Effects of spatial quantization and Rabi-shifted resonances in single and double excitation of quantum wells and wires induced by few-photon optical field

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Abstract  
We develop a fully quantum theoretical approach which describes the dynamics of Frenkel excitons and bi-excitons induced by few photon quantum light in a quantum well or wire (atomic chain) of finite lateral size. The excitation process is found to consist in the Rabi-like oscillations between the collective symmetric states characterized by discrete energy levels. At the same time, the enhanced excitation of high-lying free exciton states being in resonance with these ‘dressed’ polariton eigenstates is revealed. This found new effect is referred to as the formation of Rabi-shifted resonances and appears to be the most important and new feature established for the excitation of 1D and 2D nanostructures with final lateral size. The found new physics changes dramatically the conventional concepts of exciton formation and play an important role for the development of nanoelectronics and quantum information protocols involving manifold excitations in nanosystems.

Keywords: semiconductor nanostructures, quantum light, excitation dynamics, spatial quantization, Rabi-shifted resonances

(Some figures may appear in colour only in the online journal)

1. Introduction

Semiconductor systems and nanostructures are now very attractive for fundamental investigations and are found to be very promising for many practical applications. The interaction of such systems with few-photon quantum light opens new possibilities in development of nanoelectronics, quantum measurements, semiconductor quantum optics and quantum information science [1–3]. For the systems with reduced dimensionality the excitation by light leads to pronounced interband transitions and efficient coupling between several or only two electronic bands due to the effect of spatial quantization [4]. In such a case the theoretical treatment is usually restricted to the few-level models. However large (or even infinite) longitudinal dimensions of such systems result in quasi-continuum of additional non-resonant transitions possible for states with different initial momentum. To take into account such transitions theoretically is a separate benefit task. Moreover, the momentum conservation is fulfilled with the
accuracy up to the inverse size of the system. For this reason, in largely extended semiconductors only ‘vertical’ field-induced transitions in the momentum space are allowed and an exciton is formed with a total quasi-momentum equal to zero [5–9].

However in systems with a finite size $L$, the momentum conservation law is not strictly fulfilled, and the quasi-momentum is efficiently conserved only up to the value of $1/L$. In quantum wells and wires with size in some directions significantly larger than atomic ones, but finite, the spatial quantization results in a discretization of exciton momentum and energy states that are distributed quasi-continuously with distance between individual levels decreasing with growing $L$. Actually not the only exciton state with zero quasi-momentum but a set of states around it is involved in the transitions. This peculiarity leads to specific features of the exciton dynamics which is still not analyzed and described correctly since strictly speaking an infinite number of states should be taken into account in this case.

In our work the interaction of a quantum field mode in a high-Q resonator with a quantum well or a quantum wire of finite size is investigated and the formation of excitons and bi-excitons induced by few photon quantum field is analyzed. For such systems, the interaction with electromagnetic fields opens new possibilities to control the localized field-induced excitations and currents that are now considered as good candidates for encoding and transfer of quantum information. For this reason the performed investigations appear to be very promising for the development of nanoelectronics and quantum information technology. We focus mainly on new physical effects arising due to the finite lateral size of the considered nanostructures.

We study the formation of excitons and bi-excitons induced by one or two photons of quantum field mode. In the case of very-high-Q resonator the interaction with single photon mode can be very efficient and corresponding Rabi period can be much shorter than relaxation times of the electronic subsystem [8, 10, 11] so as we neglect the dissipation induced by electron-phonon interaction and the Auger decay of multiple exciton states.

To correctly take into account the quasi-continuum of excitonic states mentioned above and formed due to spatial quantization effects, we start from the microscopic analysis and develop a fully quantum theoretical approach which describes the field-induced dynamics of Frenkel excitons in a quantum well or wire (atomic chain) of finite lateral size. The model of Frenkel excitons is well approved for organic media, where excitons can be regarded as excitations in individual molecules coupled with each other by dipole-dipole interaction. Such model is important in organic semiconducting nanowires which are of high interest in optoelectronic devices (see, e.g. [12]). To take into account the electron-electron interaction the dipole-dipole transfer of excitation between neighboring atoms is considered. In the frame of the developed approach, a set of novel and important results is obtained. We have found the energies and eigenfunctions of collective excitonic states ‘dressed’ by quantum light in a multi-atomic system and analyze the role of spatial confinement and energy quantization effects in 1D and 2D cases. New features and peculiarities of the exciton dynamics are found. We show that evolution of the system mainly demonstrates polariton-like behavior resulting in Rabi oscillations between the non-excited initial state and the collective symmetric excitonic mode. At the same time, the enhanced excitation of high-lying free exciton states being in resonance with these ‘dressed’ polariton eigenstates is shown to be of great importance. This found new effect is referred to as the formation of Rabi-shifted resonances and cannot be described in terms of the ladder bosonic operators that are often used for theoretical treatment of exciton formation in semiconductors [13–19]. We have shown that under some conditions the excitation of these resonant states is predominant in comparison to the usually discussed polariton states. The performed research and revealed physical mechanisms are very important to provide control and managing of the quantum properties and dynamics of excitons in real nanostructures.

2. Theoretical approach

The microscopic description of the Frenkel excitons is based on consideration of a semiconductor nanostructure as a chain of $N$ two-level atoms (in 1D case) which corresponds to a nanowire or square lattice of $N \times N$ atoms (in 2D case) which describes a quantum well with the dipole-dipole excitation transfer between neighboring atoms being taken into account.

In the case of atomic chain the Hamiltonian of such system interacting with quantum field mode consists of three terms: the non-perturbed photon and exciton energies $H_{ph}$ and $H_0$ respectively and the interaction term $H_{int}$ and is given by:

$$H = H_{ph} + H_0 + H_{int}$$

$$= h\omega b^\dagger b + \left( \sum_{n=1}^{N} \varepsilon n a_n^\dagger a_n - \sum_{n=1}^{N-1} w a_{n+1}^\dagger a_n - \sum_{n=2}^{N} w a_n^\dagger a_{n-1} \right)$$

$$+ g \sum_{n=1}^{N} \left( h a_n^\dagger b^\dagger a_n + h a_n b a_n^\dagger \right).$$ (1)

Here $b^\dagger$ ($b$) stand for bosonic creation (annihilation) photon operators in a resonator mode, $a_n^\dagger$ ($a_n$) are the fermionic operators of creation (annihilation) of excitation of the atom with number ‘$n$’ with double excitation of the same atom being forbidden. The parameter $w$ characterizes the strength of dipole-dipole excitation transfer which depends on the field polarization direction in relation to the chain orientation, $\varepsilon$ and $h\omega$ stand for energies of atomic excitation and a photon respectively. The field mode is considered as a plane wave with wave vector oriented perpendicularly to the chain or the atomic lattice. For this reason the interaction of the atomic chain with photons is presented in the dipole approximation and $g$ is the coupling constant of photon-atom interaction. The rotative wave approximation is used i.e. terms with fast optical oscillations are excluded.

It should be noticed that the developed theoretical approach is based on the model of two-level atoms and the highest excited and ionized atomic states are neglected. This leads to the fact that, for example, the Auger decay of multiple exciton
states, in which two excitons pass into the ionized electron–hole pair, is not taken into account. Therefore the applicability of our approach is limited by times less than the inverse rate of the Auger decay. Nevertheless, this restriction is not fundamental since the qualitative nature of the interaction with the quantum photon field is the same even if more atomic states are involved in the excitation process.

The wave-functions of the field-free eigenstates $|1,k\rangle$ of a single exciton in the chain (nanowire) describe the excitation of the ‘$n$th’ atom with probability amplitude $\chi_{nk}^N$:

$$|0\rangle_{\text{ph}} |1,k\rangle_{\text{ex}} = \sum_{n=1}^{N} \chi_{nk}^N a_n^\dagger |0\rangle, \quad \chi_{nk}^N = \sqrt{\frac{2}{N+1}} \sin \left( \frac{\pi nk}{N+1} \right).$$

(2)

Here $\chi_{nk}^N$ can be referred to the shape of the field-free exciton mode with $k = 1,\ldots,N$. The energies of these states give rise to the exciton dispersion law typical for many semiconductors:

$$E_{1,k} = \varepsilon - 2w \cos \left( \frac{\pi k}{N+1} \right) \equiv \varepsilon_k.$$  

(3)

For large $N$ this energy turns into the usually used parabolic dependence [9] on the quasi-momentum $q = \pm \frac{\pi k}{N+1}$ around the extremum $\varepsilon_0 = \varepsilon - 2w$.

$$E_{1,k} \approx \varepsilon_0 + w \left( \frac{\pi k}{N+1} \right)^2 = \varepsilon_0 + \frac{\hbar^2 q^2}{2m_{\text{eff}}}. \tag{4}$$

Here the effective mass is given by $m_{\text{eff}} = \hbar^2/(2wd^2)$, $d$ is the distance between atoms. For our system of finite size, the minimal exciton energy slightly exceeds $\varepsilon_0$ due to quantum-confinement effects and is equal to $\varepsilon_0 + w \pi^2/(N+1)^2$. For convenience we introduce in equation (3) the set of energies $\varepsilon_k = \varepsilon_0 + w \pi^2 k^2/(N+1)^2$.

