Breaking the Carnot limit without violating the second law: A thermodynamic analysis of off-resonant quantum light generation

E. Boukobza and H. Ritsch
Institut für Theoretische Physik, Universität Innsbruck, Innsbruck 6020, Austria
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The Carnot limit, formulated in 1824, represents the maximal efficiency of a classical heat engine. In this work we present a thermodynamical analysis of a light amplifier based on a three-level atom coupled *off-resonantly* to a single quantized cavity mode and to two heat reservoirs with *positive* temperatures. Based on standard work and heat flow equilibrium, we show that for a cavity blue-detuned with respect to the atomic resonance, the system can surpass the Carnot limit. Nevertheless, the second law of thermodynamics is still obeyed, as the total entropy always increases. By analyzing a semiclassical version of the model, we derive a formula for the critical frequency for which the Carnot limit is broken and a formula for the amplifier efficiency which agrees with its quantum counterpart. In the semiclassical regime, however, the second law is not satisfied and hence it does not offer a physically acceptable description of the system. Finally, we show that breaking the Carnot limit occurs also in a blue-detuned quantum amplifier with output coupling, which represents a realistic model of a laser or maser.

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I. INTRODUCTION

The Carnot limit, formulated in 1824 [1], represents the maximal efficiency of any heat engine operating between two heat reservoirs at different temperatures. It is considered as a corollary of the second law of thermodynamics, which states that in any given process the total entropy must rise.

Thermodynamics of a three-level maser-laser was first studied by Scovil–Schulz-DuBois [2], who heuristically showed that a maser operated in resonance represents a heat engine, whose efficiency is given by a ratio of the signal frequency and the pump frequency which is bounded by the Carnot limit efficiency. Boukobza-Tannor developed a general thermodynamical formalism for bipartite systems coupled to heat reservoirs, in which one degree of freedom can be replaced by a quantized field mode [3], and applied it to a single three-level system coupled to a quantized cavity mode or a classical coherent mode [4]. They showed that under perfect resonance the laser efficiency coincides with the intuitive maser efficiency due to Scovil–Schulz-DuBois and that the quantum and semiclassical models show perfect agreement in terms of steady-state thermodynamic currents.

In 2003, Scully *et al.* [5] theoretically proposed a physical system that challenges the Carnot limit. They suggested a heat engine in which mechanical work might arise from coupling of an optical cavity to two heat reservoirs with the same temperature. In Ref. [5] a movable mirror would be pushed (work) via radiation pressure of photons in an optical cavity interacting with a stream of coherently prepared atoms which serve as one heat reservoir. In addition, a stationary mirror is coupled to a second reservoir whose temperature might be identical to the temperature of the coherent atomic cloud passing through the cavity.

In this paper we present a full quantum thermodynamical analysis of a three-level system coupled to two thermal reservoirs and to a single quantized cavity mode. The thermodynamical analysis incorporated here is based on a quantum master equation treatment for the general bipartite formalism developed in Ref. [3], which continues Alicki's analysis of systems driven by time-dependent external fields [6]. The most remarkable feature of our model is that when the cavity is detuned from atomic resonance the field can be infinitely amplified with an efficiency that surpasses the Carnot limit, while still obeying the second law, i.e., the *total* entropy production function, introduced originally by Spohn [7], is *always* positive.

II. QUANTUM TREATMENT

Consider a three-level system (which we call henceforth atom) interacting with a quantized electric field mode close to its $|1\rangle \rightarrow |2\rangle$ resonance and two thermal photonic reservoirs centered around the $|0\rangle \rightarrow |1\rangle|0\rangle \rightarrow |2\rangle$ resonances as depicted in Fig. 1. The system is governed by the following master equation:

$$\dot{\boldsymbol{\rho}}_{\boldsymbol{af}} = \mathcal{L}_h[\boldsymbol{\rho}_{\boldsymbol{af}}] + \mathcal{L}_{d1}[\boldsymbol{\rho}_{\boldsymbol{af}}] + \mathcal{L}_{d2}[\boldsymbol{\rho}_{\boldsymbol{af}}]. \tag{1}$$

where $\mathcal{L}_h[\boldsymbol{\rho}_{\boldsymbol{af}}]$ and $\mathcal{L}_{d1(2)}[\boldsymbol{\rho}_{\boldsymbol{af}}]$ are the Hamiltonian part of the Liouvillian and the two dissipative Lindblad superoperators, respectively, given by

$$\mathcal{L}_h[\boldsymbol{\rho}_{\boldsymbol{af}}] = -\frac{i}{\hbar}[\boldsymbol{H},\boldsymbol{\rho}_{\boldsymbol{af}}] = -\frac{i}{\hbar}[\boldsymbol{H}_{\boldsymbol{a}} + \boldsymbol{H}_{\boldsymbol{f}} + \boldsymbol{V}_{\boldsymbol{af}},\boldsymbol{\rho}_{\boldsymbol{af}}],$$
$$\mathcal{L}_{d1(2)}[\boldsymbol{\rho}_{\boldsymbol{af}}] = \Gamma_{1(2)}\{(n_{1(2)}+1)([\boldsymbol{\sigma}_{\mathbf{01(2)}}\boldsymbol{\rho}_{\boldsymbol{af}},\boldsymbol{\sigma}^{\dagger}_{\mathbf{01(2)}}] + \text{H.c.}) + n_{1(2)}([\boldsymbol{\sigma}^{\dagger}_{\mathbf{01(2)}}\boldsymbol{\rho}_{\boldsymbol{af}},\boldsymbol{\sigma}_{\mathbf{01(2)}}] + \text{H.c.})\}, \tag{2}$$

