Two-particle quantum correlations on nano and macro single-mode lasers

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ABSTRACT

We compare a novel quantum mechanical model of lasers, which includes all two-particle correlations, with the Coherent-Incoherent Model (CIM), where truncation eliminates these effects. Numerical results for the simple case of identical single-electron quantum dots are presented for two cases: only the laser mode is coupled to the quantum dots; the coupling is extended to non-resonant modes. We find coexistence between non-lasing and lasing states, together with a minimum number of photons required to initiate the laser action. The correlations introduce a non-zero variance in the field, which is otherwise strictly zero in the absence of interparticle correlations.

Keywords: Nanolaser, quantum model, two-particle correlations, bifurcations, variance, laser threshold

1. INTRODUCTION

The journey of laser miniaturization, inaugurated by the advent of VCSELs,[1,2] has uncovered a realm of novel phenomena within the micro- and nanoscale domains. While the laser threshold was successfully modelled as a clear phase transition,[3–6] miniaturizatin advancements definitely blurred its delineations,[7–14] giving rise to enigmatic notions such as "zero-threshold" and "thresholdless laser",[15–18] with the latter term finding a permanent place in the literature.

The notion of a thresholdless laser was criticized early on[19] on the basis of scaling arguments, which, however, could not be extended to the nanoscale. Separate efforts led to alternative pictures[20] whose physical relevance remain unclear. This failure stems from the fact that traditional rate-equation-based models, while effective in many respects, lack essential physical ingredients necessary for accurately characterizing the laser threshold at the nanoscale. Consequently, a protracted debate has unfolded over nearly three decades, grappling with a myriad of issues. Fundamental inquiries have emerged regarding the very concept of threshold and its operational definition,[21,22] as well as the challenges associated with threshold measurement and identification.[23,24] Techniques aimed at identifying threshold have been proposed and scrutinized,[25–29] contributing to the ongoing discourse surrounding this intriguing phenomenon.

The growing interest in understanding, defining, and measuring the laser threshold, as well as comprehending the underlying physics of these devices,[30–33] is propelled by the rapid technological advancements in ultrasmall sources.[34–37] This surge in interest is driven by the allure of ultralow pumping requirements,[38–42] which are conducive to integration into optical circuits,[43–45] aligning with the objectives of green photonics.[46,47]

Significant strides in technology have been paralleled by extensive modeling endeavors, with the aim of enhancing theoretical descriptions to encompass emerging features and novel physical effects.[10,48–54] While these efforts have yielded notable successes, the ongoing need for continual adjustments to existing models underscores the potential for broader enhancements. This persistent requirement for model refinement suggests opportunities

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for overarching improvements in our theoretical frameworks, indicating a dynamic and evolving landscape within the field of laser physics modeling.

A crucial element in the debate is the difficulty of experimentally characterizing the physics of nano-lasers due to their extremely small photon flux. The lack of detectors with sufficient sensitivity and bandwidth forces the use of well-developed statistical measurements of photon counting, which are routinely used in quantum optics. Unfortunately, the statistical picture provides information from which the dynamics cannot be inferred:[55] the inference from dynamics to statistics works only in this direction, but is not reversible. Thus, it is imperative to develop models that capture the essence of nanoscale threshold physics in order to make meaningful comparisons between measurements and predictions.

In this paper, we present a model that goes beyond the standard semiclassical description by incorporating correlations between photons and emitters. This model is compared to an existing framework that includes spontaneous and stimulated processes, with the goal of providing a more comprehensive understanding of nanolaser dynamics. The increased sophistication of our model not only reproduces established results, but also reveals novel phenomena, including the coexistence of non-lasing and lasing solutions and the requirement of a minimum photon threshold to initiate lasing.

Interestingly, parallels to our theoretical predictions are observed in experimental investigations of microlasers, where direct dynamical measurements have unveiled a pre-threshold regime characterized by the emission of photon bursts.[31,56,57] These phenomena, also observed in phenomenological stochastic simulations,[58,59] potentially correspond to the experimental manifestations of our theoretical conjectures. The comprehensive classification of laser scales provided in,[60] helps in contextualizing our theoretical framework within existing experimental configurations.

