Induced Bose-Einstein condensation of polaritons in crystals of various dimensionalities

V. P. Mis'ko, S. A. Moskalenko, A. Kh. Rotaru, and Yu. M. Shvera

Institute of Applied Physics, Moldavian SSR Academy of Sciences, Kishinev

(Submitted 29 June 1990)
Zh. Eksp. Teor. Fiz. **99**, 1215–1229 (April 1991)

We discuss the statistical properties of coherently excited polaritons, including the phenomenon of induced Bose-Einstein condensation, in crystals of various dimensionalities from $0D$ to $3D$. Our model system is a crystallite of finite volume which is embedded in a glassy matrix located in an optical resonator. External laser radiation with a frequency ω_L and energy flux S_L directly excites a single polariton mode of this crystallite with wave vector $\mathbf{k}_0$; the bare frequency $\omega(\mathbf{k}_0)$ of this polariton is detuned from resonance by an amount $\Delta\omega = \omega(\mathbf{k}_0) - \omega_L$. The polariton-polariton interaction leads to the removal of two quasiparticles from the mode $\mathbf{k}_0$ and the creation of two scattered polaritons with wave vectors $\mathbf{k}_0 \pm \varkappa$; this process in turn influences the original induced mode $\mathbf{k}_0$ in a self-consistent fashion. We find self-consistent steady-state solutions to the Fokker-Planck equation for the $\mathbf{k}_0$-mode polaritons and kinetic equations for the scattered quasiparticles belonging to the lower polariton branch. The nonequilibrium distribution function of the latter possesses a smeared-out threshold for stimulated Raman scattering. In this case, the coherently excited polaritons exhibit properties such as bunching, anti-bunching, and compressed states. These effects occur at small values of the product $S_L V$ (where V is the volume of the crystallite) and at different detunings from resonance. As $S_L V$ increases, the coherently excited polaritons become completely coherent; in the limit of a bulk crystal they acquire the properties of a macroscopically-occupied Bose-condensate. Among these properties is optical bistability, which is observed as a function of S_L.

1. INTRODUCTION

The first self-consistent investigations of condensed modes of excitons and photons combined with kinetic equations to describe exciton-phonon scattering appear in the papers of Ivanov, Keldysh, and Tikhodeev.[1-3] These authors noted the appearance of energy spectra of the "phonoriton" type, along with the phenomenon of stimulated Brillouin scattering. In contrast to the Bogolyubov model[4] of a weakly nonideal Bose gas, Bose-Einstein condensation (BEC) of dipole-active excitons and photons[5] or polaritons[6] leads to both absolute and convective instabilities in the spectra of the elementary excitations that lie outside the condensate.[5,6] The generation and amplification of new types of waves under steady-state conditions results in new condensed modes, a process that is equivalent to stimulated Raman scattering and is analogous to the processes that accompany laser oscillation on band-to-band transitions.[7]

In this paper we discuss the physical processes that occur during coherent excitation of a specific polariton mode by external laser radiation in crystals of various dimensionality, including bulk samples and crystallites embedded in a glassy matrix placed in a resonator. The glassy matrix serves as a heat bath; for the proper choice of the linear dimension d of the crystallite, the resonator allows a single coherent mode with wave vector $\mathbf{k}_0$ and bare frequency $\omega(\mathbf{k}_0)$ to be selectively excited. Assume that the laser radiation has a frequency ω_L and a power flux S_L in the vacuum; in general, we will also assume that the detuning $\Delta\omega = \omega(\mathbf{k}_0) - \omega_L$ from resonance with the laser is different from zero. The processes that occur in the course of establishing BEC depend on the quantities S_L, $\Delta\omega$, the polariton-polariton interaction constant g, and the crystal volume $V = d^3$.

As the dimensionality of the crystal changes from $3D$ to $0D$ there is a nontrivial alteration in the role played by quantum fluctuations in the number of quasiparticles, along with changes in the corresponding diffusion terms of the Fokker-Planck equation (FPE) for the reduced density matrix of the selected mode. When the volume of the mode $\mathbf{k}_0$ is large, i.e., $V \to \infty$, even small coherent pumping amplitudes can give rise to a macroscopically occupied state; this suppresses the diffusion term in the FPE and favors a drift, i.e., deterministic, description of the induced Bose condensate. For small finite volumes V a certain intensity of pumping is necessary to suppress the fluctuations. Under these conditions, new states and processes appear, which will be described below.

In place of a deterministic description of the condensed mode, which is valid for bulk crystals,[1-2] we will use a more general quantum-statistical description which is also correct when the approximation of a specified field is not applicable. We start with the master equation for the reduced density matrix $\hat{\rho}_{\mathbf{k}_0}(t)$ of the mode $\mathbf{k}_0$, or more specifically with the corresponding FPE. Following Ref. 6, we will investigate polariton-polariton scattering, along with the process of stimulated Raman scattering whereby two quasiparticles are simultaneously created out of the coherently excited condensed mode $\mathbf{k}_0$; we will assume that the condensed-mode polaritons belong to the lower polariton branch, and that they are converted into two scattered quasiparticles on the same branch. For simplicity we will not include the upper polariton branch.

Polaritons in $0D$ systems can exist only when their wave vectors $\mathbf{k}$ and frequencies ω satisfy the inequality $k = \omega n(\omega)/c \geqslant 2\pi/d$, where $n(\omega)$ is the index of refraction. For the lower polariton band $n(\omega)$ increases rapidly when the frequency approaches $\omega_\perp$ from below, i.e., from within the polariton gap. Size quantization of polaritons is ob-

 0038-5646/91/040676-09$03.00

served[9,10] in thin films of CdS and GaAs with thicknesses d equal to 1000 and 990–2010 Å, respectively. We expect that there is a cutoff frequency ω_1 for the lower-branch polaritons in crystallites with these values of d. For smaller values of energy and wave vector it is possible to speak of the constituent parts of the polariton individually, i.e., excitons whose centers of gravity execute a size-quantized motion and photons with a continuous energy spectrum.

Thus, we are dealing here with quasiparticles of a single type that have a mixed discrete-continuous energy spectrum. Since we are primarily interested in crystals having dimensions $d \approx 600$–2000 Å, we will not take the discreteness of the spectrum into account in explicit form. It is important primarily in the region of values of d smaller than 500 Å down to sample thicknesses on the order of the exciton diameter. In this range of d the polariton description is entirely replaced by the exciton-photon description.[11]

Strictly speaking, the law of conservation of momentum does not hold in crystallites; it only serves to indicate the most probable quantum transitions. In the next section we will build this fact explicitly into the polariton interaction Hamiltonian. Our results contradict Ref. 12, in that they indicate an additional inhomogeneous broadening of the two-quasiparticle quantum transitions involved in the scattering process. Thus, e.g., in Fig. 1 we use the two pairs of points *1–2* and *3–4* to illustrate the production of two quasiparticles from the coherently excited mode $\mathbf{k}_0$ in the crystal bulk.[6] If we do not include homogeneous broadening of the polariton spectrum, the constraints imposed by conservation of energy and momentum imply that real production is possible only when the states *1–4* are discrete. This leads to singularities in the kinetic equations of Ref. 12. In crystals there is an additional inhomogeneous broadening because states *2* and *4* (Fig. 1) are located in the continuous photon-like portion of the lower polariton band, and states *1* and *3* are not coupled to *2* and *4* through the law of conservation of momentum but only that of conservation of energy.

Our work is based on a quantum-statistical description of the process by which an induced coherent state is established in macroscopic systems with various dimensionless volumes $N \sim V$ and numbers $N_{\mathbf{k}_0} \sim V$ of coherently excited quasiparticles of the special mode $\mathbf{k}_0$. The quantity $N_{\mathbf{k}_0}$ depends not only on N but also on the dimensionless power flux $Y \sim S_L$ of the laser radiation and the distribution function of scattered quasiparticles. Although our system is more complicated than the model investigated in Ref. 13, in which a single nonlinear oscillator interacts with a heat bath, the two models are nevertheless very close with regard to the state of the individual mode. The difference between them arises because in Ref. 13 the volume of the mode is fixed ($N = 1$), whereas in our model we can track the way N influences the processes occurring in the system as it varies from 1 to ∞. By describing in detail the nonequilibrium distribution function of the scattered polaritons on an equal footing with the polaritons of the special mode, we can track how the real excitation of the mode $\mathbf{k}_0$ takes place under conditions where the detuning from resonance $\Delta\omega$ is different from zero. The existence of nonequilibrium scattered quasiparticles under steady-state conditions ensures that the law of conservation of energy is fulfilled for each microscopic quantum transition event, leading to the classical picture of forced oscillations of a damped oscillator under the action of an external periodic force.

