

# Nonequilibrium condensates and lasers without inversion: Exciton-polariton lasers

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We analyze elementary properties of exciton and polariton lasers — devices that generate coherent optical and matter waves using final-state stimulation of exciton-phonon scattering. First we discuss the relation between the conditions for the onset of equilibrium and nonequilibrium excitonic condensates. Provided that the thermal de Broglie wavelength  $\lambda_T$  exceeds the exciton Bohr radius  $a_B$ , an exciton laser operates without electronic population inversion. In contrast to previous proposals, this is a different type of *laser without inversion* which utilizes many-body coherences. When the excitonic character of the polariton branch vanishes, a polariton laser becomes indistinguishable from a photon laser. [S1050-2947(96)08506-X]

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## I. INTRODUCTION

The impressive progress in laser cooling and trapping of atoms has recently resulted in the experimental realization of a Bose-Einstein condensate (BEC) of cold rubidium, lithium, and sodium atoms [1]. Similar results suggesting the existence of condensation effects were previously reported in excitonic systems [2]. The experiments in both domains are so convincing that the topic is now considered ripe enough for applications: There are already several proposals for atom [3] and exciton (polariton) [4] lasers. In fact, recent experiments have provided evidence for the existence of polariton laser action in semiconductor microcavity structures [5], where a coherent state of exciton polaritons is generated by final state stimulation of exciton-phonon interaction. In contrast to Bose-Einstein condensates [6,7], atomic or excitonic systems are driven far from equilibrium in these *matter lasers*.

Two well-known phenomena arising directly from Bose statistics are (1) BEC of massive bosonic particles (such as  $^{87}\text{Rb}$ ) in thermal equilibrium and (2) photon lasers; i.e., coherent light generation from incoherent nonequilibrium (inverted) reservoirs. Even though both of these topics have been extensively studied, little is known about their connection. In this paper, we analyze fundamental properties of polariton lasers, which in our opinion sheds light on the relation between these two phenomena. Figure 1 shows a diagram summarizing our viewpoint: For a thermal equilibrium reservoir and a vanishing *photon character* of the exciton polaritons, one obtains a BEC of excitons. In the opposite limit of a nonequilibrium (inverted) reservoir and a vanishing exciton character, the polariton laser is indistinguishable from a photon laser.

In what follows, we discuss several basic results that pertain to polariton lasers. It is well known that the relevant elementary excitations in a strongly coupled exciton-photon system are polaritons [8]. If a single cavity mode and a single exciton band are coupled, one obtains two split polariton branches [9]

$$\hat{p}_{i,k} = u_{i,k} \hat{C}_k + v_{i,k} \hat{a}_k, \quad (1)$$

where  $\hat{p}_k$ ,  $\hat{C}_k$ , and  $\hat{a}_k$  denote the polariton, exciton, and photon annihilation operators (with wave vectors  $k$ ), respectively. The dispersion curves of the two polariton branches ( $i=1,2$ ) are determined by the exciton and cavity dispersions; a single branch may be excitonlike ( $|u| > |v|$ ) and photonlike ( $|v| > |u|$ ) for different values of the wave vector  $k$ . For condensation type effects, we are interested in the  $k \approx 0$  region of the dispersion curve; we will assume that condensation into (only) a single branch is important and discard the other branch in our discussions. The analysis presented here pertains to the low-exciton density limit where the excitons (and polaritons) satisfy bosonic commutation relations [7]. The interactions between polaritons are neglected, since the *saturation* and *phase diffusion* [4] caused

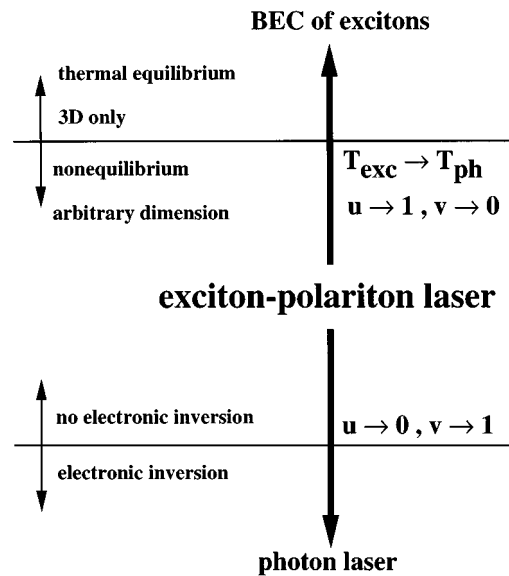


FIG. 1. Diagram comparing the exciton polariton laser to the more familiar concepts of Bose-Einstein condensate of excitons and photon lasers. The annihilation operator for the exciton polariton quasiparticles is given by  $\hat{p} = u\hat{C} + v\hat{a}$ , where  $u$  and  $v$  determine the exciton and photon character of the polariton, respectively.

by such interactions are not relevant to the present discussion. Even though we concentrate exclusively on the exciton polariton system, our results should in principle apply to other nonequilibrium bosonic systems as well.

