3 July 1998

Chemical Physics Letters 290 (1998) 335–340

CHEMICAL PHYSICS LETTERS

# Optical heterodyne detection in cavity ring-down spectroscopy

M.D. Levenson [a,b], B.A. Paldus [a,c], T.G. Spence [a], C.C. Harb [a,c], J.S. Harris Jr. [a,c], R.N. Zare [a,1]

[a] Department of Chemistry, Stanford University, Stanford, CA 94305-5080, USA
[b] M.D. Levenson Consulting, 19868 Bonnie Rige Way, Saratoga, CA 95090, USA
[c] Department of Electrical Engineering, Stanford University, Stanford, CA 94305-4070, USA

Received 30 December 1997; in final form 27 April 1998

**Abstract**

Polarization-selective optical heterodyne detection is shown to enhance the practical sensitivity of cavity ring-down spectroscopy. Initial experiments demonstrate a signal-to-noise ratio above 31 dB. Minor improvements should yield shot-noise-limited operation. © 1998. Published by Elsevier Science B.V. All rights reserved.

## 1. Introduction

Cavity ring-down spectroscopy (CRDS) is a very sensitive technique that has been used with pulsed lasers for quantitative spectroscopy of trace and weakly absorbing gas-phase species. In CRDS, a high-finesse resonator is excited by an input beam that terminates abruptly. The exponential decay time of the flux exiting the resonator is measured after the excitation terminates [1]. The inverse of the power decay time $\tau$ is linearly proportional to the total optical losses in the resonator, which can include absorption by gas-phase species:

$$\frac{1}{\tau} = \frac{(L + A)}{t_{rt}}, \qquad (1)$$

where $L$ is the round-trip (fractional) loss in the empty resonator, $A$ the absorption of the medium enclosed by the resonator cavity, and $t_{rt}$ the round-trip time of light in the resonator. Plotting $1/\tau$ as a function of wavelength yields an absorption spectrum of the species in the resonator provided that the cavity losses $L$ are constant. Advantages of CRDS over traditional absorption spectroscopies include an insensitivity to intensity fluctuations of the light source, a dramatic increase in effective pathlength, and the convenience of detecting well-collimated coherent radiation.

The cavity ring-down technique was originally developed for the characterization of low-loss mirrors [2,3]. These early studies employed lower-power CW laser sources to build up radiation in an external ring-down cavity. An optical switch could then used to rapidly deflect the laser light entering the cavity, which allows the ring-down time of the cavity to be measured [2]. Recently, several researchers have returned to the use of CW lasers in CRDS to build up light in a ring-down cavity, taking advantage of the narrower linewidths of these

[1] E-mail: rnz@leland.stanford.edu


PII: S0009-2614(98)00500-4

sources to excite only one spatial cavity mode [4–7]. For example, Paldus et al. [6] reported a method in which a three-mirror ring resonator was locked to an external cavity diode laser (ECDL), greatly reducing fluctuations caused by feedback and laser mistuning. Despite the use of very low-noise photodetectors, the major source of noise in the observed single-shot decays was ''technical noise'' introduced by the detection electronics. Indeed, this problem is common to all CRDS systems because a portion of the desired exponential signal always becomes obscured by instrumental noise, regardless of any increases in either the initial optical power or decay duration. In this letter, we report an optical heterodyne detection technique that greatly enhances the power in the detected signal, thereby reducing the impact of electronic noise. The heterodyne technique we use consists of beating together two light sources of slightly different frequencies, having orthogonal linear polarizations. One polarization carries the cavity ring-down signal; the other polarization serves as a local oscillator. Whenever there is sufficient power in the local oscillator, true shot-noise-limited measurements can be obtained, regardless of the strength of the ring-down signal itself [8].