States with several excitons are obtained in a similar way, taking into account the fact that two excitations cannot exist on the same atom. Thus, the energy of the biexciton state is given by:

$$E_{2,k_1,k_2} = 2\varepsilon - 2w \left( \cos \left( \frac{\pi k_1}{N+1} \right) + \cos \left( \frac{\pi k_2}{N+1} \right) \right), \quad k_1 = 1,\ldots,N-1, \quad k_2 = k_1 + 1,\ldots,N. \tag{5}$$

It is assumed that $k_2 > k_1$ due to the indistinguishability of excitons. The total number of biexciton states is equal to $N(N-1)/2$.

The details of the analytical description of exciton in 1D and 3D case are given in appendices A and B respectively, while the generalization to the $m$ exciton case is described in appendix C.

In contrast to wave-functions (2) another set of excitonic modes can be introduced that follow immediately from the interaction term in the Hamiltonian. Indeed, the operator of the interaction with quantum field can be rewritten in the following form:

$$\hat{H}_{\text{int}} = g \sum_{n=1}^{N} (b_{n}^{\dagger} b_{n} + b_{n}^{\dagger} b_{n}) \equiv g \left(bR^{\dagger} + b^{\dagger}R\right), \tag{6}$$

where the operator $R^{\dagger} = \sum_{n=1}^{N} a_{n}^{\dagger}$ is responsible for the creation of the excitation of one of the atoms in the chain with equal probability. It can be easily seen that this operator appears to be bosonic one and leads to the excitation of the symmetric ‘collective’ excitonic mode in the chain $1/\sqrt{N}R^{\dagger}|0\rangle$ with zero value of quasi-momentum of the exciton. At the same time the biexcitonic symmetric ‘collective’ state can be found as $1/\sqrt{2N(N-1)} R^{\dagger,2}|0\rangle$. Though the exciton dynamics in semiconductors is usually described in terms of the bosonic ladder operators and symmetric collective excitonic modes [13–19], these states differ dramatically from the field-free excitonic eigenstates.

The interaction between the atomic chain and quantum field mode presented in (1) leads to the selection rules of field-induced transitions providing excitation of states (2) only with odd quantum numbers $k = 1,3,\ldots, 2\left\lfloor \frac{N+1}{2} \right\rfloor + 1$. Such states can be called ‘active’. The oscillator strength for the transition to these states can be calculated analytically:

$$f_k = \left| \gamma_k \right|^2, \quad \gamma_k = \langle 0 | bR^{\dagger} + b^{\dagger}R | 0 \rangle_{\text{ph}} |1,k\rangle_{\text{ex}} = \sum_{n=1}^{N} \chi_{nk}^N$$

$$= \sqrt{\left(1 - (-1)^k\right)} \frac{\sqrt{\frac{2}{(N+1)^2}} \cot \left( \frac{\pi k}{2(N+1)} \right)}{\sqrt{N+1}}. \tag{7}$$

Here we introduce the set of matrix elements $\gamma_k$ which are non-zero only for odd $k$.

In case of $N >> 1$, the dipole matrix element can be shown to be inversely proportional to the quantum number $k$ of excitonic mode (see appendix A, equation (A.2)). Evidently, these oscillator strengths satisfy the sum rule $\sum_{k=1}^{N} f_k = N$.

It should be emphasized that the superposition of free exciton eigenstates $|1,k\rangle$ with amplitudes equal to the transition matrix elements (7) explicitly gives collective symmetric excitonic state:

$$\sum_{k=1}^{N} \gamma_k |0\rangle_{\text{ph}} |1,k\rangle_{\text{ex}} \equiv \frac{1}{\sqrt{N}} R^{\dagger}|0\rangle. \tag{8}$$

Thus, a direct correspondence between transitions to a set of field-free excitonic states and the excitation of a collective symmetric mode is found. Does it mean that the formation and evolution of the excitation process in these two representations are equivalent? Below we analyze the role of collective states in excitation dynamics and answer this question.
3. Results and discussion

In this section, we describe the dynamics of excitons induced in quantum wire and quantum well by few-photon quantum light in the case of the limited linear dimensions of the system (10 μm–100 μm). The revealed dynamics is compared with the results obtained in the frame of bosonic ladder creation/annihilation operators of excitation of symmetric collective states. The spatial confinement of the system is found to induce new physical features of the time-dependent exciton behavior arising due to the formation of Rabi-shifted resonances and free exciton states.

3.1. Exact quasi-energy solution and dynamics of excitation

Let us first consider a 1D atomic chain and only one photon in the quantum field mode. The interaction is supposed to be switched on at the time moment \( t = 0 \). The initial state of the system corresponds to first Fock state for the quantum field and zero number of excitons in the chain and is given by \( |0\rangle_{ph} |0\rangle_{ex} \). In this case, only ‘active’ states describing one exciton and zero number of photons in the system \( |0\rangle_{ph} |1,J\rangle_{ex} \) (with odd \( k \)) are involved in the excitation process.

The diagonalization of the total Hamiltonian including the interaction term leads to the following equation:

\[
\hat{H} |\psi\rangle = \lambda |\psi\rangle ,
\]

(9)

where \( \lambda \) stands for the eigenenergies (quasi-energies) of the joint system describing the nanostructure, ‘dressed’ by quantum field, and is shown to obey the following equation:

\[
(\hbar \omega - \lambda) = g^2 \sum_{j=1, \text{odd } j}^{N} \frac{\gamma_j}{\epsilon_j - \lambda} =: g^2 G(\lambda).
\]

(10)

In the limit of large number of atoms these energies can be found analytically (see appendix A).

The eigenvectors (quasi-energy states) corresponding to the found from (10) quasi-energies \( \lambda_m \) and presented in terms of the free excitonic basis (2) are given by:

\[
|\psi_m\rangle = P_m^{-1/2} \left( |1\rangle_{ph} |0\rangle_{ex} + \sum_{j=1, \text{odd } j}^{N} \frac{\gamma_j}{\epsilon_j - \lambda_m} |0\rangle_{ph} |1,J\rangle_{ex} \right)
\]

(11)

with normalization factor \( P_m \) found in the form:

\[
P_m \equiv 1 + g^2 \sum_{j=1, \text{odd } j}^{N} \frac{\gamma_j^2}{(\epsilon_j - \lambda_m)^2} = 1 + g^2 \frac{dG(\lambda)}{d\lambda} \bigg|_{\lambda = \lambda_m}.
\]

(12)

Figure 1(a) shows the spectrum of the found quasi-energies in dependence on detuning \( \delta = \hbar \omega - \varepsilon_0 \). Several important features are demonstrated. Instead of two Rabi-originated polariton dispersion curves well-known for two resonantly coupled states, there is only one lower polariton branch (curve 3) corresponding to the discrete quasi-energy \( \lambda_0 < \varepsilon_0 \) but a lot of the quasi-continuum energy levels with \( \lambda_m > \varepsilon_0 \) (above line 1) found for each detuning. Red dashed line (2) describes photon state for zero \( g \) (without interaction with chain corresponding quasi-energy equals to \( \hbar \omega \)). In case of only one photon we have polariton states based on two types of states w/o interaction: ‘photon without exciton’ (only one state) and ‘exciton without photon’ (the number of such states equals to the number of active states \( M \)). With account of interaction the first state converts to lowest polariton branch with strong dependence on detuning (proportional to detuning for negative detunings), whereas quasi-energies for second type of states are roughly horizontal on the plot (with deviation near the upper polariton energies).

