Optical Interruption of Quantum Cascade Laser for Cavity Ring-Down Spectroscopy

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ABSTRACT

Researchers from VTT Technical Research Centre of Finland have developed optical interruption of a mid-infrared (MIR) quantum cascade laser (QCL) using a near-infrared (NIR) laser diode for cavity ring-down spectroscopy (CRDS). The QCL is modulated from the injection of the NIR laser diode, and this shifts the frequency of the signal in the cavity generating rapid interruption of the high-finesse cavity resonance. This method is compared to the traditional electrical interruption method and shows similar performance. Optical interruption reduces the high bandwidth requirements of the QCL driver as well as lowers cost and complexity of the system. The necessary precision control was provided by Wavelength Electronics' laser and QCL drivers. With this optical interruption method, other potential applications in MIR laser spectroscopy are now possible.

CRDS BACKGROUND

Cavity Ring-Down Spectroscopy (CRDS) is one of the most sensitive laser spectroscopy methods. It is effective at detecting a variety of molecules at trace concentrations. CRDS is based on the amount of time it takes the laser beam to decay in a high finesse resonator made up of high reflectivity mirrors. This multi-pass cavity allows the effective distance traveled by the light to increase to lengths as long as a few kilometers. Longer distance traveled through a sample gas ensures better measurement precision. When the light into the cavity is interrupted, a ring-down event is started (**Figure 1**), and the decay time can be accurately measured. A detector is used after the cavity to ensure the correct threshold of light build-up in the cavity has occurred, and it can measure the light leakage out of the cavity (mirrors are not 100% reflective) to track the time decay once the light has been interrupted.[1]

CRDS allows spectroscopy measurements independent of laser power stability. Because the laser light into the cavity is interrupted when enough light has built-up in the cavity, the concern is no longer the laser itself but the decay time of the light in the resonator. The decay time of the light varies with the specific molecules trapped in the cavity. The absorption coefficient of certain molecules can be found by comparing the cavity ring-down (CRD) time when those molecules are in the cavity with the CRD time of an empty cavity. Different molecules will have unique absorption coefficients and spectra. The CRD time will be accelerated with absorbing gas inside the cavity and will only be affected by the reflectivity of the mirrors and the absorbing substance inside the cavity.[1] For trace concentration measurements, accurate calibration with an empty cavity is essential. CRDS applications include atmospheric sensing, industrial monitoring, air quality monitoring, detection of harmful compounds, and medical applications among others.

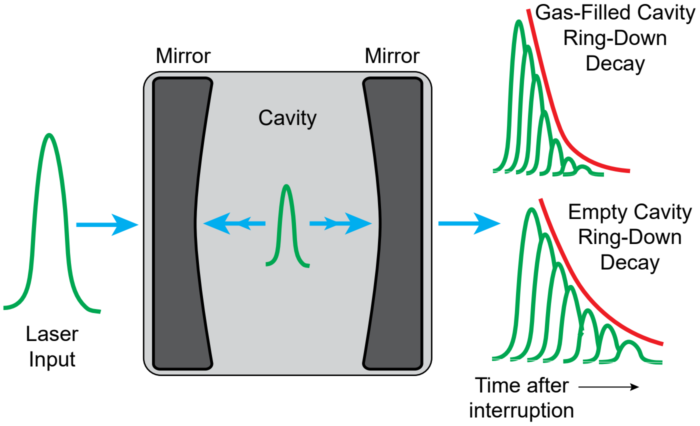

Figure 1. Cavity Ring-Down Spectroscopy Schematic

It is common for researchers to use infrared (IR) lasers for spectroscopy applications. Mid-infrared (MIR), ranging from 3 - 50 µm, consists of the best absorption transitions, specifically fundamental rotational and vibrational transitions, of many important molecules. Compared to near-infrared (NIR) and far-infrared (FIR), MIR has a cleaner spectrum with more distinct peaks and troughs for the specified absorbed sample. MIR enables trace detection in small volumes of homogeneous samples, unlike NIR and FIR. Some gases have weak transitions in NIR but are much stronger in the MIR region. CRDS utilizes the strong absorption coefficients in the MIR range for more sensitive measurements.

PROBLEMS AND GOALS

CRDS can be very effective for low concentration measurements. However, until recently MIR optics, lasers, and quick drivers were either hard to obtain or expensive to implement into the CRDS system. CRDS has slowly overcome these limitations with the development of a room-temperature Quantum Cascade Laser (QCL), prices for components at MIR wavelengths have dropped and they have become more available as have high reflectivity MIR mirrors for creating the high-finesse resonator cavity.