## 2. Optical heterodyne detection

In optical heterodyne detection, the desired signal amplitude, $\boldsymbol{E}_{\rm S}$, is made to overlap spatially with a stronger wave, the local oscillator, $\boldsymbol{E}_{\rm LO}$, on a photodetector. When the local oscillator (LO) and signal optical frequencies differ by $\delta\nu$, the total detector current, $I_{\rm D}$, which is proportional to the absolute square of the total field amplitude, can be separated into three terms, one of which oscillates at the difference frequency:

$$\begin{aligned} I_{\rm D} &\propto |\boldsymbol{E}_{\rm S} + \exp(-i(2\pi\delta\nu t + \Phi))\,\boldsymbol{E}_{\rm LO}|^2 = |\boldsymbol{E}_{\rm S}|^2 + |\boldsymbol{E}_{\rm LO}|^2 + 2\,{\rm Re}\left[\exp(-i(2\pi\delta\nu t + \Phi))\,\boldsymbol{E}_{\rm LO}^{*}\cdot\boldsymbol{E}_{\rm S}\right] \\ &\propto I_{\rm S} + I_{\rm LO} + |I_{\rm H0}|\cos(2\pi\delta\nu t + \Phi)\,. \end{aligned} \tag{2}$$

The three terms at the right of the equality sign are individually proportional to the corresponding terms on the right of the last proportionality sign. The final term $I_{\rm H}(t) = |I_{\rm H0}|\cos(2\pi\delta\nu t + \Phi) \propto 2\,{\rm Re}[\exp(-i(2\pi\delta\nu t + \Phi))\boldsymbol{E}_{\rm LO}^{*}\cdot\boldsymbol{E}_s]$ is the heterodyne current which is extracted from $I_{\rm D}$ with analog or digital electronics. The direct detection current previously employed in CRDS, $I_{\rm S}(t) = I_{\rm S0}\exp(-t/\tau)$, is much less than $|I_{\rm H}|$ when $|\boldsymbol{E}_{\rm LO}| \gg |\boldsymbol{E}_{\rm S}|$. Under ideal conditions, the heterodyne amplitude is $|I_{\rm HO}| = 2(I_{\rm LO} I_{\rm S})^{1/2}$, and the heterodyne current signal is:

$$I_{\rm H}(t) = \begin{cases} 2(I_{\rm LO} I_{\rm S0})^{1/2}\cos(2\pi\delta\nu t + \Phi) & t<0 \\ 2(I_{\rm LO} I_{\rm S0})^{1/2}\cos(2\pi\delta\nu t + \Phi)\exp(-t/2\tau) & t>0 \end{cases}, \tag{3}$$

where the phase, $\Phi$, depends on the difference between the local oscillator and the signal optical path lengths, and any transient behavior at the $t=0$ turn-off time has been ignored. The exponential decay time of the heterodyne current (which is linear in the optical amplitude) is twice that of the direct-detection ring-down power signal $I_{\rm S}(t)$.

The initial ringdown amplitude is set by the conditions existing when the driving field is turned off, and any fluctuations that this amplitude may contribute can be normalized out. When receiver noise at baseband and at the difference frequency is absent from the local oscillator beam (e.g. frequency filtering by a high finesse resonator), the electronic power signal-to-noise ratio, $S/N$, for heterodyne detection depends on the heterodyne signal $I_{\rm H}(t)$, the noise power of the detection system, $\eta\Delta\nu$, and the shot noise on the local oscillator photocurrent (or power), $2eI_{\rm LO}\Delta\nu R$, where $R$ and $\Delta\nu$ are the impedance and bandwidth of the data collection system, respectively:

$$S/N = \frac{\langle I_{\rm H}^2\rangle R}{\eta\Delta\nu + 2eI_{\rm LO}R\Delta\nu} = \frac{2I_{\rm LO}I_{\rm S}R}{\eta\Delta\nu + 2eI_{\rm LO}R\Delta\nu}\,. \tag{4}$$

