The generation of stable trains of ultrashort (femtosecond to picosecond), terahertz-frequency radiation pulses with large instantaneous intensities is an underlying requirement for the investigation of light–matter interactions for metrology and ultrahigh-speed communications. In solid-state electrically pumped lasers, the primary route to generate short pulses is through passive mode-locking; however, this has not yet been achieved in the terahertz range, defining one of the longest standing goals over the past two decades. In fact, the realization of passive mode-locking has long been assumed to be inherently hindered by the fast recovery times associated with the intersubband gain of terahertz lasers. Here we demonstrate a self-starting miniaturized short pulse terahertz laser, exploiting an original device architecture that includes the surface patterning of multilayer-graphene saturable absorbers distributed along the entire cavity of a double-metal semiconductor 2.30–3.55 THz wire laser. Self-starting pulsed emission with 4.0-ps-long pulses is demonstrated in a compact, all-electronic, all-passive and inexpensive configuration.
set-ups such as free electron lasers, hindering their use beyond basic research.

Quantum cascade lasers (QCLs) are the only miniaturized direct sources of laser radiation in the terahertz frequency range, combining chip-scale size, high (>1 W) power emission, high spectral purity and broad bandwidth. Although room-temperature operation is yet to be achieved, Fabry–Perot terahertz QCLs can operate in a compact Peltier cooler configuration up to 261 K (ref. 19) and generate trains of short pulses by active mode-locking, that is, by modulating the gain and losses in the active medium with a bias current modulation. Active techniques are favoured by the QCL ultrafast carrier dynamics, inherent to the intersubband transitions in the gain medium. The gain in a QCL recovers from its saturated value much faster than the cavity round-trip time (72 ps for a standard 3-mm-long cavity) and the photon lifetime, resulting in gain recovery times spanning from 2 ps (ref. 25) to 34–50 ps (ref. 26). This is the result of very strong electron–electron and electron–phonon interactions in the QCL polar semiconductor gain media, which induce an ultrafast non-radiative intersubband relaxation. This, in turn, prevents the successful use of conventional passive mode-locking techniques based on lumped absorbers, as local perturbations of the photon population are quickly reabsorbed by the fast gain that rapidly restores a quasi-continuous-wave emission profile. Recent approaches for short pulse emission in terahertz QCLs are therefore all based on active schemes. These include soliton compensation of the active region, leading to the generation of a train of 4-ps-long pulses, or generation of shorter isolated pulses, albeit with a duration limited by the slow electrical modulation employed in the active mode-locking. The latter, which makes inefficient use of the gain, also requires dedicated external electronics and connections, impacting its widespread adoption.

Pulse trains generated by soliton-like mode-locking were reported in quantum cascade ring lasers. Although they do not require a saturable absorber or external active modulation, they rely on the balancing action of anomalous chromatic dispersion and Kerr non-linearity to generate temporally and spectrally immutable pulses. Soliton formation was predicted and demonstrated in mid-infrared ring QCLs, but required spectral filtering at the output due to the presence of an additional dispersive wave. This approach cannot be exploited in Fabry–Perot architectures.

The realization of self-starting short pulse emission from terahertz QCLs is highly desirable for a variety of applications and for providing a compact alternative to bulky terahertz time-domain systems. Theoretical predictions of design strategies (that is, interleaved gain and absorbing periods with appropriate dipole moments) have been followed by the encouraging experimental observation of Rabi-flopping in mid-infrared QCLs; however, at terahertz frequencies, engineering intracavity semiconductor multilayers with stringent requirements of the gain and absorption faces fundamental obstacles due to the extremely small photon energies involved.

Here we demonstrate passive mode-locking in a semiconductor heterostructure laser operating at terahertz frequencies. By employing a heterogeneous gain medium and an architecture that integrates a distributed graphene saturable absorber (DGSA) on the top-surface of the double-metal QCL cavity, we achieve self-starting pulsed emission with 4.0-ps-long pulses in a compact, all-electronic, all-passive and inexpensive configuration. We take advantage of graphene saturable absorption’s high transparency modulation (~80%) and fast recovery times (2–3 ps) (faster than the gain recovery time in terahertz QCLs) while the intracavity terahertz radiation experiences saturable losses, which favours pulsed emission over naturally occurring continuous-wave emission.

