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Complex dynamics in quantum dot light emitting diodes

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Abstract. We report both experimentally and theoretical investigation the appearance of Mixed Mode Oscillations (MMOs) and chaotic spiking in a Quantum Dot Light Emitting Diode (QDLED). In the theoretical treatment the proposed model reproduces Homoclinic Chaos (HC) and MMOs. The dynamics is completely determined by the variation of the injecting bias current in the wetting layer of the QDLED. The influence of the injecting current on the transition between Mixed Mode Oscillations and chaotic behavior has been also investigated. It was found that the theoretical model verifies the experimental findings.

1 Introduction

Most light sources exhibit intensity and phase fluctuations due to the nature of the quantum transition process itself. These fluctuations are very important when the stability is a prior. In fact, every spontaneous emission event in the oscillating mode causes a phase variation of the electromagnetic field. This variation is defined as a quantum noise and it is responsible for the carrier density variation.

Semiconductor nitrides such as aluminum-nitride (AlN), gallium-nitride (GaN), and indium-nitride (InN) are commonly utilized as materials for their potential use in high-power and high-temperature optoelectronic devices. The commonly quoted value for the optical band gap of InN was 1.89 eV [1]. Nevertheless other values are obtained by changing the state [2]. The properties of III-N QDs are closely related to those of bulk materials [3]. Although bulk LEDs can be found as wurtzite, zinc blend, and rock salt structures, the wurtzite structure is more dominant in thermodynamically stable condition. III-nitride QDs are commonly the strained systems [4].

In addition to chaotic spiking, recent studies involving surface chemical reactions [5–7], electrochemical systems [8,9] neural and cardiac cells [10,11], calcium dynamics [12], and plasma physics [13] showed that oscillatory dynamics often takes place in the form of complex temporal sequences known as Mixed Mode Oscillations (MMOs) [14]. However, periodic-chaotic sequences and Farley sequences of MMOs do not necessarily involve a torus or a homoclinicorbit. They can occur also through the canard phenomenon [15,16]. Here a limit cycle born at a supercritical Hopf bifurcation experiences the abrupt transition from a small-amplitude quasi-harmonic cycle to large relaxation oscillations in a narrow parameter range. Most studies of this dynamics have been carried out in chemical systems. Nevertheless in semiconductor laser systems with optoelectronic feedback incomplete homoclinic scenarios have been recently predicted and observed [17,18].

The main goal of this work is to provide a physical model reproducing qualitatively the experimental results and showing that chaotic spiking and MMOs are a consequence of the modulating bias current of the QDLED. In self-feedback in QDLED a dramatic change in the photon statistics where a strong super-thermal photon bunching is indicative of random-intensity fluctuations associated with the spike emission of light is ignored. Our experiments reveal that spiking competition of quantum dot in the active layer enhances the influence of self-feedback and modulating perturbation thus opening up new avenues for the study of chaos in quantum systems. Theoretically, we proved that the model verifies the experimental findings.

2 Experiment

A common aspect of spiking in the output of the QDLEDs is the presence of feedback that couples the output signal partially back to the input [19]. We consider an open-loop optical system, consisting of a QDLED source emitting at

