omega_0$	◇ 10^{-6} • 10^{-12}	◇ 10^{-10} • 10^{-15}	Need Doppler-free method! Uncertainty is method dependent
Second-order Doppler	v^2/c^2	◇ 10^{-12} • 10^{-13}	◇ 10^{-17} • 10^{-18}	Requires knowledge of velocity distributions or cold atoms
Ramsey method interaction	$\Delta\phi\mathbf{k}\cdot\mathbf{v}/\omega_0$	◇ 10^{-11} • 10^{-12}	◇ 10^{-13} • 10^{-15}	Optical geometry dependent and relative Ramsey-phase $\Delta\phi$ dependent
Wavefront curvature	$v/(w_0\omega_0)$	◇ 10^{-11}	◇ 10^{-15}	Geometry dependent. Need flat wavefronts and low velocities
Power broadening	$(\gamma/\omega_0)\sqrt{(1 + I/I_s)}$	◇ 10^{-8} to 10^{-13}	◇ 10^{-8} to 10^{-13}	Need low intensity I relative to saturation intensity I_s
Light shift, (a.c.-Stark shift)	$(\mu_n\cdot E)^2 \sum_n \omega_0 h^2(\omega-\omega_n)$	◇ 10^{-9} to 10^{-16}	◇ 10^{-9} to 10^{-16}	Significant for atoms with hyperfine structure and for two-photon transitions. Need low power and isolated lines
Gas lens	$L/(kw_0^2)$	◇ 10^{-12} to 10^{-14}	◇ 10^{-12} to 10^{-14}	Significant in cells. Depends on geometry, absorption path length L, wavelength $1/k$, and mode size w_0
d.c. E&B field shifts				State dependent
Stark	$(\mu_E\cdot E)^2/\hbar\omega_0$	◇ 10^{-10} to 10^{-17}	◇ 10^{-10} to 10^{-17}	Lower lying states and S states better
Zeeman	$\mu_B\cdot B/\hbar\omega_0$	◇ 10^{-4}/T ◇ $10^{-7}/T^2$	◇ 10^{-4}/T ◇ $10^{-7}/T^2$	Use $\Delta m = 0$, or first-order field-independent transition
Collisional				
Broadening	$(N\sigma_b v)/(V\omega_0)$	◇ 10^{-10}/Pa	?	State and collision partner dependent; cross section σ_s magnitude often unknown
Shift	$(N\sigma_s v)/(V\omega_0)$	◇ 10^{-11}/Pa	?	
Transit broadening	$(v/(w_0\omega_0))$	◇ 10^{-10}	◇ 10^{-13}	Requires large beam waits w_0 or low velocities v
Adjacent transition line pulling	$\gamma_n/(\omega_0-\omega_n)$ or $\gamma_n^2/(\omega_0-\omega_n)^2$	◇ 10^{-10} to $<10^{-15}$	◇ 10^{-10} to $<10^{-15}$	Perturbation by nearby lines with transition rates γ_n. Need well-isolated lines
Recoil shift	$\hbar\omega_0/mc^2$	◇ 10^{-11}	◇ 10^{-11}	Depends on atomic mass m
Gravity acceleration, g				Horizontal laser beam. Need large mode w_0 or fountain
Broadening	$g^{1/2}w_0^{-1/2}\omega_0^{-1}$	◇ 10^{-13}	◇ 10^{-13}	
Frequency shift	$(w_0/g)^{1/2}\mathbf{k}\cdot\mathbf{g}/\omega_0$	◇ 10^{-15}	◇ 10^{-15}	

SOURCE: Parts of this table have been adapted from a similar table presented by J. L. Hall [10], and some of the data comes from Sergstock et al. [5] and Kisters et al. [6].

[a]Listed here are the general dependencies of the various physical effects, and some typical values found in neutral-atom spectroscopy. Trapped-ion systems have similar effects, but typical values can be quite different.