Detailed electromagnetic simulations of the QCL structure and light–matter interactions in the active medium corroborate our findings, confirming the robustness of our approach. The proposed scheme can be applied to any semiconductor heterostructure laser, including mid-infrared QCLs, with claims of passive mode-locking of QCLs operating in the mid-infrared (intepreted as coherent dynamic instabilities resulting from the extremely fast gain recovery time of this family of class-A lasers).

**Results**

The QCL gain medium comprises nine GaAs/AlGaAs quantum wells, which form a cascade of alternating photon and longitudinal-optical-phonon-assisted transitions between two quasi-minibands. The gain medium consists of a 17-μm-thick GaAs/AlGaAs heterostructure featuring three active regions with gain bandwidths centred at 2.5, 3 and 3.5 THz, and comparable threshold current densities. The double-metal Fabry–Perot laser cavity includes top nickel-based lossy side absorbers to suppress high-order lateral modes (Methods). A 2.9-mm-long and 85-μm-wide reference Fabry–Perot QCL was fabricated with a set of 6.5-μm-wide side absorbers (see ref. 37 for transport and optical data), whereas a 2.3-mm-long and 49-μm-wide QCL with 0.5-μm-wide side absorbers was prepared for multilayer graphene (MLG) integration. Two 4-μm-wide stripes—aligned with the waveguide longitudinal axis and centred 12 μm from each edge—were lithographically defined onto the top gold layer, exposing the active region below. The DGSA was realized by transferring a seven-layer chemical-vapour-deposition-grown MLG film on top of the etched stripes (see Fig. 1a–c, Methods, and Supplementary Figs. 1 and 2). The MLG on the gold contact outside of the stripes is removed to allow for the application of the bonding wires (Fig. 1b), whereas the MLG ensures a homogeneous covering of the active region (Fig. 1c).

Multilayer graphene is shaped as a couple of stripes to introduce an additional loss component to the QCL waveguide; however, the intracavity electric field can saturate this loss—opening a net gain window that favours pulse formation. As MLG is distributed continuously along the entire cavity, this effect is not destroyed by the 2–3 ps fast gain recovery time of the investigated terahertz QCL active region, which would happen if MLG was placed at a single point along the QCL cavity. The position of the DGSA stripes (12 μm away from the waveguide centre) is chosen to guarantee at least 50% larger waveguide losses for TM₀₀ compared with TMₐ₀. Their width (4 μm each) results from a trade-off that ensures a sufficient overlap of the electric field on MLG while avoiding excessive losses with respect to the reference QCL.

The DGSA-QCL was simulated via COMSOL Multiphysics. The MLG is included as a transition boundary condition with refractive index determined by terahertz time-domain spectroscopy (TDS) experiments (Tera K5 by MenloSystems) performed on the nominally same MLG film transferred onto an undoped GaAs substrate, resulting in a complex refractive index 16.9 + i49.2 at 3 THz (Supplementary Fig. 3). The highly doped n layer (n = 2 × 10²⁰) between the active region and the top gold contact is also included in the model. Figure 1D shows the electric field intensity distribution in a cross-section of the QCL cavity at 3 THz; MLG affects the distribution across the interface at the active region/gold top contact/air boundary at y = 17 μm, inducing the concentration of the electric field on the MLG. Refs. 38,42 showed that a strong terahertz electric field can saturate the graphene absorption, which results in a decrease of the imaginary part of its complex refractive index due to its proportionality to the absorption coefficient αₚ = 4πk/λ₀, where λ₀ is the radiation wavelength.

We then simulated the DGSA-induced waveguide losses spectra for the TM₀₀ mode at 3 THz for different values of the imaginary part of the MLG refractive index (Fig. 1E).

