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Laser diode cavity ring-down spectroscopy using acousto-optic modulator stabilization

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By using an acousto-optic modulator, we have stabilized a free-running continuous wave (CW) laser diode in the presence of strong reflections from a high finesse Fabry–Perot resonator. The laser diode linewidth can be stabilized from several MHz, for high resolution spectroscopy of species at low pressures, to several hundred MHz, for lower resolution spectroscopy of species at atmospheric pressures. We demonstrated CW cavity ring-down spectroscopy of water vapor at both 1 atm and 5 Torr. We achieved ring-down repetition rates of 10–50 kHz, and a noise level of $2 \times 10^{-8} \text{ cm}^{-1}$.

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I. INTRODUCTION

Cavity ring-down Spectroscopy (CRDS) is a high sensitivity absorption technique with potential for absolute concentration measurements of trace gases and impurities.^{1–7} CRDS is usually practiced by coupling a pulsed laser source into a high-finesse optical resonator (Fabry–Perot cavity) that encloses the sample of interest, and detecting the decay of light in the resonator. Under many conditions, the decay is exponential and a plot of the ring-down decay rate versus frequency gives the absorption spectrum. The ring-down decay rate is controlled by the resonator finesse, and changes wherever the sample absorbs. This approach is limited in spectral resolution by the laser linewidth ($>100 \text{ MHz}$ for pulsed sources),⁸ in data acquisition rates by the pulse repetition rate of the laser (ca. 10–100 Hz), and in sensitivity by the inherent trade off between resonator finesse and light throughput.

Recently, efforts have been made to overcome these limitations of pulsed CRDS by the use of narrow linewidth ($<10 \text{ MHz}$) continuous wave (CW) lasers. Lehmann,⁷ Meijer,⁹ and Romanini¹⁰ were the first to propose and implement the use of narrowband laser sources in CRDS. Lehmann⁷ first proposed coupling narrowband CW diode laser sources to ring-down cavities, in order to build up enough energy in the cavity to perform shot noise limited measurements. Meijer *et al.*⁹ measured phase shifts (PSS-CRDS), rather than intensity decay of an intensity-modulated CW light beam. They demonstrated for the first time, that acquisition rates of kHz were compatible with high-resolution CRDS, and that narrowband laser throughput, could be improved by using stable resonators with a dense transverse mode structure. In an alternative approach, Romanini *et al.*¹⁰ swept one resonant mode of the cavity through the CW laser linewidth (CW-CRDS), thereby allowing significant buildup

of light inside the ring-down cavity, then switched off the light with an acousto-optic modulator (AOM) and measured the intensity decay. Acquisition rates of several hundred Hz were thereby demonstrated, in addition to excellent light throughput, and reduced baseline noise leading to very high sensitivities. Romanini *et al.*¹¹ were also able to apply CW-CRDS to an external cavity diode laser (ECDL).

Much of current ring-down spectroscopy^{1–6} still relies on fairly costly laser sources. As solid state lasers (e.g., Ti:Sapphire lasers, Nd:Yag-pumped OPOs, and ECDLs) have gained in reliability, tuning range, and output power, they have started to replace the more traditional tunable dye lasers, although they are no less expensive. Simultaneously, semiconductor laser diodes (LDs) have also been improving in power, wavelength coverage, and reliability. The rapid growth of the communications industry in recent years has resulted in the availability of tunable near-infrared LDs at a rapidly diminishing cost ($<\$500$). In fact, owing to their compactness, low cost, durability, high wallplug efficiency, and compatibility with both fiber and silicon technologies, infrared laser diodes seem to be an ideal light source for realizing practical CRDS systems.

