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Microcontroller based resonance tracking unit for time resolved continuous wave cavity-ringdown spectroscopy measurements

Ondrej Votava,[1,a)] Milan Mašát,[1] Alexander E. Parker,[2] Chaithania Jain,[2] and Christa Fittschen[2]
[1]*Academy of Sciences of the Czech Republic, J. Heyrovský Institute of Physical Chemistry, Dolejškova 2155/3 18223 Prague 8, Czech Republic*
[2]*Laboratoire de Physico-Chimie des Processus de Combustion et de l'Atmosphère – CNRS UMR 8522, Université Lille 1 Science et Technologie, 59655 Villeneuve d'Ascq, France*

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We present in this work a new tracking servoloop electronics for continuous wave cavity-ringdown absorption spectroscopy (cw-CRDS) and its application to time resolved cw-CRDS measurements by coupling the system with a pulsed laser photolysis set-up. The tracking unit significantly increases the repetition rate of the CRDS events and thus improves effective time resolution (and/or the signal-to-noise ratio) in kinetics studies with cw-CRDS in given data acquisition time. The tracking servoloop uses novel strategy to track the cavity resonances that result in a fast relocking (few ms) after the loss of tracking due to an external disturbance. The microcontroller based design is highly flexible and thus advanced tracking strategies are easy to implement by the firmware modification without the need to modify the hardware. We believe that the performance of many existing cw-CRDS experiments, not only time-resolved, can be improved with such tracking unit without any additional modification to the experiment. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.3698061]

I. INTRODUCTION

Cavity ring-down absorption spectroscopy with continuous wave lasers (cw-CRDS) is a highly sensitive spectroscopic absorption technique that finds a wide range of applications such as trace gas analysis, high resolution spectroscopy, plasma and flame diagnostics or chemical kinetics. The method is based on measurements of photon decay rates in a high finesse Fabry-Perot resonators: when the laser is coupled to a high finesse optical cavity and the incoming beam is switched off by a fast optical switch, an exponential decay (ring-down) of intensity is observed at the resonator output.[1–3] Time constant of this exponential decay is determined by the resonator parameters – mirror reflectivities and resonator lengths – and also by absorption of the medium placed inside the resonator. Absorption sensitivities below 10^{-10} cm^{-1} have been demonstrated.[2,4,5]

The cw-CRDS method is becoming especially popular with the compact and inexpensive tunable diode lasers as the light sources, enabling construction of very compact spectrometers with high resolution and extreme sensitivity.[3,6] Success of the method with those low-power laser diodes (typically from 1 mW to 100 mW power range) depends on efficient laser radiation coupling to the resonator. This can be achieved by matching the laser frequency to one of the resonator longitudinal modes to induce the power build-up in the resonator. Typically, the resonator length is adjusted using a precision piezoelectric transducer (PZT) until the resonance condition $2l = n\lambda$ is fulfilled, where l is the resonator length, λ is the laser wavelengths, and n is an integer number. Due to constructive interference at the resonance condition significant power buildup in the resonator is achieved.

Two basic strategies have been implemented to achieve this laser-resonator frequency matching: the locked cavity and swept cavity variations. In the first approach the laser frequency is locked to the cavity mode using a feedback servoloop control. Stabilization schemes such as Pound-Drewer-Hall have been used for this purpose.[7,8] Optical feedback locking has also been used for efficient coupling.[4] While this locked cavity cw-CRDS has been successfully demonstrated by several groups and potentially provide very efficient cavity buildup and high ring-down repetition rate, the technical difficulties associated with implementaion of this techniques quite often outweight the benefits.

In the alternative strategy the cavity lengths are continually swept by applying a voltage ramp to the PZT and the ring-down measurements are initiated every time a cavity mode is matched to the laser frequency. This frequency match is manifested by a rapid increase of the transmitted laser intensity through the resonator. A threshold circuit is used to detect this sharp intensity rise and produce a signal to the fast optical switch which turns the incoming laser off at preset buildup level to observe the ring-down decay. Compared to the locked cw-CRDS the swept-cavity approach proves to be robust and easy to implement in a wide range of applications and has become rather wide-spread over past several years.

