Frequency-switched heterodyne cavity ringdown spectroscopy

M. D. Levenson and B. A. Paldus

Informed Diagonstics Corporation, 1050-IE Duane Avenue, Sunnyvale, California 94086

T. G. Spence,* C. C. Harb, and R. N. Zare

Department of Chemistry, Stanford University, Stanford, California 94305

M. J. Lawrence and R. L. Byer

Department of Applied Physics, Stanford University, Stanford, California 94305

Received November 23, 1999

When the frequency of light coupled into a cavity is suddenly shifted, the radiation emanating from the input port of the previously excited cavity can beat with the reflection of the frequency-shifted input on the surface of a photodetector. When the beat frequency is stable, the time decay of the resulting optical heterodyne signal can be used to measure intracavity absorption spectra with near quantum-limited sensitivity. © 2000 Optical Society of America

OCIS codes: 300.6360, 120.6200, 040.2840, 300.6310.

In cavity ringdown spectroscopy (CRDS) one measures the absorption of a sample inside a high-finesse optical cavity by abruptly extinguishing the light that is incident upon the cavity and measuring the decay rate of the exiting light.[1] When this direct CRDS signal is weak, optical heterodyne detection can provide additional sensitivity if a convenient optical local oscillator (LO) is available.[2] By scanning the incident laser frequency past the cavity resonance in a short time compared with the cavity decay time, Ye, Ma, and Hall showed that the strong beam reflected from the input port of a ringdown cavity can act as such a LO but with a frequency chirp.[3] In this Letter we report what we believe to be the first spectra taken with a related method, in which the incident light suddenly switches from exciting the cavity on resonance to a well-defined off-resonance frequency, resulting in a stable beat signal that can be captured and processed by use of tuned (analog) rf electronics at a 40-kHz repetition rate. We also demonstrate that this method approaches shot-noise-limited sensitivity.

Figure 1 shows the essence of this technique: The frequency of the cw field that is resonantly exciting the TEM_{00} mode of the ringdown cavity at ω is switched by $\Delta\Omega$, an amount much greater than the cavity linewidth Γ. The light reflected from the input mirror (with amplitude reflectivity R_1 and transmission T_1) and the light leaking from the optical cavity reach a photodetector system, where they beat.[4] Either the resulting single-shot output is recorded in its entirety by a digital oscilloscope or the rf power of the Fourier component at $\Delta\Omega$ is plotted as a function of time. Repetitive signals can then be averaged rapidly as in Ref. 5.

The usual equations for the slowly varying amplitudes of the i different frequency components relate the incident, reflected, and intracavity fields[6]:

$$\frac{\mathrm{d}E_{\mathrm{cav}i}}{\mathrm{d}t} + (\Gamma - i\rho\Delta\omega_i)E_{\mathrm{cav}i} = \frac{iT_1}{\tau}E_0\,. \tag{1}$$

In Eq. (1), $\Delta\omega_i = \Omega - \omega$ is the detuning of $E_{\mathrm{cav}i}$ from the laser, $\Gamma = (1-\rho)/\tau$ is the resonance width, E_0 is the external driving field that is spatially matched to the cavity mode, $\tau = p/c$ is the round-trip time around the cavity of length p, and the round-trip transmission for a three-mirror cavity is $\rho = R_1R_2R_3 \exp[-\alpha(\Omega)p]$, where $\alpha(\Omega)$ is the amplitude attenuation of the intracavity medium at the laser frequency Ω.

A coupling factor $\eta \le 1$ parameterizes the imperfect overlaps between the cavity-driving field (E_0) and the incident field (E_{inc}). In a reference frame rotating at ω, the frequency switch can then be represented as

$$E_0(t) = \eta E_{\mathrm{inc}}(t) = \begin{cases} \eta E_{\mathrm{inc1}} & t < 0 \\ \eta E_{\mathrm{inc2}} \exp(-i\Delta\Omega t) & t > 0 \end{cases}. \tag{2}$$

Before switching, the incident field E_{inc1} drives the circulating TEM_{00} cavity mode to its steady-state resonant value $E_{\mathrm{cav1}}{}^0 = i\eta T_1 E_{\mathrm{inc1}}/(1-\rho)$, and the reflected field is minimized by the destructive interference between the incident amplitude and the transmitted portion of the intracavity amplitude.