Early attempts demonstrated difficulties in applying LD sources to CRDS:^{7,12} whenever a LD beam is reflected directly back into the laser, as occurs in a linear cavity configuration, even under optical isolation, the optical feedback may result in phase fluctuations and mode hopping of the LD. In fact, at higher feedback levels, as would be typical for most practical setups, a wide variety of effects ranging from linewidth broadening to complete “coherence collapse” (linewidth $>10 \text{ GHz}$) is often observed¹³ and is illustrated in Fig. 1(a). The inherent problem is the formation of “external cavities” by reflective optics with the output facet [which has either a low reflectivity or is antireflection (AR) coated to increase output power] of the LD. The external cavity affects both the gain and phase relations of the LD.^{14–16} Thus, whenever back reflection is allowed, the lasing characteristics of the LD (frequency, number of excited modes, output power, etc.) become highly dependent on uncontrol-

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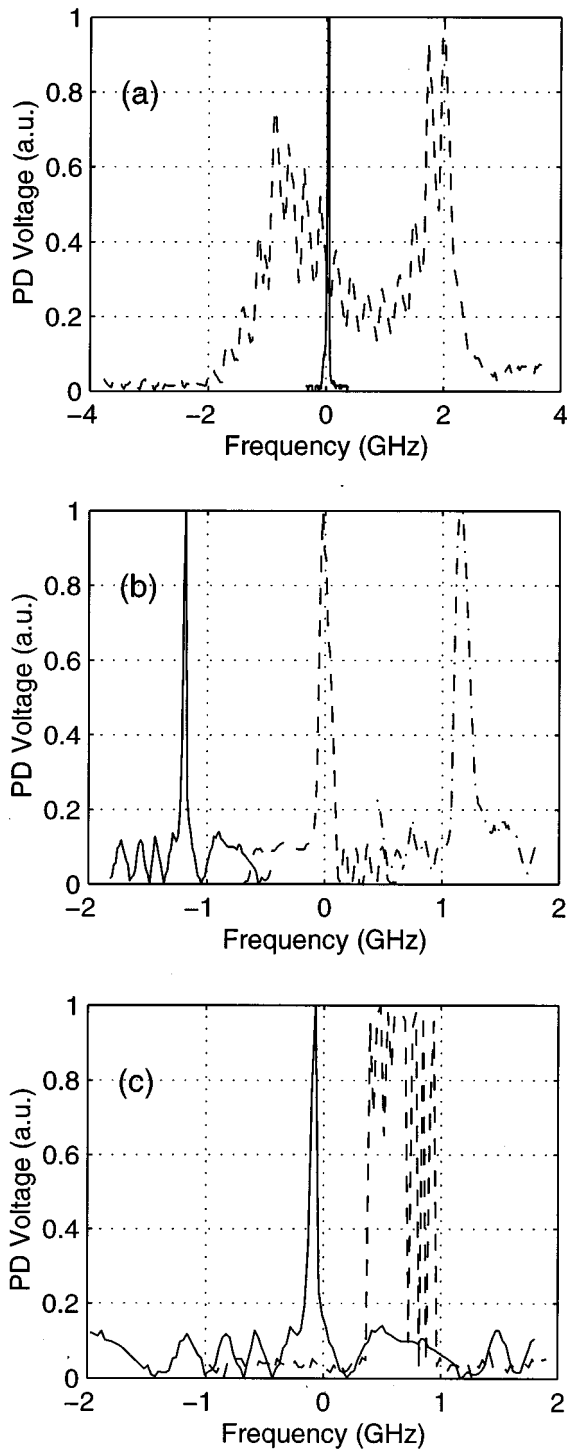


FIG. 1. (a) Linewidth for a free-running LD (solid) and for a LD under feedback (36 dB feedback) from a ring-down cavity (dashed). (b) LD linewidth as a function of external cavity length for only first order feedback: $L_{\text{ext}}=215$ cm (solid), $L_{\text{ext}}=100$ cm (dashed), and $L_{\text{ext}}=55$ cm (dashed-dotted). The feedback level is 50 dB in all three cases. (c) LD linewidth for only first order (50 dB) feedback (solid) and both first (50 dB) and zeroth (50 dB) order feedback (dashed).

lable experimental parameters, most notably the external cavity length.

