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Abstract. We review recent advances in chemical sensing applications based on surface emitting ring quantum cascade lasers (QCLs). Such lasers can be implemented in monolithically integrated on-chip laser/detector devices forming compact gas sensors, which are based on direct absorption spectroscopy according to the Beer–Lambert law. Furthermore, we present experimental results on radio frequency modulation up to 150 MHz of surface emitting ring QCLs. This technique provides detailed insight into the modulation characteristics of such lasers. The gained knowledge facilitates the utilization of ring QCLs in combination with spectroscopic techniques, such as heterodyne phase-sensitive dispersion spectroscopy for gas detection and analysis.
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Keywords: quantum cascade laser; quantum cascade detector; gas sensing; radio frequency modulation; heterodyne phase-sensitive dispersion spectroscopy.

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1 Introduction

The midinfrared spectral region exhibits numerous ro-vibrational absorption lines of various substances. Therefore, light sources emitting in this wavelength range are desirable tools for sensing and chemical fingerprinting. Due to their compact size, tailorable emission characteristics,[1,2] and high spectral brightness,[3] quantum cascade lasers (QCLs)[4] are commonly used for such absorption experiments.[5] Surface emitting QCLs possess considerable advantages compared with their edge emitting counterparts. They facilitate on-chip testing and two-dimensional array integration. Due to their rather large emission aperture, these lasers can produce narrow beam profiles.[6] The first surface emitting QCL[7] was demonstrated utilizing a second-order distributed-feedback (DFB) grating. Subsequently, further approaches have been successfully implemented: in photonic crystals[8,9] artificial periodic structures provide optical feedback and generate light beams in vertical direction. With random lasers[10,11] almost diffraction limited broadband surface emission can be achieved, while losing tunability. Manipulating beam characteristics through wave engineering has been demonstrated with vertically emitting circular DFB gratings[12] as well as graded photonic heterostructures.[13]

Surface emitting ring QCLs[14] typically consist of a circular ring waveguide with a second-order DFB grating. The latter provides single-mode selection as well as vertical light emission while the circular ring geometry creates a collimated and rotationally symmetric far field. This desirable beam profile can be modified by well-directed manipulation of a regular DFB grating.[15] Single-mode emission and continuous-wave operation at room temperature[16,17] in combination with the collimated emission beam make ring QCLs suitable tools for chemical sensing applications.[18,19] Due to their compact size and small footprint, these lasers are ideal light sources for the application in miniaturized sensing setups, such as substrate-integrated hollow waveguides.[20]

2 Surface Emitting/Detecting On-Chip Sensor

Typical state-of-the-art applications make use of conventional sensing setups, which include a light source, a light–analyte interaction region, and an external separate detector, e.g., based on the mercury-cadmium-telluride (MCT) material system. Utilization of a quantum cascade detector (QCD)[21,22] in combination with a separate QCL mounted on an individual chip demonstrates the capabilities of quantum cascade technology in spectroscopy.[23] The development of bifunctional quantum cascade heterostructures[24] enables the realization of monolithically integrated lasers (QCL) and detectors (QCD) fabricated on the same chip. Edge emitting and detecting bifunctional devices have successfully been used for sensing of liquids.[25] However, the limited length of the interaction region between laser and detector makes gas sensing impractical. One possible solution is to guide the light on the chip and thereby increase the interaction length.[26] Certainly, this approach is also limited in terms of absorption path length due to restrictions in on-chip mode guiding.

In contrast, utilization of surface emitting and detecting quantum cascade structures fabricated on the same chip in combination with an external light–analyte interaction region provides, in theory, arbitrarily long absorption paths and facilitates remote sensing experiments.[27,28] Furthermore,

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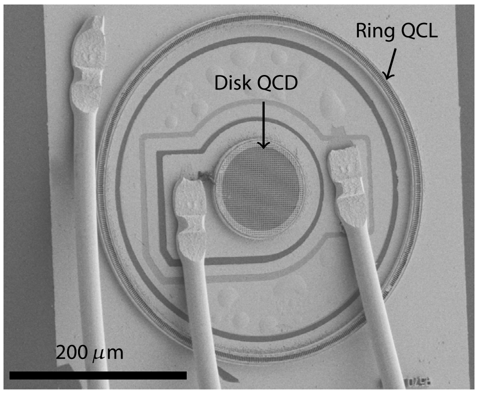

