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ABSTRACT

We present a full-wave Maxwell-density matrix simulation tool including c-number stochastic noise terms for the modeling of the spatiotemporal dynamics in active photonic devices, such as quantum cascade lasers and quantum dot structures. The coherent light-matter interaction in such devices plays an important role in the generation of frequency combs and other nonlinear and non-classical optical phenomena. Since the emergence of nonlinear and non-classical features is directly linked to the noise properties, detailed simulations of the noise characteristics are required for the development of low-noise quantum optoelectronic sources. Our semiclassical simulation framework is based on the Lindblad equation for the electron dynamics, coupled with Maxwell's equations for optical propagation in the laser waveguide. Fluctuations arising from the interactions of the optical field and quantum system with their reservoirs are treated within the quantum Langevin theory. Here, the fluctuations are included by adding stochastic c-number terms to the Maxwell-density matrix equations. The implementation in the mbsolve dynamic simulation framework is publicly available.

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I. INTRODUCTION

An optical frequency comb (OFC) describes coherent radiation with a broadband spectrum consisting of discrete, equidistantly spaced optical lines featuring a stable phase relation with low phase noise and low mode partition noise.^{1–3} Typically, such combs are used for measurements of optical frequencies in metrology and sensing, having revolutionized these fields by providing unprecedented accuracy and enabling numerous innovative applications.^{4,5} Promising semiconductor lasers (SCLs) for integrated optical frequency comb technologies in the mid-infrared and terahertz (THz)

regime are quantum dots (QDs),^{6–16} quantum dash lasers,^{17–22} interband cascade lasers,^{23–26} and quantum cascade lasers (QCLs).^{2,27–32} The active gain medium of the aforementioned low-dimensional SCLs provides a large third-order nonlinearity, which gives rise to a broadband four-wave mixing (FWM) process, resulting in mode proliferation.^{2,5} A complex interplay of parametric gain, FWM nonlinearity, chromatic dispersion, and spatial hole burning is essential for frequency comb formation.^{8,18,24,28,33–37} For a better understanding and for improving the laser performance, noise and linewidth characteristics of such devices have been extensively studied, both theoretically and experimentally.^{14,15,35,38–45} Stable and robust OFC

operation requires a narrow beatnote, which is a measure for the amount of amplitude and phase noise of the comb lines. Noise accompanying carrier transport and spontaneous emission noise can, therefore, have a significant impact on the OFC formation and the performance of SCLs.

Significant research efforts are devoted to the generation and deployment of non-classical features in optical and electronic systems.^{46–51} Recently, intensity correlations in QCL harmonic frequency combs (HFCs) have been experimentally investigated to develop a new generation of semiconductor devices for generating light with non-classical properties.⁵² Endowing commercial devices with outstanding quantum features would pave the way for practical and high-performance applications in the field of quantum networks,^{53,54} including quantum computation,^{55,56} quantum communication,⁵⁷ quantum metrology,^{58–60} and quantum simulation.^{61,62} Notably, photonic systems are quite attractive for the investigation and employment of non-classical features, such as the generation of the so-called quantum combs,^{63–65} corresponding to non-classical states of light with a multimode squeezed and/or an entangled output. As the emergence of nonlinear and non-classical features in SCLs is directly linked to the noise properties,^{52,66} the development of low-noise SCL sources based on detailed simulations is an important prerequisite.

For the modeling of the optical dynamics in nano-optoelectronic devices, the Maxwell–Bloch equations are widely used since they form a relatively compact and numerically efficient model and thus allow for spatiotemporal simulations of the laser dynamics over many optical round trips.^{67–69} Here, the Bloch equations are used for simulating the evolution of the quantum system and its coherent light–matter interaction with the optical field in the active medium. In addition, the optical field propagation is treated classically within Maxwell’s equations, where the coupling with the quantum system arises from the macroscopic polarization term.⁶⁹ The density matrix formalism can be extended and adapted by adding further quantized states in addition to the laser levels and tunneling between states.

Within the Maxwell–density matrix equations, the dissipation processes can be modeled on a microscopic level, by including relevant mechanisms such as carrier–carrier and carrier–phonon scattering. Based on the specific Hamiltonian, relaxation and dephasing rates can be derived within the second-order Born–Markov approximation.^{68,70–72} Typically, the dissipation in the Maxwell–density matrix equations is treated by phenomenological dephasing rates and electron lifetimes, which are either empirically chosen or estimated based on experimental data. Alternatively, advanced carrier transport models of different levels of complexity and numerical efficiency can be used to determine the scattering and dephasing rates self-consistently. The ensemble Monte Carlo (EMC) method, for example, is a widely used statistical method for the numerical evaluation of the carrier transport in SCLs, such as QCLs.^{73–76} Since the semiclassical EMC model cannot cover quantum coherence effects, such as resonant tunneling across thick injection barriers, e.g., in THz QCLs, quantum corrections based on the density matrix (DM) formalism have been incorporated.^{77,78} A more general quantum transport approach, which has been applied to the simulation of QCLs, is given by non-equilibrium Green’s functions (NEGFs).^{74,79–82} Both are self-consistent 3D approaches not relying on empirical or fitting parameters as an input and tak-

ing into account intrasubband processes between different kinetic energy states within a subband. In general, the NEGF approach contains the full quantum information of scattering and dephasing effects and is, therefore, superior to semiclassical methods such as the EMC method. However, the numerical simulation of the NEGF solution is expensive, if incoherent scattering within the self-consistent Born approximation is taken into account. To reduce the numerical costs, NEGF simulations are typically executed by neglecting electron–electron scattering beyond the Hartree approximation.⁷⁴

Typically, the Maxwell–density matrix equations are treated in the so-called rotating-wave approximation, where a coarser spatiotemporal grid can be used, and thus, the numerical load is greatly reduced. This approximation is, however, only valid for relatively narrowband (and not too strong) optical fields.^{67,69,83} Meanwhile, for frequency comb sources, a broadband spectrum with a high number of modes is desired, and the QCL even offers the potential for generating spectra extending over a full octave and beyond.⁸⁴ In several studies, dynamical simulations of different nonlinear optical phenomena in nanostructured devices, e.g., QCLs^{37,39,85–93} and QD lasers,^{35,94–97} were conducted. Motivated by these developments, over the recent years, our group has developed the open-source codebase *mbolve* for the numerically efficient simulation of full-wave Maxwell–density matrix-type equations, i.e., without invoking the rotating-wave approximation.^{98–100}

As pointed out above, this work aims to implement noise sources into *mbolve*, enabling a realistic simulation of the noise characteristics in low-dimensional SCLs, which is, for example, important in the context of OFC operation. In a semiclassical framework, noise can generally be included by adding stochastic terms.^{101,102} These are typically numerically implemented by using a pseudorandom number generator producing uncorrelated, Gaussian-distributed random numbers for every grid point.^{103,104} The resulting Maxwell–Bloch equations are then commonly solved numerically with the finite-difference time-domain approach.^{103,104} The magnitude of the stochastic terms can be systematically derived from the quantum Langevin equations,^{105–108} which are then represented by equivalent stochastic *c*-number equations,^{103,109–111} i.e., evolution equations for operator expectation values with additional stochastic terms. The *c*-number Langevin equations have also been used to calculate the intrinsic linewidth and estimate the intensity noise in SCLs.^{40,41,66,88,112–115} Spontaneous emission obviously plays an important role in optoelectronic devices. While the resulting recombination can simply be included by nonlinear rate terms for the carrier occupations,^{95,116} the noise contribution is not included in the Maxwell–Bloch model due to its semiclassical nature. This effect can, however, be considered in terms of a Gaussian white noise source in the optical propagation equation.^{103,117,118} In a different model, dipole fluctuations are also included by adding Langevin noise terms not only to the propagation equation but also to the off-diagonal density matrix elements.^{116,119–123} By virtue of the fluctuation–dissipation theorem, a decay of populations, coherences, or the optical field is generally accompanied by fluctuations, and a Maxwell–Bloch equation model that includes such decay-induced fluctuations has been presented.^{104,124,125} We extend this approach by including incoherent tunneling. From this, we can derive the semiclassical noise terms for our Maxwell–density matrix Langevin approach, which we then incorporate in our open-source tool

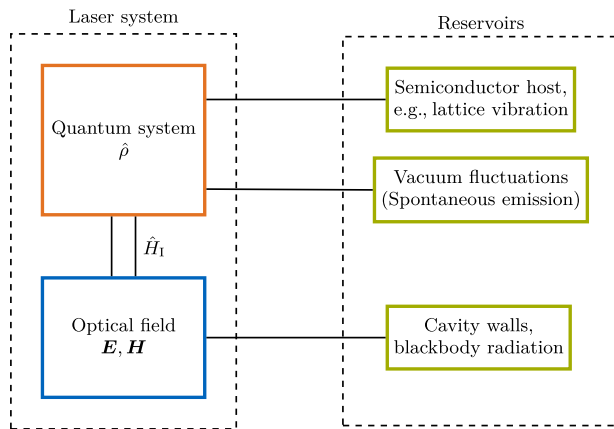


FIG. 1. Schematic illustration of the coupling of a SCL quantum system and a field system and the interaction with their associated reservoirs.

mbolve. Notably, an extension of the stochastic c-number approach to incorporate non-classical effects has been discussed,^{111,126} which might enable a direct investigation of non-classical light generation in the presence of noise.

In detail, this paper is organized as follows: In Sec. II, we calculate the stochastic noise terms of the quantum Langevin equations and derive the generalized description of the Maxwell-density matrix Langevin equations for a quantum-optoelectronic structure, such as a QCL. Our model is schematically illustrated in Fig. 1. Here, the structure is described in terms of the optical field represented by the electric and magnetic field vectors $\vec{E}$, $\vec{H}$ and the density operator $\hat{\rho}$, which are coupled to each other by the interaction Hamiltonian $\hat{H}_I$. For the calculation of drift and diffusion operators in the quantum Langevin theory, we take into account the influence of various reservoirs in our structure. Regarding the quantum system, the reservoir interactions with the semiconductor host, which, for example, includes phonons associated with (longitudinal- and transverse-optical and transverse-acoustic) thermal lattice vibrations, lattice imperfections in the form of impurities (such as dopants), interface roughness or atomic disorder in alloys, as well as vacuum fluctuations arising from spontaneous emission are considered. For the optical field, the interaction with noise arising from thermal radiation (blackbody) entering the active waveguide from the cavity walls can be taken into account by external noise sources.¹²⁷ In this paper, we rather focus on the fluctuations arising from the quantum system and dedicate the investigation of thermal noise influences, which can, for example, play a role in THz QCL active waveguides, to future studies. In Sec. III, we give an overview of the simulation tool and the implementation of the noise terms and validate the model by presenting the simulation results for a superfluorescence (SF) setup.^{104,128} Furthermore, we present the simulation results for a THz QCL harmonic frequency comb and discuss the effects of noise contributions on the comb characteristics. This paper is concluded in Sec. IV.

II. THEORETICAL MODEL

In the following, we restrict ourselves to optoelectronic devices, where the optical field can be well modeled using 1D Maxwell's

equations. This, for example, applies to semiconductor lasers with longitudinally invariant waveguide geometries, where the 3D Maxwell's equations can be reduced to an effective 1D model.⁶⁹ The focus lies on the inclusion of noise arising, for example, from spontaneous emission and fluctuations associated with the electron transport. First, we introduce the quantum Langevin equations using a simple three-level resonant tunneling QCL system. Here, the reservoir variables are eliminated and replaced by drift and fluctuation terms within the Heisenberg equation of motion. The quantum Langevin equations can be transformed into associated c-number Langevin equations, which are then used to derive the stochastic noise terms incorporated into the full-wave Maxwell-density matrix equation system.

A. The quantum Langevin equations

The quantum Langevin equations are introduced by using a simple three-level resonant tunneling QCL system as depicted in Fig. 2.^{87,129} The QCL exploits optical transitions between quantized states in the conduction band of a quantum well heterostructure, where the properties can be controlled by quantum design rather than being determined by the bulk material. This not only applies to the gain and lasing wavelength but also to the nonlinear optical properties, such as FWM. Besides confinement provided by the quantum wells, another important quantum effect is tunneling through the separating barriers, which significantly influences the carrier transport, in addition to the incoherent scattering-induced transitions due to phonons, crystal imperfections, and electron-electron interactions.^{74,77} Regarding non-stationary QCL operation as is the case for OFC emission, coherent light-matter interaction as a further quantum effect plays a significant role in the dynamic behavior, e.g., leading to Rabi flopping,¹³⁰ i.e., oscillations of the electron population between the upper and lower laser levels driven by the resonant optical field. Dephasing due to incoherent scattering has to be taken into account for a realistic description, as it greatly affects tunneling and coherent light-matter interaction.

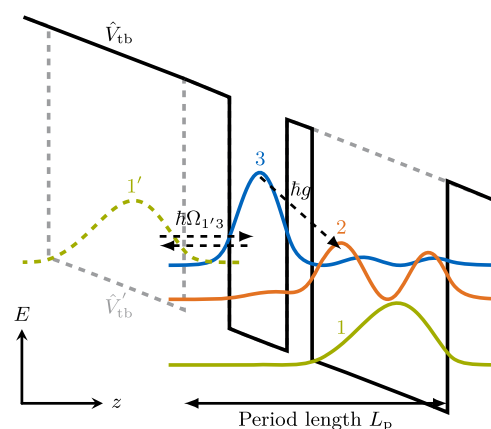


FIG. 2. Schematic conduction band profile and probability densities of a two-well THz QCL structure, where the upper laser level 3 is populated via resonant tunneling from injector level 1'. Depopulation occurs through LO-phonon scattering from lower laser level 2 to depopulation level 1.

