Remote Sensing with Commutable Monolithic Laser and Detector

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Supporting Information

**ABSTRACT:** The ubiquitous trend toward miniaturized sensing systems demands novel concepts for compact and versatile spectroscopic tools. Conventional optical sensing setups include a light source, an analyte interaction region, and a separate external detector. We present a compact sensor providing room-temperature operation of monolithic surface-active lasers and detectors integrated on the same chip. The differentiation between emitter and detector is eliminated, which enables mutual commutation. Proof-of-principle gas measurements with a limit of detection below 400 ppm are demonstrated. This concept enables a crucial miniaturization of sensing devices.

**KEYWORDS:** quantum cascade laser, quantum cascade detector, sensor, on-chip, monolithic integration

Midinfrared (MIR) spectroscopy is a powerful tool for chemical sensing applications. Molecules exhibit unique rotational and vibrational resonances, resulting in characteristic absorption patterns in the molecular “fingerprint” region. Probing these distinct resonances allows identifying and quantifying a great many chemical substances. Therefore, MIR spectroscopy is widely used in industrial and environmental monitoring as well as medical and biochemical diagnostics. Due to their small size, high spectral density, and tailorable on-demand emission wavelength, quantum cascade lasers (QCLs) are outstanding light sources for MIR applications. They are utilized in many spectroscopic techniques, such as direct absorption spectroscopy, quartz-enhanced photoacoustic spectroscopy, dual-comb spectroscopy, or chirped laser dispersion spectroscopy. Furthermore, chemical sensing with coupled cavities, substrate-integrated hollow waveguides, or micro-opto-electro-mechanical gratings demonstrates the important role of QCLs in this field. Most of these techniques share the typical spectroscopic sensing setup including a light source, an interaction region with the analyte, and a separate detector. All components have to be coordinated and aligned properly, which can be a challenging task. The increasing demand for compact and versatile sensing solutions requires reductions in total sensor size and number of components with simultaneous performance enhancement and simplification of the setup.

The demonstration of detector operation§ of QCLs initiated intensive research in the field of quantum cascade detectors (QCDs), resulting in the demonstration of diagonal optical transitions, plasmonic GaN QCDs, plasmonic lens enhanced light collection, and II−VI high-performance material systems as well as the suitability of QCDs for spectroscopy with higher operation temperatures than quantum well infrared photodetectors, enabled by their unbiased photovoltaic operation without any bias-induced dark current. Due to the different biasing conditions, the emission wavelength of the biased heterostructure is red-shifted with respect to the absorption wavelength of the same heterostructure in the unbiased case. This prevents the application of one and the same semiconductor chip in a single sensing setup. 

Here, we present a highly integrated MIR remote sensor based on bifunctional quantum cascade structures. The sensor geometry is designed to enable efficient mutual commutation between laser and detector. Two concentric ring waveguides are used for both vertical emission and detection; that is, both monolithic rings can be operated as laser and detector. Overcoming the distinction between emitter and detector, this concept facilitates a crucial miniaturization and simplification of the setup.

Received: August 13, 2016
Published: October 5, 2016

DOI: 10.1021/acsphto.6b00603
ACS Photonics 2016, 3, 1794−1798
tion of sensing devices. We introduce the working principle of the sensor and show proof-of-concept gas sensing measurements at room temperature.

Due to intersubband selection rules, devices such as QCLs and QCDs generate and detect only TM-polarized light. In order to enable laser surface emission as well as normal incidence detection, coupling schemes such as second-order distributed feedback (DFB) gratings, photonic (quasi-) crystals, or random optical media are necessary. Our sensor consists of two concentric monolithically integrated ring waveguides with second-order DFB gratings on top, as shown schematically in Figure 1. This DFB grating enables vertical emission and detection of light. Inner and outer rings have a diameter of 400 and 330 μm, respectively. Each ring is equipped with a separate top and bottom contact to filter the crosstalk between the detector and the pulsed laser. The inset in Figure 1 shows a scanning electron microscope (SEM) image of a section of the sensor with both waveguides and DFB gratings.

