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Optical interruption of a quantum cascade laser for cavity ring-down spectroscopy

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We demonstrate optical unlocking of a cavity resonance in a mid-infrared (MIR) quantum cascade laser (QCL) cavity ring-down spectrometer using a low-power multimode near-infrared (NIR) laser diode. A NIR laser with a center wavelength of 1310 nm is injected into a QCL whose amplitude and frequency are modulated as a result. The shift in frequency leads to a rapid interruption of the cavity resonance. The optical method is compared to cavity interruption with a current step by measuring ring-down times in a high-finesse optical cavity, coupled with a QCL with a center wavelength of 4.5 µm. The results indicate the comparable performance of the all-optical method with the more conventional current modulation, but with significantly reduced bandwidth requirements for the QCL driver, opening the way to other potential applications in MIR laser spectroscopy.

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Cavity ring-down spectroscopy (CRDS) is one of most sensitive laser spectroscopy methods, capable of detecting a variety of molecules at trace concentrations levels [1]. Applications of CRDS include atmospheric sensing, industrial monitoring, air quality monitoring, and detection of harmful compounds. CRDS is based on light absorption in a high-finesse optical resonator. Light injected into the resonator experiences a long, effective path length, usually of several kilometres. In contrast to direct absorption methods, in CRDS the exponential decay of light resonating inside the cavity is measured after the light injection is stopped. The decay time constant is referred to as cavity ring-down time, and it depends on the reflectivity of the mirrors and any light-absorbing substance in the cavity. By comparing the ring-down time of an empty cavity with that of a cavity containing absorbing molecules, the absorption coefficient can be inferred. The ring-down time is determined by fitting an exponential function to the signal after the injection is stopped and, thus, has the advantage of being immune to laser intensity fluctuations.

The mid-infrared (MIR) wavelength region, also referred to as the "fingerprint" region, contains the strongest fundamental rotational and vibrational absorption transitions of many important molecules. The availability and price of components at these wavelengths have been a limiting factor for both research and instrument manufacturers. MIR CRDS has seen an increase due to the recent availability of room temperature single-mode quantum cascade lasers (QCLs) and high-reflectivity MIR mirrors required for cavity enhanced spectroscopy techniques [2]. The availability of such components combined with the high sensitivity of CRDS has a great potential for compact field-capable sensitive instrumentation and in-situ detection of important molecules, approaching sensitivities of mass-spectrometer analysis.

An important step in the CRDS system is the rapid interruption of the coupled light to initiate a purely exponential ring-down event. Poor interruption of the signal results in oscillations in the decay signal that reduce the overall sensitivity or prevent the ring-down measurement altogether. With a clean decay, the signal can be fitted with a single exponential function to extract the ring-down time. Fast interruption can be achieved by fast current modulation of the laser [3], an acousto-optic modulator (AOM), or by applying a sharp step to the laser driving current to quickly shift the emission frequency of the QCL to a region absent of cavity resonances. The fast current modulation has the disadvantage of reducing the amount of light built up in the cavity and, thus, measurement may be limited by poor signal-to-noise ratio at the detection. AOMs are not always favourable due to high cost, especially in the MIR region, and added complexity. The sharp current step requires a fast QCL driver, since the ring-down times are typically between a few microseconds to hundreds of microseconds.

Recently, there has been studies on amplitude modulation (AM) and frequency modulation (FM) of MIR QCLs with near-infrared (NIR) laser diode (LD) excitation [4–8]. Both positive and negative AMs have been demonstrated, depending on the QCL structure and NIR wavelength. If the energy of the NIR photon is close to the band gap energy of the QCL active region, most of the injected NIR photons increase the electron density in the lasing sub-bands, resulting in an increase of the output power of the QCL. In contrast, with higher-energy NIR photons, electrons are mostly excited to higher-energy bands, after which they relax through electron-electron scattering without contributing to lasing, resulting in a decrease of the QCL output power (see, e.g., Refs. [9–11]). The wavelength of the QCL is blueshifted in both cases. This has been utilized in producing purified FM by using two NIR lasers with different wavelengths [10].

