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Stochastic Gross-Pitaevskii Equation for the Dynamical Thermalization of Bose-Einstein Condensates

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We present a theory for the description of energy relaxation in a nonequilibrium condensate of bosonic particles. The approach is based on coupling to a thermal bath of other particles (e.g., phonons in a crystal, or noncondensed atoms in a cold atom system), which are treated with a Monte Carlo type approach. Together with a full account of particle-particle interactions, dynamic driving, and particle loss, this offers a complete description of recent experiments in which Bose-Einstein condensates are seen to relax their energy as they propagate in real space and time. As an example, we apply the theory to the solid-state system of microcavity exciton polaritons, in which nonequilibrium effects are particularly prominent.

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Introduction.—Bose-Einstein condensates form when multiple bosonic particles relax their energy, through interaction with other particles (e.g., phonons), to collect into a low energy state. The case of cold atoms [1] is well known and examples in solid-state systems include condensates of magnons [2,3] in quantum spin gases, indirect excitons [4] in coupled quantum wells, and exciton polaritons [5–7] in semiconductor microcavities. While Bose-Einstein condensation is conventionally thought of as a macroscopic occupation of the ground state in thermal equilibrium, several theoretical works have proposed the generation of non-ground-state condensates via resonant excitation of a ground state of cold atoms [8]. This is expected to allow the study of dynamical energy relaxation processes back to the ground state.

The theoretical description of energy relaxation in non-equilibrium systems, with multiple states, is also an important issue given that thermal equilibrium is never perfectly achieved since all examples of condensing bosons have finite lifetimes. The departure from thermal equilibrium is most strongly pronounced in solid-state systems such as exciton polaritons in semiconductor microcavities, where a short lifetime of particles may make the system have a highly nonequilibrium character [9]. In this Letter we develop a kinetic description of such condensates, in which spontaneous spatial coherence has been observed [5], although not necessarily in the ground state [10,11]. While we focus on excitonic systems for their strong nonequilibrium character, our theory could also be adapted to other bosonic systems such as cold atom condensates.

In exciton or exciton-polariton condensates, scattering with acoustic phonons [12] offers a mechanism of energy relaxation. It may not offer complete thermalization, but the effect of energy relaxation is clearly seen in experiments where a potential gradient is present. The latter can be created by the application of stress [6], optically

induced potentials [13], or structural engineering [14]. In this situation, the processes of energy relaxation and condensate propagation down the gradient are closely connected. Aside from offering a clear demonstration of energy relaxation, propagating condensates have recently demonstrated a potential as optoelectronic transistors in exciton [4] and exciton-polariton systems [15].

Theoretically, the dynamics of spatially homogeneous systems have been described accounting for the exciton-phonon interaction by means of a system of semiclassical Boltzmann equations [16–20]. This approach, however, has serious drawbacks. First, the corresponding formalism is based on the assumption of full incoherence in the system; thus, quantum states are supposed to be completely uncorrelated. However, as long as the condensate has been formed, this is no longer valid. As a consequence, a number of coherent phenomena such as the onset of superfluidity [21], bistability [22], and hysteresis cannot be described. Second, the Boltzmann equations can provide us with information about the occupation numbers in reciprocal space (k space) only, whereas the real space (x space) behavior remains obscure.

On the other hand, if the processes of decoherence are fully neglected, the state of interacting particles can be treated as a classical field described by the Gross-Pitaevskii equation [21], which can be modified for incoherent pumping [9,23]. Such an approach has been successful for the description of a variety of recent experiments [24,25] in semiconductor microcavities, including, for example, experiments on the dynamics of vortices [24], spatial pattern formation [25,26], and spin textures [27,28]. However, the Gross-Pitaevskii equation conserves the particle energy and thus does not account for phonon-assisted scattering. Recent models include energy relaxation in a phenomenological way within a classical stochastic field [29] or Gross-Pitaevskii type formalism [30]. While these models

give results in agreement with experimental data, they operate with unknown phenomenological parameters.