Both rings are fabricated from the same high-performance bifunctional quantum cascade material with a spectral overlap of laser and detector operation around 6.7 μm. The active region is based on an InGaAs/InAlAs heterostructure grown by molecular beam epitaxy on InP. A more detailed description of the epitaxial layers can be found in ref 26. This design allows a ring in detector operation to sense the light emitted from a ring in laser operation. The room-temperature performance of the separate sensor elements is demonstrated in Figure 2. Inner and outer rings exhibit single-mode laser emission around 1484/cm driven by 40 kHz/100 ns pulses with a peak power of 79 and 100 mW, respectively. Both rings show broadband detector absorption spectra with a main peak from 1200 to 1700/cm. A room-temperature peak responsivity of 1.2 mA/W is found for both rings at 1481/cm. This excellent spectral overlap enables efficient coupling of the light. The coupling scheme is explicated in the remote sensing setup illustrated in Figure 3.

Surface normal light emerging from one of the rings is collimated by a lens and transmits the 10 cm long gas cell before it is back-reflected by a flat gold mirror and travels on the same path back to the sensor chip, where it is detected by the other ring. This doubles the interaction length with the analyte compared to a single-pass configuration and can be used to measure in harsh environments. The detector signal is preamplified with 1 V/200 nA and then measured using a lock-in amplifier with its trigger signal coming from the chopper, spinning at 300 Hz, to filter the crosstalk between

![Figure 1. Sketch of the remote sensor consisting of two concentric ring waveguides with a DFB grating on top. Depending on the biasing condition, each ring can be operated as laser and detector. Both rings are electrically isolated from each other and possess their own top (TC) and bottom (BC) contact in order to filter the crosstalk. The inset shows a SEM image of a section of the sensor with both waveguides and DFB gratings.](image1)

![Figure 2. Spectral performance of the sensor measured with a DTGS detector for the laser and a globar source for the detector operation. The spectra of the outer ring are shifted vertically to improve visibility. The DFB grating strongly influences the lasing procedure, eventually producing single-mode emission. In the detection scheme the DFB grating acts as a coupling element, resulting in a broadband absorption behavior. The accurate spectral overlap enables efficient coupling of light from the laser to the detector via surface emission and subsequent reflection.](image2)

![Figure 3. Remote sensing setup. Light emitted from one ring of the sensor is collimated by a lens and travels through the gas cell. At the flat gold mirror it is back-reflected and propagates along the initial path in the reverse direction back to the sensor chip, where it is detected by the other ring.](image3)
laser and detector. Using this setup the sensor can be operated in two configurations, which are inverted to each other: (i) inner ring as detector and outer ring as laser; (ii) outer ring as detector and inner ring as laser. Figure 4 shows the performance of both configurations compared to an external detection scheme. These measurements were carried out without an analyte inside the gas cell. The emission characteristics of both rings are measured with the on-chip detector, i.e., the other ring, and with the external detection scheme utilizing a calibrated DTGS detector instead of the mirror. Both configurations clearly show the onset of lasing as well as an overall good agreement between the on-chip and external measurements. Small discrepancies are attributed to different on-chip coupling efficiencies.

