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Cavity-locked ring-down spectroscopy

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We have performed cavity ring-down spectroscopy by locking a high-finesse resonator to the probe laser. We have obtained combination overtone spectra of water vapor in the ambient environment with a baseline noise of 5×10^{-9} cm^{-1} for decay constants ($R=99.93\%$ reflectors) of 1 μs. This cavity-locked approach ensures single transverse mode excitation, reduces shot-to-shot fluctuations in the decay constant to 4×10^{-3}, and eliminates oscillations in spectral backgrounds. This approach also allows ring-down decay acquisition rates limited only by the ring-down and buildup constants of the resonator, and holds the promise of offering truly shot-noise-limited cavity ring-down spectroscopy measurements. [S0021-8979(98)02208-7]

I. INTRODUCTION

Cavity ring-down spectroscopy (CRDS)[1] is a highly sensitive absorption technique that allows for absolute concentration measurements of dilute gas-phase species. In the past, this technique has primarily depended on pulsed lasers.[2–5] Typically, light from a pulsed laser is injected into a high-finesse linear resonator through the input mirror, and the decay of light within the cavity is monitored by detecting light exiting the resonator through the output mirror. The intensity of light injected into an empty cavity decays exponentially with time, t, owing to resonator losses. This ring-down is characterized by a rate, r, or lifetime, τ, given by:

$$I(t)=I_o\, e^{-rt}, \quad \tau=1/r. \tag{1}$$

For a cavity having roundtrip length l_{tot}, the ring-down rate is inversely proportional to the roundtrip time of light within the cavity, $t_r=l_{tot}/c$, and proportional to resonator losses, L_{tot}, which include transmission, T, scattering, S, and absorption of light, A by the mirrors (having reflectivity $R=1-(T+S+A)$), and absorption of light, α, by gas-phase species that might be present within the resonator:

$$L_{tot}=T+S+A+\alpha. \tag{2}$$

Therefore, by measuring r (or τ) as a function of frequency, an absorption spectrum of species present within the resonator may be generated.

As long as the pulse duration of injected light is shorter than the round-trip time within the cavity,[1] and the laser linewidth is smaller than the absorption features of interest,[6] pulsed CRDS is a straightforward and very sensitive technique for obtaining the absorption spectrum of dilute or weakly absorbing gas-phase species. The use of pulsed lasers, however, imposes several limitations on the CRDS technique.[7–11] Data acquisition rates are typically limited by pulsed laser repetition rates of hundreds of Hz, although 1–10 kHz pulsed tunable laser systems are now becoming available commercially. The intensity of the light coupled into and out of the cavity is small, as a consequence of reduced spectral overlap between cavity modes and laser linewidth (high cavity mirror reflectivity), as well as a lack of significant light buildup within the cavity. Moreover, for most practical pulsed laser systems, interference effects within the ring-down cavity preclude the use of simple models to describe the decay of light within the cavity as has been extensively demonstrated.[12–14] To overcome some of these problems, Lehmann[9] proposed the use of continuous wave (cw) laser sources for CRDS, in particular, laser diode (LD) sources. Single-mode laser diodes can be switched on and off at MHz repetition rates, and also have narrow linewidths ($<$ 100 MHz). The narrow laser linewidth increases the overlap with the ring-down cavity linewidth, which allows increased energy buildup inside the ring-down resonator.[15] Higher energy levels in the cavity translate directly to higher output light levels, which in turn increase the signal-to-noise ratios on decay wave forms and detection sensitivities.

The sensitivity limit of CRDS has been defined[2] as the ratio of experimental rms noise in the ring-down decay, $\sigma_{\ln\tau}$, the decay lifetime, τ, and the effective path length, l_{eff}:

$$\sigma_\alpha=\frac{\sqrt{2}\,\sigma_{\ln\tau}}{l_{eff}N_p}, \tag{3}$$

where $l_{eff}=c\tau=l_{tot}/L_{tot}$, $\sigma_{\ln\tau}=\sigma\tau/\tau$, and N_p is the number of photons reaching the detector. Clearly, increasing the effective path length by increasing either the cavity length or mirror reflectivity, will only decrease σ_α if $\sigma_{\ln\tau}$ is preserved. This approach to increasing CRDS sensitivity is analogous to the use of White cells in simple absorption spectroscopy, and does not fully exploit the noise filtering properties of the optical resonator that are central to CRDS. The key to significant sensitivity improvement therefore lies in the reduction of $\sigma_{\ln\tau}$. Decay constant fluctuations depend on optical

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noise in the decay wave form (i.e., shot noise depending on $\sqrt{N_p}$), noise introduced by the detection electronics (e.g., amplifier noise, thermal noise in circuit resistance, or detector dark current), noise imposed by data acquisition system (e.g., digital oscilloscope quantization) used to acquire and fit decay wave forms,[2] and variations in the experimental conditions (e.g., cavity pathlength changes or laser-cavity coupling). The smallest $\sigma_{\ln\tau}$ reported to date[2] is 0.0025 for a 1 s integration time. Furthermore, it is clear that under ideal conditions (i.e., no detection or data processing noise), σ_α ultimately depends on the cavity throughput, leading to the possibility of shot-noise-limited measurements.

