Appl. Phys. B 78, 493–501 (2004)
DOI: 10.1007/s00340-003-1360-y

**Applied Physics B**
**Lasers and Optics**

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# High-resolution cavity-tuned ringdown spectrometer using a narrow-bandwidth pulsed laser source

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**Received: 24 June 2003/Revised version: 25 September 2003**
**Published online: 11 February 2004 •** 

**ABSTRACT** A novel method of cavity ringdown spectroscopy is proposed to achieve high spectral resolution with tunable narrow bandwidth pulsed lasers. We demonstrate a cavity-tuned ringdown configuration in which only a single cavity mode is kept excited near the carrier frequency of a narrow bandwidth pulse laser. This is done simply by making a cavity resonance actively track the frequency reference served by the cw injection seed of the pulsed laser source. We present the servo mechanism used in the cavity resonance tracking, reliable procedures for transverse mode matching, and the evidence of single longitudinal mode excitation. The spectrometer performance is tested to record weak molecular overtone features of acetylene around the wavelength of 570 nm, showing cavity tracking stability within 5-MHz uncertainty which overcomes the bandwidth limit of pulsed laser sources itself.

**PACS** 42.62.Fi; 42.60.Da; 33.20.Kf

## 1 Introduction

During the past decade cavity ringdown spectroscopy (CRDS) has become one of the most popular methods in the absolute absorption measurement for its high sensitivity and technical simplicity [1]. Remarkably, the CRDS permits an excellent compatibility even with pulsed laser sources suffering considerable intensity noise and fluctuation, without any severe deterioration in the detection sensitivity [2].

In the widespread CRDS applications [3], a number of novel variations have emerged for advanced performance [4], at some cost of sacrificing the simplicity. The high spectral resolution is one of the major aims in such technical refinements. To meet the need, the CRDS has led to the use of continuous-wave (cw) lasers with much narrower linewidth than that of pulsed lasers. Incorporation with cw laser sources has required the controlled sequences to build up sizable intracavity optical power and then to interrupt the laser light being coupled to the cavity rapidly, in order to produce cavity ringdown transients reliably. Several different ways of making cavity resonance with a cw excitation laser have been proposed and gained wide acceptance to many experimentalists [5–8].

It is true that easy solutions have been made available through CRDS schemes employing cw lasers, but high spectral resolution in CRDS are also pursued by using pulsed laser excitations. This is because the CRDS using pulsed laser excitations is still required for specific applications and possesses particular merits aside from advantages in simplicity and wide wavelength coverage . Even in the presence of cw laser radiations available for a wavelength region of interest, the pulsed CRDS is promising for particular purposes such as: a study of transient sample dynamics in a pumpprobe measurement using synchronized cavity ringdown events [9–12], nonlinear CRDS techniques requiring well-characterized initial conditions of cavity excitation intensity [13, 14], high-resolution metrological CRDS that should be free of Doppler shift errors as in the typical cw-CRDS operating with a swept cavity[5, 6].

It was not until van Zee et al. [17] demonstrated the single-cavity-mode pulsed-CRDS in 1999 for the first time, that the potential of CRDS to achieve the frequency resolution offered by the extremely narrow linewidth of a high-finesse cavity was realized [18–20]. Even pulsed laser sources of finite bandwidth could permit resolutions reaching down to the linewidth of a high-finesse cavity, overcoming the bandwidth limit of laser sources themselves. This is, of course, the case only on condition that the laser bandwidth is smaller than the cavity mode spacing (free spectral range, FSR) and the frequency of a cavity resonance mode is well defined and controlled. Frequency tracking of a cavity resonance, tuned to the center frequency of a pulsed laser excitation, becomes an essential component of such high-resolution pulsed-CRDS.

However, cavity resonance tracking of an excitation laser pulse has been considered technically daunting because of the difficulty in generating frequency discrimination signals for the task. It is usually hard to measure pulsed laser frequencies relative to the cavity resonance frequency with a desirable accuracy, especially when any precise frequency reference of laser pulses does not exist and the spectrum of a pulsed laser varies shot by shot. The approach taken by van Zee et al. was to scan a ringdown cavity in parallel with the frequency tuning of a narrow-bandwidth pulsed OPO source which was frequency-stabilized to a transfer cavity; a separate but simultaneous cavity scan was carried out carefully to match changes in the light source frequency within the uncer-

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tainty as small as 3 MHz, without frequency discrimination of the light source.

This paper presents a novel experimental configuration to implement the high-resolution single-mode CRDS with pulsed laser sources. The way we puzzle out the problem is to incorporate a pulsed laser source possessing a well-defined frequency reference with a closed-loop cavity tracking mechanism: a tunable narrow-bandwidth pulsed source is constructed from the direct pulsed amplification of a tunable narrow bandwidth cw laser to offer both required power and spectral characteristics. The heart of our proposal is that the cw injection seed laser is allowed, in part, to serve as a frequency reference to which a ringdown cavity resonance is kept tuned. Unlike the previous effort by van Zee et al. [17], we employ a closed-loop feedback servo to make a ringdown cavity actively track the frequency of a cw injection seed and that of a pulse laser beam in turn. The addition of a cavity feedback mechanism not only ensures a constant, reliable coupling of a narrow-bandwidth laser pulse into a ringdown cavity via single mode resonance, but also considerably eases the technical requirement of temperature stability and linearity of cavity length scan, eliminating the need for calibration of the cavity scan characteristics which is often the critical limiting factor in open-loop controls.