The losses for each frequency have a maximum, as marked by the black dashed line on the map. Below this curve, a reduction of Im(αₚ) corresponds to lower losses, that is, the MLG stripes behave as a DGSA. Im(αₚ), as extracted from terahertz TDS, is represented by the purple dashed line in Fig. 1E. This guarantees DGSA operation at least for all frequencies >2.5 THz. The TM₀₀ and TMₐ₀ waveguide losses decrease for stronger saturation, thus leading to a net round-trip gain for pulsed over continuous wave emission. The opposite would be true if Im(αₚ)
The intermode beatnote map, plotted as a function of the driving current (Fig. 2d), shows that the comb regime in the DGSA laser persists at first for almost 100 mA (that is, the bias region of the reference laser) and then immediately before and across the negative differential resistance (NDR) region. Conversely, the reference structure behaves as a fully stabilized comb only above the onset of band alignment and for a current range of -106 mA (ref. 37), regardless of cavity dimensions. In fact, it is usually difficult to operate a QCL as a frequency comb because of the large cavity dimensions. The comb regime is only above the onset of band alignment and for a current range of -106 mA (ref. 37), regardless of cavity dimensions. In fact, it is usually difficult to operate a QCL as a frequency comb because of the large cavity dimensions.
The analysis of the beatnote linewidths shows a general agreement with the values extracted on reference lasers (2–8 kHz)12,14, whereas much smaller linewidths (600 Hz) are obtained when GVD compensation schemes are adopted37. This means that, as expected, the DGSA architecture does not affect the GVD and suggests that the appearance of a single and narrow beatnote at driving currents >570 mA is indicative of a different physical mechanism to that operated on the QCL by the GSA. Similar results (Supplementary Fig. 4) have been achieved on a second DGSA-QCL with the same active region, 15 graphene layers and different cavity dimensions (3.13 mm × 68 µm × 17 µm), also with substantial reduction of the beatnote linewidth for threshold currents >550 A cm−1.

We next measured the emission profile of the DGSA in the time domain with a coherent technique, gaining access to the amplitude and phase information of the emitted electric field. To this aim, we performed a coherent measurement based on injection seeding46, previously used to show pulsed behaviour induced in terahertz QCLs by active mode-locking45. Such a technique allows measurement of the free-running QCL emission with a temporal resolution better than 100 fs.

Figure 3a plots a 250-ps-long time-trace of the electric field emitted by the reference QCL (ref. 37) biased at 663 mA (I/ITH = 1.70), acquired 1 ns after the seeding pulse injection, that is, well into the steady-state regime where no effect of the initial pulse persists. No applied modulation at the QCL round-trip frequency is here present, as it instead occurs in the case of active mode-locking. The quasi continuous-wave profile combined with amplitude modulations on a short time scale (<5 ps) is a typical consequence15,16 of the interplay between the QCL fast gain recovery time13 and the giant Kerr non-linearity13 arising from Bloch gain12. The spectrum (Fig. 3b), obtained by Fourier-transforming the electric field time trace, shows broadband emission from 2.2 to 3.3 THz, in agreement with the FTIR spectra measured under-vacuum12,14. The latter, however, shows modes up to 3.45 THz and an even power distribution between the two bands centred at 2.8 and 3.2 THz, meaning that the higher frequency band is more attenuated in the TDS spectrum than the band at lower frequencies. This is consistent with the presence of the absorption at 3.7 THz of the ZnTe crystal50 used for coherent electro-optic sampling.

Figure 3c plots a 250-ps-long time-trace of the electric field emitted by the reference QCL (ref. 37) biased at 663 mA (I/ITH = 1.70), acquired 1 ns after the seeding pulse injection, that is, well into the steady-state regime where no effect of the initial pulse persists. No applied modulation at the QCL round-trip frequency is here present, as it instead occurs in the case of active mode-locking. The quasi continuous-wave profile combined with amplitude modulations on a short time scale (<5 ps) is a typical consequence15,16 of the interplay between the QCL fast gain recovery time13 and the giant Kerr non-linearity13 arising from Bloch gain12. The spectrum (Fig. 3b), obtained by Fourier-transforming the electric field time trace, shows broadband emission from 2.2 to 3.3 THz, in agreement with the FTIR spectra measured under-vacuum12,14. The latter, however, shows modes up to 3.45 THz and an even power distribution between the two bands centred at 2.8 and 3.2 THz, meaning that the higher frequency band is more attenuated in the TDS spectrum than the band at lower frequencies. This is consistent with the presence of the absorption at 3.7 THz of the ZnTe crystal50 used for coherent electro-optic sampling.

