Graphene saturable absorber mirror for passive mode-locking of mid-infrared QCLs

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Abstract
Passive mode-locking in quantum cascade lasers (QCLs) remains one of the huge challenges because of the fast relaxation time of the excited carriers which is typically in the range of sub-picoseconds. The use of conventional techniques such as the semiconductor saturable absorber mirror is inefficient because the spatial hole burning effect dominates the carrier dynamics. To overcome this effect, longitudinal transition structures with relaxation time around 50 ps were proposed. However, mode-locking is assured with an external modulation at a cavity roundtrip frequency. In this paper, we demonstrate that a single-layer graphene used as a saturable absorber permits to generate stable pulses in such structures. The graphene is integrated with a highly reflective mirror to increase the internal electric field and achieve the saturation intensity. The dynamic of the QCL is modeled with Maxwell-Bloch equations while Maxwell-Ampere equation is used for the graphene layer by considering a nonlinear conductivity. This system of equations is solved using the one-dimensional Finite-Difference Time-Domain (FDTD) method. To model the single-layer graphene of a 0.33 nm thickness, a specific sub-cell is implemented based on Maloney method. Simulation results of a 6.2 μm QCL with a diagonal radiative transition show a generation of isolated pulses with a peak electric field of 80 MV.m⁻¹ and a duration of 51 fs. The mode-locking remains stable for QCLs with a vertical transition having a relaxation time below 5 ps.

Keywords Quantum cascade laser · Mode-locked laser · Maxwell-bloch equations · Graphene · Numerical simulation · FDTD

1 Introduction
Quantum cascade lasers (QCL) have energy transitions located either in the conduction band or in the valence band. These transitions are obtained by engineering of quantum well structures and permit the generation of photons in mid- and far-infrared regions or in THz region Kazarinov and Suris (1971); Faist et al. (1994). The performances of QCLs have been greatly enhanced and devices are today commercially available. QCLs have become
one of the most important compact light sources in the mid-infrared region (MIR). However, the mode-locking of QCLs for the generation of ultrashort pulses remains one of the targeted challenges because of the fast gain recovery time compared to the roundtrip time of a $\sim 3$ mm cavity length. In mode-locking theory, the phase lock of the laser longitudinal modes is obtained by a modulation of losses with a period equal to the cavity roundtrip time. This modulation can be driven externally with a sinusoidal signal or passively from the intensity-dependent loss mechanism induced by the pulse energy. The active mode-locking of MIR QCLs is only obtained for a current injection close to the threshold. When the laser operates at higher current injection, spatial hole burning (SHB) effect governs the laser dynamic which induces instability in the mode-locking Wang (2009); Gkortsas (2010). In Outafat et al. (2022), we have proposed to integrate a single-layer graphene to one of the facets of the QCL structure to improve the active mode-locking. This leads to higher current injection increasing thus the pulse intensities and at the same time decreasing the pulse width. However, it is more difficult to achieve passive mode-locking in a monolithic two-section cavity because it requires specific absorbing conditions. Talukder and Menyuk (2014) gave the conditions in such two-section cavity where one section provides gain while the other one induces quantum coherent absorption acting as a saturable absorber. The disadvantage of this structure is that the ratio of dipole moments of the absorber to the gain medium must be greater than two, which makes the monolithic solution impractical. Another proposed solution concerns the interleaving of absorption and gain periods where the self-induced transparency (SIT) effect is exploited Talukder and Menyuk (2010). SIT mode-locked QCLs can produce stable pulses of the order of $\sim 100$ fs duration over a broad parameter range.

