Dark Bogolon-Excitons in a Linear Atomic Super-Lattice

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Dark and bright excitons are shown to appear naturally in a linear atomic super-lattice with two atoms per unit cell. In bringing the super-lattice into a strong coupling regime with a one-dimensional nanophotonic waveguide, bright excitons and photons are coherently mixed to form polaritons. Treating excitons as bosons implies a mechanism that forbids two excitations from being at the same atomic state, which is included here through a bosonization procedure with kinematic interactions. Interestingly these interactions couple dark and bright excitons, and which we exploit as a new tool for exciting dark states in a controllable way. We suggest a pump-probe experiment where two polaritons scatter into two dark excitons that found to be correlated and are represented as dark bogolon-excitons. The results can be adapted for any super-lattice of active materials, e.g., of organic molecules.

I. INTRODUCTION

Collective electronic excitations (excitons) have been a matter of research since their introduction by Frenkel in 1931 [1]. Frenkel excitons are of importance for fundamental physics, as they explain various electrical and optical properties in molecular crystals, e.g. in organic and noble atom crystals [2, 3]. These type of excitations have been extensively investigated and found applications for optoelectronic devices [4, 5]. The mechanism behind their appearance is through the delocalization of electronic excitations among the crystal atoms or molecules via electrostatic interactions, mainly resonant dipole-dipole interactions, and in exploiting the lattice symmetry. Such coherent states can appear spontaneously among electronic states of a huge number of atoms without the aid of external fields. Just to mention that coherence among electronic states inside the same atom implies external laser fields, as for electromagnetic induced transparency and population trapping [6].

The progress in the fabrication of optical lattices provides artificial crystals with controllable parameters [7, 8], which are adopted to simulate a wide range of complex condensed matter models and present a deep understanding of puzzling solid state effects [9]. Of importance here is ultracold atoms in optical lattices at the Mott insulator phase with one atom per site, which have been proposed for simulating Frenkel excitons and unveiled physical phenomena that are masked by thermal excitations and impurities in solid crystals [10, 11].

On the other hand optical lattices have been introduced as active material for cavity QED [10]. It was shown that electronic excitations and cavity photons in the strong coupling regime are naturally mixed to form polaritons as the system collective excitations [11]. Several configurations of optical lattices within a cavity have been suggested with diverse dimensionality and geometry [12, 13]. Of special interest is that of a one dimensional optical lattice formed by a nanophotonic waveguide [14, 15], which was realized for cesium atoms in using

tapered nanofibers [16, 17]. The optical lattice is localized outside and parallel to the nanofiber by evanescent fields of counter propagating colored fiber beams. The atomic lattice can be directly interrogated by a resonant beam that sent through the fiber and observed via linear optical spectra [18].

In the present paper we address an optical super-lattice with two sites per unit cell and that includes one atom per site, where the distance between the two on cell atoms is taken to be smaller than the distance between the centers of two neighbor unit cells. This configuration gives rise to symmetric and antisymmetric entangled states, in which an electronic excitation can be delocalized among the two atoms inside the same unit cell. The symmetric states are bright and superradiant with damping rate of twice that of a single excited atoms. While the antisymmetric states are dark with zero damping rate and decouple to the light [19]. It is a big challenge to excite dark states in an efficient and controllable way. In localizing the super-lattice parallel to a nanophotonic waveguide at a fixed distance, only the symmetric excitons coherently couple to the fiber photons in order to form one dimensional polaritons.

Excitons in an extended system behave as bosons, but then they acquire a mechanism that forbids double excitations at the same atom [20, 21]. In previous work we presented a bosonization procedure that transform two-level atoms into bosons with additional kinematic interactions that forbid two excitations from being localized on the same atom, and which found to be a good approximation at low intensity of excitations [22]. Here we apply such a procedure to optical super-lattices, and we interestingly show that kinematic interactions can couple symmetric and antisymmetric states. We exploit this mechanism as a controllable mean for exciting dark excitons. In order to achieve our goal we proposed a pump-probe experiment, where two pumped polaritons with momentum k can be scattering into two dark excitons of momenta $k + p$ and $k - p$. This parametric amplification of dark states is induced by a probe field at either

$k + p$ or $k - p$ momenta, and subjected to conservation of energy. The scattering of two polaritons into a state of two excited atoms in the same unit cell can be excluded due to conservation of energy. In the frame of Bogoliubov mean field theory the dark exciton pairs are strongly correlated and can be represented by bogolon excitations that termed here bogolon-excitons [23].