For the structure shown in Fig. 2, the lasing transition occurs between the upper laser level $|3\rangle$ and the lower laser level $|2\rangle$. Depopulation takes place via level $|1\rangle$, and electrons are injected from the depopulation level $|1'\rangle$ of the adjacent period via resonant tunneling. The resonant tunneling across thick injection barriers in THz QCLs is treated within the tight-binding model.^{74,76,78,129,131,132} Here, the tunneling between a doublet of states at the thick injection barrier is described by the coupling strength $\Omega_{ij} = -\hbar^{-1} \langle i | \hat{V}_{\text{ext}} - \hat{V}_{\text{tb}} | j \rangle$, with the extended conduction band potential $\hat{V}_{\text{ext}}$ and the tight-binding potential $\hat{V}_{\text{tb}}$. The coupling strengths Ω_{ij} between the states $|3\rangle, |2\rangle, |1\rangle$ within the active period are zero.¹³¹

In general, the QCL system is then described by the reduced system Hamiltonian,^{113,133,134}

$$\begin{aligned} \hat{H}_s &= \hat{H}_F + \hat{H}_0 + \Delta \hat{V}_{\text{tb}} + \hat{H}_I \\ &= \hbar \omega_0 \hat{a}^\dagger \hat{a} + \sum_i \epsilon_i |i\rangle \langle i| - \hbar \Omega_{1'3} (|1'\rangle \langle 3| + |3\rangle \langle 1'|) \\ &\quad + \hbar g (|3\rangle \langle 2| + |2\rangle \langle 3|) (\hat{a} + \hat{a}^\dagger), \end{aligned} \quad (1)$$

where $\hat{H}_F$ is the Hamiltonian of the optical field, $\hat{H}_0$ is the Hamiltonian of the quantum system with $\Delta \hat{V}_{\text{tb}}$ describing the coupling of electron states in two adjacent periods within the tight-binding model, and $\hat{H}_I$ constitutes the interaction Hamiltonian between the quantum system and the optical field. Here, $\hbar$ is the reduced Planck constant, ω_0 is the single mode lasing angular frequency, $\hat{a}^\dagger (\hat{a})$ denotes the creation (annihilation) operator of the radiation field, ϵ_i is the energy of level $|i\rangle$, and $\hbar \Omega_{1'3}$ the anticrossing energy gap between levels $|1'\rangle$ and $|3\rangle$. The dipole coupling constant g can be written in terms of the dipole matrix element, $\mu_{z,23} = q \langle 2 | \hat{z} | 3 \rangle$, as^{105,133}

$$g = -\sqrt{\frac{\omega_0}{2\hbar\epsilon_r\epsilon_0 V_p}} \mu_{z,23}, \quad (2)$$

where ϵ_r is the relative permittivity, ϵ_0 is the vacuum permittivity, and V_p is the volume of each quantum system associated with an active QCL period.

The Heisenberg–Langevin equation of motion for an operator $\hat{A}_\mu(t)$ reads as^{105,107,134,135}

$$\begin{aligned} \partial_t \hat{A}_\mu(t) &= -i\hbar^{-1} [\hat{A}_\mu(t), \hat{H}_s(t)] + \hat{D}_\mu(t) + \hat{F}_\mu(t) \\ &= \hat{M}_\mu(t) + \hat{F}_\mu(t). \end{aligned} \quad (3)$$

Here, the drift operator $\hat{D}_\mu(t)$ and the fluctuation operator $\hat{F}_\mu(t)$ account for the influence of the reservoirs on the system. $[\cdot, \cdot]$ denotes the commutator $[\hat{X}, \hat{Y}] = \hat{X}\hat{Y} - \hat{Y}\hat{X}$. For the drift operator $\hat{D}_\mu$, we can, under the Markovian approximation, write^{134,135}

$$\hat{D}_\mu = -\sum_{ij} \delta(\omega_i, -\omega_j) \{ [\hat{A}_\mu, \hat{Q}_i] \hat{Q}_j w_{ij}^+ - \hat{Q}_j [\hat{A}_\mu, \hat{Q}_i] w_{ji}^- \}, \quad (4)$$

where $w^\pm$ are the reservoir spectral densities and $\hat{Q}_i$ is a function of system operators. For a detailed description and derivation of this

theory together with the calculation examples for specific operators $\hat{A}_\mu$, we refer to Refs. 134 and 135.

The reservoir average of the fluctuation operator vanishes, $\langle \hat{F}_\mu^\dagger \rangle_R = \langle \hat{F}_\mu \rangle_R = 0$. The diffusion coefficient for a Markovian system is defined as

$$2\langle \hat{D}_{\mu\nu}(t) \rangle_R \delta(t - t') = \langle \hat{F}_\mu(t) \hat{F}_\nu(t') \rangle_R \quad (5)$$

and can be calculated by applying the fluctuation–dissipation theorem. Here, the δ -function indicates the very short memory period of the reservoirs. The *generalized Einstein relation* for the calculation of the diffusion coefficient is given by^{105,135,136}

$$2\langle \hat{D}_{\mu\nu}(t) \rangle_R = \partial_t \langle \hat{A}_\mu(t) \hat{A}_\nu(t) \rangle_R - \langle \hat{M}_\mu(t) \hat{A}_\nu(t) \rangle_R - \langle \hat{A}_\mu(t) \hat{M}_\nu(t) \rangle_R. \quad (6)$$

From Eq. (3) together with Eqs. (1) and (4), the quantum Langevin equations for the three-level QCL quantum system can be derived. Therefore, we introduce the electron population operators $\hat{\sigma}_{ii} = |i\rangle \langle i|$ and the coherence operators $\hat{\sigma}_{ij} = |i\rangle \langle j|$. The term $\hat{\sigma}_{32} \hat{a}^\dagger$ describes the creation of a photon accompanied by an electron transition from the lower to the higher lying energy level, and $\hat{\sigma}_{23} \hat{a}$ describes the annihilation of a photon accompanied by an electron transition from the higher to the lower lying energy level. At this point, we drop these counterrotating energy non-conserving terms in the interaction Hamiltonian $\hat{H}_I$ as in the commonly used rotating wave approximation.^{105,133} This simplifies the following calculations of the noise terms. A more complete calculation should also include more than one mode of the optical field in the system Hamiltonian. However, these concessions do not affect the form of the specific noise terms that are ultimately used in our simulations. The corresponding equations of motion are given by

$$\partial_t \hat{a}(t) = -i\omega_0 \hat{a}(t) - \frac{\kappa}{2} \hat{a}(t) - g \hat{\sigma}_{23} + \hat{F}_a(t), \quad (7a)$$

$$\begin{aligned} \partial_t \hat{\sigma}_{23}(t) &= -\frac{i}{\hbar} \Delta_{32} \hat{\sigma}_{23}(t) - \gamma_{23} \hat{\sigma}_{23}(t) + i\Omega_{1'3} \hat{\sigma}_{21'}(t) \\ &\quad + ig [\hat{\sigma}_{33}(t) - \hat{\sigma}_{22}(t)] \hat{a}(t) + \hat{F}_{23}(t), \end{aligned} \quad (7b)$$

$$\begin{aligned} \partial_t \hat{\sigma}_{31'}(t) &= -\frac{i}{\hbar} \Delta_{1'3} \hat{\sigma}_{31'}(t) - \gamma_{1'3} \hat{\sigma}_{31'}(t) + i\Omega_{1'3} [\hat{\sigma}_{33}(t) - \hat{\sigma}_{1'1'}(t)] \\ &\quad + ig \hat{\sigma}_{21'}(t) \hat{a}^\dagger(t) + \hat{F}_{31'}(t), \end{aligned} \quad (7c)$$

$$\begin{aligned} \partial_t \hat{\sigma}_{21'}(t) &= -\frac{i}{\hbar} \Delta_{1'2} \hat{\sigma}_{21'}(t) - \gamma_{1'2} \hat{\sigma}_{21'}(t) + i\Omega_{1'3} \hat{\sigma}_{23}(t) \\ &\quad + ig \hat{\sigma}_{31'}(t) \hat{a}(t) + \hat{F}_{21'}(t), \end{aligned} \quad (7d)$$

$$\begin{aligned} \partial_t \hat{\sigma}_{33}(t) &= -\frac{1}{\tau_3} \hat{\sigma}_{33}(t) + r_{32} \hat{\sigma}_{22}(t) + r_{31'} \hat{\sigma}_{1'1'}(t) \\ &\quad + ig [\hat{a}^\dagger(t) \hat{\sigma}_{23}(t) - \hat{a}(t) \hat{\sigma}_{23}^\dagger(t)] \\ &\quad - i\Omega_{1'3} [\hat{\sigma}_{31'}^\dagger(t) - \hat{\sigma}_{31'}(t)] + \hat{F}_{33}(t), \end{aligned} \quad (7e)$$

$$\partial_t \hat{\sigma}_{22}(t) = r_{23} \hat{\sigma}_{33}(t) - \frac{1}{\tau_2} \hat{\sigma}_{22}(t) + r_{21'} \hat{\sigma}_{1'1'}(t) + i g [\hat{a}(t) \hat{\sigma}_{23}^\dagger(t) - \hat{a}^\dagger(t) \hat{\sigma}_{23}(t)] + \hat{F}_{22}(t), \quad (7f)$$

$$\partial_t \hat{\sigma}_{1'1'}(t) = r_{1'3} \hat{\sigma}_{33}(t) + r_{1'2} \hat{\sigma}_{22}(t) - \frac{1}{\tau_{1'}} \hat{\sigma}_{1'1'}(t) - i \Omega_{1'3} [\hat{\sigma}_{31'}(t) - \hat{\sigma}_{31'}^\dagger(t)] + \hat{F}_{1'1'}(t), \quad (7g)$$

where κ is the cavity decay rate, Δ_{ij} denotes the energy separation between levels $|i\rangle$ and $|j\rangle$, $\tau_i^{-1} = \sum_{j \neq i} r_{ji}$ is the inverse population lifetime, $r_{ij, i \neq j}$ represents the scattering rate from level j to i , and

$$\gamma_{ij} = \frac{1}{2} \left(\frac{1}{\tau_i} + \frac{1}{\tau_j} \right) + \gamma_{ij,p} \quad (8)$$

is the dephasing rate. Here, $\gamma_{ij,p}$ is the pure dephasing rate, which for QCLs mainly consists of elastic scattering contributions due to impurity and interface roughness.⁷⁷ The equivalent equations for $\hat{a}^\dagger(t)$, $\hat{\sigma}_{32}(t)$, $\hat{\sigma}_{1'3}(t)$, and $\hat{\sigma}_{1'2}(t)$ are given by the Hermitian conjugates of Eqs. 7(a)–7(d).

Using Eq. (6), we can calculate the second-order correlation function relevant for the polarization operator as

$$\begin{aligned} \langle \hat{F}_{23}^\dagger(t) \hat{F}_{23}(t') \rangle_R &= 2 \langle \hat{D}_{3223}(t) \rangle_R \delta(t-t') \\ &= \left[\partial_t \langle \hat{\sigma}_{23}^\dagger(t) \hat{\sigma}_{23}(t) \rangle_R - \langle \hat{M}_{23}^\dagger(t) \hat{\sigma}_{23}(t) \rangle_R \right. \\ &\quad \left. - \langle \hat{\sigma}_{23}^\dagger(t) \hat{M}_{23}(t) \rangle_R \right] \delta(t-t') \\ &= \left[\left(2\gamma_{23} - \frac{1}{\tau_3} \right) \langle \hat{\sigma}_{33}(t) \rangle_R + r_{32} \langle \hat{\sigma}_{22}(t) \rangle_R \right. \\ &\quad \left. + r_{31'} \langle \hat{\sigma}_{1'1'}(t) \rangle_R \right] \delta(t-t'). \end{aligned} \quad (9)$$

A detailed derivation of the diffusion coefficient $2 \langle \hat{D}_{3223}(t) \rangle_R$ can be found in Appendix A.