In this work, we investigate the passive mode-locking of MIR QCLs emitting at a wavelength of 6.2 $\mu$m by using graphene as saturable absorber. For such laser with ultrashort gain recovery time, fast saturable absorber is required. The relaxation time of graphene is very fast compared to the gain recovery time and also to the pulse width making this solution efficient for QCL mode-locking. In addition, the saturation intensity in graphene is very low compared to the saturation intensity in semiconductor saturable absorber mirrors (SESAM). Single-layer graphene exhibits an absorbance $\alpha_\pi = 2.3\%$ Nair (2008) which is 50 times higher than the absorbance of GaAs with the same thickness Bao and Loh (2012). This means that graphene can be easily saturated due to the high optical absorption. The use of fast saturable absorber effect of graphene have been investigated in mode-locking of lasers. In Zhu (2016), the graphene was used in the cavity of a fiber laser demonstrating a slight improvement. In semiconductor laser, the graphene has also been used as a saturable absorber for mode-locking of a 1.55 $\mu$m semiconductor laser diode Mock (2017). Recently, Mezzapesa (2020) have been demonstrated the integration of graphene in THz QCL frequency comb.

The laser dynamic is modeled here using Maxwell-Bloch equations in two-level atom approximation. The resolution of these equations is performed using one-dimensional Finite-Difference Time-Domain (FDTD) method. The graphene layer has a thickness of one carbon atom Bao (2009) and the distance between two graphene layers is estimated to 0.33 nm Kumar et al. (2021). The modeling of the graphene as a single FDTD-cell requires the discretization of the space as thin as its width which could have a negative impact on the computation time. To deal with this issue, the graphene layer can be considered in a cell of larger size but with an effective value of the nonlinear conductivity Mock (2017); Mezzapesa (2020). We adopted such approach in our previous work Outafat et al. (2022) where two different methods have been used to model the nonlinear interaction of the light-wave with the graphene layer. In this paper, the single-layer graphene is modeled using a
Maloney method suitable for thin layers in FDTD method Maloney and Smith (1992). In this specific method, the graphene is localized in a sub-cell which is completed with $SiO_2$ material. The dielectric constant and the nonlinear conductivity of the graphene is calculated according to the method specifications Maloney and Smith (1992).

The remaining of the paper is organized as follows: in section II, the Maxwell-Bloch equations for QCL and the Maxwell’s equations for graphene are briefly described. In section III, the implementation of the FDTD method to solve the QCL and graphene equations is presented. Section IV describes the simulation results of a passive mode-locked QCL integrating a single-layer graphene. The last section gives a general conclusion of this work.

2 Structure modeling

2.1 Structure description

Figure 1 describes the simulated structure where the graphene layer is inserted between the QCL gain medium and the Bragg mirror. The total length of the laser cavity is 2.6 mm. The Bragg mirror is composed of 7 layers grating of a high refractive index PbSe and a low refractive index BaF$_2$. The thickness of each layer is equal to a quarter of the propagating wavelength in the related medium. The combination of the graphene and the high reflective mirror forms what is called graphene saturable absorber mirror. A $SiO_2$ layer is added between the graphene and Bragg mirror to control the overlap between the absorbing layer and the maximum field intensity. This field intensity profile is due to the interference between incident and reflected light-waves from the mirror. At the optimized graphene location, the intensity of the electric field can increase up to 400% allowing to reach the saturation intensity of graphene Zaugg (2013).

**Fig. 1** Structure of the QCL with single-layer graphene based mirror
2.2 Quantum cascade laser modeling

The QCL dynamic is modeled using Maxwell-Bloch equations in the context of the two-level atoms approximation Ziolkowski et al. (1995). This model describes the interaction of ultrashort pulse light with a nonlinear medium which couple Maxwell’s equations in one-dimensional configuration (1a)-(1b) and Bloch equations (1c)-(1e).