where $\boldsymbol{H}_{\boldsymbol{a}} = H_a \otimes \mathbb{1}_f$; $H_a = \hbar\sum_{i=0}^{2}\omega_i|i\rangle\langle i|$ is the bare atomic Hamiltonian; $\boldsymbol{H}_{\boldsymbol{f}} = \mathbb{1}_a \otimes H_f$; $H_f = \hbar\omega_f a^\dagger a$ is the bare field Hamitonian; $\boldsymbol{V}_{\boldsymbol{af}} = \lambda(\sigma_{21}\otimes a^\dagger + \sigma_{21}^\dagger \otimes a)$ is the Jaynes-Cummings Hamiltonian in the rotating wave approximation (RWA) (operators related to reduced spaces are written in regular font, whereas operators related to the full space are written in boldface); $\Gamma_{1(2)}$ are the Weisskopf-Wigner decay constants associated with the two reservoirs; and $n_{1(2)}$ are the average numbers of thermal photons within the two reservoirs. The temperature of each thermal photonic reservoir is positive

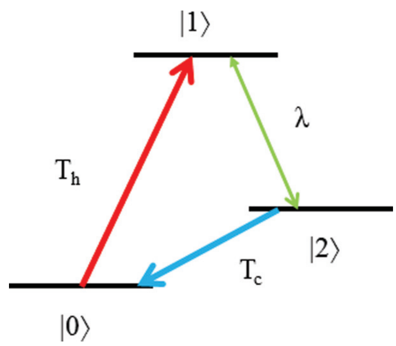

FIG. 1. (Color online) Three-level system interacting with two heat reservoirs (hot and cold) and a quantized cavity mode.

and is given by an inversion of Planck's formula for the average number of thermal photons.

We now define a new energy operator, $\tilde{\boldsymbol{H}} = \boldsymbol{H_a} + \boldsymbol{V_{af}}$, which is a quasi-semiclassical energy operator (see semiclassical treatment), for which we can calculate the energy current of the full atomic-field system, $\dot{\tilde{E}} = \mathrm{Tr}\{\dot{\boldsymbol{\rho}}_{\boldsymbol{af}} \tilde{H}\}$, which is given by

$$\begin{aligned}\dot{\tilde{E}} &= \mathrm{Tr}\{\mathcal{L}_{d(1+2)}[\boldsymbol{\rho_{af}}](\boldsymbol{H_a} + \boldsymbol{V_{af}})\} + \frac{i}{\hbar}\mathrm{Tr}\{\boldsymbol{\rho_{af}}[\boldsymbol{H_f},\boldsymbol{V_{af}}]\}\\ &= \dot{Q}_{1a} + \dot{Q}_{1V} + \dot{Q}_{2a} + \dot{Q}_{2V} - P_f = \dot{Q}_1 + \dot{Q}_2 - P_f,\end{aligned} \tag{3}$$

where $\dot{Q}_{1(2)} \equiv \dot{Q}_{1(2)a} + \dot{Q}_{1(2)V} = \mathrm{Tr}\{\mathcal{L}_{d1(2)}[\boldsymbol{\rho_{af}}]\tilde{\boldsymbol{H}}\}$ and $P_f \equiv -\frac{i}{\hbar}\mathrm{Tr}\{\boldsymbol{\rho_{af}}[\boldsymbol{H_f},\boldsymbol{V_{af}}]\}$ are heat currents and field power, respectively, as defined in Ref. [3].

In what follows we give a complete, thorough thermodynamical steady-state analysis of the $n_1 > n_2$ regime. The total entropy production function for the joint atomic-field system and the reservoirs is given by

$$\sigma = \dot{S}_{af} - \frac{\dot{Q}_1}{T_1} - \frac{\dot{Q}_2}{T_2}, \tag{4}$$

where $S_{af} = -k_B \mathrm{Tr}\{\boldsymbol{\rho_{af}} \ln \boldsymbol{\rho_{af}}\}$ is the von Neumann entropy [8] (analogous to the Gibbs entropy [9] $S = -k_B \sum_i p_i \ln p_i$). Although the entropy production function can be negative (for example, when time-dependent Hamiltonians are involved), examples of its positivity at all times are known (for example, see Refs. [4,10]). When the entropy production function is positive at all times, then one obtains the second law of thermodynamics in differential form at all times, $\sigma \geqslant 0$, without any instantaneous violations of it. If indeed the entropy production function is positive for the system in hand, and if $\dot{\tilde{E}} = 0$, one can write $\dot{Q}_2 = P_f - \dot{Q}_1$ and rearrange Eq. (4) to obtain an upper bound on the amplifier's efficiency, providing that $\dot{Q}_1 > 0$, $\dot{Q}_2 < 0$, and $\dot{S}_{af} > 0$:

$$\eta \equiv \frac{P_f}{\dot{Q}_1} \leqslant \frac{T_1 - T_2}{T_1} + \frac{\dot{S}_{af} T_2}{\dot{Q}_1}. \tag{5}$$

Equation (5) is the major motivation for this work, as the inclusion of the last term, $\frac{\dot{S}_{af}T_2}{\dot{Q}_1}$, gives a more generous bound on a heat engine's efficiency coupled to two thermal reservoirs with positive temperatures, as follows from Carnot's famous formula.