## II. EXCITON LASERS AND BOSE-EINSTEIN CONDENSATES

First, we address the connection between BEC and non-equilibrium condensates. We consider an exciton laser (i.e., the photon character  $\nu=0$ ), where we assume that all but the ground-state excitons form an excitonic reservoir. This (electrically neutral) reservoir is pumped by an external incoherent source (either electrical injection or light) and therefore is out of equilibrium with the phonon reservoir that it interacts. We assume that the excitation is weak enough that the excitons (both reservoir and ground-state) may be treated as bosonic particles; in this limit final-state stimulation of exciton-phonon interaction provides gain for the ground-state excitonic mode. A straightforward analysis has shown that to obtain a nonequilibrium condensate with a fluctuating phase (in contrast to BEC), one needs to satisfy [10,4,5]

$$\bar{n}_{exc}(k) > \bar{n}_{ph}(k), \quad (2)$$

that is, for all modes that are contributing to ground-state exciton-phonon scattering, the exciton reservoir occupancy has to exceed that of the phonons. The actual observation of population buildup in the ground excitonic state also requires that the (unsaturated) *net gain* exceeds the loss, i.e., the *laser threshold* is

$$\sum_k \Gamma_{ph}(k) [\bar{n}_{exc}(k) - \bar{n}_{ph}(k)] \geq \Gamma_{loss}, \quad (3)$$

where the sum is over all excitonic modes.  $\Gamma_{ph}(k)$  and  $\Gamma_{loss}$  denote the acoustic-phonon absorption and decay (spontaneous emission) rates of the ground-state excitons. If Eq. (3) is satisfied, a coherent state of excitons in the ground-state with a slowly diffusing phase will form; the phase fluctuations have contributions from spontaneous exciton-phonon scattering and exciton-exciton interactions, both of which remain significant at  $T_{ph}=0$  K. In contrast to BEC, the dimensionality of the excitonic system is not important in an exciton laser [11]. Throughout this work, we assume that the phonon reservoir remains in a thermal state with a well-defined temperature  $T_{ph}$ .

The laser inversion condition  $[\bar{n}_{exc}(k) > \bar{n}_{ph}(k)]$  corresponds to a minimum reservoir exciton density  $N_{exc}^{min}$  that is required to have net final state stimulated emission of ground-state excitons

$$\begin{aligned} N_{exc}^{min} &= \int_0^\infty d\omega \rho(\omega) \bar{n}_{exc}(\omega) > \int_0^\infty d\omega \rho(\omega) \bar{n}_{ph}(\omega) \\ &= \int_0^\infty d\omega \rho(\omega) \frac{1}{\exp\left[\frac{\hbar\omega}{kT_{ph}}\right] - 1} \approx 2.62\lambda_{T_{ph}}^{-3}, \end{aligned} \quad (4)$$

where  $\rho(\omega) = 3/4\pi^2(2m^*/\hbar)^{3/2}\sqrt{\omega}$  is the 3D exciton density of states and  $\lambda_{T_{ph}} = \sqrt{2\pi\hbar^2/(m^*kT_{ph})}$  denotes the thermal de Broglie wavelength of an exciton gas at temperature  $T_{ph}$  ( $=T_{exc}$  in thermal equilibrium). This is the first result of our

letter: The observation of final-state stimulation requires an exciton density which exceeds the minimum density required for BEC. However, we have made no assumptions about the functional dependence of  $\bar{n}_{exc}(\omega)$  on  $\omega$ . Extension of Eq. (4) to a two-dimensional exciton gas poses no difficulties: Since phonons with  $\omega < \omega_{min} = 2m^*c_s^2/\hbar$  do not contribute to phonon absorption or emission, the divergence of the phonon number as  $\omega \rightarrow 0$  is irrelevant in the sense that  $\bar{n}_{exc}(\omega) > \bar{n}_{ph}(\omega)$  only needs to be satisfied for  $\omega \geq \omega_{min}$ . Therefore, even a thermal distribution of reservoir excitons ( $\bar{n}_{exc} = \{\exp[(\hbar\omega - \mu)/(kT_{exc})] - 1\}^{-1}$ ) with chemical potential  $\mu < 0$  and temperature  $T_{exc} \geq T_{ph}(1 - \mu/\hbar\omega_{min})$  will be sufficient to obtain net stimulated emission of excitons.