Here we introduce a microscopic theory for the description of energy relaxation in a coherent excitonic ensemble. The exciton or exciton-polariton field is coupled to a field representing phonons in the system, which is modeled using stochastic variables. For simplicity, we consider a resonant coherent injection of excitons or exciton polaritons (models of nonresonant excitation, involving coupling to an exciton reservoir, have been considered elsewhere [9] and are compatible with our approach). We consider injection with zero in-plane momentum in a system containing a potential gradient [15]. The potential gradients accelerate the particles which undergo scattering with acoustic phonons as they propagate. The latter process leads to the energy dissipation and thermalization in the system. To give a complete description of the dynamics we fully account for the exciton-exciton scattering and losses provided by a finite lifetime.

We will use parameters corresponding to cavity exciton polaritons although we could equally well describe indirect excitons in coupled quantum wells [4], in which the control of relaxation down electrically induced gradients was recently exploited in excitonic optoelectronic transistors [31,32]. The general idea of this Letter is also applicable to the modeling of other systems in which bosons relax their energy through interactions with an incoherent gas of other particles. As another example, in the cold atom system one typically distinguishes between condensed atoms as well as an incoherent fraction of noncondensed atoms [33]. Interactions between condensed atoms and noncondensed atoms allow the condensed atoms to dynamically relax in energy.

Theory.—For simplicity, we consider a one-dimensional system, as in the case of microwires where there has been a recent experimental focus on energy relaxation of propagating condensates [13,15]. We note however that the formalism is expected to be compatible with two-dimensional systems as well. We introduce the quantum field operators for exciton polaritons $\hat{\Psi}_x$ connected with annihilation operators in reciprocal (k) space by the Fourier transforms ($\mathcal{F}$ in short notation),

$$\hat{a}_k = \mathcal{F}[\hat{\Psi}_x] = \frac{1}{\sqrt{N}} \sum_x \hat{\Psi}_x e^{-ikx}, \quad \hat{\Psi}_x = \mathcal{F}^{-1}[\hat{a}_k],$$

where N is the discretization length.

The Hamiltonian of the system reads

$$\begin{aligned} \hat{\mathcal{H}} = & \sum_k E_k \hat{a}_k^\dagger \hat{a}_k + \sum_x \mathcal{P}_x e^{i(k_p x - \omega_p t)} (\hat{\Psi}_x^\dagger + \hat{\Psi}_x) \\ & + \sum_x (V_x \hat{\Psi}_x^\dagger \hat{\Psi}_x + \alpha \hat{\Psi}_x^\dagger \hat{\Psi}_x^\dagger \hat{\Psi}_x \hat{\Psi}_x) + \sum_{\vec{q}} \hbar \omega_{\vec{q}} \hat{b}_{\vec{q}}^\dagger \hat{b}_{\vec{q}} \\ & + \sum_{\vec{q}, k} G_{\vec{q}} \hat{b}_{\vec{q}} \hat{a}_{k+q_x}^\dagger \hat{a}_k + G_{\vec{q}}^* \hat{b}_{\vec{q}}^\dagger \hat{a}_{k+q_x} \hat{a}_k^\dagger. \end{aligned} \quad (1)$$

The first two lines here correspond to coherent processes. E_k is the particle dispersion (which is nonparabolic for exciton polaritons); the last term in the first line is the coherent pumping, where $\mathcal{P}_x$ is the pump intensity, k_p is the wave

vector of the pump, corresponding to the inclination of an incident laser beam, and $\hbar \omega_p$ is the pumping energy; V_x is the potential profile in x space, and α is a constant describing the strength of particle-particle interactions.

The last line in the equation above corresponds to incoherent processes. To model the interaction with acoustic phonons, we introduce the Fröhlich Hamiltonian [12,34], where the phonons described by operators $\hat{b}_{\vec{q}}^\dagger$, $\hat{b}_{\vec{q}}$ are considered to be three dimensional. The phonon wave vector is $\vec{q} = \vec{e}_x q_x + \vec{e}_y q_y + \vec{e}_z q_z$, where $\vec{e}_x$, $\vec{e}_y$, and $\vec{e}_z$ are unit vectors: $\vec{e}_x$ is in the wire direction and $\vec{e}_z$ is in the structure growth direction. The phonon dispersion relation, $\hbar \omega_{\vec{q}} = \hbar u \sqrt{q_x^2 + q_y^2 + q_z^2}$, is determined by the speed of sound u . $G_{\vec{q}}$ is the exciton-phonon interaction strength, whose calculation can be found elsewhere [34–38] (see also the detailed derivation of the formalism in the Supplemental Material [39]).