Several solutions exist to this coupling problem. A LD with an AR coated output facet can function as a gain medium in an external cavity (formed by a feedback element

and a frequency selective element), that allows only one specific mode to exist, and that reduces (but does not completely eliminate) the sensitivity of the LD to feedback;¹¹ the feedback from a linear cavity configuration can be completely eliminated by using a ring resonator structure, as will be discussed elsewhere;¹⁷ or, the external cavity effect can be controlled by placing an AOM inside the external cavity, thereby stabilizing the time-averaged behavior of the LD. The last approach, first demonstrated by Martin *et al.*¹³ as a useful scheme for stabilizing LDs in the presence of direct back reflections, and its application to CW-CRDS, is the primary focus of this article.

II. AOM STABILIZATION OF LASER DIODES

In an AOM device, a pressure transducer creates a sound wave that modulates the index of refraction in an active non-linear crystal, thereby producing a Bragg diffraction grating that disperses an incoming light beam into multiple orders.¹⁸ By switching the radio frequency (rf) power that drives the pressure transducer, light can be rapidly deflected by the AOM into and out of the first order, so that AOMs can be used as high-speed (<(10 ns risefall time) CW light modulators. The resulting diffracted beam is also frequency shifted by an amount equal to the acoustic wave frequency.¹⁸ Hence, by placing an AOM into the external cavity formed by the LD back facet and the ring-down resonator input mirror, any light fed back from the first order becomes frequency shifted by twice the rf driver frequency. By considering the Lang and Kobayashi equations¹⁹ that model the effects of optical feedback on LD behavior, it becomes clear that the frequency shift of the AOM forces the diode to cycle through all values of phase with a period equal to the reciprocal of the frequency shift.¹³ Therefore, in order to achieve adequate stabilization, the AOM frequency must exceed the ring-down decay rate. Typically, a driver frequency greater than several MHz is sufficient for good quality ring-down reflectors ($R > 99.99\%$), and will eliminate any changes in phase shift associated with changes in length of the external cavity. Under these conditions, the effective LD linewidth becomes insensitive to both thermal (seconds time scale) and mechanical (milliseconds time scale) perturbations of the external cavity length.

By placing an AOM between the laser diode and the input mirror of the ring-down cavity, the AOM can be used not only to switch the CW beam into and out of the first order diffraction, but simultaneously control LD linewidth. The AOM driving power determines the diffraction efficiency and hence, the amount of feedback to the LD. The external cavity length fixes the maximum achievable linewidth for each feedback level [cf. Fig. 1(b)]. The first order diffraction feedback drives the LD phase and stabilizes linewidth. Finally, the linewidth can be further enhanced by introducing nonfrequency-shifted feedback (of at least the same magnitude as first order feedback), that allows multiple external cavity modes to exist. The first order feedback then cyclically chirps the LD output through these modes at twice the AOM driver frequency [cf. Fig. 1(c)]. The flexibility in achievable LD linewidth in turn enables many different CRDS applications. Pressure broadened transitions, such as

those of trace gases in the environment, can be studied using a broad LD linewidth (100 MHz to 1 GHz) with the advantage that the cavity throughput is increased with broader linewidth. Alternately, high resolution, Doppler-limited CRDS,^{10,11} can be performed with a narrowed (1 MHz to 100 MHz) LD linewidth.

In this article, we focus on the demonstration of sensitive CRDS detection of water vapor using an AOM stabilized LD with both first and zeroth order feedback. The absorption spectra were extracted from a measurement of the intensity decay of light exiting the ring-down resonator. We note, however, that the sensitivities reported here are only a starting point for this method, as the primary sources of baseline noise are not presently, completely understood. System performance will strongly depend on the application at hand. System optimization for several different applications, and a discussion of the trade offs involved in these different situations will appear elsewhere.²⁰

