Terahertz plasmonic laser radiating in an ultra-narrow beam: supplementary material

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1. MATERIALS AND METHODS

Fabrication and measurement details

The active-medium of the QCLs is based on a three-well resonant-phonon design with GaAs/Al_{0.10}Ga_{0.90}As superlattice (design RTRP3W197, wafer number VB0464), which is described in Ref. [1], and was grown by molecular-beam epitaxy. The QCL superlattice is 10 μm thick with an average n -doping of $5.5 \times 10^{15} \text{ cm}^{-3}$, and surrounded by 0.05 μm and 0.1 μm thick highly-doped GaAs contact layers at $5 \times 10^{18} \text{ cm}^{-3}$ on either side of the superlattice. Fabrication of QCLs with parallel-plate metallic cavities followed a Cu-Cu thermocompression wafer bonding technique as in Ref. [2] with standard optical contact lithography. Following wafer-bonding and substrate removal, positive-resist lithography was used to selectively etch away the 0.1 μm thick highly-doped GaAs layer from all locations where top-metal cladding would exist on individual cavities except in $\sim 5 \mu\text{m}$ wide regions at the outer rectangular boundaries of the top-metal layer due to overlapping mask layers. The removal of this layer beneath the top-metal does not impact electrical transport significantly except adding a small voltage drop at the top contact during QCL operation. Importantly, this lithography step allows radiative outcoupling from the apertures in the finally fabricated QCL cavities; and simultaneously serves to implement longitudinal and lateral absorbing boundaries as illustrated in Fig. 3(a) by leaving the highly-doped GaAs layer exposed at both longitudinal and lateral edges of the QCL cavity. The absorbing boundaries result in a highly lossy propagation of the SPP modes in those regions [3]. While the longitudinal boundaries help in DFB mode discrimination, the lateral boundaries are useful to eliminate higher-order lateral guided modes by making them more lossy in comparison to the fundamental mode [2, 4, 5].

Ti/Au metal layers of thickness 25/200 nm were used as the top metal cladding, using an image-reversal lithography mask for implementing gratings in the metal layers. Another positive-resist lithography step was used to cover the grating-metal with photoresist to be used as a mask for wet-etching of ridges in a H₂SO₄:H₂O₂:H₂O 1:8:80 solution for ~ 22 minutes. After etching the 10 μm thick superlattice active region when the bottom metal layer is exposed, an over-etch of ~ 1 minute was followed up to reduce the slope on the sidewalls of the QCL's cavities to allow for a more uniform current-density distribution through the height of the cavity (in y direction). A Ti/Au contact was used as the backside-metal contact for the finally fabricated QCL chips to assist in soldering. Before deposition of backside-metal of the wafer, the substrate was mechanically polished down to a thickness of $\sim 170 \mu\text{m}$ to improve heat-sinking.

For measurements, a cleaved chip consisting of QCLs with antenna-feedback gratings of different periods along with some Fabry-Pérot ridge QCLs and third-order DFB QCLs was In-soldered on a Cu block, the QCL to be tested was wire-bonded for electrical biasing, and the Cu block was mounted on the cold-stage of a liquid Nitrogen vacuum cryostat for measurements. Initial measurements of the fabricated QCL cavities did not yield lasing devices at 78 K. It was then realized that the $\sim 13 \mu\text{m}$ wide lateral absorbing boundaries on each side were perhaps introducing high optical losses in the cavities. The finally fabricated wafers were then etched again (post-fabrication) in a H₂SO₄:H₂O₂:H₂O 1:8:80 solution for ~ 15 seconds, which is likely to etch away most of the exposed highly-doped GaAs layer. However, $\sim 5 \mu\text{m}$ wide highly-doped GaAs layer remains unetched underneath the top-metal cladding on all four outer boundaries of the cladding, which serves to perform the role of an absorbing region. The post-etched devices lased upon re-testing, and their robust single-mode operation suggests that

Fig. S1. Full-wave 3D simulation to compute far-field radiation pattern of QCL cavities with antenna-feedback. (a) The modeled geometry for full-wave 3D simulation with the FEM solver [6]. Parallel-plate metallic terahertz QCL cavities of 1.4 mm length, 10 μm thickness, varying widths, and a grating period of $\Lambda = 21.7 \mu\text{m}$ were implemented, which excite antenna-feedback resonant-cavity modes at $\nu \sim 3.1 \text{ THz}$. (b) Simulated far-field radiation pattern of the cavity with 100 μm width. The FWHM is $\sim 7^\circ \times 7^\circ$. (c) Far-field radiation pattern of the cavity with 70 μm width. The FWHM is $\sim 6^\circ \times 5^\circ$.

both lateral and longitudinal absorbing boundaries were able to suppress undesired resonant-modes in the cavity as desired.