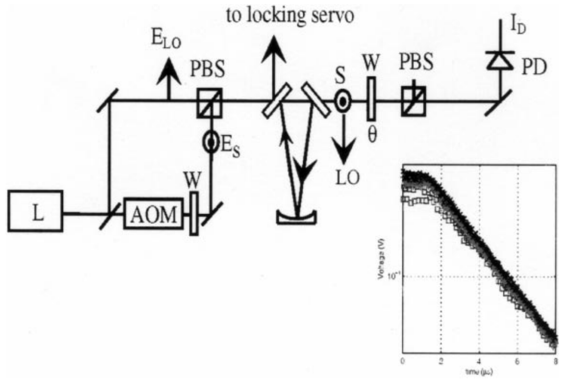


Fig. 1. Apparatus for heterodyne detected cavity ring-down spectroscopy. The ring-down cavity is described in Ref. [7]. The inset shows three successive traces of the RF current amplitude on a linear scale. The exponential decay is twice that seen by direct detection.

The electrical amplitude signal-to-noise ratio is the square root of the quantity in Eq. (4). When $eI_{LO}R \gg \eta$, $S/N$ becomes independent of the local oscillator, reaching the shot noise limit for the ring-down current:

$$S/N = I_s/(e\Delta\nu)\,. \quad (5)$$

Recently, Paldus et al. [7] reported a CW-CRDS system in which a single mode of a high-finesse ring cavity was locked to an external cavity diode laser (ECDL) while ring-down decays were simultaneously measured. In the heterodyne CW-CRDS system reported here, the p-polarized light (PPL) used to lock the cavity to the laser serves as the LO, and is mixed with the weaker s-polarized light (SPL) that bears the actual ring-down signal. The locked CW-CRDS system shown in Fig. 1 is described elsewhere [7].

## 3. Experimental method

Briefly, the output of a 10 mW ECDL is split into two orthogonally-polarized beams: PPL is used to lock one $TEM_{00}$ cavity mode to the ECDL; SPL is frequency shifted and chopped (40 kHz) with an acousto-optic modulator (AOM) and is used to excite another $TEM_{00}$ mode for the ring-down measurement. The ring resonator behaves like two decoupled resonators which share the same physical length. To achieve simultaneous resonance in both polarizations, the AOM must shift the SPL beam by the $TEM_{00}$ frequency difference, $\delta\nu \approx 282.7$ MHz.

The optical heterodyne signal was obtained by projecting part of the s- and p-polarized outputs of the cavity into a single linear polarization [9] using the birefringent wave plate, W, and the polarization beam-splitter, PBS, shown in Fig. 1. Adjusting the wave plate varies the fractions of the s- and p-polarization amplitudes that reach the detector, and the maximum heterodyne signal level is obtained when half of each is in the detected polarization. Since both waves are emitted from $TEM_{00}$ modes of the same cavity, optimal spatial overlap of the LO and signal is automatic. The cavity also filters out amplitude fluctuations of the PPL LO beam, but servo instability can introduce low-frequency variations in the relative phase $\Phi$.

The photodetector consisted of an EG&G Inc. FFD-60 silicon PIN photodiode operated at 70 V bias, and an AC-coupled broadband transimpedance amplifier. An RF spectrum analyzer (HP:8590A) was used (in the linear-scaling sample-detection mode, with zero span and 3 MHz resolution and video bandwidth) as an RF detector. The center frequency was set to 282.7 MHz and the AOM frequency fine-tuned to maximize the beat signal with both waves incident on the cavity. The video output of the spectrum analyzer produced a voltage proportional to the RF heterodyne amplitude, $(I_{LO}I_{S0})^{1/2}\exp(-t/2\tau)$. That voltage was digitized and averaged using a 10-bit digital oscilloscope (Tektronix:11402), and subsequently relayed to a computer for analysis. Because the heterodyne phase $\Phi$ fluctuates owing to transient laser-cavity detuning, mixing the photodetector current with an RF local oscillator derived from the AOM electronics proved less reliable as a detection scheme.

