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RESEARCH ARTICLE

Continuous-wave cavity ring-down technique for accurate measurement of high reflectivity

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Abstract A continuous-wave cavity ring-down (CW-CRD) technique employing a broadband diode laser is developed for high reflectivity measurement. The theory of square-wave modulated CW-CRD is presented. The spectrum of the broadband CW diode laser covers numerous free spectral ranges (FSRs) so that sufficient laser power is coupled into the cavity. Both amplitude and phase-shift of the first harmonic of the CRD signal, measured at an appropriate frequency range, are detected by a lock-in method and fitted to obtain the ring-down time and reflectivity. The measurements are repeated with five different cavity lengths and all the fitted reflectivities are in excellent agreement, indicating a high reliability of the CW-CRD technique. The reflectivity of the cavity mirror is determined statistically to be 99.70%, with an uncertainty of 0.01%.

Keywords cavity ring-down (CRD), high reflectivity, broadband, continuous-wave (CW), diode laser

1 Introduction

Cavity ring-down (CRD) spectroscopy is a highly-sensitive absorption measurement technique and has been widely used to obtain the absorption spectrum of gas-phase species [1–7]. The reflectivity of the cavity mirror can be accurately measured with an empty cavity configuration without absorbing species. In 1980, Herbelin et al. were the first to propose the use of an optical cavity for measuring the reflectivities of mirrors [8], which became the precursor of continuous-wave cavity ring-down (CW-CRD) technique. By employing an intensity-modulated CW laser and a lock-in amplifier, they accurately determined the reflectivities with an uncertainty of 0.01% by measuring the phase-shifts introduced by the ring-down cavity. Meanwhile, by abruptly switching off a CW laser when the intra-cavity intensity exceeded a predefined threshold, Rempe et al. obtained a ring-down transient from which a reflectivity as high as 99.99984% was determined [9]. Since a narrowband laser was employed in their experiment, the laser frequency and one of the cavity longitudinal modes were forced to be in resonance by scanning the cavity length via a piezoelectric transducer (PZT) [9–11].

The experimental setup of the narrowband laser based CRD is complicated and the cavity length scanning or laser wavelength tuning techniques are necessarily used to build up enough laser power in the cavity. In this paper a relatively simple CW-CRD technique employing a broadband diode laser is developed for the high reflectivity measurement. The spectrum of the broadband CW diode laser covers numerous free spectral ranges (FSR) so that sufficient laser power is coupled into the cavity which can allow for a sensitive measurement for the high reflectivity. The cavity decay time is obtained by using the amplitude-frequency and phase-frequency fitting techniques. The reflectivity of the cavity mirror is determined statistically to be 99.70% with an uncertainty of 0.01%. The measurements are repeated with five different cavity lengths and all results are in excellent agreement. Compared with the pulsed-CRD approach [12–17], the CW-CRD technique is simple, low-cost and highly precise due to the use of a CW diode laser with a high beam quality. Furthermore, this technique can be easily expanded to other wavelengths, as commercial diode lasers are available in a wide wavelength range from near ultraviolet (< 400 nm) to 2 μm.

2 Theoretical description

The schematic principle of CW-CRD method is shown in Fig. 1. The laser beam, whose intensity is square-wave

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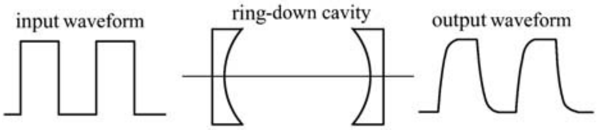

Fig. 1 Schematic diagram of CW-CRD method

modulated, is coupled into a high-finesse cavity and reflected back and forth many times in the cavity. The CRD signal has the same period as the input square-wave signal. A phase retardance and an amplitude attenuation of the CRD signal are produced by the ring-down cavity.