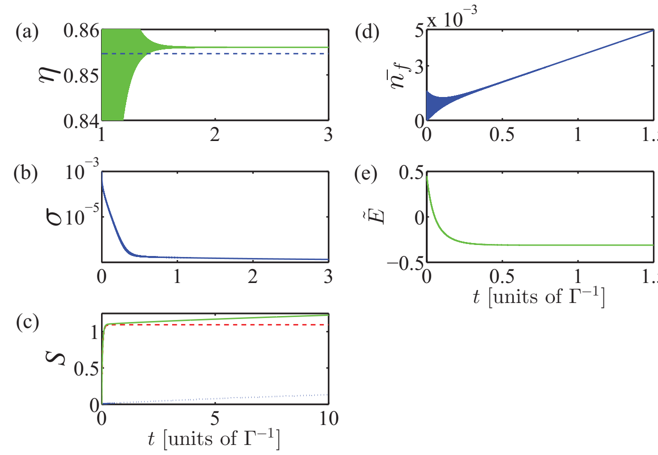

FIG. 2. (Color online) (a) Quasi-steady-state efficiency (solid green) and Carnot efficiency (dashed blue). (b) Total entropy production. (c) Atomic (dashed red), field (dotted blue), and atomic-field (solid green) entropies. (d) Average photon number. (e) Quasi-semiclassical energy, $\tilde{E}$. Parameters: $\omega_f = 50\lambda = 1.05(\omega_1 - \omega_2)$, $\frac{\omega_1-\omega_2}{\lambda} = 10^3$, $\frac{\lambda}{\Gamma_{1(2)}} = 10^3$, $n_1 = 5$, and $n_2 = 4.3$.

Figures 2(a)–2(c) are a second-law graphical summary of the presented results, in which we plot the steady-state efficiency of the light amplifier [Fig. 2(a)], the total entropy production function [Fig. 2(b)], and the various von Neumann entropies [Fig. 2(c)], for the following choice of parameters: $\omega_f = 50\lambda = 1.05(\omega_1 - \omega_2)$, $\frac{\omega_1-\omega_2}{\lambda} = 10^3$, $\frac{\lambda}{\Gamma_{1(2)}} = 10^3$, $n_1 = 5$, and $n_2 = 4.3$. The initial evolution begins with an atom in the excited state and an empty cavity (although any initial state would do to demonstrate the steady-state phenomena). The amplifier efficiency (solid green) in Fig. 2(a) oscillates at short times, but reaches a steady-state value higher than that of Carnot's (dashed blue plateau) as determined by the reservoirs' temperatures. The total entropy production function in Fig. 2(b) is positive at all times. The enhanced steady-state amplifier efficiency combined with the positivity of the entropy production function at all times is the main qualitative result of this paper. Although the field mode is a work reservoir, its entropy is not constant, as can be seen in Fig. 2(c). In most examples in the scientific literature the work reservoir has fixed entropy. However, there is no restriction in classical thermodynamics for a work source or reservoir to show a change in entropy, and the example provided here is not rare when a full quantum treatment is involved. The fact that the amplified cavity mode has an ever changing entropy will be discussed in the context of the semiclassical model later. Finally, we note that the partial (reduced) entropies of the atom and field are not extensive in this case (even at the energy steady state presented here), and hence the joint atomic-field entropy satisfies $S_{af} \neq S_a + S_f$.

Figures 2(d)–2(e) are a first-law graphical summary of the presented results, in which we plot the average photon number in the cavity, $\bar{n}_f$ [Fig. 2(d)], and the quasi-semiclassical energy, $\tilde{E}$ [Fig. 2(e)], for the same choice of parameters mentioned above. Figure 2(e) shows that $\tilde{E}$ reaches a steady-state value, while the field energy rises linearly after $t \sim 0.5\Gamma^{-1}$. We note that although $\tilde{E}$ reaches a constant value, the full atomic-field density matrix $\boldsymbol{\rho_{af}}$ is not stationary. Therefore, we call this situation an energy steady state. Moreover, unlike the resonant case, the individual thermodynamic currents $\dot{Q}_{1(2)a(V)}$ and $P_{a(f)}$ are quasiconstant off-resonantly, and they vary little over a long time scale (a maximum of a few percent in $\Delta t = 100\Gamma^{-1}$ for various choices of detuning values and thermal excitation values). We note that the growth of the energy inside the cavity is essentially unbounded, which was verified numerically up to $t = 100\Gamma^{-1}$.

The off-resonant amplifier discussed here complements in a way the saturated amplifier and attenuator discussed in Ref. [11] which is in the $n_1 < n_2$ regime (higher thermal photon excitation in the reservoir coupling levels $|0\rangle$ and $|2\rangle$) and includes detuning. However, the latter is classified either as a saturated amplifier, or an attenuator, according to the path that an initial field state follows, and not according to its steady-state behavior. Furthermore, the saturated amplifier and attenuator in Ref. [11] reach a steady state which is in fact an equilibrium state, with vanishing energy currents (including power), and hence zero steady-state entropy production.

