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Quantum analysis of luminescence of an exciton in a meso-cavity

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Abstract: Interaction of cavity modes with an exciton in a meso-cavity (the structure supporting several cavity modes separated by an energy interval comparable to Rabi-splitting of an exciton and cavity modes) has been analyzed using a quantum-mechanical approach. Simultaneous interaction of an exciton and several cavity modes results in few novel effects such as ladder-like increase of the exciton population in the system, quantum beating and non-monotonic dependence of the ground polariton state in the system on the pumping.

1. Introduction

Experimental observation in the reflectivity of a strong coupling between quantum well (QW) exciton and photon modes in semiconductor microcavities, seen as so-called Rabi splitting [1], resulted in a surge of studies of quantum microcavities of various design, where it was also important to understand how the emission can modify when the quantum wells and the optical cavity are in resonance [2]. Extensive research in this direction has formed a new paradigm in solid state physics: quantum electrodynamics on chip [3–8]. Many designs of microcavities (planar, pillar, spherical core-shell, plasmonic) [9–14] have been developed for localization of light and various material supporting exciton (semiconductor quantum wells, wires and dots, organic materials) [15–18] have been used for the observation of the strong coupling exciton and photon.

The planar structures are well studied theoretically and experimentally for high Q-factor cavities based on GaAs/AlGaAs semiconductors that are mature in terms of technology [4,8]. Additional confinement of exciton in such materials, for example in one-dimensional box, can led to stimulated emission even at relatively low optical pumping [16]. Also, microcavities containing organic semiconductors have been found suitable for achieving the strong exciton-photon coupling regime because of large oscillator strengths and thus large Rabi splitting energies despite a rather poor Q-factors [18].

In microcavities the electromagnetic fields are localized in the volume, comparable to the wavelength of light and in this case one photonic mode experience coupling with one or several excitons [6–8,16,19,20,21].

Exploration of new design of microstructures allows to predict and observe a rich variety of different interesting phenomena; for example, systems with coupled microcavities containing QWs demonstrate the optically induced splitting of bright exciton states [22]. Other structures with QWs incorporated into a Bragg mirror stack, where several photonic modes experience strong coupling to excitons, addresses a concept of Bragg polariton that can exhibit nonlinear

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behavior offering opportunities for observation of stimulated scattering, amplification, and lasing [23,24]. The QW width should be less than the Bohr radius of exciton, which can be relatively easy realized for GaAs and its alloys with Al and In and, thus, high-quality structures with microcavities can be coherently grown.

GaN, AlGaN and ZnO are wide-band gap semiconductors benefiting properties such as bright emission in UV region, large oscillator strength of excitons, chemical and thermal stability. However, employing these materials for growth of high Q-factor cavities with small volume in terms of light wavelength is yet to be develop. From this point of view, fundamental studies of exciton-photon interaction in larger resonators will pave the way for technological progress in UV nanophotonics.

In the meso-cavities with a size an order of magnitude larger than the wavelength, several photonic modes can interact with an exciton. In a work [25], GaN structures were experimentally investigated, several dominant emission lines were found in the low-temperature cathodoluminescence spectrum, and numerical analysis showed that in such structures the interaction of highly localized modes with an exciton is possible. Optical meso-cavities can be thought as a photonic analogue of electronic mesoscopic systems, where coherence length of electron wavefunction is comparable to the size of the systems [26–31]. Recently, the coupling of excitons and photons has been considered in two-dimensional meso-cavities supporting several photonic modes with separation between them much less compared to the Rabi splitting and a number of new interesting effects has been indicated [32]. It was theoretically shown that in such structures, the Q-factors of the resonator modes, as well as the strength of interaction with excitons, can differ by several orders of magnitude. The analysis of polariton modes in meso-cavity has demonstrated that the interaction of several photonic modes with an exciton can occur in the strong coupling regime, despite the high density of resonator modes due to the large sizes of the systems.