As is customary in nanolaser descriptions, we parameterize the effective coupling of radiation into the lasing mode using the well-known β parameter, defined as the ratio of the spontaneous emission rate into the lasing mode to the total spontaneous emission rate, $\beta = \left[\frac{\gamma_l}{(\gamma_{nl}+\gamma_l)}\right]$, where γ_l and γ_{nl} denote the spontaneous emission rates into the lasing and non-lasing modes, respectively. This parameterization allows for a quantitative characterization of the nanolaser's behavior in terms of its coupling efficiency.

Specifically, in a more refined framework, we introduce a laser model labeled as the Two-Particle Model (TPM), which accounts for all two-particle quantum correlations. This model is compared to the previously derived Coherent-Incoherent Model (CIM), which simplifies the physical description by neglecting these correlations. Through numerical simulations, we examine the behavior of the TPM in a basic scenario involving identical, single-electron quantum dots. We explore two distinct conditions: first, when only the laser mode is coupled to the quantum dots, and second, when the coupling extends to non-resonant modes alongside the laser mode.

2. THE MODEL

The quantized total Hamiltonian is $H = H_{\mathrm{E}} + H_{QD} + H_{int}$, where

$$H_{\mathrm{E}} = h \sum_q \nu_q \left(b_q^\dagger b_q + \frac{1}{2} \right), \tag{1}$$

$$H_{QD} = \sum_n \left(\varepsilon_{c,n} c_n^\dagger c_n + \varepsilon_{v,n} v_n^\dagger v_n \right), \tag{2}$$

$$H_{int} = -ih \sum_{n,q} \left[g_{nq} b_q c_n^\dagger v_n - g_{nq}^* b_q^\dagger v_n^\dagger c_n \right], \tag{3}$$

correspond the energies of the electric field, of the charge carriers and of the light-matter interaction in the dipole approximation, respectively. ν_q is the frequency of a q-th mode photon, $\epsilon_{c,n}$, $\epsilon_{v,n}$ are the energies and $c_n, c_n^\dagger$ and $v_n, v_n^\dagger$ the annihilation and creation operators for conduction and valence electrons of the n-th quantum dot, respectively, and g_{nq} is the strength of light-matter coupling of q-th mode in the n-th quantum dot.

The Heisenberg equations for the one and two particle operators including non radiative losses, polarization dephasing and cavity mode losses due to coupling to a Markovian bath are

$$d_t b_s = -(\gamma_s + i\nu_s) b_s + \sum_n g_{ns}^* v_n^\dagger c_n \tag{4a}$$

$$d_t v_l^\dagger c_l = -(\gamma + i\nu_{\varepsilon_l}) v_l^\dagger c_l + \sum_q g_{lq} \left[2b_q c_l^\dagger c_l - b_q\right] \tag{4b}$$

$$d_t c_l^\dagger c_l = -\gamma_{nr} c_l^\dagger c_l - \sum_q \left[g_{lq} b_q c_l^\dagger v_l + \text{h.c.}\right] \tag{4c}$$

$$d_t b_q^\dagger b_q = -2\gamma_q b_q^\dagger b_q + \sum_n \left[g_{nq} b_q c_n^\dagger v_n + \text{h.c.}\right] \tag{4d}$$

$$d_t b_s c_l^\dagger v_l = -(\gamma + \gamma_s + i\Delta\nu) b_s c_l^\dagger v_l + g_{ls}^* c_l^\dagger c_l - \sum_q g_{lq}^* \left(2b_q^\dagger b_s c_l^\dagger c_l - b_q^\dagger b_s\right) + \sum_{n\neq l} g_{ns}^* c_l^\dagger v_n^\dagger c_n v_l \tag{4e}$$

$$d_t b_s c_l^\dagger c_l = -(\gamma_s + \gamma_{nr} + i\nu_s) b_s c_l^\dagger c_l \; - b_s \sum_q \left(g_{lq} b_q c_l^\dagger v_l + h.c.\right) + \sum_{n\neq l} g_{ns}^* v_n^\dagger c_l^\dagger c_l c_n \tag{4f}$$

$$d_t b_s b_s = -2(\gamma_s + i\nu_s) b_s b_s + 2\sum_n g_{ns}^* b_s v_n^\dagger c_n \tag{4g}$$

