Effects of incomplete light extinction in frequency-agile, rapid scanning spectroscopy

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ABSTRACT

The effect of finite beam extinction ratio on the precision and accuracy of cavity ring-down decay time constant measurements was examined using the frequency-agile, rapid scanning, cavity ring-down spectroscopy (FARS-CRDS) technique. This new approach to CRDS uses a waveguide-based electro-optic phase modulator (EOM) to provide a laser beam extinction ratio as high as 80 dB: a value that is ≈30 dB greater than that typically achieved with acousto-optic-modulator-based beam switches. We find that the observed measurement precision scales inversely with extinction ratio, such that an EOM enables measurement of the cavity ring-down decay time with a relative precision of $\approx 8\times10^{-5}$. We demonstrate that insufficient extinction can be the dominant cause of statistical uncertainty for extinction ratios below 60 dB. Furthermore, insufficient extinction can result in non-exponential decays, which cause systematic measurement biases in cavity losses and absorption.

Keywords: Rapid scanning, high bandwidth, cavity ring-down spectroscopy, optical extinction, electro-optic modulator.

1. INTRODUCTION

Cavity ring-down spectroscopy (CRDS) enables ultra-sensitive measurements of molecular absorption with relatively simple instrumentation [1-3]. In a typical CRDS experiment, a gas sample of interest is placed within an optical cavity comprising two highly reflective mirrors. Laser radiation is then injected into the optical cavity and allowed to reach a threshold intensity at which point the incoming radiation is extinguished by means of a fast optical switch. The decay of optical power exiting the cavity is then measured by an external photo-receiver. For single-mode excitation of the ring-down cavity, the optical power decays exponentially with a time constant, τ, that is directly related to the absorption coefficient of the analyte. Unfortunately, while CRDS allows for quantitative measurements of weakly absorbing systems, spectrum acquisition is generally slow due to the thermal or mechanical frequency tuning of the laser source.

We have recently demonstrated an alternative, high-bandwidth approach in which frequency scanning of the probe laser is realized through the use of a waveguide-based electro-optic phase modulator (EOM). We refer to this method as frequency-agile, rapid scanning spectroscopy (FARS) [4]. The EOM is driven by a microwave source at a modulation frequency, ν_m, thus generating a series of regularly spaced sidebands about the laser carrier frequency. The optical cavity is then used to reject (*i.e.*, reflect) all but a single selected sideband which is then used to probe the gas analyte. This approach enables stepwise scanning rates exceeding 8 kHz for high finesse optical resonators and is limited only by the cavity response time itself [4]. In this scheme, ring-down decays are initiated by switching off the microwave drive frequency to the EOM, which removes the selected sideband frequency from the probe laser. This approach differs from traditional cw-CRDS in which an acousto-optic modulator (AOM) is utilized to switch off the probe laser beam.

Here we have examined the effects of finite extinction in the EOM on FARS-CRDS measurement statistics. This work is motivated by a recent study that identified finite extinction in AOMs to be a dominant noise source in many traditional cw-CRDS experiments [5].

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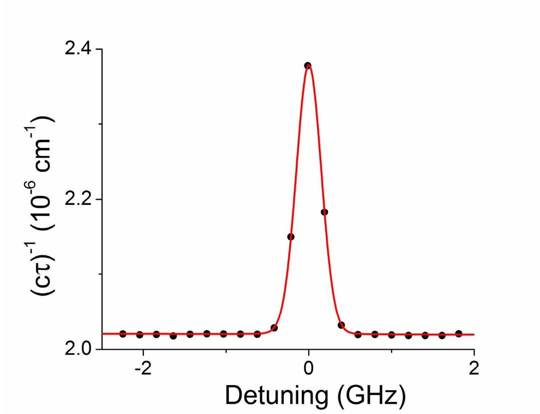

Figure 1. Example spectrum of the (31112)←(01101) *P*7e CO_2 transition located at 6350.6624 cm^{-1} measured with FARS-CRDS. The corresponding fitted Voigt line profile is also shown. This weak hot band transition has an intensity of only 3.89×10^{-25} cm molecule^{-1} [6]. It was recorded with a pure CO_2 sample at a pressure of 44 Pa and exhibited a signal-to-noise ratio of ≈500:1. This 4.2 GHz wide spectrum can be recorded in times as short as ≈2.6 ms.