The presented above description corresponds to the exact solution of the problem and we find the quasi-energies \( \lambda_m \) for this solution. Therefore, we can obtain the time-dependent state vector for any initial condition. All the found quasi-energy states contribute to the time evolution of the state vector:

\[
\varphi(t) = \sum_{m=0}^{M} b_m |\psi_m\rangle \exp(i\lambda_m t / \hbar)
\]

(13)

with coefficients \( b_m = \langle |\psi_m\rangle |1\rangle_{ph} |0\rangle_{ex} = P_m^{-1/2} \) which strongly depend on the initial condition and the value \( \lambda_m \) and can be found analytically in the limit of large number of atoms (see appendix A). We can find the time-dependent population of any required state by its projection onto the time-dependent state vector (13). For example, the initial state probability can be calculated as follows:

\[
\rho_0(t) = |\langle 1\rangle_{ph} |0\rangle_{ex} \varphi(t) \rangle|^2 = \sum_{m=0}^{M} P_m^{-1} \exp(i\lambda_m t)^2.
\]

(14)

The total probability of exciton formation in dependence on time is simply found as \( \rho_{ex} = 1 - \rho_0 \). The time dependent population \( \rho_j \) of a certain exciton state \( |0\rangle_{ph} |1,J\rangle_{ex} \) is given by (see equation (11)):

\[
\rho_j(t) = |\langle 0\rangle_{ph} |1,J\rangle_{ex} \varphi(t) \rangle|^2
\]

(15)

\[
= \sum_{m=0}^{M} g^2 \frac{\gamma_j}{(\epsilon_j - \lambda_m)} P_m^{-1} \exp(i\lambda_m t)^2.
\]

In addition, the population of the collective symmetric state \( \sqrt{N} R^+ |0\rangle \) equals to:

\[
\rho_{R^+}(t) = |\langle 0\rangle_{ph} \frac{1}{\sqrt{N}} R \varphi(t) \rangle|^2
\]

(16)

\[
= \sum_{m=0}^{M} g P_m^{-1} \exp(i\lambda_m t) \sum_{j=1, \text{odd } j}^{N} \frac{\gamma_j}{(\epsilon_j - \lambda_m)}^2.
\]
Figure 1. Simulation of 1D chain with $N = 20000$ atoms, $w = 0.25$ eV, $g\sqrt{N} = 30 \mu$eV. (a) Quasi-energies (green curves) depending on the detuning $\delta = \hbar \omega - \varepsilon_0$; the lower (curve 3) and upper (curve 4) polariton branches, $\varepsilon = \hbar \omega$—red dashed line (2), $\varepsilon = \varepsilon_0$—line (1). (b) Distribution of weights of ‘dressed’ quasi-energy states for different detuning $\delta = \hbar \omega - \varepsilon_0$. Large circles correspond to positions and weights of quasi-energies for 2-level system values $P_{-1}$ and $\sum_{m=1}^{M} P_m = 1 - P_{-1}$ (in $R^+ |0\rangle$ approximation). (c) Time-dependent population of excitons (blue) and the difference between this population and the population of the collective symmetric state $\sqrt{N} R^+ |0\rangle$ (red) calculated for negative detuning $\delta = -2g\sqrt{N} = -60 \mu$eV. The inset shows the structure of this and all following ‘population vs time’ plot with two scales in intervals 0–1.5 ns and 1.5–30 ns. (d) Square root of population $\sqrt{\rho_j(t)}$ of different excitonic states for the same parameters. (e), (f) The same as (c), (d) for zero detuning $\delta = 0$. (g), (h) The same as (c), (d) for positive detuning $\delta = g\sqrt{N} = 30 \mu$eV.
The weights $b_m^2 = P^{-1}_m$ of different quasi-energy levels with $\lambda_m > \varepsilon_0$ involved in the solution for the chosen initial condition are shown on the inset in figure 1(a) by changing color intensity of red dots representing different levels with $\lambda_m$. As a result, the dots with maximum of $P^{-1/2}_m$ for positive $m$ and different detunings form the upper polariton branch shown by curve 4 in the main part of figure 1(a). The structure of this branch is more complex than that for lower polariton curve, due to its quasi-discrete character and avoid-crossing features arising from the influence of many close neighboring quasi-energy states. All other eigenstates have negligibly small contribution of photon state and looks like pure exciton states.

Figure 1(b) shows more precisely the weights $b_m^2 = P^{-1}_m$ of quasi-energies for four values of detuning. One low quasi-energy state with $\lambda_0 < \varepsilon_0$ and a few ‘dressed’ states around some most pronounced quasi-energy $\lambda^* > \varepsilon_0$ are seen to contribute mainly to the final solution for each detuning. The two predominate quasi-energies $\lambda_0$ and $\lambda^*$ are found to form the well-known structure of the lower and upper polariton energies arising usually in the two-level system and providing the Rabi oscillations. From the structure of the eigenvector with quasi-energy $\lambda_0$ which for large $N$ can be estimated analytically as follows

$$
\psi_0 \approx P^{-1/2}_0 \left( |1\rangle_{ph}|0\rangle_{ex} + \frac{g\sqrt{N}}{\lambda_0 - \varepsilon_0} \sum_{k = 1, \text{odd}}^{N} \gamma_k |0\rangle_{ph} |1\rangle_{ex} \right)
= P^{-1/2}_0 \left( |1\rangle_{ph}|0\rangle_{ex} + \frac{g}{\lambda_0 - \varepsilon_0} R^\dagger |0\rangle \right),
$$

it can be easily understood that the found Rabi oscillations are provided between the initial state and the collective symmetric excitonic mode $\frac{1}{\sqrt{N}} R^\dagger |0\rangle$. Indeed, the arising Rabi oscillations are clearly seen on the time-dependent probability of excitons presented in figures 1(c), (e) and (g). For more convenience, the abscissa has two time scales: times below 1.5 ns are shown in more details. At the initial stage of the excitation dynamics, the above-mentioned Rabi-like oscillations take place and the red curve is close to zero. The probability of excitons together with the population of initial state gives unity. Thus the minimal probability of the system to remain in the initial state during the excitation appears to be rather high (about 50%) for negative detunings (figure 1(c)), but is found to be close to zero for the resonant case and positive detunings (figures 1(e) and (g)).

However, for larger time the found Rabi oscillations are shown to be characterized by some slow modulation. In this regime the population of collective symmetric state $\frac{1}{\sqrt{N}} R^\dagger |0\rangle$ no longer fully describes the excitation process since the population of some additional states increases with time (red curve) and starts to oscillate with frequency much lower than the Rabi frequency. These slow oscillations are found to arise due to the population of the field-free excitonic ‘active’ states with a certain energy $\varepsilon_k$ being in resonance with the ‘dressed’ eigenenergy $\lambda^*$ corresponding to the upper polariton branch. Therefore, these states are referred to as the Rabi-shifted resonance states and are characterized by a group of $k$ around a certain number of $k$ in equations (3) and (4) which increases from 25 up to about 50 with changing the detuning and growing the value of $\lambda^*$ (see 3D plots in figures 1(d), (f) and (h)). The characteristic frequency of the slow oscillations is determined by matrix element $\gamma_k$ (equation (7)) which is rather low and is inversely proportional to the mean number $\langle k \rangle$ of the populated free excitonic states. For $k$ around 50 the period of slow oscillations is estimated as about 5 nanoseconds and its half is very close to characteristic time of the population growing of the resonant states presented in figure 1(h).

At the same time the excitonic states forming the collective symmetric state $\frac{1}{\sqrt{N}} R^\dagger |0\rangle$ are characterized by rather small $k$-numbers (see figures 1(d), (f) and (h)). As a result, the population of the resonance states and the collective symmetric mode can be distinguished both in time and momentum domain.

In a ‘dressed states’ picture these ‘resonance’ states form the eigenvectors with quasi-energies neighboring to the $\lambda^*$ and characterized by rather high weights (see figure 1(b)). For this reason at least couple of quasi-energies around the peak value $\lambda^*$ should be always taken into account to describe correctly the excitation dynamics.

Thus, except the found resonances, the lower and upper polariton branches are responsible for the Rabi oscillations between the initial state and symmetric excitonic mode $\frac{1}{\sqrt{N}} R^\dagger |0\rangle$ that can be correctly described in terms of the bosonic ladder operators. However, possible resonances between the free exciton energies and the upper polariton branch appear to change significantly the dynamics of the excitation process. This effect is shown to become important at rather large times and appears to be significant at zero and positive detuning. For negative detuning or in the limit of infinitely large number of atoms (infinite size of the nanostructure) the population of these resonance states can be strongly suppressed.