$$d_t b_s v_l^\dagger c_l = -[\gamma + \gamma_s + i(\nu_{\varepsilon_n} + \nu_{\varepsilon_l})] b_s v_l^\dagger c_l + g_{ls} b_s b_s \left(2c_l^\dagger c_l - 1\right) + \sum_{n\neq l} g_{ns}^* v_n^\dagger v_l^\dagger c_l c_n \tag{4h}$$

$$\begin{aligned} d_t c_l^\dagger v_n^\dagger c_n v_l = &-[2\gamma + i(\nu_{\varepsilon_n} - \nu_{\varepsilon_l})] c_l^\dagger v_n^\dagger c_n v_l + \sum_q \Big[g_{lq}^* \left(2b_q^\dagger v_n^\dagger c_l^\dagger c_l c_n - b_q^\dagger v_n^\dagger c_n\right) \\ &+ g_{nq}\left(2b_q c_l^\dagger c_n^\dagger c_n v_l - b_q c_l^\dagger v_l\right)\Big] \end{aligned} \tag{4i}$$

$$\begin{aligned} d_t v_n^\dagger c_l^\dagger c_l c_n = &-(\gamma + \gamma_{nr} + i\nu_{\varepsilon_n}) v_n^\dagger c_l^\dagger c_l c_n + \sum_q \Big[g_{nq}\left(2b_q c_n^\dagger c_l^\dagger c_l c_n - b_q c_l^\dagger c_l\right) - g_{lq} b_q c_l^\dagger v_n^\dagger c_n v_l \\ &- g_{lq}^* b_q^\dagger v_n^\dagger v_l^\dagger c_l c_n\Big] \end{aligned} \tag{4j}$$

$$d_t c_n^\dagger c_l^\dagger c_l c_n = -2\gamma_{nr} c_n^\dagger c_l^\dagger c_l c_n - \sum_q \left[g_{nq} b_q c_n^\dagger c_l^\dagger c_l v_n + g_{lq} b_q c_l^\dagger c_n^\dagger c_n v_l + \text{h.c.}\right] \tag{4k}$$

$$\begin{aligned} d_t v_n^\dagger v_l^\dagger c_l c_n = &-[2\gamma + i(\nu_{\varepsilon_n} + \nu_{\varepsilon_l})] v_n^\dagger v_l^\dagger c_l c_n + \sum_q \Big[g_{nq}\left(2b_q v_l^\dagger c_n^\dagger c_n c_l - b_q v_l^\dagger c_l\right) \\ &+ g_{lq}\left(2b_q v_n^\dagger c_l^\dagger c_l c_n - b_q v_n^\dagger c_n\right)\Big], \end{aligned} \tag{4l}$$

where ν_s, $\nu_{\varepsilon_n} = \varepsilon_n/h$ are the frequencies of the s-th mode and the radiative transition of the n-th QD, γ_{nr} is the population decay rate due to non radiative losses, γ the decay rate of the polarization and γ_s the decay rate of the s-th mode.

These equation contain three-particle operators because the Hamiltonian H and the Lindblad diffusors included to take into account for the losses[61] give rise to an infinite hierarchy of coupled equations of motion for operators of all orders. We obtain a finite-dimensional model of quantum dots in a laser by transforming the Heisenberg equations into equations for the expectation values and by truncating the expectation value equations assuming that all quantum correlations above the second order are negligible and can be set to zero.[62] The decomposition of the expectation values of three particle operators in the sum of the three particle correlations and products of single and two particle expectation values is

$$\langle O_i O_j O_k\rangle = \delta\langle O_i O_j O_k\rangle + \langle(O_i O_j O_k)\rangle, \tag{5}$$

where

$$\langle(O_i O_j O_k)\rangle = \langle O_i\rangle\langle O_j O_k\rangle + \langle O_j\rangle\langle O_i O_k\rangle + \langle O_k\rangle\langle O_i O_j\rangle - 2\langle O_i\rangle\langle O_j\rangle\langle O_k\rangle\,. \tag{6}$$