Our work shows that the mode volume N and the average number of coherently excited particles $N_{\mathbf{k}_0}$ affect the evolution of the statistical properties of the $\mathbf{k}_0$ mode in various ways. Thus, for example, the diffusion terms in the FPE are proportional to N^{-1} while the statistical properties of the coherently excited polaritons depend on the product NY. For small values of NY the phenomena of bunching and antibunching are possible, as well as the appearance of compressed states. We can show that there are internal similarities between the way quantum fluctuations affect the statistical properties of the polaritons in the selectively-excited mode and the way many-body quantum transitions affect the creation of exciton absorption bands for various detunings $\Delta\omega$ from resonance and small intensities of excitation light. As $NY \to \infty$ a macroscopically large coherent state is formed, which is in fact an induced Bose condensate. We will describe the basic features of the phenomenon of optical bistability which occurs under these conditions.

Coherent pumping can be introduced into the equation of motion for the average value $\langle a_{\mathbf{k}_0} \rangle$ of the annihilation operator $a_{\mathbf{k}_0}$ for polaritons. To do this we must relate the intensity of the external laser radiation field to the photon component of the polariton mode $\mathbf{k}_0$, using the Maxwell-Fresnel boundary conditions at the surface of the resonator mirror.[14,15] We will introduce this source of pumping into

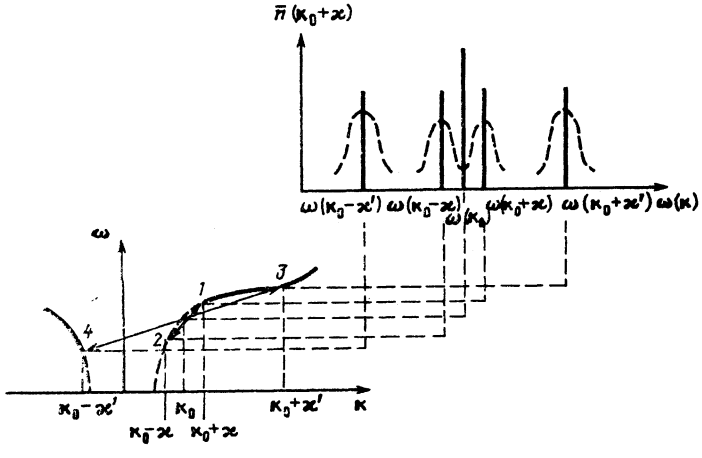

FIG. 1. Dispersion law for the lower polariton band in crystallites and distribution function of scattered polaritons.

the Hamiltonian in a way that is equivalent to the boundary conditions.

2. MODEL HAMILTONIAN, FOKKER-PLANCK EQUATION, AND KINETIC EQUATIONS

The system consisting of the polaritons (S), the heat bath (T), and the source of coherent laser radiation (L) are described by the following Hamiltonian, which is written in the rotating-wave approximation using a coordinate system rotating with frequency ω_L:

$$\mathcal{H}=\mathcal{H}_{S0}+\mathcal{H}_{SL}+\mathcal{H}_{S,int}+\mathcal{H}_T+\mathcal{H}_{ST}. \tag{1}$$

The free-polariton Hamiltonian $\mathcal{H}_{S0}$ has the form

$$\mathcal{H}_{S0}=\sum_{\mathbf{k}}\hbar(\omega(\mathbf{k})-\omega_L)a_{\mathbf{k}}^{+}a_{\mathbf{k}}, \tag{2}$$

where $a_{\mathbf{k}}^{+}$, $a_{\mathbf{k}}$ are creation and annihilation operators. The Hamiltonians for the interaction of polaritons of the $\mathbf{k}_0$ mode with the external laser radiation $\mathcal{H}_{SL}$ and among themselves $\mathcal{H}_{S,\text{int}}$ are as follows:

$$\begin{aligned}\mathcal{H}_{SL}&=i[(dE_0)a_{\mathbf{k}_0}^{+}-(dE_0)^{*}a_{\mathbf{k}_0}],\\ \mathcal{H}_{S,int}&=\frac{1}{2V}\sum_{\mathbf{pq}\sigma}g\Phi(\mathbf{k},\sigma)a_{\mathbf{p}}^{+}a_{\mathbf{q}}^{+}a_{\mathbf{q}+\mathbf{k}}a_{\mathbf{p}-\sigma}\\ &=\hat{U}+\hat{L}+\hat{F}^{+}+\hat{F}+\hat{P}^{+}+\hat{P}+\ldots,\end{aligned} \tag{3}$$

where E_0 is the amplitude of the forcing field within the resonator. Its relation to the external laser radiation will be identified by following Refs. 14 and 15; here d is the dipole moment of the transition. The interaction parameter is chosen to be a constant g for values of the wave vector that do not exceed the inverse exciton radius a_{ex}^{-1}, and to vanish for all other ranges of wave vector. The function $\Phi(\mathbf{k},\sigma)$ takes into account the the deviation from the law of momentum conservation; as $V\to\infty$ it reduces to a Kronecker δ symbol. For simplicity we will assume that the smeared-out function $\Phi(\mathbf{k},\sigma)$ retains the property $\Phi(\mathbf{k},\sigma)=\Phi(\mathbf{p}-\sigma,\mathbf{p}-\mathbf{k})$. It is expedient to write out those parts of the Hamiltonian $\mathcal{H}_{S,\text{int}}$ that correspond to the processes that are most interesting:

$$\begin{aligned}\hat{U}&=\frac{g\Phi(0,0)}{2V}\hat{A}^{+}\hat{A},\qquad \hat{L}=\frac{2\hat{N}_{\mathbf{k}_0}}{V}\sum_{\mathbf{q}\varkappa}g\Phi(\varkappa,0)a_{\mathbf{k}_0+\mathbf{q}}^{+}a_{\mathbf{k}_0+\mathbf{q}+\varkappa},\\ \hat{F}^{+}&=\frac{\hat{A}^{+}}{2V}\sum_{\mathbf{k}\varkappa}g\Phi(\mathbf{k},\varkappa)a_{\mathbf{k}_0+\mathbf{k}}a_{\mathbf{k}_0-\varkappa},\\ \hat{P}^{+}&=\frac{a_{\mathbf{k}_0}^{+}}{V}\sum_{\mathbf{pq}\varkappa}g\Phi(\mathbf{q},\varkappa)a_{\mathbf{k}_0+\mathbf{p}}^{+}a_{\mathbf{k}_0+\mathbf{p}+\mathbf{q}}a_{\mathbf{k}_0-\varkappa},\\ \hat{A}^{+}&=(a_{\mathbf{k}_0}^{+})^2,\quad \hat{N}_{\mathbf{k}_0}=a_{\mathbf{k}_0}^{+}a_{\mathbf{k}_0},\quad \hat{B}=\hat{A}^{+}\hat{A},\quad \hat{C}=\hat{A}\hat{A}^{+}.\end{aligned} \tag{4}$$

The operators $\hat{F}^{+}$ and $\hat{F}$ describe the conversion of two quasiparticles belonging to the mode $\mathbf{k}_0$ into two scattered quasiparticles with momenta $\mathbf{k}_0+\varkappa$ and $\mathbf{k}_0-\varkappa$. According to Ref. 6, for a bulk crystal the points *1* and *2* shown in Fig. 1 are located at a distance on the order of k_0 from the point $\mathbf{k}_0$, while points *3* and *4* are at a distance on the order of $2k_0$ away. The operators $\hat{P}^{+}$ and $\hat{P}$ describe scattering processes with the participation of one quasiparticle of the mode $\mathbf{k}_0$. The explicit form of the terms $\mathcal{H}_T$ and $\mathcal{H}_{ST}$ is not presented here. They are included in the standard way and give rise to phenomenological constants in the master and kinetic equations.[16,17]