The essentials of a proposed excitation laser source is the availability of a definite frequency reference offered by a tunable cw laser source that is simultaneously used for seeded generation of pulsed laser radiation. The tunable cw laser seed, however, does not necessarily have the same wavelength of the final wavelength of the pulsed laser source. This becomes more obvious when the proposal is extended to use a pulsed laser source that is constructed from either a seeded OPO or harmonically converted seeded amplification, in which the wavelength of the cw seed laser is definitely related, but different from, the target wavelength of the pulsed laser. The role of a tunable cw laser source that has been employed in the proposed CRDS scheme is to serve as a wavelength reference only, designed to infer the carrier wavelength of a pulsed laser source but, in principle, not to be used for cavity excitation itself. Any confusion should therefore be avoided on the role of a tunable cw laser source as an alternative source of cavity excitation, which might arise in our demonstration where wavelengths of a tunable cw seed laser and a pulsed source happen to be the same in the simplest experimental form of high-resolution pulsed CRDS.

Our proposed scheme of cavity-tuned pulsed CRDS does not adopt elaborate cavity locking procedures based on the Pound-Drever-Hall technique [21, 22], avoiding painstaking details involved with ultrahigh-finesse cavities used for CRDS [8]. Instead, a scan-and-seek strategy is employed where a cw frequency reference beam, a portion split from the cw injection seed for a pulse laser source, illuminates a ringdown cavity after being dithered in frequency. Cavity transmission bursts are then monitored. The frequency shift at which resonant cavity transmission occurs provides the measure of frequency detuning of the cavity from the pulsed laser frequency. Each frequency ramp produces the error signal representing the cavity detuning and the cavity can be made to correct its length toward resonance with the light source. The limit of frequency resolution in our cavity-tuned pulsed CRDS, is therefore determined by the quality of cavity frequency tracking.

We also attempt to relax the constraints on the cavity length and the mirror curvature that have been previously configured to ensure sufficiently wide cavity transverse mode spacings with respect to pulse laser bandwidths. By carrying out an efficient transverse mode optimization, the length of a ringdown cavity in this work can be further extended as long as the cavity FSR is no smaller than the full span of a pulse laser spectrum.

In the following discussion, the configuration of a tunable narrow bandwidth pulsed laser source and the cavity tracking servo are described, with emphasis on the technique of laser frequency dithering and cavity frequency discrimination. Also presented are the reliable procedures for transverse mode matching and the experimental evidence of single longitudinal mode excitation. Finally, the spectrometer performance is characterized by recording weak molecular overtone features of acetylene in the visible region.

## 2 Principle of cavity-tuned ringdown spectrometer with pulsed single cavity mode excitation

The working principle and the optical layout of the cavity-tuned pulsed CRDS is depicted in Fig. 1. The spectral light source is constructed from direct narrow-bandwidth pulsed amplification of a tunable single-frequency cw injection seed laser. The resulting pulsed laser beam works for cavity excitation to produce ringdown signals whereas a fraction of the tunable single-frequency cw laser beam is reserved for the use in cavity resonance tracking. The pulsed and cw laser beams are able to carry out their own functions simultaneously, having linear polarization states assigned orthogonal to each other. Without any disturbance from the cw frequency reference beam that is in charge of cavity tracking control, the pulsed laser can be introduced to the optical path through a ringdown cavity via a polarizing beam splitter placed before the ringdown cavity, allowing a CRD measurement procedure in the pulsed laser's polarization. During the frequency scan of the tunable cw laser, the cavity resonance is fed back to be

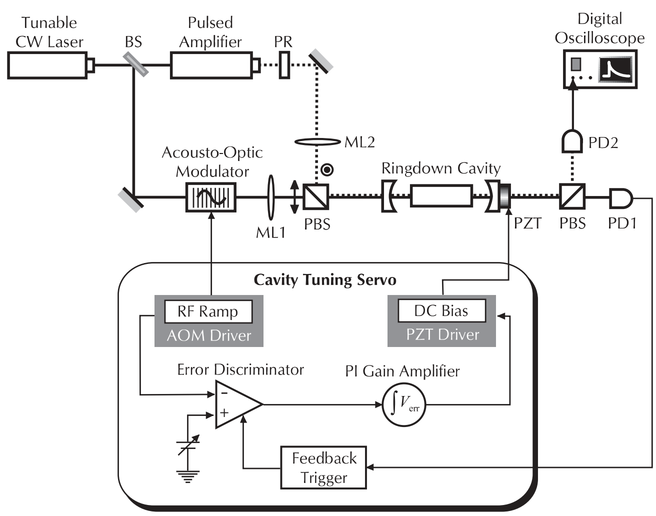


**FIGURE 1** Schematic configuration of the cavity-tuned pulsed CRD spectrometer

tuned to the cw frequency reference by a cavity tracking servo. Because of the direct pulsed amplification from the cw injection seed laser, the pulsed laser's center frequency is identical to the cw frequency reference and thus it keeps track of the cavity resonance as well.

There have been, in fact, several alternatives developed to lock an optical cavity to a frequency-stabilized cw laser beam. Conventional techniques include (i) the sideband lock on a cavity transmission peak [23], (ii) the frequency modulation technique utilizing a derivative error signal [24], and (iii) the phase modulation technique [22], performing phase-sensitive detection at the modulation sideband frequency, to produce a dispersion-shaped error signal with a steep zero-crossing and wide re-locking wings. All aforementioned optical cavity locking techniques, however, are quite painstaking and sophisticated for use with high-finesse cavities in CRDS implementations. We thus chose to devise a less demanding technique that is suitable and convenient for our high-resolution purposes in pulsed CRDS; such a tight cavity locking is not a procedure in absolute need, instead, maintaining a cavity resonance around the cw reference frequency within a certain amount of uncertainty is sufficient.