As shown in Figure 2, both rings exhibit similar detector performance because the absorption characteristics mainly depend on the quantum cascade heterostructure and the DFB grating simply couples the light to the active zone inside the cavity. On the other hand the emission in the lasing configuration strongly depends on the DFB grating and can be tailored accordingly. Our sensor features laser emission at 1484.3/cm and 1483.3/cm for the outer and inner ring, respectively. The corresponding laser spectra are shown in Figure 5 together with the absorbance spectra of the two analyte gases isobutene and isobutane, which were utilized for the proof-of-principle measurements. Isobutene possesses a linear absorbance behavior around both emission lines. Isobutane, on the other hand, exhibits a quadratic behavior with an increasing absorbance within the emission pulse of the inner ring. This should result in a stronger signal attenuation for the sensor configuration (ii). Figure 6a and b indicate the transmittance vs concentration for isobutene and isobutane, respectively. The on-chip detector signal was averaged over 120 data points with a lock-in time constant of 0.5 s. A good agreement with the theoretical prediction of the Beer–Lambert law is achieved over a concentration range of 0–16%. However, for higher concentrations a small deviation from the Beer–Lambert law is evident for the isobutane measurement. Explicit absorbance values are extracted from the measured data. They are given in the top right corner of each graph. Our data show a rather similar transmissive behavior for both gases in configuration (i). We can extract absorbance values of $A_{\text{C}_4\text{H}_8(i)} = 4.75 \times 10^{-5}$ and $A_{\text{C}_4\text{H}_{10}(i)} = 4.78 \times 10^{-5}$ for isobutene and isobutane, respectively. In contrast to that, configuration (ii) reveals a significantly stronger absorbance of $A_{\text{C}_4\text{H}_8(ii)} = 5.49 \times 10^{-5}$ for isobutene compared to $A_{\text{C}_4\text{H}_{10}(ii)} = 5.07 \times 10^{-5}$ for isobutane. This confirms the increasing absorbance curve from literature within the emission line of the inner ring laser. Altogether a good agreement between the literature and experimental absorbance values is observed. A 3σ limit of detection (LOD) of 397 ppm for isobutane in the configuration (i), without temperature stabilization of the sensor, was achieved. This performance is comparable to in-plane bifunctional sensors for detection in liquids that deal with much shorter interaction regions and directly couple the light in and out via the facet. Compared with previously demonstrated sensor geometries, the double-ring configuration exhibits an enhanced detection of the incoming light. This is attributed to the fact that the back-reflected light distribution incident on the sensor chip in the presented sensing setup is ring-shaped and can be best detected by a ring detector. Furthermore, this ring detector in turn can also be used as a laser. This increases the performance as well as the variety of applications. The demonstrated detection sensitivity is mainly limited by noise generated by electrical crosstalk. In order to improve the sensitivity, a reduction of the crosstalk is necessary, which can be realized by etching a deep trench between the inner and outer ring or by enabling continuous wave operation of the laser. A stronger tuning of the laser wavelength in combination with a time-resolved detection scheme could further improve the sensitivity of the sensor. A multichannel array of these double-ring sensors would increase the selectivity of the presented prototype.

In summary, we present a compact and efficient remote sensor concept based on bifunctional and commutable quantum cascade lasers and detectors. This concept consists of two concentric ring waveguides with DFB gratings and provides room-temperature surface lasing and detection at two different wavelengths monolithically integrated on the same
Comparison between ridge and DFB ring detector, simulated laser and detector performance as a function of the grating parameters, and Allan variance plot (PDF)

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Notes
The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

The authors acknowledge the support by the Austrian Science Fund project Next-Lite (FWF: F49-P09).