\[
\frac{\partial H}{\partial t} = -\frac{1}{\mu_0} \frac{\partial E}{\partial y} \tag{1a}
\]

\[
\frac{\partial E}{\partial t} = -\frac{1}{\varepsilon_0 \varepsilon_r} \frac{\partial H}{\partial y} - \frac{N \mu}{\varepsilon_0 \varepsilon_r \tau_2} \rho_a + \frac{N \mu_0 \omega_0}{\varepsilon_0 \varepsilon_r} \rho_b - l_0 E \tag{1b}
\]

\[
\frac{\partial \rho_a}{\partial t} = \omega_0 \rho_b - \frac{1}{T_2} \rho_a \tag{1c}
\]

\[
\frac{\partial \rho_b}{\partial t} = -\omega_0 \rho_a - \frac{1}{T_2} \rho_b + \frac{2 \mu E}{\hbar} \Delta \tag{1d}
\]

\[
\frac{\partial \Delta}{\partial t} = -\frac{2 \mu E}{\hbar} \rho_b - \frac{\Delta}{\tau_1} + I + D \frac{\partial^2 \Delta}{\partial y^2} \tag{1e}
\]

where \( E \) is the electric field along \( z \)-axis, \( H \) is the magnetic field along \( x \)-axis, \( l_0 \) is the linear loss per unit length, \( \varepsilon_0 \) is the dielectric constant of vacuum, \( \varepsilon_r \) is the relative permittivity, \( \mu_0 \) is the vacuum permeability, \( \rho_a \) and \( \rho_b \) are the existence probability of electron in state \( a \) and \( b \) respectively, with \( a \) and \( b \) referring respectively to the ground and excited states, \( \mu \) is the electric dipole moment of a two-energy level atom, \( \Delta \) is the inversion population, \( \tau_1 \) is the excited-state lifetime and \( \tau_2 \) is the coherence time, \( \hbar \) is the reduced Planck constant, \( \omega_0 \) is the transition frequency. The overlap factor between the laser mode and the active region is chosen equal to unity.

These equations are solved using 1D-FDTD method along a propagation following \( y \)-axis. The mirror reflections modeled through Fresnel reflection coefficients induce the growth of standing waves inside the cavity. This leads to SHB which affects the stability of the mode-locking.

2.3 Single-layer graphene modeling

The physical parameter of the single-layer graphene involved here is the saturable absorption. To consider this nonlinear behavior, the graphene can be modeled in different ways. In Mock (2017), two-level rate equations have been considered for the semiconductor laser (gain medium) and the graphene layer (absorbing medium) coupled to Maxwell’s equations. The dynamic of the carrier density has been described using the electric conductivity that is calculated from the saturable absorption coefficient following the equation (4). In Mezzapesa (2020), the Maxwell-Bloch equations integrating the saturable absorption have been discussed by describing the saturable absorbers as an open two-level system. The dipole moment and carrier density values have been determined from the saturation.
intensity and saturable absorption coefficient, respectively. Here, we model the graphene using the Maxwell-Ampere equation by determining the nonlinear conductivity from the graphene saturable absorption. We have compared such approach with Maxwell-Bloch equations in Outafat et al. (2022) and close results have been obtained with the two methods.

To model the graphene conductivity from the electromagnetic wave absorption coefficient, the general expression (2) is used Singh et al. (2021).

\[
\sigma = \varepsilon_0 \varepsilon_{rg} \omega \sqrt{\left( \frac{\sqrt{2} c}{\omega n_g} \alpha \right)^2 + 1} - 1 \quad (2)
\]

where \( \varepsilon_{rg} \) is the real permittivity of graphene, \( c \) is light velocity, \( n_g \) is the graphene refractive index and \( \alpha \) is the absorption coefficient.

In the frequency range of the mid-infrared region from 2 to 10 \( \mu \text{m} \), the condition \( \left( \frac{\sigma}{\varepsilon_0 \varepsilon_{rg} \omega} \right)^2 \ll 1 \) is verified and the below approximation may be used

\[
\sigma(I) \approx \frac{2 n_g}{\eta_0} \alpha(I) \quad (3)
\]

with \( \eta_0 \) the vacuum wave impedance.

The absorption coefficient of graphene depends on the light intensity following

\[
\alpha(I) = \frac{\alpha_s}{1 + \frac{I}{I_s}} + \alpha_{ns} \quad (4)
\]

where \( \alpha_s \) is the saturable absorption and \( \alpha_{ns} \) is the non-saturable absorption estimated experimentally with the z-scan technique to respectively 0.72 and 0.13 Li (2014).

\[ I = \frac{n_s}{n_0} |E|^2 \] is the field intensity and \( I_s = \frac{n_s}{n_0} |E_s|^2 \) is the field saturation intensity of graphene estimated to be 0.2 MW.cm\(^{-2}\) at 10 \( \mu \text{m} \) wavelength Vasko (2010).