The cavity resonance tracking we employ is based on a scan-and-seek approach in which the deviation of a cavity resonance frequency $\omega_{\text{res}}$ from a desired cw frequency reference $\omega_{\text{trk}}$ is determined by measuring the frequency at which a resonant coupling in cavity transmission occurs. For this purpose, a frequency-dithered cw tracking beam is generated and its transmission through the ringdown cavity is observed. When the frequencies of the cavity resonance and the dithered cw tracking beam coincide, a transmitted signal burst results, and the cavity resonance frequency is inferred from the frequency of the dithered cw tracking beam at that moment. Suppose that the frequency modulation $\omega_{\text{m}}(t)$ is carried out by an acousto-optic modulator (AOM) around the cw reference frequency at $\omega_{\text{L}}$ as,

$$\omega_{\text{m}}(t) = \omega_{\text{L}} + kV(\Omega t) + \omega_{\text{off}}\,, \tag{1}$$

where $V(t)$ is the modulation signal, $\Omega$, the modulation frequency, $k$, the proportionality factor, and $\omega_{\text{off}}$, the dc frequency offset accompanied in AOM. The position of a cavity resonance mode can be deduced as $\omega_{\text{res}} = \omega_{\text{L}} + kV_{\text{res}} + \omega_{\text{off}}$, from the voltage level of the modulation signal, $V_{\text{res}}$, at which the cavity resonantly transmits the cw tracking laser beam in each modulation period.

Note that the frequency offset $\omega_{\text{off}}$ might be comparable with or larger than the modulation depth in using AOMs and it would be more desirable for the frequency offset of a tracking laser beam from the cw frequency reference to be kept as small as possible. Such frequency offset can be completely compensated for, in principle, by use of an additional AOM in series operating at the opposite diffraction order. In our demonstrative experiment using a single AOM, however, the existence of frequency offset would cause little harm since the amount of the offset is well within the bandwidth of the pulsed laser probe beam.

If one designates $\omega_{\text{trk}} = \omega_{\text{L}} + \omega_{\text{off}}$ as the tracking target frequency a cavity feedback control, to maintain the resonance voltage $V_{\text{res}}$ at the nil tracking voltage $V_{\text{trk}} = 0$, will meet the purpose. In order to adjust the cavity to be resonant at the frequency $\omega_{\text{trk}}$, one should make a correction in the cavity length by exerting a feedback voltage $\delta V_{\text{PZT}}$ on the piezoelectric device (PZT) on which one of the cavity mirrors is mounted. For the error signal in frequency discrimination, given by $\omega_{\text{err}} = \omega_{\text{res}} - \omega_{\text{trk}} = k(V_{\text{res}} - V_{\text{trk}})$, the feedback voltage applied to PZT must be $\delta V_{\text{PZT}} = -(\omega_{\text{err}}/\omega_{\text{FSR}})V_{\text{FSR}}$, where $\omega_{\text{FSR}}$ is the free spectral range (FSR) of the cavity and $V_{\text{FSR}}$ is the voltage required for a single FSR excursion of the PZT. The practical implementation of this feedback loop can be made with electronic circuitry performing an operation of $V_{\text{PZT}} = -\int g(V_{\text{res}} - V_{\text{trk}})\,\mathrm{d}t$ with a system-dependent gain factor $g$. The bandwidth of this feedback loop and the system noise will determine the quality of cavity resonance tracking and hence the frequency uncertainty in pulsed CRDS.

## 3 Experimental apparatus

The cavity-tuned pulsed CRDS spectrometer consists of a pulsed laser source for ringdown cavity excitation, a high-finesse cavity, a sample gas chamber capable of gas feeding and evacuation, a ringdown signal detection system. With the aid of a computer controlled interface, the spectrometer automatically performs the tasks including the laser frequency scan and monitor, the cavity resonance tracking, the temperature stabilization of the sample-contained cavity, the acquisition of digitized ringdown signals, and the spectral data processing through numerical curve fitting. The optical elements and apparatus used for the construction of the cavity-tuned pulsed CRD spectrometer are described in this section.

### 3.1 *Tunable narrow-bandwidth pulsed laser source*

A pulsed dye laser source was employed for the cavity excitation, which was implemented by using a direct pulsed amplification of a tunable cw narrow-bandwidth dye laser source [25–27]. The characteristics of an amplified laser pulse output thus possess both the enhanced power and the high spectral qualities of a cw laser. The laser system of this kind is simple in principle and gives good single-mode stability during the frequency scan. In this study, a four-pass dye laser amplifier was used as depicted in Fig. 2 [28], which improved the efficiency of pulsed amplification and thus allowed a high energy output. A cw ring dye laser system (Coherent 899) was used for a tunable narrow-linewidth single-frequency radiation source in the visible region. The laser system could offer a well-characterized beam of 1 MHz bandwidth and a continuous 30 GHz frequency scan with 7.5 MHz resolution under a computer-interfaced control. This ring dye laser was pumped by a single-mode cw $Ar^{+}$-ion laser (Spectra Physics 2080A-12) with the nominal power of 5 W. A four-pass dye laser amplifier used a gain medium of $1 \times 10^{-4}$ M Rhodamine 590 in methanol contained in a dye cell similar to the prism cell developed by Bethune. The cell had a bore size of 30 mm length $\times$1.5 mm diameter. The gain medium was transversely pumped with a frequency-doubled Nd : YAG laser (Quantel YG661-10) with a repetition rate of 10 Hz. The pump laser pulse with duration of 12 ns was produced by the temporal stretch of an original pulse of 8 ns duration, which was done by splitting the pump pulse into two pulses with the ratio of 3 : 7 and recombining at the dye cell after a 5 ns delay