The time variables in Eqs. (1) and (2) pertain to the input, but light leaking out of the cavity in the direction of the reflected beam has been delayed by a round-trip time τ, allowing the incident field to advance in phase. As a result, the amplitude of the reflected field at the time corresponding to time t in Eq. (1) is[6]

$$E_{\mathrm{refl}}(t) = R_1E_{\mathrm{inc}}(t+\tau) + iT_1R_2R_3\Sigma_i E_{\mathrm{cav}i}(t)\,. \tag{3}$$

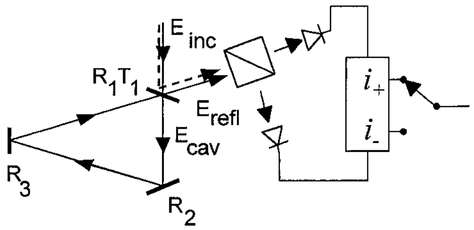

Fig. 1. Schematic of the three-mirror cavity, dual-detector experiment. The solid and dashed lines represent the cavity couplings before and after the frequency switch, respectively.

After switching, the original intracavity field decays exponentially at rate Γ. The reflected amplitude after switching becomes

$$E_{\mathrm{refl}}(t) = iT_1R_2R_3E_{\mathrm{cav1}}{}^0 \exp(-\Gamma t) + R_1E_{\mathrm{inc2}} \exp[-i\Delta\Omega(t+\tau)]. \tag{4}$$

The two right-hand terms represent the CRDS amplitude and the frequency-shifted LO necessary for optical heterodyne detection, respectively. A very small term proportional to the intracavity field driven after switching has been neglected.

The current produced by the photodetector system (in the summation mode) is proportional to the absolute square of the reflected amplitude integrated over the surface area (A) of the detector. The spatial overlaps of the intracavity fields with the reflected field are again reduced by the factor η, which can be considered a heterodyne efficiency. Because the quantum efficiency (q) is also less than unity, the destruction of $\hbar\Omega$ units of light energy produces eq electrical charge. When all the reflected light reaches the detector, the sum current is

$$\begin{aligned} i_+(t) = \frac{eqc\epsilon_0A}{2\hbar\Omega}\{&R_1{}^2|E_{\mathrm{inc2}}|^2 \\ &+ T_1{}^2R_2{}^2R_3{}^3|E_{\mathrm{cav1}}{}^0|^2 \exp(-2\Gamma t) \\ &- 2\eta T_1R_1R_2R_3 \,\mathrm{Im}[E_{\mathrm{cav1}}{}^0E_{\mathrm{inc2}}{}^* \exp(i\Delta\Omega\tau) \\ &\times \exp(i\Delta\Omega t - \Gamma t)]\}. \end{aligned} \tag{5}$$

The direct CRDS term [$i_d(t)$, second term in braces] decays as $\exp(-2\Gamma t)$, whereas the heterodyne (last) term decays at half that rate and oscillates in time as $\exp(i\Delta\Omega t)$. As $t \rightarrow \infty$, $i_+(t)$ approaches a steady-state value proportional to the optical LO power.

The experimental apparatus was nearly identical to the system published previously.[5] It employed a Lightwave Electronics 122 Nd:YAG laser to excite the high-finesse cavity at 1.064 μm. The acousto-optic modulator was suddenly switched between two oscillators separated in frequency by $\Delta\Omega$. Finally, the photodetectors collecting the s-polarized reflection from the cavity produced the signal current. Elaborate servos ensured that the cavity was initially on resonance.

In Fig. 2 we compare the experimentally measured current signal through a complete switching cycle with the prediction of Eqs. (1) and (5), using the parameters listed in Table 1 and appropriate amplification factors. The agreement between the recorded data, including the frequency of the oscillations, transient behavior just as the input is switched (shown in the inset of Fig. 2), the decay, and the ring-up, and theory is very good. Also shown on the same scale is the inferred direct detection signal [$i_d(t)$] and the current corresponding to the portion ($\sim 1 - \eta^2$) of the reflected (LO) beam that does not interfere with the cavity field.

Although the waveforms can be fitted readily by computer, it is more convenient for spectroscopy to extract the decay rates with analog electronics.[5] This extraction was accomplished most easily by use of a Hewlett-Packard 8590L electronic spectrum analyzer to measure the rf power from the photodetector within a bandwidth of $\sim 2 \times 10^7$ s^{-1} ($\Delta\nu = 3$ MHz) around $\Delta\Omega$ as a function of time. It was also convenient to use rather larger shift frequencies ($\Delta\Omega/2\pi \sim 10.5$ MHz) than in Fig. 2. The electrical power at $\Delta\Omega$, given by

$$P_{\Delta\Omega}{}^+(t) = [i_+{}^2(t)R]|_{\Delta\Omega} + N_{\mathrm{inc2}}\Delta\nu + \zeta\Delta\nu + 2ei_+(t)R\Delta\nu, \tag{6}$$