Time resolved absorption experiments such as, for example, flash photolysis kinetics experiments,[6,9–13] where concentrations of reactants, reaction intermediates and/or products are monitored as a function of time after a reaction is initiaded by laser pulse require fast sampling rates. Temporal resolution of the cw-CRDS technique is in principle limited

a)Author to whom correspondence should be addressed. Electronic mail: ondrej.votava@jh-inst.cas.cz.

by the ring-down time: if absorption coefficient changes on shorter time scale, the single-exponential character of the ring-down decay signal will be distorted. To achieve the best time resolution in cw-CRDS experiment the next cavity power build-up would have to be initiated right at the end of the previous ring-down measurement to maximize the sampling rate. In practice, however, the temporal resolution is limited by delays between the individual ring-down masuring events. Hence to achieve high temporal resolution, those delays must be minimized.

In the most commonly used implementation of the swept-cavity cw-CRDS technique a saw-tooth voltage signal is applied to the PZT to repetitavely modulate cavity length over slightly more than one free spectral range (FSR). It is therefore ensured that at least one resonance between the laser and a cavity mode is observed per sweep. Such full-FSR sweep technique is very easy to implement but has limitations in terms of the repetition rate of the ringdown events. Indeed the PZT has to sweep the distance $\lambda/2$ that is hundreds nanometers, before next resonance is encountered. This typically takes several milliseconds while the ringdown event detection window ranges between 10 and 100 μs depending on the cavity length and mirror reflectivities. The measuring duty cycle under those conditions is therefore on the order of few percent at maximum.

Much more efficient approach is based on cavity tracking. In this approach the PZT is dithered over a range that is significantly narrower than the cavity FSR with the sweep range centered at the resonance. Ring-down repetition rate then depends on the modulation amplitude and sweep rate. This approach has been first reported by Romanini *et al.*[2] and also used in other experimental setups.[12,14] To keep the resonance at the center of this narrow modulation range a feedback servoloop is used to adjust a dc offset on the PZT to compensate for slow drifts of the cavity lengths and/or for the laser tuning.

Dedicated analogue tracking circuits based on a phase-locked-loop (PLL) which detects the phase shift between the dither signal and the resonance have been used for this purpose in the past. The PLL produces an error signal with zero crossing for the resonance in the center of the dither range which is integrated to produce the dc offset voltage. Disadvantage of this approach is the slow re-locking after the tracking is lost due to, for example, sudden disturbance of the cavity length. By design, the dc offset must be adjusted at a slow rate compared to the dither frequency. Therefore, if the resonance is lost due to a sudden cavity disturbance, re-locking is slow and the ringdown events may be lost for considerable time, until the slowly sweeping dc offset moves to the next cavity resonance. Re-locking times of typically 1 s have been noted in the literature.[2] This may be a severe limitation in time resolved experiments where high repetition rate along with fast re-locking is required to maintain fast data acquisition.

In this paper we describe a new design of the cavity tracking feedback circuit for high rate cw-CRDS measurements required for time dependent absorption experiments. The technique is based on rapid control of ramp direction in vicinity of the resonance position. Such approach provides simple and robust alternative to the existing mode tracking techniques. It can be easily implemented into existing experimental setups without any need for optical layout modifications. Compared to previously used analog tracking schemes, much faster relocking is achieved when the tracking is lost due to external disturbances.

The design is based on single chip microcontroller unit (MCU) that controls both PZT dithering and tracking. This solution provides high level of flexibility: various tracking strategies can be implemented by simply modifying the microcontroller software without the need to change the circuit layout. Number of operation modes can also be pre-programmed and easily software selected during the operation. Use of the MCU based design also makes the circuit straightforward with a low part count.

II. CIRCUIT DESCRIPTION

General layout of the cw-CRDS spectrometer, demonstrating the integration of the tracking unit within the cw-CRDS experimental setup is schematically shown in Figure 1. The CRDS cavity is formed by a pair of high reflectivity concave mirrors in a stable Fabry-Perot resonator configuration. One of the mirrors is attached to the PZT transducer for resonator length control. Laser radiation is passed from the laser through an acousto-optical modulator (AOM) used as a fast optical switch. Radiation transmitted through the CRDS resonator is detected on a photodiode (PD). As the cavity length is modulated by the PZT, photodiode signal is monitored to determine when resonances with the laser radiation occur. The incoming radiation is then rapidly switched off with the AOM and intensity decay is recorded on the photodiode, sampled by the data acquisition unit and analyzed to obtain the decay rate from which the absorption coefficient is determined. The tracking unit uses the photodiode signal as an input to detect the cavity resonances and outputs both an analog signal to the PZT to modulate the cavity length as well as digital signals to the AOM and data acquisition units.