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physical mechanism at the origin of the observed phenomenon. The active region dynamics are modelled employing a Lindblad-type approach \(^{51,52}\) and the quantum system represents a QCL period with periodic boundary conditions \(^{51,52}\) (Methods). The waveguide parameters used for the simulations are presented in Table 1. The simulated optical power of the last five roundtrip times \((T_{rt})\) outcoupled at the back facet in stationary operation, and the associated power spectrum, are shown in Fig. 4a,b. As in the experiment, at ~2.5-times the threshold current a regular train of pulses is observed with a narrow beatnote (far below the numerical resolution limit) and a temporal pulse width (FWHM) of ~4.4 ps, consistent with the measured duration (4 ps). The resulting comb shows clear, narrow (far below the numerical resolution limit) and equidistant lines, with thirteen of them in a 10 dB power range. The slight asymmetry of the pulse is reproduced (compare Fig. 3h), and the optical power per comb tooth (0.15 mW for the highest, and 0.07 mW average for the teeth in a 10 dB power range) fits well to the measured ~0.12 mW. For comparison, enhancing the gain by increasing the doping density by ~2.5% reproduces the experimental breakdown of pulses at currents \(350 \text{ mA} < I < 570 \text{ mA}\), that is, above the breakdown of the initial comb state and before entering in the second stable comb regime (Fig. 4c,d), with the resulting chaotic behaviour. The simulated radiofrequency beatnote width ~400 MHz agrees very well with the experimental results in Fig. 2e.

Table 1 | Simulation parameters

| Parameter                          | Value   |
|-----------------------------------|---------|
| Loss coefficient \(a_0\)          | 10.5 cm\(^{-1}\) |
| Saturable loss coefficient \(a_1\) | 10.5 cm\(^{-1}\) |
| Saturation power \(P_s\)          | 42.5 mW |
| Saturable absorber recovery time \(\tau_r\) | 2.5 ps |
| Overlap factor                    | 0.97    |
| Centre frequency                  | 3.0 THz |
| Mirror reflectivity (power)       | 70.88%  |
| Refractive index                   | 3.6     |
| GVD factor \(\beta_2\)            | \(1.0 \times 10^{-22} \text{s}^2 \text{m}^{-1}\) |
To prove quantitatively the presence of mode-locking in the simulated optical field, we employ power and phase noise quantifiers $M_{\rho}$ and $M_{\Delta \phi}$ (ref. 53), as:

$$M_{\rho} = \frac{1}{N_{10}} \sum_{q=1}^{N_{0}} \sigma_{\rho_q} \quad M_{\Delta \phi} = \frac{1}{N_{10}} \sum_{q=1}^{N_{0}} \sigma_{\Delta \phi_q},$$

respectively, with

$$\sigma_{\rho_q} = \sqrt{\langle (P_q(t) - \langle P_q(t) \rangle)^2 \rangle} \quad \sigma_{\Delta \phi_q} = \sqrt{\langle (\Delta \Phi_q(t) - \langle \Delta \Phi_q(t) \rangle)^2 \rangle}$$

where $P_q(t)$ represents the modal amplitudes and $\Delta \Phi_q(t)$ represents the modal phase differences between one mode and the adjacent, at time $t$ for each mode $q = 1, ..., N_{0}$, with $N_{0}$ being the number of modes in the ~10 dB spectral bandwidth.