III. APPARATUS AND EXPERIMENTAL METHOD

The LD employed in these experiments was a Sharp LT017MD single-mode (V-channel current blocking) GaAlAs LD. The reflections of the facets are about 90% and 5%. Its threshold current is typically 60 mA when free-running. The LD was operated with both current and temperature stabilization (Peltier heater/cooler), provided by a laser diode controller (Melles Griot: 06DL203 and 06DTC007). The free-running diode produces about 40 mW output power at 812 nm for 26 °C and 100 mA operating current I_{op} . Wavelength scans were performed by current tuning the diode for small wavelength excursions ($0.001 \text{ nm} = 0.015 \text{ cm}^{-1}$ at 810 nm with a 0.01 nm/mA rate), and temperature tuning for larger wavelength excursions ($0.1 \text{ nm} = 0.15 \text{ cm}^{-1}$ at 810 nm with a 0.05 nm/°C rate). The LD output remained stable over 1.2 nm (812.6–813.8 nm), but only 0.6 nm (813.2–813.8 nm) were used for the spectroscopy of water vapor. The bandwidth of the free-running device was independently measured to be about 50 MHz.

The experimental setup is shown in Fig. 2. The components were mounted on a vibration-isolated movable optical table (Newport: VW Series Workstation), but otherwise no special precautions were taken against thermal motion of the optical elements, even the ring-down resonator. The output of the laser is collimated with a NA=0.25 lens, and passes through a pinhole (used to selectively block back reflections into the laser, and to protect it from back reflections from the isolator itself), and then an optical isolator (OFR: IO-5-NIR-LP) before it reaches the AOM (Brimrose: GPM-400-100-960 with FFA-400-B2-F1 driver). The laser is focused to about 60 μm at the center of the AOM crystal and is recollimated after the AOM. The rf driver of AOM is modulated with a square wave output from a function generator (HP: 8116A). The zeroth order of the AOM passes into the diagnostics of the experiment, namely a fiber-coupled wavelength meter with 0.001 nm resolution (Advantest: TQ8325), and a scanning confocal interferometer (Burleigh: 8 GHz SA^{Plus} analyzer with RG-91 driver, DA-100 detector) whose back reflection can be used for zeroth order feedback (or blocked with a pinhole on slight detuning). The beamsplitter

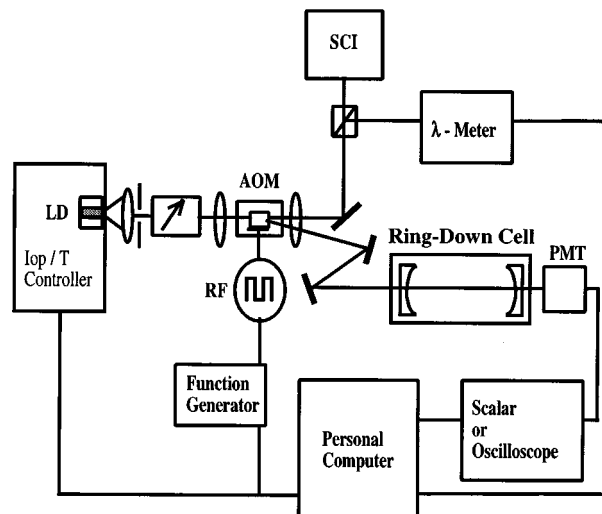


FIG. 2. Laser diode cavity ring-down spectroscopy setup using AOM feedback stabilization.

in the diagnostics achieves a 65/35 split with 65% of the light reaching the wavelength meter. The first order passes through optional beam shaping optics (beam expander and mode-matching lenses) before entering the ring-down resonator. The ring-down cavity is 45.7 cm in length, comprises two 1 in. mirrors with 1 m radius and 99.97% reflectivity (Newport: 10CV00.SR.40F), and is enclosed in a vacuum chamber with 2 in. sapphire windows. Cavity adjustment is made via flexible bellows fixed in gimbal mounts (MDC: FGC-275-M). Such a stable rather than confocal optical cavity configuration, as first proposed by Meijer *et al.*,⁹ was chosen to provide a fairly dense mode structure that was found to increase output signal levels; and reduce shot-to-shot fluctuations in cavity coupling, and hence time constant.