The excess reservoir exciton density that is required for the formation of a nonequilibrium condensate or an exciton laser depends on the loss rate  $\Gamma_{loss}$  and the frequency dependence of the phonon emission or absorption rate  $\Gamma_{ph}(\omega)$ . If we assume a hypothetical system where  $\Gamma_{ph}(\omega) \approx \Gamma_{ph}$ , then we obtain

$$N_{exc}^{thres} = \frac{2.62}{\lambda_{T_{ph}}^3} + \frac{\Gamma_{loss}}{V\Gamma_{ph}}, \quad (5)$$

as the density required to obtain a nonequilibrium exciton condensate. Here,  $V \geq \lambda_{T_{ph}}^3$  denotes the volume of the semiconductor (quantization volume) that is assumed to be free of defects and impurities. Physically,  $N_{exc}^{thres}$  corresponds to the reservoir density at which net gain equals net loss, per unit volume. In the equilibrium limit ( $\Gamma_{loss} \rightarrow 0$ ), we naturally obtain the requirement for BEC.

Before closing this discussion, we note that the density requirement of Eq. (5) (obtained for a frequency independent phonon rate) is sufficient but not necessary in many particular realizations. If we consider, for example, the case where the principal gain mechanism is the final state stimulation of LO phonon emission, then the only requirement is a large exciton reservoir occupancy at  $\omega_{exc} = \omega_{LO}$ , since the LO-phonon scattering rate is much larger than the acoustic phonon rates, i.e.,  $\Gamma_{LO} \gg \Gamma_{ph}$ . In fact, the exciton occupancy at other frequencies may be well below that of phonons so that  $N_{exc}^{min} \ll 2.62\lambda_{T_{ph}}^{-3}$ . Therefore, provided that we can keep the exciton reservoir far from equilibrium, the condensation effects may be observed at densities lower than is required for BEC.

## III. EXCITON-POLARITON LASER AS A COHERENT LIGHT SOURCE

It has been shown theoretically [4] that when the above mentioned conditions are satisfied, a coherent state of excitons with a diffusing phase is formed. The output from such a nonequilibrium condensate is either a coherent matter wave (obtained when the condensed excitons are allowed to tunnel out into another semiconductor medium) or a coherent light wave (obtained when the coherent excitons hit a physical boundary, or when they couple to a *radiation field reservoir*). Since conventional coherent light sources depend on the existence of an electronic population inversion, it is important to determine if a similar condition is necessary for an exciton (polariton) matter laser.

Semiconductor Bloch equations [12] specify the relevant

inversion operator for generation of coherent light from an incoherent (uncorrelated) electron-hole reservoir as

$$\hat{I}_{\mathbf{p}} = 1 - \hat{n}_{e,\mathbf{p}} - \hat{n}_{h,-\mathbf{p}}, \quad (6)$$

where  $\hat{n}_{e,\mathbf{p}}$  ( $\hat{n}_{h,-\mathbf{p}}$ ) denotes the electron (hole) number operator. If  $\langle \hat{I}_{\mathbf{p}} \rangle > 0$ ,  $\forall \mathbf{p}$ , then all applied (weak) electromagnetic fields will experience net loss [12].

Light generation at the ground-state exciton frequency is achieved by a superposition excitation of free electron-hole pairs, i.e., radiation field reservoir modes  $\hat{a}_k$  couple to exciton modes  $\hat{C}_k^\dagger$  with

$$\hat{C}_{\mathbf{k}}^\dagger = \frac{1}{\sqrt{V}} \sum_{\mathbf{p}} \varphi(\mathbf{p}) \hat{e}_{\mathbf{k}/2+\mathbf{p}}^\dagger \hat{h}_{\mathbf{k}/2-\mathbf{p}}^\dagger, \quad (7)$$

where  $\hat{e}_{\mathbf{p}}^\dagger$  and  $\hat{h}_{-\mathbf{p}}^\dagger$  denote the electron and hole creation operators, and