The quantum Liouville equation for the density matrix of the entire system (1) is solved by perturbation-theoretic methods in the Markov approximation that parallel the theory of quantum fluctuations in lasers.[16,18,9] Using first-order perturbation theory for the operators $\hat{U}+\hat{L}$ and second-order theory for the operators $\hat{F}^{+}+\hat{F}+\hat{P}^{+}+\hat{P}$, we find the master equation for the reduced density matrix $\hat{\rho}_{\mathbf{k}_0}(t)$ of polaritons in the mode $\mathbf{k}_0$:

$$\begin{aligned}\frac{\partial\hat{\rho}_{\mathbf{k}_0}(t)}{\partial t}&=[(dE_0/\hbar)a_{\mathbf{k}_0}^{+}-(dE_0/\hbar)^{*}a_{\mathbf{k}_0},\hat{\rho}_{\mathbf{k}_0}(t)]\\ &-i\Big\{[\Delta\omega+l/\hbar+p_1-q_1+m(\mathbf{k}_0)][\hat{N}_{\mathbf{k}_0},\hat{\rho}_{\mathbf{k}_0}(t)]\\ &+\frac{1}{2V}[g\Phi(0,0)/\hbar+f_1][\hat{B},\hat{\rho}_{\mathbf{k}_0}(t)]-\frac{g_1}{2V}[\hat{C},\hat{\rho}_{\mathbf{k}_0}(t)]\Big\}\\ &-\Big\{\frac{f_2}{2V}([\hat{A}^{+},\hat{A}\hat{\rho}_{\mathbf{k}_0}(t)]+[\hat{\rho}_{\mathbf{k}_0}(t)\hat{A}^{+},\hat{A}])\\ &+\frac{g_2}{2V}([\hat{A},\hat{A}^{+}\hat{\rho}_{\mathbf{k}_0}(t)]+[\hat{\rho}_{\mathbf{k}_0}(t)\hat{A},\hat{A}^{+}])\\ &+\Big(p_2+\frac{\gamma_1(\mathbf{k}_0)}{2}\Big)([a_{\mathbf{k}_0}^{+},a_{\mathbf{k}_0}\hat{\rho}_{\mathbf{k}_0}(t)]+[\hat{\rho}_{\mathbf{k}_0}(t)a_{\mathbf{k}_0}^{+},a_{\mathbf{k}_0}])\\ &+\Big(q_2+\frac{\gamma_2(\mathbf{k}_0)}{2}\Big)([a_{\mathbf{k}_0},a_{\mathbf{k}_0}^{+}\hat{\rho}_{\mathbf{k}_0}(t)]+[\hat{\rho}_{\mathbf{k}_0}(t)a_{\mathbf{k}_0},a_{\mathbf{k}_0}^{+}])\Big\}.\end{aligned} \tag{5}$$

The coefficients l, f_i, g_i, p_i, and q_i depend on the average occupation numbers $\bar{n}_{\mathbf{k}_0+\varkappa}$ of scattered polaritons; we will present a kinetic equation for these quantities in what follows. The quantities $m(\mathbf{k}_0)$, $\gamma_i(\mathbf{k}_0)$ are determined by interactions with the heat bath and do not depend on the state of the system S. Explicit expressions for the coefficients f_i, g_i, p_i, and q_i will be given only in those combinations that will be encountered in the FPE. The latter is obtained by using the nondiagonal P-representation based on the Glauber coherent states[20] $|\alpha\rangle$ and $|\beta\rangle$, as proposed by Drummond and Gardiner:[21]

$$\hat{\rho}_{\mathbf{k}_0}(t)=\int_C d\alpha\int_{C'}d\beta\,P(\alpha,\beta,t)\frac{|\alpha\rangle\langle\beta^{*}|}{\langle\beta^{*}|\alpha\rangle}. \tag{6}$$

Here α and β are assumed to be independent variables which vary along the contours C and C'. In our case α and β are macroscopic variables, and we will indicate their dependence on volume explicitly:

$$\alpha=N^{1/2}\xi,\quad \beta=N^{1/2}\eta,\quad E_0=N^{1/2}\mathcal{E}_0,\quad N=Vn_c. \tag{7}$$

The dimensionless volume N can be introduced in various ways. We use the concentration n_c, which will appear below in the course of finding $\bar{n}_{\mathbf{k}_0+\varkappa}$. Finally, after a few simplifications, we find the following FPE:

$$\begin{aligned}\frac{\partial P(\xi,\eta,t)}{\partial t}&=\Big\{\frac{\partial}{\partial\xi}\Big[K\xi+2\chi\xi^2\eta-\frac{d\mathcal{E}_0}{\hbar}\Big]\\ &+\frac{\partial}{\partial\eta}\Big[K^{*}\eta+2\chi^{*}\eta^2\xi-\Big(\frac{d\mathcal{E}_0}{\hbar}\Big)^{*}\Big]-\\ &-\frac{\chi}{N}\frac{\partial^2}{\partial\xi^2}\xi^2-\frac{\chi^{*}}{N}\frac{\partial^2}{\partial\eta^2}\eta^2\Big\}P(\xi,\eta,t).\end{aligned} \tag{8}$$

The basis of our simplified approach is the approximation that the density of scattered quasiparticles n_1 is small compared to the density of coherently excited polaritons n_0 (i.e., $n_1 < n_0$), where

$$n_0 = \frac{\langle \hat{N}_{\mathbf{k}_0} \rangle}{V}, \quad n_1 = \frac{1}{V} \sum_{\varkappa} \bar{n}_{\mathbf{k}_0+\varkappa}, \tag{9}$$

and the limitation to values $N \geqslant 1$. This latter condition bounds the size of the crystallites from below. These arguments allow us to discard nondiagonal diffusion terms and terms containing products of higher order. The square brackets on the right side of the FPE contain drift terms, which specify the degree of deterministic behavior of the $\mathbf{k}_0$-mode polaritons. The remaining two terms are diagonal diffusion terms, which describe quantum fluctuations of the polaritons of this mode. The coefficients K and χ that enter into the FPE are connected with the previously defined coefficients by the relations

$$K = i\overline{\Delta\omega} + \sigma, \quad \chi = i\mu + \delta, \tag{10}$$

where

$$\begin{aligned} \overline{\Delta\omega} &= \Delta\omega + \frac{l}{\hbar} + (p_1 - q_1) + m(\mathbf{k}_0), \\ \sigma &= \frac{\gamma(\mathbf{k}_0)}{2} + (p_2 - q_2), \quad \delta = \frac{n_c}{2}(f_2 - g_2), \\ \mu &= n_c \left[\frac{g\Phi(0,0)}{\hbar} + \frac{1}{2}(f_1 - g_1) \right], \\ \gamma(\mathbf{k}_0) &= \gamma_1(\mathbf{k}_0) - \gamma_2(\mathbf{k}_0) > 0, \quad m(\mathbf{k}_0) = m_1(\mathbf{k}_0) - m_2(\mathbf{k}_0). \end{aligned} \tag{11}$$

The coefficients $l, f_1 - g_1$, and $p_1 - q_1$ that enter into Eq. (11) have the following form when the deviation from momentum conservation is taken into account:

$$\begin{aligned} l(t) &= \frac{2}{V} \sum_{p \neq 0} g\Phi(0,0) \bar{n}_{\mathbf{k}_0+\mathbf{p}}(t), \\ f_1 - g_1 &= \frac{1}{V\hbar^2} \sum_{\mathbf{pq}} g^2 \Phi^2(\mathbf{p},\mathbf{q}) \mathscr{P} \frac{1}{\Omega(\mathbf{p},\mathbf{q})} (1 + \bar{n}_{\mathbf{k}_0+\mathbf{p}} + \bar{n}_{\mathbf{k}_0-\mathbf{q}}), \\ p_1 - q_1 &= \frac{2}{V\hbar^2} \sum_{\mathbf{pq}\varkappa} g^2 \Phi^2(\mathbf{q},\varkappa) \mathscr{P} \frac{1}{\Theta(\mathbf{p},\mathbf{q},\varkappa)} \\ &\times [(1 + \bar{n}_{\mathbf{k}_0+\mathbf{p}+\varkappa})(1 + \bar{n}_{\mathbf{k}_0-\varkappa}) \bar{n}_{\mathbf{k}_0+\mathbf{p}} - \bar{n}_{\mathbf{k}_0+\mathbf{p}+\mathbf{q}} \bar{n}_{\mathbf{k}_0-\varkappa} (1 + \bar{n}_{\mathbf{k}_0+\mathbf{p}})], \end{aligned} \tag{12}$$

where

$$\begin{aligned} \Omega(\mathbf{p},\mathbf{q}) &= 2\omega(\mathbf{k}_0) - \omega(\mathbf{k}_0+\mathbf{p}) - \omega(\mathbf{k}_0-\mathbf{q}), \\ \Theta(\mathbf{p},\mathbf{q},\varkappa) &= \omega(\mathbf{k}_0) + \omega(\mathbf{k}_0+\mathbf{p}) - \omega(\mathbf{k}_0+\mathbf{p}+\mathbf{q}) - \omega(\mathbf{k}_0-\varkappa). \end{aligned} \tag{13}$$

When $\Phi^2(\mathbf{p},\mathbf{q}) = \delta_{\mathbf{p},\mathbf{q}}$, Eqs. (12) and (13) reduce to Eq. (4) of Ref. 12, where $\Omega(\mathbf{p}) \equiv \Omega(\mathbf{p},\mathbf{p})$ and $\Theta(\mathbf{p},\mathbf{q}) \equiv \Theta(\mathbf{p},\mathbf{q},\mathbf{q})$. Expressions for $f_2 - g_2$ and $p_2 - q_2$ are obtained from the expressions for $f_1 - g_1$ and $p_1 - q_1$ by replacing the principle values involving the functions $\Omega^{-1}(\mathbf{p},\mathbf{q})$ and $\Theta^{-1}(\mathbf{p},\mathbf{q},\varkappa)$ by $\pi\delta(\Omega(\mathbf{p},\mathbf{q}))$ and $\pi\delta[\Theta(\mathbf{p},\mathbf{q},\varkappa)]$, respectively.