In this framework, an optical frequency comb is present for $M_{\rho} < 10^{-3}$ mW and $M_{\Delta \phi} < 2 \times 10^{-2}$ rad. Regarding the pulsed mode of Fig. 4a,b, values of $M_{\rho} = 3 \times 10^{-4}$ mW and $M_{\Delta \phi} = 3 \times 10^{-2}$ rad are obtained. We can thus confirm an ultrastable mode-locked operation in this current range. Calculating the same parameters for the results of Fig. 4c,d, yields $M_{\rho} = 0.13$ mW and $M_{\Delta \phi} = 4.91$ rad. This increase proves the absence of mode-locking for higher gain, in agreement with our theory of pulse formation, as discussed below.

Discussion

One key observation to understand pulse formation in our DGSA-QCL is that the graphene stripes provide fast saturable absorption across the resonator length, while the gain also has a recovery time that is much faster (around 3 picoseconds for the present structure) than the roundtrip time. Ref. 54 suggests that mode-locked operation only occurs if the pulses are stabilized by a net gain window, whereas the standard passive mode-locking theory does not provide self-starting mode-locked pulse solutions for instantaneous gain and saturable absorption. These assumptions are relaxed in our experiments by the fact that both the gain and saturable absorber have a finite recovery time of a few picoseconds. As also pointed out in ref. 54, the constraints for self-starting passive mode-locking are further relaxed if a second, longer gain recovery time is present, as is typically the case for bound-to-continuum QCLs due to the electron transport across the miniband. But most importantly, deviations from the analytical theory of ref. 54 arise due to the unique design of our structure with a saturable absorber distributed along the entire Fabry–Perot cavity, which gives rise to effects such as cross-saturation and spatial hole burning. Furthermore, quantum coherence and nonlinear effects of the quantum active region have to be taken into account, requiring our detailed Maxwell–Bloch-type simulations presented in the Methods. The emergence of mode-locked pulses that are in close qualitative agreement with the measured ones confirms the validity of our model. Furthermore, this theoretical model allows us to clearly identify the saturable absorber as the element enabling mode-locked operation: decreasing the saturable absorber recovery time yields shorter pulses while considerably extending the recovery time results in a breakdown of mode-locking.

The numerical pulse duration of 4.4 ps is relatively close to the Fourier limit (3.1 ps). This suggests that the main limitation in terms of achievable pulse duration is the laser bandwidth and the chromatic dispersion. Furthermore, as mentioned above, the simulations indicate that an even faster saturable absorber recovery time would enable considerably shorter pulses; thus, further optimization of the waveguide structure, adjustments to the graphene optical properties and the employment of ultrabroadband (an octave) active regions could be adopted to enhance the emission bandwidth uniformity, effectively indicating a practical path to achieve sub-picosecond terahertz pulse generation.
Conclusion
We have shown 4.0 ps pulse generation from a passively mode-locked semiconductor terahertz laser using a unique graphene-based saturable absorber that is distributed along the entirety of the terahertz QCL cavity. This has overcome a long-standing bottleneck of realizing passive pulse generation from laser systems with ultrafast dynamics, where the gain recovery time is much shorter that the photon round trip time, and could be applied to other types of laser systems beyond terahertz QCLs. Exploiting the wide spectral gain of terahertz QCLs and the lithographic capability to embed miniaturized, highly nonlinear, intracavity graphene absorbers to produce short pulses without any external source or seeding system, could allow QCLs to become an attractive alternative to present-day terahertz TDS systems in spectroscopic and metrological applications. Although this work can naturally impact frequency comb spectroscopy, further improvements could impact other domains such as the dynamics of systems that need to be pumped at targeted terahertz frequencies. This will require shorter (sub-picosecond) and higher intensity pulses separated by longer round trip times, which could be achieved through longer or external cavities to increase the round-trip time, combined with pulse compression techniques (on-chip or external) to compensate for dispersion. It should also be noted that most femtosecond commercial systems in the visible/infrared rely on passive pulse generation, as this approach should also be noted that most femtosecond commercial systems in the visible/infrared rely on passive pulse generation, as this approach permits the shortest pulses and highest powers. Our work will hence open interesting application perspectives in spectroscopy, as there is currently no passive mode-locked system for the terahertz range.