Initially, efforts to use a cw laser in CRDS were directed at optically locking a LD to a high-finesse cavity.[16] This approach was precluded, however, by a lack of long-term stability (i.e., thermal drift) in locking the laser to the same cavity mode. Romanini *et al.*[7,8] avoided this optical locking problem altogether by sweeping one resonant mode of their ring-down cavity through the cw laser linewidth. When sufficient buildup of light within the cavity was detected, the input laser beam was deflected with an acousto-optic modulator (AOM), and the cavity ring-down decay was recorded. Using this technique Romanini *et al.* achieved acquisition rates of several hundred Hz, with greatly reduced baseline noise, and extreme sensitivities. Nevertheless, Romanini *et al.* observed several limitations of this approach. In particular, the noise in the ring-down decay wave forms was far from shot-noise-limited; pulse-to-pulse variations in the ring-down decay were on the order of a few percent; baseline oscillations close to, but not equal to, the cavity free spectral range appeared and could not be systematically eliminated; and, back reflections from their linear CRD resonator strongly perturbed the external cavity diode laser (ECDL) performance, so that significant optical isolation (35 dB) was required.

We report here an alternate cw-CRDS technique that employs two orthogonally polarized laser beams from a single ECDL. One beam is used to lock continuously a high-finesse external cavity to the output of the ECDL, while the second beam is used to measure the ring-down time of the high-finesse cavity. Despite the greater complexity involved in locking a ring-down cavity (RDC) to an ECDL, there is a significant reduction of the shot-to-shot variation in the decay constant, elimination of baseline oscillations, and decreased optical feedback to the laser. In this contribution, both detection and locking systems involved are described, and their performance and limitations discussed.

II. CAVITY-LOCKED cw-CRDS

Light possesses both a direction of propagation and a polarization of the electric field. This fact has recently been exploited by Meijer and co-workers[17] to perform polarization-dependent cavity ring-down spectroscopy on species exposed to magnetic fields. In the experiment described here, we also exploit the two mutually orthogonally components of linearly polarized light: p-polarized light (PPL) and s-polarized light (SPL). We define PPL to be light whose electric field vector lies in the plane of incidence of the RDC reflectors.

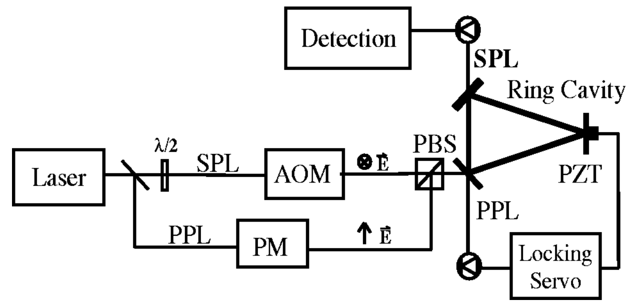

FIG. 1. cw-CRDS setup using a ring resonator locked to an external cavity diode laser source.

It is well known from the physics of optical interfaces[18] that non-normal incidence of linearly polarized light on any dielectric interface will result in different responses for PPL and SPL. Thus, any optical ring resonator constructed with a geometry using non-normal incidence reflectors (e.g., an isosceles triangle) will actually consist of two nondegenerate Fabry–Perot cavities: a p-polarization cavity (PPC), and an s-polarization cavity (SPC). Moreover, non-normal incidence dielectric mirrors typically have a lower reflectivity for PPL than they do for SPL. Consequently, a ring resonator will consist of a reduced finesse PPC, and a higher finesse SPC. The ring resonator PPC and SPC transverse mode structure remains identical, because it is determined only by geometrical considerations. PPC and PSC free spectral ranges differ by the contribution of the derivative of the mirror phase shift,[2] which was estimated to be a few tens of kHz. Moreover, the unequal phase shifts accrued during non-normal reflection of PPL and SPL will result in a fixed frequency difference between the PPC and SPC frequency responses.

Because orthogonal polarizations are easily separated with polarizing optics, the simultaneous use of PPL and SPL provides a straightforward solution for separating the cavity locking problem from the actual ring-down measurement. Constraints on locking the laser are relaxed by the use of the lower finesse PPC. Absolute wavelength accuracy at each point is determined by the PPC finesse and locking servo quality, rather than by laser linewidth or cavity mode sweep range. This improved frequency stability will enable very high resolution spectroscopy ($<$ 100 kHz) to be performed with these locked systems. Furthermore, ring cavities do not reflect light directly back into the light source, so that optical feedback problems are greatly reduced.