In numerical studies it is necessary to calculate the expectation values in the rotating frames in order to eliminate the fast frequencies. The relations between expectation values in the Heisenberg picture and in the rotating frame are:

$$\langle b\rangle = \frac{\langle b\rangle_H}{N^{1/2}} e^{i\nu t}, \tag{7a}$$
$$\langle v^\dagger c\rangle = \langle v^\dagger c\rangle_H e^{i\nu t}, \tag{7b}$$
$$\langle bc^\dagger c\rangle = \frac{\langle bc^\dagger c\rangle_H}{N^{1/2}} e^{i\nu t}, \tag{7c}$$
$$\langle v^\dagger c^\dagger cc\rangle = \langle v^\dagger c^\dagger cc\rangle_H e^{i\nu t}, \tag{7d}$$
$$\langle bv^\dagger c\rangle = \frac{\langle bv^\dagger c\rangle_H}{N^{1/2}} e^{i2\nu t}, \tag{7e}$$
$$\langle bb\rangle = \frac{\langle bb\rangle_H}{N} e^{i2\nu t} \tag{7f}$$
$$\langle v^\dagger v^\dagger cc\rangle = \langle v^\dagger v^\dagger cc\rangle_H e^{i2\nu t}, \tag{7g}$$

where we indicate with an H suffix the expectation values in the Heisenberg picture. We also rescale the coupling coefficient with the number of quantum dots N as

$$g = g_H N^{1/2}\,. \tag{8}$$

The equations for the expectation values in the rotating frame are:

$$d_t\langle b\rangle = -\gamma_c\langle b\rangle + g^*\langle v^\dagger c\rangle, \tag{9a}$$
$$d_t\langle v^\dagger c\rangle = -\left(\gamma - i\Delta\nu\right)\langle v^\dagger c\rangle + g\left(2\langle bc^\dagger c\rangle - \langle b\rangle\right). \tag{9b}$$
$$d_t\langle c^\dagger c\rangle = -\gamma_{nr}\langle c^\dagger c\rangle - \left(g\langle bc^\dagger v\rangle + \text{h.c.}\right) + r\left(1 - \langle c^\dagger c\rangle\right) - \gamma_{nl}\langle c^\dagger c\rangle, \tag{9c}$$
$$d_t\langle b^\dagger b\rangle = -2\gamma_c\langle b^\dagger b\rangle + \left(g\langle bc^\dagger v\rangle + \text{h.c.}\right), \tag{9d}$$
$$d_t\langle bc^\dagger v\rangle = -\left(\gamma + \gamma_c + i\Delta\nu\right)\langle bc^\dagger v\rangle + g^*\left[\frac{\langle c^\dagger c\rangle}{N} + 2\left\langle\left(b^\dagger bc^\dagger c\right)\right\rangle - \langle b^\dagger b\rangle\right] + \frac{(N-1)}{N} g^*\langle c^\dagger v^\dagger cv\rangle \tag{9e}$$
$$\begin{aligned} d_t\langle bc^\dagger c\rangle =& -\left(\gamma_c + \gamma_{nr}\right)\langle bc^\dagger c\rangle - g\left\langle\left(bbc^\dagger v\right)\right\rangle - g^*\left\langle\left(b^\dagger bv^\dagger c\right)\right\rangle + \frac{(N-1)}{N} g^*\langle c^\dagger v^\dagger cc\rangle \\ &+ \langle b\rangle\left[r\left(1 - \langle c^\dagger c\rangle\right) - \gamma_{nl}\langle c^\dagger c\rangle\right] \end{aligned} \tag{9f}$$
$$d_t\langle bb\rangle = -2\gamma_c\langle bb\rangle + 2g^*\langle bv^\dagger c\rangle \tag{9g}$$
$$d_t\langle bv^\dagger c\rangle = -\left[\gamma_c + \gamma - i\Delta\nu\right]\langle bv^\dagger c\rangle + g\left[2\left\langle\left(bbc^\dagger c\right)\right\rangle - \langle bb\rangle\right] + \frac{(N-1)}{N} g^*\langle v^\dagger v^\dagger cc\rangle \tag{9h}$$
$$\begin{aligned} d_t\langle c^\dagger v^\dagger cv\rangle =& -2\gamma(1+\mu)\langle c^\dagger v^\dagger cv\rangle + g^*\left[2\left\langle\left(b^\dagger v^\dagger c^\dagger cc\right)\right\rangle - \left\langle b^\dagger v^\dagger c\right\rangle\right] \\ &+ g\left[2\left\langle\left(bc^\dagger c^\dagger cv\right)\right\rangle - \langle bc^\dagger v\rangle\right] \end{aligned} \tag{9i}$$
$$\begin{aligned} d_t\langle v^\dagger c^\dagger cc\rangle =& -\left[\gamma(1+\mu) + \gamma_{nr} - i\Delta\nu\right]\langle v^\dagger c^\dagger cc\rangle + g\left[2\left\langle\left(bc^\dagger c^\dagger cc\right)\right\rangle - \langle bc^\dagger c\rangle\right] \\ &- g\left\langle\left(bc^\dagger v^\dagger cv\right)\right\rangle - g^*\left\langle\left(b^\dagger v^\dagger v^\dagger cc\right)\right\rangle + \langle v^\dagger c\rangle\left[r\left(1 - \langle c^\dagger c\rangle\right) - \gamma_{nl}\langle c^\dagger c\rangle\right], \end{aligned} \tag{9j}$$
$$\begin{aligned} d_t\langle c^\dagger c^\dagger cc\rangle =& -2\gamma_{nr}\langle c^\dagger c^\dagger cc\rangle - \left[2g\left\langle\left(bc^\dagger c^\dagger cv\right)\right\rangle + \text{h.c.}\right] \\ &+ 2\langle c^\dagger c\rangle\left[r\left(1 - \langle c^\dagger c\rangle\right) - \gamma_{nl}\langle c^\dagger c\rangle\right], \end{aligned} \tag{9k}$$
$$d_t\langle v^\dagger v^\dagger cc\rangle = -2\left[\gamma(1+\mu) - i\Delta\nu\right]\langle v^\dagger v^\dagger cc\rangle + 2g\left[2\left\langle\left(bv^\dagger c^\dagger cc\right)\right\rangle - \langle bv^\dagger c\rangle\right], \tag{9l}$$