It should be also noticed that the obtained results are valid if the dissipative processes are negligibly small. The effects of dephasing lead to suppression of the resonant state population. However in the case of efficient interaction with quantum field the found dynamics takes place at times significantly shorter than the characteristic dissipation time and the revealed physical effects are really observed. Thus, the performed analysis allows us to reveal new features of the quantum-optical excitation of nanostructures and establish the limitations and applicability of its description using the bosonic ladder operators.

### 3.2. Comparison with 3-level system

To prove the idea about an important role of the found ‘resonance’ states we consider a much simpler system characterized by two states resonantly coupled by photon with coupling constant $g$ (ground and main excited states, to be specific) and the upper state with detuning $\Delta$ above the main level with weaker interaction with light (coupling constant $g$).
$g \mu$, $\mu \ll 1$). The scheme of considered energy levels is presented in figure 2(a).

If we neglect the interaction with the upper level ($\mu = 0$), the system is characterized by two quasi-energies $\varepsilon_0 \pm g$ (see figure 2(a), left panel). When we take into account the weak interaction with the upper level which is in resonance with the upper quasi-energy $\varepsilon_0 + g$ ($\Delta = g$), this quasi-energy is split into two quasi-energies due to avoid-crossing effect (see figure 2(a), right panel). The explicit values of quasi-energies can be easily found in such a case and for $\mu \ll 1$ in linear approximation on $\mu$ appear to be equal to $\varepsilon_0 - g$, $\varepsilon_0 + g \pm g \mu / \sqrt{2}$. If at $t = 0$ system is in the ground state and there is one photon in the field mode, the populations of ground $|C_0 (t)|^2$ and main excited state $|C_1 (t)|^2$ depends on time as follows:

$$
|C_0 (t)|^2 = \frac{3}{8} + \frac{1}{2} \cos \left(2h^{-1}gt\right) \cos \left(\frac{h^{-1}g\mu t}{\sqrt{2}}\right)
+ \frac{1}{8} \cos \left(\frac{\sqrt{2}h^{-1}g\mu t}{\sqrt{2}}\right),
$$

(18)

while the population of the upper Rabi-shifted resonance level $|C_2 (t)|^2$ is given by:

$$
|C_2 (t)|^2 = \frac{1}{4} \left(1 - \cos \left(\sqrt{2}h^{-1}g\mu t\right)\right).
$$

(19)

The dynamics of these found populations is shown in figure 2(b). The evolution of this system qualitatively represents the evolution of excited states in the chain for $\delta = 0$ (compare with figure 1(e)). It is seen that initially weakly coupled upper level can efficiently exchange the energy with polariton formed by two main levels and the photon, with maximum achievable population equal to one half. This polariton state is the analog of the polariton state in the 1D chain formed by coupling of collective symmetric state $\frac{1}{\sqrt{2}} R^{+} |0\rangle$ and initial state with one photon, whereas the upper resonance level corresponds to a set of Rabi-shifted resonance levels in the chain. Since in the chain (nanowire) the resonance states contribute to more ‘dressed’ eigenvectors, the slow resonance oscillations of their population appear to be more irregular than in three-level system.

For the non-zero detuning $\delta = \hbar \omega - \varepsilon_0$ similar results can be obtained. The significant population of the upper (third) state is achieved if this level is shifted above the main one by the value $\Delta = \frac{1}{2} \left(\delta + \sqrt{4g^2 + \delta^2}\right)$. The time-dependent population of the main and upper levels is presented in figures 2(c) and (d) and is evidently very close to the obtained exciton dynamics illustrated by figures 1(c), (e) and (g).

The oscillations of the population of the upper Rabi-shifted resonance level described by equation (19) is defined by the difference $\sqrt{2}g\mu$ between two upper eigenenergies. We can compare this result with the frequency of slow oscillations in the case of 1D chain described in the previous subsection. The frequency of slow oscillations is approximately equal to $\sqrt{2}g\gamma_k$ where $k$ is the mean number of the populated free excitonic states being in resonance with the upper polariton branch (see figures 1(d), (g) and (h)).
Thus, the coincidence (resonance) of energy of free exciton states with the upper polariton branch appears to perturb dramatically the quasi-energy spectrum of the whole system and leads to significant population of these ‘resonance’ states.

3.3. Spatial distribution of excited atoms in 1D case

We can estimate the spatial distribution of excited atoms by using the excitonic spatial modes $\chi_{in}$ defined by equation (2). Figures 3(b)–(h) shows the evolution of the spatial distribution in time for specific times which corresponds to maxima and minima of exciton population (see figure 3(a)). At the first maximum of Rabi oscillations ($t = 0.033$ ns) the spatial distribution of excited atoms is almost uniform (except some boundary oscillations). The population of any atom is the same and equals to $1/N$. This distribution corresponds to collective symmetric state $\phi_{jN}$. With the increase of time the excitation of additional resonant states with higher $j$ takes place and strongly modifies the spatial distribution. This redistribution comes from the spatial interference of different excitonic modes $\chi_{in}$ of free excitonic states being in resonance with upper polariton quasi-energies and strongly populated. The population of these states provide localization of excitation in the center or in peripheral areas with significantly different distribution in comparison to the collective symmetric mode. Therefore, the spatial structure reflects the development of the population of higher Rabi-resonant states.

3.4. Quantum well

Let us consider now a quantum well with monatomic thickness. The formulas which describe fragment of crystal with simple cubic structure and $N_x \times N_y \times N_z$ atoms are presented in appendix B. In the case of significantly large lateral dimensions with $N_x \gg 1$ and $N_y \gg 1$ the field-free exciton states $|1, \{k_x, k_y\}\rangle$ are characterized by two quantum numbers $k_x$ and $k_y$ with the correspondent energy written for large $N_x, N_y$ in the form:

$$E_{1,\{k,k\}} = \varepsilon - 2w_x \cos \left( \frac{\pi k_x}{N_x + 1} \right) - 2w_y \cos \left( \frac{\pi k_y}{N_y + 1} \right)$$

$$\approx \varepsilon - 2w_x - 2w_y + \left( \frac{\pi k_x}{N_x + 1} \right)^2 + \left( \frac{\pi k_y}{N_y + 1} \right)^2$$

for $k_x << N_x, k_y << N_y$. (20)

For simplicity we suppose that dipole moments of interacting atoms are directed perpendicular to the well. In this case the dipole-dipole transfer integrals are the same: $w_x = w_y = w > 0$.

The dipole matrix element for field-induced transitions depends on both $k_x, k_y$ and is given by:

$$\gamma_{\{k,k\}} = 4 \sqrt{\left(1 - (-1)^{k_x}\right)\left(1 - (-1)^{k_y}\right)} \frac{(N_x + 1)(N_y + 1)}{\pi^2 k_x k_y}.$$

(21)

The number of such ‘active’ states approximately equals to $N_x N_y/4$, since $\gamma_{\{k,k\}} \neq 0$ only for odd $k_x$ and $k_y$.

The theoretical analysis of the excitation in quantum well is rather similar to the case of the atomic chain using equation for quasi-energies similar to equation (10). Unfortunately the sums in this equation cannot be evaluated analytically. Function $G(\lambda)$ can be evaluated using Kramers–Kronig relations from the absorption of the system proportional to $\sum_{k_x = 1}^{N_x} \sum_{k_y = 1}^{N_y} \gamma^2_{\{k,k\}} \delta \left(\varepsilon_{\{k,k\}} - \hbar \omega\right)$. In case of quantum well main terms come from poles with $k_x = 1$ or $k_y = 1$. The total input from terms with $k_x \neq 1$ and $k_y \neq 1$ is about $1 - \frac{8}{27} \approx 0.19$. All poles are two-fold degenerated except one with $k_x = k_y = 1$. Thus, the free excitonic states with $k_x = 1$ or $k_y = 1$ appear mostly to be populated and determine the evolution of excitation process.