[b]The magnitudes of the fractional frequency shifts and broadening coefficients are indicated by ◇, and the uncertainties designated with a •. These values are not for a specific system, but are given for order-of-magnitude comparisons.

frequently try to reduce the noise even further. One approach is to sample the laser's amplitude noise with a beam splitter and photodetector, then to feedback to the injection current with an electronic servo to reduce the amplitude noise. This has the appearance of simplicity and does work, but the results depend on the degree to which the sampled beam represents the experiment beam. In the limit where the sampled beam is simply some fraction of the experiment beam, the fluctuations of the experiment beam, expressed in decibels above the shot-noise limit, may be reduced to $-10\log S$. S is the fraction of the power sampled (e.g., with a 50% beam splitter, a servo can put the experiment beam noise within 3 dB of the shot-noise limit) [15]. To reliably attain this level of noise reduction, care must be taken to ensure that the sampled beam accurately represents the experiment beam. Many lasers are packaged with an internal photodiode, which is used to detect the beam out of the laser's rear facet. Caution should be exercised in using this signal for the sampling channel, because it is not an accurate representation of the intensity fluctuations on the front facet beam.

3.3.3 Frequency Stabilization

Because diode laser cavities are small and facet reflectances are low, the residence time for a photon in the cavity is short ($\lesssim 1$ ns), which results in a substantial amount of frequency noise and hence broad spectral linewidths. Linewidths of solitary diode lasers can range from approximately 10 MHz for the near-infrared AlGaAs lasers, up to 200 MHz for the visible AlGaInP lasers. Good discussions of the various contributions to diode laser noise are given in Yamamoto et al. [16–19] .Of particular interest is the paper by Telle [19], which gives a clear and concise overview of diode laser frequency noise and stabilization. Also Zhu and Hall [20] give a good detailed description of how laser frequency noise contributes to a laser's spectral line shape.

Quite a lot of useful information about the spectral quality of diode lasers can be inferred from the measurements of the frequency-noise spectral density, an example of which is shown in Figure 3.1. The frequency noise on solitary lasers is largest at low frequencies and decreases monotonically toward higher frequencies,

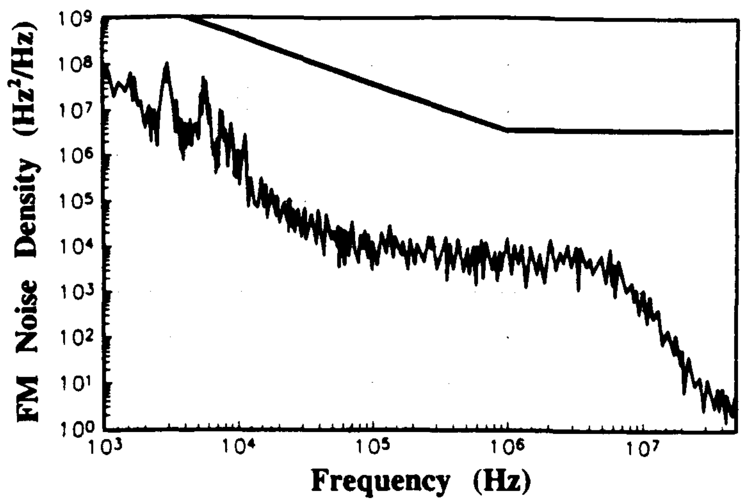

FIGURE 3.1 Frequency-noise spectral density of diode lasers. The frequency noise of an ECDL was measured by using the transmission fringe of a Fabry–Perot cavity as a frequency discriminator. The bandwidth of the cavity fringe is responsible for the roll-off in the measured noise near 8 MHz. The data was taken with an InGaAlP laser (657 nm) and was adapted from Fox et al. [21]. For comparison, the approximate noise level of a solitary diode laser is indicated as a solid line.

reaching a plateau that extends from approximately 1 MHz out to many gigahertz. For typical AlGaAs lasers the level of this plateau in the frequency noise is on the order of $10^7\ Hz^2/Hz$. In the case of extended cavity lasers the spectral characteristics depend on the cavity length, optical feedback ratio, and mechanical stability of the cavity [16–19, 21]. The frequency-noise density is reduced by a large factor (typically in the range of 100 to 1,000) from the noise level of a solitary laser. However, we still find that the laser's frequency fluctuations are largest at low frequencies, and then they level off to a plateau at intermediate frequencies (a typical spectral density is approximately $10^4\ Hz^2/Hz$ between 50 kHz and 100 MHz). For extended cavities approximately 10 cm long, the fast linewidth will be near 50 kHz. Low-frequency fluctuations of the cavity length that will modulate the inherent fast linewidth, effectively broadening it. The low-frequency vibrations show up as resonant peaks in the frequency noise as can be easily seen in Figure 3.1. As with any oscillator that exhibits increasing noise at lower frequencies, the actual linewidth will depend on the averaging time that is used to measure it, making linewidth statements very ambiguous.