has contributions from noise processes as well as from the signal current in Eq. (5). In Eq. (6), R represents the nominal impedance of the detection system, N_{inc2} and ζ are the spectral densities of the reflected laser and electronic noise (after the frequency switch), respectively, and the final term represents the shot noise on the detected light. Trace (a) in Fig. 3 shows a single experimental trace of $P_{\Delta\Omega}{}^+(t)$ measured in [dBm] and recorded from the video output of the spectrum analyzer set at $\Delta\Omega$. An exponential decay of the heterodyne signal through >4 orders of magnitude is evident from $t \sim 13$ μs to $t \sim 35$ μs. The plateau at -63 dBm represents the noise floor that is due to the last three terms in Eq. (6).

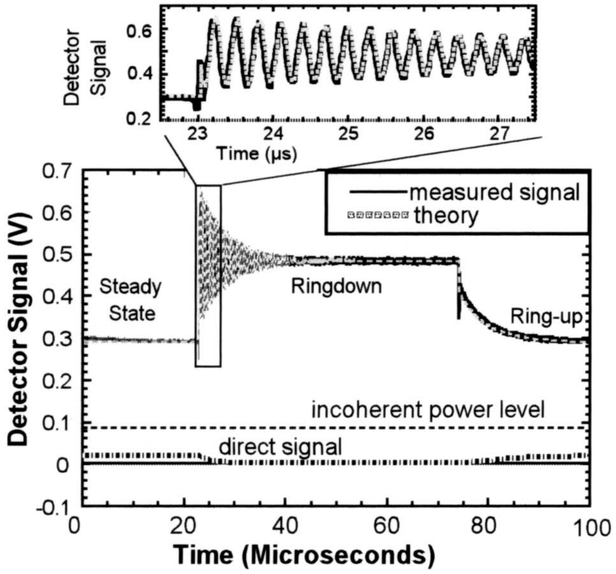

Fig. 2. Comparison of experimental (solid) and theoretical (gray dashed) traces of photodetector output as a function of time through an entire ringdown cycle. The inset shows the excellent agreement at short times after the frequency switch. Also shown are the incoherent background level $\propto (1 - \eta^2)i_+(\infty)R$ and the inferred direct signal $i_d(t)R$.

Table 1. Experimental Parameters

T_1	7×10^{-3}
$R_1 = R_2 = R_3$	$1 - (4.5 \times 10^{-6})$
τ	1.4×10^{-9} s
Γ	3.51×10^5 s^{-1}
ρ	0.9985
p	42 cm
$E_{\mathrm{inc1}}/E_{\mathrm{inc2}}$	0.81
η	0.91
q	0.64

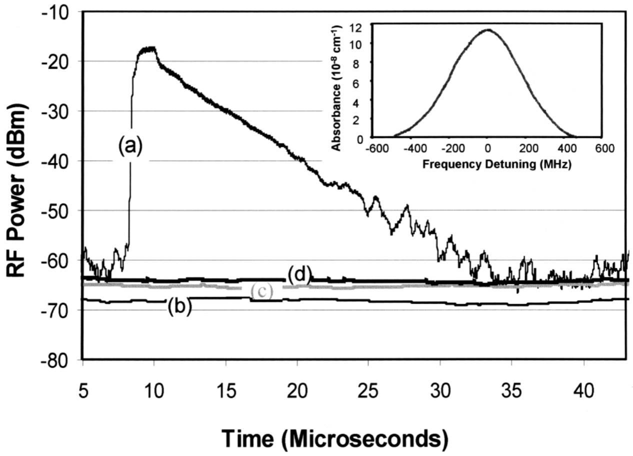

Fig. 3. rf power [in dBm] (a) produced by frequency-switched heterodyne detection of the CRDS amplitude, compared with the averaged dark-noise level (b), detector difference noise power (c), and steady-state LO noise (d). The inset shows an absorption trace of the $(0, 0^0, 0) \rightarrow (2, 0^0, 3)$ overtone transition of 2 Torr of CO_2 at 1.064 μm as recorded by this system.