These studies suggest interesting application possibilities within the scope of laser absorption techniques such as FM spectroscopy [12]. One potential application is the use of NIR-induced FM in a MIR QCL to interrupt the light injection in CRDS to trigger a clean ring-down event, which is experimentally demonstrated in this Letter. The use of an all-optical interruption has the advantage of fewer requirements for the QCL driver bandwidth. As an example, a recently introduced passive QCL driver [13] could then be used in CRDS setups. Another potential advantage is the fast and more precise return to original frequency, compared to electrical modulation, allowing acquisition of ring-down events at an increased rate if the cavity is locked on a resonance [14]. Optical interruption of light injection has been demonstrated in a NIR LD based CRDS system [15], where the main laser frequency is shifted with a second LD with comparable NIR wavelength. The NIR emission from the secondary LD induces AM, as well as FM of the main LD, resulting in sharp interruption of light build-up in the cavity. Nonetheless, it is particularly relevant to demonstrate that a similar approach can be achieved with QCLs, as they are increasingly used in various laser spectroscopy applications.

We present here the experimental results for optical interruption in MIR CRDS, based on a room temperature QCL emitting at 4.5 μm. We use a low-cost multimode NIR laser with a center wavelength of 1310 nm and an average output power of 5 mW to modulate the QCL upon cavity build-up. The performance of the method is compared to the interruption using a fast current interruption.

The experimental setup is shown in Fig. 1. It is similar to the one used in an earlier work [16], except for the cavity mirrors, which were replaced with a set of silicon mirrors with higher reflectivity from LohnStar Optics Inc. A MIR QCL (Hamamatsu L12004-2209H-C) is mode matched to an optical resonator with a linewidth of 3.5 kHz and a mirror separation of 0.38 m corresponding to a free spectral range (FSR) of 395 MHz. This results in empty cavity ring-down times on the order of 46 μs. An optical isolator (EOT MESOS) is used to prevent feedback from the cavity. The light after the cavity is detected with a mercury-cadmium-telluride (MCT) detector (Vigo PVI-3TE-5). The signal is routed through an analog pulse detection circuit, which provides a timing signal for the data acquisition, as well as for the interruption. The ring-down signal is digitized at 50 MHz and sent to a PC for signal analysis. The timing signal is sent to either the QCL driver (Wavelength Electronics QCL1000) via a summing circuit

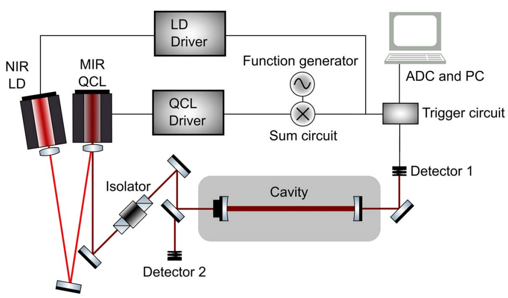

Fig. 1. Schematics of the experimental setup; more details are given in the text.

for electrical interruption or to the LD driver (Wavelength Electronics LD200) for optical interruption. A signal generator is used to scan the QCL frequency around the cavity resonances. The NIR laser (Thorlabs ML725B8F) is focused onto the QCL from the side with a ∼10 deg angle. The QCL output power is monitored with a separate MCT detector, measuring the light leaked from the final mirror before the cavity. The recorded traces are fitted in real time with an exponential function implemented in LabView.

For alignment of the NIR laser, a 1 kHz square wave modulation is applied to the NIR laser, and the emission of the QCL is observed on detector 2. For the results presented in this Letter, the AM depth is around 0.7% of the total QCL output power. It is noted that no significant attempts were made to optimize the modulation depth, since only a small shift is sufficient for the purpose. The observed AM corresponded to an approximate frequency shift of 68 MHz, as later discussed. The small frequency shift is enough for interruption of the light in a high-finesse cavity, having a linewidth of a few kilohertz. Due to the high reflectivity of the QCL collimating lens (Thorlabs C028TME-E) with NIR wavelengths, only 10% of the total NIR power reaches the QCL chip, after transmission through the collimating lens and the QCL window. Moreover, due to multiple spatial modes, part of this light is lost as well on focusing. In our case, the 1310 nm NIR modulation results in positive AM and in a blueshifted wavelength, similar to what has been observed in earlier work.