One of the most important aspects of CRDS is the laser interruption. It is important to accomplish this quickly and efficiently to ensure the CRD event will be rapid and purely exponential. If the interruption is of poor quality it can result in oscillations in the CRD decay signal. This could decrease the sensitivity of the system or prevent the CRD event measurement altogether.[1]

There are three common ways to enable rapid interruption. All three methods have their own disadvantages.

1. *Fast current modulation of the laser* - Modulating the current of the laser can potentially reduce the amount of light build up in the cavity. In this case, the signal-to-noise ratio is poor, decreasing the sensitivity of the spectrometer.
2. *Acousto-Optic Modulator (AOM)* - AOMs are not ideal due to high cost in the MIR range. Problems also can be created due to difficulties in alignment of the laser and overall added complexity to the system design.
3. *Sharp Current Step* - This method can be very effective in interrupting the laser to the cavity by shifting the frequency to a region absent of cavity resonances. However, this requires a fast QCL driver. The CRD events are on the order of a few microseconds to a couple of hundred of microseconds. The current step needs to be fast or the poor interruption will generate oscillations in the CRD event, decreasing the precision and effectiveness of the measurement. Finding a fast QCL driver can be expensive.[1]

Researchers at the VTT Technical Research Centre of Finland have developed a new optical interruption design using a laser diode to modulate a QCL upon cavity build up.

METHOD

The NIR emission allows for amplitude modulation (AM) and frequency modulation (FM) of the laser diode to create a fast interruption of the QCL signal into the resonator. This method, compared to the current method of electrical interruption, benefits from lower cost, lower required QCL driver bandwidth, and the potential to return to the original frequency quickly and with greater precision. A faster return to the original frequency allows acquisition of ring-down events at an increased rate if the cavity is locked on a resonator. **Figure 2** shows the schematic of the optical interruption of a laser diode pumped QCL.

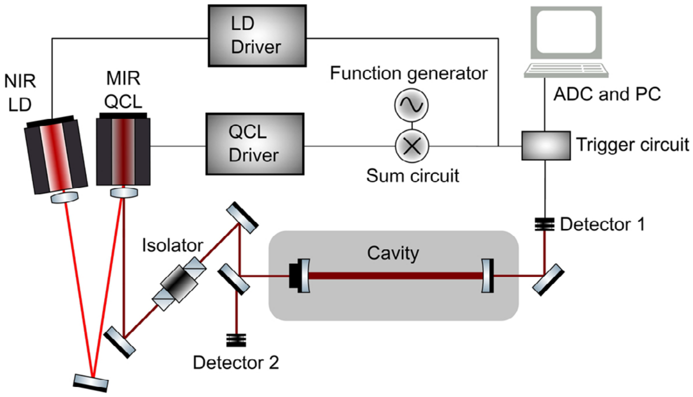

Figure 2. Schematics of the experimental setup; more details are given in the text. Reprinted with permission from Reference 1 © The Optical Society.

The laser diode is in the NIR wavelength range (1310 nm) and induces modulation in the QCL in the MIR wavelength range (4.5 μm). The injection of NIR laser diode photons increases the electron density in the lasing sub-bands of the QCL if the energy of the photons is close to the bandgap energy of the QCL active region. This will increase the output power of the QCL, although if the photon energies are higher, it will not increase the power or contribute to lasing. It will actually decrease the output power. In either case, the QCL wavelength is shifted.

The noise of the laser diode and the driver could potentially affect the linewidth of the QCL. Low noise drivers are critical for narrow linewidth and wavelength stability. Stable current to the laser diode and QCL is necessary to efficiently conduct the experiment and measurement.

The QCL is mode matched to an optical resonator cavity with a linewidth of 3.5 kHz, and the mirror separation inside the cavity is 0.38 m and corresponds to a free spectral range (FSR) of 395 MHz. This will give typical CRD event times in an empty cavity on the order of 46 μs.