Detection at the cavity output is performed with a photomultiplier tube (Hamamatsu: R2658P with SRS PS 325 high-voltage supply) and a photon-counting scalar (SRS: SR430). It should be noted, that the cavity output is attenuated with a ND = 2 filter in order to prevent overflow of the scalar's counting bins. Later, the attenuator was removed, and the output signal of the photomultiplier was directly digitized on an oscilloscope (HP: 54510A). The ring-down decay obtained from either the scalar or the scope is fitted on a personal computer using the Levenberg–Marquardt algorithm, with the initial guess being provided by a linear least squares fit of the logarithm of the signal. This scheme, first proposed by Naus *et al.*,²¹ correctly fits exponential decays with a constant offset, as is typical for a photomultiplier output. This becomes especially important when a large dynamic range in ring-down constant is required, i.e., both very weak and very strong lines are simultaneously present in a spectrum, or large fluctuations in concentration for a given species are being measured. Our effective data acquisition rate is presently limited to several hundred Hz by the oscilloscope and scalar response rates.

Cavity pressures are measured with both a baratron (MKS: PDR-D-1) and a convection gauge (Granville–Phillips: Model 275). Before any sample is introduced for

measurement, the cavity is purged below 1 mTorr using a mechanical pump with a liquid nitrogen trap. All background scans are also taken at this pressure. For low pressure measurements, a constant water vapor pressure is established in the cavity, by adjusting both the cavity evacuation rate and the water vapor inflow rate. Stable flows ranging from 20 mTorr to 10 Torr in cavity pressure were thus established. When measuring water vapor in room air, the pump was removed, and the valve simply opened to let air into the cavity.

IV. RESULTS AND DISCUSSION

Using the AOM stabilization scheme for a laser diode source, shown in Fig. 2, we were able to perform CRDS on water vapor present in ambient air or in an evacuated optical cavity. LD linewidth control was performed with feedback from both first and zeroth orders, with levels of -48 and -47.6 dB, respectively, worst case, and -52 and -51.7 dB, respectively, best case (the isolator has 36–40 dB isolation) for broad linewidth measurements. These feedback levels were estimated with the help of a beamsplitter placed immediately at the LD output. A total full width at half-maximum linewidth ranging from 500 MHz at lower tuning temperatures (30°C) to 240 MHz at higher tuning temperature (40°C) was observed, and is illustrated in Fig. 3. This laser linewidth assured that at least one longitudinal mode, and its associated transverse modes, were always excited in the ring-down resonator.⁹ Similar observations can be made for low pressures scans made with narrower linewidths (180–240 MHz).

The decrease in linewidth, i.e., the number of excited external cavity modes, appears to indicate that the overall LD dynamics depend strongly on temperature. Changes in carrier density noise, index of refraction or carrier confinement, all of which affect the LD phase, appear to be fairly negligible for a 10°C temperature change, and hence not directly responsible for this substantial linewidth change. Nonoptimal packaging, and hence nonuniform LD heating could produce strain on the output facet, thereby changing its reflectivity, and hence the feedback coupling. More detailed investigations are necessary in order to determine the origin and to control this dependence.²⁰ For small temperature excursions (ca. 2°C) and correspondingly small wavelength excursions (ca. 0.1 nm), however, the linewidth remained constant, as expected. Thus, for monitoring one or two lines of a given species, this stabilization scheme is more than adequate. Because minimization of baseline noise remains critical, a comparative study on the effects of using zeroth order feedback versus external cavity length to achieve broad linewidth LD output is necessary.²⁰

The cavity output decay train as seen on an oscilloscope is shown in Fig. 4(a), while that recorded by the scalar for a single decay is shown in Fig. 4(b). Figures 4(a) and 4(b), show the actual averaged signals that were fitted to obtain the ring-down decay rates. Over a millisecond time interval, the stabilized LD is coupled into the unstabilized resonator every time that the AOM diffracts light into the first order, because our linewidth was much larger than that used by Meijer.⁹ Effectively, the AOM modulation square wave at 10