After the alignment, the square wave modulation is changed to constant emission of the NIR laser. A 10 Hz rising saw-tooth ramp is applied to the current of the QCL to scan over the cavity resonances. It is important for the slow ramp to produce a wavelength shift in the same direction as the interruption in order to prevent potential recoupling to the same resonance. The NIR laser is rapidly stopped (<1 μs) to redshift the QCL frequency, when detector 1 is above a certain threshold corresponding to sufficient cavity build-up. The ring-down rate was relatively low (10–15/s). With a 120 μs long trigger signal, this corresponds to a NIR duty cycle (DC) around 98.8%. When the NIR laser was tested with positive modulation with 0.02% DC, a significantly slower response time (of several hundreds of microseconds) of the NIR laser was observed due to a slow thermal response. Thus, this mode of operation is not suitable to interrupt the cavity resonance. The NIR laser noise, in theory, could affect the linewidth of the MIR QCL; however, we did not observe a significant difference in the amount of light coupled to the optical cavity with and without NIR injection.

An oscilloscope trace of both detectors during the ring-down measurement is shown in Fig. 2(a) with and without optical interruption. The AC signal from detector 2 shows a decrease in signal due to the release of the NIR injection.

To estimate the QCL frequency shift resulting from the NIR injection, ring-down times were plotted in a scatterplot against the QCL modulation voltage shown in Fig. 2(b). The laser current was scanned over five cavity FSRs, while the cavity length was kept constant. The cavity resonances are clearly visible, and the FSR of the cavity can be used as a frequency scale. From the FSR value of 395 MHz, the frequency shift is estimated to be 68 MHz. The frequency shift can be easily adjusted by either aligning the NIR laser or by adjusting the NIR emission power. While simultaneously observing the standard deviation of the fitted ring-down decays, one can find the

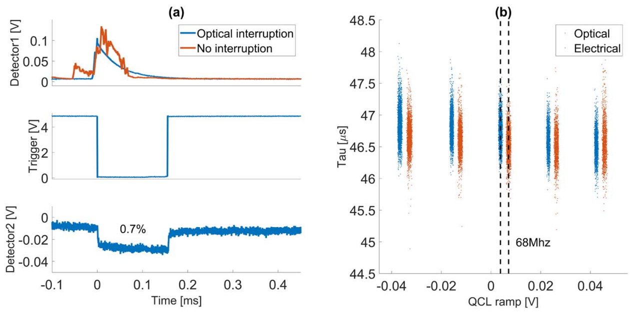

Fig. 2. (a) Typical cavity ring-down traces from detector 1 with and without active interruption. Timing signal that triggers the interruption and laser power measured with the detector 2 (AC), showing the effect of NIR modulation. (b) Scatterplot of the ring-down measurements against the QCL modulation voltage.

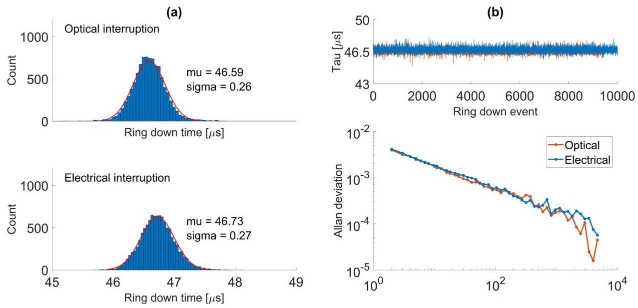

Fig. 4. (a) Histogram plot of the fitted ring-down times with optical and electrical interruption. (b) Relative Allan deviations of the time series of ring-down events measured with optical and electrical interruptions.

minimum frequency shift, which will still produce clean ring-down events. This is likely very dependent on the overall experimental setup. In our case, we have reduced the frequency shift down to 17 MHz without decrease in performance, which is expected since it is three orders of magnitude more than the cavity linewidth of 1.7 kHz.

An example of the fitted ring-down traces measured with the digitizer and fitted in real time with LabView is shown in Fig. 3. The digitized signal is cut at 85% of its maximum amplitude, as the beginning of the ring-down event is usually not clean. Both electrical and optical methods show clear single exponential ring-down signals. In the case of no active interruption, oscillations are observed, even in the cleanest events. Most of the ring-down events recorded without interruption exhibit even higher amounts of residual, as shown in Fig. 2(a). The amplitude of the electrical shift was empirically optimized to a current value of 0.6 mA, corresponding to a frequency shift of 220 MHz.