Figure 4 shows the dynamics of the excitations in case when only one photon is in the field mode and atoms in the well are unexcited at $t = 0$. In order to compare the cases of the chain and quantum well, we should keep the total response of the chain $(g^2 N)$ and the well $(g^2 N^2)$ to be the same (here $g$ is the coupling constant for the well). In this case the branches of lower and upper polaritons obtained for the 2D well and 1D chain behave in a similar way (compare figures 1(a) and 4(a)), while the number of quasi-energies is $N$ times higher in the case of the well, and the set of quasi-energies is much denser. The structure of weights of quasi-energies also appears to be similar for both cases, except the larger number of quasi-energies close to the upper polariton energy obtained for the quantum well (compare figures 1(b) and 4(b)). Some non-monotonous character of the weights of quasi-energies in a quantum well results from the mentioned above peculiarity of $\gamma_{\{k,k\}}$ which have larger values for $k_x = 1$ or $k_y = 1$. In the 2D case two different types of resonant states are populated which are close in energy but are characterized by different spatial modes and therefore by different coupling with the field. As a result the oscillations of their populations are characterized by very different frequencies and interfering with each other provide irregular temporal dynamics very soon.

The excitation dynamics in a quantum well is found to be very similar to the case of 1D chain. It consists in the Rabi oscillations between the initial state and the symmetric collective mode

$$\frac{1}{\sqrt{N_x N_y}} R^\dagger \left| 0 \right> = \frac{1}{\sqrt{N_x N_y}} \sum_{n_x = 1}^{N_x} \sum_{n_y = 1}^{N_y} a_{n_x n_y}^\dagger \left| 0 \right>$$

$$= \sum_{k_x = 1}^{N_x} \sum_{k_y = 1}^{N_y} \gamma_{\{k,k\}} \left| 1, \{k, k\} \right>,$$

(22)

accompanied by the slow population of the exciton states which are in resonance with the upper polariton branch. These ‘resonant’ states have almost the same energy proportional to $k_x^2 + k_y^2$ and their distribution over 2D quasi-momentum
Figure 3. Spatial distribution of excited atoms for 1D chain with $N = 20000$ atoms, $w = 0.25 \text{ eV}$, $g \sqrt{N} = 30 \mu \text{eV}$ for zero detuning $\delta = 0$ for different time values. Panel (a) shows the time-dependent population of excitons (equivalent to figure 1(c)) with points corresponded to chosen set of time values.
Figure 4. Simulation of 2D well with $N \times N = 20000 \times 20000$ atoms, $w = 0.25$ eV. $g_w N = 30 \mu$eV. (a) Quasi-energies (green curves) depending on the detuning $\delta = \hbar \omega - E_0$. (b) Distribution of weights of quasi-energies for different detunings $\delta$. Large circles correspond to positions and weights of quasi-energies for 2-level system ($R^+ |0\rangle$ approximation). (c) Time-dependent population of excitons (blue) and the difference between this population and the population of the collective symmetric state $1/\sqrt{N} R^+ |0\rangle$ (red) calculated for negative detuning $\delta = -2g_w N = -60 \mu$eV. (d) Sixth root of population $\left(\rho(t)\right)^{1/6}$ of different excitonic states for the same parameters at $t = 0.6$ ns. (e), (f) The same as (c), (d) for zero detuning $\delta = 0$. (g), (h) The same as (c), (d) for positive detuning $\delta = g_w N = 30 \mu$eV.
obtained at a certain instant of time forms a ring-shaped structure in figures 4(d), (f) and (h) with significant maxima pronounced at $k_1 = 1$ or $k_2 = 1$ due to specific features of matrix elements $\gamma(k_n, k_o)$. 

In contrast to 1D chain, in the case of 2D quantum well a set of free excitonic states with energy close to the upper polaron branch are characterized by rather large matrix element and can be populated with rather high weights. As a result, the contribution of the resonance states to the exciton dynamics are mostly pronounced in the case of quantum well and are seen to increase gradually even being averaged in time as it is presented in figures 4(c), (e) and (g) (red curves).

### 3.5. Interaction of two photons with 1D chain

Let us consider the case when there are initially $m$ photons in the photon mode. In this case the sequential excitation of the nanowire is possible up to the production of $m$ excitons in the system. The detailed theoretical analysis of such process is given in the appendix C. Here we focus on the two photon initial state of the field mode $|2, \phi \rangle |0 \rangle_{ex}$ providing exciton and bi-exciton production. For the chain with $N$ atoms there are $N$ excitonic and $N(N - 1)/2$ bi-excitonic free states denoted $|1, \phi \rangle |1, \phi \rangle_{ex}$ and $|0, \phi \rangle |2, k_1, k_2 \rangle_{ex}$ respectively. Due to Pauli principle and non-distinguishability of excitons $k_2$ should be larger than $k_1$. The energy of excitonic free state is

$$E_{1,1,k} = \hbar \omega + \varepsilon - 2w \cos \left( \frac{\pi k}{N + 1} \right),$$

whereas the energy of bi-exciton state equals to

$$E_{2,k_1,k_2} = 2\varepsilon - 2w \left( \cos \left( \frac{\pi k_1}{N + 1} \right) + \cos \left( \frac{\pi k_2}{N + 1} \right) \right).$$

For large $N$ and $w > 0$ the minimal energy of bi-exciton equals to $2\varepsilon_0 + 5w \pi^2/(N + 1)^2$ (achieved for $k_1 = 1$ and $k_2 = 2$) and exceeds the doubled exciton energy by $3w \pi^2/(N + 1)^2$. This fact means that the Pauli principle results in repulsion of two excitons, since the confinement effect is stronger for the second exciton and limits its localization between the primary exciton and one of the chain ends. This effect is mostly associated with the Frenkel-type excitons, while for the widely spread Wannier excitons the attraction effects can take place. Nevertheless, the repulsive excitons are also observed experimentally in real nanoplatelets [20].

The matrix elements of photon-induced transitions between these states and found selection rules are discussed in appendix C.

The spectrum of the ‘dressed’ states and their weights found in the case of initially unexcited system are presented in figures 5(a) and (b) respectively. The obtained polaron structure is found to consists now of 3 branches: lower (curve 4), intermediate (curve 5) and upper (curve 6) polaron branches which can be easily obtained in the frame of boson ladder operators and for zero detuning correspond to the quasi-energies $\varepsilon_0 - 2g$, $\varepsilon_0$ and $\varepsilon_0 + 2g$ respectively.

The correspondent collective symmetric modes are found as

$$\frac{1}{\sqrt{N}} R^+ |1, \phi \rangle |0 \rangle_{ex} = \frac{1}{\sqrt{N}} \sum_{n=1}^{N} a_n^\dagger |1, \phi \rangle |0 \rangle_{ex}$$

for excitonic state and

$$\frac{1}{\sqrt{N(N-1)}} (R^+)^2 |0 \rangle = \sqrt{\frac{2}{N(N-1)}} \sum_{n_1=1}^{N-1} \sum_{n_2=n_1+1}^{N} a_n^\dagger a_{n_2}^\dagger |0 \rangle$$

for bi-excitons. In case of two photons we have three base states without interaction: ‘two photons without exciton’ (only one state), ‘photon with exciton’ ($M$ states) and ‘bi-exciton without photon’ (about $M^2/2$ states). If we take the interaction into account, we have single lower polaron band (with asymptotic $2\delta$ for negative detunings), then $M$ quasienergies for states ‘photon with exciton’ (with asymptotic $\delta$ for negative detunings) and $M^2/2$ almost horizontal states for bi-excitons.

The structure of the found ‘dressed’ states can be seen from the figures 5(d), (f) and (h) where the contribution of different free exciton and bi-exciton states is presented. The states with very small energies above the threshold $\varepsilon_0$ and $2\varepsilon_0$ for exciton and bi-exciton channels respectively are found to contribute strongly to the collective symmetric modes. The relative contributions of these two channels are found to be significantly changed with detuning. Indeed, the dominance of excitons is found at negative detuning while for zero-detuning this channel is significantly suppressed and the prevail role of bi-excitons is revealed. At the same time, the additional contribution of free exciton and bi-exciton states with higher energies appears to be important that are not involved in the collective symmetric mode and can be referred to as ‘resonant’ states discussed in the previous sections. The oscillations show irregular character like in the case of the quantum well due to the same mechanism: the appearance of set of resonant states with different probability of excitation. In contrast to the 2D case, the number of such states is larger: both exciton and bi-exciton states with different coupling with the field contribute simultaneously.