It is very clear from the Figure 3.1 curves that the frequency noise on solitary diode lasers is large in magnitude and broadband. The ECDLs have a much lower level of frequency noise, but it still extends to fairly high Fourier frequencies. As is usually the case, the magnitude of the frequency noise diverges toward low frequencies. Fluctuations in carrier density, localized temperature fluctuations, and mode competition noise are believed to be the main contributions to the frequency noise at low frequencies; whereas spontaneous emission [enhanced by a factor of $(1+\alpha^2)$, with $2 < \alpha < 12$] contributes throughout.

In the literature we often find simple theoretical models (such as the phase diffusion model) of diode laser frequency noise, which are then used to declare that diode lasers have Lorentzian line-shapes. This is not our experience in the laboratory. Actual diode laser line shapes are complicated by fundamental interactions that introduce excess frequency noise (some mentioned above) as well as other less interesting, but important, technical details (like even small amounts of optical feedback) that dramatically alter the spectral character of diode laser light. The lasing frequency of ECDLs is also perturbed by thermal and vibrational changes in the cavity length, which lead to peaks in the frequency noise as can be seen in Figure 3.1. Only in hypothetical cases where the frequency-noise spectral density depends in a simple way on Fourier frequency can the line shape be described by simple analytical functions. In most practical diode laser systems, the frequency noise does not satisfy these conditions and the resulting line-shape is complicated.

As mentioned above, we are usually faced with the situation that even the laser's line width depends on how long an observation time we use to measure it. Nonetheless, it is still useful to have some simple, qualitative, one- or two-parameter measure of the spectral width of a laser. Because the actual linewidth depends on the observation time, one approach is to define a fast linewidth and then lump fluctuations that occur at lower frequencies into frequency jitter and drift (which are often conveniently neglected). Although imprecise, one prescription for determining the fast linewidth comes from using the spectrally flat portion of the frequency-noise spectral density (e.g., the region between approximately 50 kHz and 5 MHz in Figure 3.1). It turns out that, if the frequency noise is spectrally flat, then the line shape is really Lorentzian and the linewidth is given by $\Delta\nu = \pi S_f(f)$. So applying this definition to the ECDL data in Figure 3.1 gives a fast linewidth of approximately 30 kHz, and a fast linewidth of approximately 15 MHz for the solitary laser. Alternative methods to measure the line shape are to make a beat note with a narrow linewidth laser, or, in the case of spectrally broad lasers, an optical delay line can be used for a self-heterodyne measurement. These techniques can also be used in the time domain by measuring the amount of time it takes for the optical phase to become ambiguous by π radians; this serves as another indicator of laser linewidth.

The usual models of semiconductor laser gain predict that the plateau of frequency noise at high frequencies extends out to the laser's relaxation frequency (typically a few GHz) and then drops rapidly toward higher frequencies [16]. Although we have not carefully measured the frequency (phase) noise out to very high Fourier frequencies, we have inferred from noise measurements in atomic absorption experiments that the frequency noise density can be almost flat to frequencies as large 9 GHz away from the laser carrier. This admittedly indirect measurement implies that these lasers AlGaAs Fabry–Perot lasers operating at $I/I_{th} \approx 3.0$) have frequency noise that is almost spectrally flat (for frequencies between 1 and 9 GHz) with a spectral density that is approximately $-40\,dB_c$ in a 1 MHz bandwidth. Although it has not been proven, we could speculate that this level is dominated by the background spontaneous emission in the spatial mode of the coherent field of the laser.