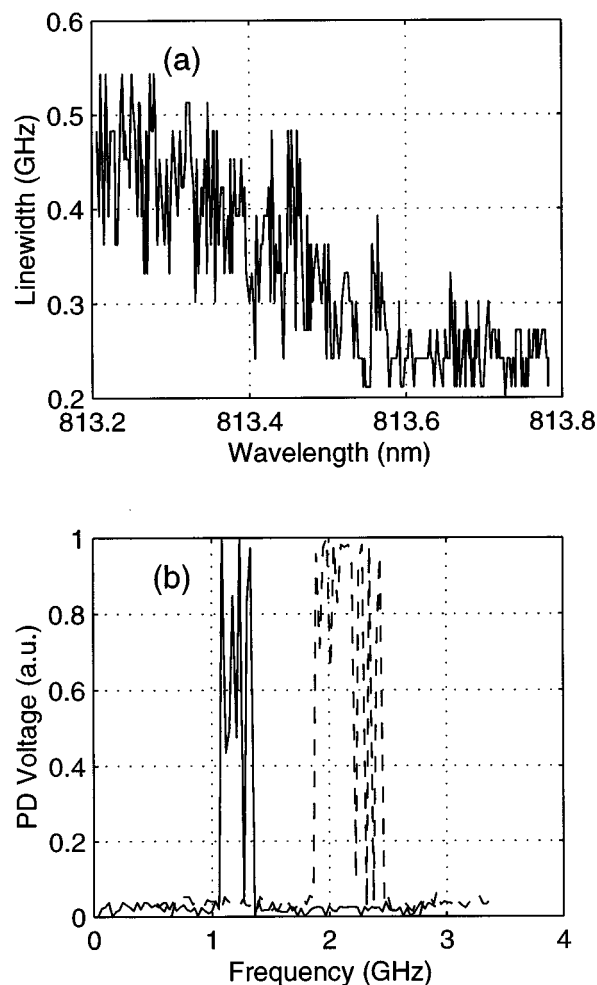


FIG. 3. (a) LD linewidth as a function of wavelength (50 dB feedback); (b) LD spectral output at 813.2 nm (dashed) and 813.6 nm (solid).

kHz is convolved with the cavity response, producing an exponential signal build up (ring-up) and an exponential decay (ring-down) with equal time constants. For sufficiently fast sampling electronics, both ring-down and ring-up time constants could be measured, and time constant acquisition rates on the order of kHz (depending on the quality of the cavity mirrors) could be achieved. Of course, ring-up time constants have the possibility of additional laser noise, which seems to be the case in the present setup.

On closer inspection of the coupling, however, it was found that there was a variation (about 5%) in the intensity of the coupled light. This root-mean-square (rms) variation corresponds to different numbers of cavity modes being excited by the input radiation. In fact, this cavity mode number variation was probably a major source of our baseline noise, which proved to be about 1% from shot to shot at a fixed wavelength in an evacuated environment, and about 4% from shot to shot in room air. By averaging over 300 shots on the oscilloscope and 5000–10 000 shots on the scalar, this variation could be significantly reduced. Signal levels for both scalar and scope detection remained high, so that actual noise in the waveform itself remained below 1%, allowing accurate numerical fitting.