With a square-wave modulation, the normalized intensity of the laser beam can be expanded into a Fourier series as

$$I_{\mathrm{sqr}}(t)=\frac{1}{2}+\frac{2}{\pi}\sum_{k=1}^{\infty}\frac{1}{k}\sin\frac{k\pi}{2}\cos(k\omega_0 t), \tag{1}$$

where ω_0 is the angular modulation frequency. The impulse response of the ring-down cavity is a single exponential decay function. The CRD signal is proportional to the convolution of the input square-wave function and the impulse response function, which can be expressed as [18]

$$\begin{aligned} I(t)&=\frac{1}{\tau}\int_{-\infty}^{t}I_{\mathrm{sqr}}(t')\mathrm{e}^{-(t-t')/\tau}\mathrm{d}t' \\ &=\frac{1}{2}+\frac{2}{\pi}\sum_{k=1}^{\infty}\frac{1}{k\left(1+k^2\omega_0^2\tau^2\right)}\sin\frac{k\pi}{2} \\ &\quad\cdot[\cos(k\omega_0 t)+k\omega_0\tau\sin(k\omega_0 t)]. \end{aligned} \tag{2}$$

The cavity decay time is given by [19, 20]

$$\tau=\frac{L}{c(1-R+\alpha L)}, \tag{3}$$

with $R=\sqrt{R_1R_2}$, R_1, R_2 are the reflectivities of the two cavity mirrors, respectively. α is the intra-cavity loss, including the absorption and scattering in the cavity. L is the cavity length and c is the velocity of light. The CRD signal can be resolved into a series of odd orders harmonics as

$$I_k(t)=I_1\cos(k\omega_0 t+\phi). \tag{4}$$

Here the amplitude and phase-shift are given by

$$I_1=\frac{2}{k\pi\sqrt{1+k^2\omega_0^2\tau^2}}, \tag{5}$$

$$\tan\phi=-k\omega_0\tau, k=1,3,5,\ldots, \tag{6}$$

respectively. Equation (6) was previously given by Engeln [18] with $k=1$. In this paper the cavity decay time is obtained by fitting the experimental frequency dependence of the amplitude and phase-shift of the first harmonic of the CRD signal according to Eqs. (5) and (6). Then the reflectivity of the cavity mirror is calculated by Eq. (3).

With high reflectivity cavity mirrors, the ring-down cavity acts as a frequency-comb filter with a frequency period of ν_{FSR} ($=c/(2L)$, i.e., the free spectral range). The theoretical intensity transmission will approach 100% if the laser frequency is in resonance with one of the longitudinal eigenmodes of the cavity. However, when a narrowband laser (with a line width of $\Delta\nu<\nu_{\mathrm{FSR}}$) is used, a PZT should be employed to scan the cavity length and obtain a matching between the laser frequency and one of the cavity modes. The cavity length scanning approach makes the experimental setup of the narrowband laser based CRD complicated. One of the advantages of the broadband diode laser based CW-CRD is that the laser spectrum covers numerous FSRs, so that the CW laser power can always be continuously injected into the cavity. The intensity fluctuation, introduced by the cavity mirror vibration and laser wavelength drifting, as well as its influence on the high reflectivity measurement, is reduced due to the use of a broadband laser.

3 Experimental setup

The schematic diagram of the experimental CW-CRD setup for the high reflectivity measurement is shown in Fig. 2. A CW broadband diode laser (Melles Griot, Model 56ICS115/HS) is used, whose wavelength is centered at 828 nm with an effective line width of 3 nm. The intensity of the diode laser is square-wave modulated by a function generator, whose output also serves as the reference signal for the lock-in detection.

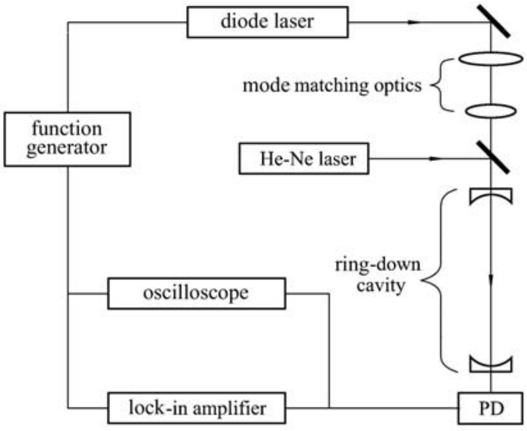

Fig. 2 Schematic diagram of experimental CW-CRD setup for high reflectivity measurement