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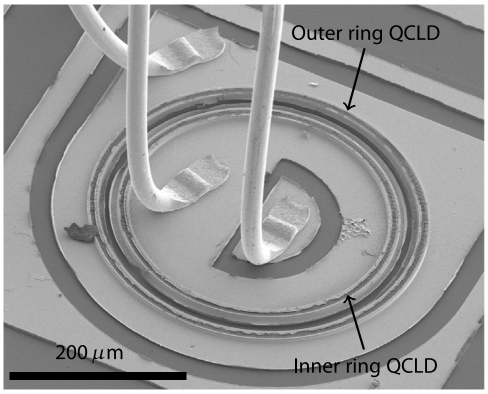
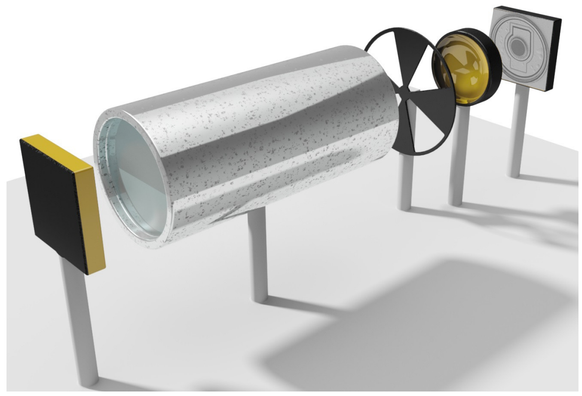


(b)

Fig. 1 SEM images of surface emitting and detecting quantum cascade sensors. (a) Ring QCL with a centered disk QCD.[27] (b) Two concentric DFB ring quantum cascade laser/detector (QCLD) devices.[28] Each ring can be used for light emission and detection.



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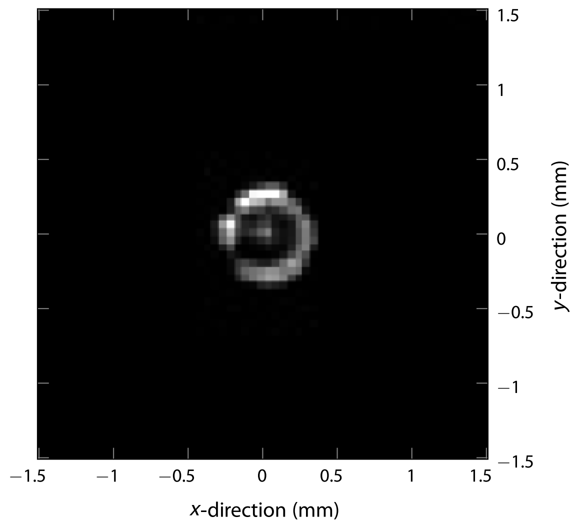


(b)

Fig. 2 (a) Sketch of the experimental setup for remote sensing based on a monolithically integrated surface emitting and detecting on-chip sensor. Light is emitted from the sensor chip, traverses the gas cell, and is back-reflected by a flat gold mirror.[28] (b) Beam profile measured with a bolometer camera after two identical lenses. This pattern corresponds to the intensity distribution incident on the sensor chip.

the small footprint of such sensors enables the realization of compact sensing systems. Figure 1(a) shows a scanning electron microscope (SEM) image of a surface emitting ring QCL with a surface detecting disk QCD in its center.[27] Vertical light emission and detection are provided by the second-order DFB grating (QCL) and the metal hole grating (QCD), respectively. A sketch of the corresponding sensing setup is shown in Fig. 2(a).

Light emitted from the ring QCL is collimated by a lens and modulated by a chopper in order to filter out the high-frequency electrical cross talk before it propagates through the gas cell. A flat gold mirror at the end of the cell reflects the light back toward the chip. After inverse propagation along its initial path, the light is focused onto the sensor chip where it is detected by the disk QCD. In this setup, an image of the laser source is projected back onto the sensor chip. Figure 2(b) shows the incident beam pattern, which was recorded in a mirrored setup with two identical lenses preceding a bolometer camera. The extent of the measured intensity pattern indicates that only a fraction of the emitted light is impinging on the QCD. An enhanced coupling between laser and detector can be achieved by adapting the detector geometry to the shape of the laser beam. Such an approach is depicted in Fig. 1(b), which shows two monolithically integrated concentric ring structures. The implemented second-order DFB gratings facilitate vertical light emission and detection, which enables mutual commutation of both rings, i.e., inner and outer ring can be used as laser and as detector, respectively.[28] Both concepts, with the disk and ring detector, are based on conventional absorption spectroscopy according to the Beer–Lambert law.