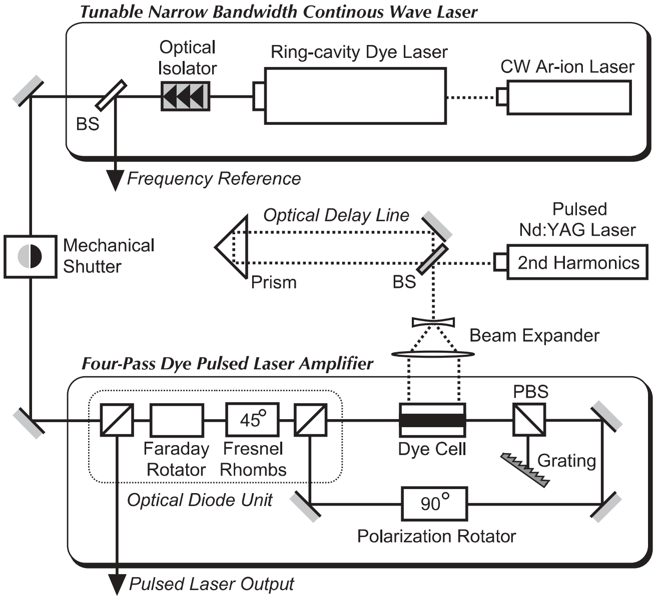


**FIGURE 2** Optical layout of the tunable narrow-bandwidth pulsed dye laser source consisting of a stabilized cw seed laser and a pulsed amplification unit

of the stronger one. This improved the bandwidth of the amplified output pulses by both temporal stretching and lowering of the peak electric field strength present in the cell. The cw injection seed beam was chopped before the amplifier entrance, using a fast mechanical shutter with an open interval of 1 ms, to prevent the cw beam from remaining in the dye cell during most of time interval when it was not pumped.

The four-pass amplifier produced a 1.5 mJ output pulse in the duration of 7 ns by the direct pulsed amplification of a cw 100 mW dye laser near 570 nm using a doubled Nd : YAG pumping energy of 5.6 mJ. As characterized in the previous work [28], the energy output characteristics was measured as a function of pumping energy, and the optimal gain reaching $\sim 10^6$ and the efficiency of 27% was obtained. The output spectrum was observed by the single-grating monochromator (Jobin-Yvon THR 1000) with resolution of $< 0.05$ nm, implying the successful amplification of a cw injection seed laser in high spectral quality. The portion of amplified spontaneous emission (ASE), in the presence of the cw injection seed laser, was kept below $\sim 1.5\%$ of the total energy of a pulsed laser output. To estimate the bandwidth of a pulsed output, a confocal scanning Fabry–Perot interferometer (Burleigh) with 3 GHz FSR and a finesse of 125 was used. The pulse bandwidth was determined as 130 MHz in full width at half maximum (FWHM), which is narrow enough for the cavity-tuned CRD experiment in this study using a ringdown cavity with the resonance mode spacing of larger than 500 MHz.

### 3.2 *Ringdown cavity*

A Fabry–Perot cavity was constructed on a stainless-steel body, serving simultaneously as a sample gas chamber with a gas feed line and a vacuum pump system. Supermirrors (Research Electro Optic, Inc.) used were high-reflectivity ($R > 0.9999$) mirrors which were stated to have 4 m radii of curvature with 0.5 inch diameters. Following the consideration given for a ringdown cavity, dummy surface sides of the mirrors were worked further to have wedge angles of $\gtrsim 20$ mrad with the surface of mirror coatings. This procedure was needed to eliminate the cavity mirror substrate etaloning, otherwise a ringdown time undulation with an oscillation period of about 17 GHz and of amplitude 3% would occur. The cavity mirror separation was set to be $L = 27.5$ cm and fine control of the cavity length for maintaining cavity resonance was made by applying a high voltage to a hollow cylindrical piezoelectric transducer (PZT) on which the cavity rear mirror sat. The ringdown cavity had an FSR of 545.4 MHz with transverse mode separations of 64.7 MHz (0.12 FSR), leaving the possibility of coupling of our pulsed laser source of 130 MHz FWHM bandwidth through several transverse modes.

### 3.3 *Cavity resonance tracking servo*

To quantify the erroneous cavity detuning from a resonance designated with the reference laser frequency at $\omega_L$, a tracking laser beam of about 10 mW was adopted from the cw injection seed laser beam and then frequency modulated before coupling into the cavity. An acousto-optic modulator (AOM) from Neos Technologies, Model N23080, was used to modulate the frequency of the tracking laser beam with the modulation depth of $\Delta\omega = 20$ MHz-pp and an offset of 80 MHz. The modulation was synchronized to a sweep function generator that delivered 100 Hz triangular waves in 10 $V_{pp}$ to the AOM driver. The tracking laser could thus be resonant with a detuned cavity at a certain moment when the shifted frequency became equal to a resonance frequency of the detuned cavity. A momentary optical transmission was detected by a 125 MHz-bandwidth photodiode (New Focus 1801) with a built-in signal amplifier providing the conversion gain of $2.4 \times 10^4$ V/W. The transmission signal burst then triggered the feedback servo to acquire the erroneous resonance frequency, which was obtained in terms of the modulation ramp voltage $V_{res}$ read from the sweep function generator. Since the destination frequency of tracking can be related to the tracking reference voltage $V_{trk}$ as well, the amount of correction needed for the length of a detuned cavity is proportional to $V_{res} - V_{trk}$. Thus the cavity was given a feedback applied for the PZT bias correction by $g(V_{res} - V_{trk})$ with a properly adjusted gain $g$. For the purpose of preliminary testing of this study, the feedback mechanism was simply implemented on a personal computer with analog input/output ports through the A/D & D/A interface board (National Instrument Lab-PC$^+$; 12 bit resolution, 50 kHz bandwidth). A servo control program was running on this computer to read error signals and produce the analog outputs for biasing the PZT through the high voltage amplification offered by the PZT driver from Burleigh (Model RC 45). Having a frequency modulation depth of 20 MHz which is much smaller than the cavity FSR of $\sim 545$ MHz, the feedback servo should operate with the help of a resonance search loop. The PZT required a bias voltage of 160 V for the cavity length scan of a single free spectral range (FSR), and the cavity resonance tracking was performed over a bias voltage range of 400 V, corresponding to a cavity frequency excursion of 2.5 FSR.