In this paper, we consider the general case of a meso-cavity, regardless of its shape or dimension, under the following conditions: a system of several modes interacts with one exciton, moreover the strengths of interaction of different modes with an exciton are comparable, and the interval between cavity modes is comparable to Rabi splitting. Such a system has not previously been studied, and we show that a rather complex interaction pattern leads to a number of interesting fundamental effects. Since the main tool of experimental studies of various photonic structures is optical spectroscopy, our paper is aimed at theoretical studies of the luminescence spectra in meso-cavities experiencing strong coupling with exciton.

2. Formalism

We consider a model system consisting of one exciton mode with energy $\hbar\omega_0$ and several photonic modes with energies $\hbar\omega_k$. In order not to complicate the system and focus on the effect associated with several photonic modes with an exciton mode, in this work we neglected the interaction with electrons and phonons of the system. Taking these effects into account, of course, would give spectra close to real ones, but such an approach would not allow a complete description of the discovered effects. The Hamiltonian describing the exciton-exciton interaction and the interaction of the exciton in the meso-cavity with the optical modes of the meso-cavity reads [33]:

$$\hat{H} = \hbar\omega_0\hat{x}^+\hat{x} + \sum\nolimits_k \hbar\omega_k\hat{a}_k^+\hat{a}_k + \sum\nolimits_k \hbar g_k(\hat{a}_k^+\hat{x} + \hat{a}_k\hat{x}^+) + \hbar U\hat{x}^+\hat{x}^+\hat{x}\hat{x}, \tag{1}$$

where $\hat{x}^+$, $\hat{a}_k^+(\hat{x}, \hat{a}_k)$ are the operators of creation (annihilation) of exciton and photons, respectively, obeying the commutation rule for bosons $[\hat{x}, \hat{x}^+] = 1$ and $[\hat{a}_k, \hat{a}_k^+] = 1$ [34]. The exciton is treated as a bosonic mode with weak nonlinearity. This consideration is applicable in the case of quantum wells or large quantum dots at a low excitation density. The term $\hbar g_k(\hat{a}_k^+\hat{x} + \hat{a}_k\hat{x}^+)$ in the Hamiltonian (Eq. (1)) describes the interaction of the exciton and photon modes with the interaction energy $\hbar g_k$, while the term $\hbar U\hat{x}^+\hat{x}^+\hat{x}\hat{x}$ describes the exciton-exciton interaction with the energy $\hbar U$ ($U \ll g_k$) for weakly interacting excitons [33].

In presence of dissipation, the system is upgraded to a Liouvillian description, with a quantum dissipative master equation for the density matrix. Hence, the full system state is described by the density matrix $\hat{\rho}$ and its evolution is determined by the equation [35]:

$$\partial_t \hat{\rho} = \hat{\mathcal{L}} \hat{\rho}, \tag{2}$$

where $\hat{\mathcal{L}}$ is Liouvillian with the Lindblad terms, taking into account the damping of excitons γ_0 and the damping of photons γ_k [36]:

$$\hat{\mathcal{L}}\hat{\rho} = \frac{i}{\hbar}[\hat{\rho}, \hat{H}] + \frac{\gamma_0}{2}(2\hat{x}\hat{\rho}\hat{x}^+ - \hat{x}^+\hat{x}\hat{\rho} - \hat{\rho}\hat{x}^+\hat{x}) + \sum\nolimits_k \frac{\gamma_k}{2}(2\hat{a}_k\hat{\rho}\hat{a}_k^+ - \hat{a}_k^+\hat{a}_k\hat{\rho} - \hat{\rho}\hat{a}_k^+\hat{a}_k). \tag{3}$$