$$\varphi(p) = 8 \sqrt{\pi a_B^3} \frac{1}{[1 + (pa_B)^2]^2} \quad (8)$$

is the Fourier transform of the hydrogenic  $1s$  wave function. Since we are interested in coherent light generation from excitons, we evaluate the expectation value of the average inversion operator  $\hat{I} = (1/V) \sum_{\mathbf{p}} |\varphi(\mathbf{p})|^2 \hat{I}_{\mathbf{p}}$ . We find

$$\langle \hat{I} \rangle \geq 1 - 2a_B^3 \left( N_{exc} + \frac{n_0}{V} \right), \quad (9)$$

where  $n_0$  is the average ground-state exciton occupancy. Provided that  $(N_{exc} + n_0/V)a_B^3 \ll 1$ , there is no electronic inversion in the system (i.e.,  $\langle \hat{I} \rangle > 0$ ). This result is not unexpected since  $\hat{I} = [\hat{C}, \hat{C}^\dagger]$  and the bosonic character of the excitons implies absence of electronic inversion.

As we have already seen, coherent light generation from an exciton laser in most cases requires  $N_{exc} \lambda_{T_{ph}}^3 > 2.62$ . By choosing a low enough temperature, we can guarantee that  $\lambda_{T_{ph}} \gg a_B$ . In this limit,  $N_{exc} a_B^3 \ll 1$  will be satisfied. To evaluate the contribution to electronic inversion from the ground-state excitons, we recall that despite the finite phase fluctuations the condensate wave function may be approximated by [7]

$$|\psi\rangle = \frac{1}{\sqrt{\mathcal{N}}} e^{\alpha \hat{C}^\dagger} |0\rangle = \frac{1}{\sqrt{\mathcal{N}}} \prod_{\mathbf{p}} \left[ 1 + \frac{\alpha \varphi(\mathbf{p})}{\sqrt{V}} \hat{e}_{\mathbf{p}}^\dagger \hat{h}_{-\mathbf{p}}^\dagger \right] |0\rangle, \quad (10)$$

where  $|\alpha| = \sqrt{n_0}$  is determined by the net gain and saturation mechanisms and  $\mathcal{N}$  is the normalization constant. Since  $\varphi(p) \leq \varphi(0) = 8 \sqrt{\pi a_B^3}$ , for small enough coherent state amplitudes (i.e.,  $\alpha < \sqrt{V/64\pi a_B^3}$ ) we obtain  $|\alpha|^2 |\varphi(p)|^2 / V < 1$ . This condition implies that the occupancy of the state  $\hat{e}_{\mathbf{p}}^\dagger \hat{h}_{-\mathbf{p}}^\dagger |0\rangle$  satisfies  $\alpha \varphi(p) / \sqrt{V + |\alpha|^2 |\varphi(p)|^2} < 0.5 \forall \mathbf{p}$ , i.e., all optically active electron-hole states are noninverted. In this limit, we have  $2a_B^3 n_0 / V \ll 1$ .

Since a coherent exciton state [Eq. (10)] generates coherent light by spontaneous radiative recombination, an exciton laser may be viewed as a *photon laser without inversion* when inversion is defined as  $\langle \hat{I} \rangle < 0$ . There is a significant difference between the exciton laser described here and the

earlier proposals for II-VI exciton lasers, where a BCS-type state of excitons is utilized for coherent light generation [13]; a BCS state of excitons where  $|\alpha|^2 |\varphi(p)|^2 / V \gg 1$  is electronically inverted according to the definition we have introduced.

So far we have considered an exciton laser, which is the limiting case of a polariton laser when the photon character of the quasiparticle  $v \rightarrow 0$ . In the opposite limit of  $u \rightarrow 0$ , the polariton laser should be indistinguishable from a photon laser. Next, we consider the transition between these two limits and its implication for the electronic population inversion requirement.

For a photonlike exciton polariton, we expect two gain mechanisms to be significant: Final-state stimulation of exciton-phonon scattering and stimulated photon emission. Due to the presence of band-gap renormalization and Coulomb enhancement of interband transitions [12] together with many-body coherences, it is extremely difficult to find a general analytic expression for net gain. To address the question of electronic population inversion, however, it is sufficient to consider final-state stimulation of phonon scattering, since stimulated photon emission requires inversion and will not be effective unless there is inversion. Therefore, for a non-inverted electron-hole system the lasing condition is given by