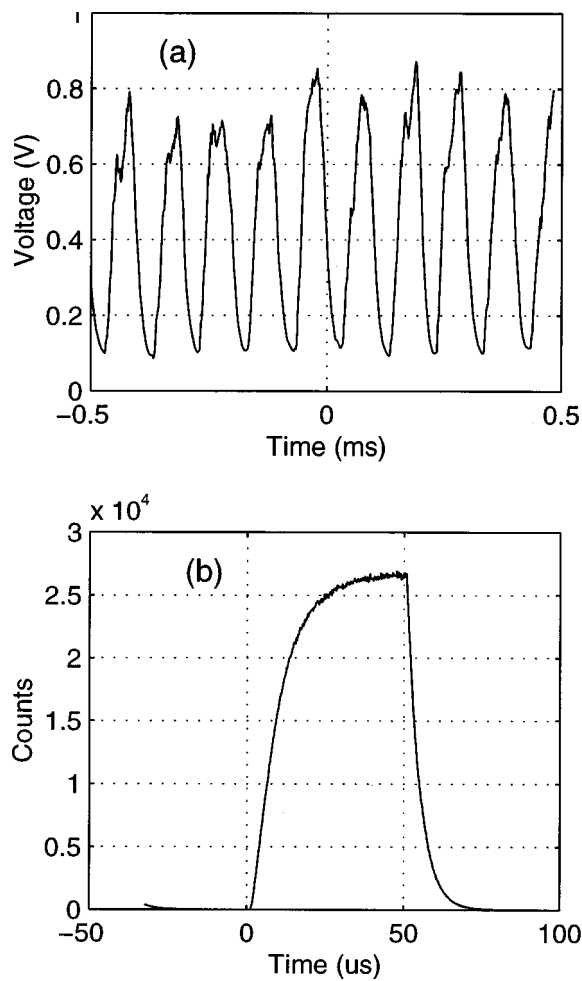


FIG. 4. Ring-down waveforms showing cavity throughput as recorded on (a) an oscilloscope (300 shots averaged); and (b) a multichannel scalar (5000 shots averaged).

Spectra of water vapor in room air and at 5 Torr are given in Fig. 5. Spectra were obtained in one continuous scan. Spectra at ambient pressure used maximum zeroth order feedback (47.6 dB) to achieve the largest possible linewidth (240–500 MHz) and cavity coupling. Spectra at low pressures (<100 Torr) used less zeroth order coupling (58 dB) to achieve a narrower laser linewidth (180–240 MHz), and to avoid convolution of the laser line with the absorption line.⁸ Scan step size in both cases remained limited to 0.001 nm resolution by the current step resolution (0.1 mA) of the LD driver. No baseline adjustments have been made, but the overall baseline noise exceeds that reported by Romanini *et al.*,^{10,11} and results from the excitation of multiple transverse modes in the cavity, which were used to improve light throughput. Our detection limit was $2 \times 10^{-8} \text{ cm}^{-1}$ for 99.985% reflectors. Our quoted detection limit is calculated as the rms baseline noise in an evacuated cell, divided by the quantity $L/(1-R)$, where L is the path length and R is the mirror reflectivity. Its value is comparable to most pulsed CRDS studies,^{2,4-6} and is presently limited by the quality of our mirrors.

Our spectra compare very favorably, in absolute frequency, linestrength and linewidth, to those generated by the

