Terahertz sources based on Čerenkov difference-frequency generation in quantum cascade lasers

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Room-temperature terahertz (THz, \( \lambda = 30–300 \) \( \mu \)m) semiconductor lasers are highly desired for terahertz applications. Despite the progress with THz quantum cascade lasers (QCLs), existing devices still require cryogenic cooling. We have previously demonstrated that dual-wavelength mid-infrared QCLs with giant optical nonlinearities may be used to produce THz output via intra-cavity difference-frequency generation (DFG). This approach has recently led to the realization of room-temperature QCLs operating in the 4–5 THz range with approximately 3.5–5 \( \mu \)W/W\(^2\) mid-infrared-to-terahertz power conversion efficiency for multi-mode generation and up to 13 \( \mu \)W of THz power output. However, these sources are still highly inefficient because THz radiation produced further than \( \sim 100 \mu \)m away from the output facet is completely absorbed by free carriers. In this report we demonstrate that a Čerenkov DFG scheme may be used to create THz QCL sources in which THz radiation is extracted along the whole length of the waveguide. Reported devices have approximately one order of magnitude higher conversion efficiency for multi-mode operation compared to that in Refs. 7 and 8 and provide room temperature THz output in 1.2–4.5 THz range.

Previous THz DFG QCL sources rely on modal phase matching. Their waveguide is designed to provide confinement for both mid-IR and THz modes within the laser active region. The active region is designed to provide dual-color mid-infrared emission and optical nonlinearity. It needs to have a sufficiently high doping density \( n \approx 5 \times 10^{16} \) cm\(^{-3}\) is used in Refs. 6–8) to provide high-power mid-IR output and high optical nonlinearity; waveguide cladding layers need to have similar doping to provide sufficient conductivity. This doping leads to high optical losses (\( x \)) at THz frequencies. Detailed waveguide calculations for 4–5 THz sources reported give a value for 1/\( x \) of \( \sim 40 \) \( \mu \)m (Ref. 7) and \( \sim 67 \) \( \mu \)m. THz DFG occurs along the whole waveguide length in these devices. However, high THz losses imply that for a 3-mm-long THz DFG QCL, over 97% of THz light generated at 4–5 THz is absorbed in the QCL waveguide. The situation is significantly worse for generation at lower THz frequencies, due to increased free carrier absorption. This problem has been noticed in the very first report of THz DFG in QCLs, and a surface-emission scheme using second-order gratings was proposed to extract THz radiation along the laser waveguide. For this scheme to work efficiently, the second-order grating must not affect mid-IR modes and have the THz mode out-coupling coefficient \( x \) similar to \( z \). It is difficult to implement such a strong grating in practice and experimental implementation did not lead to an output power increase.

Here we demonstrate that an intra-cavity Čerenkov DFG scheme may be used to efficiently extract THz radiation along the whole length of a QCL waveguide and produce THz QCL sources with broadband directional THz output. Čerenkov THz DFG emission occurs when the nonlinear polarization wave propagates at a higher phase velocity compared to that of the THz radiation. In terms of propagation constants, this means that the propagation constant of the nonlinear polarization wave \( (k_{\text{oa}}) \) is smaller compared to that of the THz radiation \( (k_{\text{THz}}) \). In this case, generated radiation is emitted at the Čerenkov angle

\[
\theta_C = \cos^{-1}(k_{\text{oa}}/k_{\text{THz}})
\]

relative to the direction of \( k_{\text{oa}} \), as shown schematically in Fig. 1(a). Experimentally, Čerenkov nonlinear emission was first observed for second harmonic generation (SHG) and later for DFG. Since then, it has been observed in a wide range of devices and processes, including SHG in GaAs/AlGaAs mid-IR QCLs.

In the case of DFG in QCLs, the propagation constant of the nonlinear polarization wave in the active region is given as \( \beta_{\text{oa}} = \beta_{\text{oa}}(\omega_1) \), where \( \beta_{\text{oa}} = n_{\text{eff}}(\omega_1) \alpha/\epsilon \) with \( n_{\text{eff}}(\omega_1) \) being the effective refractive index of the mid-IR pump mode at frequency \( \omega_1 \). Since the two mid-IR pump frequencies are close, \( \omega_1 \approx \omega_2 \), one can write...
FIG. 1. (a) Schematic of Čerenkov THz DFG emission in a QCL. Waveguide cladding layers are shown in light gray, top gold contact layer is shown in yellow. Čerenkov THz radiation emitted into the substrate is shown with arrows. It may also be emitted towards the top contact layer (show with dashed arrows) and reflected to the substrate. (b) Facet-view schematic. Gold contact layers are shown as yellow, insulating SiN layers are shown as blue, InP layers are shown as grey and labeled, active region with two sections designed to emit mid-IR pumps at \( \omega_2 \) and \( \omega_3 \) is shown in brown. (c) Vertical refractive index profile in our devices for mid-IR \((\lambda = 9\mu m, \text{grey line, right axis})\) and 4.5 THz \((\text{black line, right axis})\) light. Position \( x = 0 \) corresponds to the middle of the active region, see (b). Also shown is the computed mid-IR waveguide mode. Colored regions indicate waveguide cladding layers (dark grey), active region (red), and substrate (light grey).

