## **Supplementary Information**

## Terahertz semiconductor laser chaos

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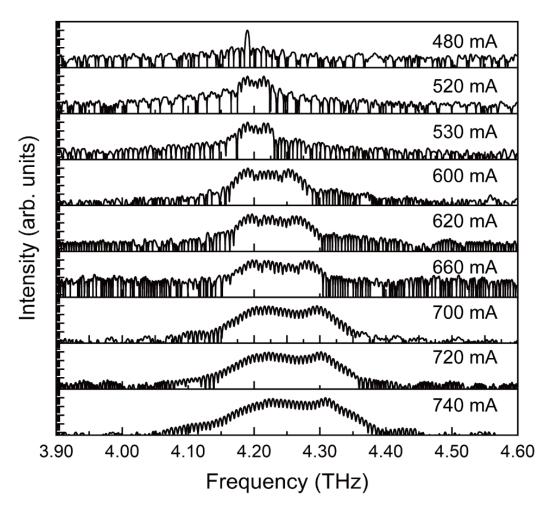
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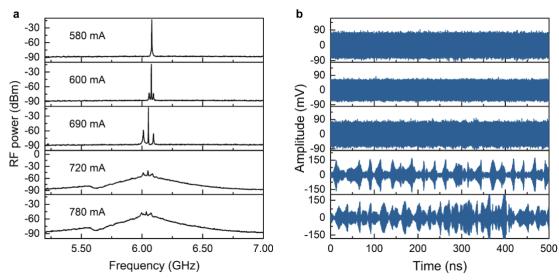
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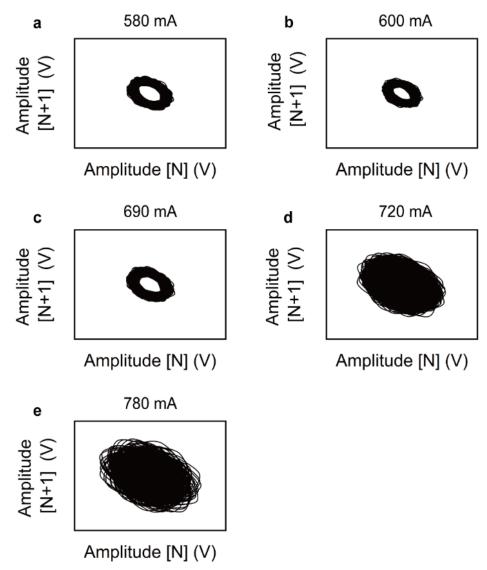
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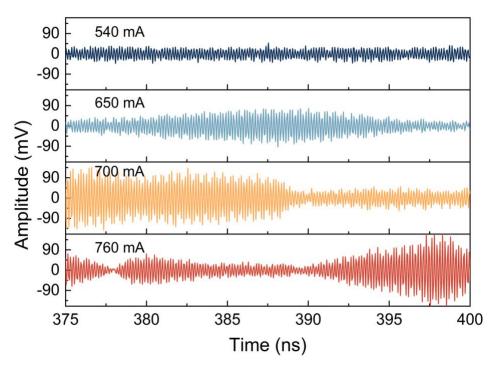
**Fig. S1.** Terahertz emission spectra at various currents measured using a Fourier-transform infrared (FTIR) spectrometer (Bruker, v80) with a spectral resolution of 3 GHz. To reduce the water absorption loss as much as possible, the FTIR chamber is pumped down to 3 mbar and the optical beam path outside the FTIR is purged with dry air. The terahertz light is coupled into the FTIR chamber using a parabolic mirror.