where r is the pumping rate and $\gamma\mu$, with $\mu \geq 0$, is the dephasing rate due to phonon scattering.[63] γ_{nl} is a decay rate that takes into account the effect of the non lasing modes, which have been adiabatically eliminated.

3. NUMERICAL RESULTS

In this section we compare the TPM model given in the previous section with the semi-classical Coherent-Incoherent Model (CIM)[64–66] derived including only slowly varying quantum correlations of the type $\delta\langle bC^\dagger v\rangle$.

The CIM equations are (9a) to (9e) after a suitable truncation. We reproduce the CIM equations here for convenience:

$$d_t\langle b\rangle = -\gamma_c\langle b\rangle + g^*\langle v^\dagger c\rangle, \tag{10a}$$

$$d_t\langle v^\dagger c\rangle = -(\gamma - i\Delta\nu)\langle v^\dagger c\rangle + g\langle b\rangle\left(2\langle c^\dagger c\rangle - 1\right). \tag{10b}$$

$$d_t\langle c^\dagger c\rangle = -(\gamma_{nr}+\gamma_{nl})\langle c^\dagger c\rangle - \left(g\langle bc^\dagger v\rangle + \text{h.c.}\right) + r\left(1-\langle c^\dagger c\rangle\right), \tag{10c}$$

$$d_t\langle b^\dagger b\rangle = -2\gamma_c\langle b^\dagger b\rangle + \left(g\langle bc^\dagger v\rangle + \text{h.c.}\right), \tag{10d}$$

$$d_t\langle bc^\dagger v\rangle = -(\gamma+\gamma_c+i\Delta\nu)\langle bc^\dagger v\rangle + g^*\left[\frac{\langle c^\dagger c\rangle}{N} + \langle b^\dagger b\rangle\left(2\langle c^\dagger c\rangle - 1\right)\right] + \frac{N-1}{N}g^*\langle c^\dagger v\rangle\langle v^\dagger c\rangle. \tag{10e}$$

Plots of the power of the laser light versus the pumping rate are easily obtained experimentally and have been extensively considered in investigations of the laser threshold. The light power is proportional to the number of photons, which are generated by both coherent and incoherent processes: as a result, plots of the same lasers calculated with different models give very similar results, as shown in Figure 1. The dependence of the total number of photons on the pump depends on γ_{nl}, i.e. the number of non resonant modes coupled to the QDs, but it is virtually unaffected by two-particle quantum correlations even for lasers with a low number of emitters. The number of photons scales as the number of QDs, which makes the slope of the rising part of the curves steeper for high γ_{nl}.