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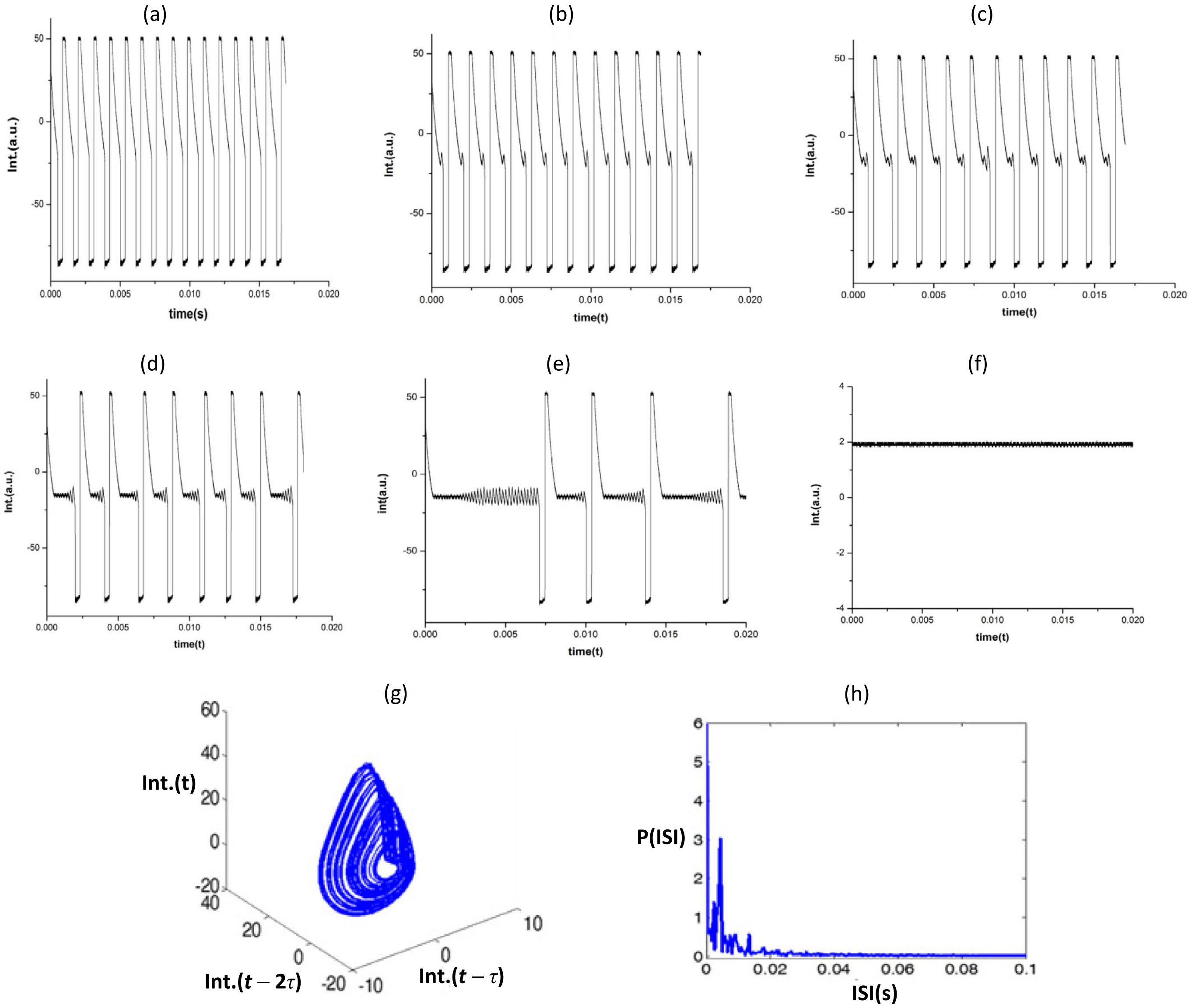

Fig. 1. Transition from periodic to chaotic spiking to and steady state as the dc-pumping current is varied (a) 6.2 mA, (b) 5.52 mA, (c) 5.35 mA, (d) 5.21 mA, (e) 5.01 mA, (f) 4.700 mA; (g) experimental reconstruction of the phase portrait through the embedding technique; (h) corresponding experimental ISI probability distribution for the chaotic spiking regime.