■ REFERENCES

(1) Faist, J.; Capasso, F.; Sivco, D. L.; Sirtori, C.; Hutchinson, A. L.; Cho, A. Y. Quantum Cascade Laser. Science 1994, 264, 553–556.
(2) Hancock, G.; van Helden, J. H.; Peev, R.; Ritchie, G. A. D.; Walker, R. J. Direct and wavelength modulation spectroscopy using a cw external cavity quantum cascade laser. Appl. Phys. Lett. 2009, 94, 201110–201112.
(3) Lewicki, R.; Wysocki, G.; Kosterev, A. A.; Tittel, F. K. QEPAS based detection of broadband absorbing molecules using a widely tunable, cw quantum cascade laser at 8.4 µm. Opt. Express 2007, 15, 7357–7366.
(4) Villares, G.; Hugi, A.; Blaser, S.; Faist, J. Dual-comb spectroscopy based on quantum-cascade-laser frequency combs. Nat. Commun. 2014, 5, 5192–5200.
(5) Wysocki, G.; Weidmann, D. Molecular dispersion spectroscopy for chemical sensing using chirped mid-infrared quantum cascade laser. Opt. Express 2010, 18, 26123–26140.
(6) Fuchs, P.; Seufert, J.; Koeth, J.; Semmel, J.; Höfling, S.; Worschech, L.; Forchel, A. Widely tunable quantum cascade lasers with coupled cavities for gas detection. Appl. Phys. Lett. 2010, 97, 181111–181113.
(7) Türüncü, E.; Kokoric, V.; Szedlak, R.; MacFarland, D.; Zederbauer, T.; Detz, H.; Andrews, A. M.; Schrenk, W.; Strasser, G.; Mizakoff, B. Analyt. Chem. 2016, Advance Article, 10.1021/acsanw110130F.
(8) Wagner, J.; Ostendorf, R.; Grahmann, J.; Merten, A.; Hugger, S.; Jarvis, J.-P.; Fuchs, F.; Bokovic, D.; Schenck, H. Widely tunable quantum cascade lasers for spectroscopic sensing. Proc. SPIE 2015, 9370, 937012–937017.
(9) Hofstetter, D.; Beck, M.; Faist, J. Quantum-cascade-laser structures as photodetectors. Appl. Phys. Lett. 2002, 81, 2683–2685.
(10) Giorgetta, F. R.; Baumann, E.; Graf, M.; Yang, Q.; Manz, C.; Kohler, K.; Beere, H. E.; Ritchie, D. A.; Linfield, E.; Davies, A. G.; Fedoryshyn, Y.; Jackel, H.; Fischer, M.; Faist, J.; Hofstetter, D. Quantum Cascade Detectors. IEEE J. Quantum Electron. 2009, 45, 1039–1052.
(11) Reiningher, P.; Schwarz, B.; Gansch, R.; Detz, H.; MacFarland, D.; Zederbauer, T.; Andrews, A. M.; Schrenk, W.; Strasser, G. Quantum cascade detector utilizing the diagonal-transition scheme for high quality cavities. Opt. Express 2015, 23, 6283–6291.
(12) Pesach, A.; Sakr, S.; Giraud, E.; Sorias, O.; Gal, L.; Tchernycheva, M.; Orenstein, M.; Grandjean, N.; Julien, F. H.; Bahir, G. First demonstration of plasmonic GaN quantum cascade detectors with enhanced efficiency at normal incidence. Opt. Express 2014, 22, 21069–21078.
(13) Harrer, A.; Schwarz, B.; Gansch, R.; Reiningher, P.; Detz, H.; Zederbauer, T.; Andrews, A. M.; Schrenk, W.; Strasser, G. Plasmonic lens enhanced mid-infrared quantum cascade detector. Appl. Phys. Lett. 2014, 105, 171112–171115.

Figure 6. Measured (dots) transmittance of (a) isobutene and (b) isobutane as a function of the concentration and associated fits (lines) according to the Beer–Lambert law. Left and right y-axis show the same data in linear and logarithmic representation, respectively. The extracted absorbance values for both configurations and gases are displayed in the top right corner of each graph. These values show a good agreement with the literature absorbance values in Figure 5.

chip. Two operation configurations are possible, with the inner ring as the detector and the outer ring as the laser and vice versa. We present proof-of-principle gas sensing experiments and reach a LOD of below 400 ppm for isobutane with our prototype. Quantum cascade lasers enable tailoring of the emission wavelength for many applications and various traceable analytes. Potential array integration facilitates multicolor spectroscopy. This sensor combines the versatility of quantum cascade heterostructures and favorable qualities of surface emission and detection to achieve long analyte interaction lengths with the compactness of a monolithically integrated remote sensor. Therefore, this concept could be suitable for compact hand-held remote sensors in a variety of fields.

■ ASSOCIATED CONTENT

Supporting Information
The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsphotonics.6b00603.
(14) Ravikumar, A. P.; De Jesus, J.; Tamargo, M. C.; Gmachl, C. F. High performance, room temperature, broadband II-VI quantum cascade detector. Appl. Phys. Lett. 2015, 107, 141105−141108.

(15) Hofstetter, D.; Giorgetta, F. R.; Baumann, E.; Yang, Q.; Manz, C.; Köhler, K. Mid-infrared quantum cascade detectors for applications in spectroscopy and pyrometry. Appl. Phys. B: Lasers Opt. 2010, 100, 313−320.