### 3.4 *Signal detection and spectral data acquisition*

For the sensitive and high-speed detection of ringdown signals, a photomultiplier tube (Hamamatsu R955) was employed as a photodetector, providing the conversion gain of $6.8 \times 10^5$ A/W with the dark current level at 3 nA and the bandwidth of 35 MHz using a 600 Ω load resistance. The analog ringdown signals were digitized by the A/D converter for appropriate bandwidth and bit resolution. In our CRD measurement that dealt with relatively long decay times in the order of $\sim 10\,\mu$s, a PC built-in 12 bit data acquisition board (Gage Applied Sciences, CompuScope 1012) with a bandwidth of 10 MHz and a sampling rate of 20 Ms/s was used. The transferred ringdown signals were then fitted to exponential decay curves numerically by a least-chi-squares fit based on the Levenberg–Marquardt method. The resultant ringdown times were processed to determine the cavity loss at each step of a frequency scan. The 30 GHz scan of a pulsed laser probe was performed by applying an external control voltage variation to the cw ring dye laser with a minimum frequency step of 7.5 MHz. Compensating the minute discrepancy between the external frequency control voltage and the scanned laser frequency, we used a wavemeter (Burleigh WA-1500) to calibrate the actual laser frequency of the scan. At each step of a frequency scan, the control voltage and the frequency reading from the wavemeter were recorded and, after the scan, their relation was numerically fitted to a polynomial to be used in compensating for nonlinearity and discarding of abrupt frequency jumps.

## 4 Results and discussions

### 4.1 *Performance of cavity resonance tracking*

The cavity resonance tracking servo was operated and examined for its feasibility and stability. As displayed in Fig. 3, resonantly coupled transmission signals were monitored with respect to the frequency modulation signal. The 10-$V_{pp}$ triangular ramp signal corresponds to the linear frequency modulation of a tracking laser beam from 70 MHz to 90 MHz with the modulation center frequency of 80 MHz. In this experiment, the tracking target voltage was set at the nil ramp voltage $V_{trk} = 0$, which means that a cavity resonance is kept tuned to the tracking laser beam shifted by 80 MHz from that of the cw injection seed laser. The quality of the cavity resonance tracking was satisfactory in the presence of mechanical noise and thermal drifts in usual laboratory environments; the cavity resonance could be kept within the frequency jitter of $\sim 5$ MHz-rms and the tracking at a given target frequency was observed not to fail within several hundreds of seconds.

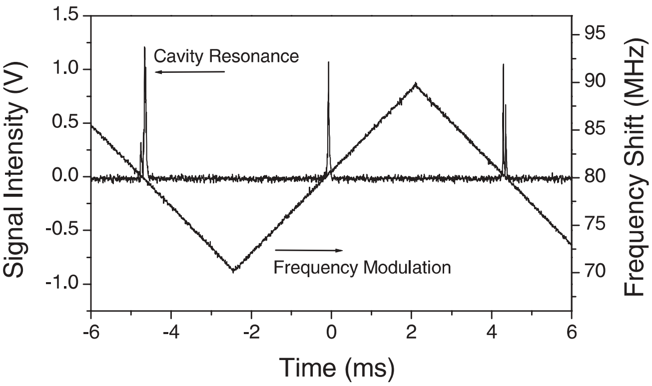


**FIGURE 3** Cavity resonant signals from a frequency-tracked cavity with frequency-modulated tracking laser input

### 4.2 *Optimization of transverse mode matching*

The transverse mode matching of a pulsed laser output is of importance in two respects: (i) the unambiguous determination of a ringdown time and hence the minimum uncertainty in measuring the cavity loss, are only warranted for CRD signals in single exponential decay, and (ii) the extreme resolution limit achievable with the pulsed CRDS can be realized on condition that a single cavity mode is excited by a finite-bandwidth pulsed laser through lowest-order transverse mode matching. Nevertheless, this is not so trivial because a convenient and reliable way to do the mode matching has not yet been well developed for pulsed CRDS.

In this experiment, the transverse mode matching was given special care in three steps: Firstly, the mode matching optics were chosen and put in place according to the theoretical prediction based on the beam parameters of the pulsed laser beam which were carefully measured with a CCD camera. Secondly, the cavity mirrors were best aligned to the cw tracking laser beam which was concomitantly transformed and aligned following the conventional mode matching procedure for cw laser beams. The pulsed laser beam was then aligned to overlap perfectly with the optical path taken by the cw tracking laser beam which could be presumed to be in the best mode match with an ideal cavity alignment. The pulsed laser beam was further transformed by adjusting the mode matching optics to let the beam parameters be similar to those of the cw tracking laser beam at arbitrary positions. Thirdly, the mode matching was optimized to the finest degree by taking the strategy developed recently by Lee et al. [29], in which the non-degeneracy in the transverse mode eigenfrequencies is brought about by the intentional asymmetry induced from a slightly misaligned cavity, and the non-degenerate transverse mode beating obviously reveals the signature of even a small transverse mode excitation.

By carrying out such an optimization of transverse mode matching, the length of a ringdown cavity can be extended up to the point where the cavity FSR becomes equal to the full span of a pulse laser spectrum. We also attempt to relax the constraints on the cavity length and the mirror curvature that have been previously configured to give sufficiently wider cavity transverse mode spacings compared with given pulse laser bandwidths.