To find the average values of the number of particles in the exciton and photon modes, as well as in their mixed states, we have to solve a system of nonlinear differential equations, which is obtained from the following relation $\partial_t\langle\hat{O}\rangle = Tr(\hat{O}\partial_t\hat{\rho}) = Tr(\hat{O}\hat{\mathcal{L}}\hat{\rho})$, where $\hat{O}$ is an arbitrary operator (for example operator of number of particles). The system of equations for our case is as follows:

$$\partial_t n_{xx} = \sum\nolimits_k ig_k(n_{kx} - n_{xk}) - \gamma_0 n_{xx} + I(n_{xx} + 1), \tag{4a}$$

$$\partial_t n_{ij} = i(\omega_i - \omega_j)n_{ij} - ig_j n_{ix} + ig_i n_{xj} - \frac{1}{2}(\gamma_i + \gamma_j)n_{ij}, \tag{4b}$$

$$\partial_t n_{xi} = i(\omega_0 - \omega_i)n_{xi} + \sum\nolimits_k ig_k n_{ki} - ig_i n_{xx} - \frac{1}{2}(\gamma_0 + \gamma_i)n_{xi} + 2iUn_{xx}n_{xi}, \tag{4c}$$

where $n_{xx} = \langle\hat{x}^+\hat{x}\rangle$ is the mean value of the number of excitons, $n_{xi} = \langle\hat{x}^+\hat{a}_i\rangle = n^*_{ix}$ and $n_{ij} = \langle\hat{a}_i^+\hat{a}_j\rangle$are the mixed states, which correspond to the coherence between the system modes, and the term $I(n_{xx} + 1)$ describs the pumping of excitons into the system [37–39]. Note that the number of particles in pure photonic state is given by $n_{ii} = \langle\hat{a}_i^+\hat{a}_i\rangle$. If $U = 0$, then the system of differential equations is closed, but when exciton-exciton interaction appears, the system ceases to be closed, since the $\langle\hat{x}^+\hat{x}\hat{x}^+\hat{a}_i\rangle$ term appears. However, when considering a rather weak interaction between excitons, we can make the following assumption$\hbar U\hat{x}^+\hat{x}^+\hat{x}\hat{x} \approx \hbar U\hat{x}^+\hat{x}\langle\hat{x}^+\hat{x}\rangle \approx \hbar Un_{xx}\hat{x}^+\hat{x}$ [40] (see Supplement 1).

Excitons and cavity modes form mixed states of photons and excitons (polaritons), and it is convenient to describe the system in the polariton basis (see Supplement 1):

$$p_i = \langle\hat{p}_i^+\hat{p}_i\rangle = c_{0i}^2 n_{xx} + \sum\nolimits_k c_{0i}c_{ki}(n_{xk} + n_{kx}) + \sum\nolimits_{k,k'} c_{ki}c_{k'i}n_{kk'} \tag{5}$$

where p_i is the average number of particles in polariton states with index "i" and c_{ij} are the elements of the transition matrix to the polariton basis.

Figure 1(a) shows a schematic representation of the meso-cavity. All quantities in this work are in exciton energy units ω_0. The length of the cavity is about 10 wavelengths of exciton radiation. In Fig. 1(b) shows the eigenmodes of a typical meso-cavity (21 photon modes with an energy interval between the modes of $14.3 \cdot 10^{-3}\omega_0$ and dumping of $1.88 \cdot 10^{-3}\omega_0$) and eigenmodes of a system of coupled exciton-photon states (polaritons). The energies of polaritons are found using the diagonalization of Hamiltonian (1) (see Supplement 1), the dumping for polaritons are found using the following relation $\gamma_{p_i} = \gamma_0 c_{0i}^2 + \sum\limits_k \gamma_k c_{ki}^2$. The coupling constant is found from the known relation $g_k = \sqrt{\omega_k/2\epsilon_0\epsilon\hbar V}d_{eg}$ [8], where d_{eg} is a dipole moment, V is a volume of quantization, $\epsilon_0\epsilon$ is the dielectric constant. In our case, we chose the unknown quantities so that the coupling constant was close in magnitude to the coupling constant in GaN $g_k \approx 50meV$ [41] if we take the exciton energy $\omega_0 = 3.5{\cdot}\mathrm{eV}$ close to the exciton energy in GaN.