Using the Heisenberg equations of motion for the operators, one can write the formal solution for the phonon field as follows:

$$\begin{aligned} \hat{b}_{\vec{q}}(t) = & \hat{b}_{\vec{q}}(0) e^{-i\omega_{\vec{q}} t} \\ & - \frac{i}{\hbar} \int_0^t G_{\vec{q}}^* \sum_k \hat{a}_{k+q_x}^\dagger(t') \hat{a}_k(t') e^{-i\omega_{\vec{q}}(t-t')} dt'. \end{aligned} \quad (2)$$

Remembering that phonons represent an incoherent thermal reservoir, we can replace the term $\hat{b}_{\vec{q}}(0) e^{-i\omega_{\vec{q}} t}$ by a stochastic classical variable $b_{\vec{q}}(t)$ (and a similar replacement can be made for the conjugate field). This represents the analogue of the Markov approximation within the Langevin approach, when phonons are assumed to have a randomly varying phase [35]. The stochastic variables are complex numbers with real and imaginary components normalized as follows:

$$\begin{aligned} \langle b_{\vec{q}}^*(t) b_{\vec{q}'}(t') \rangle &= n_{\vec{q}} \delta_{\vec{q}\vec{q}'} \delta(t - t'), \\ \langle b_{\vec{q}}(t) b_{\vec{q}'}(t') \rangle &= \langle b_{\vec{q}}^*(t) b_{\vec{q}'}^*(t') \rangle = 0, \end{aligned} \quad (3)$$

where $n_{\vec{q}}$ is the number of phonons in the state with wave vector $\vec{q}$ determined by the temperature of the system.

The exciton-polariton field dynamics can then be determined solely by the exciton-polariton operators and the stochastic terms. Further, within the mean field approximation, the field operator $\hat{\Psi}_x$ can be treated as a classical variable for condensed exciton polaritons, $\psi_x = \langle \hat{\Psi}_x \rangle$ (with the Fourier image ψ_k). Then, physical observables are calculated over multiple realizations of the evolution dynamics with the stochastic variables $b_k(t)$. The corresponding equation of motion reads (see the Supplemental Material [39] for the details of the derivation)

$$\begin{aligned} i\hbar \frac{d\psi_x}{dt} = & \mathcal{F}^{-1} [E_k \psi_k + \mathcal{S}_k(t)] + \left[V_x + \alpha |\psi_x|^2 - i \frac{\hbar \gamma}{2} \right] \psi_x \\ & + \mathcal{P}_x e^{ik_p x} e^{-i\omega_p t} + \sum_k \{ \mathcal{T}_{-k}(t) + \mathcal{T}_k^*(t) \} e^{-ikx} \psi_x, \end{aligned} \quad (4)$$

where we have introduced phenomenologically the decay term $-i\hbar\gamma\psi_x/2$ to account for the radiative decay of particles [21]. The constant α describing polariton-polariton interactions can be estimated as [37] $\alpha \approx E_b a_B^2 / (L_y \Delta x)$, where L_y is the lateral size of the microwire and $\Delta x = L_x / N$ is the discretization unit.

One can see that interaction with phonons leads to the appearance of two types of terms. First, one has a term

$$\mathcal{S}_k(t) = \sum_{q_x} \psi_{k+q_x}(t) \left(\int_0^t \mathcal{A}_{q_x}(t') \mathcal{K}_{q_x}(t-t') dt' \right), \quad (5)$$

where $\mathcal{A}_{q_x}(t) = \sum_{k'} \psi_{k'+q_x}^*(t) \psi_{k'}(t)$. The term is proportional to the cube of the polariton field and does not directly include a stochastic term. It can be thus interpreted as the term corresponding to the emission of phonons by a condensate stimulated by the polariton density. The convolution integral is responsible for energy conservation. Note that the function

$$\begin{aligned} \mathcal{K}_{q_x}(t) &= - \sum_{q_y, q_z} |G_{\vec{q}}|^2 (e^{-i\omega_{\vec{q}} t} - e^{i\omega_{\vec{q}} t}) \\ &\rightarrow 2i \frac{L_z}{2\pi} \frac{a_B}{2\pi} \iint |G(\vec{q})|^2 \sin[\omega(\vec{q})t] dq_y dq_z \end{aligned} \quad (6)$$

is approximately independent of q_x in the range of $q_x \in (-10^8, 10^8) \text{ m}^{-1}$, and thus in our calculations we put $\mathcal{K}_{q_x}(t) \approx \mathcal{K}_0(t)$.