Thus, the system of equations that describes the interaction of electromagnetic field with graphene is as follows

\[
\frac{\partial H}{\partial t} = -\frac{1}{\mu_0} \frac{\partial E}{\partial y} \quad (5a)
\]

\[
\frac{\partial E}{\partial t} = -\frac{1}{\varepsilon_0 \varepsilon_{rg}} \frac{\partial H}{\partial z} - \frac{\sigma(I)}{\varepsilon_0 \varepsilon_{rg}} E \quad (5b)
\]

From (3) and (5b), the Maxwell-Ampere equation becomes

\[
\frac{\partial E}{\partial t} = -\frac{1}{\varepsilon_0 \varepsilon_{rg}} \frac{\partial H}{\partial z} - \frac{2 n_g}{\varepsilon_0 \varepsilon_{rg} \eta_0} \left( \frac{\alpha_{ns} + \alpha_s}{1 + \left( \frac{E}{E_s} \right)^2} \right) E \quad (6)
\]

In our previous work Outafat et al. (2022), the single-layer graphene has been modeled by one cell of FDTD grids. The nonlinear conductivity has been determined from the normalized absorption coefficient considering a cell spatial step of 31 nm. However, we use here
the realistic thickness of single-layer graphene which is typically 0.33 nm. The FDTD cell size cannot be chosen at such a small value because the structure dimension is much larger and the time step \( \Delta t \) would be very short. Then, a specific method proposed in Maloney and Smith (1992) is used. The graphene is located in a thin part of the FDTD cell and the rest of the cell is completed with another material (see Fig. 1). We chose the \( \text{SiO}_2 \) material to complete the cell size. The implementation of this method requires a new conductivity \( \sigma_M \) and a dielectric constant \( \varepsilon_M \) of the considered cell as following

\[
\sigma_M(I) = d \sigma(I) \tag{7a}
\]

\[
\varepsilon_M = (1 - d) \varepsilon_0 \varepsilon_{r\text{SiO}_2} + d \varepsilon_0 \varepsilon_{rG} \tag{7b}
\]

where \( d = \frac{L_g}{\Delta y} \) is the ratio between the graphene thickness \( (L_g) \) and the cell size \( (\Delta y) \), and \( \varepsilon_{r\text{SiO}_2} \) is the \( \text{SiO}_2 \) relative dielectric constant.

From these new defined parameters, we can rewrite equation (6) as

\[
\frac{\partial E}{\partial t} = \frac{1}{\varepsilon_M} \frac{\partial H}{\partial z} - \frac{2d}{\varepsilon_M \varepsilon_0} \left[ \alpha_{ns} + \alpha_s \left( 1 + \left[ \frac{E}{E_s} \right]^2 \right)^{-1} \right] E \tag{8}
\]

3 Numerical implementation

The differential equations are solved using the finite-difference time-domain method. This method was proposed by Yee in 1966 Yee (1966) to solve complex RF-structures and was generalized to photonic structures and light/matter dynamic interaction analyses Taflove et al. (2005). More recently, the FDTD was used to simulate the passive mode-locking of semiconductor diode laser Mock (2017) and THz frequency comb QCL Mezzapesa (2020) based on graphene as saturable absorber. As FDTD involves the discretization of time and space to form cells, its application to solve equations (1a)-(1e) is not trivial because of the mutual parameters dependence. Specially, the expression (1b) includes variables computed at the same time steps. This description represents an explicit scheme which requires additional numerical methods. A complete nonlinear model for wave propagation in a two-level atom system has been demonstrated in Ziolkowski et al. (1995) using the FDTD with an iterative predictor-corrector method to solve the Maxwell-Bloch equations. This model is able to predict saturation effects as well as self-induced transparency. In addition, the numerical setup is simple compared to numerical simulations with the semiclassical traveling wave model. This model is accomplished by introducing in equations (1a)-(1e) the slowly varying amplitude method which requires the counter-propagating waves Vukovic et al. (2020).