To calculate the emission spectra of meso-cavities, we used the formalism described in [19]. Since we are considering only weakly interacting excitons, the exciton-exciton interaction will

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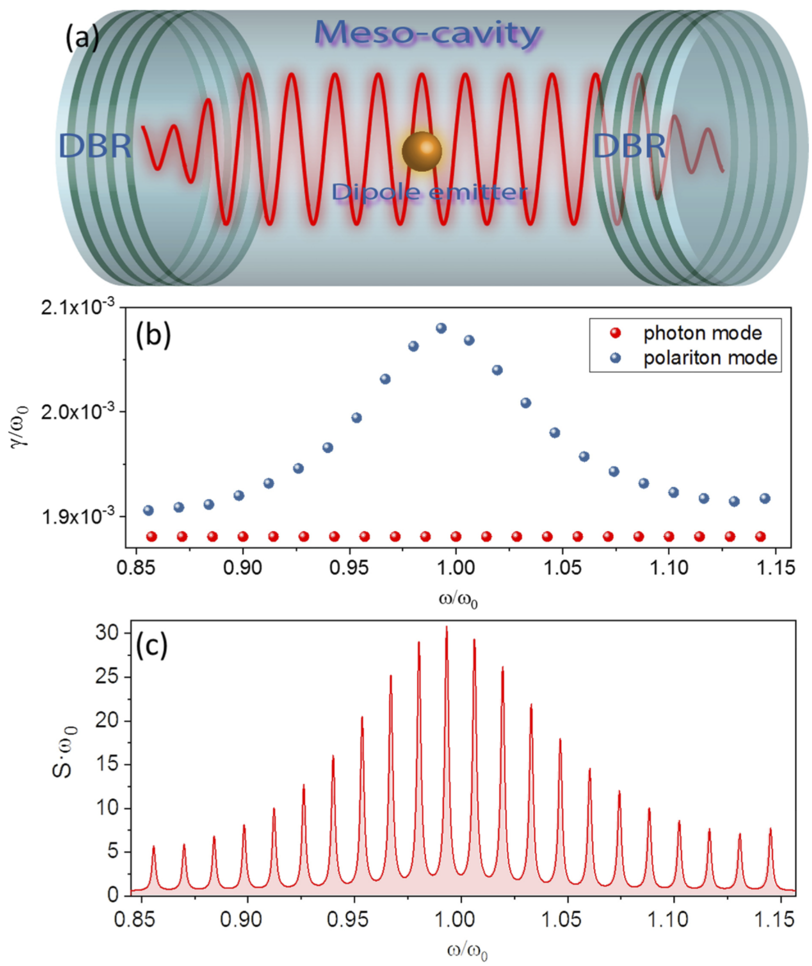

Fig. 1. (a) Schematic representation of a meso-cavity with an emitter. The size of the cavity between two distributed Bragg reflectors (DBR) is about 10 radiation wavelengths in the material. (b) Energy and damping cavity modes (red circles) and polariton modes (blue circles). (c) Emission spectra of meso-cavity. Damping of exciton is $\gamma_0 = 2.26 \cdot 10^{-3}\omega_0$, exciton-photon coupling constant $g_k \approx 14.3 \cdot 10^{-3}\omega_0$, number of photon modes is $N = 21$.

have little effect on the emission spectrum [42], therefore, for the calculation of the emission spectra, we neglect exciton-exciton interaction. The mean number of photons in the system with frequency ω for cavity mode i is found using the following:

$$s_i(\omega) = \langle \hat{a}_i^+(\omega)\hat{a}_i(\omega)\rangle. \tag{6}$$

The emission spectrum for the system (the density of probability that a photon emitted by the system has frequency ω), taking into account normalization, will look as follows:

$$S(\omega) = \frac{\sum_i s_i(\omega)}{\sum_i \int_0^\infty n_{ii} dt}. \tag{7}$$