650 nm wavelength. The QDLED operates with variable driven current. The light is sent to a photodetector whose output current is proportional to the optical intensity. The corresponding signal is sent to a variable gain amplifier characterized by a nonlinear transfer function of the form $f(w) = Aw/(1 + \beta w)$, where A is the amplifier gain and βa saturation coefficient, then to a fast (500 MHz) digital oscilloscope. By decreasing the dc-pumping current which plays good control parameter to see several behaviors for our system, we observe the dynamical sequence shown in Figures 1a–1f. In Figure 1a we will see regular spiking at the current (6.2 mA), more controlling to control parameter, transition from periodic dynamic passing to MMOs will happen as shown in Figures 1b–1d, corresponding to the lowest current (5.01 mA), the detected optical power is in a chaotic spiking regime where large intensity pulses are separated by irregular time intervals in which the system displays small-amplitude chaotic oscillations (Fig. 1e). Typical time traces are characterized by a mixture of large-amplitude relaxation spikes (L) followed by small-amplitude (S) quasi-harmonic oscillations, where oscillations intermediate between L and S do not occur). At even larger current decrease (4.7 mA), the firing vanishes and eventually a steady state is established as shown in Figure 1f. This scenario is qualitatively similar to Homoclinic Chaos (HC) as previously observed in semiconductor lasers and LEDs with optoelectronic feedback [17]. In HC signals, the pulse duration (associated with a precise orbit in the phase space) is uniform, while the interpulse times vary irregularly. This is shown by the corresponding interspike interval (ISI) probability distribution (Fig. 1e). However, there are no external perturbations

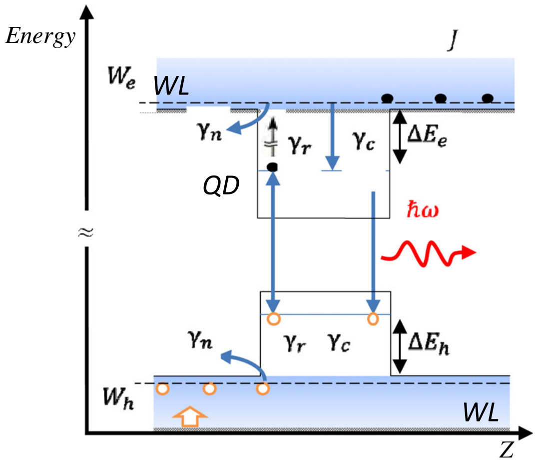

Fig. 2. Energy diagram illustrates the two recombination mechanisms considered in this work of the active layer QDLED; recombination radiative and non-radiative via deep level and reabsorption recombination processes.

that mean our system is autonomous system, the small chaotic background is clearly larger than the residual electronic noise, and the ISI histogram displays a structure of sharp peaks that could correspond to unstable periodic orbits embedded in the chaotic attractor [10]. We will show in the next section that a fully deterministic theoretical model explains such a distribution.

3 The model

In this study, we propose a rate equations model of the three-level system. Hereby we will model QDLED structures by rate equations (Fig. 2). In the QDLED system the electrons are first injected into the wetting layer (WL) before they are captured by the QDs. We consider a system made up of upper and lower electronic levels.

The equations describe the dynamics of the total population of carriers in the upper levels, and the number of photons in the optical mode s, as follows

$$\begin{aligned}
\dot{s} &= An_{QD} - ds - \Gamma s,\\
\dot{n}_{QD} &= \gamma_c n_{wl}\left(1 - \frac{n_{QD}}{2N_d}\right) - \gamma_r n_{QD} - (An_{QD} - ds),\\
\dot{n}_{wl} &= \frac{J}{e} - \gamma_n n_{wl} - \gamma_c n_{wl}\left(1 - \frac{n_{QD}}{2N_d}\right),
\end{aligned} \quad (1)$$

here, A is the spontaneous emission rate into the optical mode, γ_r and γ_n are the non-radiative decay rates of the population of carriers in the upper levels and WL respectively, N_d is the two-dimensional density of dots; J is the injection current, e is electron charge, γ_c is the capture rate from WL into an empty dot, and d and Γ are the absorption and output coupling rate of photons in the optical mode, respectively.

For a two-level atomic system where the transition is homogeneously broadened, it can be shown from the Einstein relation that [20].

$$d = An_0 \quad (2)$$

where n_0 is the occupation number of the lower level. In such a case, the spontaneous emission coefficient and absorption coefficient possess identical line shapes. For realistic material systems, such as semiconductor or organic emitters, both the lower and the upper levels can be considered as inhomogenously broadened. Population distributions in the lower and upper levels have to be taken into account explicitly in order to determine the correct relation between absorption and spontaneous emission spectra [21].