3.1 Quantum cascade laser

In order to keep the formulation of equations explicit, the weak coupling approach is used to solve the system of QCL equations. The Maxwell-Ampere equation (1b) and Bloch equations (1c)-(1e) are separated in time as described in Outafat et al. (2022) for the QCL including self-induced transparency and in Outafat et al. (2022) for the active
mode-locking of MIR QCL. The advantage of this method is the implementation simplicity of Maxwell-Bloch equations using the one-dimensional FDTD method. Thus, the electric field is computed at time steps \( n \Delta t \) while Bloch equations and the magnetic field are computed at time steps \( \left( n + \frac{1}{2} \right) \Delta t \). However, a wise choice of time steps is to be considered because it has been demonstrated in Petropoulos (1994) that inconsistent time centering of the discretized derivatives results in a more restrictive stability condition than the original FDTD formulation for dispersive media. The discretized Maxwell-Bloch equations in one-dimensional problem become then

\[
H_{m+\frac{1}{2}}^{n+\frac{1}{2}} = H_{m+\frac{1}{2}}^{n-\frac{1}{2}} - \frac{\Delta t}{\mu_0 \Delta y} (E_{m+1}^n - E_m^n) \tag{9a}
\]

\[
E_m^{n+1} = E_m^n - \frac{\Delta t}{\varepsilon \Delta y} \left[ H_{m+\frac{1}{2}}^{n+\frac{1}{2}} - [H_{m+\frac{1}{2}}^{n-\frac{1}{2}}] - \frac{\Delta t N \mu_0}{\varepsilon T_2} \rho_{a_m}^{n+\frac{1}{2}} \right]
+ \frac{\Delta t N \mu_0}{\varepsilon} \rho_{a_m}^{n+\frac{1}{2}} \Delta t I_0 \frac{E_m^n + E_m^{n+1}}{2} \tag{9b}
\]

\[
\rho_{a_m}^{n+\frac{1}{2}} = \rho_{a_m}^n - \frac{\Delta t}{2 T_2} \left[ \rho_{a_m}^{n+\frac{1}{2}} + \rho_{a_m}^{n-\frac{1}{2}} \right] + \frac{\Delta t \omega_0}{2} \left[ \rho_{b_m}^{n+\frac{1}{2}} + \rho_{b_m}^{n-\frac{1}{2}} \right] \tag{9c}
\]

\[
\rho_{b_m}^{n+\frac{1}{2}} = \rho_{b_m}^n - \frac{\Delta t \omega_0}{2} \left[ \rho_{a_m}^{n+\frac{1}{2}} + \rho_{a_m}^{n-\frac{1}{2}} \right] - \frac{\Delta t}{2 T_2} \left[ \rho_{b_m}^{n+\frac{1}{2}} + \rho_{b_m}^{n-\frac{1}{2}} \right] \tag{9d}
\]

\[
\Delta_m^{n+\frac{1}{2}} = \Delta_m^{n-\frac{1}{2}} - \frac{\Delta t \mu E_m^n}{\hbar} \left[ \rho_{b_m}^{n+\frac{1}{2}} + \rho_{b_m}^{n-\frac{1}{2}} \right] - \frac{\Delta t}{2 T_1} \left[ \Delta_m^{n+\frac{1}{2}} + \Delta_m^{n-\frac{1}{2}} \right]
+ \Delta t I + \frac{\Delta t}{\Delta y^2} D \left[ \Delta_m^{n+\frac{1}{2}} - 2 \Delta_m^{n-\frac{1}{2}} + \Delta_m^{n-\frac{1}{2}} \right] \tag{9e}
\]

where index \( m \) corresponds to spatial locations and index \( n \) to time steps.