The tracking electronics therefore consists of three major subunits: the threshold detector, the ramp generator, and

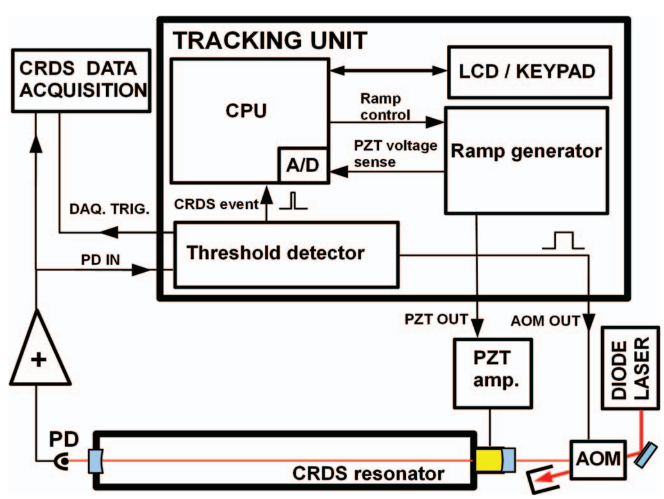

FIG. 1. Schematics of the tracking unit integration into a cw-CRDS experiment.

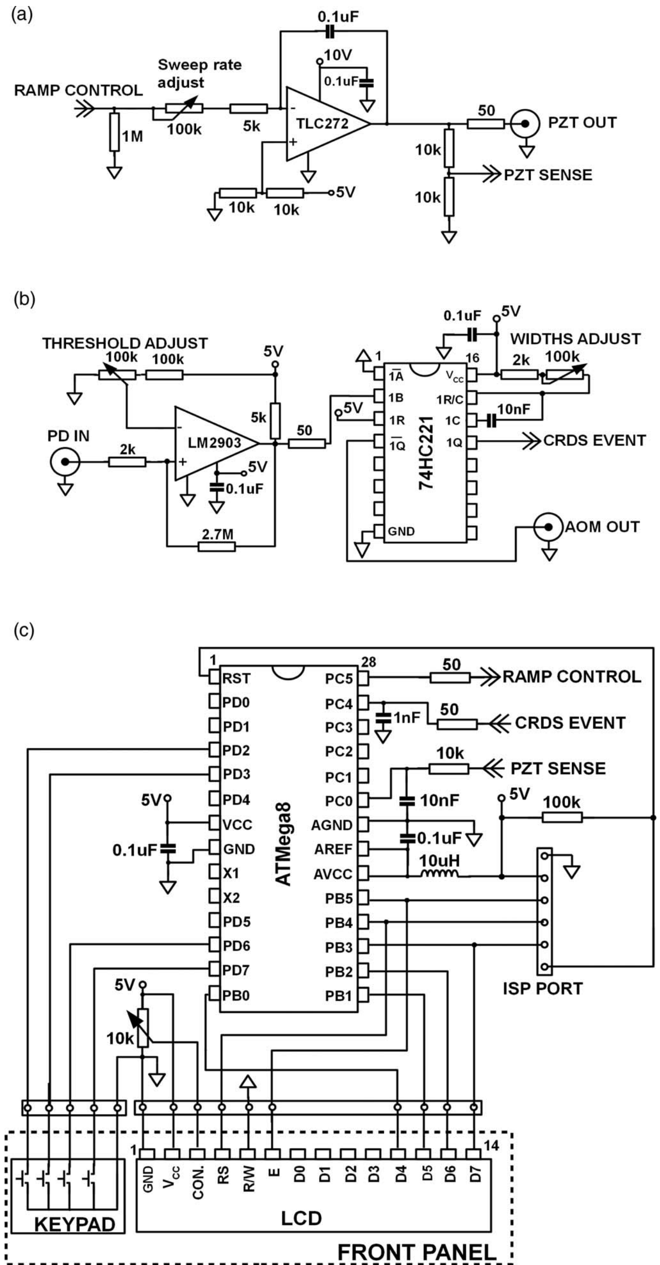

FIG. 2. Circuit diagrams of the tracking electronics sub-units: (a) ramp generator, (b) threshold generator, (c) CPU microcontroller unit.

the microcontroller unit (MCU) as indicated in the Figure 1, and detailed in Figures 2(a)–2(c). The ramp generator circuit produces up/down voltage ramp for the PZT transducer. It is based on operational amplifier integrator with direction of the ramp controlled by a transistor-transistor logic (TTL) level on the integrator input. The threshold detector unit is used to detect the CRDS events and produces a CRDS-event-trigger TTL pulse, which is send both to the MCU unit and to the outside data acquisition board. This part of circuit is realized using an analog comparator IC (LM2903) with the threshold level adjustable by a potentiometer. A single shot timer IC (74HC221) is used to produce the output pulse of desired polarity and duration for the AOM switching.