\[
|\beta_1 - \beta_2| \approx \frac{n_g \omega_{THz}}{c},
\]

where \( n_g = n_{eff}(\omega_1) + \omega_1 \frac{\partial n_{eff}}{\partial \omega} \bigg|_{\omega = \omega_1} \) is the group effective refractive index at \( \omega_1 \) and \( \omega_{THz} = \omega_1 - \omega_2 \) is the THz difference-frequency. In order to produce Čerenkov DFG emission into the substrate, the substrate refractive index at \( \omega_{THz} \) must be larger than \( n_g \). We have realized that this condition is satisfied throughout the 1–5 THz spectral range for InP/GaInAs/AlInAs QCLs grown on semi-insulating InP. As a result, efficient broadband THz QCL sources based on Čerenkov DFG can easily be implemented.

The schematic of proof-of-principle devices is shown in Fig. 1(b). The laser structure is virtually identical to that of the first room-temperature THz DFG QCLs, except that here we used a semi-insulating InP substrate and a lateral current extraction scheme (similar to that employed in GaAs/AlGaAs QCLs with semi-insulating waveguides\(^2\)). The refractive index profile for mid-IR and THz frequencies in our laser is shown in Fig. 1(c). In mid-IR, the refractive index of the substrate is low and allows for good mode confinement. In the 1–5 THz range, due to the Reststrahlenband at 8–10 THz, the refractive index of undoped InP is high and the Čerenkov condition is fulfilled. The waveguide calculations for our devices give \( n_g \approx 3.37 \) in mid-IR. Given the refractive index of undoped InP of 3.6 (virtually independent of frequency in 1–5 THz range),\(^{10} \) we obtain a Čerenkov angle \( \theta_C \approx 21^\circ \) for DFG in the whole 1–5 THz range. Once in the substrate, THz radiation propagates towards the facet. Since undoped InP is virtually lossless over 1–5 THz, the Čerenkov emission scheme allows for efficient extraction of THz radiation along the whole length of the QCL waveguide. To avoid total internal reflection of the THz Čerenkov wave at the front facet, the substrate has to be polished at a 20°–30° angle as shown in Fig. 1(a).

Devices were grown by molecular beam epitaxy (MBE) on 350-μm-thick semi-insulating InP substrates. InGaAs/AlInAs heterostructures lattice-matched to InP was used. The growth started with a 3-μm-thick current extraction layer made of InP doped to \( n = 1 \times 10^{17} \text{cm}^{-3} \), followed by 33 repetitions of the “bound-to-continuum” active region with integrated optical nonlinearity and 27 repetitions of the “double-phonon resonance” active region, separated by a 100-nm-thick InGaAs spacer region doped to \( n = 5 \times 10^{16} \text{cm}^{-3} \).

To be able to see the advantage of the Čerenkov DFG scheme over a modal phase matching scheme,\(^7\) the bandstructure design of the two active region sections in our devices was chosen to be identical to that used in Refs. 7 and 8. The top cladding layer consists of 3.5-μm-thick InP n-doped to \( 5 \times 10^{16} \text{cm}^{-3} \) followed by a 200-nm-thick InP n-doped to \( 2 \times 10^{18} \text{cm}^{-3} \).

The material was processed into 35-μm-wide ridge waveguides via dry etching. The sidewalls of the ridges were insulated with a 600-nm-thick layer of SiN, followed by a Ti/Pt/Au (30 nm/60 nm/1000 nm) contact layer deposition. Figure 1(b) shows the cross section of the processed ridge laser. The wafer was then cleaved into laser bars with 1-mm-long devices. A high reflectivity coating of Al₂O₃/Ti/Au (100 nm/5 nm/50 nm) was then evaporated onto the back facets of devices. The 350-μm-thick InP substrate associated with the exit facet of the device was then mechanically polished to the desired angle with a combination of SiC lapping compound and Al₂O₃ lapping film. Special care was taken to ensure that the laser waveguide facets are unaffected by polishing. Devices were then indium soldered onto copper holders and wire bonded. A high resistivity silicon micro lens was attached to the THz exit facet with Epoxy adhesive in order to improve outcoupling and collection efficiency of the THz beam.

Spectral emission of both the mid-infrared and terahertz beams was measured using a Fourier-transform infrared spectrometer. A helium-cooled silicon bolometer and deuterated L-alanine doped triglycine sulfate (DLaTGS) detector were used for the THz and mid-infrared spectral measurements, respectively. The power output of these devices was measured using a calibrated thermopile detector for the mid-infrared beams and a calibrated helium-cooled silicon bolometer for the THz beams. All data presented in the paper is corrected for the collection efficiency. The power collection efficiency for the output beams in our optical setup is approximately 70% for the mid-infrared emission and approximately 50% for the THz emission. Appropriate optical filters were used to differentiate between the mid-infrared pump beams and to separate mid-infrared and THz emission.