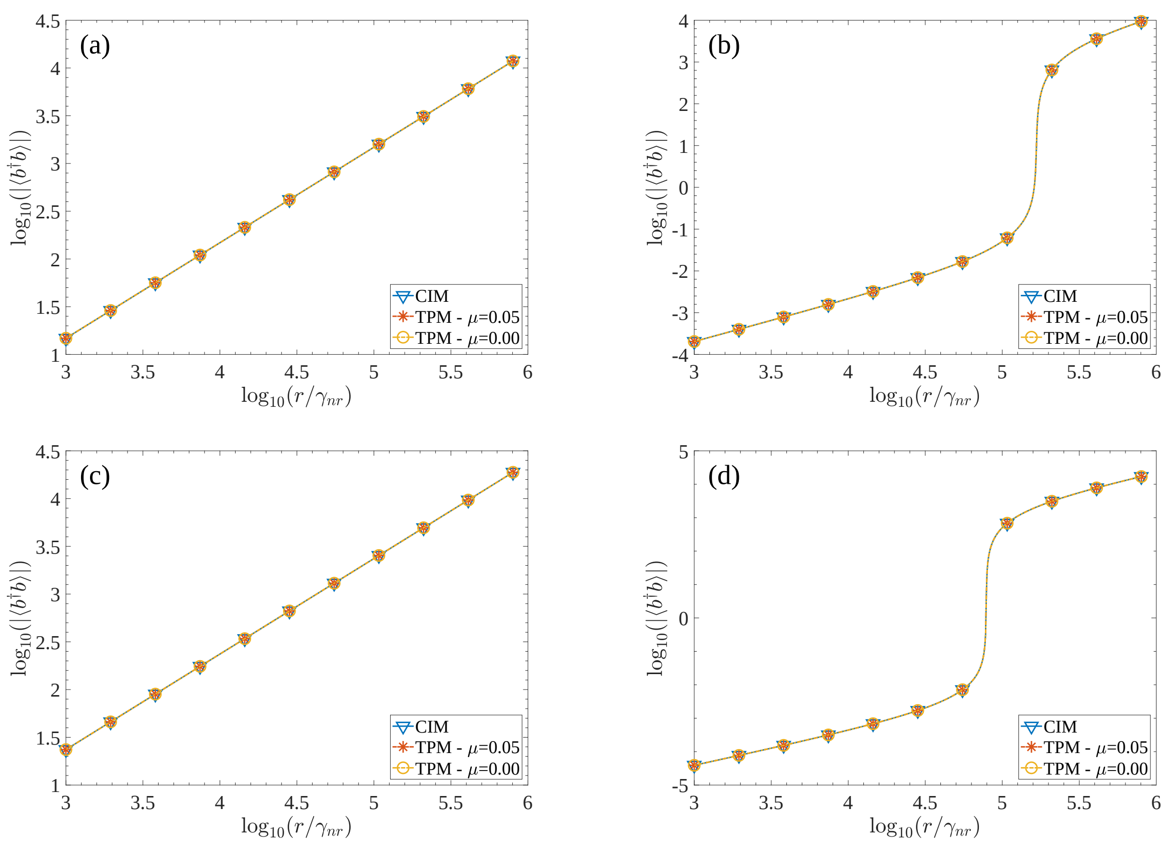

Figure 1. Log-log plot of the number of photons divided by the number of QDs vs the pumping rate r/γ_{nr} for CIM model (blue curve) and TPM with $\mu = 0$ (no phonon scattering, yellow curve) and $\mu = .05$ (red curve). For all figures $g_H = 70$, $\Delta\nu = 0$, $\gamma = 10^4$, $\gamma_c = 10$, $\gamma_l = 0.968$; (a) $N = 50$, $\gamma_{nl} = 0$, (b) $N = 50$, $\gamma_{nl} = 7\times10^4$, (c) $N = 350$, $\gamma_{nl} = 0$, (d) $N = 350$, $\gamma_{nl} = 7\times10^4$. Differences in the numbers of photons for these theories are very small.