The stochastic functions $\mathcal{T}_{q_x}$ and $\mathcal{T}_{q_x}^*$ in the last line of Eq. (4) are defined by the correlators

$$\begin{aligned} \langle \mathcal{T}_{q_x}^*(t) \mathcal{T}_{q'_x}(t') \rangle &= \sum_{q_y, q_z} |G_{q_x, q_y, q_z}|^2 n_{q_x, q_y, q_z} \delta_{q_x, q'_x} \delta(t-t'), \\ \langle \mathcal{T}_{q_x}(t) \mathcal{T}_{q'_x}(t') \rangle &= \langle \mathcal{T}_{q_x}^*(t) \mathcal{T}_{q'_x}^*(t') \rangle = 0. \end{aligned} \quad (7)$$

These thermal terms contain the phonon field and so are strongly temperature dependent. The proportionality of the thermal part to the first power of ψ_x in Eq. (4) means that the scattering processes proceed at a rate proportional to the first power in exciton-polariton density. Consequently, this term corresponds to the absorption of the phonons by the polariton ensemble and their emission which is stimulated by the final state phonon occupancy, but not the exciton-polariton density. The latter processes are instead represented by the stimulated term [Eq. (5)] considered above.

Results.—We considered an InGaAlAs alloy-based microcavity and in computations used the following set of parameters: speed of sound $u = 5370 \text{ m/s}$ [34], $\gamma = 1/18 \text{ ps}^{-1}$ [15]. The function $V_x = V_0 - \beta x$ is the exciton-polariton potential in x space, composed of the potential defining the walls of the exciton-polariton wire V_0 and a potential gradient $\beta = 9 \text{ meV/mm}$. We consider a linear potential gradient as in the experiment of Ref. [15]. Our formalism would however be compatible with any potential shape and could be applied to, for example, the parabolic [6] or staircase [14] potentials studied recently in experiments. The exciton-polariton dispersion was calculated using a two oscillator model with a cavity photon effective mass of 4×10^{-5} of the free electron mass, a

Rabi splitting of 10 meV, and an exciton-photon detuning of -2.5 meV at zero in-plane wave vector.

The main phenomenon we investigated was the relaxation of energy of exciton polaritons (thermalization) caused by the interaction with acoustic phonons. Exciton polaritons were introduced to the system by a localized coherent pump with energy coinciding with the bottom of the lower polariton branch (zero detuning case) which guarantees that we are outside the bistable regime.

The stochastic equation [Eq. (4)] can be solved numerically. The results of the modeling are presented in Figs. 1 and 2(a)–2(f). Figure 1 illustrates the propagation of particles created by a short pulse along the one-dimensional wire. The pumping is switched on during the first 20 ps of the theoretical experiment, and then it is switched off and we observe the decay of intensity due to the finite particle lifetime. The quantity $|\psi_x|^2$ is depicted for different times: 40, 60, and 80 ps (t is the parameter). One can see that the wave packet of exciton polaritons propagates along the channel with time, disperses, and accumulates at the right-hand side. Note, that reflection from the potential jump at the end of the wire produces the pronounced interference fringes clearly visible in recent experiments [40].

Figure 2 illustrates the energy relaxation in the system for three time intervals: 0–50 ps, 50–100 ps, and 100–150 ps, correspondingly. It is clearly seen that exciton polaritons pumped at $x = 0, k = 0$ [see Figs. 2(a) and 2(b)] propagate along the potential slope losing their energy [see Figs. 2(c) and 2(d)] and accumulate on the right-hand side of the wire [see Fig. 2(e)] thus condensing at $k = 0$ in reciprocal space [see Fig. 2(f)]. The plots [see Figs. 2(a), 2(c), and 2(e)] provide snapshots of the x space dynamics of the system; the k space dynamics is

FIG. 1 (color online). Propagation of particles along the potential slope in the quantum wire due to phonon-assisted relaxation. The curves correspond to the particle concentration profile in x space for different times: 40 (red or dotted line), 60 (green or dashed line), and 80 ps (blue or solid line). The particles are introduced by a coherent Gaussian pump of the duration of 20 ps of the theoretical experiment. Particles propagate along the wire (green and blue curves), suffering losses caused by their finite lifetime and reflecting from the end of the wire. The interference of the incoming and reflected waves produces fringes clearly visible at 80 ps.