The MCU part of the circuit is based on Atmel Mega8 microcontroller. There are many possible choices of eligible microcontroller units that would serve well for this purpose from number of suppliers. The choice of this particular unit is given by the minimal external parts needed for the operation, sufficient number of programmable input/output lines, easy and straight forward in-system-programming capabilities, on-chip 10-bit analog-to-digital (A/D) convertors, and availability in the DIL package for easy prototyping. All those factors combine in an extremely universal device that is easy to work with even in rather modestly equipped electronic workshops. Indeed, the complete MCU board shown in Figure 2(c) is rather universal unit useful in number of other applications around the laboratory. The microcontroller monitors the PZT voltage via one of the A/D inputs and also senses when the digital CRDS-event-trigger is detected by the threshold unit and controls the direction of the PZT voltage ramp via one of the programmable digital output lines. The MCU unit utilizes other digital input/output lines to communicate with an external alphanumeric LCD display and uses four-button keypad for user-friendly interface.

We have implemented and tested two basic modes of operation in the microcontroller firmware that can be selected at run-time: (i) the full sweep mode and (ii) the tracking mode, respectively. In the full-sweep mode the PZT voltage sweeps between preset minimum (U_{MIN}) and maximum (U_{MAX}) voltage values. The sweep rate is set by the ramp generator hardware. Values of U_{MIN} and U_{MAX} are software selectable and therefore the amplitude of ramp can be adjusted to be slightly larger than one FSR of the CRDS cavity.

In the tracking mode the PZT is dithered around the resonance to produce high frequency of CRDS events. This is achieved by switching the ramp direction every time resonance has been detected: the microcontroller monitors when the resonance occurs via the signal obtained from the threshold detector and switches the direction of PZT ramp at a set delay after the event to opposite direction to sweep back towards the resonance. By this simple procedure the PZT voltage oscillates around the resonance position up and down switching the sweep direction each time the resonance is reached.

Figures 3 and 4 show the timing diagrams and corresponding control software logic flow-charts for those two basics modes of operation, the full sweep and tracking, respectively. In the full sweep regime the PZT voltage is swept using the ramp generator subunit and the MCU monitors its instantaneous value via the PZT SENSE line. When the voltage reaches either lower or upper bounds of the sweep range determined by user defined values U_{MIN} and U_{MAX}, respectively, direction of the MCU unit switches the PZT ramp direction via the RAMP CONTROL digital line. Values of U_{MIN} and U_{MAX} are chosen such that the resonator is tuned over more than full FSR. CRDS event detection subunit provides digital output for the AOM switch and the data acquisition card when a resonance is detected, but the MCU unit does not utilize the signal in this mode. Under typical experimental conditions the PZT sweeps the full FSR in approximately 4.5 ms yielding up to 220 CRD events/s sampling rate. The sweep rate is limited by the PZT and mirror assembly mechanical resonant frequency. Furthermore, while increasing the sweep rate could increase the CRD sampling

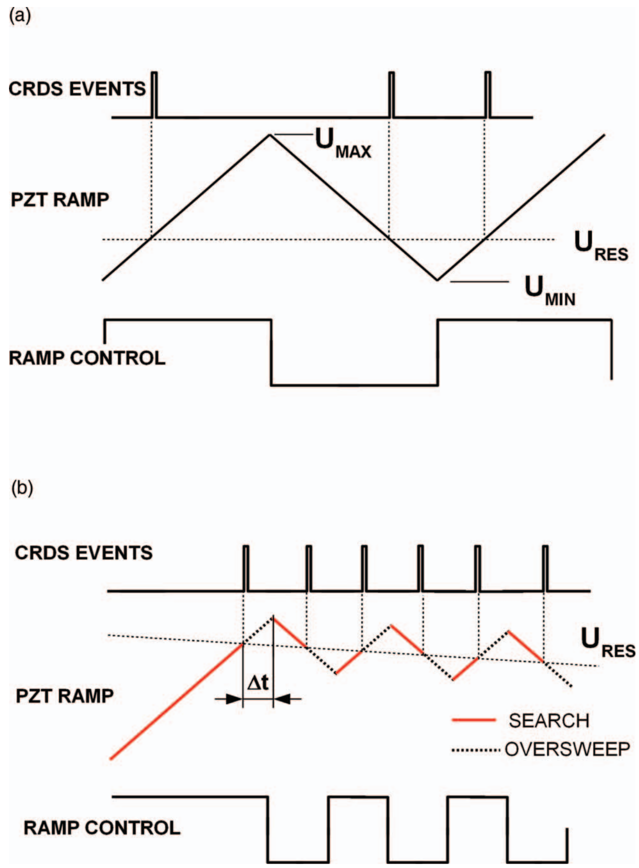

FIG. 3. Tracking unit timing diagrams for (a) the full sweep mode and (b) for tracking mode. See text for details.

