Broadly tunable terahertz difference-frequency generation in quantum cascade lasers on silicon

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Abstract. We report broadly tunable terahertz (THz) sources based on intracavity Cherenkov difference-frequency generation in quantum cascade lasers transfer-printed on high-resistivity silicon substrates. Spectral tuning from 1.3 to 4.3 THz was obtained from a 2-mm long laser chip using a modified Littrow external cavity setup. The THz power output and the midinfrared-to-THz conversion efficiency of the devices transferred on silicon are dramatically enhanced, compared with the devices on a native semi-insulating InP substrate. Enhancement is particularly significant at higher THz frequencies, where the tail of the Reststrahlen band results in a strong absorption of THz light in the InP substrate. © 2017 Society of Photo-Optical Instrumentation Engineers (SPIE) [DOI: 10.1117/1.OE.57.1.011020]

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1 Introduction

The terahertz (THz) spectral region from 0.3 to 10 THz hosts a wide range of applications, including noninvasive security imaging and industrial processes screening, spectroscopy and chemical sensing, biomedical diagnosis, and others.1 Compact mass-producible semiconductor sources of THz radiation with room temperature operation, electrical pumping, high power, and wide tunability are highly desired for the development of compact low-cost THz systems. THz difference-frequency generation quantum cascade laser sources (THz DFG-QCLs) are currently the only monolithic electrically pumped mass-producible semiconductor devices that can generate broadly tunable coherent THz emission at room temperature.2 The active regions of THz DFG-QCLs are designed to provide both optical gain for midinfrared (mid-IR) pumps and giant second-order nonlinearity $\chi^{(2)}$ for efficient DFG of THz waves inside of the laser cavity.3,4 Since the DFG process does not rely on population inversion and mid-IR QCLs can operate at room temperature with watt-level optical powers, THz DFG-QCLs also operate at room temperature, unlike traditional THz QCLs.1,2

Significant improvement in the THz output power and tunability of THz DFG-QCLs has been made after the implementation of the Cherenkov emission scheme for THz extraction.3,6 The initial version of the Cherenkov THz DFG-QCLs used a semi-insulating (SI) InP substrate with its refractive index in the 1- to 6-THz range higher than the effective index of the THz nonlinear-polarization wave propagating in the QCL active region. Such configuration makes the DFG-QCL to emit the THz waves generated in the active region into the SI InP substrate that provides significantly lower THz loss than that of the QCL active region. As a result, Cherenkov DFG-QCLs produced THz power output approximately two orders of magnitude above that achievable with edge-emitting DFG-QCLs.2 An automatic phase-matching condition provided by the Cherenkov scheme allowed for demonstrating widely tunable THz DFG-QCL sources with a tuning range of 4.5 THz in an external cavity (EC) setup7 and a tuning range of up to ~2.5 THz in various monolithic configurations.8,9 Recently, the epi-up-mounted continuous-wave Cherenkov DFG-QCL with very low threshold current density (1.3 kA/cm²) has been realized at room temperature as well.10 Continuous-wave THz DFG-QCLs have THz emission linewidth below 1 MHz,11 which makes them suitable for operation as local oscillator sources for heterodyne detectors and for high-resolution spectroscopy.

However, despite no free-carrier absorption, SI InP substrates are not completely transparent for THz frequencies. Detailed investigations into the optical properties of SI InP12 show that strong absorption associated with impurity levels, differential phonon absorption, and tails of optical phonon absorption bands is present, particularly above 2 THz as shown in Fig. 1(a). The SI InP absorption exceeds 15 cm⁻¹ for THz frequencies above 4.3 THz, which implies that for a typical 3-mm long THz DFG-QCL on the SI InP substrates, only about 10% of the THz power produced in the active region can reach the output facet of the device. Additionally, high refractive index dispersion in SI InP implies that the Cherenkov DFG emission from a tunable THz DFG-QCL device will occur at different angles for different THz frequencies.6

In this paper, we demonstrate that the THz power and out-coupling efficiency of Cherenkov THz DFG-QCLs can be dramatically improved by implementing the III-V-on-Si hybrid laser concept to the DFG-QCLs. The improvements are particularly significant for THz frequencies above 3.5 THz. High-resistivity float-zone (HR-FZ) Si has
extremely low loss and nearly constant refractive index in the THz spectral range as shown in Fig. 1(a). Experimentally, we achieve up to 8 times improvement in the mid-IR-to-THz conversion efficiency and up to 5 times improvement in THz output power for a 3.48 THz Cherenkov DFG-QCLs transfer-printed on HR FZ Si. We also demonstrate beam steering free EC tuning of a DFG-QCL on HR-FZ Si in 1.3 to 4.3 THz spectral range. The tuning range is primarily limited by the residual mid-IR reflectivity at the device facets due to imperfection of antireflection coating.