In this section we investigate how to predict unstable behavior and spiking generation from QDLED equations without external feedback. To do so, we rescale the system (1) to a set of dimensionless equations. Defining new variables and dimensionless parameters by: $x = s$.

$$\begin{aligned}
&y = \frac{A}{\Gamma}(n - n_0 s),\ w = \frac{n_{wl}\gamma_c}{A},\ \gamma_n = \frac{t}{t^2},\\
&\gamma = \frac{\Gamma}{\gamma_n},\ \gamma_1 = \frac{A}{\gamma_n}, \gamma_2 = \frac{A}{\Gamma},\ \gamma_3 = \frac{\gamma_r}{\gamma_n},\ \gamma_4 = \frac{\gamma_c}{\gamma_n},\\
&N_d \equiv a, n_0 \equiv b \text{ and } \delta_0 = \frac{J}{Ae}.
\end{aligned}$$

Equations (1) can be rewritten in a dimensionless form as the following

$$\begin{aligned}
\dot{x} &= \gamma(y - x)\\
\dot{y} &= \gamma_1\gamma_2 w(1 - bx/a) - \gamma_1 y(1 + w/a)\\
&\quad - b\gamma_1(x - y) - \gamma_3(y + \gamma_2 x)\\
\dot{w} &= \gamma_4\delta_0 - w(1 - y\gamma_4/a\gamma_2) - \gamma_4 w(1 - bx/a).
\end{aligned} \quad (3)$$

Here prime means differentiation with respect to t^2 and the bias current density is represented by δ_0.

4 Direct current modulation

The developed QDLED rate equation model predicts unstable behavior, we also investigate the transition between periodic and chaotic states and analyze the effects of direct and periodic modulating injection current on the chaotic attractors using the same theoretical model by adding the perturbation term. The output of the system will be chaotic. There are several possibilities to accomplish these conditions [22]. The simplest one is by modulating the injection current periodically with a modulating frequency f_m. The injection current in the pumping term $\frac{J}{e}$ equations (1) and (3) has to be replaced by a source of injection current like

$$J = I_0 + I_\mathrm{ac}\sin(2\pi f_\mathrm{m} t), \quad (4)$$

where I_0 is the dc part of the injection current and I_ac is the amplitude of the ac part of the injection current.