In this experiment, the output of a cw laser is split into two arms (cf. Figure 1): the PPL arm is used to lock the laser to a single mode of the lower finesse PPC of the ring resonator, while the SPL arm is switched on and off with an AOM to perform CRDS using the high finesse SPC of the ring resonator. The AOM also provides the appropriate frequency shift to the SPL, so that it can couple into the same transverse mode that is used to lock the PPC. Because frequency fluctuations in the laser source appear identically in both polarization arms, if the PPC mode is locked to the laser, then the SPL will consistently couple into the corresponding SPC mode. Both polarization arms can be identically mode matched to the cavity. The ring resonator presented in this paper is locked to an ECDL by means of the Pound–Drever–Hall technique.[19] The observed reduction of shot-to-shot fluctuations in the ring-down time constant, and improved baseline stability lend promise to this approach.

III. EXPERIMENTAL SETUP

A. Ring cavity cw-CRDS optical setup

The light source employed in this experiment is a commercial ECDL (New Focus: 6226-H032), tunable from 833.2 nm to 862.5 nm. Its output power varies from about 9 to 13 mW for a 60 mA drive current over its entire tuning range. Although it is possible to electronically lock the ECDL to the RDC, it was found that the laser would not reliably tune as a single-mode device because of residual back reflections from optical elements normal to the beam path. This behavior was corrected by placing a 25–35 dB isolator (New Focus: 5568) at the output of the laser head. The half-wave plate of this isolator was adjusted to obtain PPL. A Faraday cage was constructed around the laser, controller, and connecting cable to shield the laser from stray EMI that causes instability in the laser's temperature controller.

The reflectors of the ring-down cavity[20] form an isosceles triangle with a long arm length of 19.5 cm and a base length of 2.0 cm. The plano/plano mirrors of the ring (CVI:TLM2-800-45S-1037) have 99.93% reflectivity for SPL at 833 nm, and 99.3% for PPL at 833 nm. The 1 m radius of curvature, concave/plano reflector (REO: run C628, 7.75 mm, 840–880 nm, 1° wedge) had a 99.95% reflectivity at 833 nm for both polarizations. The curved mirror is glued to a piezoelectric transducer so that the cavity length can be modulated over an entire free spectral range. CVI mirror absorption and scattering losses on the order of 400 ppm/pass were measured for the PPL and SPL, using a high-precision power meter to detect the reflected and transmitted power from the mirror at 45° (i.e., for a 10 mW input at 830 nm, 9.993 mW were reflected, and 3 nW were transmitted). REO mirror absorption was quoted to not exceed 100 ppm for either polarization.

The silicon photodiode detectors are based on designs by Harb *et al.*[21] The photodetectors used to detect the transmitted and reflected PPL locking signals are designed to maximize bandwidth (90 MHz) rather than gain, to render the locking sidebands with minimal distortion and noise. Shot-noise-limited sideband signals were readily attainable for optical powers of hundreds of microwatts in the p-arm. The SPL photodetector is maximized for gain rather than bandwidth (25 MHz), because SPL cavity output light levels are low (several microwatts), and ring-down decays exceed hundreds of nanoseconds (even for the strongest absorption lines).

The optical setup is shown in Figure 2(a). All components are mounted on a vibration-isolated optical table (Newport: VW Series Workstation). It was found that the quality and duration of the locking improved dramatically with optical mount stability, and isolation from room air currents.