The mode matching of a pulsed laser beam was finely adjusted in such a way that the beat oscillation manifested at an eigenmode splitting frequency showing minimum amplitude, as illustrated in Fig. 4. The cavity alignment was then recovered to its original condition where the beat oscillation could vanish completely. To avoid inaccuracy in recovering the cavity alignment, the ideal cavity alignment was double-checked by observing the cavity transmission of the cw laser beam input from the ringdown cavity whose resonance frequency

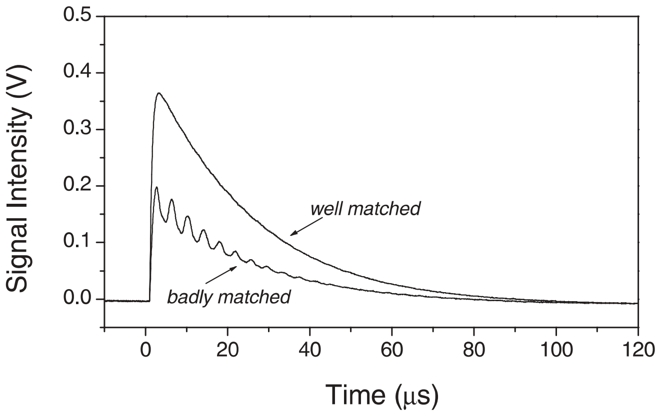


**FIGURE 4** Non-degenerate transverse mode beating signatures observed for the optimization of transverse mode matching

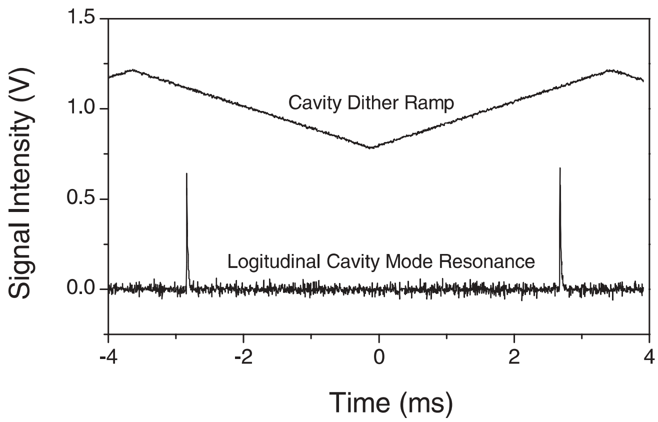


**FIGURE 5** Cavity transmission of a cw laser beam through a dithered ringdown cavity. Resonant coupling is permitted only for the fundamental transverse mode, which indicates both the perfect mode matching and the cavity alignment

was dithered over an entire FSR. As confirmed in Fig. 5, the cavity alignment was nearly perfect in the experiment to produce single resonance transmission through the lowest-order transverse mode. The transverse mode matching procedure we performed here will ensure single cavity mode excitation, even for pulsed laser inputs of relatively wide bandwidth that spans over several higher-order transverse mode frequencies. The better the traverse mode matching achieved, the less stringent the conditions required for the cavity FSR and transverse mode spacings are.

### 4.3 *Cavity-tuned ringdown signal measurement*

Acquisition of cavity-tuned ringdown signals was performed with the cavity transmission output in the vertical polarization state, which was isolated from the procedure of cavity resonance tracking done in the horizontal polarization.

With the 130 MHz-bandwidth pulsed laser excitation by an energy of 5 μJ per pulse, we could record ringdown traces of single exponential decay as displayed in Fig. 6. In practice, the ringdown signals were averaged to enhance the measurement sensitivity; the 16 shot averaged signals could reduce the single-shot noise of a ringdown signal from −23 dB to −29 dB and, in turn, the resulting uncertainty involved in the determination of the ringdown time. By fitting the ringdown

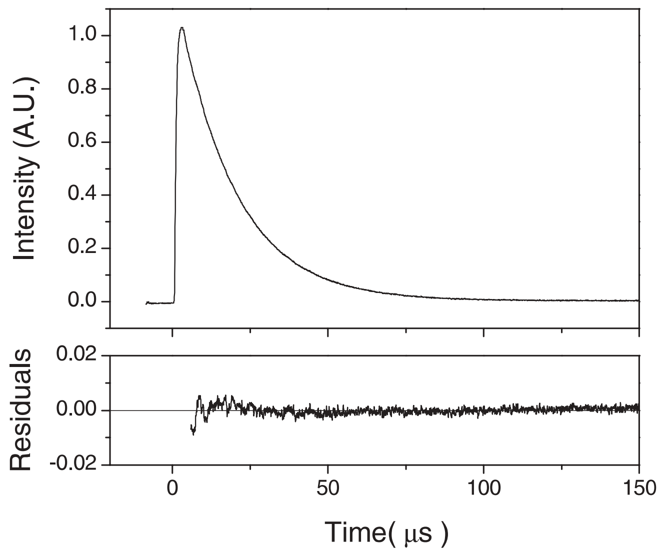


**FIGURE 6** Typical 16-shot-averaged ringdown signal waveform experimentally measured. The curves of exponential decay fit to the experimental and the resulting residuals are plotted in the inset of each figure

signals, the average of the ringdown time for the empty cavity was determined to be about 20 μs, which directly led to the single-pass cavity loss of $\mathfrak{L}_c = 4.58 \times 10^{-5}$ and a cavity finesse of about 68 500.