To have a spectrally narrow laser that is also precisely tunable relative to a stable reference frequency, we follow the general approach developed some time ago by J. L. Hall and others. With this method, a nontunable Fabry–Perot cavity is used to stabilize the frequency of a reference laser; a second laser is then phase-locked to the reference laser but with a frequency offset. The frequency offset is

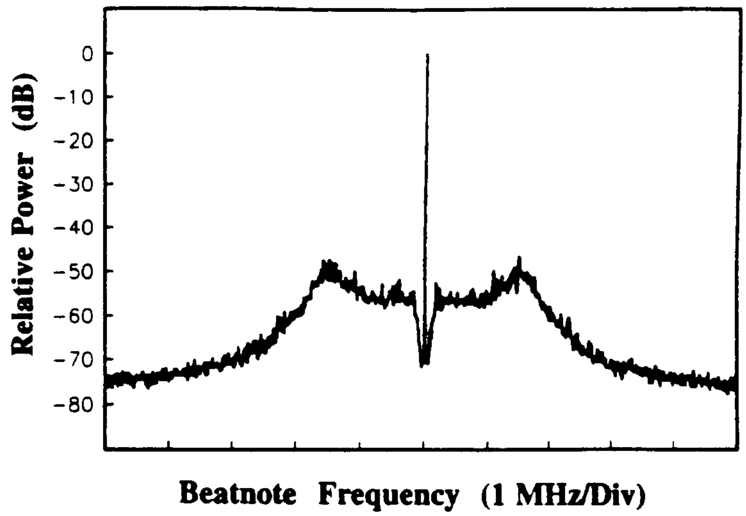

FIGURE 3.2 Beat-note between phase-locked diode lasers operating at 657 nm. Curve shows the log of the power spectral density of the beat note between the reference laser and the frequency offset laser when the system is phase locked. Noise close into the carrier is down $-60\,dB_c$ from the peak. The resolution bandwidth is 1 kHz and the horizontal axis has a full span of 10 MHz. The frequency offset of the phase-locked laser, relative to the reference laser, is continuously tunable between 10 MHz and 2 GHz with sub-Hertz precision.

provided by a RF-microwave synthesizer, which thus allows precise tunability of the laser frequency. Figure 3.2 shows the beat note between the reference laser and the tunable phase-locked ECDL under locked conditions.

To achieve the highest accuracy in the lock of the reference laser to the Fabry–Perot cavity we use the Pound–Drever–Hall optical-heterodyne technique. Frequency-modulated sidebands at approximately 20 MHz are generated with an external, resonant, phase modulator. Two feedback loops are used to frequency stabilize the reference ECDL, a low-frequency path to a piezo-crystal (PZT) that controls the ECDL length, and a high-frequency path to an AD*P phase modulator within the laser cavity. Servo-loop bandwidths of approximately 1 MHz result in a fast linewidth of approximately 500 Hz for the stabilized reference laser. More details on the stabilized reference laser can be found in Fox et al. [21].

In the laser phase locked to the reference laser, there are also two feedback channels, a slow one to a PZT, and a faster feedback channel to the laser's injection current [22, 23]. In this ECDL, the injection current tunes the frequency of the laser with a coefficient of approximately 40 MHz/mA. Unfortunately, changes in the laser's injection current also change the laser's output power, with a coefficient of approximately 0.09 mW/mA. It was not clear a priori whether the injection-current loop for frequency stabilization would reduce the amplitude noise or increase it. In studying a few of these ECDL systems, we have found that using the injection current to stabilize the laser's frequency always degrades the laser's amplitude noise. This can be understood from the tuning coefficients and the relative magnitudes of the frequency- and amplitude-modulated noise on ECDLs. The diode laser's amplitude noise is very small, but the frequency noise is significant. Thus, the injection current that removes the frequency noise causes an increase in the amplitude noise. The dependence of the relative phase between the amplitude and the frequency modulation for ECDLs (also a function of Fourier frequency) is an interesting question, but it is almost irrelevant (because of the difference in magnitude) in the usual regime of frequency stabilization by using the injection current. An example of this effect is the degradation of the amplitude noise of the red diode laser when it is phase locked to the reference laser. Figure 3.3 shows the amplitude noise of the laser with, and without, the phase lock. This amplitude-modulation data corresponds to the same lasers and conditions as the beat note curve in Figure 3.2.