At the same time, we can obtain the function $s_i(\omega)$ through the first-order correlation function [19]:

$$s_i(\omega) = \frac{1}{\pi}\mathcal{R} \int_0^\infty \int_0^\infty G_i^{(1)}(t,\tau)e^{i\omega\tau} d\tau dt, \tag{8}$$

where

$$G_i^{(1)}(t,\tau) = \langle \hat{a}_i^+(t)\hat{a}_i(t+\tau)\rangle. \tag{9}$$

To find the correlation function, we must use the following relation:

$$V_i(t,t+\tau) = e^{\boldsymbol{M}\tau} V_i(t,t), \tag{10}$$

where

$$V_i(t,t+\tau) = \begin{bmatrix} \langle a_i^+(t)x(t+\tau)\rangle \\ \langle a_i^+(t)a_1(t+\tau)\rangle \\ \vdots \\ \langle a_i^+(t)a_i(t+\tau)\rangle \\ \vdots \\ \langle a_i^+(t)a_j(t+\tau)\rangle \end{bmatrix} \tag{11}$$

and matrix M couples first order correlation functions at moment "t" and at moment "$t+\tau$". We can find the matrix M using the quantum regression theorem $tr(C_{\{\eta\}}\mathcal{L}(\rho\Omega)) = \sum_{\{\lambda\}} M_{\{\eta\lambda\}} tr(C_{\{\lambda\}}\rho\Omega)$. In our case $C_{\{\eta\}} = \hat{x}, \hat{a}_i$ and $\Omega = \hat{a}_i^+$. After these calculations, the matrix M is obtained as follows (for N optical modes):

$$\boldsymbol{M} = \begin{bmatrix} -i\omega_0 - \frac{\gamma_0}{2} & -ig_1 & -ig_2 & \cdots & -ig_N \\ -ig_1 & -i\omega_1 - \frac{\gamma_1}{2} & 0 & \cdots & 0 \\ -ig_2 & 0 & -i\omega_2 - \frac{\gamma_2}{2} & \cdots & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ -ig_N & 0 & 0 & \cdots & -i\omega_N - \frac{\gamma_N}{2} \end{bmatrix} \tag{12}$$

In Fig. 1(c) shows the luminescence spectrum of the system calculated by this method (see Supplement 1). The luminescence spectrum correlates with the distribution of polariton modes shown in Fig. 1(b). The peaks in the spectrum are located at the energies of the polariton modes, and the height of the peaks is proportional to the exciton contributions to the polariton states (Hopfield coefficients).

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3. Results and discussions

The resulting polaritonic spectrum is defined by the ratio of the interval between the cavity modes and the Rabi-splitting between exciton and cavity mode. Further, in the entire part of the results, we took $\hbar = 1$. When the separation between cavity modes Δ is less than the value of the coupling constant $\Delta < g_k$, the two modes on the edges of the polariton spectrum have maximal excitonic contribution (Hopfield coefficient), and consequently, could provide (depending on the population of polariton states) most pronounced corresponding emission lines, as shown in Figs. 2(a) and 2(d).