The emitted optical power was measured with a deuterated triglycine sulfate pyroelectric detector (DTGS) pyroelectric detector and calibrated with a terahertz thermopile powermeter (ScienTech model AC2500H) without any optical component or cone collecting optic inside the cryostat to improve collection of radiated power in the case of DFB QCLs. (A Winston cone was used to collect power, but only for the Fabry-Pérot-cavity QCL reported in Fig. S3.) The spectra were measured at 78 K in linear-scan mode with a resolution of 0.2 cm^{-1} (6 GHz) using a Bruker Fourier-transform infrared spectrometer (FTIR) under vacuum, with a room-temperature DTGS detector placed inside the FTIR. Far-field radiation patterns were measured with a DTGS pyroelectric cell detector mounted on a computer-controlled two-axis ($x - y$ plane) moving stage in the end-fire (z) direction at a distance of 65 mm from the QCLs' end-facets. The detector's size leads to spatial averaging of the measured beam with a step-size of 1.7° , which has a negligible impact on the measured beam-width.

Modeling

Finite-element (FEM) simulations were performed using a commercially available software [6]. The eigenmodes of the DFB cavities are computed using an eigenfrequency solver. The loss in absorbing boundaries of the cavity is implemented using a complex dielectric constant computed using Drude-model [2]. The imaginary part of the computed eigenfrequencies is used to compute the propagation loss of the resonant-cavity modes in the units of inverse of length. The active region is modeled as lossless and the metal is modeled to be perfect electrical conductors, in which case, the computed loss is the sum of loss at absorbing boundaries as well as that due to radiation (outcoupling). The thickness of the metallic layer is 400 nm in simulation. The relative losses of various resonant modes for the DFB cavities are not changed and neither are the mode-shapes and the corresponding resonant frequencies when using lossy metal, or when the thickness of the metallic film are altered (as long as it is kept significantly thinner than that of the active region). For the 2D simulations (cavities of infinite-width) the far-field boundaries are modeled as absorbing layers as described in Ref. [2]. For 3D simulations, the in-built perfectly matched layers in the FEM solver were utilized to model the far-field boundaries.

2. FULL-WAVE 3D SIMULATION FOR RADIATION-PATTERNS

The narrow-beam emission from plasmonic lasers with antenna-feedback is due to a combination of two factors that are related to the radiative behavior of phased-array antennas. In addition to the *array-factor* that leads to narrower beams for more number of elements in a phased-array (i. e. in this case, a longer length of the spaser's cavity), the far-field radiation pattern is additionally narrowed owing to an *element-factor*, which is defined as the far-field radiation pattern due to an individual emitter of the phased-array [7]. The large spatial extent of the single-sided SPP wave in the surrounding medium results in a narrower element-factor compared to an omnidirectional point-source emitter.

As measured experimentally and shown in Fig. 4, the antenna-feedback terahertz QCL cavities with a narrower width of 70 μm resulted in a beam with smaller divergence as compared to the 100 μm wide cavity. This seemingly unique behavior from antenna-feedback QCLs serves to further validate the concept of the specific feedback scheme, in which the cavity radiates like a phased-array antenna. A narrower cavity causes greater lateral spread of the single-sided SPP mode (in the x dimension) in the surrounding medium, especially when the cavity's width is sub-wavelength. A single-sided SPP mode with a broader cross-section in the $x - y$ plane will lead to a more directional far-field radiation pattern owing to a narrower element-factor for the phased-array antenna structure.