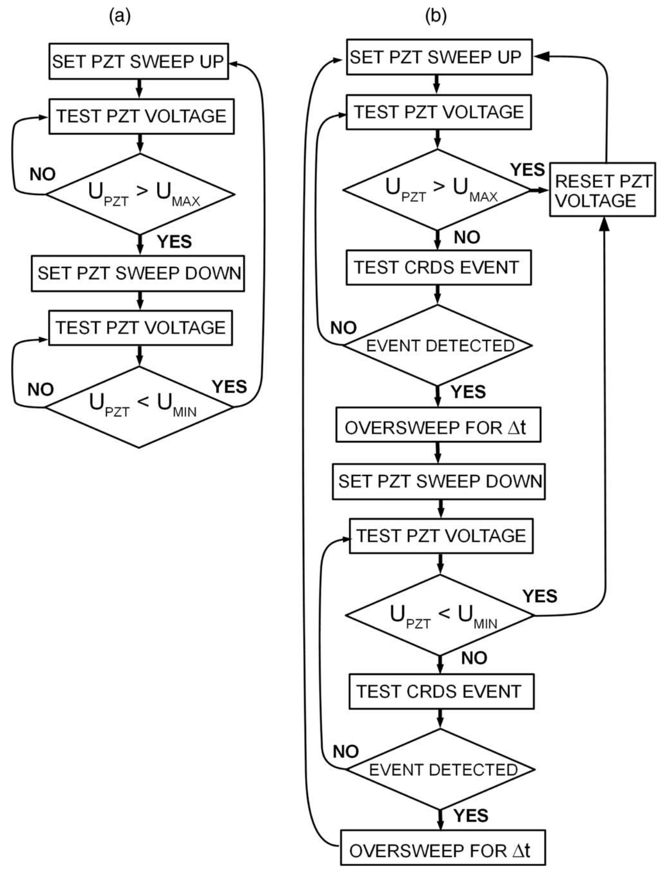

FIG. 4. Logical flowcharts of software implemented in the microcontroller to perform the full sweep (a) and tracking (b) modes as introduced in the diagrams of Figures 3(a) and 3(b), respectively.

rate, this also reduces the resonance time and thus leads to less efficient cavity filling.[15] Thus, a compromise between the cavity filling efficiency and the CRD sampling rate must often be achieved under specific experimental conditions.

The tracking mode timing diagram is depicted in Figure 3(b) and the flowchart of the corresponding tracking software is in Figure 4(b). When the CRDS cavity is far from resonance the PZT voltage is swept in the same way as for the full-sweep mode until the first resonance is detected by the threshold unit. From this point the PZT is swept in the same direction for a preset oversweep delay Δt, user adjustable via the software. At the end of this delay the PZT ramp direction is switched via the RAMP CONTROL digital line. Now the cavity moves back towards the recently detected resonance. This procedure is repeated, once the resonance is again detected. As a result the cavity now oscillates in a narrow range around the resonance. Amplitude of those oscillations is determined by the oversweep delay Δt between the resonance detection and ramp direction switching and by the PZT sweep rate. If the position of the resonance changes in time, the search period between the PZT ramp switching and detection of next resonance (red line in Figure 3(b)) is alternatively longer and shorter than Δt for the up/down sweeps, respectively, and thus center of the sweep range changes to track the resonance as depicted in Figure 3(b). By this procedure the resonance is tracked as long as its drift is slower than the PZT sweep rate.

III. PERFORMANCE

Figure 5 shows the PZT ramp and the output of the photodiode over 15 ms in both, full sweep (a) and tracking mode (b). Top traces show the PZT voltage. The cavity resonances appear as the intensity spikes on the photodiode signal (lower traces). For the full sweep example, the PZT is modulated with 110 Hz, i.e., a maximum of 220 ring-down events can be obtained per second in this configuration with one resonance per up/down ramp.

In the tracking mode it can be seen that the ramp frequency is increased and the amplitude is decreased. The ring-down repetition rate due to the tracking mode has increased by more than a factor of 3 in this particular example compared to the full sweep mode.