To assess the performance of the two methods, ring-down events of an empty cavity were measured for 15 min with each method. A total of 10640 ring-down signals were collected and fitted in real time with the optical modulation and 11246 with the electrical method. The laser current was scanned with a positive sawtooth ramp at a 10 Hz rate over five cavity FSRs, and the cavity length was kept constant after acquisition; some ring-down measurements were discarded based on the mean residual value of the exponential fit ($>6\,10^{-7}$). A total of 1204 (11%) and 681 (6%) ring-down measurements were discarded for the optical and electrical methods, respectively. The discarded events consist of events where laser light was coupled into the cavity during the ring-down event due to random overlap of the detuned laser frequency with higher-order transverse cavity modes. Note that the proportion of rejected events greatly depends on the alignments of the system. The histograms of the measurements with a normal distribution fit are shown in Fig. 4(a), and the relative Allan-deviation plot of the same dataset is shown in Fig. 4(b). The filtered ring-down events with both methods fit the normal distribution well and show ideal averaging based on the Allan-deviation plot.

Finally, we used the optical interruption method to measure a CO_2 spectrum centered on 2209.08 cm^{-1} and shown in Fig. 5. The spectrometer used in this Letter is designed to probe elevated levels of radiocarbon in carbon dioxide [16]; however, in this demonstration, standard CO_2 was used. The pressure inside the cavity was 20 mbar. The temperature of the cavity was not actively regulated; however, the laboratory space had a controlled temperature of 23°C. The spectra were acquired using both electrical and optical methods for signal interruption. The acquisition time was 300 s, which results in 3386 and 3980 ring-down events for optical and electrical, respectively. The QCL modulation ramp was extended to cover 10 FSRs. In addition, the cavity length is slowly scanned with piezo-actuators connected to one of the cavity mirrors to in-

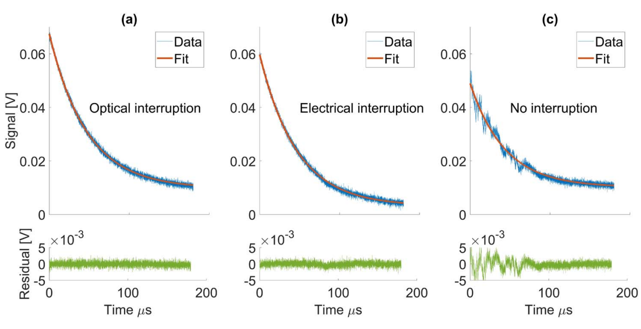

Fig. 3. Typical ring-down traces and fit residuals with (a) optical interruption, (b) electrical interruption, and (c) no interruption.

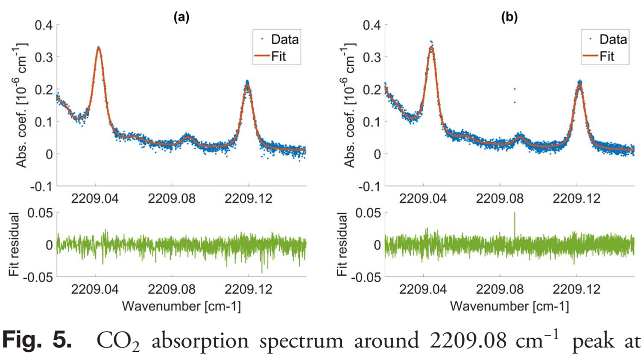

Fig. 5. CO_2 absorption spectrum around 2209.08 cm^{-1} peak at 20 mbar pressure and approximately 23°C. The CO_2 amount fraction was close to 100%. Spectra measured with (a) optical interruption and (b) electrical interruption.

crease the number of points along the wavelength axis, which is calibrated using a 25.4 mm long germanium etalon. The data are fitted by a sum of Voigt profiles using a nonlinear least-square-fitting routine. Based on the fit residuals, no differences between the two methods can be observed, which demonstrates the applicability of the all-optical CRDS method for detection of trace gases using a MIR QCL.

In summary, optical interruption of a MIR QCL using a low-cost low-power NIR diode laser was experimentally demonstrated by various tests against the more conventional method of modulating the current driving the QCL. We have shown that the optical approach can provide fast interruption of laser coupling into the cavity to induce clean ring-down events in a repeatable manner. Both optical and electrical interruption methods produce very comparable results. Further work on this topic and optimization of different parameters such as optical power, frequency, and alignment of the NIR LD can potentially lead to even better performances. The main advantage of the optical method is the significant reduction of the bandwidth needed for the QCL driver, which can lead to reduction on both cost and complexity of the electronics. We also hypothesize, that acquiring ring-down events rapidly by locking the laser to the cavity modes could be achieved with the optical interruption due to faster recovery after each ring-down event. Demonstrating this is, however, outside of the scope of this Letter.

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