With the same procedure, we determine the other non-vanishing second-order correlation functions,^{134,135}

$$\langle \hat{F}_a^\dagger(t) \hat{F}_a(t') \rangle_R = \kappa n_{\text{th}}(\omega_0) \delta(t-t'), \quad (10a)$$

$$\langle \hat{F}_a(t) \hat{F}_a^\dagger(t') \rangle_R = \kappa (n_{\text{th}}(\omega_0) + 1) \delta(t-t'), \quad (10b)$$

$$\begin{aligned} \langle \hat{F}_{23}(t) \hat{F}_{23}^\dagger(t') \rangle_R &= \left[r_{23} \langle \hat{\sigma}_{33}(t) \rangle_R + \left(2\gamma_{23} - \frac{1}{\tau_2} \right) \right. \\ &\quad \left. \times \langle \hat{\sigma}_{22}(t) \rangle_R + r_{21'} \langle \hat{\sigma}_{1'1'}(t) \rangle_R \right] \delta(t-t'), \end{aligned} \quad (10c)$$

$$\begin{aligned} \langle \hat{F}_{31'}^\dagger(t) \hat{F}_{31'}(t') \rangle_R &= \left[r_{1'3} \langle \hat{\sigma}_{33}(t) \rangle_R + r_{1'2} \langle \hat{\sigma}_{22}(t) \rangle_R \right. \\ &\quad \left. + \left(2\gamma_{1'3} - \frac{1}{\tau_{1'}} \right) \langle \hat{\sigma}_{1'1'}(t) \rangle_R \right] \delta(t-t'), \end{aligned} \quad (10d)$$

$$\begin{aligned} \langle \hat{F}_{31'}(t) \hat{F}_{31'}^\dagger(t') \rangle_R &= \left[\left(2\gamma_{1'3} - \frac{1}{\tau_3} \right) \langle \hat{\sigma}_{33}(t) \rangle_R \right. \\ &\quad \left. + r_{32} \langle \hat{\sigma}_{22}(t) \rangle_R + r_{31'} \langle \hat{\sigma}_{1'1'}(t) \rangle_R \right] \delta(t-t'), \end{aligned} \quad (10e)$$

$$\begin{aligned} \langle \hat{F}_{21'}^\dagger(t) \hat{F}_{21'}(t') \rangle_R &= \left[r_{1'3} \langle \hat{\sigma}_{33}(t) \rangle_R + r_{1'2} \langle \hat{\sigma}_{22}(t) \rangle_R \right. \\ &\quad \left. + \left(2\gamma_{1'2} - \frac{1}{\tau_{1'}} \right) \langle \hat{\sigma}_{1'1'}(t) \rangle_R \right] \delta(t-t'), \end{aligned} \quad (10f)$$

$$\begin{aligned} \langle \hat{F}_{21'}(t) \hat{F}_{21'}^\dagger(t') \rangle_R &= \left[r_{23} \langle \hat{\sigma}_{33}(t) \rangle_R + \left(2\gamma_{1'2} - \frac{1}{\tau_2} \right) \right. \\ &\quad \left. \times \langle \hat{\sigma}_{22}(t) \rangle_R + r_{21'} \langle \hat{\sigma}_{1'1'}(t) \rangle_R \right] \delta(t-t'), \end{aligned} \quad (10g)$$

$$\begin{aligned} \langle \hat{F}_{33}(t) \hat{F}_{33}(t') \rangle_R &= \left[\frac{1}{\tau_3} \langle \hat{\sigma}_{33}(t) \rangle_R + r_{32} \langle \hat{\sigma}_{22}(t) \rangle_R \right. \\ &\quad \left. + r_{31'} \langle \hat{\sigma}_{1'1'}(t) \rangle_R \right] \delta(t-t'), \end{aligned} \quad (10h)$$

$$\begin{aligned} \langle \hat{F}_{22}(t) \hat{F}_{22}(t') \rangle_R &= \left[r_{23} \langle \hat{\sigma}_{33}(t) \rangle_R + \frac{1}{\tau_2} \langle \hat{\sigma}_{22}(t) \rangle_R \right. \\ &\quad \left. + r_{21'} \langle \hat{\sigma}_{1'1'}(t) \rangle_R \right] \delta(t-t'), \end{aligned} \quad (10i)$$

$$\begin{aligned} \langle \hat{F}_{1'1'}(t) \hat{F}_{1'1'}(t') \rangle_R &= \left[r_{1'3} \langle \hat{\sigma}_{33}(t) \rangle_R + r_{1'2} \langle \hat{\sigma}_{22}(t) \rangle_R \right. \\ &\quad \left. + \frac{1}{\tau_{1'}} \langle \hat{\sigma}_{1'1'}(t) \rangle_R \right] \delta(t-t'). \end{aligned} \quad (10j)$$

Here, $n_{\text{th}}(\omega_0) = \left[\exp \left(\frac{\hbar \omega_0}{k_B T} \right) - 1 \right]^{-1}$ is the number of thermal photons in the lasing mode at temperature T , where k_B denotes the Boltzmann constant.

B. The c-number Langevin equations

In order to derive the stochastic noise terms for the semiclassical Maxwell-density matrix equations, the operator Langevin equations have to be converted into the associated c-number Langevin equations.

The quantum Langevin equation for the operator $\hat{A}_\mu(t)$ in chosen order is given by

$$\partial_t \hat{A}_\mu(t) = -i \hbar^{-1} [\hat{A}_\mu(t), \hat{H}_s(t)]^c + \hat{D}_\mu^c(t) + \hat{F}_\mu^c(t), \quad (11)$$

where we make use of the commutation relation $\hat{A}_\mu^\dagger \hat{A}_\nu = \hat{A}_\nu \hat{A}_\mu^\dagger - [\hat{A}_\nu, \hat{A}_\mu^\dagger]$ to bring the equation into the chosen order. We use the superscript c to highlight that we have put the operators in chosen order. To explain this formulation in more detail, we use the fluctuation operator $\hat{F}_\mu(t)$ as an example, but the following description holds for the other operators in the same way. For a chosen order $\hat{A}_1, \dots, \hat{A}_\mu$, we can write

$$\hat{F}_\mu = \hat{F}_\mu^c(\hat{A}_1, \dots, \hat{A}_\mu), \quad (12)$$

where the fluctuation operator $\hat{F}_\mu^c$ in the chosen order is, of course, equal to the fluctuation operator $\hat{F}_\mu$ in the original order. The associated c-number fluctuation term $F_\mu^c(\hat{A}_1, \dots, \hat{A}_\mu)$ is obtained using c-numbers A_ν . By defining a linear chosen ordering operator $\hat{C}$, we can further indicate^{134,137}

$$\hat{F}_\mu^c(\hat{A}_1, \dots, \hat{A}_\mu) = \hat{C}(F_\mu^c(A_1, \dots, A_\mu)), \quad (13)$$

where the operator $\hat{C}$ has the function of replacing each A_ν by the corresponding operator $\hat{A}_\nu$ and bringing all terms into chosen order.

If we now convert the quantum Langevin equation into the equivalent c-number Langevin equation, we may write

$$\begin{aligned} \partial_t A_\mu(t) &= L_\mu(t) + D_\mu(t) + F_\mu^c(t) \\ &= M_\mu(t) + F_\mu^c(t), \end{aligned} \quad (14)$$

with $L_\mu(t)$ being the coherent term corresponding to the commutation of $\hat{A}_\mu(t)$ with the system Hamiltonian $\hat{H}_s$ and $D_\mu(t)$ denoting the drift term. Furthermore, by the use of Eq. (14), we obtain the c-number equation,

$$\begin{aligned} \partial_t [A_\mu(t)A_\nu(t)] &= A_\mu(t)\partial_t A_\nu(t) + A_\nu(t)\partial_t A_\mu(t) \\ &= A_\mu(t)M_\nu(t) + A_\nu(t)M_\mu(t) \\ &\quad + A_\mu(t)F_\nu^c(t) + A_\nu(t)F_\mu^c(t). \end{aligned} \quad (15)$$

In analogy to the reservoir average in the operator case, we may write the c-number equation as

$$\partial_t \langle A_\mu(t)A_\nu(t) \rangle_R = \langle A_\mu(t)M_\nu(t) \rangle_R + \langle A_\nu(t)M_\mu(t) \rangle_R + 2\langle D_{\mu\nu}(t) \rangle_R, \quad (16)$$

where we can make use of the following relation under the Markovian approximation:^{105,134}

$$2\langle D_{\mu\nu}(t) \rangle_R = \langle A_\mu(t)F_\nu^c(t) + A_\nu(t)F_\mu^c(t) \rangle_R. \quad (17)$$

The diffusion coefficients in the c-number Langevin equations may differ from the ones in the quantum Langevin equations, as the c-numbers commute, whereas the operators do not. By requiring the equivalence of Eqs. (16) and (6) in both c-number and quantum Langevin theory, it can be shown that, in general,

$$2\langle \hat{D}_{\mu\nu}(t) \rangle_R \neq 2\langle \hat{C}(D_{\mu\nu}(t)) \rangle_R. \quad (18)$$

By taking our chosen ordered operator representation of the system operators $\hat{a}^\dagger, \hat{\sigma}_{23}^\dagger, \hat{\sigma}_{31'}^\dagger, \hat{\sigma}_{21'}^\dagger, \hat{\sigma}_{33}, \hat{\sigma}_{22}, \hat{\sigma}_{1'1'}, \hat{\sigma}_{21'}, \hat{\sigma}_{31'}, \hat{\sigma}_{23}, \hat{a}$, we obtain the corresponding c-numbers $a^*, \sigma_{23}^*, \sigma_{31'}^*, \sigma_{21'}^*, \sigma_{33}, \sigma_{22}, \sigma_{1'1'}, \sigma_{21'}, \sigma_{31'}, \sigma_{23}, a$. Therefore, we derive the c-number second-order moments and obtain for the populations differing terms compared to the operator case,

$$\begin{aligned} \langle F_{33}(t)F_{33}(t') \rangle_R &= \left\{ \frac{1}{\tau_3} \langle \sigma_{33}(t) \rangle_R + r_{32} \langle \sigma_{22}(t) \rangle_R + r_{31'} \langle \sigma_{1'1'}(t) \rangle_R \right. \\ &\quad + i g [\langle a^*(t) \sigma_{23}(t) \rangle_R - \langle a(t) \sigma_{23}^*(t) \rangle_R] \\ &\quad \left. + i \Omega_{1'3} [\langle \sigma_{31'}^*(t) \rangle_R - \langle \sigma_{31'}(t) \rangle_R] \right\} \delta(t-t'), \end{aligned} \quad (19a)$$

$$\begin{aligned} \langle F_{22}(t)F_{22}(t') \rangle_R &= \left\{ r_{23} \langle \sigma_{33}(t) \rangle_R + \frac{1}{\tau_2} \langle \sigma_{22}(t) \rangle_R + r_{21'} \langle \sigma_{1'1'}(t) \rangle_R \right. \\ &\quad \left. + i g [\langle a^*(t) \sigma_{23}(t) \rangle_R - \langle a(t) \sigma_{23}^*(t) \rangle_R] \right\} \delta(t-t'), \end{aligned} \quad (19b)$$

$$\begin{aligned} \langle F_{1'1'}(t)F_{1'1'}(t') \rangle_R &= \left\{ r_{1'3} \langle \sigma_{33}(t) \rangle_R + r_{1'2} \langle \sigma_{22}(t) \rangle_R \right. \\ &\quad + \frac{1}{\tau_{1'}} \langle \sigma_{1'1'}(t) \rangle_R + i \Omega_{1'3} [\langle \sigma_{31'}^*(t) \rangle_R \\ &\quad \left. - \langle \sigma_{31'}(t) \rangle_R] \right\} \delta(t-t'). \end{aligned} \quad (19c)$$

As an example, we provide a detailed derivation of the diffusion term from Eq. (19a) in Appendix A. In addition, we obtain diffusion coefficients absent in the quantum Langevin theory, e.g.,

$$\langle F_{23}(t)F_{23}(t') \rangle_R = 2i g \langle a(t) \sigma_{23}(t) \rangle_R \delta(t-t'), \quad (20a)$$

$$\langle F_{31'}(t)F_{31'}(t') \rangle_R = -2i \Omega_{1'3} \langle \sigma_{31'}(t) \rangle_R \delta(t-t'). \quad (20b)$$

The complete diffusion matrix $\mathbf{D}(\vec{A}, t)$, including all relevant cross correlation terms of the three-level QCL system with the c-number vector $\vec{A} = [a^*, a, \sigma_{23}^*, \sigma_{31'}^*, \sigma_{21'}^*, \sigma_{33}, \sigma_{22}, \sigma_{1'1'}, \sigma_{21'}, \sigma_{31'}, \sigma_{23}]^T$, is illustrated in Appendix B.

In the literature,^{138,139} it has been shown that a set of Ito stochastic differential equations (SDEs) can be derived for the given c-number vector and can serve as an efficient basis for numerical simulations. The equivalent Ito SDEs to the Langevin theory are given by

$$\partial_t \vec{A}(t) = \vec{M}(t) + \vec{F}(t) = \vec{M}(t) + \mathbf{B}(\vec{A}, t) \cdot \vec{\xi}(t), \quad (21)$$

where $\vec{\xi}(t)$ is a vector with real, independent Gaussian random numbers. Here, a semi-definite and symmetric diffusion matrix $\mathbf{D}(\vec{A}, t)$ is required, which can then be factorized into the following form:^{111,138,140}

$$\mathbf{D}(\vec{A}, t) = \mathbf{B}(\vec{A}, t) \mathbf{B}^T(\vec{A}, t), \quad (22)$$

where the derived noise matrix $\mathbf{B}(\vec{A}, t)$ is not necessarily symmetric.

To calculate the full noise matrix $\mathbf{B}(\vec{A}, t)$ for the three-level QCL system, we can divide the diffusion matrix $\mathbf{D}(\vec{A}, t)$ into four different submatrices, where a correlation between the corresponding terms is identified. The given subvector $\vec{A}_\nu$ and the submatrices $\mathbf{B}_\nu(\vec{A}_\nu, t)$ and $\mathbf{D}_\nu(\vec{A}_\nu, t)$ are illustrated in Table I. Here, we include correlations between three states by taking into account a tunneling transition followed by an optical transition. This leads to a substantial extension of the initially derived quantum theory of propagation of non-classical radiation in a two-level system¹¹¹ and is of essential importance for the description of quantum fluctuations in THz QCL systems, where electron transport across thick barriers is mediated by tunneling between closely aligned energy levels. A detailed symbolic derivation of the noise submatrices $\mathbf{B}_\nu(\vec{A}_\nu, t)$ and the resulting noise matrix $\mathbf{B}(\vec{A}, t)$ for the three-level QCL system can be found in the GitHub project mbsolve.¹⁴¹

TABLE I. Division of the diffusion matrix into submatrices $\mathbf{D}_v(\vec{A}_v, t)$ and the corresponding c-number subvectors $\vec{A}_v$ and noise submatrices $\mathbf{B}_v(\vec{A}_v, t)$. In order to preserve the physical properties of the quantum system description, we have to interpret noise matrices $\mathbf{B}_v(\vec{A}_v, t)$ differently for occupation and coherence terms. The differing matrix expressions for the coherence terms are highlighted in red.