In an analogous manner to locking to a Fabry–Perot cavity, an ECDL can also be stabilized to an atomic or molecular resonance such as absorption, fluorescence, saturated absorption, or an optogalvanic signal. The easiest way to lock to the peak of a resonance is to use a modulation lock [24]. In this case, the laser's frequency is modulated by dithering either the PZT that controls the cavity length or the injection current. The optical power transmitted through the resonance is detected, demodulated with a locking amplifier, and fed

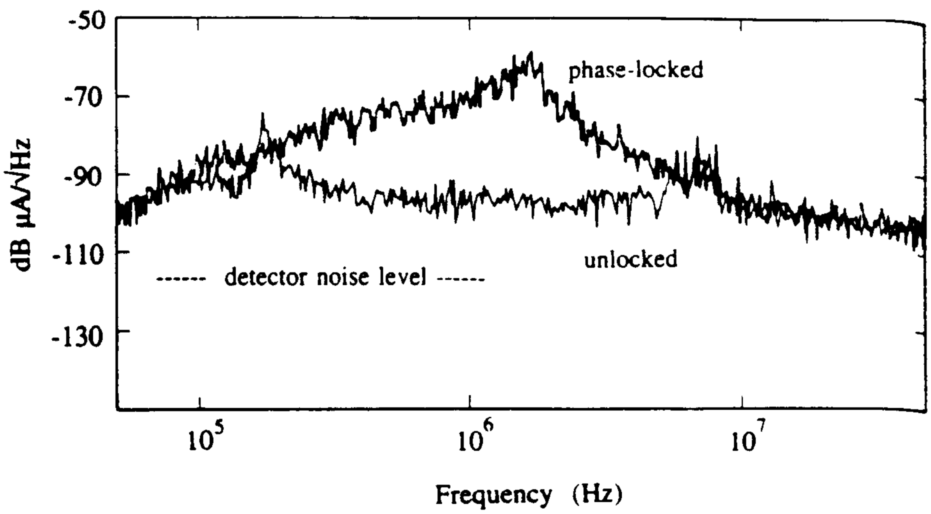

FIGURE 3.3 Amplitude-noise of the phase-locked laser for the lock conditions shown in Figure 3.2. The phase-lock servo to the injection current of the ECDL removes the frequency noise, but it simultaneously degrades the laser's amplitude noise. The upper curve shows the amplitude noise on the phase-locked laser when the system is phase locked, and the lower trace shows the intrinsic amplitude noise on the laser when the system is not locked. The equivalent shot-noise for the detected photo current of 360 μA is approximately $-100\,\mathrm{dB}_\mu\mathrm{A}/\sqrt{\mathrm{Hz}}$. Adapted from Fox et al. [23].

back to the PZT to control the laser frequency. These lockin systems that require modulation of the laser's frequency are often contaminated by unwanted amplitude modulation that comes along with the frequency modulation. In many cases, this is not significant, but the problem has to be addressed for high-accuracy frequency control. For some applications that require a spectrally clean output beam the frequency modulation can itself be a problem. In these cases, it is possible to lock to the side of a resonance without using modulation, or it is sometimes possible to derive an error signal by modulating the atomic resonance rather than the laser's frequency. A good example of this is the use of an a.c. magnetic field to Zeeman modulate atomic resonances.

3.4 CALCIUM SPECTROSCOPY WITH DIODE LASERS

The calcium intercombination line at 657 nm has been thoroughly studied using dye lasers with atomic beams for use as a wavelength-frequency reference [2, 6, 25]. The availability of red diode lasers at this wavelength, and also high-power diode lasers at 846 nm has stimulated renewed interest in calcium as a wavelength-frequency reference [26]. The whole system can be greatly simplified by use of diode lasers; with reduced size and power requirements a compact, transportable, transfer standard seems feasible. The 846 nm lasers can be frequency-doubled efficiently to produce the 423 nm light that is required for laser cooling and trapping. Similar research efforts and results in high-resolution spectroscopy of calcium with diode lasers have also been reported from the PTB group in Germany [27], and by Kurosu et al. at NRLM in Japan [28].