### 3.2 Single-layer graphene highly reflective mirror

The highly reflective mirror is composed of a single-layer graphene and a Bragg mirror. This mirror is formed by cascading layers of high and low refractive index materials. The thickness of each layer is equal to a quarter wavelength \( \Delta \), where the sub-index \( q \) refers to the considered material. The Bragg mirror is modeled using Maxwell equations as given by expressions (5a) and (5b) with the conductivity set to zero.

As for the QCL gain medium, the electric field is calculated at time steps \( n \Delta t \) and the magnetic field is determined at time steps \( \left( n + \frac{1}{2} \right) \Delta t \). The discretization of Maxwell equations for the graphene effective layer is
\[ H_{m+\frac{1}{2}}^{n+\frac{1}{2}} = H_{m+\frac{1}{2}}^{-\frac{1}{2}} - \frac{\Delta t}{\mu_0 \Delta y} (E_{m+1}^n - E_m^n) \] (10a)

\[ E_{m+1}^n = \frac{1}{B} \left[ A E_m^n - \frac{\Delta t}{\varepsilon_0 \varepsilon_{ri} \Delta y} \left( H_{m+\frac{1}{2}}^{n+\frac{1}{2}} - H_{m-\frac{1}{2}}^{n+\frac{1}{2}} \right) \right] \] (10b)

where

\[ A = 1 - \frac{n}{\varepsilon_\infty \eta_0} \Delta t \left[ \alpha_{ns} + \alpha_s (1 + \left( \frac{E_{m+1}^n + E_m^n}{2 E_s} \right)^2)^{-1} \right] \]

\[ B = 1 + \frac{n}{\varepsilon_\infty \eta_0} \Delta t \left[ \alpha_{ns} + \alpha_s (1 + \left( \frac{E_{m+1}^n + E_m^n}{2 E_s} \right)^2)^{-1} \right] \]

The expression (10b) cannot be solved directly due to the square term of the electric fields at time steps \( n\Delta t \) and \((n+1)\Delta t\). An additional numerical method is needed to deal with these two equations and the fixed-point method has been chosen. This method consists of an iterative resolution of nonlinear equations by numerical approximation of the root, here the unknown electric field at time step \((n+1)\Delta t\), in the form \( E_{m+1}^n = f\left(E_m^{n+1}\right) \) Dlala et al. (2008).

## 4 Simulation parameters

Table 1 summarizes the physical parameters of graphene Mock (2017) and Bragg mirror used in simulation. The Bragg mirror is composed of 3 layers of PbSe and 2 layers of BaF\(_2\) materials. The QCL parameters are the same as those given in Wang (2009); Gkortzas (2010) and Outafat et al. (2022).

For the highly reflective mirror, the thicknesses of PbSe and BaF\(_2\) layers are respectively fixed to \( \frac{d}{4} \). The thickness of the SiO\(_2\) layer permits to control the overlap of the maximum field intensity with graphene. A field intensity enhancement factor up to 400% has been obtained using such distance from the reflective mirror in Zaugg (2013). Because of the standing waves establishment due to the incoming and reflected waves at the mirror, the field intensity depends on the distance \( y \) from the mirror location. Thus, the position of the graphene layer is chosen to maximize the interaction between the field intensity and the graphene layer. The initial value of SiO\(_2\) is set to \( \frac{d_{SiO2}}{4} \) and thereafter changed to analyze its effect on the mode-locking stability. The field intensity is shown in Fig. 2 where the point

| Table 1 Physical parameters used in simulation |
|-----------------------------------------------|
| **Parameters**                        | **Symbol** | **Value** |
| Saturation intensity                 | \( I_s (\text{MW/cm}^2) \) | 0.2       |
| Saturable loss                       | \( \alpha_s (m^{-1}) \) | 2.32 \times 10^7 |
| Non-saturable loss                   | \( \alpha_{ns} (m^{-1}) \) | 2.09 \times 10^6 |
| Relative dielectric constant PbSe   | \( \varepsilon_{PbSe} \) | 5         |
| Relative dielectric constant BaF\(_2\) | \( \varepsilon_{BaF2} \) | 1.4       |
| Relative dielectric constant SiO\(_2\) | \( \varepsilon_{SiO2} \) | 1.5       |
y = 0 represents the graphene location. As it is shown, the field intensity is maximum for a thickness of SiO$_2$ layer equal to $\frac{\lambda_{SiO_2}}{4}$.