The paper is organized as follows. In section 2 we introduce dark and bright excitons into an optical super-lattice. An atomic super-lattice strongly coupled to a nanophotonic waveguide is presented in section 3 by using the language of polaritons. Kinematic interactions are derived in section 4 with emphasize on the symmetric-antisymmetric mixing. Pump-probe experiments appear in section 5 where a process for exciting dark states is proposed. Section 6 includes Bogoliubov mean field theory with dark bogolon-excitons. A summary is given in section 7.

II. COLLECTIVE EXCITATIONS IN LINEAR SUPER-LATTICES

We consider a system of one-dimensional optical lattice with a basis of two sites (super-lattice). Namely, we have a lattice with unit cell of two trapping minima, as seen in figure (1). The lattice constant is a, which is the distance between the center of two neighbor unit cells, and the distance between the two sites within the same unit cell is R. The optical super-lattice can be realized in using counter propagating laser beams to get a standing wave super-lattice [14, 15]. Ultracold or cold atoms are loaded into such a system, where the atoms experience optical super-lattice potential. We treat the case of one atom per site with two atoms per unit cell. Here we concentrate in a system of nanophotonic waveguide, as beside the off-resonance light used to produce the super-lattice it provides a strong resonant coupling of light to the atomic super-lattice. The recent set-up of tapered nanofibers have the potential for realizing such a system [16, 17].
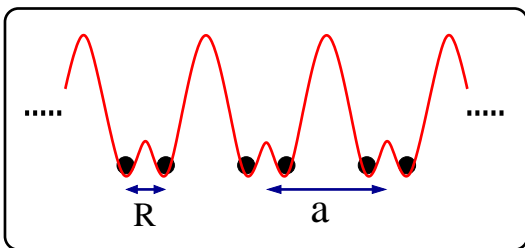

FIG. 1: Optical super-lattice with two sites per unit cell and one atom per site. The on-site atoms are separated by R, and neighbor unit cells by a.

The atoms are taken to be of two-level systems with atomic transition energy E_A. In using the second quan-

tization language, the excitation Hamiltonian reads

$$H_{ex} = E_A \sum_{n,\alpha} S_n^{\alpha\dagger} S_n^{\alpha} + \sum_{nm,\alpha\beta} J_{nm}^{\alpha\beta} \; S_n^{\alpha\dagger} S_m^{\beta}, \quad (1)$$

where $S_n^{\alpha\dagger}$ and S_n^{α} are the creation and annihilation operators of an electronic excitation at atom α in site n, respectively. Here $(\alpha, \beta = 1, 2)$ stand for the two atoms at the same unit cell, and $(n, m = 0, \pm1, \cdots, \pm M)$ for the summation over the super-lattice sites, with $N = 2M + 1$ the number of unit cells.

The Hamiltonian includes resonant dipole-dipole interactions between the atoms that allow for energy transfer, in which an excited atom decays to the ground state and other atom is excited. The interaction parameter among α and β atoms at sites n and m is $J_{nm}^{\alpha\beta}$, which is

$$J_{nm}^{\alpha\beta} = \frac{\mu^2}{4\pi\epsilon_0 (R_{nm}^{\alpha\beta})^3} \left(1 - 3\cos^2\theta\right), \quad (2)$$

where μ is the transition dipole that has an angle θ with the super-lattice axis. The interatomic distance is $R_{nm}^{\alpha\beta} = |R_n^{\alpha} - R_m^{\beta}|$, where R_n^{α} is the position of atom (n, α) in the super-lattice.

We write the Hamiltonian as a sum of two terms, the first includes interactions among atoms in the same unit cell, and the second among different unit cells. We can write $H_{ex} = \sum_n H_n + \sum'_{nm} H_{nm}$, where

$$H_n = E_A \sum_{\alpha} S_n^{\alpha\dagger} S_n^{\alpha} + \sum_{\alpha\beta}' J_n^{\alpha\beta} \; S_n^{\alpha\dagger} S_n^{\beta}, \quad (3)$$

and

$$H_{nm} = \sum_{\alpha\beta} J_{nm}^{\alpha\beta} \; S_n^{\alpha\dagger} S_m^{\beta}. \quad (4)$$

The primes indicate that in the summations we have $(n \neq m)$ and $(\alpha \neq \beta)$.