The effect of quantum correlations is clear in Figure 2, where we plot $|\langle b\rangle|$ – corresponding to the complex amplitude of the classical field – $|\langle b\rangle|^2/\langle b^\dagger b\rangle$ – the ratio of energy due to coherent emission over the total energy of the field – and the generalized variance $|\langle bb\rangle - \langle b\rangle^2|$ – which is zero for classical fields and provides an estimate of the importance of quantum effects – as functions of the non dimensional pump parameter r/γ_{nr}. We show results for a nanolaser with only the laser mode coupled to the QDs ($\gamma_{nl} = 0$, top row) and a macrolaser with a laser mode and many non resonant modes coupled to the QDs ($\gamma_{nl} = 7 \times 10^4$, bottom row) both with $N = 50$. From the plots of $\langle b\rangle$ – Figure 2(a) and Figure 2(d) – we can see that in the semi-classical CIM stable laser emission grows from zero through a pitchfork bifurcation. The two particle quantum correlations change the nature of the bifurcation: in these models the laser emission always starts with a finite amplitude in a saddle-node bifurcations that also creates a second, unstable laser solution (not shown). In these models lasing can be achieved only if triggered by a finite amplitude fluctuation of the coherent variables and stable lasing and non-lasing solutions can coexist. Figure 2(b) and Figure 2(e) show that at threshold the intensity of the coherent emission, $|\langle b\rangle|^2$, is a significant fraction of the photon number for the TPM, while it is zero for the CIM. Finally, the generalized variance, – Figure 2(c) and Figure 2(f) – which is zero for the semi-classical CIM, is instead different from zero for the TPM, as expected in quantum systems where measurements are associated to probability densities. Furthermore, the generalized variance is always larger at the bifurcation, as are fluctuations due to noise in classical systems.

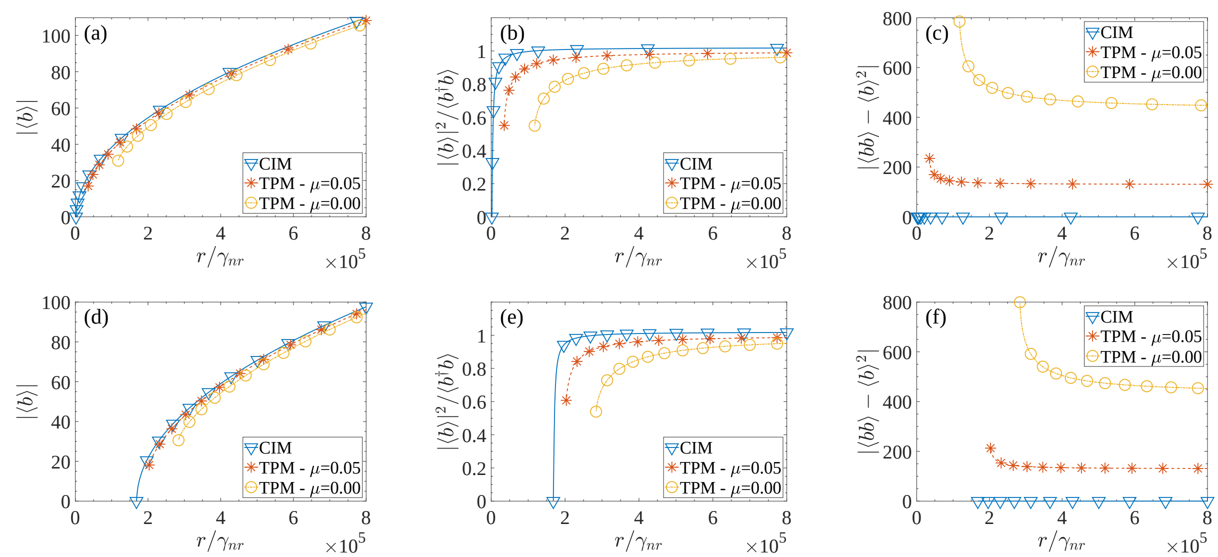

Figure 2. $N = 50$: Nanolaser with same parameter as in Figure 1(a) in (a), (b), (c); macroscopic laser with same parameter as in Figure 1(b) in (d), (e), (f).