A transverse-mode-matching system is employed to the coupling of the laser beam into the TEM_{00} mode of the ring-down cavity. To align the highly-reflective cavity mirrors, a He-Ne laser visible at 632.8 nm is employed. The ring-down cavity is formed by two identical plano-concave mirrors with a radius of curvature $r = -100$ cm. In addition, TiO_2/SiO_2 dielectric multi-layers are deposited on the concave surface of both mirrors to improve their reflectivity near 830 nm. The theoretical reflectivity of the cavity mirror is calculated to be 99.94% by using the film-design software and experimentally measured to be $99.9 \pm 0.3\%$ with a spectrophotometer (Lambda-900). The laser beam is reflected back and forth many times in the cavity and attenuated with a fixed loss factor after each round trip. The light that leaks out of the cavity is detected by a silicon photo-detector (PD) module. The output of the detector module is sent to a lock-in amplifier to measure the amplitude and phase-shift of the first harmonics of the CRD signal.

4 Results and discussion

The temporal waveform of the CW-CRD signal and corresponding reference signal are recorded and shown in Figs. 3(a) and 3(b) with a modulation frequency of 10 and 50 kHz, respectively. The CRD signal has the same period as the input square-wave signal. As the modulation frequency increases, the phase-shift of the laser beam leaking out of the cavity increases, and the temporal waveform of the cavity ring-down signal evolves from a square-wave function into a triangle function.

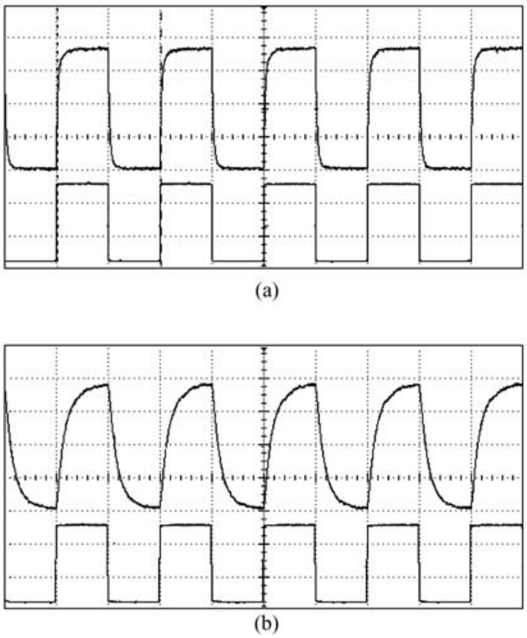

Fig. 3 Temporal waveforms of CW-CRD signal and corresponding reference signal. (a) 10 kHz modulation frequency; (b) 50 kHz modulation frequency

The frequency dependences of the phase-shift and amplitude of the first harmonic of the CRD signal are recorded by a lock-in amplifier and shown in Figs. 4 and 5, respectively. Corresponding theoretical best-fits are also presented. As predicted by Eq. (6) and also shown in Fig. 4, the tangency of phase-shift is directly proportional to the angular modulation frequency and the slope is just the cavity decay time. According to Eq. (6), the cavity decay time can be calculated directly from the phase-shift at a single modulation frequency. However, in this paper several phase-frequency data points are measured at each cavity length and used to fit the cavity decay time, leading to an averaged result. The amplitude of the first harmonic of the CRD signal is shown in Fig. 5 as a function of the angular modulation frequency. The amplitude is normalized by the intensity of the incident beam. The lock-in measurement is repeated at five different cavity lengths, and the excellent agreements

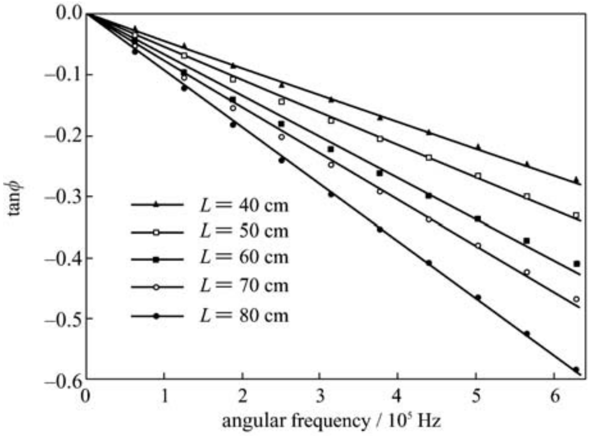

Fig. 4 Angular frequency dependence of tangency of phase-shift of the first harmonic of CRD signal