We have been working to improve diode laser technology to the performance level required for very-high-precision spectroscopy of calcium. This includes precise tuning, amplitude noise reduction, and frequency stabilization. The laser that probes the 657 nm line needs to be narrower than the width of the transition, and so requires precision in the frequency control that is approximately $\lesssim 1/10$th of the 400 Hz natural width, i.e., approximately 40 Hz. Our present system does not achieve this required level of performance, but rather has a fast linewidth of 500 Hz, and residual frequency jitter that causes laser frequency excursions of approximately 5 kHz at acoustical frequencies. In addition, the system is presently plagued by frequency drifts in the reference cavity that amount to approximately 1 kHz/s at the optical frequency. The jitter and the drift are solely due to instabilities in the reference cavity. The laser frequency is locked tightly to the cavity resonance and so follows the fluctuations inherent in the cavity resonance frequency. With a better cavity and using the vibration isolation and stabilization techniques that have already been demonstrated with dye lasers [3], we can expect to reach the required diode laser frequency stability.

At 657 nm, the diode lasers are a little challenging (large initial linewidths and relatively low powers) but most of the difficult problems have now been solved. In terms of ultrahigh-resolution spectroscopy, we have not done quite as well with the diode lasers as the previous results with dye lasers, but the diodes are fairly new and improving rapidly. The semiconductor lasers themselves do not seem to present a fundamental limit, and we expect equal performance will be achieved with diodes or dye lasers. The one remaining challenge is that we would like to have more optical power than is readily available from single-frequency tunable ECDLs. This is particularly relevant for time-domain Ramsey spectroscopy of cold-trapped atoms. In this case, the optimum power at 657 nm will be approximately 50 mW delivered to the atoms in a repetitive pulse sequence. The power limitation of diode lasers should not be a serious long-term problem, because there are at least two possible solutions: (a) higher power single-mode lasers in the red appear to be on the near term horizon, and (b) we can use injection locking of broadband lasers to increase the usable single-mode power.

We have constructed two very different vacuum systems to do the saturated absorption in calcium. In earlier work [26], we developed a high-flux atomic beam-cell that allowed detection of the saturated absorption signal by optical heterodyne saturation spectroscopy. That system is relatively compact and achieves good spectral resolution with an exceptionally good signal-to-noise ratio. The detected saturated-absorption signal was not large (approximately 1% of the laser power), but the linewidth was narrow (approximately 60 kHz FWHM) and, using heterodyne detection, gave a background noise that was near shot-noise limited. The linewidth and noise-limited signal-to-noise ratio gave a projected frequency stability of $3 \times 10^{-14}\tau^{-1/2}$. With this level of short-term stability, it may even be possible to lock directly to the calcium resonance without the use of any reference cavity.

The second calcium system that we constructed is a more traditional atomic-beam optical Ramsey-fringe spectrometer. Although the present system is not designed to be portable, it is reasonably compact (total length ~ 1.0 m), has good power efficiency (requiring only 20 W for the beam source), and the vacuum is maintained with a single 20 l/s ion pump. With this system, we have observed four-zone saturated-absorption, optical Ramsey-fringes with high sensitivity on the 657 nm line (Figure 3.4). To capture the narrow Ramsey-fringe structure and at the same time display the whole