5 Obtained results

We start the analysis with a MIR QCL of a diagonal optical transition characterized by a relaxation time $T_1$ around 50 ps which is in the order of the roundtrip time of the laser cavity. Fig. 3 shows simulation results of the passive mode-locking integrating the single-layer graphene and the highly reflective mirror. We can observe the presence of a single pulse per roundtrip time for the injection pumping rate $a_p = 1.1$. The pumping rate is defined as the ratio of the injected current to the threshold value. The pulse is characterized by a peak field intensity of 12.8 MV.m$^{-1}$ and a duration of 56.7 fs. This maximum electric field is higher than the field obtained for the active mode-locking with the same structure parameters Outafat et al. (2022) where the maximum field for a low operation pumping rate has been equal to 4 MV.m$^{-1}$ which gives an enhancement coefficient of 4. The pulse duration has been determined at 100 fs which is almost the double of the value obtained here. This is due to the highly reflective mirror where the light power is totally reflected at this laser facet. The pulse duration is also shortened with the added graphene-based mirror. The related spectrum is represented in Fig. 4 which is centred at a wavelength of $2.5 \mu m$.

The increase of pumping rate $a_p$ until 2 maintains the generation of single pulses per roundtrip as is illustrated in Fig. 5a. The effect of SHB is negligible because the saturable absorber (i.e. graphene layer) favors the propagation of high intensity pulses and absorbs pulse wingdings of low intensities. The effect of SHB outweighs the absorption once increasing pumping from $a_p = 3$. In this case, the intensity of the side peaks of laser pulses becomes strong inducing the apparition of multiple pulses per roundtrip. Fig. 5b describes the output field for $a_p = 3$ where 2 pulses per roundtrip are obtained.
As shown in Fig. 6, the laser spectrum narrows as the pumping current increases. The stability of the mode-locking is affected due to SBH effect.

**Fig. 3** Time evolution of the electric field for a pumping ratio $a_p = 1.1$ a two pulses and b zoom of one pulse
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Fig. 4 Output spectrum of the QCL emitting at 6.2 $\mu$m for $a_p = 1.1$

Fig. 5 Time evolution of the electric field for pumping ratio (a) $a_p = 2$ and (b) $a_p = 3$

Fig. 6 Output spectrum of the QCL emitting at 6.2 $\mu$m for $a_p = 3$
To optimize the structure so that the effect of SHB can be lowered by the absorbing mirror, we move the position of the graphene layer by changing the thickness of SiO$_2$ layer. In Zaugg (2013), the relation between the absorption of graphene and the thickness of SiO$_2$ layer was given by the field intensity enhancement factor $\xi_{ab}$ as following

$$
\xi_{ab}(d_{SiO_2}) \approx \frac{4}{1 + n_{SiO_2}^2 \cot^2 \left( \frac{2\pi}{\lambda_{SiO_2}} n_{SiO_2} d_{SiO_2} \right)}
$$

(11)

We exploit this expression to determine the position that maximizes this factor. For $d_{SiO_2} = \frac{\lambda_{SiO_2}}{4}$, the enhancement factor is $\xi_{ab}(\frac{\lambda_{SiO_2}}{4}) = 4$. This means that the graphene absorption is $\xi_{ab}(d_{SiO_2}) \times 2.3% = 9.2%$. For $d_{SiO_2} = \frac{\lambda_{SiO_2}}{8}$, the absorption is $\xi_{ab}(d_{SiO_2}) \times 2.3% = 2.83%$ while it is close to zero for $d_{SiO_2} = \frac{\lambda_{SiO_2}}{2}$.