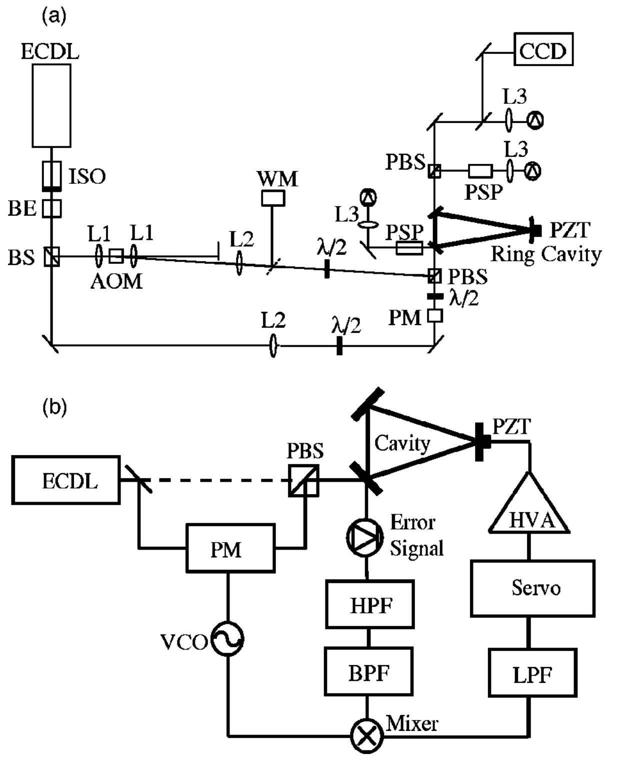

FIG. 2. cw-CRDS experimental setup: (a) optical system and (b) cavity locking system. In (a) note the following abbreviations: (AOM) acousto-optic modulator, (BE) beam expander, (BS) beam splitter, (ISO) isolator, (λ/2) halfwave plate, (L1) lens f=50 mm, (L2) lens f=1270 mm, (L3) lens f= 60 mm, (PBS) polarizing beam splitter, (PM) phase modulator, (PSP) polarization separation prism, (WM) wavelength meter. In (B) note the following abbreviations: (BPF) band-pass filter, (HPF) high-pass filter, (LPF) low-pass filter, (PM) phase modulator, (PZT) piezoelectric actuator, (VCO) voltage controlled oscillator.

The output beam of the laser isolator is first circularized before it is separated into two arms. One quarter of the light is used in the locking arm (PPL), while three quarters enter the detection arm (SPL). The locking PPL beam is mode matched to the TEM_{00} cavity mode with a 127 cm focal length lens, and travels through an electro-optic resonant phase modulator (PM, New Focus: 4001, 58.5 MHz), a polarizing cube beamsplitter and a 1.0 mm aperture, before entering the ring-down cavity. The PPL reflected from the cavity is used to lock the cavity to the laser, as described in the following section.

The detection beam is focused to a spot size of approximately 60 μm at the center of an AOM (Brimrose: GPM-400-100-960) crystal and recollimated using 5 cm focal length lenses. The AOM is driven by a voltage-controlled oscillator (VCO) with a frequency range of 300–535 MHz. The VCO rf output is modulated using a 50 kHz square wave from a function generator (SRS: DS345) and a rf switch. Thus, the AOM not only switches the SPL beam on and off, but also frequency shifts the SPL beam by the 320 MHz difference between SPC and PPC modes of the ring resonator. The first-order output passes through an aperture that blocks the zeroth order. A flipper mount may be used to direct the first-order beam into a fiber coupler for wavelength detection by a wavelength meter with 0.001 nm resolution (Advantest: TQ8325). The beam then passes through a 127

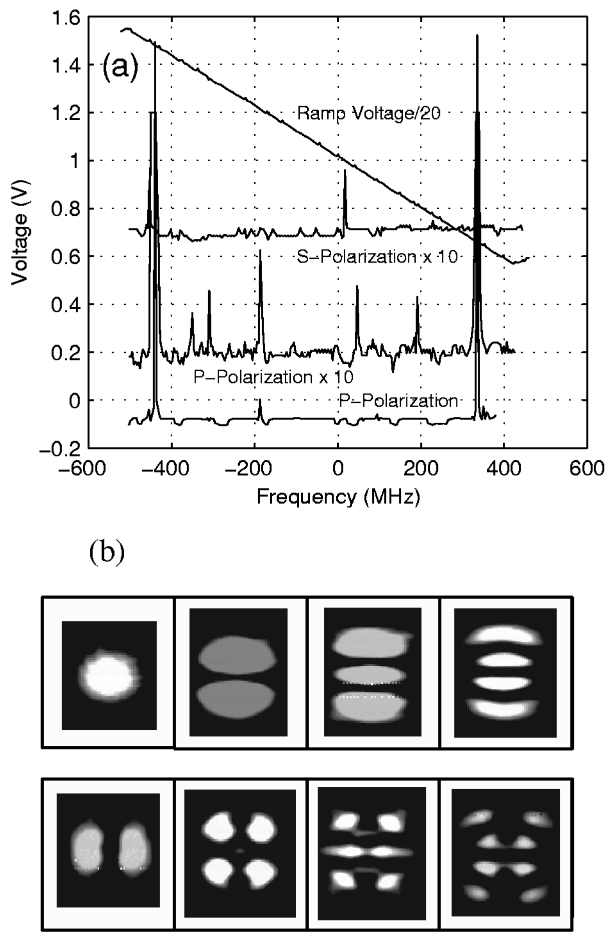

FIG. 3. (a) Frequency response of the mode cleaner cavity design obtained by sweeping the cavity length through one free spectral range, and (b) CCD images of transverse modes for unoptimized cavity alignment.

cm focal length lens for mode matching (identical to the PPL), and through a half-wave plate to convert it to SPL. It is reflected by a polarizing cube beamsplitter through an aperture before it enters the ring-down cavity.