Subvector $\vec{A}_v$	Submatrix $\mathbf{B}_v(\vec{A}_v, t)$	Submatrix $\mathbf{D}_v(\vec{A}_v, t)$
$\begin{bmatrix} \sigma_{23}^* \\ \sigma_{33} \\ \sigma_{22} \end{bmatrix}, \begin{bmatrix} \sigma_{31}^* \\ \sigma_{33} \\ \sigma_{22} \end{bmatrix}, \begin{bmatrix} \sigma_{21}^* \\ \sigma_{33} \\ \sigma_{22} \end{bmatrix},$ $\begin{bmatrix} \sigma_{23}^* \\ \sigma_{22} \\ \sigma_{1'1'}^* \end{bmatrix}, \begin{bmatrix} \sigma_{31}^* \\ \sigma_{33} \\ \sigma_{22} \end{bmatrix}, \begin{bmatrix} \sigma_{21}^* \\ \sigma_{33} \\ \sigma_{22} \end{bmatrix}$	$\begin{bmatrix} a & -ia \\ -b & -ic \\ b & ic \\ a^* & ia^* \end{bmatrix}$	$\begin{bmatrix} 0 & -ab - ac & ab + ac & 2 a ^2 \\ -ab - ac & b^2 - c^2 & -b^2 + c^2 & -a^*b + a^*c \\ ab + ac & -b^2 + c^2 & b^2 - c^2 & a^*b - a^*c \\ 2 a ^2 & -a^*b + a^*c & a^*b - a^*c & 0 \end{bmatrix}$
$\begin{bmatrix} \sigma_{33} \\ \sigma_{22} \\ \sigma_{23}^* \end{bmatrix}, \begin{bmatrix} \sigma_{33} \\ \sigma_{1'1'}^* \\ \sigma_{31}^* \end{bmatrix}, \begin{bmatrix} \sigma_{22} \\ \sigma_{1'1'}^* \\ \sigma_{31}^* \end{bmatrix}$	$\begin{bmatrix} a \\ -a \end{bmatrix}, \begin{bmatrix} a \\ a^* \end{bmatrix}$	$\begin{bmatrix} a^2 & -a^2 \\ -a^2 & a^2 \end{bmatrix}, \begin{bmatrix} a^2 & a ^2 \\ a ^2 & (a^*)^2 \end{bmatrix}$
$\begin{bmatrix} \sigma_{23}^* \\ \sigma_{31}^* \\ \sigma_{31'}^* \\ \sigma_{23} \end{bmatrix}, \begin{bmatrix} \sigma_{23}^* \\ \sigma_{21}^* \\ \sigma_{21'}^* \\ \sigma_{23} \end{bmatrix}, \begin{bmatrix} \sigma_{23}^* \\ \sigma_{21}^* \\ \sigma_{21'}^* \\ \sigma_{23} \end{bmatrix}$	$\begin{bmatrix} a & ia \\ b & -ib \\ c & ic \\ a^* & -ia^* \end{bmatrix}$	$\begin{bmatrix} 0 & 2ab & 0 & 2 a ^2 \\ 2ab & 0 & 2bc & 0 \\ 0 & 2bc & 0 & 2a^*c \\ 2 a ^2 & 0 & 2a^*c & 0 \end{bmatrix}$
$\begin{bmatrix} \sigma_{23}^* \\ \sigma_{23} \end{bmatrix}, \begin{bmatrix} \sigma_{31}^* \\ \sigma_{31'}^* \end{bmatrix}, \begin{bmatrix} \sigma_{21}^* \\ \sigma_{21'}^* \end{bmatrix}$	$\begin{bmatrix} a & ia \\ a & -ia \end{bmatrix}$	$\begin{bmatrix} 0 & 2a^2 \\ 2a^2 & 0 \end{bmatrix}$

By calculating the operator expectation value in the Schrödinger picture, we can demonstrate that the c-numbers representing the quantum system can be replaced by the density matrix elements $\rho_{23}, \rho_{31}, \rho_{21}, \rho_{33}, \rho_{22}, \rho_{1'1'}, \rho_{1'2}, \rho_{1'3}, \rho_{32}$. The expectation value can be written as

$$\begin{aligned} \langle \hat{\sigma}_{ij} \rangle &= \text{Tr} \{ |i\rangle \langle j| \hat{\rho}(t) \} \\ &= \text{Tr} \left\{ |i\rangle \langle j| \sum_{j', i'} \rho_{j' i'}(t) |j'\rangle \langle i'| \right\} = \rho_{ji}(t). \end{aligned} \quad (23)$$

Furthermore, we can write the interaction Hamiltonian $\hat{H}_I$ of the quantum system and the optical field as

$$\hat{H}_I = -\hat{\mu}_z \hat{E}_z = -\vec{\mu}_{z,23} \hat{E}_z (\hat{\sigma}_{32} + \hat{\sigma}_{23}), \quad (24)$$

where the electrical field operator $\hat{E}_z$ is defined as

$$\hat{E}_z = \sqrt{\frac{\omega_0}{2\hbar\epsilon_r\epsilon_0 V_p}} (\hat{a}^\dagger + \hat{a}) \vec{e}_z. \quad (25)$$

Here, $\vec{e}_z$ denotes the unit vector in the z -direction. For devices in which the intraband transitions between quantized states occur within the conduction band, e.g., the QCL quantum well heterostructure, only the dipole matrix element $\vec{\mu}_z$ for polarization in the growth direction z is nonzero and relevant.

C. Generalized Maxwell-density matrix Langevin equations in 1D

In the following, we derive the generalized Maxwell-density matrix Langevin equations with additional microscopic fluctuation

terms and characterize the influence of spontaneous emission noise on the optical field evolution. For the description of the coherent carrier dynamics and the incoherent relaxation processes, as well as the interaction with the classical optical field, the generalized full-wave Maxwell-density matrix equations constitute a compact semiclassical model. By combining it with the Langevin approach, the microscopic noise characteristics can be fully taken into account. Here, the carrier dynamics in a SCL system are described in the density matrix formulation using the Lindblad equation,

$$\partial_t \hat{\rho} = -i\hbar^{-1} [\hat{H}_0 + \Delta \hat{V}_{\text{tb}} + \hat{H}_I, \hat{\rho}] + \mathcal{D}(\hat{\rho}) + \mathcal{F}(\hat{\rho}), \quad (26)$$

which is coupled to Maxwell's equations in one dimension,

$$\partial_t E_z = \epsilon^{-1} (-\sigma E_z - \partial_t P_{z,\text{class}} - \Gamma \partial_t P_{z,\text{qm}} + \partial_x H_y), \quad (27a)$$

$$\partial_t H_y = \mu^{-1} \partial_x E_z, \quad (27b)$$

where $\mathcal{D}(\hat{\rho})$ is the dissipation superoperator, $\mathcal{F}(\hat{\rho})$ is an additional Langevin fluctuation superoperator, and the other operators have their usual meanings. The permittivity is given by the product $\epsilon = \epsilon_0 \epsilon_r$, σ is the material conductivity, μ is the permeability, and the confinement factor $\Gamma \in [0, 1]$ gives the spatial overlap of the transverse optical field mode with the quantum system. As we focus in this work on optoelectronic devices with an invariant transverse field distribution, the reduction to a one-dimensional model for optical propagation in the waveguide is justified.⁶⁹ The Lindblad equation is the general form of a time-local and Markovian linear master

equation for a quantum system, described by its completely positive trace-preserving density matrix, interacting with an environment. Obviously, the conventional Bloch equations, corresponding to a two-level system, describe the interaction of the laser levels with the optical field E_z and constitute a special case of the Lindblad equation given in Eq. (26). The interaction with the environment is here modeled by scattering and dephasing rates, r_{ij} and γ_{ij} . Further levels can be considered in Eq. (26), and additional effects, such as tunneling, are included in the Hamiltonian. Moreover, quantum fluctuations are considered in the model given by Eq. (26) by adding a suitable Langevin fluctuation superoperator $\mathcal{F}$. Maxwell's equations capture the optical propagation through the waveguide resonator, where the coupling with the quantum system is described by the macroscopic polarization $P_{z,qm}$ arising from the contributions of the dipole matrix elements. The expectation value of the dipole moment operator $\hat{\mu}_z$ is calculated by averaging over a large ensemble of quantum systems within an adequate volume V_p around the position z , and we can write for the macroscopic polarization

$$P_{z,qm} = n_{3D} \text{Tr}\{\hat{\mu}_z \hat{\rho}\} = n_{3D} (\mu_{z,23} \rho_{32} + \mu_{z,32} \rho_{23}) \\ = n_{3D} \mu_{z,23} (\rho_{32} + \rho_{23}), \quad (28)$$

where n_{3D} is the carrier number density. The two classical contributions, $P_{z,class} = \epsilon_0 \chi E_z$ and σE_z , account for the polarization caused by bulk and waveguide dispersion as well as material losses.⁹⁹

Finally, the update equations of the density matrix elements for the QCL system depicted in Fig. 2 can be written as

$$\partial_t \rho_{32}(t) = -\frac{i}{\hbar} \Delta_{32} \rho_{32}(t) - \gamma_{23} \rho_{32}(t) + i \Omega_{1'3} \rho_{1'2}(t) \\ + \frac{i}{\hbar} \mu_{z,23} E_z [\rho_{33}(t) - \rho_{22}(t)] + F_{23}(t), \quad (29a)$$

$$\partial_t \rho_{1'3}(t) = -\frac{i}{\hbar} \Delta_{1'3} \rho_{1'3}(t) - \gamma_{1'3} \rho_{1'3}(t) + i \Omega_{1'3} [\rho_{33}(t) \\ - \rho_{1'1'}(t)] + \frac{i}{\hbar} \mu_{z,23} E_z \rho_{1'2}(t) + F_{31'}(t), \quad (29b)$$

$$\partial_t \rho_{1'2}(t) = -\frac{i}{\hbar} \Delta_{1'2} \rho_{1'2}(t) - \gamma_{1'2} \rho_{1'2}(t) + i \Omega_{1'3} \rho_{32}(t) \\ + \frac{i}{\hbar} \mu_{z,23} E_z \rho_{1'3}(t) + F_{21'}(t), \quad (29c)$$

$$\partial_t \rho_{33}(t) = -\frac{1}{\tau_3} \rho_{33}(t) + r_{32} \rho_{22}(t) + r_{31'} \rho_{1'1'}(t) \\ - 2\hbar^{-1} \mu_{z,23} E_z \Im(\rho_{32}(t)) \\ - 2\Omega_{1'3} \Im(\rho_{1'3}(t)) + F_{33}(t), \quad (29d)$$

$$\partial_t \rho_{22}(t) = r_{23} \rho_{33}(t) - \frac{1}{\tau_2} \rho_{22}(t) + r_{21'} \rho_{1'1'}(t) \\ + 2\hbar^{-1} \mu_{z,23} E_z \Im(\rho_{32}(t)) + F_{22}(t), \quad (29e)$$

$$\partial_t \rho_{1'1'}(t) = r_{1'3} \rho_{33}(t) + r_{1'2} \rho_{22}(t) - \frac{1}{\tau_{1'}} \rho_{1'1'}(t) \\ + 2\Omega_{1'3} \Im(\rho_{1'3}(t)) + F_{1'1'}(t). \quad (29f)$$

Via the macroscopic polarization $P_{z,qm}$, the quantum fluctuations added to the coherence term of Eq. (29a) have an influence on the evolution of the classical optical field. The quantum-mechanical fluctuations are based on the symbolic evaluations derived in Sec. II B and can be found in Appendix C. For the reduction to a two-level system, we obtain similar noise terms as derived by Drummond and Raymer.¹¹¹ However, unlike Drummond and Raymer, we can assure the preservation of the physical properties of the density matrix, i.e., positive definiteness and unit trace. This is accomplished by a suitable choice of the submatrices $B_v(\vec{A}_v, t)$ depicted in Table I.