The average occupation number of scattered polaritons is found by solving the following system of kinetic equations:

$$\begin{aligned} \frac{\partial \bar{n}_{\mathbf{k}_0+\varkappa}(t)}{\partial t} &= -\gamma(\mathbf{k}_0+\varkappa) \bar{n}_{\mathbf{k}_0+\varkappa} + \frac{2\pi}{\hbar^2 V^2} \sum_{\mathbf{q}} g^2 \Phi^2(\varkappa,\mathbf{q}) \\ &\times \delta(\Omega(\varkappa,\mathbf{q})) [\langle \hat{B} \rangle (1 + \bar{n}_{\mathbf{k}_0+\varkappa})(1 + \bar{n}_{\mathbf{k}_0-\mathbf{q}}) - \langle \hat{C} \rangle \bar{n}_{\mathbf{k}_0+\varkappa} \bar{n}_{\mathbf{k}_0-\mathbf{q}}] \\ &+ \frac{4\pi}{\hbar^2 V^2} \sum_{\mathbf{q}\sigma} g^2 \Phi^2(\mathbf{q},\sigma) \delta(\Theta(\varkappa,\mathbf{q},\sigma)) \\ &\times [(1 + \langle \hat{N}_{\mathbf{k}_0} \rangle)(1 + \bar{n}_{\mathbf{k}_0+\varkappa}) \bar{n}_{\mathbf{k}_0+\varkappa+\mathbf{q}} \bar{n}_{\mathbf{k}_0-\sigma} - \langle \hat{N}_{\mathbf{k}_0} \rangle \\ &\times \bar{n}_{\mathbf{k}_0+\varkappa} (1 + \bar{n}_{\mathbf{k}_0+\varkappa+\mathbf{q}})(1 + \bar{n}_{\mathbf{k}_0-\sigma})] + \frac{8\pi}{\hbar^2 V^2} \sum_{\mathbf{pq}} g^2 \Phi^2(\mathbf{q},-\varkappa) \\ &\times \delta(\Theta(\mathbf{p},\mathbf{q},-\varkappa)) [\langle \hat{N}_{\mathbf{k}_0} \rangle \bar{n}_{\mathbf{k}_0+\mathbf{p}} (1 + \bar{n}_{\mathbf{k}_0+\varkappa})(1 + \bar{n}_{\mathbf{k}_0+\mathbf{p}+\mathbf{q}}) \\ &- (1 + \langle \hat{N}_{\mathbf{k}_0} \rangle)(1 + \bar{n}_{\mathbf{k}_0+\mathbf{p}}) \bar{n}_{\mathbf{k}_0+\varkappa} \bar{n}_{\mathbf{k}_0+\mathbf{p}+\mathbf{q}}]. \end{aligned} \tag{14}$$

The first term on the right side of system (14) contains damping constants $\gamma(\mathbf{k}_0 + \varkappa)$ of the same type as $\gamma(\mathbf{k}_0)$. The second term describes the conversion of two polaritons of the $\mathbf{k}_0$ mode into two scattered polaritons. The last two terms correspond to scattering processes in which only one out of the four quasiparticles involved in the scattering—two in the initial state and the two in the final state—belongs to the $\mathbf{k}_0$ mode.

To conclude this section, we specify the relation between the constant $d\mathscr{E}_0/\hbar$ for the source of coherent pumping and the power flux of the laser radiation S_L, using the example of a ring resonator with two semitransparent mirrors with reflection coefficients R and two opaque mirrors. In the space between the first two mirrors we place a glassy matrix in the form of a film of thickness L containing microcrystallites. Following Ref. 14, we then write boundary conditions that relate the electromagnetic field outside the resonator to the field inside it. We depart from Ref. 14 only in our replacement of the field within the resonator by the photon component of the mode-$\mathbf{k}_0$ polaritons. The relation we are looking for is the following:

$$Y = \left| \frac{d\mathscr{E}_0}{\hbar\gamma_{ef}} \right|^2 = \frac{S_L}{S_c} \frac{\Omega_{res}^2}{\gamma_{ef}^2} \zeta. \tag{15}$$

Here γ_{ef} has the sense of an effective attenuation for the polariton level and the following notation is introduced:

$$\begin{aligned} S_c &= \frac{\hbar\omega_L n_c \varepsilon_\infty}{2k_0}, \quad \Omega_{res}^2 = \frac{c^2(1-R)}{L^2\varepsilon_\infty}; \\ \zeta &= \frac{[\omega_{ex}(\mathbf{k}_0) - \omega(\mathbf{k}_0)]^2}{[\omega_{ex}(\mathbf{k}_0) - \omega(\mathbf{k}_0)]^2 + |\varphi_{\mathbf{k}_0}/\hbar|^2}, \quad \left| \frac{\varphi_{\mathbf{k}_0}}{\hbar} \right|^2 = \frac{\omega_T \omega_{LT} \varepsilon_\infty}{2}. \end{aligned} \tag{16}$$

The factor ζ indicates the fraction of photon component that enters into a polariton of frequency $\omega(\mathbf{k}_0)$; $\omega_{\text{ex}}(\mathbf{k}_0)$ is the exciton frequency, ε_∞ is the high-frequency dielectric constant of the crystallite, and c is the velocity of light in vacuum. Let us now turn to an investigation of the stationary states of this system.

3. STATIONARY SELF-CONSISTENT SOLUTION

Integrodifferential equations like (14) can only be solved approximately. In Eq. (14) we have explicitly taken into account the inhomogeneous broadening of the energy spectrum of scattered quasiparticles associated with the loss of quasimomentum conservation in finite-volume crystallites. We have also investigated a variant of Eq. (14) that

takes into account homogeneous broadening of the polariton energy due to two-particle losses from the mode $\mathbf{k}_0$ in a phenomenological way; our treatment of this equation is described in full in Ref. 12. Despite the differences in the two approaches, the qualitative properties of the solutions obtained and the conclusions that follow from them coincide.

The stationary solution to Eq. (14), i.e., for which the left-hand part equals zero, is given by the expression

$$\bar{n}_{\mathbf{k}_0+\varkappa}=\frac{[D_0(\varkappa)+G(\varkappa)]\mathscr{T}(-\varkappa)+D_0(\varkappa)[G(-\varkappa)-G(\varkappa)]}{\mathscr{T}(\varkappa)\mathscr{T}(-\varkappa)-D_0(\varkappa)[\mathscr{T}(\varkappa)+\mathscr{T}(-\varkappa)]}. \tag{17}$$