Approximately 90% of the laser carrier light is coupled into the TEM_{00} mode of the cavity for PPL, and 85% for SPL. In both arms, the remaining light consists mainly of the TEM_{01} mode (9%) with less than 1% of the total light in the TEM_{10}, TEM_{02}, and TEM_{11} modes. The spatial coupling efficiency remained unchanged over several days. Moreover, the coupling efficiency was constant for 0.3 nm of continuous tuning of the laser wavelength. A scan of the cavity frequency response is given in Figure 3(a) where it can be seen that the SPC resonance frequencies are shifted by 282 MHz relative to the PPC, and that most of the laser light in both arms couples into the TEM_{00} cavity mode. We note that when the cavity modes overlap an absorption line of water, the total amount of dispersion between SPL and PPL changes by several tens of kHz, requiring that the SPL frequency offset be adjusted by changing the VCO frequency driving the AOM.

PPL and SPL exiting the ring-down cavity are resolved using a polarizing cube beamsplitter. PPL is further divided by a 10/90 beamsplitter with 10% going to a charge coupled device (CCD) camera to visualize the transverse mode either on a TV screen or using laser beam diagnostics software (Coherent: BeamView™), and 90% going to the transmitted p-arm detector. Cavity modes recorded on a CCD are given in Figure 3(b). For the TEM_{00} mode, a beam waist of 341 μm was measured, which is only slightly larger than the expected 328 μm TEM_{00} eigenmode waist of the ring resonator.

SPL passed through a Glan–Thomson prism to isolate the detection arm photodetector from PPL. The ring-down decay wave form issued by this detector was first amplified with a low-noise, high-speed amplifier (Stanford Research Systems: SR445) before digitization on a 10-bit, 1 GHz oscilloscope (Tektronix: 11402). The decay wave form obtained on the scope was fitted with 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, as first proposed by Naus *et al.*[22] Our effective data acquisition rate is presently limited to several hundred Hz by data transfer rates between the oscilloscope and personal computer. The oscilloscope itself can acquire and average wave forms at kHz rates.

All spectral scans were performed in room air, at atmospheric pressure and with a water vapor partial pressure of 12 Torr. After assembly, the ring-down cell's only aperture was sealed to prevent dust and particulates from entering the cavity (the only other inlet into the cavity are oversized holes in the piezo mount for electrical wires to the transducer). To obtain spectra, the ECDL was scanned by varying the piezoelectric voltage for fine frequency (0.001 nm) adjustments and the grating position for larger frequency adjustments (0.01 nm).

B. Locking using the Pound–Drever–Hall technique

The laser and cavity were locked with a technique devised by Drever, Hall and co-workers,[19] which generates an ''error signal'' proportional to the difference between the laser and the cavity line centers. In the experimental setup, shown in Figure 2(b), the optical phase modulator was driven by a VCO set to 58.5 MHz. The optical signal reflected from the ring-down cavity passed through a Glan–Taylor prism, so that only PPL arrived at the photodetector. The photodetector electrical output passed through a high-pass filter and 60 MHz band-pass filters before entering a mixer where it was heterodyned with the 58.5 MHz VCO output. The phase shift between the rf and the photodetector output was optimized by adjusting BNC cable length. The mixer output passed through a low-pass filter to generate the Pound–Drever–Hall error signal.

The error signal was then processed by a feedback servo circuit consisting of a polyimide (PI) controller and a high voltage amplifier. The piezoelectric transducer exhibited a resonance at 52 kHz which was eliminated by a twin-T passive notch filter at 52 kHz center frequency, having 48 dB maximum attenuation, and a 3 dB bandwidth of 2 kHz. The notch filter dramatically improved the feedback loop stability and sensitivity to external perturbations, such as acoustic noise. The complete servo circuit (including the notch filter) had a dc gain of 60 dB, and a 3 dB frequency of 455 Hz. The

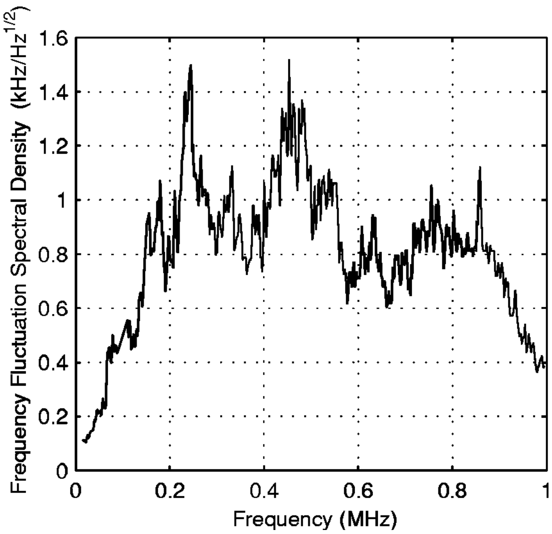

FIG. 4. Spectral density of laser-cavity frequency fluctuation response for a locked system.