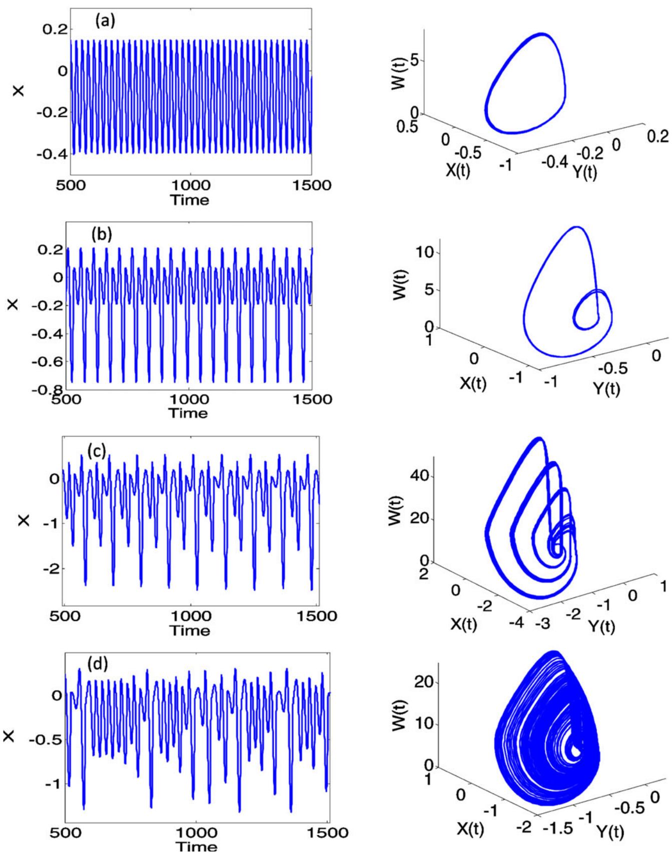

Fig. 3. Numerical simulations of the model equations. Left panels: time series for the light intensity. Right panels: reconstructed phase space for the chaotic spiking regime for selected bias current (a) $\delta_0 = 6$, (b) $\delta_0 = 4$, (c) $\delta_0 = -0.5$, (d) $\delta_0 = 0.1$ correspond to $\delta_0 = 6$–0.1 as indicated in the first bifurcation. The other parameters are: $x_0 = 0.066$, $y_0 = 0.99$, $w_0 = 0.0049$, $\gamma = 0.127$, $\gamma_1 = 0.144$, $\gamma_2 = 0.03$, $\gamma_3 = 0.07$, $\gamma_4 = 0.087$, $a = 1.04$ and $b = 3.838$.

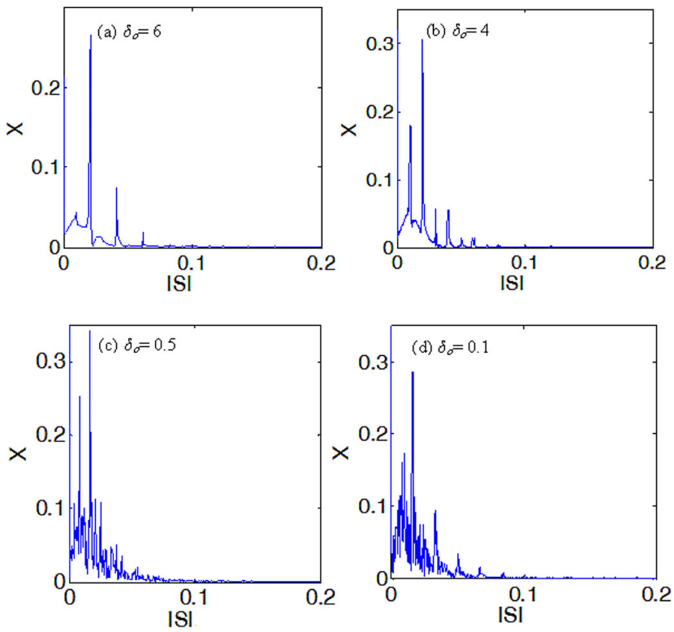

Fig. 4. The ISI distribution of the conditions as in Figure 2, when (a) $\delta_0 = 6$, (b) $\delta_0 = 4$, (c) $\delta_0 = 0.5$, (d) $\delta_o = 0.1$. The other parameters are: $x_0 = 0.066$, $y_0 = 0.99$, $w_0 = 0.0049$, $\gamma = 0.127$, $\gamma_1 = 0.144$, $\gamma_2 = 0.03$, $\gamma_3 = 0.07$, $\gamma_4 = 0.087$, $a = 1.04$ and $b = 3.838$.

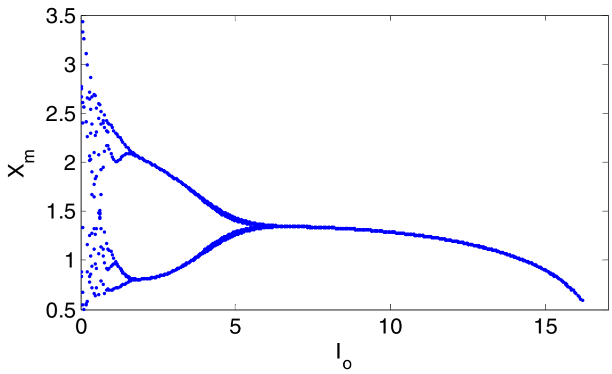

Fig. 5. Bifurcation diagram for the model equations. The parameter values are set as in Figure 3.