(16) Schneider, H.; Schönbein, C.; Walther, M.; Schwarz, K.; Fleissner, J.; Koidl, P. Photovoltaic quantum well infrared photodetectors: The four-zone scheme. Appl. Phys. Lett. 1997, 71, 246–248.

(17) Schwarz, B.; Reinerger, P.; Detz, H.; Zederbauer, T.; Maxwell Andrews, A.; Kalchmair, S.; Schrenk, W.; Baumgartner, O.; Kosina, H.; Strasser, G. A bi-functional quantum cascade device for same-frequency lasing and detection. Appl. Phys. Lett. 2012, 101, 191109−191112.

(18) Schwarz, B.; Reinerger, P.; Ristanić, D.; Detz, H.; Andrews, A. M.; Schrenk, W.; Strasser, G. Monolithically integrated mid-infrared lab-on-a-chip using plasmonics and quantum cascade structures. Nat. Commun. 2014, 5, 4085−4091.

(19) Harrer, A.; Szedlak, R.; Schwarz, B.; Moser, H.; Zederbauer, T.; MacFarland, D.; Detz, H.; Andrews, A. M.; Schrenk, W.; Lendl, B.; Strasser, G. Mid-infrared surface transmitting and detecting quantum cascade device for gas-sensing. Sci. Rep. 2016, 6, 21795−21800.

(20) Szedlak, R.; Holzbauer, M.; MacFarland, D.; Zederbauer, T.; Detz, H.; Andrews, A. M.; Schrenk, W.; Strasser, G. The influence of whispering gallery modes on the far field of ring lasers. Sci. Rep. 2015, 5, 16668−16675.

(21) Bai, Y.; Tsao, S.; Bandyopadhyay, N.; Slivken, S.; Lu, Q. Y.; Caffey, D.; Pushkarsky, M.; Day, T.; Razeghi, M. High power, continuous wave, quantum cascade ring laser. Appl. Phys. Lett. 2011, 99, 261104−261106.

(22) Boyle, C.; Sigler, C.; Kirch, J. D.; Lindberg, D. F.; Earles, T.; Botez, D.; Mawst, L. J. High-power, surface-emitting quantum cascade laser operating in a symmetric grating mode. Appl. Phys. Lett. 2016, 108, 121107−121111.

(23) Colombelli, R.; Srinivasan, K.; Troccoli, M.; Painter, O.; Gmachl, C. F.; Tennant, D. M.; Sergent, A. M.; Sirco, D. L.; Cho, A. Y.; Capasso, F. Quantum Cascade Surface-Emitting Photonic Crystal Laser. Science 2003, 302, 1374−1377.

(24) Vitiello, M. S.; Nobile, M.; Ronzani, A.; Tredicucci, A.; Castellano, F.; Talora, V.; Li, L.; Linfield, E. H.; Davies, A. G. Photonic quasi-crystal terahertz lasers. Nat. Commun. 2014, 5, 5884−5891.

(25) Degl’Innocenti, R.; Shah, Y. D.; Masini, L.; Ronzani, A.; Pitanti, A.; Ren, Y.; Jessop, D. S.; Tredicucci, A.; Beere, H. E.; Ritchie, D. A. Hyperuniform disordered terahertz quantum cascade laser. Sci. Rep. 2015, 6, 19325−19331.

(26) Schwarz, B.; Ristanić, D.; Reinerger, P.; Zederbauer, T.; MacFarland, D.; Detz, H.; Andrews, A. M.; Schrenk, W.; Strasser, G. High performance bi-functional quantum cascade laser and detector. Appl. Phys. Lett. 2015, 107, 071104−071107.

(27) Brandstetter, M.; Genner, A.; Schwarz, C.; Mujagic, E.; Strasser, G.; Lendl, B. Time-resolved spectral characterization of ring cavity surface emitting and ridge-type distributed feedback quantum cascade lasers by step-scan FT-IR spectroscopy. Opt. Express 2014, 22, 2656−2664.