2. RESULTS AND DISCUSSION

Measurements were made using the Pound-Drever-Hall-locked, FARS-CRDS spectrometer at the National Institute of Standards and Technology (NIST) in Gaithersburg, MD. This instrument will be described in more detail in a forthcoming publication [7]. An example spectrum can be found in Figure 1, in which a weak near-infrared CO_2 transition was recorded. In addition, a schematic of the instrument can be found in Figure 2. Briefly, the laser beam is separated into two legs: a probe leg and a locking leg. The two legs have orthogonal polarizations and can thus be readily separated. The locking leg is utilized to Pound-Drever-Hall [8] offset frequency lock the probe laser to the optical cavity. Generally, the ring-down decays are initiated through the use of a high speed reflective microwave switch with an extinction of >80 dB and a switching speed of <20 ns. The measured optical power extinction ratio has been shown to exceed 70 dB. For the present measurements, the microwave switch utilized to initiate the ring-down decays was replaced with the components shown in Figure 2 in order to produce a switch whose extinction can be readily controlled. This switch is similar to the one found in Ref. [5].

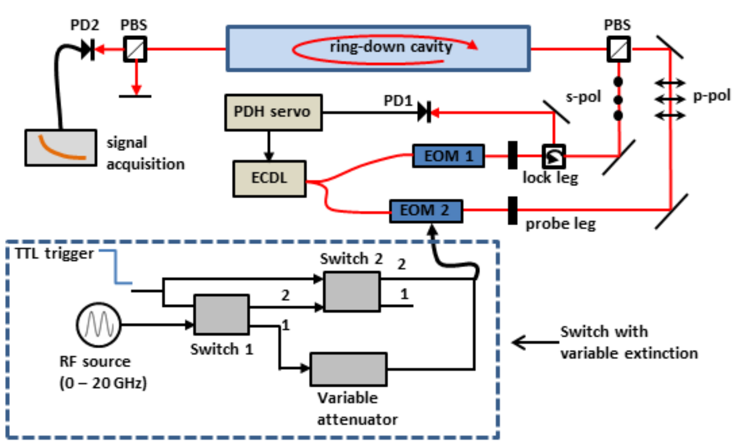

Figure 2. Schematic of the Pound-Drever-Hall (PDH) locked, FARS-CRDS spectrometer. A microwave switch with variable extinction was constructed in order to quantify the effects of finite extinction on the instrument's sensitivity. The shown components include photodiodes (PD1, PD2), polarizing beam splitters (PBS), external-cavity diode laser (ECDL), and fiber waveguide-based electro-optic phase modulators (EOM1, EOM2).

In an analysis of cw-CRDS experiments, Huang and Lehmann showed that residual leakage of the probe laser beam through an optical switch interferes with the decaying field, thus affecting the statistics and functional form of the ring-down decay signal [5]. To investigate this phenomenon in our system, we recorded 1000 ring-down decays and fit single-exponentials to these data for power extinction ratios, 10 log(r), between 11 and 77 dB. The results are summarized in Figure 3 where we present the relative variances [$(\sigma_\tau/\tau)^2$] for our measurements (circles) and those of Huang and Lehmann [5] (triangles) . We note that the present results correspond to a probe laser that is phase-locked to the ring-down cavity with a relative line width of ≈130 Hz [7], whereas those of Huang and Lehmann were obtained with a distributed-feedback (DFB) diode laser which has a Lorentzian spectrum with a width of a few MHz. This distinction is important as shown below, because the excitation spectrum affects the resulting interference between the decaying field and leakage field, and hence the dependence of the statistics on extinction ratio.

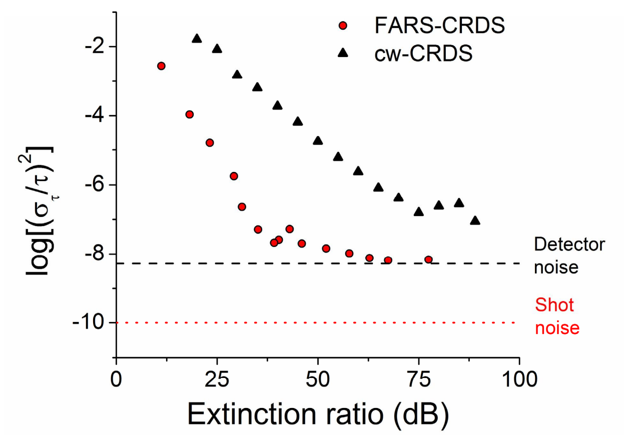

Figure 3. Relative variance [$(\sigma_\tau/\tau)^2$] as a function of extinction ratio for FARS-CRDS (present study) and traditional cw-CRDS [5]. Note the logarithmic scale. For the shown data the uncertainties in the ordinate values is smaller than the symbols, whereas the uncertainties in the abscissa values is ≈1 dB. For the present study, the cavity is pumped by a laser that is phase locked to the ring-down cavity resonance, whereas in traditional cw-CRDS, the cavity is pumped by an unlocked DFB laser with a spectrum bandwidth that is much wider than the ring-down cavity line width. In both cases, as the extinction is increased, $(\sigma_\tau/\tau)^2$ decreases until it reaches a noise limit. In the FARS-CRDS experiment this limit was the detector noise, whereas in the traditional cw-CRDS experiment it was a combination of detector noise and cavity drift.