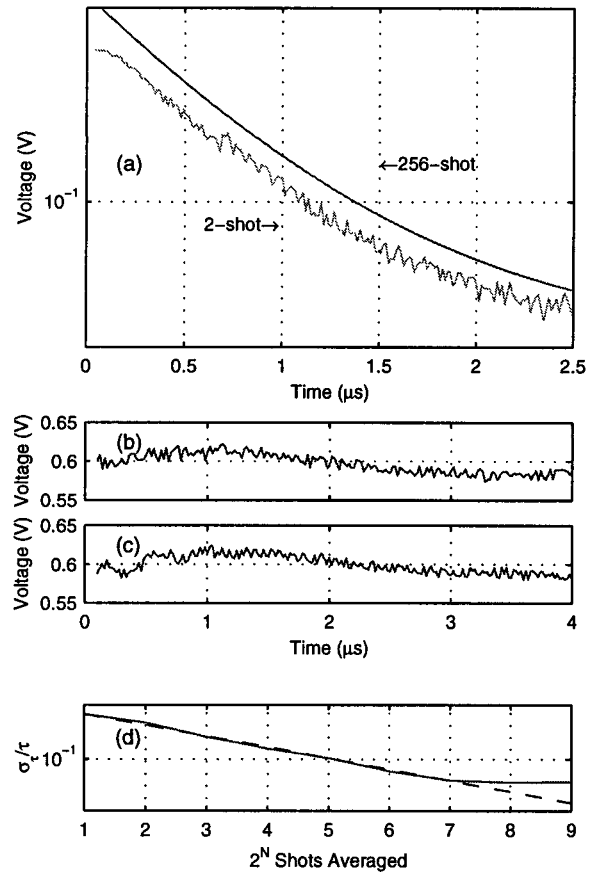

FIG. 5. (a) Ring-down decays recorded on an oscilloscope with two shots and 256 shots, (b) residual from an exponential fit of a 256-shot wave form (magnified 100 times), (c) residual from a simulated exponential decay of a low-pass filter (magnified 100 times), and (d) dependence of $\sigma_{\ln\tau}$ on the number of shots averaged (measured =solid line, predicted = dashed line).

processed error signal from the servo directly controlled the piezoelectric actuator and hence cavity length. Thus, the servo closed the loop for locking the ring-down resonator to the laser.

To lock the cavity to the laser, a manual offset on the high-voltage piezo driver was scanned until the cavity TEM_{00} mode frequency would enter the locking range of the servo loop. The cavity would then remain continuously locked to the laser for up to 10 min (typically 5 min), before outside mechanical or acoustic vibrations produced a system excursion beyond the servo range. Long-term laser-frequency drift was easily compensated with the manual offset voltage on the high-voltage amplifier, which was also used to bring the system within the locking range after larger laser wavelength changes ($>$ 30 MHz) during spectral scans.

The locked system response was monitored using either the error signal and/or the transmitted p-arm signal: when the system was locked, these signals were recorded on a signal analyzer to determine their fluctuations. By using the discriminator slope of the error signal, which converts voltage to optical frequency, fluctuations in the error signal were converted into residual differences between the laser frequency and the cavity resonance frequency. A plot of the frequency fluctuation spectral density is given in Figure 4, which shows the reduction in laser noise produced by servo locking at lower frequencies. The total jitter between laser and cavity resonances was 1.1 MHz, slightly less than the free-running laser linewidth. Laser and cavity overlap were guaranteed by the servo for the ''average'' laser line shape, i.e., the cavity line was locked to the center of the average laser line shape. A much faster and complex feedback servo would have been required to dynamically lock the cavity to the laser. The effective laser and cavity linewidth difference accounted for the significant reduction in SPL throughput. Overall, SPL suffered modematching losses of 70%, a linewidth ratio loss of 86%, and mirror absorption/scattering losses on the order of 850 ppm per pass over 3100 passes, while PPL suffered respective losses of 40%, 30%, and 850 ppm per pass over 300 passes. Thus for a 900 μW SPL input, 2.3 μW were expected at the output, and 1.9 μW were measured. Similarly, 1.0 mW of PPL was injected, 325 μW of output were expected, and 315 μW were actually obtained.

Shot-to-shot fluctuations of about 4% in the fitted decay wave form intensities at each wavelength indicated that laser-cavity coupling was sufficiently reproducible to perform CRDS. Thus, the servo adequately ensured that laser light coupled only into the TEM_{00} mode of the cavity, and that coupling occurred for every trigger of the AOM. Therefore, the maximum potential data acquisition rate was 50 kHz, the AOM on/off modulation rate. As mentioned above, our actual data acquisition rate remained limited by detection electronics to only several hundred Hz.