We now analyze in detail the quasi-semiclassical energy, $\tilde{E}$. The field power P_f can be written as follows:

$$\begin{aligned} P_f &= \lambda[(\omega_1 - \omega_2) + \Delta]\mathrm{Tr}\{\boldsymbol{\rho_{af}}(i\sigma_{21}^\dagger \otimes a + \mathrm{H.c.})\} \\ &= -P_a + P_\Delta, \end{aligned} \tag{6}$$

where $\Delta \equiv \omega_f - (\omega_1 - \omega_2)$ is the detuning from resonance, $P_a = \lambda(\omega_1 - \omega_2)\mathrm{Tr}\{\boldsymbol{\rho_{af}}(i\sigma_{21}^\dagger \otimes a + \mathrm{H.c.})\}$, and $P_\Delta = \lambda\Delta\mathrm{Tr}\{\boldsymbol{\rho_{af}}(i\sigma_{21}^\dagger \otimes a + \mathrm{H.c.})\}$. At the energy steady state both $\dot{\tilde{E}} = 0$ and $\dot{E}_a \equiv \mathrm{Tr}\{\rho_a H_a\} = \dot{Q}_{1a} + \dot{Q}_{2a} + P_a = 0$. Therefore, Eq. (3) rearranges to

$$\dot{\tilde{E}}^{ss} = \dot{Q}_{1V} + \dot{Q}_{2V} - P_\Delta = 0, \tag{7}$$

where ss stands for steady state. If one writes $\dot{Q}_{1(2)V}$ and P_Δ explicitly one finds that

$$\begin{aligned} \dot{Q}_{1(2)V} &= -2\lambda\Gamma_{1(2)}(n_{1(2)} + 1)\sum_m \sqrt{m+1}\,\mathrm{Re}\left(\boldsymbol{\rho_{af}^{1m,2(m+1)}}\right) \\ P_\Delta &= 2\lambda\Delta\sum_m \sqrt{m+1}\,\mathrm{Im}\left(\boldsymbol{\rho_{af}^{1m,2(m+1)}}\right), \end{aligned} \tag{8}$$

hence one finds that at the energy steady state

$$\cot^{\mathrm{ss}}(\theta) = \frac{\sum_m \sqrt{m+1}\,\mathrm{Re}\left(\boldsymbol{\rho_{af}^{1m,2(m+1)}}\right)}{\sum_m \sqrt{m+1}\,\mathrm{Im}\left(\boldsymbol{\rho_{af}^{1m,2(m+1)}}\right)} = -\frac{\Delta}{B}, \tag{9}$$

where $B = \Gamma_1(n_1 + 1) + \Gamma_2(n_2 + 1)$. Equation (9) represents the semiclassical relative phase as we show in Sec. III. The phase, however, *cannot* be recovered from a naive partial trace over the field or the atom, as both the atomic and field reduced density matrices are diagonal at steady state. To see this we note that the full atomic-field bipartite density matrix assumes the following form after $t > \Gamma^{-1}$:

$$\boldsymbol{\rho_{af}} = \left(\begin{array}{c|c|c} \boldsymbol{P_{0,0}} & \mathbf{0} & \mathbf{0} \\ \hline \mathbf{0} & \boldsymbol{P_{1,1}} & \boldsymbol{C} \\ \hline \mathbf{0} & \boldsymbol{C}^\dagger & \boldsymbol{P_{2,2}} \end{array}\right), \tag{10}$$

where $\mathbf{0}$ is an $m \times m$ zero matrix, $\boldsymbol{P_{i,i}}$ are diagonal matrices ($i = 0,1,2$) whose elements are $\boldsymbol{\rho^{im,im}}$, and $\boldsymbol{C}$ is an $m \times m$ correlation matrix whose elements are all zero except the elements above the main diagonal $\boldsymbol{\rho_{af}^{1m,2(m+1)}}$. Clearly, tracing over the field degree of freedom at the energy steady state will yield a 3×3 diagonal atomic (reduced) density matrix with no coherence whatsoever. To emphasize this point further, if one would design a local atomic phase measurement based on ρ_a at steady state and any linear combination of σ_x and σ_y (or $\boldsymbol{\rho_{af}}$ together with a linear combination of $\sigma_x \otimes \mathbb{1}_f$ and $\sigma_y \otimes \mathbb{1}_f$), the obtained result would be zero. Indeed, the three-level light amplifier presented in Ref. [3] was shown to have a quasi-Poissonian photon statistics and no intrafield coherence. Does this mean that amplified light has no coherence? No. The observation that the field has no internal coherence is a manifestation of Mølmer's conjecture [12], that pure optical coherence in an interacting light-matter system is a convenient fiction. The coherence is a joint atomic-field property as Eqs. (7) and (9) suggest (they include the Jaynes-Cummings atomic-field interaction Hamiltonian and the atomic-field density matrix elements which are absent from a partial trace over the atom or field). Therefore, we conclude that the semiclassical relative phase, θ, associated with coherence of light amplification can be derived from fundamental steady-state thermodynamic currents of the full quantum model and is in fact an inter-atomic-field property rather than a local atomic or field property.

III. SEMICLASSICAL TREATMENT

Consider now the three-level system as depicted in Fig. 1, but where the quantized electric field mode is replaced by a classical field. The system is governed by a dissipative Liouvillian similar to the quantum case (written only in the reduced atomic space) and by a semiclassical RWA JCM Hamiltonian: $H = H_a + V(t)$, $V(t) = \lambda(\sigma_{21}\exp^{i\omega_f t} + \sigma_{21}^\dagger \exp^{-i\omega_f t})$.