The ringdown signals were produced reliably with constant amplitudes by virtue of cavity resonance tracking to a narrow-bandwidth pulsed excitation. The effective probe frequency, equal to the frequency of a cavity resonance mode, was set at a frequency apart from the center of a pulse laser bandwidth by a fixed amount. To verify the feature of single-cavity-mode coupling of a pulsed laser input, the cavity resonance tracking mechanism was intentionally disabled and ringdown signals were observed by varying the cavity resonance frequency. As implied by the result given in Fig. 7, the amplitude of ringdown signals clearly showed a dependence on the cavity detuning, which means the bandwidth of a laser pulse localized well within a cavity longitudinal mode spacing. Reaching the exact resonance condition, the amplitude of ringdown signals becomes nearly twice that of cavity-tuned ringdown signals obtainable with the cavity tracking servo

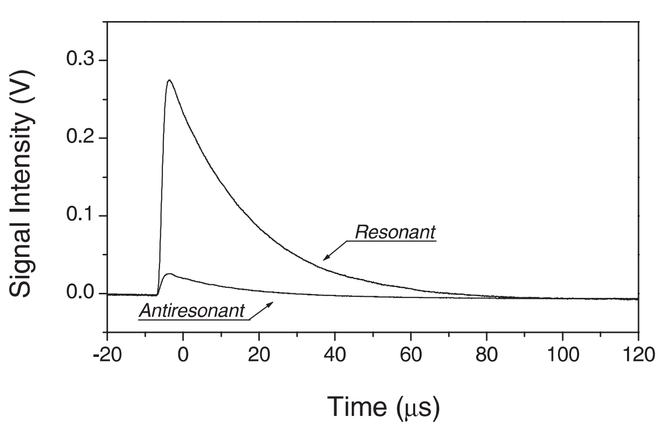


**FIGURE 7** Comparison of ringdown signals with different coupling amplitudes depending on the location of a cavity mode frequency

running for an 80 MHz-shifted target frequency. On the other hand, for anti-resonance, ringdown signal amplitudes are considerably reduced by a factor of about 15.

In the detuning behavior observed, undesirable coupling via more than one cavity mode was found to take place slightly. Since the possibility of higher-order transverse mode excitation can be excluded based on our careful efforts given to the mode matching, coupling through the nearest neighbor longitudinal cavity modes might be one of the strong suspects. Though the bandwidth of an excitation laser pulse was 130 MHz in FWHM, the wide sideband tails of a Lorentzian-like excitation spectrum might be somewhat broad to yield such residual transmission in a cavity with 545 MHz FSR. From a rough estimation, however, the residual sideband transmission in case of exact cavity resonance, can be further suppressed and be no higher than 2% with respect to the major coupling through a desired longitudinal cavity mode.

### 4.4 *Spectral recording of molecular overtone transitions*

The performance of our CRD spectrometer was tested by measuring the absorption spectra of the molecular overtones of acetylene ($C_2H_2$) near 570 nm. The same spectra have been recorded previously by Romanini et al. [5], and Hahn et al. [6], using the cw-CRDS measurements. Acetylene gas was filled up in the evacuated ringdown cavity up to a pressure of 50 Torr. The ringdown time measurement was made for a 30 GHz scan of the laser frequency by a resolution of 75 MHz, beginning from the wavenumber of 17 552.61 cm$^{-1}$. The ringdown time was measured at each frequency step and we could obtain the absorption spectrum of the gas after subtracting the ringdown time baseline of an empty cavity.

As plotted in Fig. 8, the spectrum obtained by the cavity-tuned pulsed CRDS is being overlapped and compared with that recorded by Hahn et al. [6]. The comparison indicates that the absorption spectra obtained by the cavity-tuned pulsed CRDS is in good quantitative agreement with that of cw-CRDS. The detection sensitivity achievable with this prototype cavity-tuned pulsed CRDS system was found to be

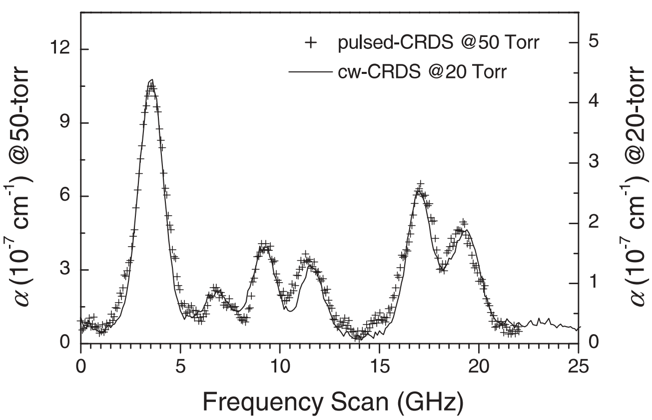


**FIGURE 8** Absorption spectra acquired for the molecular overtone transitions of acetylene ($C_2H_2$) beginning from the wavenumber of 17 552.9 cm$^{-1}$. The spectrum obtained by the cavity-tuned pulsed CRDS is overlaid with the spectrum recorded by cw-CRDS for the comparison. Note that the sample gas pressure for the spectra in the two experiments are different

$\sim 2\times10^{-6}$ of per-pass absorption loss, which was still relatively poor in performance compared with the state-of-the-art CRDS.

The degraded detection sensitivity, however, is not attributed to the cost of enhancing the frequency resolution of pulsed CRDS. Instead, some technical imperfections of our own experiment, such as the residual excitation of neighboring longitudinal cavity modes and the accidental transverse mode mismatch, are thought to be responsible for the excess noise components in the measured spectra. Further improvement in the detection sensitivity along with the frequency resolution can be expected in other more careful experiments.

A ringdown cavity shorter in length than one used in this experiment would reduce the frequency noise in spectral measurements, by eliminating the residual excitation through adjacent cavity modes with an extended separation. In addition, the non-stationary cavity misalignment that is likely to occur in the course of cavity scanning by PZT tube should be compensated for to avoid the fluctuation of ringdown time baseline. The primary cause of the cavity misalignment is inferred in the fact that biasing PZT not only changes the tube length but also accompanies a minute cantilever deformation from the optical axis. The non-stationary cavity misalignment induces a transverse mode mismatch and, in turn, a variation in the effective cavity finesse due to the parasitic transverse modes. The measurement of the ringdown time fluctuation in vacuum as a function of the PZT bias voltage monitored in cavity resonance tracking, exhibited evidence for the dependence of ringdown times on the PZT bias voltage. Since noise of this kind is quite system-dependent and is not reproducible, choosing an ideal PZT of either minimal bending deformation or three-segment sectioned biasing seems to be the only alternative.