5 Numerical results

The rate equations (3) are solved numerically using the fourth -order Runge-Kutta method with 0.01 ns step time size. The parameter values used in the simulations are assigned as $x_0 = 0.066$, $y_0 = 0.99$, $w_0 = 0.0049$, $\gamma = 0.127$, $\gamma_1 = 0.144$, $\gamma_2 = 0.03$, $\gamma_3 = 0.07$, $\gamma_4 = 0.087$, $a = 1.04$ and $b = 3.838$. The injection parameters, i.e., the dc bias current, I_0, or the dc bias strength, the modulation current, I_{ac}, or modulation depth, and modulation frequency, f_{m}, have been chosen carefully. The chaotic spiking regime arises from the interplay of the large phase-space orbits mentioned before and a period doubling route to chaos occurring in the vicinity of the turning point (Fig. 3) shows the time series and the corresponding attractors and ISI (Fig. 4) for different current injection current δ_0 values.

For WL, since it has a large number of states (it is considered as a continuum state), its carrier number is always higher than zero as. There are a limited number of states in QD structure, then the ground state is filled and emptied due to the capturing and relaxation processes (positive γ_c and negative γ_r parts in the second equation of the system, Eqs. (1)). Thus, the behavior of the carrier number in QD goes between positive and negative values as shown in the left panel of Figure 3. So, the negative part of photon number shown in Figure 3 is due to both absorption and emission and it is also shown in experimental works in bulk LED.

As the model, the emitted photon which depends entirely on the spontaneous emission shows that the extent of absorption plays a bigger role in the output.

In addition to the chaotic behavior in the QDLED output MMOs also have been found. In MMOs mixture of L large amplitude relaxation spikes followed by S small-amplitude quasi-harmonic oscillations.

The bifurcation diagram (Fig. 5) is computed by varying δ_0 over a small interval contiguous to the initial Hopf bifurcation. As δ_0 approaches the turning point (when its

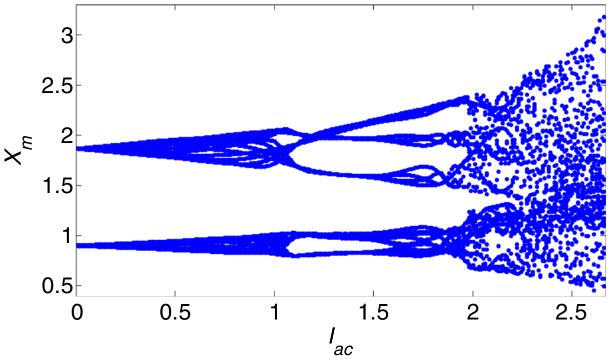

Fig. 6. Bifurcation diagram of the photon density as a function of modulation depth ($I_{\rm ac}$) for the dc bias strength value $\delta_0 = 0.6$ and modulation frequency $f_{\rm m} = 2.4 \times 10^{-2}$.

value becomes less than 4) the chaotic amplitude fluctuations are sufficiently large to trigger fast dynamics. This results in an erratic-sensitive to initial condition-sequence of homoclinic spikes on top of a chaotic background.

We have eventually studied the chaotic spiking dynamics as the parameter modulation depth ($I_{\rm ac}$) is increased. As in the experiment, the increasing of the modulation depth by around 2 leads to faster chaotic spiking regimes until the duration of the slow-fast pulses becomes of the order of the chaotic background characteristic timescale (Fig. 6). On the basis of the analysis in the previous section, it is now clear that the period of the phase-space orbit is fully determined by the QDLED timescales. Sequences of this type are ubiquitous in nature and are originally observed in chemical systems more than 100 years ago [23], with the Belouzov-Zhabotinsky reaction being the most thoroughly studied example [24–27].

In conclusions, we have demonstrated experimentally and theoretically the existence of chaotic behavior and mixed mode oscillations sequences in the dynamics of a quantum dot light emitting diode with optoelectronic feedback. The timescale of these dynamics is fully determined by the ISI probability distribution of the QDLED output.

Remarkably, on all occasions, the control by changing the bias current works successfully. We eventually provided a feasible minimal model reproducing qualitatively the experimental results and allowing an interpretation in terms of chaotic and mixed mode oscillations dynamics. In order to improve the quantitative agreement between theory and experiments several modifications of the rate equations have been proposed considering intrinsically quantum effects such as non-irradiative decay rates of the population of carriers in the WL. Experiments concerning synchronization phenomena in QDLEDs arrays are currently in preparation and will be the subject of the future work.

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