The energy currents of the three-level system $\dot{E} = \mathrm{Tr}\{\rho_a[H_a \dot{+} V(t)]\}$ are given by

$$\begin{aligned} \dot{E} &= \mathrm{Tr}\left\{\frac{\partial\rho_a}{\partial t}[H_a + V(t)]\right\} + \mathrm{Tr}\left\{\rho_a\frac{\partial V}{\partial t}\right\} = \dot{Q} + P \\ &= \dot{Q}_{1a} + \dot{Q}_{1V} + \dot{Q}_{2a} + \dot{Q}_{2V} + P = \dot{Q}_1 + \dot{Q}_2 + P, \end{aligned} \tag{11}$$

where $\dot{Q} \equiv \mathrm{Tr}\{\frac{\partial\rho_a}{\partial t}[H_a + V(t)]\} = \mathrm{Tr}\{\mathcal{L}_{d(1+2)}[\rho_a][H_a + V(t)]\}$ and $P \equiv \mathrm{Tr}\{\rho_a\frac{\partial V}{\partial t}\}$ are heat currents ($\dot{Q}_{1(2)a}$ and $\dot{Q}_{1(2)V}$ are defined in a way similar to that of the bipartite treatment) and power, respectively, as defined in accordance with the general thermodynamical formalism for open forced unipartite systems due to Alicki [6]. The steady-state efficiency of the light generator is derived from the steady-state semiclassical density matrix given by (in the Schrödinger picture)

$$\rho = \frac{1}{F}\begin{pmatrix} 1 - A_1 - B_1 & 0 & 0 \\ 0 & A_1 & C_1\exp^{-i\omega_f t} \\ 0 & C_1^*\exp^{i\omega_f t} & B_1 \end{pmatrix}, \tag{12}$$

where $A_1 = f(\Gamma_1,\Gamma_1,n_1,n_2,\Delta) > 0$, $B_1 = f(\Gamma_1,\Gamma_1,n_1,n_2,\Delta) > 0$, and $C_1 = -\Gamma_1\Gamma_2\lambda(\Delta - iB)(n_1 - n_2)$. If one calculates the ratio between the real and imaginary parts of the coherence density matrix element (without the modulus 1 exponential which vanishes in the rotating frame) in Eq. (12) one recovers exactly the phase as appears in Eq. (9) in Sec. II. Diagonalization of this density matrix yields time-independent eigenvalues given by

$$\lambda_0 = \frac{1 - A_1 - B_1}{F}, \quad \lambda_{1,2} = \frac{A_1 + B_1 \pm \sqrt{(A_1 - B_1)^2 + 4|C_1|^2}}{2F}. \tag{13}$$

In order to find the steady-state thermodynamic currents, one transforms to a rotating (field frequency) frame, finds the steady-state solution of the density matrix, transforms back to the Schrödinger picture, and finally calculates the thermodynamic currents, which are given by

$$\begin{aligned}
\dot{Q}^{ss}_{1a} &= \frac{AB}{F}(n_1 - n_2)(\omega_1 - \omega_0),\\
\dot{Q}^{ss}_{1V} &= \frac{A}{F}\Gamma_1(n_1 + 1)(n_1 - n_2)\Delta,\\
\dot{Q}^{ss}_{2a} &= -\frac{AB}{F}(n_1 - n_2)(\omega_2 - \omega_0),\\
\dot{Q}^{ss}_{2V} &= \frac{A}{F}\Gamma_2(n_2 + 1)(n_1 - n_2)\Delta,\\
P^{ss} &= -\frac{AB}{F}(n_1 - n_2)\omega_f,
\end{aligned} \tag{14}$$

where $A = 2\Gamma_1\Gamma_2\lambda$ and $F = f(\Gamma_1,\Gamma_1,n_1,n_2,\Delta) > 0$. The steady-state efficiency of the light generator is obtained by dividing the power term, $-P^{ss}$, by the hot reservoir heat current term, $\dot{Q}^{ss}_1 = \dot{Q}^{ss}_{1a} + \dot{Q}^{ss}_{1V}$, which yields

$$\eta \equiv -\frac{P^{ss}}{\dot{Q}^{ss}_1} = \frac{\omega_s}{\omega_p + \alpha'\Delta}, \tag{15}$$

where $\omega_s = \omega_f$ is the signal frequency, $\omega_p = \omega_1 - \omega_0$ is the pump frequency, and $0 < \alpha' = \frac{\Gamma_1(n_1+1)}{\Gamma_1(n_1+1)+\Gamma_2(n_2+1)} < 1$. We found that Eq. (15) perfectly agrees with the numerical efficiency of the quantum treatment. In perfect resonance, $\Delta = 0$, we recover the Scovil–Schulz-DuBois result [2].

Now we investigate the two regimes of detuning. If the light is red-detuned from the atomic resonance, $\Delta < 0$, then

$$\begin{aligned}
\frac{\omega_s}{\omega_p} &= \frac{\omega_{res} + \Delta}{\omega_p} < \eta = \frac{\omega_{res} + \Delta}{\omega_p + \alpha'\Delta} < \frac{\omega_{res}}{\omega_p} < \eta_{Carnot}\\
&= \frac{\omega_p - \beta'(\omega_p - \omega_{res})}{\omega_p},
\end{aligned} \tag{16}$$

where $\omega_{res} = \omega_1 - \omega_2$ is the atomic resonance frequency and $0 < \beta' = \frac{\ln(1+1/n_1)}{\ln(1+1/n_2)} < 1$ (for $n_2 < n_1$).

