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Vibrational Spectroscopy

### Ring quantum cascade lasers with grating phase shifts and a light collimating dielectric metamaterial for enhanced infrared spectroscopy



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#### 1. Introduction

#### A great many substances exhibit rotational and vibrational resonances in the mid-infrared, the so-called fingerprint region of the electromagnetic spectrum. Industrial process control, health and environmental sciences as well as many other scientific and technological fields demand powerful and reliable light sources in order to analyze these resonances. Quantum cascade lasers [1] (QCLs) have proven to be favorable instruments for non-destructive identification and concentration measurements of a variety of chemical compounds [2-5]. In contrast to conventional Fourier transform infrared spectrometers (FTIR), QCLs are small, versatile and show a much higher spectral density. Medical diagnostics including non-invasive blood sugar monitoring [6-8] and the detection and analysis of exhaled human breath [9,10] using QCLs have been developed in the last years. Likewise, QCLs started to play an important role in industrial and environmental applications like on-chip [11] absorption spectroscopy of liquids [12] and trace gas sensing [13] with continuous-wave QCLs [14,15] or novel

#### ABSTRACT

Brighter and more efficient light sources are desired to further increase the sensitivity of spectroscopic measurements. Simultaneously, a strong trend towards miniaturization and compactness is ubiquitous. In the last years quantum cascade lasers have become powerful and reliable tools for infrared spectroscopy. We report on ring quantum cascade lasers with two distinct features which increase the efficiency of these devices and provide enhanced performance for spectroscopic applications. First, the distributed feedback grating exhibits two  $\pi$ -phase shifts. This provides far fields with a central intensity maximum. Second, a gradient index metamaterial is fabricated into the substrate. This collimates the emitted light and can replace external optics.

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techniques like chirped laser dispersion spectroscopy [16] or quartz-enhanced photoacoustic spectroscopy [17]. For these applications, typically, edge-emitting Fabry-Pérot (FP) resonators are used. Recently, we introduced surface-emitting quantum cascade based ring lasers [18] with a second order distributed feedback (DFB) grating for vertical light outcoupling. These devices provide high-power performance [19] and exhibit many advantages compared to FP-lasers [20] including a much more collimated emission beam due to the larger emitting area. In this paper we combine ring QCLs having two  $\pi$ -phase shifts in the DFB grating with a dielectric gradient index [21,22] metamaterial layer on the substrate side of the chip. While the phase shifts provide a far field with a central intensity maximum, the metamaterial is increasing the emitting area and collimating the emission beam [23]. Compared to other light collimation techniques using plasmonic antennas [24] or nanopores [25], our dielectric metamaterial shows a high efficiency and does not suffer from scattering losses.

#### 2. Theory and methods

Corresponding author. E-mail address: rolf.szedlak@tuwien.ac.at (R. Szedlak). The laser is based on an  $In_{0.52}Al_{0.48}As/In_{0.53}Ga_{0.47}As$  [26] active region grown on an InP substrate. The ring resonator has an outer

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diameter of 400  $\mu$ m, a width of 10  $\mu$ m and a height of 7  $\mu$ m. The metamaterial on the other side of the chip exhibits a width of 80  $\mu$ m and lies exactly below the laser as illustrated in Fig. 1.

Processing steps of the laser are according to previous publications [27,28].

#### 2.1. Phase shifts

Surface emitting ring lasers with a regular second order distributed feedback grating for light outcoupling exhibit an inherent intensity minimum in the center of the far field [29]. High performance spectroscopic applications though require a light beam with as much intensity in the beam center as possible. For straight DFB lasers a central intensity maximum can be achieved via absorption of the antisymmetric mode [30]. Furthermore, it has been shown for ridge [31] and ring lasers [28] that grating phase shifts can produce a central-lobe emission beam as well. Fig. 2 shows a comparison between a standard and a phase shifted grating for ring lasers.

Opposing sides of the standard ring show an antiparallel orientation of the emitted electric field vectors which causes destructive interference in the center of the far field. The  $\pi$ -phase shift compensates for this effect and opposing sides of the ring exhibit a parallel orientation of the field vector. This results in constructive interference and a central intensity maximum. From the area of the phase shifts itself no light is emitted due to local destructive interference of the clockwise and counter-clockwise oriented electric field vectors.

#### 2.2. Metamaterial

For the metamaterial a 500 nm thick SiN layer is deposited on the substrate. A polymethylmethacrylate film on top of the SiN layer is exposed by electron beam lithography and serves as an etch mask for the SiN which in turn forms the etch mask for the substrate. Prior to the exposure the coordinates of the ring laser with respect to the edges of the chip are determined using scanning electron microscopy. This is necessary in order to correctly position the metamaterial on the back side. The collimating metamaterial consists of differently sized sub-wavelength holes which form a refractive index gradient in radial direction. The nominal etch depth of 4.2  $\mu$ m is reduced for holes with a diameter less than 1.5  $\mu$ m due to aspect ratio dependent etching (ARDE). These holes were designed to be accordingly larger to match the required refractive index given by

$$n(r) = n_0 + n_0 \frac{d_{\rm sub}}{d_{\rm met}} \left( 1 - \frac{\sqrt{d_{\rm sub}^2 + r^2}}{d_{\rm sub}} \right)$$
(1)

with the radial parameter  $r = r' - r_0$ , the refractive index of InP  $n_0 = 3.06$ , the substrate thickness  $d_{sub}$  and the metamaterial thickness  $d_{met}$ . The refractive index can also be expressed with the hole diameter b(r) and the hole period  $d_r = 1.8 \,\mu\text{m}$  as follows

$$n_0(d_r - b(r)) + b(r) = d_r n(r)$$
 (2)

The corrected hole diameter is given by equalizing Eqs. (1) and (2) using the variable etch depth  $d_{met} = u + vb$ . The ARDE parameters u = -0.79 and v = 3.08 were extracted from the inspection of vertical cross sections through the hole array which were prepared by focused ion beam milling [23]. This results in a quadratic equation with the following solution for the corrected hole diameter

$$b(r) = \frac{-B \pm \sqrt{B^2 + 4AC}}{2A}$$

$$A = v - n_0 v$$

$$B = u - n_0 u - d_r v$$

$$C = d_r u + \left(1 - \frac{\sqrt{d_{sub}^2 + r^2}}{d_{sub}}\right) d_r d_{sub} n_0$$

The working principle of the metamaterial is based on the idea that light rays emitted from the ring laser under different angles propagate through material with different refractive index. The higher the emission angle the lower the refractive index. The metamaterial is designed as such that all rays emitted under a critical angle  $\alpha_c$  cover the same optical path length inside the substrate and metamaterial layer. In other words, the spherical wavefront is deformed and planarized within the metamaterial. From this point, according to the Huygens-Fresnel principle, the light emerges again in a spherical wavefront but from a much larger emitting area, namely the complete surface area of the metamaterial. The emitted beam is therefore more collimated than the initial beam of the ring laser.

The refractive index shape of the metamaterial is fixed and can only be altered by adjusting the etch depth which corresponds to the nominal thickness  $d_{met}$  of the metamaterial. The larger the etch



**Fig.1.** Sketch of the front (left) and back (right) side of the laser device. The latter exhibits a gradient index metamaterial layer which collimates the light emitted by the ring laser on the front side. The center of the waveguide is located at the radial distance *r*<sub>0</sub>.

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