As a result, the time-dependent probability of excitons and bi-excitons in the system presented in figures 5(c), (e) and (g) is mainly caused by dynamics of population of the collective symmetric modes as well as involved ‘resonant’ states. The population of these resonant states is generally higher than in the case of one photon interacting initially with the chain. However their role can be suppressed by growing the number of atoms, i.e. by passing to the limit of the infinitely long chain. It should be also noticed that the dynamics of the system excitation is revealed to be very sensitive to the field frequency and the detuning value. For negative detuning the contribution of ‘resonant’ states appear to be rather small and the excitation process is correctly described in terms of the bosonic ladder operators. In this case the probability of exciton formation dramatically exceeds the bi-exciton population. In addition, significant population is found for the system to remain in the initial unexcited state. In contrast, for zero and positive detunings the system tends to be excited with the predominant role of bi-exciton production. The population of excitons tends to be suppressed due to the destructive interference of transitions involving these states. Moreover, the contribution of the ‘resonant’ states to the bi-exciton formation increases dramatically with growing detuning and at large positive values is responsible for almost whole bi-exciton probability.
Figure 5. Simulation of 1D chain with $N = 20000$ atoms, $w = 0.25$ eV, $g \sqrt{N} = 30$ µeV interacting with 2 photons in the field. (a) Quasi-energies (green curves) depending on the detuning $\delta = \hbar \omega - \varepsilon_0$. (b) Distribution of weights of quasi-energies for different detunings $\delta$. (c) Time-dependent population of excitons (gray) and bi-excitons (green) as well as the difference between these populations and the population of correspondent collective symmetric states (red and violet respectively) obtained for negative detuning $\delta = -2g \sqrt{N} = -60$ µeV. (d) Population of different field-free excitonic (blue) and bi-excitonic states (yellow) for the same parameters averaged over first 3 nanoseconds. The abscissa shows the energy above the threshold $\varepsilon_0$ and $2\varepsilon_0$ for free exciton and bi-exciton states respectively. (e), (f) The same as (c), (d) for zero detuning $\delta = 0$. (g), (h) The same as (c), (d) for positive detuning $\delta = g \sqrt{N} = 30$ µeV.
4. Conclusions

In this work, we investigate the interaction of a quantum photon field in a high-Q resonator with nanostructures of finite size, depending on the detuning of the resonator mode with respect to the minimal exciton transition energy. Starting from the microscopic analysis we develop a fully quantum theoretical approach which describes the field-induced dynamics of Frenkel excitons in a finite size quantum well or wire modeled by a square lattice or chain of two-level atoms respectively. The dipole-dipole transfer of excitation between neighboring atoms is taken into account providing realistic dispersion law typical for many semiconductor nanosystems. The advantage of the developed approach consists in the analytical solution of the problem which allows to reveal the physical nature and mechanisms of the found effects. The eigenenergies and eigenvectors of the 'dressed' excitonic and bi-excitonic states induced by light in these multilatonic systems are found. The spatial confinement arising in large size 1D and 2D systems is demonstrated to provide energy quantization effects and discretization of exciton states that are distributed quasi-continuously. All these states are shown to play an important role in the excitation process and are explicitly taken into account in our approach.

In the frame of the developed approach, a set of novel important results is obtained and new physical effects arising due to the finite lateral size of the considered nanostructures are revealed.

The dynamics of the excitation is found to be characterized by the Rabi oscillations between the initial state and the collective symmetric excitonic mode \( \frac{1}{\sqrt{N}} R_+ |0\rangle \) which can be correctly described in terms of the bosonic ladder operators. At the same time, the enhanced excitation of high-lying free exciton states being in resonance with these ‘dressed’ polariton eigenstates is shown to be of great importance. This found new effect is referred to as the formation of Rabi-shifted resonances and appears to be the most important and new feature established for the excitation of 1D and 2D nanostructures with final lateral size. The physical mechanism of the efficient population of such resonant states and its dynamics is revealed and confirmed using a restricted three level model.

We have shown that under some conditions the excitation of these resonant states is predominant in comparison to the usually discussed polariton states. The probability to find the system in these excited ‘resonant’ states becomes significant mostly at rather large times and is very sensitive to the detuning of the field from the minimal exciton transition frequency. Substantial enhancement of the population of such states is found in the case of quantum well in comparison to the quantum wire as well as for multiple excitations in the nanosystem including the bi-exciton production. The contribution of the ‘resonant’ states to the bi-exciton formation is found dramatically large for positive detuning and appears to be the major part of the bi-exciton probability. Thus, these possible resonances between the free excitonic energies and the upper polariton branch appear to be the key point of the found new physical effects and are shown to change dramatically the dynamics of the excitation process. Nevertheless, the role of such resonant states can be significantly suppressed by growing the number of atoms in the structure, i.e. by passing to the limit of the infinitely long chain and large lattice.

It should be emphasized that the revealed new physical effects represent common features of the excitation process of nanostructures induced by quantum light. Though the obtained dynamics is observed if the dissipative processes are negligibly small, the limitation is not critical. The fastest dissipations come from the exciton-phonon interaction which is characterized by sub-picosecond times. However in the case of efficient interaction with quantum field easily achieved in high-Q micro-resonators, the found excitation takes place at more shorter times and the revealed physical effects are really observed.

The performed research and revealed physical mechanisms are very important to provide control and managing of the quantum properties and dynamics of excitons in real nanostructures and therefore can be considered as a basis for development of quantum information algorithms and protocols involving manifold excitations in nanostructures. The predicted effects, possible to observe in real nanostructured systems, especially organic semiconducting nanowires and nanowells (nanoplatelets), will stimulate future planned and carried out experiments. These results can be also interesting for developing of single-photon optoelectronics.

Data availability statement

Any data that support the findings of this study are included within the article.

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Appendix A. Details of analytical calculation in 1D case

The diagonalization of the Hamiltonian in terms of this basis leads to the zero value of the following determinant:

\[
\begin{vmatrix}
\hbar \omega - \lambda & g\gamma_1 & \cdots & g\gamma_N \\
g\gamma_1 & \varepsilon_1 - \lambda & 0 & 0 \\
\vdots & 0 & \ddots & 0 \\
g\gamma_N & 0 & 0 & \varepsilon_N - \lambda
\end{vmatrix} = 0,
\]

(A1)

where \( \varepsilon_k \) and \( \gamma_k \) with only odd indexes are included. This Hamiltonian matrix has \((M + 1) \times (M + 1)\) dimensions, where \( M \) is the total number of ‘active’ excitonic states. In the one-dimensional case, \( M = \lfloor (N+1)/2 \rfloor \). The secular equation
which determines the quasi-energies of quantum polariton or the energies of exciton dressed by one-photon quantum field, can be written in the form (10) from the main text.

For large \( N \) one can obtain the oscillator strength \( f_k = \frac{4}{\pi \varepsilon_k^2} (N + 1 + o \left( \frac{1}{N} \right)) \), and the dipole matrix element \( \gamma_k = \frac{2}{\pi \varepsilon_k^2} \sqrt{N + 1 + o \left( \frac{1}{N} \right)} \) for odd \( k \). This result can be obtained by replacing the sum in equation (7) by the integral:

\[
\gamma_k \approx \sqrt{2(N+1)} \int_0^1 \sin(\pi k x) dx = \frac{\sqrt{2(N+1)} (1 - (-1)^k)}{\pi k}.
\]

(A2)

This dependence of the matrix element on the quantum number of the exciton state is due to the fact that the law of conservation of wavevector \( q = \pi k / d (N + 1) \approx \pi k / L \) in the interaction of a photon with a zero wavevector with a system with finite linear size \( L \) is satisfied with an accuracy of \( \Delta q \approx 1 / L \).