III. SIMULATION

The density matrix equations can be used to model the carrier dynamics in low-dimensional SCLs.¹⁴² In QCLs, for example, optical intersubband transitions occur between quantized energy levels in the conduction band of a multiple quantum well heterostructure. The quantized energy state with index i refers here to a subband and is characterized by a wavefunction ψ_i . Due to the 1D-confinement in the growth direction, the free electron motion is restricted to the in-plane directions and is in carrier transport simulations characterized by an in-plane wavevector $\vec{k} = [k_x, k_y]^T$. Various 1D and 3D density matrix models for QCL structures are available in the literature.^{71,72,100,129–132,143–145} In THz QCLs with small energy level spacings, we can typically neglect nonparabolicity effects and obtain $\vec{k}$ independent optical transition frequencies ω_{ij} . In addition, Pauli blocking and Hartree–Fock renormalization effects, which occur in interband SCLs, play a subordinate role in QCLs due to the typically low doping levels.^{69,146} Since dynamic simulations of nonlinear optical effects, such as the generation of frequency combs, have to run over thousands of cavity round trips to achieve convergence, a complete 3D Maxwell-density matrix simulation is not feasible from a numerical point of view. If we take into account the $\vec{k}$ conservation of the dipole matrix elements, we can sum over $\vec{k}$, which yields density matrix equations of the form in Eq. (29). Under the assumption of moderate temporal variations in intersubband electron distributions $\rho_{jj,\vec{k}}$, the averaged scattering rates r_{ij} can be calculated by^{69,76}

$$r_{ij} = \sum_{\vec{k}, \vec{k}'} r_{jk' \rightarrow i\vec{k}} \rho_{jj,\vec{k}}^0 / \sum_{\vec{k}} \rho_{jj,\vec{k}}^0 \quad (30)$$

where $\rho_{jj,\vec{k}}^0$ is the steady-state electron distribution in the subband j . The $\vec{k}$ dependent scattering rates $r_{jk' \rightarrow i\vec{k}}$ can be extracted, e.g., from self-consistent density matrix-EMC (DM-EMC) simulations where they are calculated using Fermi's golden rule.⁷⁴ In QCLs, the electron distributions within one subband can often be well described by Fermi–Dirac or Maxwell–Boltzmann distributions. Here, the characteristic subband electron temperatures can significantly exceed the lattice temperatures.^{74,147} The effective dephasing rates γ_{ij} are typically obtained by averaging over the inversion between the subbands as^{148,149}

$$\gamma_{ij} = \sum_{\vec{k}} \gamma_{ij,\vec{k}} |\rho_{ii,\vec{k}}^0 - \rho_{jj,\vec{k}}^0| / \sum_{\vec{k}} |\rho_{ii,\vec{k}}^0 - \rho_{jj,\vec{k}}^0|. \quad (31)$$

As mentioned above, the Maxwell-density matrix equations are commonly treated in the so-called rotating-wave/slowly varying amplitude approximation to reduce the numerical load, which is only valid for relatively narrowband (and not too strong) optical fields.^{67,69,83} However, as discussed in the Introduction, in the framework of this paper, a broadband frequency comb with many modes is highly beneficial, and QCLs even offer the potential for generating spectra extending over a full octave and beyond.⁸⁴ None of the available open-source platforms are suitable for our purposes, mostly because they employ the rotating-wave approximation. Thus, we have developed our own open-source project mbsolve, allowing for numerically extensive simulations of multilevel systems based on the full-wave Maxwell-density matrix equations.^{98,141} In detail, the development of the codebase has been based on various principles. Here, the generalized Lindblad equation [Eq. (26)] instead of the usual, quite restrictive two-level Bloch equation model is used. We have developed numerical methods that preserve physical properties, such as the complete positivity and trace preservation of the density matrix.^{69,150} This is especially important in the context of long-term simulations, as required for frequency comb modeling. Furthermore, a computational speedup is obtained by using parallelization techniques,¹⁵¹ which is also especially important for long-term simulations of rather complex quantum systems, e.g., intracavity THz frequency comb difference frequency generation by mid-IR QCLs.¹⁰⁰ Our scientific software package mbsolve is developed following sustainable software engineering strategies and includes all common and essential best software engineering practices.^{151,152} It is based on C++ for performance reasons and features an easy-to-use Python interface facilitating the setup and active quantum system of the low-dimensional optoelectronic structures. The central part of the software is the mbsolve-lib base library, providing a framework for defining a simulation setup and the infrastructure to add solver and writer components. Importantly, mbsolve supports different numerical methods for solving the Lindblad equation,¹⁵⁰ as well as different parallelization techniques, e.g., OpenMP for shared memory systems. For a detailed package description, the reader is referred to Ref. 98.

As pointed out above, we extended our code base considering vacuum fluctuations due to spontaneous emission and fluctuations associated with the electronic transport.^{103,104,124} The solver class `solver_cpu_fdttd` has the method `run`, which executes the simulation loop by updating the magnetic field H_y , electric field E_z , density matrix ρ , and polarization $P_{z,qm}$ for all spatial and temporal grid points in the Yee grid. This update procedure is explained in detail in Ref. 98. We add a new density matrix algorithm class, which is based on the Strang operator splitting method,^{150,153} to account for fluctuations accompanying the electronic transport and vacuum fluctuations. The density algorithm class `algo_lindblad_reg_cayley_qnoise` contains the method `propagate_fluctuation`, which calculates the fluctuations for an update step by adding the product of the fluctuation superoperator and the time interval $\Delta t \cdot \mathcal{F}(\rho)$ to the updated density matrix $\rho^{n+1/2}$. Here, we investigate the active SCL gain media in lasing operation above threshold. In the simulations, we have to find a balance between

numerical efficiency and modeling accuracy. Most of the fluctuation terms given in Sec. II arise from the operator ordering when reducing the operator equations to c-number Langevin equations. As it was proven in the literature,^{104,122} these terms are negligible in the lasing regime above threshold with strong optical fields in the laser cavity. The fluctuation terms for a N -level system featuring diagonal elements F_{ii} and off-diagonal elements $F_{ij} = F_{ji}^\dagger$ can thus be significantly reduced for the numerical treatment and are described by

$$F_{ii}^j = -F_{jj}^i = \xi_{1,ij} \sqrt{\frac{r_{ji}\rho_{ii} + r_{ij}\rho_{jj}}{N_{\text{cell}}}}, \quad (32a)$$

$$F_{ii} = \sum_{j \neq i} F_{ii}^j, \quad (32b)$$

$$F_{ij} = (\xi_{2,ij} + i\xi_{3,ij}) \sqrt{\frac{-\tau_j^{-1}\rho_{jj} + \sum_{n \neq j} r_{jn}\rho_{nn} + 2\gamma_{ij}\rho_{jj}}{2N_{\text{cell}}}} \quad (32c)$$

for $i > j$,

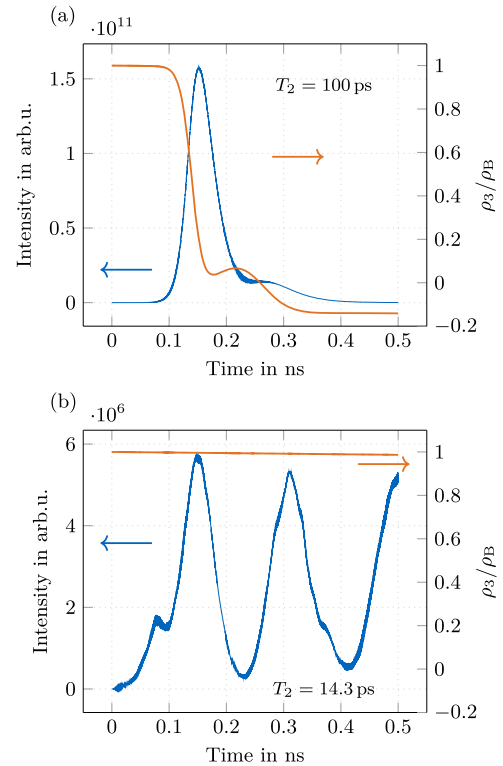


FIG. 3. Simulation results for a superfluorescence test setup¹⁰⁴ in an initially inverted two-level system using the Maxwell-density matrix Langevin equations. (a) Cooperative emission characteristic of superfluorescence for the dephasing time $T_2 = 100$ ps. (b) Amplified spontaneous emission pulse for the dephasing time $T_2 = 14.3$ ps.

where N_{cell} is the number of carriers in one grid cell. $\xi_{1,ij}$, $\xi_{2,ij}$, and $\xi_{3,ij}$ are real Gaussian random numbers and fulfill the correlation function,

$$\langle \xi_{k,ij}(t) | \xi_{l,mn}(t') \rangle = \delta_{kl} \delta_{im} \delta_{jn} \delta(t - t'). \quad (33)$$

For future applications, in which a more detailed fluctuation treatment would be beneficial, it might be necessary to extend our numerical model by additional noise terms derived in Sec. II C. Furthermore, we have implemented a class `ic_density_random_2lvl`, which represents random initial conditions for the common Maxwell-Bloch two-level system. As the dipole moment operators σ_{12}, σ_{21} and the atomic operators σ_{11}, σ_{22} do not commute, we have to take into account a non-vanishing initial stochastic value for the polarization term following the uncertainty principle.^{104,154} The tipping angle θ is obtained by drawing a random number from a Gaussian distribution with a standard deviation $\sigma = 2N_{\text{cell}}^{-1/2}$, and the angle ϕ in the xy-plane is obtained by drawing a random number from a uniform distribution.¹²⁴

A. Superfluorescence and amplified spontaneous emission

The system was tested with a superfluorescence (SF) setup in a two-level configuration.^{104,128} This setup describes the spontaneous buildup of a macroscopic coherent dipole moment in an initially inverted system, resulting in a collective emission of a superfluorescent pulse. This behavior can be reproduced numerically within our mbsolve framework by simulating an ensemble of excited ions and using a dephasing time $T_2 = 100$ ps. All other parameters required for the simulation are taken from Ref. 104. There, an excited state's lifetime $T_1 = 76$ ns, carrier number density $n_{3D} = 8.53 \times 10^{19} \text{ m}^{-3}$, carrier numbers per cell $N_{\text{cell}} = 3 \times 10^4$, transition frequency $f = 477$ THz, dipole length $d = 6.875 \times 10^{-2}$ nm, and equilibrium inversion $w_0 = -1$ are specified. We further investigate a device with length $L = 7$ mm using a grid discretization $\Delta x = 70$ nm.

The simulated SF pulse is illustrated in Fig. 3(a) and compares well with previous numerical and experimental findings.^{104,128,155} By increasing the collisional dephasing rate within the system, the SF pulse is significantly disturbed and gets broadened until the spontaneous buildup of the coherent dipole moment is prevented. For a dephasing time $T_2 = 14.3$ ps below the critical point, the SF pulse is replaced by amplified spontaneous emission (ASE). The increased noise amplitude accompanying the smaller dephasing time is crucial for the modeling of ASE, which cannot be reproduced otherwise. The ASE simulation results are presented in Fig. 3(b).

Furthermore, the degree of decoherence is studied using the quantity ρ_3/ρ_B , where $\rho_B = \sqrt{\rho_1^2 + \rho_2^2 + \rho_3^2}$ is the length of the Bloch vector. When the dephasing time T_2 is high [Fig. 3(a)], the population inversion ρ_3 is quickly depleted through the spontaneous buildup of the macroscopic dipole moment and the SF emission, which clearly surpasses the decay of ρ_1 and ρ_2 and results in a rapid drop of ρ_3/ρ_B . In the second case [Fig. 3(b)], we have used a smaller dephasing time T_2 , which prevents the macroscopic dipole moment buildup and limits the radiative decay. This decoherence state indicates a very slow decay of ρ_3/ρ_B , which stays close to one.

B. Noise characteristics in THz QCL harmonic frequency comb emission

Concerning the experimental investigation of intensity correlations in QCLs,⁵² we aim to characterize the noise properties of a self-starting THz QCL HFC setup. Here, the THz QCL active region is based on a homogeneous four-quantum well design with a diagonal transition.¹⁵⁶ The charge carrier transport in the active gain medium at a bias of 50 kV/period is analyzed using our in-house Monaco framework, consisting of a Schrödinger-Poisson solver and a density matrix-ensemble Monte Carlo modeling tool.^{74,76,77,157,158} For an appropriate description of the physical properties, we consider five wavefunctions in the active quantum well heterostructure. Furthermore, one incoherent tunneling transition from the injector state into the upper laser level and one optical transition are specified for the quantum mechanical description of the QCL system in the dynamical simulation. The Python script `forrer_2021_50mVperperiod.py` with the simulation setup to start the mbsolve simulation is given in Listing 1 of Appendix D and is furthermore included in the GitHub repository.¹⁴¹ Here, all input parameters for the full description of the quantum system are extracted from self-consistent DM-EMC simulations.

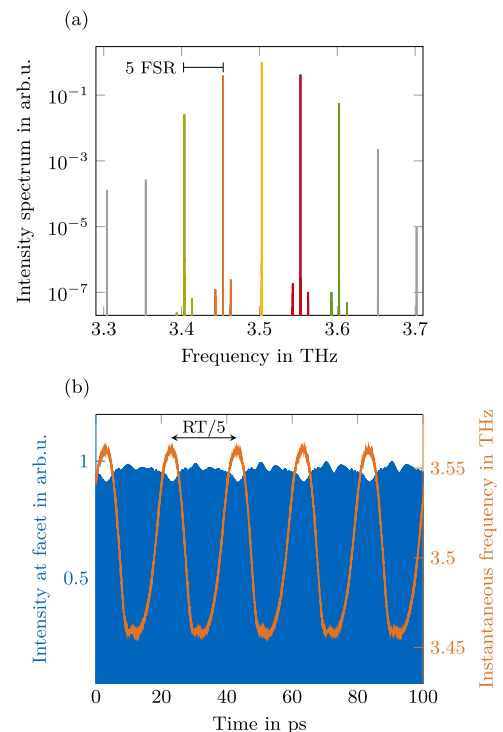


FIG. 4. Maxwell-density matrix Langevin simulation results of HFC emission with a mode spacing of 5 FSR in a 4 mm long THz QCL device with a metal-metal waveguide at 80 K and for $V = 50$ mV/period. (a) Intensity spectra of the optical radiation at the facet. (b) Simulated instantaneous intensity at the facet and calculated instantaneous frequency from the Hilbert transform of the simulated electric field over a single round trip time (RT).

In the following, we present simulation results for a 4 mm long double-metal THz QCL with a free spectral range (FSR) of 9.94 GHz. The intensity spectrum of the THz HFC at 3.5 THz with a mode spacing of 5 FSR is illustrated in Fig. 4(a). The THz QCL emits a broadband HFC with a cavity repetition rate of 49.7 GHz. In Fig. 4(b), the temporal evolution of the intensity at the facet and the calculated instantaneous frequency are depicted. We can identify a regular field pattern, which shows a periodic repetition with five times the round trip time. Here, only the three strongest modes are involved in the temporal evolution of the instantaneous frequency, as their intensities are of similar magnitude and contribute most to the overall comb emission power.