The results are shown in Fig. 2 for a 1-mm-long laser with the substrate polished at 20°. The mid-IR emission was observed at two groups of frequencies centered around 1020 cm⁻¹ (\( \lambda_1 = 9.8 \mu m \)) and 900 cm⁻¹ (\( \lambda_2 = 11.1 \mu m \)) as...
shown in the inset of Fig. 2. Experimentally, the mid-IR pump wavelengths are slightly longer compared to design wavelengths of 8.9 and 10.5 µm. Since QCLs with an identical active region design operated as designed earlier,7,8 the difference in the current devices is attributed to growth calibration. Power measurements were performed using band-pass filters to separate two groups of frequencies. The results are shown in Fig. 2 along with the current-voltage characteristic of the laser. We emphasize that the mid-IR pumps are unaffected by substrate polishing since the ridge facet and mid-IR waveguide structure remains intact during polishing. The improvement in the THz power output is thus entirely due to improved outcoupling of THz radiation. The room-temperature THz emission spectrum of the same device is displayed in Fig. 3(a). To maximize THz collection efficiency, a high-resistivity silicon hyperhemisphere lens was affixed to the polished substrate facet for spectral measurements. Optical filters were used to block mid-IR output. The spectrum shows three THz peaks in perfect agreement with the mid-IR pump frequency spacing shown in Fig. 2. Because of the large emitter area (whole laser waveguide), the Čerenkov THz wave is expected to be well-collimated in the direction normal to the waveguide layers. Figure 3(b) shows far-field THz emission profiles of three devices: 1-mm-long lasers with the substrate output facet as cleaved (vertical), polished at 20°, and polished at 30°. The far field pattern of the unpolished sample is very broad and weak. In these lasers the Čerenkov wave experiences total internal reflection, and only optical nonlinearity near the exit facet contributes to THz output, similar to devices reported in Refs. 6–8. Contrastingly, the samples polished to 20° and 30° had highly directional emission and strong output. For the case of 20° polishing, we see a sharp emission peak approximately 22.5° below normal incidence. In the case of 30° polishing, the beam emits almost normal to the exit facet of the ridge, with a peak 7° above normal incidence. In both cases, taking into account the refraction that occurs at the air-semiconductor interface, this corresponds to an internal emission angle at approximately 20.5° which is in good agreement with the theoretical value of θ_C ≈ 21°. Difference in the widths of the emission profiles in the case of 20° and 30° polishing in Fig. 3(b) is most likely due to curvature or other artifacts of the polished facet introduced during manual polishing.

Figure 3(a) displays the THz power output for two samples: one with an unpolished exit facet and the other one with exit facet polished to 20°. High resistivity silicon hyper-hemisphere lenses were affixed to the THz exit facet for both devices. The unpolished sample produced peak THz power output of only 0.07 µW at 300 K, which corresponds to a mid-IR to terahertz conversion efficiency of ≈7 µW/W². The 20° polished sample produced 0.5 µW of THz output power at 300 K, which corresponds to a conversion efficiency of ≈45 µW/W², which is approximately a factor of 10 above the conversion efficiencies in multi-mode THz DFG QCLs reported earlier.7,8 More detailed analysis of power distribution in mid-IR pumps and THz output may be performed by integrating areas below peaks in mid-IR (Fig. 2(a)) and THz (Fig. 3(a)) spectra. This analysis yields conversion efficiencies of 70, 40, and 8.4 µW/W² for THz emission lines at 3.3, 4.5, and 1.2 THz, respectively.

To evaluate the mid-IR-to-THz conversion efficiency in our devices theoretically, we treat Čerenkov emission as a leaky THz waveguide mode with the propagation constant along the waveguide direction fixed at β_{pol} − β_{zol}, following an approach described in Ref. 22. Assuming a constant nonlinear susceptibility χ^{(2)} = 4 × 10^4 pm/V in the bound-to-bound transition.
continuum section of the active region and neglecting optical nonlinearity elsewhere, we obtain an expected conversion efficiency of 3.3, 0.7, and 0.1 mW/W² for THz DFG at 4.5, 3.3, and 1.2 THz, respectively. The discrepancy with experimental results may be due to the fact that the theoretical results are sensitive²¹ to the values of refractive indices of different waveguide layers in the THz, and these are not known precisely, especially, for 4–5 THz frequencies which are close to the Reststrahlenband. Furthermore, the mid-IR pumps may be partly operating in higher-order waveguide modes as described in Ref. 22. Lasing in these high-order modes may be suppressed, e.g., by implementing DFB gratings for the TM₀₀ pumps as shown in Ref. 8. Single mode operation of the pumps is desirable, and it is beyond the scope of this paper to describe the methods that ensure it. But one possible way would be to use narrow ridge widths on the order of ~12 µm.²²

Future work will focus on improving the output power of devices. Besides optimization of the bandstructure and waveguide design, THz power scaling may be produced by extracting THz radiation along longer waveguide sections. This may be achieved, e.g., by growing lasers on thicker substrates to ensure that the Čerenkov emission does not get clipped by the backside of the substrate. We also expect to boost THz output power by applying a high-reflectivity front facet coating that enhances the intra-cavity mid-IR power and depositing anti-reflection THz coating on the polished side of the substrate. These improvements will be implemented in future work.

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