Other elements in the design include the isolator, two detectors, function generator, and a trigger circuit. The isolator prevents feedback from the cavity. Detector 1 (after the cavity) collects the light leaking out of the cavity. When enough build up is detected in the cavity, the trigger circuit will stop the NIR laser and shift the frequency of the QCL, starting the ring-down event. The timing signal, from the trigger circuit, is sent to the QCL driver for electrical interruption or to the laser diode driver for optical interruption. Detector 2 (before the cavity) monitors the QCL output after the final mirror before the cavity and is recorded and fitted to an exponential curve in real time. Both detector signals can be seen in **Figure 3**. The function generator is used to modulate the NIR laser for the alignment portion of the design, but for measurements the laser diode is changed to constant emission. The QCL is modulated with a 10 kHz rising saw-tooth ramp to scan over the cavity resonances. This slow ramp, from the QCL driver, produces a wavelength shift in the same direction as the interruption to avoid potential recoupling to the same resonance.

When the laser diode is stopped by the trigger circuit, the interruption begins and starts the CRD event (**Figure 3**). There is a dramatic change in signal from detector 2 due to the change in NIR laser diode output. **Figure 3** also shows the desired exponential decay with the optical interruption compared to the residual effects without interruption. This effect can further be seen in **Figure 4**.

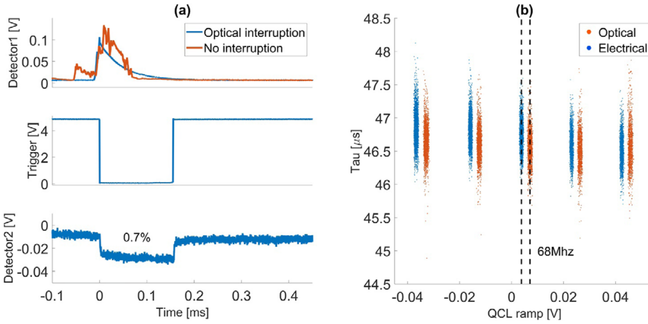

Figure 3. (a) Typical cavity ring-down traces from detector 1 with and without active interruption. Timing signal that triggers the interruption and laser power measured with detector 2 (AC), showing the effect of NIR modulation. (b) Scatterplot of the ring-down measurements against the QCL modulation voltage. Reprinted with permission from Reference 1 © The Optical Society.

RESULTS

When compared to electrical interruption, optical interruption of a QCL shows little difference. **Figure 4** shows the CRD events with exponential decay fit curves as well as residual voltages.

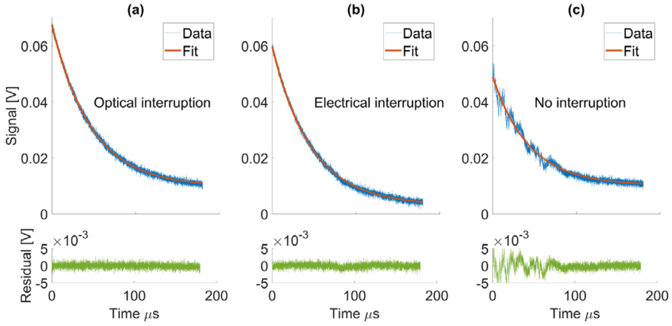

Figure 4. Typical ring-down traces and fit residuals with (a) optical interruption, (b) electrical interruption, and (c) no interruption. Reprinted with permission from Reference 1 © The Optical Society.

It is clear that without interruption the CRD event is overcome by oscillations in the signal and large residuals, even in the cleanest events. To compare the electrical and optical interruptions, 10640 and 11246 CRD events for optical and electrical, respectively, were recorded. These measurements were recorded in 15 minutes and collected into a histogram plot with a normal distribution fit. **Figure 5** shows the histogram plots of both methods, and both fit the normal distribution well. They also show ideal averaging based on the Allan-deviation plot.

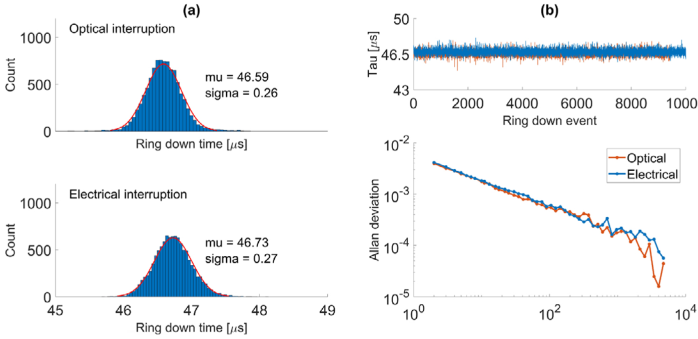

Figure 5. (a) Histogram plot of the fitted ring-down times with optical and electrical interruption. (b) Relative Allan deviations of the time series of ring-down events measured with optical and electrical interruptions. Reprinted with permission from Reference 1 © The Optical Society.