For a large number of atoms, one can use equations (4) and (A2) and use the relation

\[
\sum_{j=1}^{\infty} \frac{1}{(2j-1)^2} = \frac{\pi}{x^{3/2}}
\]

(A3)

Then, introducing dimensionless variables \( x = (\lambda - \varepsilon_0)^{N+1/2} / \sqrt{\pi} \), \( \delta = (\omega - \varepsilon_0)^{N+1/2} / \sqrt{\pi} \), one can obtain a transcendental equation for determining the dimensionless eigenvalues

\[
(\delta - x) = \beta \left( \frac{2 \tan(\pi \sqrt{x} / 2) - \pi \sqrt{x}}{\pi x^{3/2}} \right), \beta \equiv g^2 (N+1)^5 / \omega^2 \pi^4
\]

(A4)

for positive \( x > 0 \) and

\[
(\delta - x) = -\beta \left( \frac{2 \tanh(\pi \sqrt{-x} / 2) - \pi \sqrt{-x}}{\pi (-x)^{3/2}} \right).
\]

(A5)

for negative \( x < 0 \). The poles of the function \( G^{(1)}(x) \) correspond to the squares of odd numbers, and all the eigenvalues \( x_m \) are between them. The exception is the first eigenvalue \( x_0 \), which can be positive less than one or negative. It can be obtained from equation (A5) that for \( \beta > 1 \) the negative dimensionless quasi-energy \( x_0 \) satisfies the equation

\[
(\delta + |x_0|) = \beta \left( \frac{1}{|x_0|} - \frac{2}{\pi |x_0|^{3/2}} \right).
\]

(A6)

In case \( |x_0| > 1 \) we get

\[
|x_0| \approx \frac{1}{2} \left( -\delta - \frac{2 \beta^{1/4}}{\pi} + \sqrt{4 \beta + \left( \delta + \frac{2 \beta^{1/4}}{\pi} \right)^2} \right)
\]

\[
\approx \frac{1}{2} \left( -\delta + \sqrt{4 \beta + \delta^2} \right).
\]

(A7)

or \( \lambda_0 \approx \frac{1}{\pi} \left( \varepsilon + \hbar \omega - \sqrt{4 g^2 (N+1) + (\varepsilon - \hbar \omega)^2} \right) \). This is the fundamental oscillation frequency of the population of the photonic mode, taking into account the Rabi frequency. The positive values of the dimensionless quasi-energies are determined by the relation

\[
x_m = \left( 2m + \frac{2}{\pi} \arctan \left( \frac{\pi (2m)^3}{2 \beta} \left( \delta - 4m^2 + \frac{\beta}{4m^2} \right) \right) \right)^2.
\]

(A8)

The dimensionless quasi-energy \( x_m \) is slightly lower than \( (2m + 1)^2 \) for numbers \( m < \pi |x_0| \), otherwise it is slightly higher \( (2m - 1)^2 \). The quasi-energy \( x_m \) is close to \( 4m^2 \) in the case of equality \( m = \sqrt{|x_0|} / 2 \).

\[
P_m \equiv 1 + g^2 \sum_{j=1}^{M} \frac{\gamma_j^2}{(\varepsilon_j - \lambda)^2} = 1 + g^2 \frac{dG^{(1)}(\lambda)}{d\lambda} \bigg|_{\lambda=\lambda_m}.
\]

(A9)

One can calculate \( P_m \) for large \( M \) using the relation

\[
\sum_{j=1}^{\infty} \frac{1}{(2j-1)^2} = \pi \right)

(A10)

Using the eigenvalue equations (A4) and (A5), it can be shown that \( P_m^{-1} \) equals to (for all \( x_m \), including negative \( x_0 \))

\[
P_m^{-1} = \frac{8 \beta x_m}{\pi^2 (\beta - x_m (x_m - \delta))^2 + 4 \beta (5x_m - 3\delta)}.
\]

(A11)

It is evident from equation (11) that the amplitude of photon component in the eigenfunction equals to \( P_m^{-1/2} \). When \( \beta \) is large, the maximum value of \( P_m^{-1/2} \) is reached in the region where the denominator is minimal, that is, at points near roots of \( (\beta - x_m (x_m - \delta))^2 = 0 \). These roots equal to \( x_0 = \left( \delta - \sqrt{4 \beta + \delta^2} \right) / 2 \) for one (positive root) and a group of quasi-energies around \( x_{max} = \left( \delta + \sqrt{4 \beta + \delta^2} \right) / 2 \) with width about \( \Delta x \approx \sqrt{3 / \beta^{1/4} / \pi} \) (quasi-resonance with account for Rabi frequency). Negative root \( x_0 \) corresponds to lower polariton state (see figure (1a)). At zero detuning the weight of phonon state in lower polariton state equals to \( P_0^{-1} = 1 / 2 \) (taking into account the terms of the order \( \beta^{1/4} / \pi \) in equation (A7)). For positive detuning \( |\delta| > 2 \sqrt{\beta} / \beta \) this weight is its contribution equals to \( P_0^{-1} \approx 1 - 4 \beta / \delta^2 \). Thus, the eigenfunction with index zero describes the photon-like state of the polariton. For large positive detuning, the weight of photon component becomes small \( P_0^{-1} \approx 4 \beta / \delta^2 \) and lower polariton branch has large exciton component.

The structure of exciton component in lower polariton eigenstate can be estimated from equation (11) if we take into
account that for large $\beta$ and $m = 0$ we can write $\varepsilon_j - \lambda_0$ in the denominators of all exciton components. In this case

$$\hat{H}_0 = P_{m}^{-1/2} \left\{ 1, \ldots, \frac{\sqrt{\beta}}{|\lambda_0|} \gamma_j, \ldots \right\}^T \tag{A12}$$

and we can introduce full-symmetry state as the sum of $|1, 2j - 1\rangle$ with weights $\gamma_j$:

$$\sum_{k=1}^{N} \gamma_k |1, k\rangle = \frac{1}{\sqrt{N}} \sum_{m=1}^{N} \left( \sum_{k=1}^{N} \frac{2}{N+1} \cos \left( \frac{\pi k}{N+1} \right) \sin \left( \frac{\pi k N}{N+1} \right) \right) a_0^\dagger |0\rangle = \frac{1}{\sqrt{V}} \sum_{m=1}^{N} a_0^\dagger |0\rangle = R^+ |0\rangle \tag{A13}$$

(with account for $\sum_{m=1}^{\infty} |1, k\rangle \sin \left( \frac{\pi k N}{N+1} \right) = 1$ for $1 \leq n \leq N$, $N >> 1$). It can be checked that equation (A13) is correct for all $N$. The state $(N)^{-1/2} R^+ |0\rangle$ describes the collective excitation of all atoms in the chain with equal probability and the same phase.

The maximum of $P_m^{-1/2}$ for positive $m$ corresponds to upper polariton state. This group corresponds to the exciton levels with number $j$ around $j_{max} = 1 / 2(\sqrt{1 + 2/\beta} - 1/\beta)^{1/2}$, and for $\beta^{1/4} >> 1$ only one or two levels participate in formation of the maximum of $P_m^{-1/2}$ for upper polariton solution. The structure of this state is more complex than for lower polariton state, since we cannot neglect the resonance exciton state when $\varepsilon_j$ is close to $\lambda_0$. Therefore, upper polariton is a mixture of a photon state (with weight $\sum_{m=1}^{\infty} P_m^{-1} = 1 - P_0^{-1}$, $R^+ |0\rangle$ and one or two exciton states with $k_{max} = 2 j_{max} - 1$ which are in quasi-resonance with the frequency of upper polariton state.

**Appendix B. Excitons in 3D case**

Let us consider 3D simple orthorhombic crystal lattice in approximation of hoping only to the nearest neighbors from the first coordination sphere. In this case the Hamiltonian equals to

$$\hat{H}_0 = \sum_{n \in V} \varepsilon_n a^\dagger_n a_n - \sum_{n, \Delta \in V} \sum_{s=\{x, y, z\}} w_{n, \Delta} a^\dagger_{n+s} a_{n+\Delta, s}$$

$$\Delta = \{x, -x, y, -y, z, -z\}, \tag{B1}$$

where $x, y, z$—unit vectors along the directions of lattice translations, $n = (n_x, n_y, n_z)$—coordinates of nodes in the volume $V = [1, \ldots, N_x] \times [1, \ldots, N_y] \times [1, \ldots, N_z]$, $s = \{x, y, z\}$—three possible directions of dipole momenta of an excited atom, $w_{n, \Delta}$—matrix elements for transfer of the excitation of the atom in node $n$ with dipole momentum in direction $\Delta$ to the neighbor atom located in the node $n + \Delta$. If we consider only one polarization of the photon field directed along one of the crystal axis (we assume this restriction below), the system will be characterized by only three values of matrix elements $\{w_{1, x}, w_{1, y}, w_{1, z}\}$. In case of cubic crystal these values equal to $w_{xy}$ or $w_{xz}$ depending on the direction of the electric field of the photon mode. Crystals with more complicated crystal structure (e.g. face-centered cubic lattice) the components appears which mix different directions of the dipole momentum. In case if the number of atoms in one direction is small ($N_x << N_y, N_z << N_x$), such system describes the quantum well of finite size.