The data in Figure 5(b) also demonstrate the behavior of the servoloop when the tracking is disturbed. The cavity resonance marked with (*) falls below the threshold for ring-down detection (indicated by the dashed horizontal line). The direction of the ramp therefore does not change after this resonance, because it is not recognized by the threshold unit as a ring-down event. The PZT voltage instead continues to ramp down at the same sweep rate, until it reaches the lower bounds of the sweep range as set by the U_{MIN} parameter. Then the direction of the ramp is changed by the MCU and the cavity sweeps back towards the resonance position. Relocking takes place in approximately 5 ms in this case – a typical

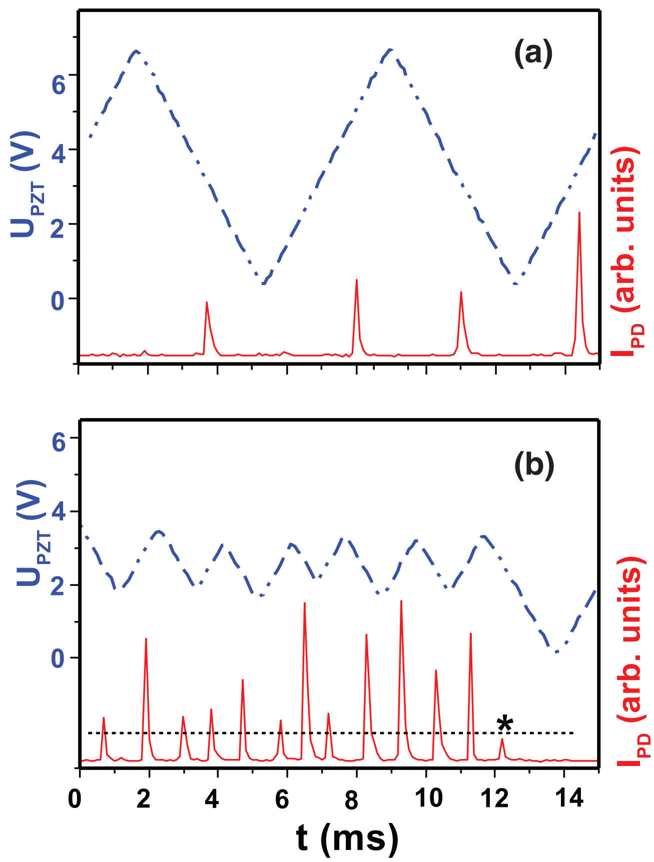

FIG. 5. Demonstration of the servoloop performance. PZT voltage (dashed line) and photodiode signal (solid line) for (a) the full sweep mode and (b) the tracking mode, respectively. The fine-dashed line in the locked mode indicates the ringdown event threshold level. Resonance marked with asterisk (*) does not reach this threshold and thus the PZT ramp direction does not change and the lock is temporarily lost.

time delay between two ring-down events in the full-sweep mode. Consequently, the average rate of the ring-down events increases using the tracking procedure compared to the full-sweep mode, even in the cases when the tracking is lost frequently due to, for example, a large frequency noise on the excitation laser which results in random amplitude fluctuations of the observed resonances.

To illustrate the advantages of this cavity tracking in time resolved cw-CRDS, Figure 6 shows typical data obtained in a flash photolysis experiment with cw-CRDS detection of HO_2 radical intermediates. Details of the experimental setup can be found in recent papers,[6,10] and thus only brief description is given here: HO_2 radicals are generated by 248 nm photolysis of H_2O_2

$$H_2O_2 + h\nu\,(248\text{ nm}) \rightarrow 2\ OH$$

$$OH + H_2O_2 \rightarrow H_2O + HO_2$$

HO_2 is a radical intermediate and will disappear typically in our experiments on the timescale of milliseconds in subsequent reactions, mainly,