If the light is blue-detuned from the atomic resonance, $\Delta > 0$, then

$$\frac{\omega_{res}}{\omega_p} < \eta < \frac{\omega_f}{\omega_p}, \tag{17}$$

and there can be a detuning window for which the Carnot limit is broken, as $\frac{\omega_f}{\omega_p}$ may be bigger than η_{Carnot}. By requiring that $\eta > \eta_{Carnot}$ [where η is the right-hand-side expression in Eq. (15) and where η_{Carnot} is the right-hand-side expression in Eq. (16)] one obtains the critical field frequency for breaching the Carnot limit:

$$\omega_f \geqslant \frac{(1 - \beta')\omega_p^2 - \alpha'\beta'\omega_{res}^2 + (\alpha'\beta' + \beta' - \alpha')\omega_p\omega_{res}}{(1 - \alpha')\omega_p + \alpha'\beta'(\omega_p - \omega_{res})}. \tag{18}$$

The immediate consequence of breaking the Carnot limit in the semiclassical treatment is that the entropy production function is negative. This follows from the fact that the three eigenvalues of the semiclassical density matrix at steady state are time independent, and hence the unipartite atomic system entropy production, $\frac{\partial S_a}{\partial t}$, is zero. This is contradicted by the quantum treatment for which $\frac{\partial S_{af}}{\partial t} > 0$ and also compensates for the negative entropy production of the reservoirs. Therefore, it is expected that the joint atomic-field entropy varies with time, and in the long run we even see that the main contribution comes from the field as Fig. 2(c) clearly shows. From this comparison we learn that the drawback of the semiclassical treatment is that it does not account for the change of the field's entropy (amplified noise), and hence the second law is erroneously violated for certain detuning values. Moreover, it seems that if one accounts for the field dynamics directly (second quantization of the field), there is no need to amend the dissipative part of the Liouvillian as was done, for example, in Refs. [13,14].

Since the interaction heat currents $\dot{Q}_{1V}$ and $\dot{Q}_{2V}$ depend on the detuning, they can change the thermodynamical picture as $\dot{Q}_{1(2)} = \dot{Q}_{1(2)a} + \dot{Q}_{1(2)V}$ may become negative (positive) for red (blue)-detuned light. The limiting red-detuning is obtained when $\dot{Q}_1 \leqslant 0$ and is given by

$$\omega_f \leqslant \omega_{res} - \left(1 + \frac{\Gamma_2(n_2 + 1)}{\Gamma_1(n_1 + 1)}\right)\omega_p < 0, \tag{19}$$

which is unacceptable physically. The limiting field frequency for blue-detuned light is obtained when $\dot{Q}_2 \geqslant 0$ and is given by

$$\omega_f \geqslant \omega_p + \frac{\Gamma_1(n_1 + 1)}{\Gamma_2(n_2 + 1)}(\omega_p - \omega_{res}), \tag{20}$$

which means that the field frequency should be bigger than the pump frequency (very hard to implement in a realistic physical system).

We note that the individual analytical semiclassical steady-state energy currents for off-resonant excitation do not agree with their numerical counterparts in the fully quantized treatment (with a deviation that increases with the absolute value of the detuning). However, the semiclassical efficiency that stems from the relevant energy currents does show excellent agreement with its quantum counterpart. The analytical prediction of Eq. (15) in the paper agrees with the numerical steady-state efficiency presented in Fig. 2(a), up to five significant digits, which in any case represents a higher precision of the exemplified breaking of the Carnot limit. This is because all the thermodynamical currents differ from their quantum counterpart by a constant factor.

Finally, we note that in Ref. [15] it is indicated that Spohn's entropy production function can be negative for certain detuning values. In order to circumvent the negativity of the entropy production function, Boukobza and Tannor [15] introduced a new entropy production function (which relies on energy currents of the bare atomic Hamiltonian), $\bar{\sigma}$, that is always positive. A nonideal property of this entropy production function is that the efficiency that stems from it is constant, regardless of detuning.

IV. DAMPED QUANTUM AMPLIFIER

The final stage of this work is to thermodynamically characterize a damped quantum amplifier, which represents a more realistic model for a laser or maser, when compared with the idealized amplifier described in Sec. II. A three-level damped light amplifier is governed by the following master equation:

$$\dot{\boldsymbol{\rho}}_{\boldsymbol{af}} = \mathcal{L}_h[\boldsymbol{\rho}_{\boldsymbol{af}}] + \mathcal{L}_{d1}[\boldsymbol{\rho}_{\boldsymbol{af}}] + \mathcal{L}_{d2}[\boldsymbol{\rho}_{\boldsymbol{af}}] + \mathcal{L}_{df}[\boldsymbol{\rho}_{\boldsymbol{af}}]. \quad (21)$$

where $\mathcal{L}_h[\boldsymbol{\rho}_{\boldsymbol{af}}]$ and $\mathcal{L}_{d1(2)}[\boldsymbol{\rho}_{\boldsymbol{af}}]$ are the Hamiltonian part of the Liouvillian and the two atomic dissipative Lindblad superoperators described in Sec. II. $\mathcal{L}_{df}[\boldsymbol{\rho}_{\boldsymbol{af}}]$ is the (zero temperature) field dissipative damping term, given by

$$\mathcal{L}_{df}[\boldsymbol{\rho}_{\boldsymbol{af}}] = \Gamma_f\{([\boldsymbol{a}\boldsymbol{\rho}_{\boldsymbol{af}}, \boldsymbol{a}^\dagger] + \text{H.c.}), \quad (22)$$

where Γ_f is the Weisskopf-Wigner field decay constant associated with the reservoir of electromagnetic field modes outside the cavity that are coupled to the cavity mirrors.