IV. RESULTS AND DISCUSSION

Using the locked cavity stabilization scheme described above, we were able to obtain a spectrum of water vapor present in the ambient air. Figure 5(a) shows two-shot and averaged SPL ring-down wave forms. Figure 5(b) presents the residual obtained after fitting an exponential decay to the averaged wave form. Single-shot decays exhibited high frequency intensity noise whose phase was random with respect to the AOM (and oscilloscope) triggering signal. Hence, this noise was reduced to less than 0.05% of the decay wave form intensity by averaging 64 or more decay wave forms. Typically, 256 shots were averaged using the digital oscilloscope to obtain an averaged decay wave form (ADW), and to ensure that the only noise remaining on the ADW was scope quantization noise. Possible sources for this high frequency noise include amplifier noise and pickup, high-voltage amplifier and servo loop noise, as well as imperfectly compensated piezoelectric transducer electrical and mechanical resonances.

Although averaging greatly reduced the amplitude of high frequency residuals in the decay wave form, it enhanced a low frequency, quasi-sinusoidal residual [seen in Figure 5(b)] whose phase remained fixed with respect to the triggering signal. It was found that the amplitude and period of this residual depended only on the intensity and period of the triggering signal. In fact, this residual could be reproduced exactly by fitting artificially generated (low-pass filter on a square wave output of a function generator) decay wave forms to an exponential decay, as shown in Figure 5(c). Thus, it was finally determined that this regular residual was caused by input channel cross-talk within the digital oscilloscope. During subsequent data analysis, this reproducible residual was subtracted from the averaged ring-down decay wave forms prior to exponential decay fitting.

The dependence of $\sigma_{\ln\tau}$ on the number of shots averaged, N, is given in Figure 5(d), and shows a clear $\sqrt{N}$ dependence up to 64 shots. When more than 64 shots were averaged, the overall noise on the decay wave form could not be further reduced, and was observed to be limited by digitization noise imposed by the oscilloscope. Single shot $\sigma_{\ln\tau}$ was less than 4×10^{-3} and could be reduced to less than 5×10^{-4} for 64 shot ADWs which is comparable to recent results by Romanini *et al.*[23] It should be noted that $\sigma_{\ln\tau}$ for a given series of shots never exceeded the fit uncertainty in each shot by more than 5%. However, to guarantee that all wave forms were indeed digitization limited, 256 shot ADWs were used, even though they did not result in any sensitivity gain over 64 shot ADWs. Ten ADWs were then recorded at each wavelength. Each spectral element was taken to be an average of 7–10 ADW ring-down decay constants, because ADWs that showed a significant decrease in intensity when the apparatus was perturbed by external vibrations during the experiment were discarded. The $\sigma_{\ln\tau}$ of averaged decay constants was 2×10^{-4}. The noise producing these variations in the ADW decay constant was primarily excess technical noise in the detection electronics and not oscilloscope quantization noise, so that under true shot-noise-limited detection conditions, these decay constant variations could be further reduced.

Figure 6 shows an absorption spectrum of water vapor present in ambient room air. The resolution of the reported spectrum is 0.002 nm, except about the absorption peaks where resolution was increased to 0.001 nm. Ten ADWs were recorded at each wavelength. Each spectral element represents an average of 7–10 ADW ring-down. The rms baseline noise of 5×10^{-9} cm^{-1} in our spectra is comparable to that initially reported by Romanini *et al.*,[7] and is comparable to most pulsed CRDS studies.[1–3,24] It exceeds,

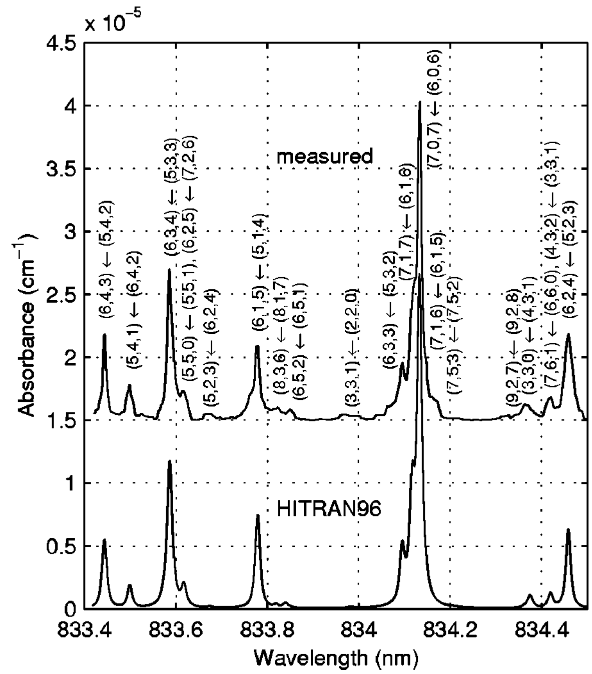

FIG. 6. Spectrum of water vapor in room air obtained using a ECDL locked to the ring-down cavity. Spectrum based on HITRAN96 is shown in dashed lines. Weakest lines occur at 833.206 nm and 833.674 nm. Transitions are labeled as $(J',K_a',K_c')\leftarrow(J,K_a,K_c)$.