3 Modulation Characteristics and Dispersion Spectroscopy

In contrast, ring QCLs can also be used in combination with sophisticated spectroscopic techniques. Knowledge of the laser's current-tuning characteristics[29] is important for a variety of sensing techniques, such as wavelength- and frequency-modulation spectroscopy,[30,31] intrapulse absorption spectroscopy,[32] chirped laser dispersion spectroscopy,[33] and heterodyne phase-sensitive dispersion spectroscopy (HPSDS).[34,35] The latter is a molecular dispersion technique that probes a sample's refractive index spectrum, rather than its absorption, via the phase of radio frequency-modulated laser light.[36] Its sensitivity to the signal's phase makes

HPSDS immune to power fluctuations and therefore enables calibration-free sensor operation.

In the following, an HPSDS spectrometer is implemented and experimentally validated. Therefore, a surface emitting single-mode ring QCL [Fig. 3(a)] is sinusoidally modulated at radio frequencies and swept across a well-known absorption line. This technique allows a thorough investigation of the laser's modulation characteristics. A detailed description of the quantum cascade heterostructure can be found in Ref. 37. In this study, characteristic laser tuning parameters are fitted to a known sample's absorption profile using the model described in Ref. 38. Based on this characterization, HPSDS gas sensing experiments are performed.

The ring laser is operated at liquid nitrogen temperatures, and its emitted radiation is measured with an MCT detector after passing through a 10-cm-long gas cell filled with 0.4% CH_4 in N_2 at a pressure of 24 mbar. A sketch of the corresponding setup is shown in Fig. 3(b). Different modulation frequencies between 1 and 150 MHz are utilized in combination with four DC current values (65, 101, 150, and 200 mA). The corresponding laser submount temperatures (118, 110, 100, and 88 K) are reduced in order to stabilize the emission wavelength of the laser. All operating points are well above threshold, which is found to be at 27 mA (80 K) and at 43 mA (110 K). For each DC current, the laser is slowly tuned over the two absorption features at 1322.08/cm and 1322.15/cm. The tuning frequency of the current ramp is around 50 MHz with a peak-to-peak amplitude of 11.8 mA. Subsequently, the signal of the MCT is fed into a mixer and downmixed to 100 kHz in order to measure the amplitude and phase with a lock-in amplifier.[35] Figure 4 shows the amplitude (a) and phase (b) of the downmixed signal at a modulation frequency of 100 MHz and a DC current of 200 mA. For all DC currents and modulation frequencies, only the larger feature at 1322.08/cm is fitted and used for the following analysis.

According to the notation in Ref. 38, Fig. 5(a) shows the ratio $\Delta f/m = \Delta f/(\Delta P/P_0)$ between absolute frequency modulation (FM) and intensity modulation (IM) index. In Fig. 5(b), the FM–IM phase shift Θ is provided as a function of the modulation frequency. The region between 30 and 150 MHz is characterized by a rather flat behavior of $\Delta f/m$ and Θ. However, below 20 MHz, the rise of the