We have introduced the following notation and approximations:

$$D_0(\varkappa)=\frac{2\pi\langle\hat{B}\rangle}{\hbar^2V^2}\sum_{\mathbf{q}}g^2\Phi^2(\varkappa,\mathbf{q})\delta(\Omega(\varkappa,\mathbf{q}))\approx\frac{2\pi g^2}{\hbar^2}b\tilde{\delta}(\Omega(\varkappa)),$$

$$b=\frac{\langle\hat{B}\rangle}{V^2},$$

$$\Omega(\varkappa)=2\omega(\mathbf{k}_0)-\omega(\mathbf{k}_0+\varkappa)-\omega(\mathbf{k}_0-\varkappa),$$

$$D_1(\varkappa)=\frac{2\pi b}{\hbar^2}\sum_{\mathbf{q}}g^2\Phi^2(\varkappa,\mathbf{q})\delta(\Omega(\varkappa,\mathbf{q}))\bar{n}_{\mathbf{k}_0-\mathbf{q}}\approx D_0(\varkappa)\bar{n}_{\mathbf{k}_0-\varkappa},$$

$$D_1'(\varkappa)=\frac{4\pi(2\langle\hat{N}_{\mathbf{k}_0}\rangle+1)}{\hbar^2V^2}\sum_{\mathbf{q}}g^2\Phi^2(\varkappa,\mathbf{q})\delta(\Omega(\varkappa,\mathbf{q}))\bar{n}_{\mathbf{k}_0-\mathbf{q}}$$

$$\approx\frac{4\pi g^2}{\hbar^2}\left(\frac{2\langle\hat{N}_{\mathbf{k}_0}\rangle}{V^2}+\frac{1}{V^2}\right)\tilde{\delta}(\Omega(\varkappa))\bar{n}_{\mathbf{k}_0-\varkappa},$$

$$G(\varkappa)=G_1(\varkappa)+G_2(\varkappa)=\frac{8\pi\langle\hat{N}_{\mathbf{k}_0}\rangle}{\hbar^2V^2}\sum_{\mathbf{pq}}g^2\Phi^2(\mathbf{q},-\varkappa)$$

$$\times\,\delta(\Theta(\mathbf{p},\mathbf{q},-\varkappa))\,\bar{n}_{\mathbf{k}_0+\mathbf{p}}(1+\bar{n}_{\mathbf{k}_0+\mathbf{p}+\mathbf{q}})+\frac{4\pi(1+\langle\hat{N}_{\mathbf{k}_0}\rangle)}{\hbar^2V^2}$$

$$\times\sum_{\mathbf{q}\sigma}g^2\Phi^2(\mathbf{q},\sigma)\,\delta(\Theta(\varkappa,\mathbf{q},\sigma))\,\bar{n}_{\mathbf{k}_0+\mathbf{q}+\varkappa}\bar{n}_{\mathbf{k}_0-\sigma}.$$

(18)

The smoothing function $\tilde{\delta}(\Omega(\varkappa))$ approximates the result of integrating the product of the singular function $\delta[\Omega(\mathbf{q})]$ and a weight function $\Phi^2(\varkappa,\mathbf{q})$. We assume that $\tilde{\delta}[\Omega(\varkappa)]$ is Lorentzian in form. The quantity $4\hat{N}_0+2$ is the difference between the operators $\hat{C}$ and $\hat{B}$. The total attenuation $\mathscr{T}(\varkappa)$ consists of three terms caused by the heat bath, two-particle losses, and one-particle losses of polaritons from the condensed $\mathbf{k}_0$-mode respectively:

$$\mathscr{T}(\varkappa)=\gamma(\mathbf{k}_0+\varkappa)+\Gamma(\varkappa)+D_1'(\varkappa). \tag{19}$$

The function $\Gamma(\varkappa)$ has the form

$$\Gamma(\varkappa)=\Gamma_0(\varkappa)+\Gamma_1(\varkappa)+\Gamma_2(\varkappa)$$

$$=\frac{4\pi\langle\hat{N}_{\mathbf{k}_0}\rangle}{\hbar^2V^2}\sum_{\mathbf{q}\sigma}g^2\Phi^2(\mathbf{q},\sigma)\,\delta(\Theta(\varkappa,\mathbf{q},\sigma))$$

$$\times(1+\bar{n}_{\mathbf{k}_0+\mathbf{q}+\varkappa}+\bar{n}_{\mathbf{k}_0-\sigma})+\frac{8\pi\langle\hat{N}_{\mathbf{k}_0}\rangle}{\hbar^2V^2}$$

$$\times\sum_{\mathbf{pq}}g^2\Phi^2(\mathbf{q},-\varkappa)\,\delta(\Theta(\mathbf{p},\mathbf{q},-\varkappa))\cdot$$

$$\times(\bar{n}_{\mathbf{k}_0+\mathbf{p}+\mathbf{q}}-\bar{n}_{\mathbf{k}_0+\mathbf{p}})+\frac{8\pi}{\hbar^2V^2}\sum_{\mathbf{pq}}g^2\Phi^2(\mathbf{q},-\varkappa)$$

$$\times\,\delta(\Theta(\mathbf{p},\mathbf{q},-\varkappa))\,\bar{n}_{\mathbf{k}_0+\mathbf{p}+\mathbf{q}}(1+\bar{n}_{\mathbf{k}_0+\mathbf{p}})$$

$$-\frac{4\pi}{\hbar^2V^2}\sum_{\mathbf{q}\sigma}g^2\Phi^2(\mathbf{q},\sigma)\,\delta(\Theta(\varkappa,\mathbf{q},\sigma))\,\bar{n}_{\mathbf{k}_0+\mathbf{q}+\varkappa}\bar{n}_{\mathbf{k}_0-\sigma}.$$

(20)

The functions $G(\varkappa)$ and $\Gamma(\varkappa)$ are supplied with subscripts indicating that the quantities under the integral sign are independent, depend linearly, or depend quadratically, on the average occupation number $\bar{n}_{\mathbf{k}_0+\mathbf{p}}$ of scattered quasiparticles, respectively. Equation (17) contains the terms $D_0(\varkappa)$ and $G(\varkappa)$ in the numerator, corresponding to spontaneous two-particle and one-particle losses of polaritons from the mode $\mathbf{k}_0$. The single-particle loss $G(\varkappa)$ involves the participation of a scattered quasiparticle in the initial state and the formation of two quasiparticles in the final state. The first term leads to a peak-like structure in the nonequilibrium $\mathbf{k}$-space distribution function, while the second leads to a continuous background. The difference between them is the same as the difference between the shapes of exciton absorption bands for direct-gap and indirect-gap semiconductors. For the case of strong size quantization, the quantity $D_0(\varkappa)$ may in fact be smaller than $G_1(\varkappa)$ on the upper portion of the lower polariton band. On the other hand, within a rather small range of $\varkappa$ ($\varkappa<k_0$), $G_1(\varkappa)$ can have a peak-like structure of the form $(8\pi g^2/\hbar^2)n_0n_1\cdot\delta[\Omega(\varkappa)]$. These cases require a special investigation. The denominator of Eq. (17) is a difference of two terms, each of which depends on $\mathscr{T}(\varkappa)$ and varies with increasing density of scattered polaritons. However, the first term is a product $\mathscr{T}(\varkappa)\mathscr{T}(-\varkappa)$, and therefore will change more rapidly than the sum $\mathscr{T}(\varkappa)+\mathscr{T}(-\varkappa)$.

For qualitative estimates let us choose the smoothing function $\tilde{\delta}(\Omega(\varkappa))$ in the form of a Lorentzian

$$\tilde{\delta}(\Omega(\varkappa))=\frac{1}{\pi}\frac{q(\varkappa)}{\Omega^2(\varkappa)+q^2(\varkappa)}. \tag{21}$$

The quantity $q(\varkappa)$ is determined by inhomogeneous broadening in this variant or homogeneous broadening $\gamma_{\text{ef}}(\varkappa)$ in Ref. 12. In practice it is necessary to choose the larger of the two. Let us replace the function $\tilde{\delta}(\Omega[\varkappa])$ by its maximum value $1/\pi q(\varkappa)$ in the denominator of Eq. (17), and retain terms of zeroth and first order in the density of scattered polaritons n_1 inclusively. We write

$$\mathscr{T}_0(\varkappa)=\gamma(\mathbf{k}_0+\varkappa)+\Gamma_0(\varkappa),\quad \mathscr{T}_1(\varkappa)=\Gamma_1(\varkappa)+D_1'(\varkappa)$$

and note that $\Gamma_1(\varkappa)\sim n_1$, $D_1(\varkappa)\sim\bar{n}_{\mathbf{k}_0-\varkappa}$. If we remove a common factor $4g^2/q(\varkappa)\hbar^2$, the remaining portion of the denominator has the form

$$n_c^2(\varkappa)\left(1+\frac{\mathscr{T}_1(\varkappa)}{\mathscr{T}_0(\varkappa)}+\frac{\mathscr{T}_1(-\varkappa)}{\mathscr{T}_0(-\varkappa)}\right)-b\left(1+\frac{\mathscr{T}_1(\varkappa)+\mathscr{T}_1(-\varkappa)}{\mathscr{T}_0(\varkappa)+\mathscr{T}_0(-\varkappa)}\right), \tag{22}$$

where $n_c^2(\varkappa)$ is the square of the critical concentration:

$$n_c^2(\varkappa)=\frac{\hbar^2}{2g^2}\frac{\mathscr{T}_0(\varkappa)\mathscr{T}_0(-\varkappa)q(\varkappa)}{\mathscr{T}_0(\varkappa)+\mathscr{T}_0(-\varkappa)}. \tag{23}$$

Equation (22) equals $n_c^2(\varkappa) - b$ if $\mathscr{T}_1(\pm\varkappa) = 0$. In this case it is possible for the nonequilibrium distribution function $\bar{n}_{k_0+\varkappa}$ to exhibit a threshold dependence on the value of b averaged over the state of the coherently excited mode. A dependence of this sort is analogous to the threshold dependence encountered in stimulated Brillouin scattering as described in Refs. 2 and 3.