however, the baseline noise reported by Romanini *et al.*[8] for the same model ECDL by a factor of 25 because not only was our cavity half the length of that used by Romanini *et al.*, but our mirrors had only a 99.93% reflectivity compared to the 99.999% reflectivity used by Romanini *et al.*, i.e., our empty cavity losses were 70 times larger.

By locking the RDC to the laser source, the shot-to-shot fluctuations in decay constant were reduced by a factor of almost 100 compared to a free-running, multi-mode laser diode system.[11] Shot-noise-limited detection was precluded by the quality of our detectors, by the laser linewidth, and by the limited SPC throughput. This illustrates, in fact, the tradeoff involved when mirror reflectivity is used to enhance sensitivity. While higher mirror reflectivity increases the empty cavity decay constant, thereby making detection of relative changes in the decay constant easier (slower detector and electronics can be used), the resulting decrease in the intensity of the detected signal limits the signal-to-noise ratio on the output decay wave forms. The increased noise on the decay wave forms leads to greater uncertainty in the fitted exponential, and hence greater uncertainty in the decay constant, which decreases sensitivity. Perhaps the use of a very narrow linewidth laser such as a cw Nd:YAG laser would allow very sensitive, shot-noise-limited measurements to be made, owing to their extremely narrow linewidths (<100 Hz).

Our spectra compare very favorably, in absolute frequency, line strength, and linewidth, to those generated by the HITRAN96 database[25] (http://www.hitran.com). The lines presented here belong to rotation transitions within the $3\nu_1+\nu_2$, $2\nu_1+\nu_2+\nu_3$, and $\nu_1+3\nu_2+\nu_3$ vibrational overtone bands.[26] The weakest peaks, (recorded in HITRAN96), that are measurable with our system are located at 833.674 nm and 833.206 nm, and correspond to line strengths of 4.126×10^{-26} cm^{-1}/(molecule/cm^2) and 1.259×10^{-26} cm^{-1}/(molecule/cm^2), respectively. These are somewhat weaker than previously reported water lines recorded with a 45.7 cm long linear cavity mirrors having $R=99.97\%$.[11] The nominal sensitivity of this system to water vapor is ppm at 1 atm pressure.

V. CONCLUSIONS

In this contribution we demonstrate that it is possible to use a single-beam, dual-arm approach to CRDS to continuously lock a narrow-band external-cavity diode laser to a high-finesse resonator with one arm, while performing CRDS on the locked system using a second, orthogonally polarized arm. By using a ring resonator as the cavity ring-down cell, the two orthogonal polarizations see different mirror reflectivities at non-normal incidence and, therefore, a single resonator can be simultaneously used as a low-finesse resonator for laser locking, and as a high-finesse resonator for CRDS. By frequency-shifting one arm of a split laser beam with an acousto-optic modulator and then rotating its polarization, the two arms of the laser beam (one s- and the other p-polarized with respect to the ring-down cavity mirrors) can become simultaneously resonant in the cavity. In this manner, the cavity may remain continuously locked to the laser output, ensuring that both arms will couple efficiently into a single mode of the cavity, while the light in one polarization is switched on and off to perform decay time measurements. Presently, this measurement technique remains limited by cavity throughput, so that determination of the ring-down decay constant is limited by technical noise in the photodetectors.

We have performed cw-CRDS using such a cavity-locked system achieving ring-down repetition rates of 50 kHz, and a baseline noise level on the order of 10^{-9} cm^{-1}. We have obtained combination overtone spectra of water vapor in the ambient environment between 833.4 and 834.5 nm, with nominal sensitivities of 5 ppm at 1 atm. This sensitivity can be dramatically improved by choosing a stronger absorption band of water vapor (such as 1.3647 μm), a narrower linewidth laser source (e.g., a cw IR-OPO) and allowing the use of better reflectors and better designed photodetectors. The realization of these improvements, leading to shot-noise-limited measurements with estimated noise levels of 10^{-12} cm^{-1} to even 10^{-13} cm^{-1} and spectral resolution on the order of a hundred Hz, may make CRDS one of the most sensitive, high-resolution spectroscopic tools.

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