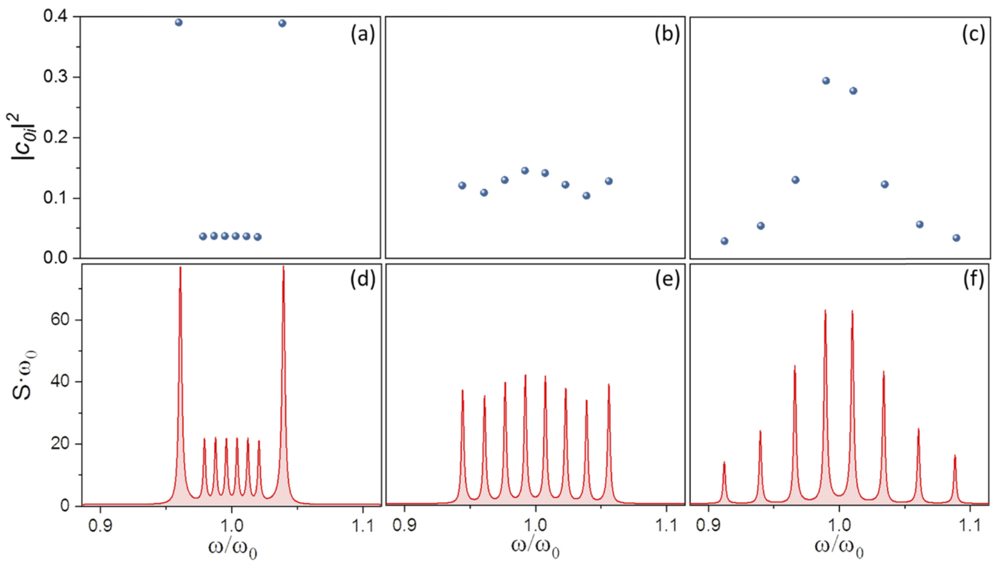

Fig. 2. (a-c) Hopfield coefficients illustrating exciton contribution to polariton modes and emission spectra of meso-cavity (d-f). The parameters used in calculation are the following the number of photon modes $N = 7$, damping of exciton is $\gamma_0 = 3.76 \cdot 10^{-3}\omega_0$, photons decay are $\gamma_i = 1.88 \cdot 10^{-3}\omega_0$, exciton photon coupling strength $g_k \approx 14.3 \cdot 10^{-3}\omega_0$, exciton-exciton interaction constant $U = 0$. The separation of cavity modes Δ are $8 \cdot 10^{-3}\omega_0$ (a, d); $17 \cdot 10^{-3}\omega_0$ (b, e); and $30 \cdot 10^{-3}\omega_0$ (c, f).

When Δ and g_k are comparable to each other, there is more or less a uniform exciton contribution to polaritonic modes and similar intensities of emission peaks corresponding to different polariton modes, see Figs. 2(b) and 2(e). In the case of a large interval between cavity modes $\Delta > g_k$, the distribution of the Hopfield coefficients has a bell-like envelope function centered near the frequency of exciton (Fig. 2(c)), and the emission spectrum of the cavity (Fig. 2(f)) mimics the distribution of Hopfield coefficients.

The interaction of exciton with several cavity modes is qualitatively similar to the behavior of the interacting cavity mode and inhomogeneously broadened exciton described by Houdre et al. [43].

It is interesting to analyze the temporal dynamics of the exciton population in meso-cavities. Figures 3(a) and 3(b) show a number of excitons in the system after instant injection of 1000 excitons. In this case, we considered a linear model. That is, we took U = 0. Therefore, the magnitude of the injected excitons does not affect the population dynamics. In Supplement 1, we considered the effect of the number of injected excitons on the population dynamics. It can be seen, that there is pulsed oscillation of the number exciton in the system, and the frequency of the beating is equal to the frequency separation between cavity modes Δ. Pulsed build-up

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of the exciton population (and excitonic polarization of the system) could manifest itself in various experiments studying non-linear behavior of system, in particular in four-wave mixing experiments. Note that the magnitude of oscillation is reduced due to radiative leakage and non-radiative damping of excitons, and it can be seen, and beating decay during the time interval corresponding to radiative damping of the exciton. We took the values for dissipation for all considered cases close to the values considered in the works [33,44,45].