For the DFB cavity excitation, interference between the two fields (corresponding to the leaking probe laser and the decaying field, respectively) is influenced by random phase fluctuations in the former. This effect adds noise to the decay signal, which is manifest by statistical variations in the fitted decay time constants. However in the present FARS-CRDS case, the long coherence time of the leakage field (relative to τ) does not introduce substantial phase noise into the decay signal. Indeed for large extinction ratios, our fit residuals are dominated by the detector noise. We note that as the extinction ratio is reduced below ≈50 dB, we see the emergence of systematic, non-exponential signals (see Figure 4) which are observable due to the long coherence time of the PDH-locked laser. Nevertheless, as the non-exponential character of the signal increases with reduced extinction ratio, the fit statistics exhibit increased variation, which leads to decreased measurement precision. For both cases shown in Figure 3, the relative variance in τ decreases with extinction ratio and approaches a noise limit at high extinction ratios, although our values exhibit a far steeper slope. Furthermore, for an extinction ratio of ≈70 dB our measurements attain a minimum relative variance in τ that is about ten times less than that of Huang and Lehmann [5].

In order to quantify biases in measurements of decay time constants caused by leakage from an idealized optical switch, we modeled the ring-down signal, $y(t)$, caused by the sum of two interfering fields exiting the cavity: a dominant decaying component and a weak cw component (*i.e.* the leak-through field). Assuming that the switch occurs at time t=0 and considering only slow dependence (relative to τ) for the leak-through beam intensity, we find

$$y(t) = y_0 + A[e^{-t/\tau} + 2\sqrt{I_l(t)/I_d}\,e^{-t/(2\tau)} + I_l(t)/I_d] \qquad (1)$$

where, $I_l(t)/I_d$ is ratio of the leak-through beam intensity to the decaying field intensity, y_0 is a constant baseline signal, and A is the product of photo-receiver responsivity and initial decay signal intensity, I_d. Note that the power extinction ratio, r, is given by $I_d/I_l(0)$. Assuming a constant leak-through intensity, this model predicts bi-exponential decays that have components at τ (the infinite extinction time constant) and 2τ. Single exponential decays were fitted to simulated signals (assuming constant I_l), and the fractional deviation of the fitted time constant relative to the simulated value, $\Delta\tau_f/\tau$, was calculated as a function of extinction ratio. Here $\Delta\tau_f = \tau_f - \tau$, where τ is the infinite extinction time constant and τ_f is the time constant that results when a single exponential decay is fitted to the bi-exponential curve given by Eq. 1. This quantity represents a systematic bias in the measured time constant.

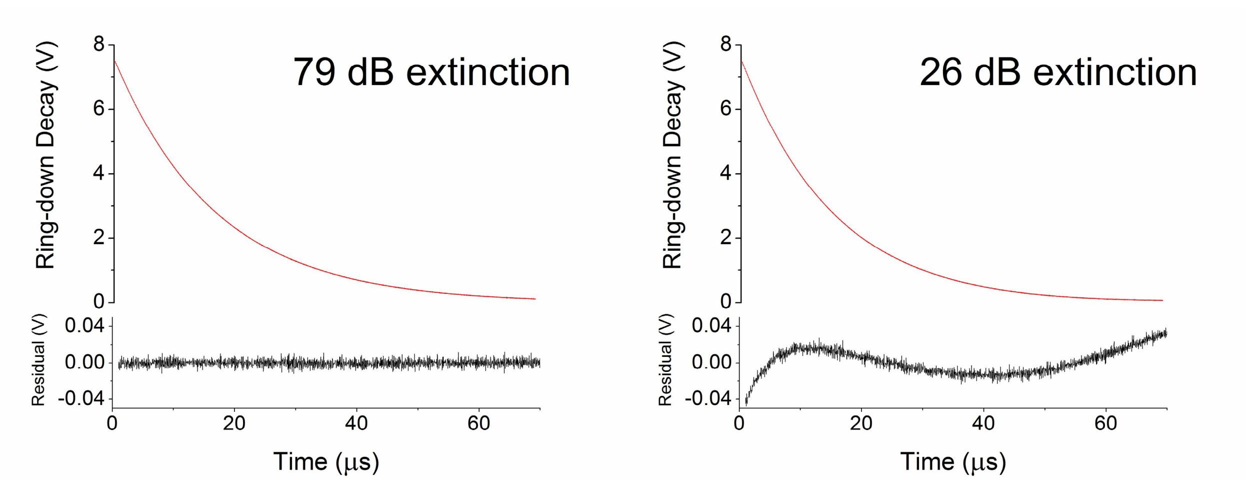

Figure 4. Measured ring-down decay curves and fit residuals (fit – measurements) from a single-exponential fit for two different extinction ratios. Three parameters were floated in the fits: baseline, decay time, and signal amplitude at t=0. The left panel was recorded at an extinction ratio of 79 dB and exhibits a maximum signal-to-noise ratio of ≈2000:1. The right panel was recorded at an extinction ratio of 26 dB and clearly exhibits non-exponential behavior because of poor extinction. This distortion leads to a fitted ring-down time constant which is ≈10% less that recorded at high extinction.