To further validate the experimental results of Fig. 4, full-wave 3D FEM simulations were carried out for terahertz QCL cavities with antenna-feedback. Figure S1 shows the computed far-field radiation patterns for band-edge DFB mode for terahertz QCL cavities implemented with antenna-feedback gratings in the top metal cladding. The in-built perfectly-matched layers (PMLs) in the FEM solver [6] serve as an effective absorbing boundary for computation of the radiation pattern. The PMLs' parameters are coarsely adjusted to achieve minimum reflection at terahertz frequencies and transform the propagating waves to exponentially decaying waves. The PMLs are placed far enough to avoid interaction with the single-side SPP standing-wave in the near-field of the cavity. Considering the computer's memory limitations, laterally absorbing boundaries are not modeled in the cavity, which does not impact the simulation's results since the DFB mode could always be found, even though it is not the

lowest loss mode in the eigenmode spectrum of the cavity due to other higher order lateral modes that can have even lower propagation loss. All metal layers are modeled as perfectly conducting surfaces to limit the mesh size in the geometry. In order to set up a far-field calculation, a far-field domain node is implemented that is a single closed surface surrounding all radiating apertures in the cavity. The distribution of far EM field is based on the Fourier transform of the near-field as implemented in the FEM solver. Simulated 3D beam patterns demonstrate single-lobed beams with narrow divergence for the band-edge mode with antenna-feedback as shown in the figure. The shape and FWHM of the computed radiation-pattern is in close agreement with the experimentally measured beams that are shown in Fig. 4. The FWHM of simulated beam patterns is slightly larger than that of the measured results. The difference are likely due to the relative simplification of implemented 3D model, which does not account for the sloped sidewall profile of the ridges and also the fact the ground plane around the QCL's cavity is different for the measured chip as compared to that in the simulated geometry.

Fig. S2. Lasing characteristics of a terahertz QCL with third-order DFB without phase matching. Light-current characteristics of a terahertz QCL with third-order DFB in pulsed operation. The QCL is fabricated on the same chip, and the cavity dimensions are similar to the QCLs with antenna-feedback for which data is presented in Fig. 3. The inset shows the measured far-field radiation-pattern with the same angular definitions as in Fig. 4(a). The QCL emitted predominantly in single-mode at $\lambda = 103.0 \mu\text{m}$ ($\nu = 2.91 \text{ THz}$) with grating period $44 \mu\text{m}$.

3. EXPERIMENTAL CHARACTERISTICS OF THIRD-ORDER DFB QCL AND A FABRY-PÉROT-CAVITY QCL

Figure S2 shows L - I characteristics of a representative terahertz QCL with third-order DFB without well defined phase-matching condition that was fabricated on the same chip by standard lithography to pattern grating on top metallic layer, and with similar dimensions as that of the antenna-feedback QCLs whose data is reported in Fig. 3. Using the phase condition $\lambda = 2n_a\Lambda/p$ as determined from equation 1 for $p = 3$ (third-order DFB), an effective-mode index of $n_a = 3.51$ is estimated for the resonant-cavity SPP mode in the active medium of the cavity. For ideal phase-matching, a mode-index value of 3.0 is required, hence the phased-matching condition is not satisfied for this QCL. Conse-

Fig. S3. Lasing characteristics of a terahertz QCL with Fabry-Pérot cavity. Light-current characteristics of a QCL with Fabry-Pérot cavity at different heat-sink temperatures in pulsed operation. Output power was collected with a Winston cone placed close to laser facet due to the highly divergent beam pattern of Fabry-Pérot type THz QCLs. The inset shows lasing spectra at different bias at $\sim 78 \text{ K}$.

quently, the measured FWHM of the far-field radiation pattern $\sim 12^\circ \times 16^\circ$ for this QCL is not as good as that for phased-matched third-order DFB QCLs with reduced effective mode index [5, 8, 9]. Elongated and multi-lobed beam pattern for third-order DFB when refractive index is close to 3.6 is observed in Ref. [5]. The peak output power for the third-order DFB QCL without phase matching is $\sim 0.45 \text{ mW}$ at 78 K , which is about three times less than that obtained for the antenna-feedback QCL with similar dimensions lasing at similar frequency. The output power slope efficiencies at 78 K are $4 \text{ mW}/\text{A}$ and $13 \text{ mW}/\text{A}$ respectively for the third-order DFB QCL and antenna-feedback QCL respectively. The optical power was measured without cone collecting optics. The relative differences in the measured optical power are in good agreement with the radiative losses estimated via 2D FEM simulations for similar cavity dimensions as in Fig. 2. The maximum operating temperature of this QCL is almost similar to that of the antenna-feedback QCL, which suggests that radiative loss is relatively a small fraction of the overall waveguide loss in both types of DFB QCL cavities. This also suggests that there is a scope for enhancing radiative efficiency to increase optical power output without causing a significant degradation in temperature performance of terahertz QCLs with antenna-feedback. Calculation of surface-outcoupling efficiency as in Ref. [10] is outside the scope of this work due to large uncertainty in the absolute loss contribution of metal layers, absorbing boundaries, as well as the active region itself. The optimized phase-matched third-order DFB [9] with bound-to-continuum design shows lower current-density and higher efficiency without absorbing regions. Since the feedback of laser mode is achieved by SPP mode propagating on the surface for antenna-feedback, technique of covering waveguide sidewall with metal [2] or lateral corrugated grating [9], which does not absorb effective outcoupled laser radiation from antenna-feedback, can replace lateral absorber layer here and will further enhance the outcoupling efficiency. L - I characteristics at different heat-sink temperature of a representative terahertz QCL with Fabry-Pérot (FP) cavity is shown in Fig. S3. Exposed highly-doped lossy contact layer serving as absorber introduces $\sim 15 \text{ K}$ temperature degradation for DFB cavity. Be-