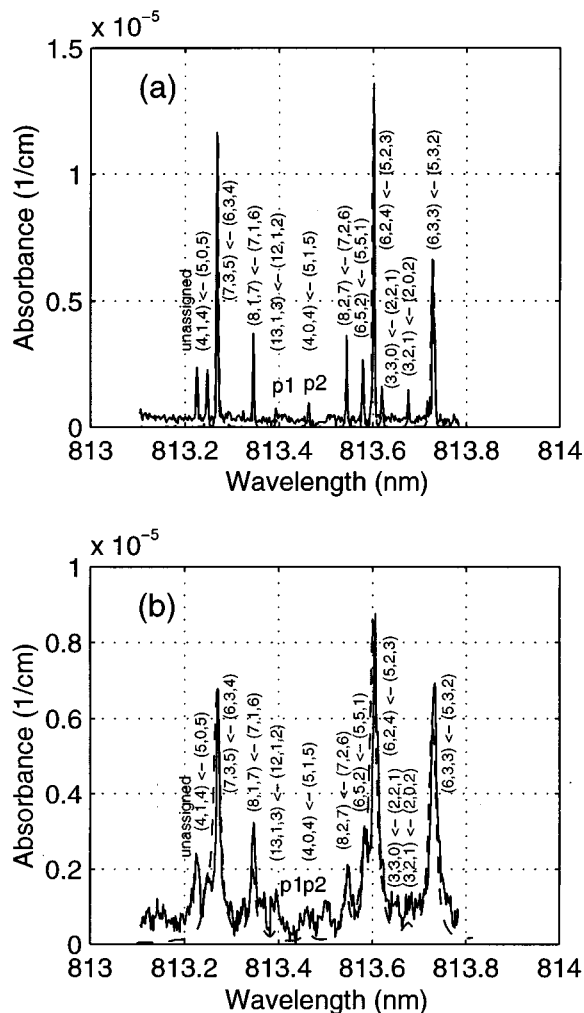


FIG. 5. Spectrum of (a) water vapor in room air and (b) 5 Torr water vapor in a cell previously evacuated to below 1 mTorr. Spectra based on HITRAN96 are shown as dashed lines. Weakest lines occur at p1 (813.391 nm) and p2 (813.452 nm). Transitions are labeled as $(J', K'_a, K'_c) \leftarrow (J, K_a, K_c)$.

HITRAN96 database.²² HITRAN is an acronym for High Resolution Transmission molecular Absorption database (<http://www.hitran.com>). The peak located at 813.224 nm, which consistently appeared in our spectra, is not listed in HITRAN96, but can be found in Table III of Ref. 23, where it is unassigned. The lines presented here belong to rotational transitions within the $2\nu_1 + \nu_2 + \nu_3$, $3\nu_1 + \nu_2$, and $\nu_1 + \nu_2 + 2\nu_3$ vibrational bands. The weakest peaks recorded in HITRAN96 were measurable with our system. These peaks, located at 813.452 nm (p2 in Fig. 5) and 813.391 nm (p1 in Fig. 5) correspond to linestrengths of $6.33 \times 10^{-26} \text{ cm}^{-1}/(\text{molecule}/\text{cm}^2)$, and $3.40 \times 10^{-26} \text{ cm}^{-1}/(\text{molecule}/\text{cm}^2)$, respectively. The nominal sensitivity of this system to water vapor was 20 ppm in low pressure flows, and 200 ppm at 1 atm pressures.

For the “open air” CRDS measurements, the baseline, noise, and overall sensitivity were significantly degraded from those of the evacuated cell (the time constant decreased

from 10 to 6 μs). This degradation is most likely caused by the deposition of dust and other airborne particles or droplets existing in the environment, because it was reproducibly eliminated by evacuating the ring-down cell. Such contamination of the high reflectors that are fundamental to CRDS sensitivity, may limit the ultimate sensitivity of trace species detection systems for the ambient environment.

V. CONCLUSIONS

We have demonstrated that it is possible to not only stabilize a free-running laser diode in the presence of strong reflections from a ring-down cavity, but also control the linewidth of the laser diode. It is also possible to stabilize the laser diode to only several MHz, if high resolution is required. In our application, we are mainly interested in water vapor concentrations at near-atmospheric pressures. Consequently, we used linewidths on the order of a few hundred MHz, comparable to those of pulsed lasers. We have performed CW-CRDS with ring-down repetition rates of 10–50 kHz, and have achieved a noise level of $2 \times 10^{-8} \text{ cm}^{-1}$, comparable to pulsed CRDS.

The nominal sensitivity (20 ppm at low pressure, 200 ppm at 1 atm) of this system to water vapor can be improved by not only choosing a stronger absorption band of water at 1.3647 μm , but by designing fast analog detection electronics, averaging over more waveforms, obtaining better reflectors, and adding some cavity tracking or feedback to allow mode matching and controlled mode coupling. The sensitivity enhancement factor of each of these improvements is estimated at 1000, 5, 3, 10, and 10, respectively, leading to detection levels of sub ppb of water vapor under atmospheric conditions (and tens of ppt for low-pressure conditions). The realization of these detection limits in nonevacuated environments, however, will require means to be found to keep the reflectivity of the mirrors at a high constant value during operation of the instrument.

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