In spite of unwanted noisy components observed in the measured spectra, the single cavity mode operation of pulsed CRDS was not obstructed during the data acquisition, as evidenced by experimental data given. A spectral degradation of instrumental line broadening associated with the spectral uncertainty of the pulsed laser source, did not appear to take place either, which means that the spectral recording was not perturbed by either the bandwidth or the frequency jitter of the pulsed laser source. Otherwise, a sizable spectral broadening, on the order of 130 MHz associated with the pulsed laser bandwidth, must have manifested on the spectral shape of the absorption features. The acquisition of ringdown signals would also have been unstable and suffered from the fluctuation of ringdown signal amplitudes, which, however, was not observed to be so. The experimental results indicate that the cavity-tuned pulsed CRD spectrometer is capable of recording high-resolution spectra containing absorption features of narrow linewidths, comparable with the bandwidth of a probing pulsed laser source.

## 5 Conclusions

As a refinement of CRDS for high resolution, a new cavity-tuned ringdown setup using pulsed laser excitation has been proposed. Distinguished from conventional pulsed CRDS configurations, our experimental implementation has employed a novel design of a narrow bandwidth pulsed laser source with a well-defined frequency reference, a closed-loop cavity resonance tracking servo, and a reliable procedure of transverse mode matching optimization. A pulsed laser beam of 130 MHz bandwidth generated from direct amplification of a tunable cw injection seed laser, has been reliably tuned to a single resonance mode of a ringdown cavity with 545 MHz FSR. Single cavity mode operation within the frequency tracking error of less than 5 MHz has been successfully accomplished during the frequency scan of the cw injection seed, overcoming the conventional resolution limit imposed by the frequency bandwidth of a pulsed laser excitation.

The spectrometer implementation has been tested by the spectral recording of the ultraweak molecular absorption features of acetylene, around a wavelength of 570 nm. This has demonstrated that the proposed cavity-tuned pulsed CRD spectrometer is feasible for measuring high-resolution spectra of a narrow absorption feature comparable with the bandwidth of a probing pulsed laser source. The absorption spectra measured by the cavity-tuned pulsed CRDS has been found to be in good quantitative agreement with the previous result of cw-CRDS measurements, allowing a prototype performance in the detection sensitivity of $\sim 2\times10^{-6}$ for per-pass absorption loss. The demonstrated detection sensitivity in the presence of some technical imperfections of our own experiment has been found, as evidenced by experimental data, to neither, be a cost of the procedure enhancing the frequency resolution, nor to obstruct successful single cavity mode ringdown events. In a more careful experimental setup, the detection sensitivity will be further improved along with the frequency resolution.

What we suggest for the excitation source of high-resolution pulsed CRDS is any pulsed laser radiation of bandwidth narrower than a cavity FSR, generated from a tunable cw laser source that is able to serve simultaneously as a definite reference for frequency discrimination. Our experimental demonstration with an excitation source based on direct pulsed amplification, where wavelengths of a tunable cw seed laser and a resulting pulsed source are the same, is one of the simplest forms of such novel pulsed laser sources. The proposal in this study is thus readily extended to using pulsed laser sources constructed by either harmonic conversion after the seeded amplification or seeded optical parametric oscillation (OPO). The final wavelength of these pulsed lasers is different from that of cw seed lasers, but has a definite wavelength relation so that the cavity resonance tracking procedure can be carried out by using a cavity feedback servo with a simple addition for frequency offset of the tracking frequency reference. Therefore high-resolution CRDS studies are expected to become more feasible in the spectral region ranging from ultraviolet (UV) to mid-infrared (IR).

Finally, we summarize promising features of the high-resolution pulsed CRDS. As a matter of course, high-resolution spectral measurements can be conducted in the spectral region where tunable cw lasers are not directly available as in the UV or mid-IR region [9–12]. Even when excellent cw lasers are available at the wavelength of interest, advantages of the pulsed CRDS scheme are many. To incorporate CRDS with nonlinear absorption techniques for sub-Doppler resolution [15, 16], phenomena such as saturation, and two-photon absorption, are exploited and one needs to specify exactly the time evolution of optical intensity in the sample-contained cavity, as well as the dynamics of the absorber population being analyzed. In this sense, pulsed excitation schemes are more accessible and reliable than the cw ones since pulsed lasers could permit experimentally well-defined initial conditions of the intracavity optical intensity for the quantitative analysis of entire ringdown events. A swept-cavity or a locked-cavity CRDS using cw lasers, on the other hand, might be liable to ambiguities associated with practical fluctuations in both the optical intensity and the absorbing population density at the forefront part of every ringdown event. For a dynamic study of transient samples [13, 14], cavity ringdown events triggered by excitation laser pulses are easily synchronized to pump–probe measurements, while swept-cavity cw-CRDS schemes usually suffer a timing jitter at the moment of resonance coupling of an excitation laser. The pulsed CRDS scheme is also promising from the view point of metrology, because the swept-cavity CRDS schemes using cw lasers might be problematic due to the spectral degradation by intracavity Doppler shifts of the order of up to a few hundred MHz [5, 6]. Our high-resolution pulsed CRDS is free from the intracavity Doppler error since the cavity resonance issue is resolved through a modulation of tracking laser frequency rather than a cavity length, offering the steady nature of the cavity mirrors. In many respects, as mentioned, the high-resolution pulsed CRDS that is based on a reliable single cavity mode operation will be able to play an increasing role as a workhorse in quantitative absorption measurements, complementary to high-sensitivity cw-CRDS techniques.

**ACKNOWLEDGEMENTS** This work was supported by the grant No. R01-2000-000-00041-0 from the Korea Science and Engineering Foundation.

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