FIG. 2 (color online). Relaxation of energy of exciton polaritons along the potential gradient in the regime of *cw* excitation in a quantum wire due to phonon-assisted processes in *x* and *k* space for different time ranges: 0–50 ps [(a), (b)], 50–100 ps [(c), (d)], and 100–150 ps [(e), (f)]. White curves show the model potential profile in the real space and free exciton-polariton dispersion in the reciprocal space used in calculations. Coherent pumping represents a Gaussian pump in *x* space centered around *x* = 0. The inclination angle of the pump is zero, thus $k_p = 0$ and $\hbar\omega_p = E(0)$. Polaritons created at the pumping spot (a) propagate along the channel (c) and accumulate on its right-hand side (e). The *k* space behavior shows the decrease of the condensate energy [(b),(d),(f)].

depicted in plots [see Figs. 2(b), 2(d), and 2(f)]. The white dashed curves in the figures correspond to the potential profile (in *x* space) and free polaritons dispersion (in *k* space). It should be noted, that the relaxation with phonons is an efficient process; thus, particles rapidly move towards the bottom of the slope and accumulate there, and their concentration near the pumping spot soon becomes lower than the concentration in the signal point.

Discussion.—Comparing with previous theoretical models aimed at a description of energy relaxation in coherent condensates, we would like to stress that all the parameters, including phonon scattering rates, are calculated microscopically rather than being treated in a phenomenological way. Furthermore our approach allows us the possibility to treat large regions of direct and reciprocal space with

reasonably low computational cost. This is in contrast to the theory developed in Ref. [30] where the need to calculate an evolving particle spectrum in a coarse time window was extremely memory demanding. In the approach of Ref. [29] the analysis was restricted to a single quantum state and the energy relaxation was introduced phenomenologically. Very recently, in Ref. [41], an approach merging the Boltzmann equations with a Gross-Pitaevskii treatment was developed where Boltzmann scattering rates are dynamically calculated from the mean-field wave functions. The theory was successful in the description of micropillars with a small number of confined states, but it is not obvious how efficient the theory would be in the description of many different modes in a large reciprocal space.

Comparing our results to the Monte Carlo approach developed in Ref. [42], it should be noted that we are seeking to address a different problem. In Ref. [42] the focus was on how spontaneous emission introduces decoherence in parametric oscillators. While a white noise term was introduced, it did not describe energy-relaxation processes. These require coupling to an incoherent (phonon) field to which the energy is transferred. Unlike in Ref. [42], our theory is temperature dependent and allows a dynamic description of spatially dependent energy relaxation. To address coherence properties, an approach based on a matrix of correlators would be more appropriate [43,44]. Unfortunately, such an approach is very demanding computationally, requiring matrices of size $N \times N$, where *N* represents the number of states in reciprocal space.

Finally, while we have considered the energy relaxation of exciton polaritons via phonon interactions, we should note that another relaxation mechanism is provided by scattering processes involving hot excitons with large momentum [20]. Such hot excitons are typically created in nonresonant or incoherently pumped systems [40] but can be neglected under the resonant coherent excitation which we consider in this manuscript. In principle, the description of scattering with high momentum excitons can be accommodated within our formalism. This would allow the theoretical study of the interplay between both exciton mediated and phonon mediated scattering processes in extended systems and is an important direction for future research.

Conclusion.—We have derived a stochastic Gross-Pitaevskii equation, where the energy relaxation of bosons is provided by coupling to an incoherent field, treated as a stochastic variable. As an example, we applied the theory to the modeling of exciton polaritons in a one-dimensional microwire with a potential gradient. The partial thermalization of exciton polaritons is observed, together with their trapping in real space. This result is of particular relevance to a variety of recent experiments in exciton and exciton-polariton systems where the energy relaxation of propagating Bose-Einstein condensates was reported.

By replacing the Frölich exciton-phonon interaction part of the Hamiltonian with a suitable interaction between

condensed and noncondensed atoms [33], dynamical energy relaxation could also be modeled with our approach in cold atom condensates.

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