2 Theoretical Description

Cherenkov emission in THz DFG-QCLs occurs when the phase velocity of the THz wave in the device substrate is faster than the phase velocity of the difference-frequency nonlinear-polarization wave in the device active region. The schematic of the Cherenkov DFG emission in a QCL using the slab-waveguide approximation is shown in Fig. 1(b). The propagation angle of the Cherenkov THz emission with respect to the QCL cavity is defined as

$$\theta_c = \cos^{-1}(n_g/n_s),$$

where $n_g$ and $n_s$ are the group indices of mid-IR pumps and the THz refractive index of the substrate, respectively.\textsuperscript{5,6} Since the mid-IR group index in QCLs has a nearly constant value of ~3.37 in the 8- to 12-μm spectral region,\textsuperscript{13} any substrate with a THz refractive index higher than ~3.37 can fulfill the condition of the Cherenkov DFG emission.

Previously, SI InP substrates were used for Cherenkov THz DFG-QCLs since high-performance mid-IR QCLs with InGaAs/AlInAs active regions can be grown on InP. However, SI InP has relatively high THz absorption loss that limits the propagation length of Cherenkov THz emission to ~2 mm and below for the frequencies above 2 THz [see Fig. 1(a)]. SI InP also has a relatively strong THz refractive index dispersion that results in the Cherenkov emission angle $\theta_c$ changing from 22.5 deg at 4.5 THz to 17 deg at 1 THz according to Eq. (1) and the data are shown in Fig. 1(a). The variation of the Cherenkov angle in the substrate will translate to over 15 deg THz beam steering in free space.\textsuperscript{7}

In contrast to SI InP, HR-FZ Si has a nearly constant value of the refractive index and nearly two orders of magnitude lower THz absorption in the THz spectral range as shown in Fig. 1(a). The refractive index of HR-FZ Si is higher than the group index of the mid-IR pumps, which makes Cherenkov DFG emission possible. Figure 2(a) displays the ratio of the THz power outcoupled to free space through the polished substrate facet for the THz DFG-QCL devices on HR-FZ Si and SI InP substrates ($W_{Si}$ and $W_{InP}$, respectively) for different values of SI InP absorption coefficient. The output power is calculated as

$$W = \eta_{ext}I_0 \int_0^{L_{cav}} e^{-\alpha_{sub}x} \cos \theta_c \, dx,$$

where $\eta_{ext}$ and $I_0$ are, respectively, the outcoupling efficiencies of the Cherenkov radiation through the facet (assumed to be 30% for the SI InP substrate and 45% for the HR-FZ Si, see the discussion further below) and the linear power density (W/m) of the THz Cherenkov wave emitted from the laser waveguide into the substrate (the values of $I_0$ are assumed to be the same for the devices on SI InP and HR-FZ Si), $L_{cav}$ is the laser cavity length, and $\alpha_{sub}$ is the absorption coefficient of the substrate. The power output of the SI InP devices stops increasing significantly after the length of the laser cavity exceeds the absorption length of the THz radiation in the SI InP substrate. In contrast, the power output of the hybrid QCL-on-Si increases linearly with the device cavity length in the 1- to 10-mm range, owing to negligibly small THz absorption in HR-FZ Si.

Fig. 1 (a) The refractive indices (left) and optical absorption (right) of HR-FZ Si (solid red) and SI InP (dashed black) as a function of THz frequency. The value of the mid-IR group refractive index ($n_g$) for a typical QCL is shown in dot-dashed blue. (b) Schematic of the Cherenkov DFG in a QCL. The QCL active region is shown in orange, cladding layers are in gray, top metal contact is in yellow, and the SI InP substrate is in black. Solid arrows show Cherenkov THz DFG emission from the active region produced by difference-frequency mixing of the forward-going mid-IR pumps; dashed arrows show Cherenkov THz DFG emission produced by the backward-going mid-IR pumps. The front facet of the substrate is polished to facilitate the outcoupling of the THz radiation to free space.\textsuperscript{5,6}
We note that the expression in Eq. (2) does not include the backward-going Cherenkov wave that is reflected from the back facet of the device [cf. Fig. 1(b)]. This wave is nearly entirely absorbed in devices on the SI InP substrates, but it can reach the front facet without any loss in the devices on HR-FZ Si. If we assume that 100% of the backward-going Cherenkov wave is reflected from the back device facet, the effective length of the device \( L_{\text{eff}} \) in Eq. (2) and Fig. 2(a) is doubled and the advantage of HR-FZ Si substrates is even more significant.