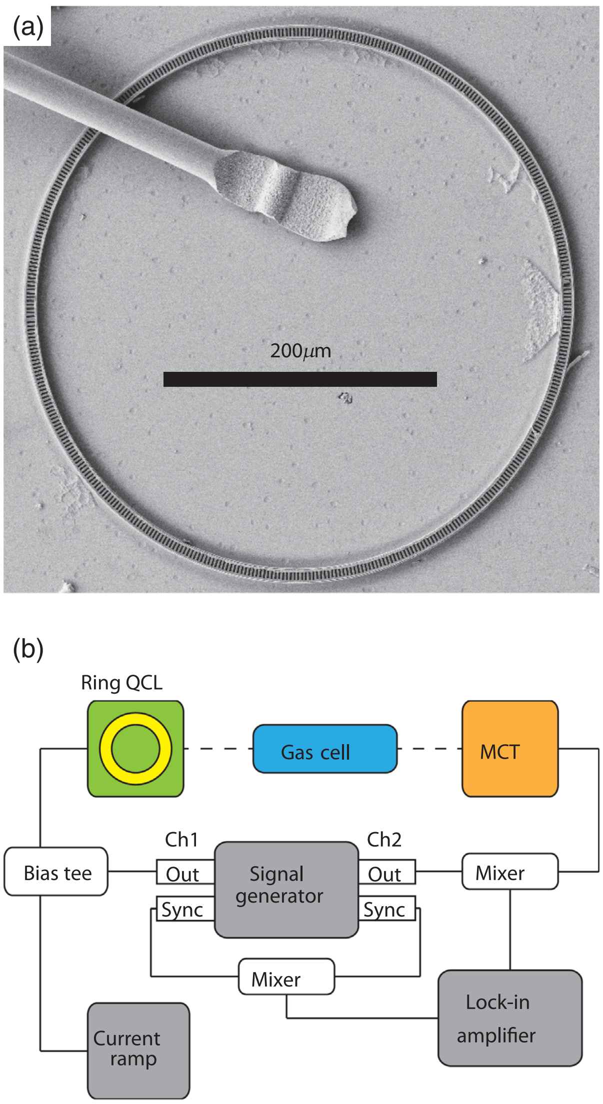


Fig. 3 (a) SEM image of a surface emitting ring QCL with a second-order DFB grating for surface emission. (b) Schematic of the experimental setup used for the characterization of the laser's modulation properties.

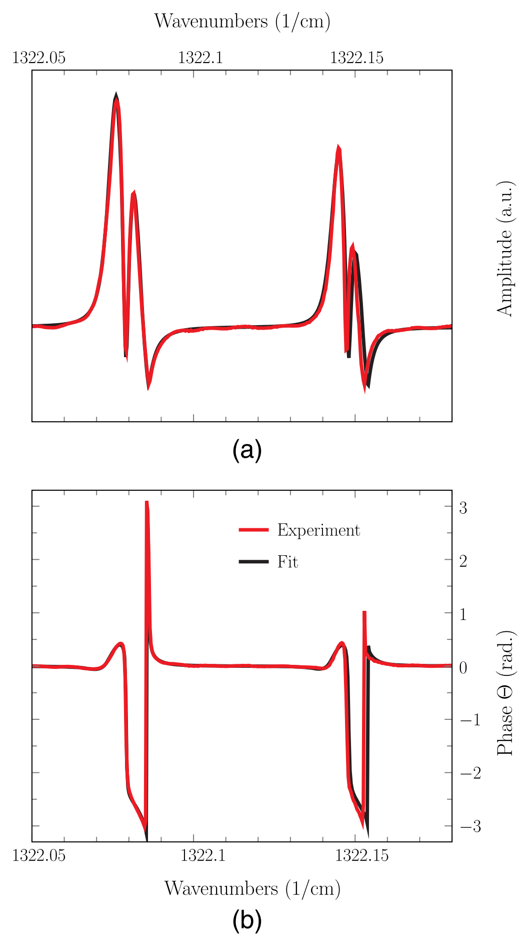
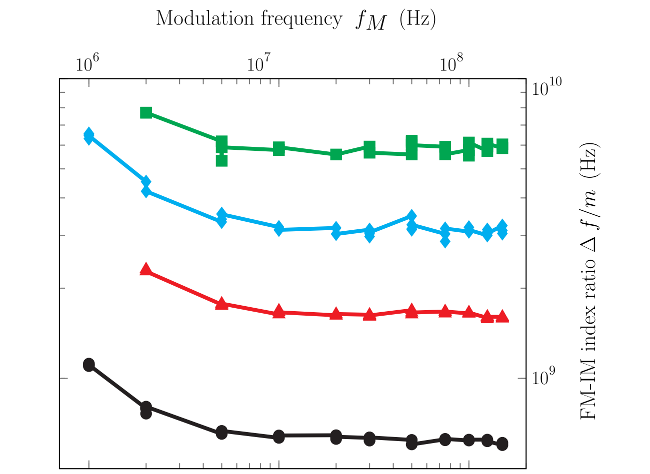


Fig. 4 (a) Amplitude and (b) FM–IM phase of an exemplary measurement with a modulation frequency of 100 MHz and a DC current of 200 mA. Experimental data (red) and fit (black) are shown.