Since the field mode is now coupled directly to reservoir modes, the energy currents directly associated with it will include both a power term and a heat current term, unlike the ideal amplifier described here and in Ref. [4]. To see this we expand the average value of the field energy in differential form:

$$\dot{E}_f \equiv \text{Tr}\{\dot{\rho}_f H_f\} = \text{Tr}\{\mathcal{L}_{df}[\boldsymbol{\rho}_{\boldsymbol{af}}]\boldsymbol{H}_{\boldsymbol{f}}\} - \frac{i}{\hbar}\text{Tr}\{\boldsymbol{\rho}_{\boldsymbol{mf}}[\boldsymbol{H}_{\boldsymbol{f}}, \boldsymbol{V}_{\boldsymbol{mf}}]\}$$
$$= \dot{Q}_f + P_f, \quad (23)$$

where $\dot{Q}_f \equiv \text{Tr}\{\mathcal{L}_{df}[\boldsymbol{\rho}_{\boldsymbol{af}}]\boldsymbol{H}_{\boldsymbol{f}}\}$. Equation (23) represents the first law of thermodynamics from the field mode perspective. At steady state (and since the field is also damped, there will be a global steady state, $\dot{\boldsymbol{\rho}}_{\boldsymbol{af}} = 0$), from the field perspective all the positive field power is turned into heat ($\dot{Q}_f < 0$). However, one would still be able to calculate the efficiency of the amplifier, which is the efficiency of maintaining a positive constant power inside the cavity. In Fig. 3(a) we plot the amplifier efficiency for the following choice of parameters: $\omega_f = 50\lambda = 1.05(\omega_1 - \omega_2)$, $\frac{\omega_1-\omega_2}{\lambda} = 10^3$, $\frac{\lambda}{\Gamma_{1(2)}} = 10^3$, $n_1 = 5$, $n_2 = 4.3$, and $\frac{\Gamma_{1(2)}}{\Gamma_f} = 10^3$, and for extended times. The calculation was carried out by direct integration of Eq. (21) in Liouville space, $\hat{\hat{\rho}}_{af}(t) = e^{\mathcal{L}}\hat{\hat{\rho}}_{af}(0)$, using a diagonal time-independent total Liouvillian superoperator ["translated" from the total Liouvillian that appears in Eq. (21)]. It is clearly visible that the damped amplifier also breaches the Carnot limit for identical detuning and atomic reservoirs conditions as the nondamped oscillator. The redefined energy, $\tilde{\boldsymbol{H}}$, is now decomposed into

$$\dot{\tilde{E}} = \dot{Q}_{1a} + \dot{Q}_{1V} + \dot{Q}_{2a} + \dot{Q}_{2V} + \dot{Q}_{fV} - P_f, \quad (24)$$

where $\dot{Q}_{fV} \equiv \text{Tr}\{\mathcal{L}_{df}[\boldsymbol{\rho}_{\boldsymbol{af}}]\boldsymbol{H}_{\boldsymbol{f}}\}$. We note that at extended times and when $\Gamma_f << \Gamma_{1(2)}$, amplification is not artificial. This is seen by the average number of photons inside the cavity for $\boldsymbol{\rho}_{\boldsymbol{af}} = (|0\rangle\langle 0|)_a \otimes (|0\rangle\langle 0|)_f$, $\omega_f = 50\lambda = 1.05\omega_{\text{res}}$, and pure Hamiltonian dynamics [Fig. 3(c)], as compared to the case where both the atomic and field dissipation is dropped [Fig. 3(d), JCM Hamiltonian dynamics]. The low excitation in the cavity is attributed to the detuning value, which is

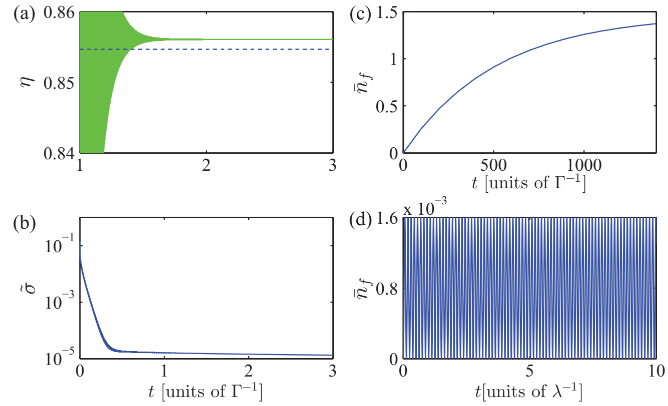

FIG. 3. (Color online) (a) Steady-state efficiency (solid green) and Carnot efficiency (dashed blue). (b) Entropy production without the (positive) contribution of the reservoir coupled to the cavity mirrors. (c) Average photon number of the damped amplifier. (d) Average photon number of the detuned JCM (without damping and amplification). Parameters: $\omega_f = 50\lambda = 1.05(\omega_1 - \omega_2)$, $\frac{\omega_1-\omega_2}{\lambda} = 10^3$, $\frac{\lambda}{\Gamma_{1(2)}} = 10^3$, $n_1 = 5$, $n_2 = 4.3$, and $\frac{\Gamma_{1(2)}}{\Gamma_f} = 10^3$.

3 orders of magnitude lower than the amplification scenario in Fig. 3(b).