In addition to having lower optical loss, HR-FZ Si offers an additional advantage over SI InP due to its lower value of the THz refractive index. Since the width of a typical THz DFG-QCL waveguide (typ. 10 to 30 \( \mu \)m) is smaller than the THz radiation wavelength, the Cherenkov THz emission is generated in a conical shape as shown schematically in Fig. 2(b). The Cherenkov DFG emission cone angle is \( \sim 20 \) deg in SI InP. Since the critical angle at the substrate facet/air interface is 16 deg, a significant portion of the Cherenkov cone emitting in the lateral direction in SI InP devices cannot be outcoupled to free space, despite substrate polishing. The value of the refractive index in HR-FZ Si is 3.4 and it matches well with the group index \( n_g \approx 3.37 \), resulting in the Cherenkov cone angle of only about 8 deg. As a result, the devices on the HR-FZ Si substrate are expected to have a higher proportion of the incoming THz wave transmitted through the polished facet.

Figure 2(c) shows the results of three-dimensional (3-D) COMSOL simulations obtained for a 200-\( \mu \)m long Cherenkov THz DFG-QCL with a 22-\( \mu \)m wide ridge width on an SI InP substrate. The nonlinear-polarization source in the QCL active region is modeled by setting the TM-polarized dipoles in the laser active region to oscillate at 3.5-THz frequency with the phase relationship corresponding to the mid-IR pumping with forward-propagating waves (direction to the facet). We assume the substrate facet-polishing angle of 20 deg for the device on the SI InP substrate and the substrate polishing angle of 10 deg for the device on the HR-FZ Si substrate in the simulation. The facet outcoupling efficiency in each case is estimated by comparing the incoming THz power onto the substrate facet to the outcoupled THz power on the air screen as shown in Fig. 2(c). Our simulations show that the facet outcoupling efficiency of the device on the SI InP substrate is \( \eta_{\text{ext}} \approx 30\% \) and that of the device on the HR-FZ Si substrate is about \( \eta_{\text{ext}} \approx 45\% \).

### 3 Quantum Cascade Laser Processing and Transfer-Printing on High-Resistivity Float-Zone Si

The laser structure was grown on a 660-\( \mu \)m thick SI InP substrate by a commercial foundry (IQE Inc.). The laser structure is identical to that of devices reported in Refs. 9 and 15. The laser growth started with a 200-nm thick \( \text{In}_{0.53}\text{Ga}_{0.47}\text{As} \) current injection/etch-stop layer followed by a 3-\( \mu \)m thick InP cladding layer and a 200-nm thick \( \text{In}_{0.53}\text{Ga}_{0.47}\text{As} \) waveguide layer. Then, two 25-repetition stacks of \( \text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{In}_{0.52}\text{Al}_{0.48}\text{As} \) active regions, the bottom one designed for the peak gain at 8.5 \( \mu \)m and the top one designed for the gain peak at 9.5 \( \mu \)m, were grown. Both stacks are based on the bound-to-continuum design modified to maximize optical nonlinearity for DFG. The active regions were capped with a 300-nm thick \( \text{In}_{0.53}\text{Ga}_{0.47}\text{As} \) waveguide layer, a 3-\( \mu \)m thick InP cladding layer, a 100-\( \mu \)m thick InP contact layer, and finally a 10-\( \mu \)m thick \( \text{In}_{0.53}\text{Ga}_{0.47}\text{As} \) capping layer.

Devices were processed into distributed feedback (DFB) lasers with the side current injection scheme following conventional dry-etching ridge waveguide fabrication steps. Two sections of the first-order surface gratings with the grating pitches for selecting the two mid-IR pump frequencies were formed using e-beam lithography and dry etching. The grating pitches for the 9.0-\( \mu \)m short-wavelength \( (\lambda_{\text{short}}) \) and the 10.1-\( \mu \)m long-wavelength \( (\lambda_{\text{long}}) \) pumps were 1.408 and 1.584 \( \mu \)m, respectively. The completely processed wafer die was cleaved into two sections, one section for the characterization of the reference devices on the SI InP substrate and the other second section for transfer on a 1-mm thick HR-FZ Si wafer.