## 4. Results and discussion

The inset in Fig. 1 shows three successive two-shot traces of the heterodyne detected ring-down current, obtained using a logarithmic scale on the signal analyzer, as measured on the oscilloscope. The traces were obtained at an optical LO power of 150 μW, and an initial ring-down power of 0.9 μW at the detector. The exponential decay time of 2.200 ± 0.002 μs is twice that observed under the same conditions by direct detection [7]. The plateau for $t<0$ permits data normalization so that fluctuations in $(I_{LO} I_{S0})^{1/2}$ can be effectively suppressed. The inset in Fig. 2 shows a 36 dB signal-to-noise ratio for this plateau for a 1 MHz resolution bandwidth. The baseline noise power at −72 dBm is due entirely to the photodetector system; at this LO power, the shot noise level would be ∼ 15 dB lower.

Fig. 2 compares a 256 trace-averaged heterodyne-detected ring-down decay with a 256 trace-averaged direct-detected decay on a semi-logarithmic plot, where the time scale of the former has been halved to equalize the slopes. The square of the heterodyne current signal is proportional to the direct detection signal, and has the same decay time $\tau$. The ring-down decay obtained via direct detection exhibits linearity over three ring-down lifetimes before becoming obscured by background photocurrent. The heterodyne power is linear over six lifetimes before plateauing at the receiver noise level of the system. Since the photodetection noise $\eta\Delta\nu$, is relatively constant in time, its level could be subtracted out, extending the linear regime. However, because of their wide dynamic range, the heterodyne signals exhibit quantization noise caused by the limited (10-bit) resolution of the digital oscilloscope. The heterodyne detection was limited by the scope quantization when more than 8 traces were averaged, compared to the direct detection case, where it was necessary to average 64 traces before detection became scope limited.

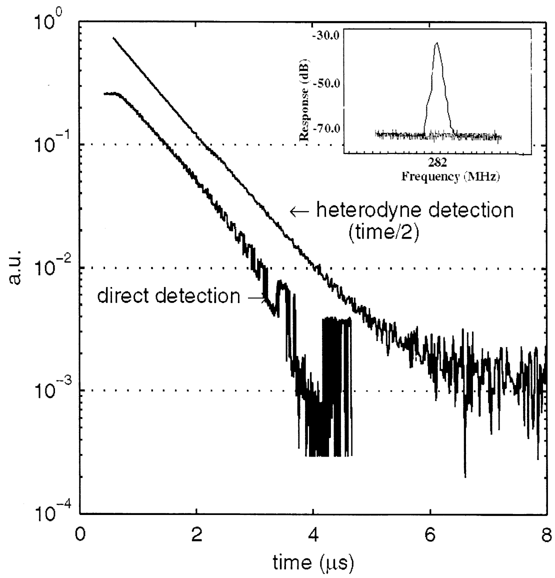


Fig. 2. Comparing the averages of 256 ring-down decays for direct detection, and heterodyne detection, with the time scale of the latter trace halved, to equalize decay times. The 10-bit digital oscilloscope shows least-significant-bit error at the bottom. The inset shows signal power levels for single detector unoptimized heterodyne detection.

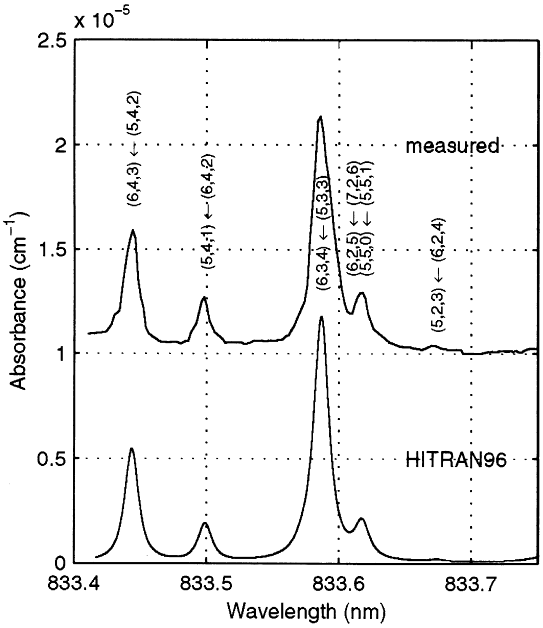


Fig. 3. Spectrum of water vapor in room air obtained using a ECDL locked to the ring-down cavity, and single detector polarization-dependent heterodyne detection. Spectrum based on HITRAN96 is shown in dashed lines. Transitions are labeled as $(J',K_a',K_c') \leftarrow (J,K_a,K_c)$.