cause sub-wavelength mode confinement in FP cavities results in highly divergent output beams of THz QCLs with parallel-plate metallic cavities, output power is measured with a Winston cone and the detector was placed adjacent to the cryostat window. Diameter of the circular opening of cone is ~ 1.9 mm. Frequency of gain medium covers ~ 2.8 THz to ~ 3.2 THz at ~ 78 K.

4. ANALYSIS OF ANTENNA-FEEDBACK SCHEME BY 2D AND 3D SIMULATIONS

Fig. S4. Energy-density profiles along the length of the metallic cavities (in z direction) for the band-edge DFB modes for (a) third-order DFB cavity, and (b) cavity with antenna-feedback respectively.

Figure S4 shows energy-density profile for the resonant band-edge modes of the cavities with third-order DFB and antenna-feedback schemes respectively, along the entire length of the cavity (~ 1.4 mm). Energy-density is calculated and summed up along the cavity's height ($10 \mu\text{m}$) from 2D simulations (that effectively model cavities of infinite width). The eigenmode spectra and electric-field distributions for the cavities are shown in Fig. 2. Both third-order DFB and antenna-feedback schemes show non-uniform envelope shapes, which gradually decrease from the center to the end-facets of cavity and provide indication of typical DFB action due to coupling of propagating waves along the length of cavity as per the corresponding schematics in Fig. 1.

A thin highly-doped GaAs layer below the top-metal cladding was left unetched at the regions close to longitudinal facets, serving as the longitudinal absorbing boundary to ensure the excitation of the desired antenna-feedback mode as the lowest-loss lasing mode. Firstly, such absorbing boundaries reduce the end-facet reflectivities that strengthens the distributed-feedback coupling for the antenna-feedback scheme. Secondly, the longitudinal absorber boundaries play an important role in providing the necessary mechanism for mode-discrimination,

Fig. S5. Role of longitudinal absorbing boundaries in resonant-cavities with antenna-feedback. (a) Eigenmode spectrum of QCL metallic cavity with antenna-feedback gratings in the top metallic layer, but without longitudinal absorbing boundaries at the two ends of the cavity. The dimensions of the cavity are otherwise similar to that for the result in Fig. 2(b). (b) Energy density profile along the length of the ~ 1.4 mm long cavity and dominant electric field E_y of a Fabry-Pérot cavity type mode that exists sufficiently away from the photonic bandgap. (c) Energy density profile and electric-field profile along the length of the cavity for the “desired” antenna-feedback mode that exists at the edge of the photonic bandgap. The z direction is along the length of laser ridge, as marked in Fig. S4.

to selectively excite the desired band-edge antenna-feedback mode for lasing. Figure S5(a) shows that eigenmode spectrum of antenna-feedback scheme without longitudinal absorbing boundaries. There exist the Fabry-Pérot type modes with uniform energy density profile along the whole length of laser cavity and large electric field distribution close to end facets of the cavity, as shown in Fig. S5(b). However, the loss of such modes is small due to highly reflective end-facets. The desired antenna-feedback mode, located at the lower bandedge, shows non-uniform energy-density profile along the length of cavities as an indicator of DFB action and strong coupling between plasmonic mode on top of metallic layer and guided mode in the active core, as seen from Fig. S5(c). However, its radiative loss is higher due to the improved and large radiative field of antenna-feedback mode and hence, it cannot be excited for lasing in the absence of longitudinal absorbing boundaries. When longitudinal absorbing boundaries are introduced, the relative loss of the modes with large intensity closer to longitudinal ends of cavity, which are further away from the photonic bandgap, will be selectively and significantly increased. Therefore, the antenna-feedback mode is excited for lasing as the robust lowest-loss mode as seen from the eigenmode spectrum in Fig. 2(b).