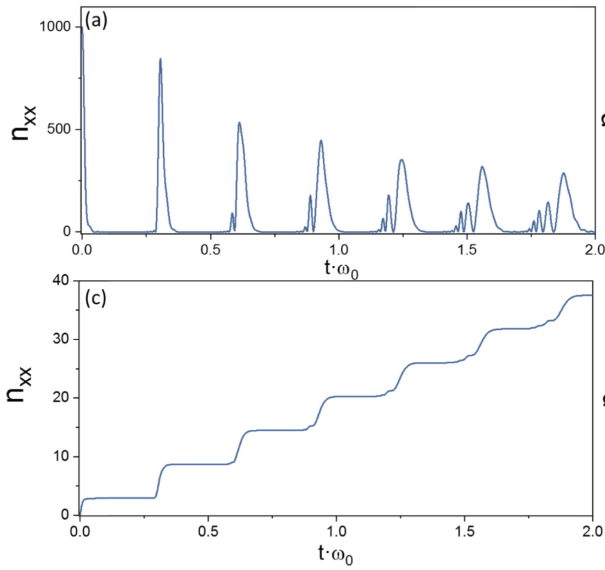

Fig. 3. Temporal dependence of the number of particles in the exciton mode on time. For CW pumping $I = 0.2\omega_0$ (c, d) and temporal dependence of the population of the excitonic mode after single pulsed excitation providing occupancy of excitonic mode $n_{xx}{}^0 = 1000$ (a, b), shown for small temporal interval $2\omega_0^{-1}$ ps (a, c) and "long" interval $80\omega_0^{-1}$ ps. The damping of exciton is $\gamma_0 = 0.02 \cdot 10^{-3}\omega_0$, photons decay are $\gamma_i = 0.02 \cdot 10^{-3}\omega_0$, exciton-photon coupling constant $g_k \approx 14.3 \cdot 10^{-3}\omega_0$, exciton-exciton interaction constant $U = 0$, number of photon modes is $N = 21$, energy distance between the modes is $\Delta = 14.3 \cdot 10^{-3}\omega_0$.

In the case of CW exciton injection, there is a ladder-like increase of the exciton population, as shown in Figs. 3(c) and 3(d). The width (duration) of each step is defined by the separation of the cavity modes Δ, and is identical to the interval between pulses shown in Fig. 3(a).

The step-like increase of the population has been demonstrated in the cavity, where a set of polaritons interacts with the THz mode [11].

Polariton bistability [33,38,44–49] is one of the most fascinating effects in the polaritonics, which often regarded as a basing principle of the construction of future polaritonic information processing systems [44,45,48,49]. It is a platform for studying multichannel entanglement arising from multiple optical branches and the nature of polaritons with large optical nonlinearities [50,51]. This effect manifests itself as hysteresis-like dependence of the population of polariton states as a function of pumping and occurs in the case of non-linear interaction of polaritons, for example, due to exciton-exciton interaction. Since the system has bistability properties, its response depends on the prehistory of the process, which leads to the possibility of observing the photoluminescence intensity hysteresis. The interaction of polaritons results in a blue shift [52] and leads to an abrupt increase in the polariton population. Significant progress is also observed in the experimental part of the study of polariton bistability. For example, a novel type of bistable behavior controlled by the phase twist across the fluid is experimentally evidenced in [53]. Similar to the case of interaction of single polariton and single exciton, meso-cavity where several photonic modes interact with exciton demonstrate bistability effect. Figure 4 shows the dependencies of the population of polariton levels in meso-cavity, supporting three cavity modes interacting with an exciton in the presence of exciton-exciton interaction characterized by interaction constant U = $12.86 \cdot 10^{-6}\omega_0$ (If the exciton energy is $\omega_0 = 3.5 \cdot$ eV, then U = 45μeV [34,49,54]). The behavior of the population of polariton modes from pumping demonstrates a threshold behavior. At sufficiently low pumping values, the dependence of the population of polariton MOs on pumping is very weak. When the pump reaches a certain threshold value, the occupancy of the polariton modes increases sharply and then grows much faster. This behavior is similar to the behavior of populations, which is obtained by solving the semiclassical Boltzmann equations. The magnitude of the hysteresis loop strongly depends on the magnitudes of the dissipations of the exciton and photon modes. With an increase in dissipation, the hysteresis loop decreases. This behavior is described in sufficient detail in [38]. It can be seen, that all four polaritonic modes demonstrate hysteresis-like dependence, but for ground state polariton, there is non-monotonic behavior of the population on the pumping. Such additional non-linear feature be used to enhance the range of non-linear application of polaritons.