Fig. 3 shows an absorption spectrum of water vapor present in ambient room air. Using these very weak transitions, the present sensitivity to water vapor at atmospheric pressures is about 1 part per million. RMS baseline noise is $\Delta\alpha = 1.7 \times 10^{-9}$ cm$^{-1}$, and corresponds to a RMS deviation $\sigma_\tau/\tau$ of $6 \times 10^{-5}$ for an average of 256 shots, i.e., a shot-to-shot fractional deviation of $1 \times 10^{-3}$. This value of $\sigma_\tau/\tau$ is about a factor of 50 above the expected noise level for a truly shot-noise-limited measurement at this PPL power level, without quantization error.

The present single-detector heterodyne detection system wastes half of the light emanating from the cavity. A dual-differential-heterodyne-detection scheme (with photodetectors at both outputs of the polarized beam splitter, and detector outputs differenced electronically) collects all the light exiting the cavity, and produces twice as much ring-down current. When properly balanced, such a system can also reject whatever noise might be present on the LO beam [10]. We demonstrated the increase in signal in such a system, but also found an increase in technical noise (from having twice the number of transimpedance amplifiers), which reduced its advantage.

## 5. Conclusions

We have reported here a heterodyne detection technique which resulted in a signal-to-noise increase of 31 dB over direct detection schemes employing the same cavity ring-down setup. Straightforward improvements in the electronics, laser power, and cavity throughput should allow true shot-noise-limited sensitivity for heterodyne-detected ring-down spectroscopy. Under shot-noise-limited conditions, the sensitivity and precision with which gas-phase absorption coefficients can be measured depends only on the cavity length, mirror reflectivity, and laser noise statistics. With current cavity technology (mirror reflectivity of 99.999%, and maximum throughput of hundreds of microwatts), the absorption sensitivity and precision are expected to be $\Delta\alpha = 10^{-13}$ cm$^{-1}$ for a single-shot decay measurement. With such an absorption sensitivity it should be possible to detect only a few thousand molecules inside the volume of the laser beam on typical IR absorption features.

## References

[1] A. O'Keefe, D.A.G. Deacon, Rev. Sci. Instrum. 59 (1988) 2544.
[2] D.Z. Anderson, J.C. Frisch, C.S. Masser, Appl. Opt. 23 (1984) 1238.
[3] J.M. Herbelin, J.A. McKay, M.A. Kwok, R.H. Ueunten, D.S. Urevig, D.J. Spencer, D.J. Benard, Appl. Opt. 19 (1980) 144.
[4] D. Romanini, A.A. Kachanov, N. Sadeghi, F. Stoeckel, Chem. Phys. Lett. 264 (1997) 316.
[5] D. Romanini, A.A. Kachanov, N. Sadeghi, F. Stoeckel, Chem. Phys. Lett. 270 (1997) 538.
[6] B.A. Paldus, J. Martin, J. Xie, J.S. Harris, R.N. Zare, J. Appl. Phys. 82 (1997) 3199.
[7] B.A. Paldus, C.C. Harb, T.G. Spence, B. Willke, J. Xie, J.S. Harris, R.N. Zare, J. Appl. Phys. 83 (1998) 3991.
[8] M.D. Levenson, S. Kano, Introduction to Nonlinear Laser Spectroscopy (Academic Press, Boston, MA, 1988).
[9] M.D. Levenson, G.L. Eesley, Appl. Phys. 19 (1979) 1.
[10] B.L. Schumaker, Opt. Lett. 9 (1984) 189.