To specify the degree of coherence of the obtained HFC and for comparison with the experimental findings, we investigate the radio frequency (RF) spectrum using an observation time window of 2 μ s. The obtained simulation results are shown in Fig. 5(a), and

the clear appearance of the harmonic beatnote proves the purity of the harmonic state. The linewidth is substantially below the numerical frequency resolution of 500 kHz, which is confirmed by the zoom on the extremely narrow harmonic beatnote in the inset of Fig. 5(a). In addition, we can identify sub-beatnotes, which arise due to the beating of the center mode with the sub-comb lines. These sub-comb lines are generated by FWM processes, where the strong harmonic side modes act as pump modes and generate weak sidebands with a frequency spacing of 1 FSR from the corresponding pump modes. As can be seen in Fig. 4(a), the intensities of the sub-modes are at least ~ 5 orders of magnitude smaller than those of the pump modes.

To further analyze the noise characteristics of the THz QCL HFC setup, we calculate the relative intensity noise (RIN) for the total output power and for the power of the five harmonic comb lines contributing most to the HFC emission. Here, the RIN spectrum can be calculated by

$$RIN_i(f) = \lim_{T \rightarrow \infty} \frac{1}{T} \frac{|\int_0^T [P_i(t) - \langle P_i(t) \rangle] e^{-i2\pi f t} dt|^2}{\langle P_i(t) \rangle^2}, \quad (34)$$

where T denotes the simulation time and P_i is either the power of a specific mode i or the total power P_{all} . By numerically filtering the electric field at the facet $E_{\text{facet}}(t)$ using a filter with a 3 dB bandwidth of 20 GHz, we can extract the temporal electric field components $E_i(t)$ of the individual modes. The RIN results are depicted in Fig. 5(b) for the total power P_{all} and the power of the five central harmonic modes P_i with indices $i = 1, \dots, 5$. The total power RIN is around -180 dBc/Hz, while for the three central harmonic modes having a similar power, a RIN around -155 dBc/Hz is calculated. For the remaining two weaker modes 1 and 5, a higher RIN is obtained. This is in very good agreement with the experimental findings of a three-mode mid-IR HFC QCL setup.⁵² For increasing power, the RIN of the side modes decreases to that of the central mode, while sidebands closer to threshold exhibit a noisier behavior. Furthermore, we identify an overlapping RIN for side modes featuring a comparable power level, which indicates a comparable noise level. A similar result could be retrieved from the mid-IR HFC RIN measurements.⁵²

IV. CONCLUSION

In this paper, we have theoretically derived a c-number Langevin approach for a three-level quantum system from the non-classical operator description within the Heisenberg–Langevin equations. Our approach is an extension of the well-known two-level quantum theory by Drummond and Raymer,¹¹¹ where we additionally take into account incoherent tunneling injection into the upper laser level. Within the generalized Maxwell-density matrix Langevin equations, we can ensure the preservation of the physical properties of the density matrix, i.e., positive definiteness and unit trace. Furthermore, by including the derived noise terms into our open-source simulation tool mbsolve, we can model the fluctuations accompanying electronic transport and spontaneous emission in the dynamical simulations of the light–matter interaction in multilevel quantum optoelectronic systems, such as QCLs and QD lasers. The simulation approach is tested using a superfluorescence setup, where we prove the validity of our implementation by obtaining an excellent match

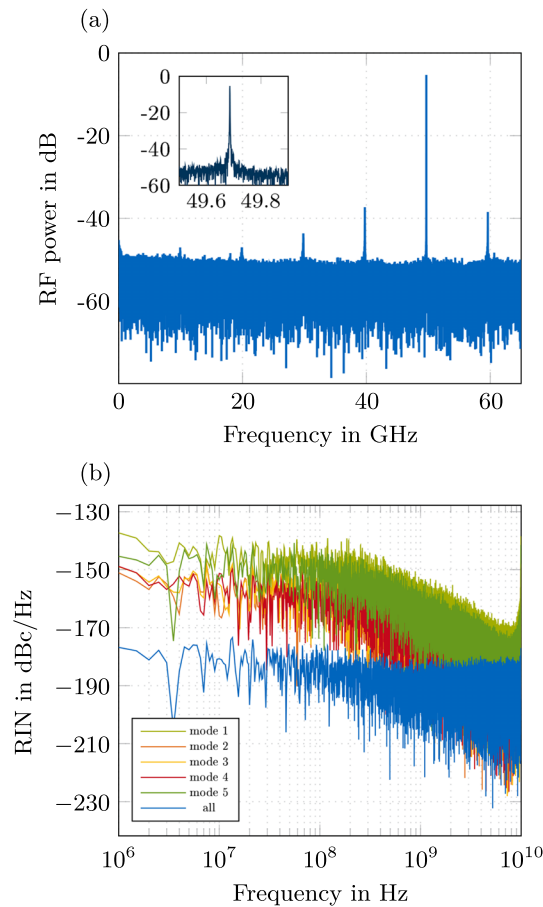


FIG. 5. (a) Simulated RF spectrum of the THz QCL HFC setup with a clear beat-note signal at 49.7 GHz. Inset: zoom on the harmonic beatnote, indicating a narrow linewidth below the numerical frequency resolution (500 kHz). (b) Calculated RIN spectra associated with the total power P_{all} (blue) and the modal power P_i of each of the five harmonic modes contributing most to the HFC emission. The colors of the individual RIN spectra correspond to those of the individual comb lines in Fig. 4(a).

with previous experimental and theoretical results.^{104,124,128,155} In addition, we have characterized the noise properties of a coherent THz QCL HFC setup and obtained a good match with experimental findings.^{52,156} Our modeling approach based on the generalized Maxwell-density matrix Langevin equations shows great potential for the theoretical investigation of intermodal intensity correlations in photonic devices and the development of low-noise integrated light emitters, also with regard to the generation of non-classical light.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Johannes Popp: Conceptualization (lead); Data curation (lead); Formal analysis (lead); Investigation (lead); Methodology (lead); Software (lead); Validation (equal); Visualization (lead); Writing – original draft (lead); Writing – review & editing (lead).

Johannes Stowasser: Conceptualization (supporting); Formal analysis (supporting); Investigation (supporting); Methodology (supporting); Validation (supporting); Writing – review & editing (equal). **Michael A. Schreiber:** Data curation (supporting); Formal analysis (supporting); Methodology (supporting); Validation (supporting); Writing – review & editing (supporting). **Lukas Seitzer:** Conceptualization (supporting); Data curation (supporting); Formal analysis (supporting); Investigation (supporting); Software (supporting); Validation (supporting); Writing – review & editing (supporting). **Felix Hitzelhammer:** Conceptualization (supporting); Formal analysis (supporting); Methodology (supporting); Validation (supporting); Writing – review & editing (supporting). **Michael Haider:** Conceptualization (supporting); Data curation (supporting); Funding acquisition (supporting); Investigation (supporting); Project administration (supporting); Software (supporting); Supervision (supporting); Validation (supporting); Writing – review & editing (supporting). **Gabriela Slavcheva:** Conceptualization (supporting); Formal analysis (supporting); Funding acquisition (supporting); Methodology (equal); Project administration (supporting); Supervision (supporting); Validation (equal); Writing – review & editing (equal). **Christian Jirauschek:** Conceptualization (supporting); Data curation (supporting); Formal analysis (supporting); Funding acquisition (lead); Investigation (supporting); Methodology (equal); Project administration (lead); Resources (lead); Software (equal); Supervision (lead); Validation (supporting); Writing – original draft (supporting); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are openly available in GitHub.

APPENDIX A: CALCULATION OF THE DIFFUSION COEFFICIENT WITHIN THE QUANTUM AND THE C-NUMBER LANGEVIN TREATMENT

By taking into account the orthogonality of the levels $\langle i|j \rangle = \delta_{ij}$, we obtain

$$\hat{\sigma}_{23}^\dagger \hat{\sigma}_{23} = (|3\rangle\langle 2|2\rangle\langle 3|) = (|3\rangle\langle 3|) = \hat{\sigma}_{33}. \quad (\text{A1})$$

The diffusion coefficient in Eq. (9) is calculated by using the *generalized Einstein relation* from Eq. (6). The detailed calculation is given by

$$\begin{aligned} 2\langle \hat{D}_{3223}(t) \rangle_R &= \partial_t \langle \hat{\sigma}_{33}(t) \rangle_R - \langle \hat{M}_{23}^\dagger(t) \hat{\sigma}_{23}(t) \rangle_R - \langle \hat{\sigma}_{23}^\dagger(t) \hat{M}_{23}(t) \rangle_R \\ &= \left\langle -\frac{1}{\tau_3} \hat{\sigma}_{33}(t) + r_{32} \hat{\sigma}_{22}(t) + r_{31'} \hat{\sigma}_{1'1'}(t) + i g \left[\hat{a}^\dagger(t) \hat{\sigma}_{23}(t) - \hat{a}(t) \hat{\sigma}_{23}^\dagger(t) \right] - i \Omega_{1'3} \left[\hat{\sigma}_{31'}^\dagger(t) - \hat{\sigma}_{31'}(t) \right] \right\rangle_R \\ &\quad - \left\langle \left\{ \frac{i}{\hbar} \Delta_{32} \hat{\sigma}_{23}^\dagger(t) - \gamma_{23} \hat{\sigma}_{23}^\dagger(t) - i g \left[\hat{\sigma}_{33}(t) - \hat{\sigma}_{22}(t) \right] \hat{a}^\dagger(t) - i \Omega_{1'3} \hat{\sigma}_{1'2}(t) \right\} \hat{\sigma}_{23}(t) \right\rangle_R \\ &\quad - \left\langle \hat{\sigma}_{23}^\dagger(t) \left\{ -\frac{i}{\hbar} \Delta_{32} \hat{\sigma}_{23}(t) - \gamma_{23} \hat{\sigma}_{23}(t) + i g \left[\hat{\sigma}_{33}(t) - \hat{\sigma}_{22}(t) \right] \hat{a}(t) + i \Omega_{1'3} \hat{\sigma}_{21'}(t) \right\} \right\rangle_R \\ &= \left(2\gamma_{23} - \frac{1}{\tau_3} \right) \langle \hat{\sigma}_{33}(t) \rangle_R + r_{32} \langle \hat{\sigma}_{22}(t) \rangle_R + r_{31'} \langle \hat{\sigma}_{1'1'}(t) \rangle_R. \end{aligned} \quad (\text{A2})$$

In addition, we prove the difference in diffusion coefficients, which arises through the transition from operator to c-number Langevin equations. Therefore, we calculate the diffusion coefficient $D_{3333}(t)$ based on the theoretical description presented in Sec. II B. By the use of Eqs. (6), and 7(e), we obtain

$$\begin{aligned} \partial_t \langle \hat{\sigma}_{33}(t) \hat{\sigma}_{33}(t) \rangle_R = & -\frac{2}{\tau_3} \langle \hat{\sigma}_{33}(t) \hat{\sigma}_{33}(t) \rangle_R + r_{32} \left[\langle \hat{\sigma}_{22}(t) \hat{\sigma}_{33}(t) \rangle_R + \langle \hat{\sigma}_{33}(t) \hat{\sigma}_{22}(t) \rangle_R \right] + r_{31'} \left[\langle \hat{\sigma}_{1'1'}(t) \hat{\sigma}_{33}(t) \rangle_R \right. \\ & \left. + \langle \hat{\sigma}_{33}(t) \hat{\sigma}_{1'1'}(t) \rangle_R \right] + ig \left[\langle \hat{a}^\dagger(t) \hat{\sigma}_{23}(t) \hat{\sigma}_{33}(t) \rangle_R - \langle \hat{a}(t) \hat{\sigma}_{23}^\dagger(t) \hat{\sigma}_{33}(t) \rangle_R + \langle \hat{a}^\dagger(t) \hat{\sigma}_{33}(t) \hat{\sigma}_{23}(t) \rangle_R \right. \\ & \left. - \langle \hat{a}(t) \hat{\sigma}_{33}(t) \hat{\sigma}_{23}^\dagger(t) \rangle_R \right] - i\Omega_{1'3} \left[\langle \hat{\sigma}_{31'}^\dagger(t) \hat{\sigma}_{33}(t) \rangle_R - \langle \hat{\sigma}_{31'}(t) \hat{\sigma}_{33}(t) \rangle_R + \langle \hat{\sigma}_{33}(t) \hat{\sigma}_{31'}^\dagger(t) \rangle_R \right. \\ & \left. - \langle \hat{\sigma}_{33}(t) \hat{\sigma}_{31'}(t) \rangle_R \right] + 2 \langle \hat{D}_{3333}(t) \rangle_R. \end{aligned} \quad (\text{A3})$$

Here, the terms that are underlined in red are not in the chosen order defined in Sec. II B. The commutation relations are used to bring these terms into chosen order, and by exploiting the level orthogonality similar to Eq. (A1), we derive

$$[\hat{\sigma}_{22}(t), \hat{\sigma}_{33}(t)] = \hat{\sigma}_{22}(t) \hat{\sigma}_{33}(t) - \hat{\sigma}_{33}(t) \hat{\sigma}_{22}(t) = 0, \quad (\text{A4a})$$

$$[\hat{\sigma}_{23}(t), \hat{\sigma}_{33}(t)] = \hat{\sigma}_{23}(t), \quad (\text{A4b})$$

$$[\hat{\sigma}_{33}(t), \hat{\sigma}_{23}^\dagger(t)] = \hat{\sigma}_{23}^\dagger(t), \quad (\text{A4c})$$

$$[\hat{\sigma}_{31'}(t), \hat{\sigma}_{33}(t)] = -\hat{\sigma}_{31'}(t), \quad (\text{A4d})$$

$$[\hat{\sigma}_{33}(t), \hat{\sigma}_{31'}^\dagger(t)] = -\hat{\sigma}_{31'}^\dagger(t). \quad (\text{A4e})$$