Figure S6 shows lateral mode shape of the antenna-feedback mode at approximately the longitudinal center of the cavity from 3D simulation. The SPP mode in surrounding medium extends in both lateral and vertical directions significantly, and the extent of the mode in lateral (x) dimension is greater than the width of the cavity itself. The FWHM of the mode shape along lateral (x) dimension of cavity at a distance of $\sim 10 \mu\text{m}$ above top gratings (picked arbitrarily to keep it slightly away

Fig. S6. Electric-field profile of the SPP mode in surrounding medium of the plasmonic laser's cavity with antenna-feedback gratings from 3D simulation. A parallel-plate metal cavity with GaAs medium of $100\ \mu\text{m}$ width, $10\ \mu\text{m}$ thickness, and $1.4\ \text{mm}$ length is simulated as in Fig. S1. The frequency of the band-edge resonant-cavity mode is $\sim 3.15\ \text{THz}$, which is the desired “antenna-feedback” mode. Inset: Mode shape along lateral x dimension of cavity at a distance of $10\ \mu\text{m}$ above the top metal cladding; the FWHM of the lateral electric-field profile is $\sim 130\ \mu\text{m}$.

from location where finite-element mesh in simulation changes abruptly) is $\sim 130\ \mu\text{m}$. This large effective wavefront created on top of the waveguide leads to a narrow far-field, i.e. a narrow “element-factor” in terminology of the radiation from a phased-array antenna. While the narrowing of “array-factor” due to constructive interference of radiation from all apertures in end-fire direction is also at play; it is not the dominant effect to cause narrow beaming for terahertz QCLs in this work for short-length cavities ($< 2\ \text{mm}$). In contrast, for third-order DFB QCLs with phase-matching, beam narrowing is primarily due to the narrow array-factor only, which is why very long cavities ($< 5\ \text{mm}$) are needed to achieve beam divergence values of less than $\sim 10^\circ$ [8].

Fig. S7. Comparison of electric-field squared modulus $|E|^2$ profile in conventional DFB (third-order DFB as an example) and antenna-feedback scheme. (a) $|E|^2$ profile for third-order DFB cavity plotted along perpendicular y direction (cavity's height direction). (b) $|E|^2$ profile for antenna-feedback scheme with an expanded view close to top metal cladding.

Figure S7 shows the electric-field squared modulus $|E|^2$ profile inside the cavity and above the metal cladding for both conventional DFB and antenna-feedback scheme from 2D simulations. Layer sequence follows active region with $10\ \mu\text{m}$ thickness, top metal layer with $400\ \text{nm}$ thickness (lossless metal) and air region. At the location above metal cladding, the mode-intensity in conventional DFB immediately decays to zero. In contrast, for antenna-feedback scheme, the sub-wavelength plasmonic resonant-optical mode inside the cavity is phased-locked with a SPP mode that travels on top of the metal cladding out-

side the cavity. The SPP wave is relatively tightly bound to the top metal cladding and decays in the length of the order of the wavelength in vacuum. The decay length is independent of the loss in metal or thickness in metal or various duty cycles of grating (which was verified via finite-element simulations). The establishment of a hybrid SPP mode on top of the cavity in vacuum is unique in antenna-feedback scheme. In a third-order DFB terahertz QCL, even with effective mode index getting close to 3.0, the feedback mechanism remains same as that in conventional DFB lasers, i.e. distributed-feedback couples propagating modes inside the cavity without involving the propagating SPP waves in the surrounding medium. Correspondingly, no standing SPP wave (with large amplitude) is established in the surrounding medium in a third-order DFB QCL's cavity as it does in antenna-feedback scheme. This could be verified by simulating a terahertz QCL cavity structure with third-order DFB gratings, but with the refractive index of the active region arbitrarily set to a value of ~ 3.0 to satisfy the so-called “phase-matching” condition in simulation.

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