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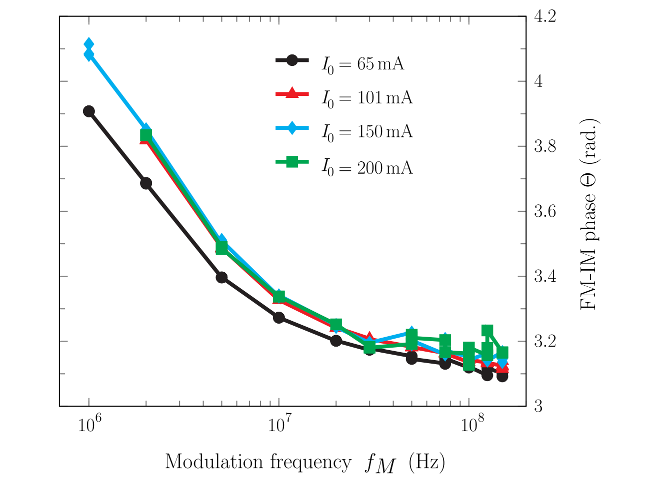

(b)

Fig. 5 (a) FM–IM ratio and (b) FM–IM phase as a function of the modulation frequency for four different DC currents.

FM–IM ratio and phase indicates the onset of thermal tuning, which is a figure of merit in ring QCLs. Based on these tuning characteristics, HPSDS gas sensing measurements were performed using a different absorption line of CH_4. The laser characterization confirmed that a low bias current and a high modulation frequency are beneficial for HPSDS. On the other hand, a large optical intensity is advantageous for a high signal-to-noise ratio. Operating the laser at a DC current of 92 mA (120 K) and 183.5 mA (100 K) with a modulation frequency of 150 MHz, different gas concentrations are investigated. Figure 6 shows the corresponding normalized phase in a concentration range from 1,000 to 12,500 ppm. The different linear ranges for the two bias current setpoints are clearly visible. This can be attributed to the different FM/IM indices for the two setpoints [compare Fig. 5(a)].[35] The low bias current setpoint corresponds to a predominantly amplitude modulated case (small FM/IM index), yielding a large dynamic range, while a high bias current favors frequency modulation and gives larger phase shifts at the expense of limited linearity. The extracted 3σ limit of detection, defined as the noise-equivalent concentration, for DC currents of 92 and 183.5 mA is extrapolated from gas measurements with low peak absorption and amounts to 16 and

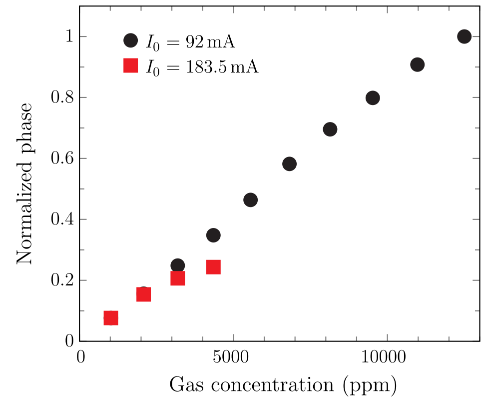

Fig. 6 Gas sensing results based on HPSDS. Normalized phase for different CH_4 concentrations obtained for a DC current of 92 mA (black) and 183.5 mA (red), both at 150 MHz.

2 ppm, respectively. The noise floor σ is measured with no CH_4 in the gas cell.

4 Conclusion

We present recent advances in the field of ring QCLs for chemical sensing applications. These light sources have been implemented in monolithically integrated on-chip laser/detector devices for gas sensing. Prototype devices featuring a disk and a ring detector have been realized. The latter enables mutual commutation between laser and detector. Furthermore, an introductive characterization of the radio frequency modulation characteristics up to 150 MHz of surface emitting ring QCLs is demonstrated. This knowledge paves the way for an implementation of these compact and versatile midinfrared light sources in gas sensing applications exploiting efficient spectroscopic techniques, such as HPSDS.

Disclosures

The authors declare no competing financial interest.

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