As shown in Figure 5, our numerical simulations reveal that significant bias in τ can occur when bi-exponential decays caused by the light leakage are fitted with a simple single-exponential function. Specifically, we find that ($\Delta\tau_f/\tau$) is nominally proportional to $r^{-1/2}$, with a 0.36% bias in τ for an extinction ratio of 50 dB. We emphasize that unlike the measurements discussed in Figure 3, this calculation captures systematic deviation in time constant determination caused by the non-exponential character of the ring-down signal. Because this effect is systematic, the uncertainty cannot be reduced by signal averaging. However, we note that the non-exponential fit residuals shown in Figure 4 are not bi-exponential, suggesting that residual time dependence in I_l also needs to be taken into account. This may be caused by non-idealities in the microwave switch or variations in the leaking field amplitude. We believe that these non-idealities are the cause of the large biases that we observe in our ring-down decays at very low extinction values (see Figure 6).

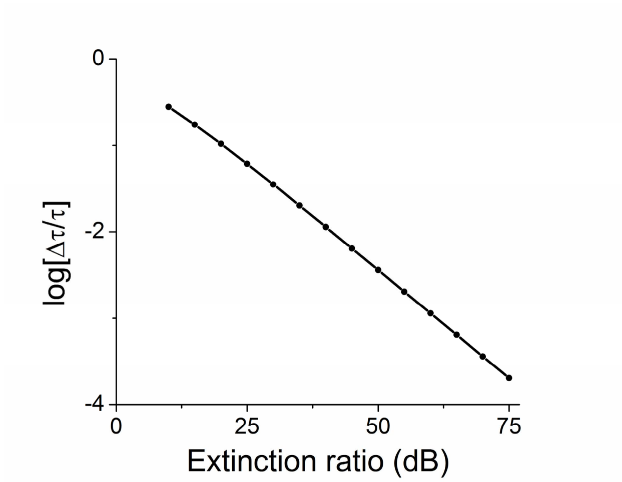

Figure 5. Calculated relative bias in measured τ caused by fitting a single exponential function to a bi-exponential decay signal according to Eq. 1. The calculations correspond to a fitting window of approximately 5τ.

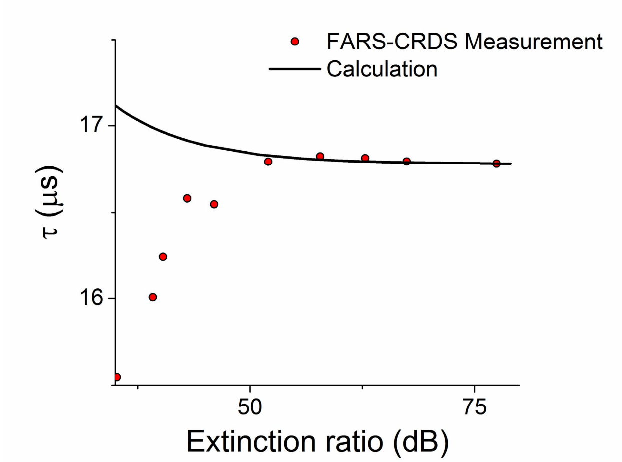

Figure 6. Measured and calculated bias in the fitted ring-down time constant, τ, due to incomplete extinction of the probe laser beam. At low extinction levels such as 11 dB the observed ring-down decay curves become non-exponential, resulting in deviations from the true optical cavity time constant of as much as 25%. While the model presented in Eq. 1 captures the observed behavior at high extinction, it diverges from the measurements for extinction ratios below 50 dB. We attribute this divergence to either non-idealities in the optical switching or fluctuations in the leak field amplitude.

3. CONCLUSIONS

We have demonstrated that extremely high extinction ratios (up to 80 dB) can be achieved in FARS-CRDS measurements through the use of a high-bandwidth, microwave-switched, electro-optic phase modulator. This high extinction ratio is comparable to that achieved in previous cw-CRDS experiments that use AOMs [5] or semiconductor optical amplifiers [9] as optical switches. With regard to the fitted time constant, we can achieve a relative precision of 8×10^{-5} at high extinction ratios, and are able to observe systematic biases caused by non-exponential decays. These biases should be extremely relevant to CRDS experiments where high fidelity line shapes are required, such as in optical determination of the Boltzmann constant [10] and measurements of isotopic ratios [11].

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