Online content
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Methods
Fabrication and graphene preparation
Fabry–Perot laser bars are fabricated in a metal–metal waveguide configuration via Au–Au thermocompression wafer bonding of a 17-µm-thick active region onto a highly doped GaAs substrate. This is followed by the removal—through a combination of mechanical lapping and wet etching—of the host GaAs substrate of a material grown by molecular beam epitaxy. An AlAs/GaAs stop layer is then removed using HF etching. Vertical sidewalls are defined by inductively coupled plasma etching of the laser bars to provide uniform current injection. A Cr/Au (10 nm/150 nm) top contact is then deposited along the centre of the ridge surface, leaving a thin region uncovered along the ridge edges and two stripes uncovered 12 µm from the centre of the cavity; 3-µm-wide nickel (5-nm-thick) side absorbers were then deposited over the lateral uncovered region using a combination of optical lithography and thermal evaporation. These lossy side absorbers are intended to inhibit lasing of the higher order lateral modes by increasing their threshold gain.\(^\text{19}\)

The multilayer graphene sample is prepared and transferred on the stripes, using a wet transfer technique: A4-950K (methyl-methacrylate) polymer (PMMA) is spin coated at 2,000 r.p.m. on the surface of a single-layer graphene (SLG) sample (1 cm × 1 cm) grown on copper via chemical-vapour-deposition. After 1 min on a hot plate at 90 °C, the sample is placed in a solution of 1 g of ammonium persulfate and 40 ml of deionized water to etch the copper substrate. Once the copper etching is complete, the PMMA-SLG film is transferred in a beaker with deionized water and then lifted with a second copper-graphene square to obtain a bilayer graphene sample. This sample is left to dry overnight and finally the PMMA is removed with acetone. The copper of the bilayer graphene is etched with the same technique, and then lifted by another SLG on copper. This process is repeated until the desired MLG thickness is reached, in our case seven layers. The MLG is transferred onto the QCL top contact and then removed from the sides of the laser cavity and from the top gold contact by oxygen reactive ion etching.

The backside of the substrate is then lapped down to 150 µm for thermal management and enable continuous-wave operation. Laser bars (50 µm wide and 2.2 mm long) are then cleaved and the device mounted on a copper bar, wire bonded, and then mounted onto the cold finger of a helium continuous-flow cryostat. The fabrication flowchart is presented in Supplementary Fig. 1.

Model
The active region dynamics is modelled based on a Lindblad-type approach, where the quantum system represents a QCL period with periodic boundary conditions.\(^\text{2,3}\) We performed self-consistent carrier transport simulations of the active region to extract the corresponding eigenenergies, optical dipole moments as well as the scattering and dephasing rates. For simplicity, the dynamical model only takes into account the 2.5 THz design, yielding agreement with experiments. The dephasing rates.\(^\text{51}\) For simplicity, the dynamical model only takes into account the 2.5 THz design, yielding agreement with experiments. The dephasing rates for simplicity.\(^\text{51}\)

\[ \frac{\partial}{\partial t} a_\pm (z,t) = -\frac{P}{\tau_E} a_\pm - \frac{a_\pm - a_s}{\tau_s} \]  

(2)

with the optical power \(P = |E|^2 + |\beta|^2\) and saturable absorber recovery time \(\tau_s\). Electromagnetic simulations yield \(\alpha_s = \alpha_s = 10.5 \text{ cm}^{-1}\) (Fig. 1c).

Data availability
The data presented in this study are available on reasonable request from the corresponding author.

Code availability
The relevant computer codes supporting this study are available from the authors on reasonable request.

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Author contributions
M.S.V. and V.P. conceived the concept. E.R. fabricated the devices, set up the transport and optical experiments. V.P. performed numerical simulations and interpreted the data. E.R., S.S.D., S.K. and A.D.V. provided equipment and acquired the experimental data. L.L, A.G.D. and E.H.L grew by the molecular beam epitaxy. An Al0.5Ga0.5As etch stop layer is then removed through \(26,000\) roundtrips to ensure convergence to steady state.\(^\text{11}\)

Competing interests
The authors declare no competing interests.

Additional information
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