The total entropy production function for the joint atomic-field system and the reservoirs when the cavity mode is damped is given by

$$\sigma = \dot{S}_{af} - \frac{\dot{Q}_1}{T_1} - \frac{\dot{Q}_2}{T_2} - \frac{\dot{Q}_3}{T_3}, \quad (25)$$

where $\dot{Q}_3 = \dot{Q}_f + \dot{Q}_{fV}$ and $T_3 = 0$ as no external pumping of the cavity is included. We note that at all times, $\dot{Q}_3 < 0$, and hence the contribution of reservoir 3 (which is coupled to the cavity mirrors) to the entropy production function is always positive ($\sigma_3 = -\frac{\dot{Q}_3}{T_3} > 0$). Numerically, however, division by zero will yield $+\infty$ contribution, and an analytical expansion for the limit $T_3 \to 0$ should be sought. Nevertheless, one may calculate numerically $\tilde{\sigma} = \dot{S}_{af} + \sigma_1 + \sigma_2$, which is plotted in Fig. 3(b). Since $\tilde{\sigma}$ is positive at all times, and since $\sigma_3 > 0$ *always*, the entropy production function is positive at all times, also for the damped amplifier. In a forthcoming publication, we will give a full analysis of the various operation regimes of the atomic-field system when $T_3 > 0$, where the heat engine picture might be reversed.

V. CONCLUSION

In this work we have given a complete thermodynamical analysis of an off-resonant light amplifier. We have shown that the off-resonant light amplifier represents a model of a heat engine (for the $n_1 > n_2$ regime) with an efficiency that surpasses the Scovil–Schulz-DuBois limit [2] for a red-detuned cavity and under certain conditions can also surpass the efficiency given by the famous Carnot formula for a blue-detuned cavity, while still satisfying the second law with respect to positive entropy production. Other significant aspects of this work can be summarized in the following eight points.

First, we have presented an analytical formula for the amplifier's efficiency based on thermodynamic energy currents at steady state. The efficiency alters with detuning and incorporates the true signal field frequency, $\omega_f = \omega_{\text{res}} + \Delta$. This feature is absent in other work, for example, Refs. [2,4,15]. This formula was obtained in the semiclassical treatment and agrees perfectly with the efficiency that stems from an exact numerical solution of the fully quantized treatment.

Second, we provided a critical field frequency formula, which may give a reasonable physical bound on breaching the Carnot limit, for which (a) the atomic-field coupling is still substantial; (b) the RWA still holds; and (c) thermodynamically, the positive heat current comes from the hot reservoirs (positive higher temperature), while the negative heat current comes from the cold reservoir (positive lower temperature).

Third, we have shown the importance of a full quantum treatment of the problem in hand. While the semiclassical treatment predicts correctly the efficiency formula of the light amplifier, it does not capture at all the field dynamics. This turns out to be a crucial aspect, as the entropy production function becomes negative for certain detuning values in the semiclassical treatment. However, in the fully quantum treatment, the entropy production function is positive in total due to a substantial entropy change of the field degree of freedom, which represents a work reservoir with a nonfixed entropy. Moreover, this work suggests that altering the dissipative terms of the Liouvillian to have a positive entropy production function is unnecessary, as full quantization of the field may resolve this issue naturally.

Fourth, the heat engine described here can be compared with the four-stroke heat engine due to Scully *et al.* [5]. The heat engine described in the present work (a) does not require an *a priori* preparation of either the cavity field or atom; (b) runs continuously; (c) does not break the Carnot paradigm of extracting work from two reservoirs with different temperatures; (d) does not reach a steady state in terms of the atomic-field, or field state; and (e) shows that surpassing the Carnot limit stems from a detailed thermodynamical balance (of heat currents and field power) which explicitly accounts for the atom, cavity mode, and heat reservoirs, using a full quantum description. Moreover, the heat engine described in Ref. [5] disregards atomic-microwave cavity correlation and entanglement and does not explicitly thermodynamically account for the microwave cavity.

Fifth, the heat engine described here can also be compared with the laser and photocell continuous quantum heat engines recently described by Scully *et al.* [16]. In Ref. [16], enhanced power in a laser or photocell is achieved by noise-induced coherence of a doubly split ground state. Whereas the enhanced power (relative to the hot reservoir pump) of the quantum optical heat engine described here is achieved via off-resonant excitation.

Sixth, we showed that the semiclassical relative phase associated with light amplification can be derived from fundamental thermodynamic currents of the fully quantized model, in a way that demonstrates that coherence is an inter-atomic-field property rather than a local atomic or field property.

Seventh, we have also demonstrated that a damped off-resonant light amplifier, which represents a more realistic model for a laser or maser, operates as a heat engine that can (under certain conditions) surpass the Carnot limit, with total positive entropy production at all times.

Finally, it is appropriate to ask how is the quantum optical amplifier discussed in this paper different from classical or Carnot heat engines discussed in standard thermomodynamics textbooks (besides the obvious fact that classical heat engines are operated in strokes and not continuously). Moreover, can the quantum optical amplifier discussed in this paper be analyzed as a heat engine at all? We believe that the answer to the latter question is yes. However, two differences between the quantum amplifier discussed here and a classical Carnot heat engine should be pointed out. In the undamped case, although the atomic working fluid might reach a steady state, the field work reservoir does not, and hence the distinction with a standard Carnot engine arises from the fact that the blue-detuned amplifier is not periodic, and does not reach a steady state. In the damped amplifier case, both the atomic working fluid and field work reservoir reach a steady state, but the distinction with a standard Carnot engine arises from the fact that now the atomic-field system is coupled to three heat reservoirs (as opposed to two in a standard Carnot heat engine).

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