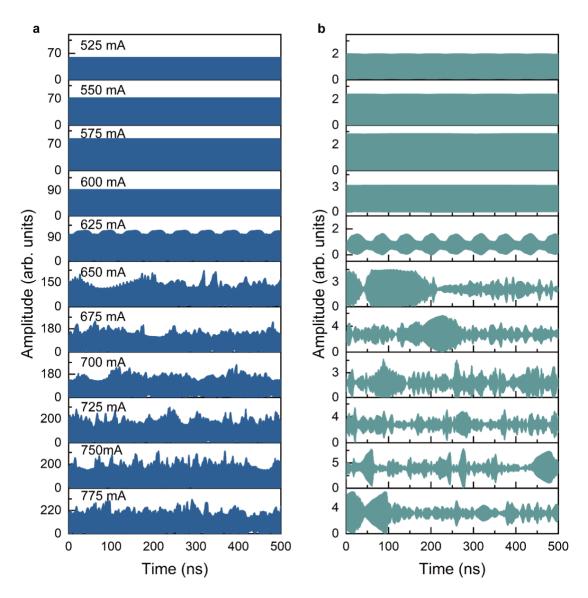
**Fig. S2.** (a), Electrical spectra at different currents, measured with a RBW of 10 kHz and a VBW of 1 kHz. (b), Evolution of corresponding time traces. As the current is increased from 580 to 780 mA, inter-mode BN spectra show clear transitions from single narrow peak, multi-peak and broad peak. At 580 mA, the time trace exhibits a sinusoidal period time series. As the current is increased to 600 and 690 mA, a modulation on the sinusoidal time trace is not clearly observed. The reason is that the signal-to-noise ratio of the sidebands is too low. When we further increase the current to 720 or 780 mA, typical chaotic time traces without any accurate and predictable periodicity are observed showing the laser enters the chaotic regime.



**Fig. S3.** Phase portraits at various currents with times series of 500 ns. A periodic system normally shows an inner circle in its two-dimensional phase portrait at 580, 600 and 690 mA. The phase portraits become more complex at 720 and 780 mA, and much stronger oscillations result in larger trajectories in phase portraits.



**Fig. S4.** Evolution of experimental time traces with a length of 25 ns at various currents.



**Fig. S5.** Simulated results. Due to the high sampling rate in numerical calculation, the minimum time interval is 0.002 ns. To avoid the influence of higher harmonics on calculated results, we use a digital filtering method to obtain pure BN signal. (a), Time series without filtering and (b), after filtering at various currents.

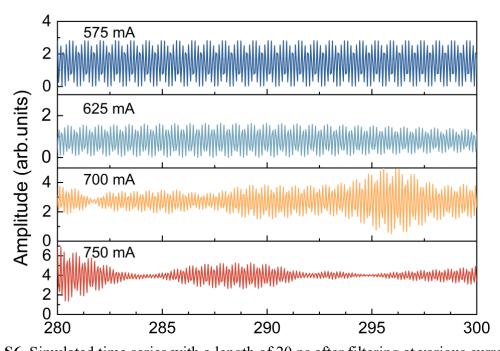
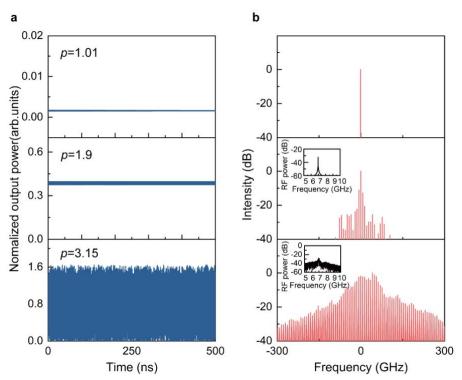


Fig. S6. Simulated time series with a length of 20 ns after filtering at various currents.



**Fig. S7.** Role of current p parameter to QCL's behaviors at  $\alpha$ =1.15 and k"=0. (a), Time series at p=1.01, 1.9, 3.15. (b), Corresponding optical spectra. The insets in b are inter-mode BN spectra. The simulated results highlight the current parameter as a critical determinant of QCL stability. When the p parameter is set to 1.01, a single-mode emission spectrum and stable time series are observed. When the p parameter is set to 1.9, the optical frequency combs emission is observed. And the QCL enters chaotic regimes when the p parameter is set to 3.15.

Table S1 Parameters used in the full model

Parameter	Quantity
Cavity length	6 mm
Background refractive index of the active medium	3.6
Waveguide losses	3.5 cm <sup>-1</sup>
Volume of the active medium	100µm*6mm*10µm
Reflectivity of the QCL facets	0.32
Number of stages of the cascading scheme	76
Central emission frequency	4.2 THz
$\delta_{hom}$	500 GHz
Carrier lifetime	50 ps
α factor	0.1
Confinement factor of the field in the active medium	0.25
Threshold current	390 mA
$f_0$	4.5*10 <sup>-6</sup> µm <sup>3</sup>

Table S2 Parameters used in the reduced model

Parameter	Quantity
Cavity length	6 mm
Background refractive index of the active medium	3.6
Waveguide losses	3.5 cm <sup>-1</sup>
Reflectivity of the QCL facets	0.32
$\delta_{hom}$	500 GHz
α factor	various