With this, we can restructure Eq. (A3) as follows:

$$\begin{aligned} \partial_t \langle \hat{\sigma}_{33}(t) \hat{\sigma}_{33}(t) \rangle_R = & -\frac{2}{\tau_3} \langle \hat{\sigma}_{33}(t) \hat{\sigma}_{33}(t) \rangle_R + 2r_{32} \langle \hat{\sigma}_{33}(t) \hat{\sigma}_{22}(t) \rangle_R + 2r_{31'} \langle \hat{\sigma}_{33}(t) \hat{\sigma}_{1'1'}(t) \rangle_R \\ & + 2ig \left[\langle \hat{a}^\dagger(t) \hat{\sigma}_{33}(t) \hat{\sigma}_{23}(t) \rangle_R - \langle \hat{a}(t) \hat{\sigma}_{23}^\dagger(t) \hat{\sigma}_{33}(t) \rangle_R \right] - 2i\Omega_{1'3} \left[\langle \hat{\sigma}_{31'}^\dagger(t) \hat{\sigma}_{33}(t) \rangle_R - \langle \hat{\sigma}_{33}(t) \hat{\sigma}_{31'}(t) \rangle_R \right] \\ & + 2 \langle \hat{D}_{3333}(t) \rangle_R + ig \left[\langle \hat{a}^\dagger(t) \hat{\sigma}_{23}(t) \rangle_R - \langle \hat{a}(t) \hat{\sigma}_{23}^\dagger(t) \rangle_R \right] + i\Omega_{1'3} \left[\langle \hat{\sigma}_{31'}^\dagger(t) \rangle_R - \langle \hat{\sigma}_{31'}(t) \rangle_R \right]. \end{aligned} \quad (\text{A5})$$

Here, the additional terms resulting from the operator ordering are underlined in green. With the use of Eqs. (14) and (15), we can derive the corresponding c-number equation,

$$\begin{aligned} \partial_t \langle \sigma_{33}(t) \sigma_{33}(t) \rangle_R = & -\frac{2}{\tau_3} \langle \sigma_{33}(t) \sigma_{33}(t) \rangle_R + 2r_{32} \langle \sigma_{33}(t) \sigma_{22}(t) \rangle_R + 2r_{31'} \langle \sigma_{33}(t) \sigma_{1'1'}(t) \rangle_R + 2ig \left[\langle a^*(t) \sigma_{33}(t) \sigma_{23}(t) \rangle_R \right. \\ & \left. - \langle a(t) \sigma_{23}^*(t) \sigma_{33}(t) \rangle_R \right] - 2i\Omega_{1'3} \left[\langle \sigma_{31'}^*(t) \sigma_{33}(t) \rangle_R - \langle \sigma_{33}(t) \sigma_{31'}(t) \rangle_R \right] + 2 \langle D_{3333}(t) \rangle_R. \end{aligned} \quad (\text{A6})$$

If we now require the equivalence of the left-hand sides of Eqs. (A5) and (A6), we end up with the diffusion coefficient from Eq. 19(a).

APPENDIX B: COMPLETE DIFFUSION MATRIX FOR A THREE-LEVEL THz QCL SYSTEM

The complete diffusion matrix for a three-level THz QCL system with incoherent tunneling injection is derived within the framework of the c-number Langevin theory and is given by

$$A = \begin{bmatrix}
 0 & \sqrt{n_{\text{th}}\kappa} & 0 & 0 & 0 \\
 \sqrt{n_{\text{th}}\kappa} & 0 & 0 & 0 & 0 \\
 0 & 0 & -2iga^* \sigma_{23}^* & iga^* \sigma_{31'}^* & -iga^* \sigma_{21'}^* \\
 0 & 0 & iga^* \sigma_{31'}^* & 2i\Omega_{1'3} \sigma_{31'}^* & 0 \\
 0 & 0 & -iga^* \sigma_{21'}^* & 0 & 0 \\
 0 & 0 & -r_{32} \sigma_{23}^* & -iga \sigma_{21'}^* + (r_{23} + r_{1'3}) \sigma_{31'}^* & -r_{32} \sigma_{21'}^* \\
 0 & 0 & (r_{32} + r_{1'2}) \sigma_{23}^* & iga \sigma_{21'}^* - r_{23} \sigma_{31'}^* & (r_{32} + r_{1'2}) \sigma_{21'}^* \\
 0 & 0 & -r_{1'2} \sigma_{23}^* & -r_{1'3} \sigma_{31'}^* & -r_{1'2} \sigma_{21'}^* \\
 0 & 0 & (\gamma_{23} + \gamma_{1'2} - \gamma_{1'3}) \sigma_{31'}^* & 0 & (2\gamma_{1'2} - r_{21'} - r_{31'}) \sigma_{1'1'}^* \\
 & & & & + r_{1'2} \sigma_{22} + r_{1'3} \sigma_{33} \\
 0 & 0 & 0 & (2\gamma_{1'3} - r_{21'} - r_{31'}) \sigma_{1'1'}^* + \\
 & & & r_{1'2} \sigma_{22} + r_{1'3} \sigma_{33} & 0 \\
 0 & 0 & (2\gamma_{23} - r_{23} - r_{1'3}) \sigma_{33} & 0 & (\gamma_{1'2} + \gamma_{23} - \gamma_{1'3}) \sigma_{31'}^* \\
 & & + r_{32} \sigma_{22} & & \\
 0 & 0 & 0 & 0 & 0 \\
 0 & 0 & 0 & 0 & 0 \\
 -r_{32} \sigma_{23}^* & (r_{32} + r_{1'2}) \sigma_{23}^* & -r_{1'2} \sigma_{23}^* \\
 -iga \sigma_{21'}^* + (r_{23} + r_{1'3}) \sigma_{31'}^* & iga \sigma_{21'}^* - r_{23} \sigma_{31'}^* & -r_{1'3} \sigma_{31'}^* \\
 -r_{32} \sigma_{21'}^* & (r_{32} + r_{1'2}) \sigma_{21'}^* & -r_{1'2} \sigma_{21'}^* \\
 (r_{23} + r_{1'3}) \sigma_{33} + r_{32} \sigma_{22} + r_{3'1} \sigma_{1'1'}^* & -r_{32} \sigma_{22} - r_{23} \sigma_{33} & i\Omega_{1'3} (\sigma_{31'}^* - \sigma_{31'}^*) \\
 +ig(a^* \sigma_{23} - a \sigma_{23}^*) + i\Omega_{1'3} (\sigma_{31'}^* - \sigma_{31'}) & +ig(a \sigma_{23}^* - a^* \sigma_{23}) & -r_{31'} \sigma_{1'1'}^* - r_{1'3} \sigma_{33} \\
 -r_{32} \sigma_{22} - r_{23} \sigma_{33} & r_{23} \sigma_{33} + (r_{32} + r_{1'2}) \sigma_{22} & -r_{21'} \sigma_{1'1'}^* - r_{1'2} \sigma_{22} \\
 +ig(a \sigma_{23}^* - a^* \sigma_{23}) & +r_{21'} \sigma_{1'1'}^* + ig(a^* \sigma_{23} - a \sigma_{23}^*) & r_{1'2} \sigma_{22} + (r_{21'} + r_{31'}) \sigma_{1'1'}^* \\
 i\Omega_{1'3} (\sigma_{31'}^* - \sigma_{31'}) & -r_{21'} \sigma_{1'1'}^* - r_{1'2} \sigma_{22} & +i\Omega_{1'3} (\sigma_{31'}^* - \sigma_{31'}) + r_{1'3} \sigma_{33} \\
 -r_{1'3} \sigma_{33} - r_{31'} \sigma_{1'1'}^* & (r_{32} + r_{1'2}) \sigma_{21'}^* & -r_{1'2} \sigma_{21'}^* \\
 -r_{32} \sigma_{21'}^* & (r_{32} + r_{1'2}) \sigma_{21'}^* & -r_{1'2} \sigma_{21'}^* \\
 iga^* \sigma_{21'}^* + (r_{23} + r_{1'3}) \sigma_{31'}^* & -iga^* \sigma_{21'}^* - r_{23} \sigma_{31'}^* & -r_{1'3} \sigma_{31'}^* \\
 -r_{32} \sigma_{23} & (r_{32} + r_{1'2}) \sigma_{23} & -r_{1'2} \sigma_{23}
 \end{bmatrix} \quad (B1)$$

$$\begin{bmatrix}
0 & 0 & 0 \\
0 & 0 & 0 \\
(\gamma_{23} + \gamma_{1'2} - \gamma_{1'3})\sigma_{31'} & 0 & (2\gamma_{23} - r_{23} - r_{1'3})\sigma_{33} \\
0 & (2\gamma_{1'3} - r_{2'1} - r_{31'})\sigma_{1'1'} \\
& + r_{1'2}\sigma_{22} + r_{1'3}\sigma_{33} & 0 \\
(2\gamma_{1'2} - r_{21'} - r_{31'})\sigma_{1'1'} & 0 & (\gamma_{23} + \gamma_{1'2} - \gamma_{1'3})\sigma_{31'}^* \\
+ r_{1'2}\sigma_{22} + r_{1'3}\sigma_{33} & & \\
-r_{32}\sigma_{21'} & i g a^* \sigma_{21'} + (r_{23} + r_{1'3})\sigma_{31'} & -r_{32}\sigma_{23} \\
(r_{32} + r_{1'2})\sigma_{21'} & -i g a^* \sigma_{21'} - r_{23}\sigma_{31'} & (r_{32} + r_{1'2})\sigma_{23} \\
-r_{1'2}\sigma_{21'} & -r_{1'3}\sigma_{31'} & -r_{1'2}\sigma_{23} \\
0 & 0 & i g a \sigma_{21'} \\
0 & -2i\Omega_{1'3}\sigma_{31'} & -i g a \sigma_{31'} \\
i g a \sigma_{21'} & -i g a \sigma_{31'} & 2i g a \sigma_{23}
\end{bmatrix}
\begin{bmatrix}
a^* \\
a \\
\sigma_{23}^* \\
\sigma_{31'}^* \\
\sigma_{21'}^* \\
\sigma_{33} \\
\sigma_{22} \\
\sigma_{1'1'} \\
\sigma_{21'} \\
\sigma_{31'} \\
\sigma_{23}
\end{bmatrix}.$$

APPENDIX C: QUANTUM MECHANICAL FLUCTUATION TERMS WITHIN THE GENERALIZED MAXWELL-DENSITY MATRIX LANGEVIN EQUATIONS

The quantum-mechanical fluctuation terms for the three-level QCL quantum system are derived within the framework of the Langevin theory. In this paper, we have calculated the full diffusion matrix resulting from the c-number Langevin equations. Exploiting the positive semi-definiteness of the diffusion matrix, one can show that there exists a set of Ito stochastic differential equations

equivalent to the Langevin equations. We can factorize the diffusion matrix to obtain a noise matrix that can be directly integrated into the Maxwell-density matrix approach for numerical modeling of fluctuations in dynamical optoelectronic devices. With a suitable choice of the noise matrix, one can guarantee a completely positive trace-preserving update map for long-term simulations. For the three-level QCL system, the fluctuation terms will fully account for the influence of the reservoirs and the properties of the nonlinear coupling between the QCL system and the optical field, including the incoherent tunneling transition, and can be represented as follows:

$$\begin{aligned}
F_{23}(t) = & \xi_{11}(t)\sqrt{r_{32}} + \xi_{14}(t)\sqrt{r_{1'2}} - \xi_{24}(t)\sqrt{2i\mu_{z,23}E_z(t)\rho_{32}(t)} - \xi_{31}^*(t)\frac{i\mu_{z,23}E_z(t)}{2} + \xi_{32}^*(t)\frac{i\mu_{z,23}E_z(t)}{2} \\
& + \xi_{33}^*(t)\sqrt{\frac{\gamma_{1'2} - \gamma_{1'3} + \gamma_{23}}{2}} + \xi_{41}^*(t)\left[\frac{\gamma_{1'3} - \gamma_{1'2} - \gamma_{23}}{2} + \frac{r_{32}\rho_{22}(t)}{2} + \frac{2\gamma_{23} - r_{1'3} - r_{23}}{2}\rho_{33}(t) \right. \\
& \left. - \frac{\mu_{z,23}^2 E_z(t)^2}{2} - r_{1'2} - r_{32} + \mu_{z,23}E_z(t)|\rho_{32}(t)|\right]^{1/2}, \quad (C1a)
\end{aligned}$$

$$\begin{aligned}
F_{31'}(t) = & \xi_{12}(t)\sqrt{r_{1'3}} + \xi_{15a}(t)\sqrt{r_{23}} - \xi_{15b}(t)i\mu_{z,23}E_z(t) - \xi_{25}(t)\sqrt{-2i\Omega_{31'}\rho_{1'3}(t)} + \xi_{31}(t)\rho_{1'3}(t) \\
& + \xi_{42}^*(t)\left[\frac{2\gamma_{1'3} - \gamma_{1'2} - \gamma_{23}}{2}\rho_{1'1'}(t) + \frac{r_{1'2}}{2}\rho_{22}(t) - |\rho_{31'}(t)|^2 + \frac{r_{1'3}}{2}\rho_{33}(t) - \mu_{z,23}^2 E_z(t)^2 - r_{1'3} - r_{23} + \Omega_{31'}|\rho_{31'}(t)|\right]^{1/2}, \quad (C1b)
\end{aligned}$$