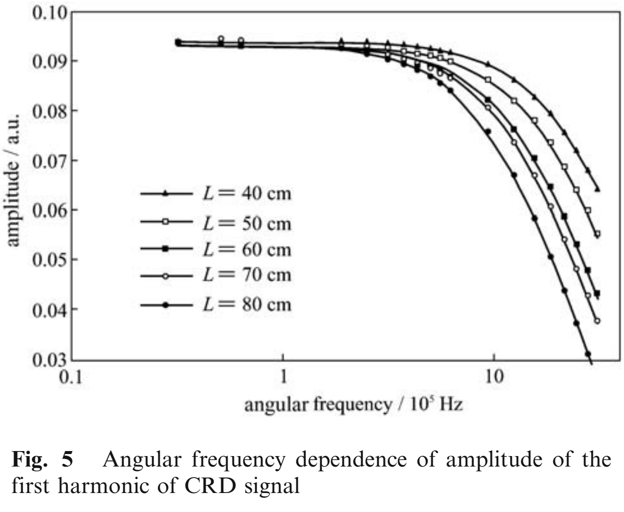

Fig. 5 Angular frequency dependence of amplitude of the first harmonic of CRD signal

are obtained between the experimental data and the theoretical predictions.

All results including the cavity decay time and the reflectivity obtained by fitting the frequency dependences of the amplitude and phase-shift of the first harmonic of the CRD signal are listed in Table 1. The fitted results obtained at five different cavity lengths and by different fitting methods are in excellent agreement, indicating that the influence of possible mis-adjustment [17] can be neglected in this experiment. The reflectivity of the cavity mirror is statistically determined to be 99.70%, with an uncertainty of 0.01%. According to Eq. (3), the cavity decay time is directly proportional to the cavity length and the slope is $1/[c(1-R)]$. The experimental τ-L curve is linearly fitted and the reflectivity is calculated to be 99.700%, with an uncertainty of 0.006%.

Table 1 Cavity decay time and reflectivity of cavity mirror fitted by frequency dependences of amplitude and phase-shift of the first harmonic of CRD signal

cavity length/cm	fitted by amplitude		fitted by phase-shift	
	ring-down time /μs	R/%	ring-down time /μs	R/%
40	0.43	99.69	0.44	99.70
50	0.53	99.69	0.54	99.69
60	0.69	99.71	0.67	99.70
70	0.77	99.70	0.76	99.69
80	0.96	99.72	0.93	99.71

The CRD technique is a sensitive method for the high reflectivity measurement [12]. Worth mentioning is the inherece in all CRD methods of higher reflectivity mirrors' capability to provide a longer cavity decay time which can lead to a more sensitive measurement. Assuming that the intra-cavity absorption and scattering can be neglected, the theoretical uncertainty of the reflectivity can be expressed as $\Delta R=(1-R)(-\Delta L/L+\Delta\tau/\tau)$, with $R=1-L/(c\tau)$. In this experiment the cavity decay time and the cavity length are measured with an uncertainty of approximately 2.0% and 0.5%, respectively. With a reflectivity of 99.70%, the uncertainty of the reflectivity is calculated to be 7.5×10^{-5}, which is in good agreement with the experimental result. If a couple of cavity mirrors with a reflectivity of 99.99% are used, the uncertainty of the reflectivity will be reduced to 2.5×10^{-6} or even lower, for the uncertainty of the cavity decay time is lower.

5 Conclusions

The broadband diode laser based CW-CRD technique was developed for the high reflectivity measurement of cavity mirrors. Both the amplitude and phase-shift of the first harmonic of the CRD signal, measured at an appropriate frequency range, were recorded by the lock-in method and fitted to obtain the cavity decay time and the reflectivity. The measurements were repeated with five different cavity lengths and the reflectivities obtained with different cavity lengths and by different fitting methods were in excellent agreement, indicating the high reliability of the CW-CRD technique. The reflectivity of the cavity mirror was determined statistically to be 99.70%, with an uncertainty of 0.01%, which was in good agreement with the measurement by the spectrophotometer (Lambda 900). Compared with the pulsed-CRD and the narrowband laser based CW-CRD approaches, the broadband diode laser based CW-CRD technique was simple, low-cost and highly precise.

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