$$2\ HO_2 \rightarrow H_2O_2 + O_2$$

$$OH + HO_2 \rightarrow H_2O + O_2$$

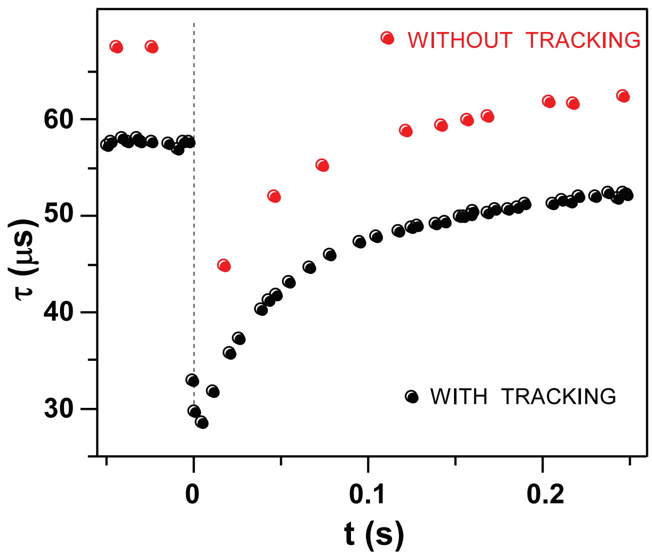

FIG. 6. Demonstration of data acquisition rate increase using the resonance tracking unit in laser photolysis experiment. Kinetics data recorded with (a) full sweep mode, (b) the tracking mode, respectively.

Figure 6 shows the evolution of the ring-down time with respect to the photolysis pulse (at time t = 0 s). Therefore, such a signal can be transformed into a concentration time profile using the following equation:

$$[HO_2]_t = \frac{R_L}{c \times \sigma}\left(\frac{1}{\tau_t} - \frac{1}{\tau_0}\right), \qquad (1)$$

where σ is the absorption cross section, R_L is the ratio between the cavity length L, i.e., the distance between the two cavity mirrors, to the length L_A over which the absorber is present (in our case the overlap of photolysis beam and cw-CRDS absorption path), c is the speed of light. Average of all ring-down events that have occurred before the photolysis pulse is the τ_0, i.e., the ring-down time in the absence of HO_2, and τ_t are the time dependent ring-down times recorded after the photolysis pulse. Both traces have been obtained after single photolysis shots: the upper trace has been obtained without the tracking system and is composed of only 12 ring-down events having passed the threshold and distributed over the 250 ms measurement time, the lower trace (shifted down by 10 μs for better visibility) has been obtained using the tracking system and is composed of a total of 49 ring-down event, 4 times more than without tracking. A clear increase in ring-down event rates is observed with the tracking system, reducing the time needed for comparable data accumulation by a factor of 4 or more, or increasing substantially the signal-to-noise ratio for the same measurement time.

IV. DISCUSION

Using the cavity tracking we have demonstrated increase of ring-down events rate in the kinetics cw-CRDS experiment by a factor of 4 compared to the full sweep mode with present experimental setup. While this already is a significant improvement, it might be useful to examine the limiting factors of this technique, as further narrowing the dither range and increasing the PZT sweep rate would be desirable for the time resolved measurements. First of all, the cavity mirror sweep rates are limited by the mechanics of the PZT-mirror assembly. The mirror is repeatedly accelerated and decelerated during the dithering. Therefore, inertia of the assembly and the force provided by the PZT limit the maximum sweep rates. To control the period of each sweep as is required for in the described tracking procedure, the dither frequency must fall below the mechanical resonance of the PZT and mirror assembly, which typically is on the order of $\sim 10^4$ Hz, depending on mirror assembly mass and electromechanical properties of the PZT.

Furthermore, there is always a compromise between the cavity sweep rate and the power buildup efficiency. The time constant for intracavity power buildup is given by the cavity response time which is the same as cavity ringdown time.[15] Therefore, only partial buildup will be achieved if the resonance time between the cavity mode and the laser line during the PZT sweep is short compared to the cavity response time. To illustrate this point, let us consider a typical example for a cavity mode spacing 300 MHz (mirror separation 0.5 m) and laser line widths 1 MHz. If the PZT sweeps the full FSR range in 5 ms the resonance time will be effectively $\delta t \sim 16\ \mu s$, which is already comparable to or shorter than typical ring-down times that range from 10 μs to well over 100 μs depending on the mirror reflectivity. Clearly, once the resonance time becomes shorter than the cavity response the buildup efficiency will decrease with increasing sweep rate.[15] It has been, for example, noted that the buildup efficiency is lower for the extended cavity diode lasers compared to the DFB lasers, because the instantaneous line widths of DFB lasers is significantly broader ($\sim$1 MHz) than for the ECDL ($\sim$100 kHz).[4] Depending on the laser linewidth and power, as well as the cavity rigngdown time, the sweep rate may need to be decreased to obtain sufficient power buildup. This will ultimately limit the ringdown repetition frequency.

Finally, the resonance tracking technique is more sensitive to external mechanical disturbances compared to the full-sweep approach. If a sudden disturbance shifts the cavity resonance outside the sweep range of the PZT dither the tracking is lost. Higher amplitude of the dither makes the tracking less sensitive to such external disturbances but at the same time leads to lower frequency of the ringdown events. Depending on the mechanical stability of the CRDS resonator and the sources of possible disturbances in the experiment the dither amplitude needs to be adjusted.