$$\begin{aligned}
F_{21'}(t) = & \xi_{13}(t)\sqrt{r_{1'2}} + \xi_{16}(t)\sqrt{r_{32}} + \xi_{32}(t)\rho_{1'2}(t) + \xi_{33}^*(t)\sqrt{\frac{\gamma_{1'2} - \gamma_{1'3} + \gamma_{23}}{2}}\rho_{1'3}(t) + \xi_{43}^*(t)\left[\frac{2\gamma_{1'3} - \gamma_{1'2} - \gamma_{23}}{2} \right. \\
& \left. \times \rho_{1'1'}(t) + \frac{r_{1'2}}{2}\rho_{22}(t) - |\rho_{21'}(t)|^2 + \frac{r_{1'3}}{2}\rho_{33}(t) - r_{1'2} - r_{32} - \frac{\gamma_{1'3} - \gamma_{1'2} - \gamma_{23}}{2}|\rho_{31'}(t)|^2\right]^{1/2}, \quad (C1c)
\end{aligned}$$

$$\begin{aligned}
F_{33}(t) = & -\xi_{11}^*(t) \frac{\sqrt{r_{32}}\rho_{32}(t)}{2} - \xi_{11}(t) \frac{\sqrt{r_{32}}\rho_{23}(t)}{2} + \xi_{12}^*(t) \frac{\sqrt{r_{1'3}}\rho_{1'3}(t)}{2} + \xi_{12}(t) \frac{\sqrt{r_{1'3}}\rho_{31'}(t)}{2} + \xi_{15a}^*(t) \frac{\sqrt{r_{23}}\rho_{1'3}(t)}{2} \\
& + \xi_{15a}(t) \frac{\sqrt{r_{23}}\rho_{31'}(t)}{2} - \xi_{15b}^*(t) \frac{\rho_{1'2}(t)}{2} - \xi_{15b}(t) \frac{\rho_{21'}(t)}{2} - \xi_{16}^*(t) \frac{\sqrt{r_{32}}\rho_{1'2}(t)}{2} - \xi_{16}(t) \frac{\sqrt{r_{32}}\rho_{21'}(t)}{2} \\
& + \xi_{21}(t) \{-(r_{32}+1)|\rho_{1'2}(t)|^2 + r_{23}[\rho_{33}(t) - |\rho_{1'3}(t)|^2] + r_{32}[\rho_{22}(t) - |\rho_{32}(t)|^2] + i\mu_{z,23}E_z(t)[\rho_{32}(t) \\
& - \rho_{23}(t)]\}^{1/2} + \xi_{22}(t) \{i\Omega_{31'}[-\rho_{1'3}(t) + \rho_{31'}(t)] + r_{31'}\rho_{1'1'}(t) + r_{1'3}[\rho_{33}(t) - |\rho_{1'3}(t)|^2]\}^{1/2}, \quad (C1d)
\end{aligned}$$

$$\begin{aligned}
F_{22}(t) = & \xi_{11}^*(t) \frac{\sqrt{r_{32}}\rho_{32}(t)}{2} + \xi_{11}(t) \frac{\sqrt{r_{32}}\rho_{23}(t)}{2} + \xi_{13}^*(t) \frac{\sqrt{r_{1'2}}\rho_{1'2}(t)}{2} + \xi_{13}(t) \frac{\sqrt{r_{1'2}}\rho_{21'}(t)}{2} + \xi_{14}^*(t) \frac{\sqrt{r_{1'2}}\rho_{32}(t)}{2} \\
& + \xi_{14}(t) \frac{\sqrt{r_{1'2}}\rho_{23}(t)}{2} - \xi_{15a}^*(t) \frac{\sqrt{r_{23}}\rho_{1'3}(t)}{2} - \xi_{15a}(t) \frac{\sqrt{r_{23}}\rho_{31'}(t)}{2} + \xi_{15b}^*(t) \frac{\rho_{1'2}(t)}{2} + \xi_{15b}(t) \frac{\rho_{21'}(t)}{2} \\
& + \xi_{16}^*(t) \frac{\sqrt{r_{32}}\rho_{1'2}(t)}{2} + \xi_{16}(t) \frac{\sqrt{r_{32}}\rho_{21'}(t)}{2} - \xi_{21}(t) \{r_{32}\rho_{22}(t) + r_{23}\rho_{33}(t) + i\mu_{z,23}E_z(t)[\rho_{32}(t) - \rho_{23}(t)] \\
& - (r_{32}+1)|\rho_{1'2}(t)|^2 - r_{32}|\rho_{32}(t)|^2 - r_{23}|\rho_{1'3}(t)|^2\}^{1/2} + \xi_{23}(t) \{r_{21'}\rho_{1'1'}(t) - r_{1'2}[|\rho_{1'2}(t)|^2 - \rho_{22}(t) + |\rho_{23}(t)|^2]\}^{1/2}, \quad (C1e)
\end{aligned}$$

$$\begin{aligned}
F_{1'1'}(t) = & -\xi_{12}^*(t) \frac{\sqrt{r_{1'3}}\rho_{1'3}(t)}{2} - \xi_{12}(t) \frac{\sqrt{r_{1'3}}\rho_{31'}(t)}{2} - \xi_{13}^*(t) \frac{\sqrt{r_{1'2}}\rho_{1'2}(t)}{2} - \xi_{13}(t) \frac{\sqrt{r_{1'2}}\rho_{21'}(t)}{2} - \xi_{14}^*(t) \frac{\sqrt{r_{1'2}}\rho_{32}(t)}{2} \\
& - \xi_{14}(t) \frac{\sqrt{r_{1'2}}\rho_{23}(t)}{2} - \xi_{22}(t) \{i\Omega_{31'}[-\rho_{1'3}(t) + \rho_{31'}(t)] + r_{31'}\rho_{1'1'}(t) + r_{1'3}[\rho_{33}(t) - |\rho_{1'3}(t)|^2]\}^{1/2} \\
& - \xi_{23}(t) \{r_{21'}\rho_{1'1'}(t) - r_{1'2}[|\rho_{1'2}(t)|^2 - \rho_{22}(t) + |\rho_{23}(t)|^2]\}^{1/2}, \quad (C1f)
\end{aligned}$$

$$\begin{aligned}
F_{1'2}(t) = & \xi_{13}^*(t) \sqrt{r_{1'2}} + \xi_{16}^*(t) \sqrt{r_{32}} + \xi_{32}^*(t) \rho_{21'}(t) + \xi_{33}(t) \sqrt{\frac{\gamma_{1'2} - \gamma_{1'3} + \gamma_{23}}{2}} \rho_{1'3}(t) + \xi_{43}(t) \left[\frac{r_{1'2}}{2} \rho_{22}(t) \right. \\
& \left. + \frac{2\gamma_{1'3} - \gamma_{1'2} - \gamma_{23}}{2} \rho_{1'1'}(t) - |\rho_{21'}(t)|^2 + \frac{r_{1'3}}{2} \rho_{33}(t) - r_{1'2} - r_{32} - \frac{\gamma_{1'3} - \gamma_{1'2} - \gamma_{23}}{2} |\rho_{31'}(t)|^2 \right]^{1/2}, \quad (C1g)
\end{aligned}$$

$$\begin{aligned}
F_{1'3}(t) = & \xi_{12}^*(t) \sqrt{r_{1'3}} + \xi_{15a}^*(t) \sqrt{r_{23}} + \xi_{15b}^*(t) i\mu_{z,23}E_z(t) + \xi_{25}(t) \sqrt{2i\Omega_{31'}\rho_{31'}(t)} + \xi_{31}^*(t) \rho_{31'}(t) + \xi_{42}(t) \left[\frac{r_{1'2}}{2} \rho_{22}(t) \right. \\
& \left. + \frac{2\gamma_{1'3} - \gamma_{1'2} - \gamma_{23}}{2} \rho_{1'1'}(t) - |\rho_{31'}(t)|^2 + \frac{r_{1'3}}{2} \rho_{33}(t) - \mu_{z,23}^2 E_z(t)^2 - r_{1'3} - r_{23} + \Omega_{3'1} |\rho_{31'}(t)| \right]^{1/2}, \quad (C1h)
\end{aligned}$$

$$\begin{aligned}
F_{32}(t) = & \xi_{11}^*(t) \sqrt{r_{32}} + \xi_{14}^*(t) \sqrt{r_{1'2}} + \xi_{24}(t) \sqrt{-2i\mu_{z,23}E_z(t)\rho_{23}(t)} + \xi_{31}(t) \frac{i\mu_{z,23}E_z(t)}{2} - \xi_{32}(t) \frac{i\mu_{z,23}E_z(t)}{2} \\
& + \xi_{33}(t) \sqrt{\frac{\gamma_{1'2} - \gamma_{1'3} + \gamma_{23}}{2}} + \xi_{41}(t) \left[\frac{\gamma_{1'3} - \gamma_{1'2} - \gamma_{23}}{2} + \frac{r_{32}\rho_{22}(t)}{2} + \frac{2\gamma_{23} - r_{1'3} - r_{23}}{2} \rho_{33}(t) \right. \\
& \left. - \frac{\mu_{z,23}^2 E_z(t)^2}{2} - r_{1'2} - r_{32} + \mu_{z,23}E_z(t) |\rho_{32}(t)| \right]^{1/2}. \quad (C1i)
\end{aligned}$$

Here, the terms $\xi_{11}, \xi_{12}, \xi_{13}, \xi_{14}, \xi_{15a}, \xi_{15b}, \xi_{16}, \xi_{31}, \xi_{32}, \xi_{33}, \xi_{41}, \xi_{42}, \xi_{43}$ are complex, while $\xi_{21}, \xi_{22}, \xi_{23}, \xi_{24}, \xi_{25}$ are real.

LISTING 1. Code snippet of the Python script for the THz HFC QCL setup in Ref. 156.

```

import mbsolve.lib as mb
import mbsolve.solvercpu
import mbsolve.writerhdf5
import mbsolve.readerhdf5

import math
import time

# Hamiltonian
energies = [ 0.0097 * mb.E0, 0.0082 * mb.E0, -0.0047 * mb.E0,
             -0.0083 * mb.E0, -0.0097 * mb.E0 ]
off_diagonals = [ 0.0005 * mb.E0, 0, 0, 0, 0, 0, 0, 0, 0, 0 ]
H = mb.qm_operator(energies, off_diagonals)

# dipole moment operator
off_dipoles = [ 0, -2.9500e-09 * mb.E0, 0, 0, 0, 0, 0, 0, 0, 0 ]
diag_dipoles = [ 0, 0, 0, 0, 0 ]
u = mb.qm_operator(diag_dipoles, off_dipoles)

# relaxation superoperator
# scattering rate matrix R
rates = [ [ 0, 1.8815e+09, 2.1290e+10, 4.0984e+09, 5.6000e+09 ],
           [ 3.5006e+09, 0, 3.2437e+08, 2.2854e+10, 2.0029e+12 ],
           [ 6.5578e+10, 6.2829e+08, 0, 8.0333e+11, 6.1577e+09 ],
           [ 6.8416e+09, 3.6845e+08, 6.6107e+11, 0, 4.7378e+12 ],
           [ 5.2192e+08, 6.7259e+10, 4.7554e+09, 4.7726e+12, 0 ] ]

# pure dephasing rates
pure_deph = [ 3.5857e+12, 9.3257e+11, 0, 0, 0, 0, 0, 0, 0, 0 ]
relax_sop = mb.qm_lindblad_relaxation(rates, pure_deph)

# initial density matrix
rho_init = mb.qm_operator([ 0.3705, 0.4937, 0.0741, 0.0333, 0.0285])

h = 15e-6
w = 60e-6
A = h*w
N = 6.35e21
N_x = 2000
l_device = 4e-3
d_x = l_device / N_x
f_noise = 1
N_cell = N * A * d_x * f_noise

qm = mb.qm_description(N, N_cell, H, u, relax_sop)
loss = 760
mat_ar = mb.material("AR_Forrer", qm, 12.96, 1, loss, 1.0)
mb.material.add_to_library(mat_ar)

dev = mb.device("51v1")

dev.add_region(mb.region("Active region", mat_ar, 0.0, l_device))

# Scenario
ic_d = mb.ic_density_const(rho_init)
ic_e = mb.ic_field_const(0.0)
sce = mb.scenario("hc_noise_forrer2021", N_x, 2200e-9, ic_d, ic_e, ic_e)
sce.add_record(mb.record("e1", 0.4e-3))

# run solver
sol = mb.solver.create_instance("cpu-fdtd-51v1-reg-cayley-qnoise", dev, sce)
print('Solver ' + sol.get_name() + ' started')
tic = time.time()
sol.run()
toc = time.time()
print('Solver ' + sol.get_name() + ' finished in ' + str(toc - tic) + ' sec')

# write results
wri = mb.writer.create_instance("hdf5")
outfile = dev.get_name() + "_" + sce.get_name() + "." + wri.get_extension()
results = sol.get_results()
wri.write(outfile, sol.get_results(), dev, sce)
outfile_autosave = dev.get_name() + "_" + sce.get_name() + \
    "_autosave." + wri.get_extension()
sim_data = sol.get_sim_data()
wri.autosave(outfile_autosave, sim_data, dev, sce)

```

APPENDIX D: mbsolve SIMULATION SETUPS: THz HFC QCL SETUP

Here, the Python script for setting up and running the mbsolve simulation of the THz HFC QCL setup is given. The script includes all input parameters for the description of the quantum system and the simulation scenario. The quantum mechanical description comprises the level occupations ρ_{ii} , the system Hamiltonian $\hat{H}_s$ with eigenenergies E_i and anticrossing energies $\hbar\Omega_{ij}$, the dipole moment operator $\hat{d}_z$, the scattering rates r_{ij} , and the dephasing rates γ_{ij} . For the one-dimensional dynamic Maxwell-density matrix simulations, the energy-resolved dephasing rates are simulated within the EMC approach and have to be averaged over the population inversions of the involved subbands, as it is described in detail in Refs. 77 and 76.

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