In Figure 3 we show for a nanolaser and a macrolaser with $N = 350$ the same type of curves as in Figure 2. The larger number of QDs reduces the thresholds for the CIM and the TPM with weak phonon scattering, while the threshold increases in the absence of phonon scattering. This is due to the competition between the increase in the light matter coefficient g, which scales as $N^{1/2}$, and the increase in the electron-electron correlations, which acts as an increased mass in an oscillator. The variance is reduced in presence of weak phonon scattering.

4. CONCLUSIONS

In conclusion, two particles quantum correlations change the nature of the bifurcation leading to the emergence of continuous laser emission that, for all lasers, appears through a finite amplitude perturbation, as opposed to the infinitesimal amplitude perturbation of semi-classical theories. The effect of quantum correlations on the laser threshold depends on the presence of damping mechanisms such as phonon scattering that produce a fractional increase in the damping rate of two-electron correlations with respect to the dumping rate of the polarization. When these mechanisms are present, we observe a small increase in the threshold and decrease in

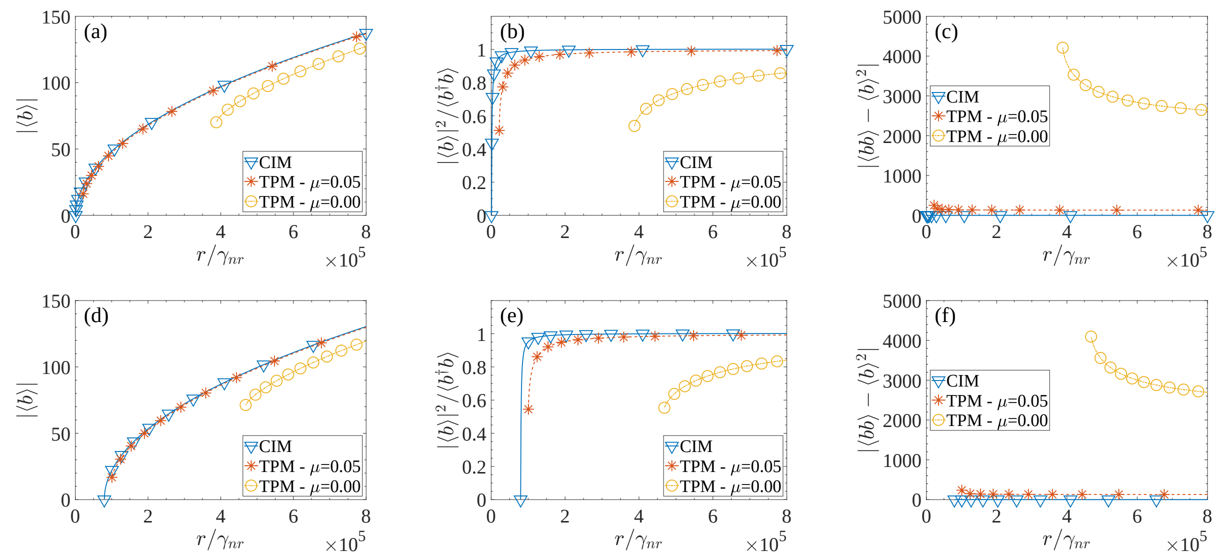

Figure 3. $N = 350$: Nanolaser with same parameter as in Figure 1(c) for (a), (b), (c); macroscopic laser with same parameter as in Figure 1(d) for (d), (e), (f).

the amplitude of the coherent field with a low number of emitters; both effects becomes harder to detect as the number of QDs increases. These effects are more pronounced when two-electron correlations decay at the same rate as the polarization and are actually stronger for larger numbers of QDs. This is associated to an increase in the generalized variance, which is associated to the larger role of quantum correlations in systems without extra damping for two photon correlations.

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