The fast re-locking feature of the presented tracking servoloop (on millisecond time scale) minimizes data loss caused by a lock disturbance. It should, however, be noted that the current servoloop scheme is at present more sensitive to isolated missing resonances compared to the PLL based analog versions. Indeed, single resonance that falls below the detection threshold breaks the lock. In the PLL servoloop on the other hand, the error signal is effectively averaged over several sweeps and is thus less sensitive to individual missing resonance. In the case of a real disturbance, due to cavity vibration and/or laser frequency jitter, both servoloops will loose the lock and then the fast relocking of the digital servoloop becomes significant advantage. We, however, feel that here still is a room for improvement of the current digital servoloop scheme. The sensitivity to missing resonance can be reduced by including software control of the resonance threshold levels. This could especially prevent loosing the lock on strong absorption features, where the resonances are lower due to increased cavity losses.

Clearly, an appropriate combination of PZT sweep rate and dither amplitude (determined by the oversweep period Δt) must be determined experimentally considering the above-mentioned factors. In general, we have found that it is advantageous to keep the dither amplitude low and to tolerate occasional lock loss due to mechanical disturbances. Under the real-life conditions of the flash kinetics experiment described here we achieved ringdown repetition rates up to 0.8 kHz, compared to $\sim$0.2 kHz, in the full-sweep mode, factor of 4$\times$ improvement as demonstrated in presented data. However, preliminary tests with highly mechanically rigid resonator construction indicate that even higher enhancement factors (up to 25$\times$) can be achieved with the same tracking electronics when resonator mechanical vibrations are minimized. Under such conditions the dither amplitude can be further reduced without compromising the servoloop stability.

One of the major advantages of the microcontroller based tracking is the high level of flexibility offered by the programmable control unit. This can be demonstrated on the example of resetting the PZT voltage when a limit is reached. If the laser frequency is tuned, the servoloop has to either extend or shrink the cavity length to track the laser and thus eventually the end of travel range will be reached and the PZT voltage has to be reset to next cavity mode. If, for example, the top of the range is reached the voltage should be reset to minimum and from that point the PZT should sweep up until the first resonance is reached. On the other hand, if the laser scans in opposite direction the voltage should be reset to the maximum value and swept down from that point. Alternatively, if the laser frequency is fixed and the tracking compensates for random drift in the cavity length it may be most practical to reset the PZT voltage to the middle of the range regardless if the bottom or top of the range has been reached. All those options can be easily implemented via the MCU firmware. Indeed, all those scenarios can be pre-programmed simultaneously and appropriate option is then chosen from a system of "menus" accessible via the user interface at the run-time.

Another demonstration of the system flexibility is the possibility to modify the tracking strategy to adequately respond to specific disturbances that affect the tracking. If an external disturbance suddenly shifts the resonance outside the dither range, the tracking is lost and servoloop must switch to a search mode until the next resonance is located. In the case of systematic disturbances, such as sudden gas heating due to laser pulse the direction of the resonance shift can be often predicted. The tracking strategy is easily modified by the MCU software to always search in the direction of the shift and thus to minimize the re-locking time.

The implementation of the microcontroller based tracking electronics has been primarily guided by simplicity of the circuit and flexibility for testing the new tracking concept.

Many improvements are still possible with only moderate additional increase of the circuit complexity. The resonance threshold control option has already been discussed above. Another useful modification would be to include a digital-to-analog convertor for sweep rate control by the microcontroller in addition to the sweep direction control used in current design. This would allow advanced PZT voltage control, such as increasing the sweep rate during the search mode to further reduce the search time and lowering the sweep rate in tracking regime to maximize the resonator power coupling.

V. CONCLUSION

We have presented new tracking servoloop electronics for cw-CRDS. The tracking unit significantly increases the repetition rate of the CRDS events and thus increases effective time resolution in kinetics studies with cw-CRDS, improves effective measurement sensitivity and/or decreases data acquisition time. The tracking servoloop uses novel strategy to track the cavity resonances that results in fast relocking after a disturbance (on the order of few milliseconds, as opposed to a ∼1 s re-locking times with previous designs). The microcontroller based design is highly flexible and thus advanced tracking strategies are easy to implement by the firmware modification without the need to modify the hardware. At the same time the servoloop electronics is rather simple with very low part count, and based on our experience can be easily constructed even in fairly modestly equipped electronics workshop. We believe that performance of many existing cw-CRDS experiments can be improved with such tracking unit without any additional modification to the experiment.

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