The second section of the completely processed QCL wafer was transfer-printed on HR-FZ Si using the following steps described in more detail in Ref. 15. The wafer was attached to a glass slide with the laser ridges facing the glass using a crystal glue (Crystalbond 509). The InP substrate was selectively removed using an HCl-based wet etchant, with the InGaAs current injection layer used as an etch stop. The exposed surface of the current injection layer was
then bonded to the HR-FZ Si wafer using an SU-8 photore sist as adhesive. To achieve a thinner than standard SU-8 thickness, we diluted SU-8 2000.5 (MicroChem Corp.) photore sist in the SU-8 thinner (MicroChem Corp.) in the proportion of 1 to 2. The mixture produces a 100-nm thick film when spin-coated at 3000 rpm on an Si wafer. Bonding was performed in the Applied Microengineering, Ltd. wafer bonding machine at a pressure of ~1 MPa and a temperature of 180°C for 15 min. Finally, the glass slide was removed by dissolving the crystal glue with acetone, and the hybrid QCL-on-Si wafer was diced into laser bars using a dicing saw. The front facets of the Si-hybrid and reference InP devices were polished at 15 deg and 30 deg, respectively, to outcouple THz Cherenkov emission in the direction approximately parallel to the laser cavity.

Figure 3 shows the scanning electron microscope images of the facets of the devices on SI InP and HR-FZ Si substrates. The somewhat roughened surface of the facet of the QCL-on-Si originates from the dicing process (devices on SI InP were cleaved). The thickness of the SU-8 bonding layer is estimated to be about 100 nm as shown in Fig. 3(b). According to our simulations, a 100-nm thick SU-8 layer has about 90% transmission of the THz Cherenkov radiation into the Si substrate. Devices were indium soldered to copper blocks in the episide-up configuration and wire bonded for testing.

4 Experimental Results

The mid-IR power output of the lasers was measured using a thermopile detector with a metal pipe closely adjacent to the laser output facet to maximize the collection efficiency. The THz power output was collected using two 2-in. diameter 90-deg off-axis parabolic mirrors in a nitrogen-purged environment. The first parabolic mirror had a focal length of 2 in. and was used to collimate the THz DFG-QCL emission; the second parabolic mirror had a focal length of 4 in. and was used to focus the THz output onto a calibrated liquid helium-cooled Si bolometer. The mid-IR power measurement setup was estimated to have nearly 100% collection efficiency, whereas the collection efficiency of the THz setup was estimated to be below 50%, based on the theoretical analysis of the far-field distribution of THz emission from the Cherenkov THz DFG-QCL devices. The mid-IR and THz power output data shown in this paper were not corrected for the collection efficiency. A longpass interference filter with the cut-on frequency of 1000 cm\(^{-1}\) was inserted in the small gap between the pipe and the detector to separate the two mid-IR pump powers. Spectra were measured using a Fourier transform infrared spectrometer equipped with a deuterated triglycerine sulfate detector (for mid-IR measurements) or an Si bolometer (for THz measurements). Devices were driven in pulsed mode using 40-ns current pulses at 15-kHz repetition frequency at 20°C of a heat sink temperature.

Figure 4(a) displays the mid-IR and THz spectra measured from the Si-hybrid device. Stable dual single-mode mid-IR pump operation was obtained at the peak wavelengths of 994 and 1110 cm\(^{-1}\). The peak positions of the mid-IR pumps were identical for devices on Si and InP. THz emission frequency of both devices is 3.48 THz in agreement with the frequency separation of the mid-IR pumps. To compare the performance of the InP and Si devices, best-performing devices of each type with the identical dimensions (22-μm wide and 4.2-mm long cavity length) were selected. For the operating conditions employed in our measurement setup, the mid-IR pumps power outputs of the Si-transferred devices were only slightly lower compared to that of the devices on their native SI InP substrates as shown in Fig. 4(b). THz peak power and the mid-IR-to-THz conversion efficiency (defined as the ratio of the measured THz peak power output to the product of the two measured peak powers of the mid-IR pumps) of both devices are plotted in Fig. 4(c). The maximum THz power of the reference SI InP devices was about 50 μW and the mid-IR-to-THz conversion efficiency was about 60 μW/W\(^2\) at the maximum THz power output. The best-performing Si-hybrid device reached THz power output of about 270 μW with the conversion efficiency of 480 μW/W\(^2\) at the maximum THz power output. Data indicate a factor of five improvements in THz power and a factor of eight improvements in the conversion efficiency for the devices transferred on HR-FZ Si.