$$\begin{aligned} \Gamma_{gain}(u) &= \sum_k |u|^2 \Gamma_{ph}(k) [\bar{n}_{exc}(k) - \bar{n}_{ph}(k)] \\ &\geq \Gamma_{loss} = |u|^2 \Gamma_{rad} + |v|^2 \Gamma_{cav}, \end{aligned} \quad (11)$$

where we assumed that reservoir exciton-polaritons are excitonlike ( $|u_k|^2 \gg |v_k|^2$ ). The factor  $|u|^2$  multiplying the phonon emission or absorption rate appears due to the fact that it is the excitonic character of the ground-state polaritons that determines the phonon emission or absorption. The polariton decay rate  $\Gamma_{loss}$  is determined by a combination of excitonic spontaneous emission to all other radiation field modes ( $\Gamma_{rad}$ ) and the cavity decay rate of the photon mode ( $\Gamma_{cav}$ ). For simplicity, we assume that these two rates are equal, eliminating the  $u, v$  dependence of the loss.

As we discussed earlier, the threshold of the polariton laser is obtained when  $\Gamma_{gain}(u) = \Gamma_{loss}$ . By assuming once again that  $\Gamma_{ph}(k) \sim \Gamma_{ph}$ , we obtain a simple expression for the *threshold* reservoir exciton density [14]

$$N_{exc}^{thres,u} \simeq \frac{2.62}{\lambda_{T_{ph}}^3} + \frac{1}{|u|^2 V} \frac{\Gamma_{loss}}{\Gamma_{ph}}. \quad (12)$$

For  $|u|^2 < (a_B^3/V) \Gamma_{loss}/\Gamma_{ph}$ , laser action will take place only when  $N_{exc} a_B^3 \sim 1$ : As the exciton character of the polariton becomes negligible, an electronic inversion is required for the formation of a coherent state of polaritons by stimulated scattering. We remark that the weakly interacting boson gas model we have been using for excitons breaks down at this density limit and it is not even correct to talk about stimulated exciton-phonon scattering. Our result only implies that as the excitonic character of the polaritons diminishes, it becomes impossible to obtain gain without electronic inversion.

At the Mott density limit  $[(N_{exc} + n_0/V)a_B^3 \sim 1]$ , the exciton binding energy diminishes [15], due to the combined

(and comparable) effects of screening [12] and anharmonic ground-state exciton-exciton interactions [7,4]. For  $N_{exc}a_B^3 \gg 1$ , an (inverted) electron-hole plasma forms. In this limit, the cavity (i.e., polariton with  $|u|^2 \ll a_B^3/V$ ) mode energy is larger than the band-gap energy, but smaller than the sum of the electron and hole chemical potentials [15]. Stimulated photon emission into the cavity mode is allowed under these conditions and we obtain a semiconductor (photon) laser that operates with electronic population inversion [12].

#### IV. CONCLUSION

Can we regard exciton-polariton matter lasers (with  $1 \gg |u|^2 \gg a_B^3/V$ ) as photon lasers without electronic inversion? For a below-threshold laser where  $0 < \Gamma_{gain} \leq \Gamma_{loss}$ , an applied weak probe field will experience net gain despite the fact that there is no electronic population inversion. In this limit, the exciton-polariton laser can be regarded as a regenerative (photon) laser amplifier without inversion, which utilizes a many-body coherence within the electron-hole system. This is in contrast to previous inversionless laser schemes which depend on single-particle coherences [16]. We reiterate, however, that *amplification* in the exciton-polariton laser is possible only when the exciton-phonon system is inverted, i.e., Eq. (2) is satisfied. Even though the

strongly coupled (bulk) exciton-microcavity system has been recently demonstrated [17], experimentally the more relevant scheme is that of a two-dimensional microcavity exciton-polariton system [5]; the extension of the present discussion to this scheme is straightforward.

In summary, we have presented three basic results pertaining to exciton and exciton-polariton lasers. We found that the sufficient condition for the formation of a nonequilibrium condensate (or net gain in an exciton matter laser) is a direct extension of the condition for the onset of equilibrium BEC. In the second part, we demonstrated that an exciton laser does not require an electronic population inversion (i.e.,  $N_{exc}a_B^3 \ll 1$ ) provided  $a_B \ll \lambda_{T_{ph}}$ . Since it is only in this limit that excitons may be treated as bosons, an exciton laser is a well-defined concept as long as there is no inversion. Finally, we showed that if the coherence in the matter system vanishes, as is the case when polaritons become dominantly photonlike, an electronic population inversion becomes necessary for coherent light generation.

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