Single exciton wavefunctions for the system under consideration are characterized by three quantum numbers $k_i = 1, \ldots, N_i, \ k_i = 1, \ldots, N_i, \ k_i = 1, \ldots, N_i$, which form the integer-valued vector $k$ and can be represented as

$$|1, k\rangle = \sum_{n \in V} N_x N_y N_z \lambda_{n,k} \lambda_{n,k} \lambda_{n,k} a_0^\dagger |n, \Delta\rangle, \tag{B2}$$

with the corresponding energies

$$E_{1, k} = \varepsilon - 2 w_x \cos \left( \frac{\pi k_x}{N_x + 1} \right) - 2 w_y \cos \left( \frac{\pi k_y}{N_y + 1} \right) - 2 w_z \cos \left( \frac{\pi k_z}{N_z + 1} \right). \tag{B3}$$

The total number of single-exciton states with definite direction of dipole momentum equals to $N_x N_y N_z$.

In the same manner $m$-exciton states are characterized by the set of quantum numbers $k_i = (k_{ix}, k_{iy}, k_{iz}), \ i = 1 \ldots m$. In order to exclude the appearance of the equivalent sets, these vectors should be ordered in some way, for instance in the incremental order of the function $\text{Ord} (k) = k_x + N_x (k_y - 1) + N_x N_y (k_z - 1)$: $\text{Ord}(k_1) < \cdots < \text{Ord}(k_m) < \text{Ord}(k_0)$. The total number of such states with account for indistinguishability of excitons equals to $\binom{(N_x N_y N_z)^m}{m}$, and their energy is

$$E_{m, k_1, \ldots, k_m} = m \varepsilon - 2 w_x \cos \left( \frac{\pi k_{ix}}{N_x + 1} \right) + \cdots + \cos \left( \frac{\pi k_{mx}}{N_x + 1} \right)$$

$$- 2 w_y \cos \left( \frac{\pi k_{iy}}{N_y + 1} \right) + \cdots + \cos \left( \frac{\pi k_{my}}{N_y + 1} \right)$$

$$- 2 w_z \cos \left( \frac{\pi k_{iz}}{N_z + 1} \right) + \cdots + \cos \left( \frac{\pi k_{mez}}{N_z + 1} \right). \tag{B4}$$

**Wavefunctions for these states can be written as**

$$|m, k_1, \ldots, k_m\rangle = \sum_{n_1 \in V} \cdots \sum_{n_m \in V} \psi(n_1, \ldots, n_m; k_1, \ldots, k_m)$$

$$\times a_{n_1}^\dagger \cdots a_{n_m}^\dagger |0\rangle. \tag{B5}$$
where Slater determinant is based on the products $\chi_{n_1,k_1}^N \chi_{n_2,k_2}^N \ldots \chi_{n_m,k_m}^N$

$$
\psi(n_1 \ldots n_m, k_1 \ldots k_m) = \begin{vmatrix}
\chi_{n_1,k_1}^N & \chi_{n_1,k_2}^N & \ldots & \chi_{n_1,k_m}^N \\
\chi_{n_2,k_1}^N & \chi_{n_2,k_2}^N & \ldots & \chi_{n_2,k_m}^N \\
\vdots & \vdots & \ddots & \vdots \\
\chi_{n_m,k_1}^N & \chi_{n_m,k_2}^N & \ldots & \chi_{n_m,k_m}^N
\end{vmatrix}.
$$ (B6)

In 3D case matrix elements for transition between vacuum state $|0\rangle$ to a one-exciton state equals to

$$
\gamma(k_i,k_j) = \sum_{n \in Y} \chi_{n,k_i}^N \chi_{n,k_j}^N \chi_{n,k_m}^N
$$

$$
= \sqrt{\frac{(1-(−1)^k_N)(1-(−1)^k_N)(1-(−1)^k_N)}{(N_+ + 1)(N_+ + 1)(N_+ + 1)}}
\times \cot \left( \frac{\pi k_1}{2(N_+ + 1)} \right) \cot \left( \frac{\pi k_i}{2(N_+ + 1)} \right)
\times \cot \left( \frac{\pi k_i}{2(N_+ + 1)} \right)
$$ (B7)

and non-zero in the case when all quantum numbers $(k_i, k_j, k_m)$ are odd.

**Appendix C. Multi-exciton states in 1D case. Bi-exciton matrix elements**

The field-free solution (2), presented in the main text, can be easily found by periodic extension of the chain with inserting the $(N+1)$th atom, which cannot receive any excitation.

The energy of the state with $m$ excitons equals to

$$
E_{m,k_1,\ldots,k_m} = m \varepsilon - 2w \left( \cos \frac{\pi k_1}{N+1} + \ldots + \cos \frac{\pi k_m}{N+1} \right),
$$

$$
k_1 = 1, \ldots, N - m + 1, \quad k_2 = k_1 + 1, \ldots, N - m + 2, \ldots,
$$

$$
k_m = k_{m-1} + 1, \ldots, N.
$$ (C1)

The total number of such states is $\frac{N!}{m!(N-m)!}$, and their wave-functions can be written as

$$
|m,k_1,\ldots,k_m\rangle = \sum_{n_1=1}^{N-m} \ldots \sum_{n_m=k_{m-1}+1}^{N} \psi(n_1 \ldots n_m, k_1 \ldots k_m) a_{n_1}^\dagger \ldots a_{n_m}^\dagger |0\rangle,
$$

$$
k_1 = 1, \ldots, N - m + 1, \quad k_2 = k_1 + 1, \ldots, N - m + 2, \ldots,
$$

$$
k_m = k_{m-1} + 1, \ldots, N.
$$ (C2)

where Slater determinant is

$$
\psi(n_1 \ldots n_m, k_1 \ldots k_m) = \begin{vmatrix}
\chi_{n_1,k_1}^N & \chi_{n_1,k_2}^N & \ldots & \chi_{n_1,k_m}^N \\
\chi_{n_2,k_1}^N & \chi_{n_2,k_2}^N & \ldots & \chi_{n_2,k_m}^N \\
\vdots & \vdots & \ddots & \vdots \\
\chi_{n_m,k_1}^N & \chi_{n_m,k_2}^N & \ldots & \chi_{n_m,k_m}^N
\end{vmatrix}.
$$ (C3)

(please note the absence of the factor $1/\sqrt{m!}$ since the sum in (C3) is limited by the region with ordered values $n_1 < n_2 < \ldots < n_m$).

For $m = 2$ the wave functions can be written as

$$
|2, k_1, k_2 \rangle = \sum_{n_1=1}^{N-1} \sum_{n_2=n_1+1}^{N} \left( \chi_{n_1,k_1}^N \chi_{n_2,k_2}^N - \chi_{n_2,k_2}^N \chi_{n_1,k_1}^N \right) a_{n_1}^\dagger a_{n_2}^\dagger |0\rangle,
$$

$$
k_1 = 1, \ldots, N - 1, \quad k_2 = k_1 + 1, \ldots, N.
$$ (C4)

Again, due to the indistinguishability of excitons, the summation is carried out for $n_2 > n_1$.

The states of the non-interacting basis of a photon mode and 1D chain can be written as $|n;0\rangle = (b^\dagger)^m |0\rangle$, $|m-1; 1, k\rangle = (b^\dagger)^{m-1} |1, k\rangle$, $|m-2; 2, k_1, k_2\rangle = (b^\dagger)^{m-2} |2, k_1, k_2\rangle$ etc. The excitons are indistinguishable, therefore $k_1 < k_2$ in the bi-exciton state.

Absorption of photons can result in transition from state with $m-1$ excitons to state with $m$ excitons. In case of two photons in initial state there are transitions from non-excited state of the chain to single-exciton state, and then from single-exciton state to bi-exciton state. Matrix elements for the latter transitions can be evaluated analytically for long 1D chain ($N >> 1$) using the replacement of the sums by the integrals:

$$
\langle 1, k | b R^\dagger + b^\dagger R \mid 2, k_1, k_2 \rangle
$$

$$
= \sqrt{2(N+1)} \left( \frac{\pi}{1 + (-1)^{k_1}} \right) \delta_{k,k_1} + \frac{1 + (-1)^{k_2}}{k_2} \delta_{k,k_2}
$$

$$
+ \left( \frac{1}{k_2} - \frac{1}{k_1} \right) \delta_{k,k_1+k_2} + \frac{1}{k_2} + \frac{1}{k_1} \delta_{k,k_1-k_2}
$$

$$
- \left( \frac{1}{k_2} + \frac{1}{k_1} \right) \delta_{k,-k_1+k_2}.
$$ (C5)

The last but one term disappears since $k_1 < k_2$. So, the transitions from ‘active’ exciton states with odd $k$ (which can be created by a longitudinal photon from the ground state of the chain) can occur only to bi-exciton states with odd $k_1 + k_2$.
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