To further show the effectiveness of optical interruption, a CO_2 spectrum was measured centered on 2209.08 cm^{-1}. Acquisition time was 300 seconds, resulting in 3386 and 3980 CRD events for optical and electrical, respectively. **Figure 6** shows the measurement results of the CO_2 data with both methods.

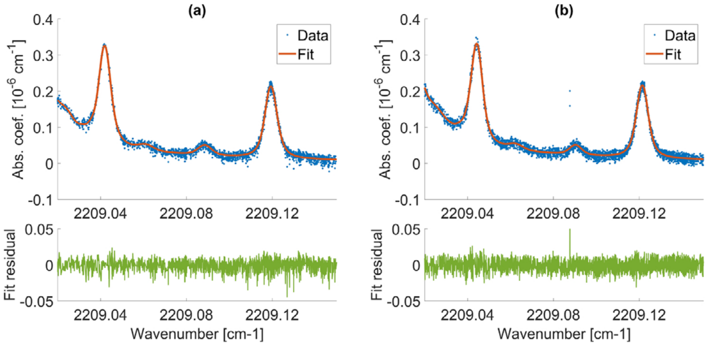

Figure 6. CO_2 absorption spectrum around 2209.08 cm^{-1} peak at 20 mbar pressure and approximately 23°C. The CO_2 amount fraction was close to 100%. Spectra measured with (a) optical interruption and (b) electrical interruption. Reprinted with permission from Reference 1 © The Optical Society.

"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."[1] Optical interruption of the MIR QCL allows for lower cost using a low-power NIR diode laser. It also reduces the bandwidth needed from the QCL driver (further reducing cost) and provides a fast interruption of the laser coupling into the cavity. The researchers 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."[1] Clean ring-down events can be generated and accurately repeated. A less complex design of the electronics can lead to very comparable results and expand the range of applications.

WAVELENGTH'S ROLE

Stability is crucial for proper operation and repeatable results for multiple lasers in the optical interruption for CRDS. Low noise from the laser diode ensured that the linewidth of the QCL was not affected.

Wavelength Electronics' LDD200 (**Figure 7**) provides a current limit of 200 mA with multiple features and benefits. In this application it drove the current of the laser diode with noise as low as 5 µA (RMS) and adjustable current limit. This low-cost and compact laser driver provided the laser stability necessary for injection current into the QCL.

Figure 7. LDD200 Laser Diode Driver

The LDD200 laser driver operates from a single power supply between 5 and 12 V. The limit current trimpot can be adjusted to protect the laser diode from exceeding its maximum current rating even when modulating the laser diode. An evaluation board is available to speed implementation of the LDD200 in the appropriate application.

Wavelength's QCL1000 OEM driver (**Figure 8**) provides a current limit of 1 A with low noise (0.7 µA) needed for spectroscopy applications. It has modulation bandwidth capabilities of up to 3 MHz, average current noise density of 2 nA/√Hz, and current limit controls on-board. The QCL driver minimizes linewidth, drift, and jitter of the laser system.

Figure 8. QCL1000 OEM QCL Driver

The patented QCL driver enables rapid integration and dual supply operation. Standard compliance voltage to the laser is 16 V, and other features (setpoint, limit, delay, and ramp) protect the laser.

REFERENCES

1. Teemu Kääriäinen and Guillaume Genoud, "Optical interruption of a quantum cascade laser for cavity ring-down spectroscopy," *Opt. Lett.* 44, 5294-5297 (2019). https://doi.org/10.1364/OL.44.005294

USEFUL LINKS

- LDD200 Product Page
- QCL1000 OEM Product Page

PERMISSIONS

Figures 2, 3, 4, 5, & 6 and data used for this case study were obtained from Reference 1. Permission was granted for use of the images and data from Optical Society of America and the corresponding author of Reference 1.

No changes were made to the images. They are presented here in their original form.

PRODUCTS USED
QCL1000 OEM, LDD200

KEYWORDS
Optical interruption, quantum cascade laser, cavity ring-down spectroscopy, CRDS, laser diode, mid-infrared, near-infrared, laser injection, bandwidth, frequency shift, laser diode driver, QCL driver

REVISION HISTORY

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REVISION	DATE	NOTES
A	January 2021	Initial Release