In our case the increase in b is accompanied by simultaneous increases in the quantities n_0 and $n_c^2(\varkappa)$. Furthermore, for positive values of $\mathscr{T}_0(\pm\varkappa)$ and $\mathscr{T}_1(\pm\varkappa)$ the correction to $n_c^2(\varkappa)$ that is linear in n_1 is larger than the corresponding correction to the quantity b for arbitrary dependences on $\varkappa$. The pure shift of the threshold value with increasing n_1 equals

$$n_c^2(\varkappa)\left[\frac{\mathscr{T}_1(\varkappa)}{\mathscr{T}_0(\varkappa)}+\frac{\mathscr{T}_1(-\varkappa)}{\mathscr{T}_0(-\varkappa)}-\frac{\mathscr{T}_1(\varkappa)+\mathscr{T}_1(-\varkappa)}{\mathscr{T}_0(\varkappa)+\mathscr{T}_0(-\varkappa)}\right]>0. \tag{24}$$

Thus, the effective threshold value, which increases with increasing n_0, b, and n_1, is smeared out to the point of being inaccessible in practice. The system is continuous at the point $b = n_c^2(\varkappa)$ because the denominator is finite there and equals (24).

Even when these approximations are made, Eq. (17) is nonlinear with respect to the average occupation numbers $\bar{n}_{k_0+\varkappa}$, because the latter enters into $D_1'(\pm\varkappa)$ in the following way:

$$\frac{D_1'(\varkappa)+D_1'(-\varkappa)}{\mathscr{T}_0(\varkappa)+\mathscr{T}_0(-\varkappa)} = \frac{2\mathscr{T}_0(\varkappa)\mathscr{T}_0(-\varkappa)}{[\mathscr{T}_0(\varkappa)+\mathscr{T}_0(-\varkappa)]^2}\left[\frac{2n_0}{n_c(\varkappa)N}+\frac{1}{N^2}\right](\bar{n}_{k_0+\varkappa}+\bar{n}_{k_0-\varkappa}). \tag{25}$$

It is easy to see that this ratio is smaller than unity for $n_0 < n_c$, $N \geqslant 1$, $\bar{n}_{k_0+\varkappa} < 1$, i.e., when we impose the same limitations that we used to derive the FPE. Let us linearize Eq. (17) with respect to $\bar{n}_{k_0+\varkappa}$ as follows: we will neglect the expressions $D_1'(\pm\varkappa)$ in the denominator of (22), and omit various $\varkappa$-dependences, specifically setting $\mathscr{T}_0(\varkappa) = q(\varkappa) = \gamma_{ef}$. Furthermore, we will replace the portions of the functions $\Gamma_1(\varkappa)$ and $G(\varkappa)$ that are proportional to n_0 by expressions of the type $n_1 n_0$, and the rest of the function $\Gamma_1(\varkappa)$ by $\varepsilon n_c n_1/N$. Making these simplifications, we find that the peak part of the nonequilibrium distribution function for scattered quasiparticles that rises above the continuous background is the following:

$$\bar{n}_{k_0+\varkappa} = \frac{\pi}{2}\frac{(b+4n_0n_1)\gamma_{ef}\tilde{\delta}(\Omega(\varkappa))}{n_c^2+\sigma n_0 n_1+\varepsilon n_c n_1/N-b}, \tag{26}$$

where

$$n_c = \hbar\gamma_{ef}/2|g|, \tag{27}$$

while the unknown constants σ and ε are positive. The term $\varepsilon n_c n_1/N$ has an additional dependence on $1/N$, and in Ref. 12 it was omitted.

In order to determine the average values n_0 and b that enter into Eqs. (9) and (18), we must find a stationary solution to the FPE. For the special case $N = 1$ such a solution was found in Ref. 13. By generalizing this solution to arbitrary finite values of N we are led to the following expression for the stationary quasiprobability $P_{ss}(\xi,\eta)$:

$$P_{ss}(\xi,\eta) = \frac{1}{I(c,z,N)}\xi^{Nc-2}\eta^{Nc^*-2}\exp\left(\frac{Nz}{\xi}+\frac{Nz^*}{\eta}+2N\xi\eta\right),$$

$$c = \frac{K}{\chi}, \quad z = \frac{d\mathscr{E}_0}{\hbar\gamma}, \quad 1 \leqslant N < \infty. \tag{28}$$

The normalization constant entering into (28) has the form

$$I(c,z,N) = -4\pi^2(Nz)^{Nc}(Nz^*)^{Nc^*}\frac{{}_0F_2(Nc,Nc^*,2N|zN|^2)}{|Nz|^2\Gamma(Nc)\Gamma(Nc^*)}. \tag{29}$$

By using (28) and (29) we can find the average values $\langle a_{k_0}^{+p}a_{k_0}^{q}\rangle$ from the operators for the coherently excited mode. These quantities can be expressed in terms of the generalized hypergeometric series ${}_0F_2(a,b,x)$ and the gamma function $\Gamma(x)$ in the following way:

$$\langle a_{k_0}^{+p}a_{k_0}^{q}\rangle = N^{(p+q)/2}(Nz)^q(Nz^*)^p \times \frac{\Gamma(Nc)\Gamma(Nc^*)\,{}_0F_2(Nc+q,Nc^*+p,2N|zN|^2)}{\Gamma(Nc+q)\Gamma(Nc^*+p)\,{}_0F_2(Nc,Nc^*,2N|zN|^2)}. \tag{30}$$

The second-order correlation function $g^{(2)}(t)$ at the instant of time $t = 0$ equals

$$g^{(2)}(0) = \frac{\langle a_{k_0}^{+2}a_{k_0}^{2}\rangle}{\langle a_{k_0}^{+}a_{k_0}\rangle^2} = \frac{|Nc|^2\,{}_0F_2(Nc+2,Nc^*+2,2N|zN|^2)\,{}_0F_2(Nc,Nc^*,2N|zN|^2)}{|Nc+1|^2[{}_0F_2(Nc+1,Nc^*+1,2N|zN|^2)]^2}. \tag{31}$$

This function determines the required statistical properties of the coherently excited polaritons for various values of the dimensionless volume of the crystallite N. Expression (31) depends on the coefficients c and z. It follows from (11) and (28) that these in turn depend on the nonequilibrium distribution function $\bar{n}_{k_0+\varkappa}$. Thus, the problem is selfconsistent, and can in principle be solved using Eqs. (11), (12) and (17)–(30).

Further analysis is not possible without a number of simplifications and approximations. We have already mentioned some of these; the others are presented below:

$$\frac{l}{\hbar\gamma_{ef}} = \frac{g}{|g|}v, \quad \frac{(f_1-g_1)n_c}{\gamma_{ef}} = -\frac{p}{4}\Phi(u,v,w), \quad \frac{(f_2-g_2)n_c}{\gamma_{ef}} = \frac{q}{2}\Phi(u,v,w), \quad p_i-q_i \approx 2n_1(f_i-g_i). \tag{32}$$

Here we have used the dimensionless polariton density and introduced the notation

$$u = \frac{n_0}{n_c}, \quad v = \frac{n_1}{n_c}, \quad w = \frac{b}{n_c^2}, \quad \Phi(u,v,w) = 1+\frac{w+4uv}{2(1-w+4uv)}. \tag{33}$$

Within the framework of the approximations we have made we find the following regular dependence of v on u and w:

$$v=\frac{1}{2\sigma u}[(1-w-2qu)^2+2\sigma quw]^{1/2}-\frac{1}{2\sigma u}(1-w-2qu). \tag{34}$$

Likewise, the quantities u and w themselves depend on (v, w) and (u, v) respectively. The constants p, q, σ are unknown parameters of the theory and can be estimated using a series expansion of the function $\omega(\mathbf{k}_0+\varkappa)$ for $\varkappa<k_0$ or some other simplification of the dispersion law for the exciton-like and photon-like portions of the polariton curve. For simplicity, ε will be set equal to zero.