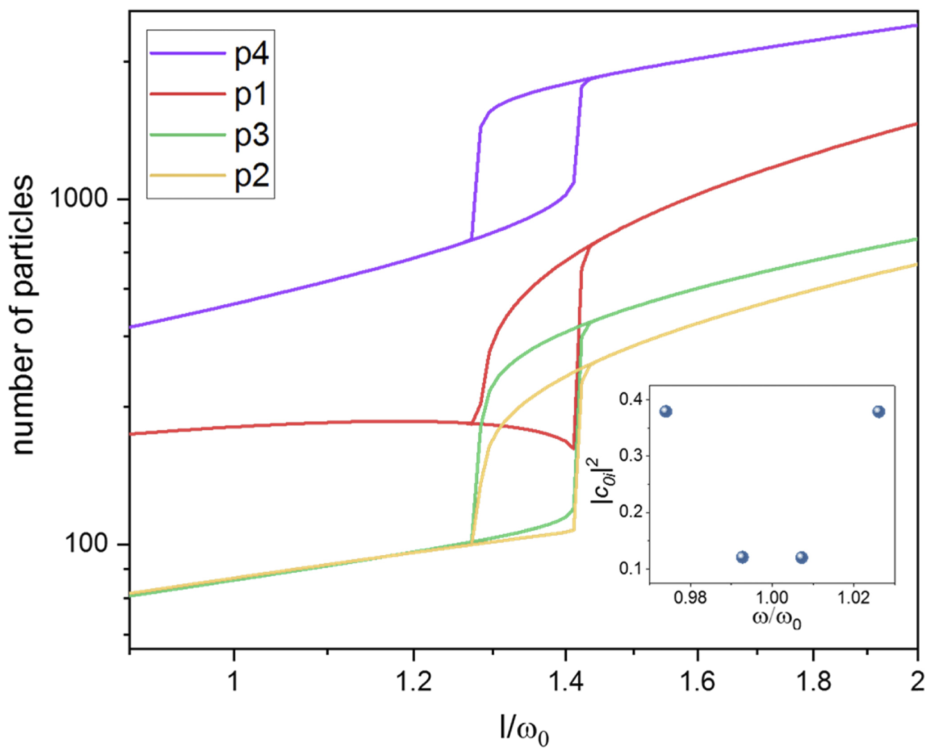

Fig. 4. Dependence of the population of polariton modes on pumping. The low-energy mode (p1) has an anomalous bistability loop with non-monotonic dependence of occupancy on pumping. Inset below: Hopfield coefficient illustrating excitonic contribution to polariton modes. The parameters of the system are the following: damping of exciton is $\gamma_0 = 0.85 \cdot 10^{-3}\omega_0$, photons decay are $\gamma_i = 5 \cdot 10^{-3}\omega_0$, exciton-exciton interaction constant $U = 12.86 \cdot 10^{-6}\omega_0$, exciton-photon coupling strength $g_k \approx 14.3 \cdot 10^{-3}\omega_0$, number of photon modes is $N = 3$, energy distance between the modes is $\Delta = 14.3 \cdot 10^{-3}\omega_0$.

4. Conclusion

We have considered the interaction of excitons and cavity modes in the structure, supporting several cavity modes with separation corresponding to the strength of exciton-photon interaction. We have demonstrated a step-like increase of the excitonic population with CW pumping and quantum beating in the case of pulsed injection of excitons. In the presence of exciton-exciton interaction, polaritons in meso-cavity demonstrate bistability accompanied by non-monotonic dependence of population of ground state polariton vs pumping, which enriches the pattern of non-linear effects in polaritonics.

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Disclosures. The authors declare no conflicts of interest.

Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

Supplemental document. See Supplement 1 for supporting content.

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