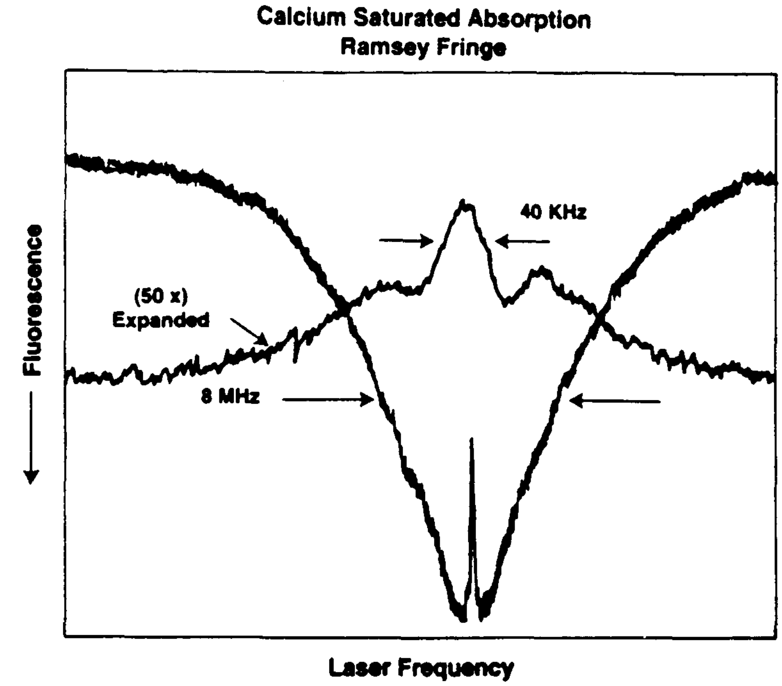

FIGURE 3.4 Calcium Ramsey-fringes taken with a diode laser at 657 nm. The lower of the two traces is a broad scan over the entire calcium resonance in the atomic beam. The laser interaction region in the atomic beam was configured in the usual way for four-zone, saturated-absorption Ramsey-fringes. Expanding this same data set by a factor of 50 along the horizontal axis produces the upper trace, where the Ramsey interference fringes now becomes apparent. In this example, the width of the central Ramsey-fringe is 40 kHz.

Doppler-broadened lineshape this spectra was taken with a scan time of 10 s but used a detection bandwidth of 10 kHz. Lower detection bandwidths give very clean Ramsey-fringe lineshapes.

Higher spectral resolution is achieved in the traditional atomic-beam system than is obtained in the beam-cell system because of the longer interaction time provided by the Ramsey interrogation method. However, there is some penalty in the signal size. The narrowest lines that we have observed up to now are 10 kHz FWHM, which is sufficient to resolve the photon-induced atomic recoil splitting of 23 kHz. Unfortunately, as is well known, the Ramsey line shape becomes quite complicated in the higher resolution limit where the effects of atomic recoil, second-order Doppler, and Ramsey-fringe resolution are all interconnected [29]. A very important simplification of the spectrum (usually at the expense of a more complex apparatus) can be achieved by eliminating one of the atomic recoil components as has been described in previous work [5, 11, 25, 30]. However, the limitation in the accuracy of this system will be the same as that previously realized in other fast atomic-beam systems. The ultimate accuracy will likely be limited by imprecise knowledge of the atomic velocity distribution and hence the second-order Doppler shift and also by the atomic velocity-dependent optical-phase sensitivity of the Ramsey interferometer. Projected accuracy of these systems is approximately 500 Hz, $(\delta\nu)/\nu \sim 1 \times 10^{-12}$.

Real improvements in the frequency accuracy require low-velocity atoms. Fortunately laser cooling and trapping of calcium has already been demonstrated with dye lasers, and, recently, high-resolution spectra of cold neutral atoms have been achieved [5, 6]. It is also now feasible to do the trapping and cooling of calcium with frequency-doubled diode lasers. We have built a doubling system that uses $KNbO_3$ in a resonant ring-cavity. Starting with an 846-nm grating-tuned extended-cavity laser, we injection lock a 150 mW, single-, spatial-mode, diode laser. The resulting high-power beam is collimated and sent through an anamorphic prism pair and optical isolator before it is mode-matched into the ring build-up cavity. The resonance of the ring is electronically locked to the laser frequency. With 110 mW of 846-nm light incident on the ring we have been able to generate as much as 45 mW of useful output at 423 nm. This should be sufficient for laser cooling and trapping of calcium. Thus far, we have demonstrated one-dimensional transverse-cooling of the calcium beam, which enhances the Ramsey-fringe signal by a factor of two to three.

It is also interesting that some other transitions in calcium are accessible with diode laser sources. Some of these may play an important role in improving the performance of this standard in the future. For instance, we have used a 672-nm diode in conjunction with the 423-nm light to probe the calcium $^1D(4s3d)$ to $^1P(4s5p)$

transition. This gives us additional information about (and may enhance) the 423-nm cooling process and may also be useful to suppress the large background (657 nm) emission that is present in the cold trapped-atom systems. Other transitions in calcium are also compatible with diode lasers, including the 612-nm transition that can be used to suppress a recoil component. Similarly, 423- or 444-nm light, from double diodes, could be used to improve the trapping or to suppress a recoil component.

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