The simulations with these SiO$_2$ thicknesses have been done. For the initial thickness $d_{SiO_2} = \frac{\lambda_{SiO_2}}{4}$, a stable mode-locking has been obtained with a peak electric field magnitude equal to 12.8 MV.m$^{-1}$ (Fig. 3). When the thickness of SiO$_2$ is decreased to $\frac{\lambda_{SiO_2}}{8}$, stable mode-locking remains with a peak electric field decreasing to 10.26 MV.m$^{-1}$ and a duration of 51.8 fs as is shown in Fig. 7a. Thus, we can observe the dependence of the pulse peak with graphene location. For $d_{SiO_2} = \frac{\lambda_{SiO_2}}{2}$, the mode-locking becomes unstable and continuous wave operation governs the laser dynamic. The output field of the QCL is represented in Fig. 7b which reveals that the graphene layer does not have any effect.

We then consider the case of a MIR QCL of a vertical radiative transition that is associated to a shorter relaxation times of 5 ps. Simulation results show that the SHB effect is strong due to the short gain recovery time and leads to multiple pulse generation per roundtrip as highlighted in Fig. 8a for $a_p = 1.1$. The intensity of the generated pulses is greater than the saturation intensity of graphene making its role ineffective. It is evident that the result is more pronounced with shorter relaxation time typically in the range 0.5 – 1 ps for high efficient devices. However, as the SHB effect depends on the induced grating of carrier population it can be controlled through the cavity length. Fig. 8b shows a passive mode-locking improvement once the cavity length is reduced to...
1.8 mm because losses are reduced with a shortened cavity. A single pulse per roundtrip is obtained with a peak electric field of 18 MV.m$^{-1}$. The pulse duration is slightly lower with a value of 51 fs. However, further simulations have shown that the reduction of the relaxation time to 1 ps disturbs again the mode-locking stability and shorter cavity length is required for the generation of isolated pulses per roundtrip.

6 Conclusion

We demonstrate in this paper the passive mode-locking capability of MIR QCLs by incorporating a highly reflective graphene-based mirror. The graphene is used here as a saturable absorber layer and the highly reflective mirror is employed to increase the field intensity within the laser cavity. The overlap between the maximal electric field and graphene layer can be controlled through a thin layer of SiO$_2$ material. The QCL modeling is performed thanks to the well-known Maxwell-Bloch equations which are solved using 1D-FDTD method. The graphene and Bragg grating are modeled with Maxwell’s equations according to the nonlinear conductivity of the single-layer graphene related to the intensity-dependent absorption coefficient. The real thickness of the graphene layer has been simulated without increasing the FDTD simulation time by adapting the FDTD scheme to the presence of this thin sheet.

The QCL structure with a diagonal transition is firstly simulated because the effect of SHB is weaker compared to the QCL structure with a vertical transition. Simulation results for $T_1 = 50$ ps have shown a stable passive mode-locking when the pumping rate $a_p$ is less than 3. Beyond this value, a secondary pulse appears and more when the pumping rate increases. However, the location of the graphene layer from the reflective mirror has to be carefully chosen as the graphene layer is more effective when it is placed at the maximal intensity position. We have shown that an optimized location corresponds to a distance of a quarter wavelength $\frac{\lambda}{4}$ between the Bragg mirror and the graphene layer for which the electric field intensity attains the maximum value. Without graphene, the QCL operates in a continuous waves mode which means that the mode-locking fails. For a QCL structure with a vertical transition, SHB effect dominates because of the fast recovery time compared to the cavity roundtrip time. With $T_1 = 5$ ps, multiple pulses per roundtrip are obtained in a
2.6 mm cavity length. However, isolated pulses are obtained by reducing the cavity length to 1.8 mm. Thus, passive mode-locking of the MIR QCL is obtained by integrating a graphene reflective mirror but requires shorter cavity length for $T_1$ in the range $0.5 - 1 \, \text{ps}$.

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**Declarations**

**Competing interests** The authors declare no competing interests.

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