The coefficients $|c|^2$, $c+c^*$, and $|z|^2$, which determine the values of the correlation function $g^{(2)}(0)$, have the form

$$|c|^2=\frac{L(u,v,w)}{M(u,v,w)},\quad \frac{c+c^*}{2}=\frac{K(u,v,w)}{M(u,v,w)},\quad |z|^2=\frac{Y}{M(u,v,w)},\quad Y=\frac{|d\mathscr{E}_0|^2}{(\hbar\gamma_{ef})^2}, \tag{35}$$

where K, L, M are the following functions:

$$M(u,v,w)=\left[\frac{1}{2}\frac{g}{|g|}\Phi(0,0)-\frac{p}{8}\Phi(u,v,w)\right]^2+\left[\frac{q}{4}\Phi(u,v,w)\right]^2,$$

$$L(u,v,w)=\left[\frac{\Delta\omega}{\gamma_{ef}}+\frac{g\Phi(0,0)}{|g|}v-\frac{pv}{2}\Phi(u,v,w)\right]^2+\left[\frac{1}{2}+qv\Phi(u,v,w)\right]^2, \tag{36}$$

$$K(u,v,w)=\left[\frac{\Delta\omega}{\gamma_{ef}}+\frac{g\Phi(0,0)}{g}v-\frac{pv}{2}\Phi(u,v,w)\right]\left[\frac{1}{2}\frac{g}{|g|}\Phi(0,0)-\frac{p}{8}\Phi(u,v,w)\right]+\frac{q}{4}\Phi(u,v,w)\left[\frac{1}{2}+qv\Phi(u,v,w)\right].$$

To summarize, we can write down two transcendental equations for u and w using the general expression (30). The third equation for v is given by (34). For a given N these three equations determine the three quantities u, v, w we require as functions of the parameter Y of the external pump, which is connected with the energy flux of the external laser radiation S_L by relation (15). There are still three unknown parameters p, q, σ in the theory whose values can be estimated from various models.

4. STATISTICAL PROPERTIES OF COHERENTLY EXCITED POLARITONS

Analytic expressions for the functions ${}_0F_2(a,b,x)$ can be used in two limiting cases: for $x<ab$ and $x\to\infty$ (Ref. 22). The case $x<ab$, where

$${}_0F_2(a,b,x)=\sum_{n=0}^{\infty}\frac{x^n}{n!}\frac{\Gamma(a)\Gamma(b)}{\Gamma(a+n)\Gamma(b+n)}\approx 1+\frac{x}{ab}, \tag{37}$$

is the most interesting. Since $x=2N|zN|^2$ and one of the values ab equals $|c|^2$, we can find a bound on the product NY of the two dimensionless quantities—the volume of the crystallite and the density of power from the coherent pump Y—in the form

$$NY<{}^1/_2 L(u, v, w). \tag{38}$$

As $v\to 0$ the expression for $L(u, v, w)$ takes the form $(\Delta\omega/\gamma_{ef})^2+\frac{1}{4}$. Based on this bound we can obtain an analytic result within a rather small region of Y, for which the maximum value Y_{max} is inversely proportional to N.

In the region of values of NY that satisfy Eq. (38), the quantities u, w, and $g^{(2)}(0)$ are as follows:

$$u=\frac{|z|^2[|Nc|^2+1+N(c+c^*)+2N^3|z|^2]}{[|Nc|^2+1+N(c+c^*)][|c|^2+2N|z|^2]},$$

$$w=\frac{[|Nc|^2+4+2N(c+c^*)+2N^3|z|^2]}{[|Nc|^2+1+N(c+c^*)][|Nc|^2+4+2N(c+c^*)]}\times\frac{N^2|z|^4}{[|c|^2+2N|z|^2]} \tag{39}$$

$$g^{(2)}(0)=\frac{w}{u^2}.$$

Analysis shows that for $(c+c^*)N+1>0$ the correlation function $g^{(2)}(0)<1$ and the polaritons of the mode $\mathbf{k}_0$ exhibit the property of anti-bunching. When the further inequality $(c+c^*)N+3<0$ is fulfilled we have $g^{(2)}(0)>1$, which is a sign that the coherently excited polaritons are bunched when condition (38) holds.[23] Using (30), it is easy to calculate the mean-square deviation $\langle(\Delta\hat{X}_i)^2\rangle=\langle\hat{X}_i^2\rangle-\langle\hat{X}_i\rangle^2$ of the quadrature phase operator $\hat{X}_i$ in the form

$$\hat{X}_1=a_{\mathbf{k}_0}+a_{\mathbf{k}_0}^+,\quad \hat{X}_2=i(a_{\mathbf{k}_0}-a_{\mathbf{k}_0}^+). \tag{40}$$

The mean-square deviation is subject to the uncertainty relation $\langle(\Delta\hat{X}_1)^2\rangle\langle(\Delta\hat{X}_2)^2\rangle\geqslant 1$.

In single-photon quantum optics the minimum noise moment satisfies $\langle(\Delta\hat{X}_i)^2\rangle=1$. This moment, which is referred to as the zero point, is achieved in a coherent state. In two-photon quantum optics compressed states are possible, in which the noise moment in one of the quadrature phases is smaller than the zero-point noise, e.g., $\langle(\Delta\hat{X}_1)^2\rangle<1$, because of the increased moment in the conjugate operator $\langle(\Delta\hat{X}_2)^2\rangle>1$ (Ref. 24). Analogous states of coherently excited polaritons are possible because the two-particle loss from the $\mathbf{k}_0$ mode behaves like to a two-photon transition in the case of degenerate modes. Analysis shows that compressed states occur in the presence of both bunching and anti-bunching when certain relations between the phases φ and ψ of the complex quantities c and z are satisfied. These novel statistical properties of polaritons in crystallites have not been discussed previously in the literature. They are sensitive to increases in N and Y, and disappear as $NY\to\infty$ when the condition (38) is violated.

Numerical estimates based on Eq. (39) were made for three values $N=1$, 10, 50, and five values $\Delta\omega/\gamma_{ef}=0$, ± 3, ± 5, for $p, q=10^{-1}$, $\sigma=10^{-5}$–10^{-4}, and $\varepsilon=0$. We chose the constant $g=10^{-32}$ erg·cm^3, $\gamma_{ef}=10^{11}$ sec^{-1}, and $n_c=5\cdot 10^{15}$ cm^{-3}, which are close to the crystal CdS. These

particular values of N and n_c correspond to crystallite dimensions $d \approx 600$–2200 Å, which fall within the bounds presented earlier. The calculation shows that for $N = 1$ the effects of bunching and antibunching are such that $g^{(2)}(0)$ deviates from 1 by 20–50%. For $N = 10$ the difference comes to 2–3%, while for $N = 50$ it is only 0.3–0.6% in all. For $N \geqslant 100$ the effect under discussion is vanishingly small and polaritons of the $\mathbf{k}_0$ mode are found to be in a purely coherent state. At the boundaries of the crystallite the polaritons are converted into light, which is subject to analysis by the method of photon counting. The extent of its bunching and anti-bunching can be evaluated following Ref. 25. The degree of compression of the fluctuations, i.e., the departure of $\langle(\Delta\hat{X}_i)^2\rangle$ from 1 for $N = 1$ comes to 10% under conditions of anti-bunching and 26% under bunching conditions. For $N = 10$ the degree of compression decreases by an order of magnitude and comes to 2.4–2.6%.