Tunability of a 2-mm long, 22-μm wide Si-hybrid device has been tested in a modified Littrow EC setup described in Ref. 7. The device was diced so that only one DFB section (fixing the pump wavelength at 995 cm\(^{-1}\)) was present. The back facet of the device was coated with an antireflection coating. The other mid-IR pump position was tuned by an EC diffraction grating. The Si substrate of the device was polished with an angle of 15 deg to direct the THz output beam approximately parallel to the laser cavity. Figure 5(a) shows the spectra of the mid-IR pumps selected by the DFB grating and EC diffraction grating at a bias current of 5.2 A, near the rollover point. The wavelength of the EC mode was tuned from 1045 to 1145 cm\(^{-1}\). The peak power outputs of the two pumps as a function of the EC grating position are also plotted in Fig. 5(a).

Figure 5(b) shows the THz performance of the EC device at the same 5.2 A bias current. We observe that there is a
noticeable difference between THz power output versus THz frequency of the Si EC device reported here and that of the SI-InP EC devices with similar active regions reported previously. While the maximum THz power output of the THz DFG-QCLs on SI InP is attained in the 3.3- to 3.8-THz range, the THz peak output power of THz DFG-QCLs on Si increases monotonically up to the maximum THz DFG frequency of 4.3 THz attained in our setup. This trend is in agreement with the $\omega^2$ dependence of THz DFG efficiency, assuming constant values of $\chi^{(2)}$ and neglecting frequency-dependence of the losses and phase-matching conditions. The reason for the drop in the THz power output of the Cherenkov THz DFG-QCL devices on SI InP beyond 3.8 THz is the onset of strong optical absorption in SI InP as shown in Fig. 1(a). We note that a more detailed analysis of the device performance should include the analysis of the frequency dependence of $\chi^{(2)}$, waveguide losses, and phase-matching conditions. This analysis, however, is beyond the scope of this report.

THz tuning from 1.3 to 4.3 THz was obtained. The tuning range was limited principally by the quality of the antireflection coating and the bandwidth of the mid-IR gain spectrum. We note that the EC tuning range of nearly 1 to 6 THz has been previously demonstrated with other Cherenkov THz DFG-QCL devices. The rough facet surface of the Si devices [see Fig. 3(b)] makes it more challenging to produce high-quality antireflection coatings and achieving high-efficiency EC feedback. This problem is also responsible for the multimode tuning spectra in Fig. 5. We expect that an improved device processing/dicing and an optimized facet antireflection coating will improve the spectral quality and the tuning bandwidth of Cherenkov THz DFG-QCLs on Si to match that of Cherenkov THz DFG-QCL devices on SI InP. The THz power output at 4.3 THz of the Si-hybrid device exceeded 0.5 mW—a record power for all EC THz DFG-QCL sources—that corresponds to the conversion efficiency of 0.55 mW/W². The EC device was 2.1 times shorter than the DFB device reported in Fig. 4 and the power output of the Cherenkov THz DFG-QCL sources on HR-FZ Si is expected to grow linearly with the device length because of very small THz absorption in HR-FZ Si. This point can also be supported as we experimentally observe nearly twice as high power in twice longer device by comparing the 2-mm long EC device [~120 µW in Fig. 5(b)] and the 4.2-mm long DFB device [~270 µW in Fig. 4(c)] at the same THz frequency of 3.48 THz. Thus, even higher output powers are expected for longer devices in the EC setup.
Since the THz refractive index of HR-FZ Si is nearly constant, the tunable Cherenkov THz DFG-QCL on Si avoids strong THz beam steering present in THz DFG-QCLs on SI InP [cf. Eq. (1)]. This has been confirmed by measuring the far field of the THz emission from EC HR-FZ Si devices. Figure 6 displays the far-field profiles in the direction perpendicular to the device plane of an EC THz DFG-QCL on HR-FZ Si. Data were measured at different THz output frequencies. The peak far-field intensities are maintained at nearly the same direction of ~10 deg with a full width at half maximum of 12 deg.

5 Conclusion
We have demonstrated that the use of HR-FZ Si substrates instead of SI InP substrates results in a dramatic improvement in the THz power output and the mid-IR-to-THz conversion efficiency in Cherenkov THz DFG-QCLs. The 22-μm width and 4.2-mm long ridge-waveguide THz DFG-QCLs transfer-printed on HR-FZ Si produced the peak THz power output of 270 μW with the mid-IR-to-THz conversion efficiency of 480 μW/W² at 3.48 THz, whereas the reference SI InP device with the same dimensions and operating frequency produced 50 μW with the conversion efficiency of 60 μW/W² at the maximum bias current. Even higher power output improvements are expected at higher THz frequencies owing to the presence of strong THz absorption in SI InP at frequencies above ~3.5 THz. We also demonstrate broadband spectral tuning of the THz DFG-QCL on silicon with virtually no beam steering of the THz emission.

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References

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