When the detector system is set to report the difference in currents between the two balanced detectors, all but ~1.6% of the ringdown signal and the incident laser noise cancel out, leaving the electronic and the quantum noise as the dominant contributions.[4] In the steady state, the difference power is

$$P_{\Delta\Omega}^{-}(\infty) = \zeta\Delta\nu + 2ei_+(\infty)R\Delta\nu\,. \tag{7}$$

Trace (b) in Fig. 3 is a time average of the electronic noise taken by blocking all the incident light and represents a measurement of $\langle\zeta\rangle\Delta\nu$. Before averaging, the dark-noise trace had submicrosecond fluctuations comparable to those seen in the plateau of trace (a) near $t = 40$ μs. Trace (c) in Fig. 3 is a similarly averaged plot of $P_{\Delta\Omega}^{-}(\infty)$ with 0.36 mW incident upon each detector. The 3.0 ± 0.6 dB increase in the level of trace (c) over trace (b) indicates that the shot-noise level was comparable to the dark noise. Trace (d) is an averaged plot of $P_{\Delta\Omega}^{+}(\infty)$ taken under the same conditions. The 1.2 ± 0.6 dB increase of this sum noise level over the difference implies that the contribution of the laser and modulator system (i.e., $\langle N_{\mathrm{inc2}}\rangle\Delta\nu$) to the noise floor is small and comparable to the uncertainty in the noise levels.

The fact that the noise floor of trace (a) overlays trace (c) indicates that the heterodyne detection system is operating within ~3 dB of the fundamental shot-noise limit. When the optical LO [first term in braces in Eq. (5)] is the dominant component at the detector the signal-to-noise ratio for heterodyne detection is

$$S/N|_{\mathrm{SNL}} = [i_+^2(t)R]|_{\Delta\Omega}/2ei_+(\infty)R\Delta\nu$$

$$= \eta^2 i_d(t)/2e\Delta\nu\,, \tag{8}$$

which approaches the shot-noise limit for direct detection as $\eta \rightarrow 1$. Thus Figs. 2 and 3 imply that the sensitivity of detection of very weak (<20-pW) ringdown signals (i.e., near 30 μs) by this method is within a factor of ~2 of fundamental limits. At higher (>50-nW) signal levels (i.e., earlier in the decay), rf electronics artifacts limit the absorption sensitivity of this early demonstration to values lower than in conventional CRDS, where microwatt powers may be available.[5]

The inset in Fig. 3 shows an absorption line that is due to the weak $(0, 0^0, 0) \rightarrow (2, 0^0, 3)$ overtone transition of 2 Torr of CO_2 at 1.064 μm as measured by this technique.[5] The absorbance scale was calibrated by use of the HITRAN database value of 7.6×10^{-6} cm^{-1} at 760 Torr for this transition. Reducing the pressure of the gas proportionately reduced the deviation of the decay time from empty-cavity value to ~0.1 Torr, at which point the absorbance that was detectable in 100 ms was $\sim 1 \times 10^{-9}$ cm^{-1}.

In conclusion, frequency-switched heterodyne cavity ringdown spectroscopy promises to be a sensitive and convenient spectroscopic tool. It is compatible with the low-quantum-efficiency, modest-bandwidth detectors that are available in the infrared region. The cavity need not transmit any power at all; large beat signals appear in reflection, making single-port cavities useful in CRDS for the first time. Suppressing rf nonlinearities and artifacts should yield an absorbance sensitivity at least as good as that of the best direct detection and previous heterodyne cw-CRDS techniques.[2,5]

This work was partially inspired by Richard G. Brewer and partly funded by the U.S. Air Force Office of Scientific Research under contract F49620-92-J-0074. M. D. Levenson's e-mail address is muddle@aol.com.

*Present address, Department of Chemistry, Loyola University, New Orleans, Louisiana 70118.

References

1. A. O'Keefe and D. A. G. Deacon, Rev. Sci. Instrum. **59,** 2544 (1988).
2. M. D. Levenson, B. A. Paldus, T. G. Spence, C. C. Harb, J. S. Harris, Jr., and R. N. Zare, Chem. Phys. Lett. **290,** 335 (1998).
3. J. Ye, "Ultrasensitive high resolution laser spectroscopy and its application to optical frequency standards," Ph.D. dissertation (Department of Physics, University of Colorado, Boulder, Colo., 1997), pp. 119–122.
4. A system in which the output current can be set to correspond to either the sum or the difference of the optical outputs from a 50–50 beam splitter facilitates noise analysis; see B. L. Schumaker, Opt. Lett. **9,** 189 (1984).
5. T. G. Spence, C. C. Harb, B. A. Paldus, R. N. Zare, B. Willke, and R. L. Byer, Rev. Sci. Instrum. **71,** 347 (2000).
6. M. J. Lawrence, B. Willke, M. E. Husman, E. K. Gustafson, and R. L. Byer, J. Opt. Soc. Am. B **16,** 523 (1999).