The connection between these effects and detuning from resonance is the following. Let us assume that the frequency of the laser radiation ω_L is larger than the bare frequency of the polariton mode $\omega(\mathbf{k}_0)$ such that $\Delta\omega/\gamma_{ef} = -3, -5$, and that its intensity is small and bounded by condition (38). Under these conditions the coherent pumping is not capable of creating a purely coherent polariton mode in a crystallite of finite volume. The frequency excess of the photons above the frequency of the polaritons $\omega(\mathbf{k}_0)$ causes a partial randomization of the latter, whose statistical properties are now reminiscent of the properties of thermal radiation and a tendency towards bunching is observed. When the excitation frequency of the radiation does not exceed the bare frequency of the mode $\omega(\mathbf{k}_0)$, there arises a deficit of photon energy below that required to excite the $\mathbf{k}_0$ polaritons. Therefore, when $\Delta\omega/\gamma_{ef} = 0, 3, 5$ the quantum effects of the forced oscillations are more evident. As we noted previously, this rather low-intensity excitation radiation cannot excite a purely coherent polariton mode $\mathbf{k}_0$; however, in this case the polaritons are observed to exhibit the phenomenon of anti-bunching, which is a characteristic of quantum states of Fock type.[23] The different behavior of coherently excited polaritons as a function of the sign of the detuning from resonance $\Delta\omega$ for rather small pumping intensities correlates with the properties of an exciton absorption band when the fundamental mechanism for energy dissipation is exciton-exciton interactions.[26]

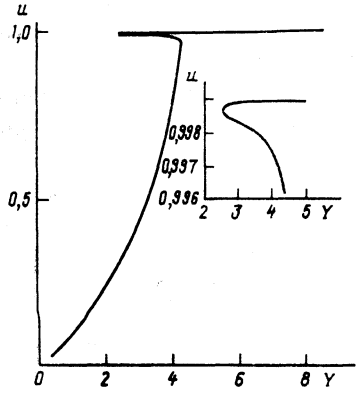

FIG. 2. Density of coherent polaritons u as a function of pump intensity Y for a detuning from resonance of $\Delta\omega/\gamma_{ef} = -3$.

Let us now estimate the laser radiation energy S_L required to observe these phenomena. According to (38), Y varies in the interval $(0, Y_{max})$, where $Y_{max} \approx (1/2N)\left[(\Delta\omega/\gamma_{ef})^2 \tfrac{1}{4}\right]$ as $v \to 0$. For all the cases that we investigated $Y \leqslant 10$ holds. We chose the following parameters: for the photons, $\omega_L = 3 \cdot 10^{15}$ sec^{-1}; for the resonator, $L = 1\,\mu$m and $R = 0.5$; for the excitons, polaritons, and exciton-photon interactions, $\omega_{ex} = ck_0$, $\omega_{ex}(\mathbf{k}_0) - \omega(\mathbf{k}_0) = \frac{1}{2}|\varphi_{k_0}/\hbar|$, $\gamma_{ef} = 10^{11}$ sec^{-1}, and $n_c = 5 \cdot 10^{15}$ cm^{-3}. According to Eqs. (15) and (16) we find that $\Omega_{res}^2/\gamma_{ef} = 10^6$, $S_c = 10^{15}$ erg/cm^2·sec and $\zeta = 0.2$. The maximum laser radiation power needed to observe these effects came to 5 kW/cm^2 for resonance excitation of crystallites of CdS type.

Let us briefly discuss the case of a bulk crystal, i.e., $V \to \infty$. The asymptotic expressions of Ref. 22 do not apply to the case of a bulk crystal for low coherent pump intensities. A more general solution to the FPE as $N \to \infty$ in steady state is

$$P_{ss}(\xi, \eta) \approx \text{const} \cdot \delta(\xi - \xi_1)\delta(\eta - \eta_1), \tag{41}$$

where ξ_1 and η_1 satisfy the equations

$$\begin{gathered}\eta_1 = \xi_1^*, \quad |\xi|^2 = u, \quad w = u^2, \quad g^{(2)}(0) = 1, \\ c\xi_1 + 2|\xi_1|^2\xi_1 = z, \\ 4u^3 + 2(c + c^*)u^2 + |c|^2 u = |z|^2.\end{gathered} \tag{42}$$

Using the expressions for $c + c^*$ and $|c|^2$ from Eqs. (35) and (36), along with Eq. (34) for v in which we replace w by u^2, we find the function $u(Y)$ shown in Fig. 2. The coherent macroscopic state possesses the property of optical bistability, with a hysteresis loop that is not very pronounced; the curve $u(Y)$ increases slowly in the quasithreshold region $u = 1$.

In conclusion, we the authors are deeply grateful to L. V. Keldysh and the participation of the Moscow State University Seminar, to V. F. Eselin, I. B. Levinson, G. E. Pikus, and E. I. Rashba, as well as colleagues in the Institute of Applied Physics of the Moldavian SSR Academy of Sciences, for discussions of the work and for their comments.

[1] A. L. Ivanov and L. V. Keldysh, Zh. Eksp. Teor. Fiz. **84**, 404 (1983) [Sov. Phys. JETP **57**, 234 (1983)].
[2] L. V. Keldysh and S. G. Tikhodeev, Zh. Eksp. Teor. Fiz. **90**, 1852 (1986) [Sov. Phys. JETP **63**, 1086 (1986)].
[3] L. V. Keldysh and S. G. Tikhodeev, Zh. Eksp. Teor. Fiz. **91**, 78 (1986) [Sov. Phys. JETP **64**, 45 (1986)].
[4] N. N. Bogolyubov, *Selected Works in Three Volumes*, Vol. 2, p. 351 [in Russian], Naukova Dumka, Kiev, 1970.
[5] S. A. Moskalenko, M. F. Miglei, M. I. Shmiglyuk *et al.*, Zh. Eksp. Teor. Fiz. **64**, 1786 (1973) [Sov. Phys. JETP **37**, 902 (1973)].
[6] M. I. Shmiglyuk and V. N. Pitei, *Coherent Polaritons in Semiconductors* [in Russian], ShTIINTsA, Kishinev, 1989.
[7] V. F. Eselin and Yu. V. Kopaev, Zh. Eksp. Teor. Fiz. **72**, 334 (1977) [Sov. Phys. JETP **45**, 177 (1977)].
[8] V. F. Eselin and Yu. V. Kopaev, Zh. Eksp. Teor. Fiz. **63**, 1447 (1972) [Sov. Phys. JETP **36**, 7677 (1972)].
[9] M. I. Strashnikova, V. Y. Reznichenko, and V. V. Cherniy, Ukr. Fiz. Zh. **32**, 187 (1985).
[10] J. Kusano, Y. Segawa, M. Mihara *et al.*, Solid State Commun. **72**, 215 (1989).
[11] A. D'Andrea and R. Del Sole, Phys. Rev. B **41**, 1413 (1990).
[12] V. R. Misko, S. A. Moskalenko, A. H. Rotaru, and Yu. M. Shvera, Phys. Status Solidi (b) **159**, 477 (1990).
[13] P. D. Drummond and D. F. Walls, J. Phys. A **13**, 725 (1980).
[14] R. Bonifacio and L. A. Lugiato, Lett. Nuovo Cimento **21**, 517 (1987).
[15] V. A. Zalozh, S. A. Moskalenko, and A. H. Rotaru, Zh. Eksp. Teor. Fiz.

95, 601 (1989) [Sov. Phys. JETP **68**, 338 (1989)].
[16] F. Arecchi, M. Scalli, H. Haken, and W. Weidlich, *Quantum Fluctuations in Laser Radiation*, p. 143 [in Russian], Mir, Moscow, 1974.
[17] S. A. Moskalenko, A. H. Rotaru, and Yu. M. Shvera, *Cooperative and Nonequilibrium Processes in High-Density Excitonic Systems*, p. 169 [in Russian], ShTIINTsA, Kishinev, 1989.
[18] S. A. Moskalenko, A. H. Rotaru, and Yu. M. Shvera, *Quantum Statistical Properties of High Densities of Excitons*, p. 3 [in Russian]. ShTIINTsA, Kishinev, 1988.
[19] S. A. Moskalenko, A. H. Rotaru, and Yu. M. Shvera, preprint, Inst. Appl. Phys. Moldavian SSR, ShTIINTsA, Kishinev, 1987.
[20] G. Klauder and E. Sudarshan, *Fundamentals of Quantum Optics*, Benjamin, New York, 1968 (Mir, Moscow, 1970).
[21] P. D. Drummond and C. V. Gardiner, J. Phys. A **13**, 2353 (1980).
[22] O. I. Marichev, *Handbook of Integral Transforms of Higher Transcendental Functions*, Halsted, New York (1983).
[23] H. Faul, Rev. Mod. Phys. **54**, 1061 (1982).
[24] C. M. Caves and B. L. Schumaker, Phys. Rev. A **31**, 3068 (1985).
[25] S. Friberg and L. Mandel, Opt. Commun. **46**, 141 (1983).
[26] A. V. Lelyakov and S. A. Moskalenko, Fiz. Tverd. Tela (Leningrad) **11**, 3260 (1969) [Sov. Phys. Solid State **11**, 2642 (1969)].

Translated by Frank J. Crowne