The H_n Hamiltonian can be diagonalized in using the transformation

$$S_n^s = \frac{S_n^1 + S_n^2}{\sqrt{2}}, \; S_n^a = \frac{S_n^1 - S_n^2}{\sqrt{2}}, \quad (5)$$

which yields

$$H_n = E_s \; S_n^{s\dagger} S_n^s + E_a \; S_n^{a\dagger} S_n^a, \quad (6)$$

where $E_s = E_A + J_0$ and $E_a = E_A - J_0$, with

$$J_0 = \frac{\mu^2}{4\pi\epsilon_0 R^3} \left(1 - 3\cos^2\theta\right). \quad (7)$$

Note that due to the dependence of J_0 on θ, the polarization direction can provide a control parameter for the symmetric-antisymmetric splitting energy.

In using the above transformation into symmetric and antisymmetric operators in H_{nm}, we get explicitly

$$
\begin{aligned}
H_{nm} = \frac{1}{2} \Big\{ &\left(J_{nm}^{11} + J_{nm}^{22} + J_{nm}^{12} + J_{nm}^{21} \right) \ S_n^{s\dagger} S_m^{s} \\
+ &\left(J_{nm}^{11} + J_{nm}^{22} - J_{nm}^{12} - J_{nm}^{21} \right) \ S_n^{a\dagger} S_m^{a} \\
+ &\left(J_{nm}^{11} - J_{nm}^{22} - J_{nm}^{12} + J_{nm}^{21} \right) \ S_n^{s\dagger} S_m^{a} \\
+ &\left(J_{nm}^{11} - J_{nm}^{22} + J_{nm}^{12} - J_{nm}^{21} \right) \ S_n^{a\dagger} S_m^{s} \Big\}.
\end{aligned} \quad (8)
$$

Let us assume interactions only among nearest neighbor unit cells, which is a good assumption in optical lattices. Between the n and $n+1$ sites we have

$$
\begin{aligned}
J_{n(n+1)}^{11} &= J_{n(n+1)}^{22} = \frac{\mu^2}{4\pi\epsilon_0 a^3} \left(1 - 3\cos^2\theta \right), \\
J_{n(n+1)}^{12} &= \frac{\mu^2}{4\pi\epsilon_0 (a+R)^3} \left(1 - 3\cos^2\theta \right), \\
J_{n(n+1)}^{21} &= \frac{\mu^2}{4\pi\epsilon_0 (a-R)^3} \left(1 - 3\cos^2\theta \right).
\end{aligned} \quad (9)
$$

We assume now the limit of $a \gg R$, then $H_{nm} \approx 2J \ S_n^{s\dagger} S_m^{s}$, where

$$
J = \frac{\mu^2}{4\pi\epsilon_0 a^3} \left(1 - 3\cos^2\theta \right). \quad (10)
$$

The antisymmetric states are on-site localized, and the mixing terms between the symmetric and antisymmetric states vanish. Only the symmetric states can be delocalized among the supper-lattice sites.

In the limit of $R \ll a$, the Hamiltonian reads

$$
H_{ex} = \sum_n \left\{ E_s \ S_n^{s\dagger} S_n^{s} + E_a \ S_n^{a\dagger} S_n^{a} \right\} + {\sum_{nm}}' 2J \ S_n^{s\dagger} S_m^{s}. \quad (11)
$$

At this point we aim to diagonalize the Hamiltonian relative to the site indices, in applying the transformation

$$
S_n^{\nu} = \frac{1}{\sqrt{N}} \sum_k e^{-ikz_n} S_k^{\nu}, \ (\nu = s, a), \quad (12)
$$

where the position of the unit cell center is $z_n = an$, and the wavenumber k takes the values $k = \frac{2\pi}{Na}p$ with $p = 0, \pm 1, \cdots, \pm M$. Here, an electronic excitation is delocalized in the lattice where it has the same probability to appear at every lattice site. We get

$$
H_{ex} = \sum_k E_s(k) \ S_k^{s\dagger} S_k^{s} + E_a \sum_k S_k^{a\dagger} S_k^{a}, \quad (13)
$$

which contains symmetric and antisymmetric excitons. The antisymmetric states are dispersion-less with the energy $E_a = E_A - J_0$. The symmetric states have the dispersion relation $E_s(k) = E_s + J(k)$, where $J(k) = 2J \sum_z e^{ikz}$. For nearest neighbor interactions we obtain $J(k) = 4J \cos(ka)$, and the symmetric dispersion now reads $E_s(k) = E_A + J_0 + 4J \cos(ka)$. In place of the discrete energy levels we get an energy band. Excitons

are collective electronic excitations that behave as quasi-particles [2, 3]. The combination of electrostatic interactions and lattice symmetry gives rise to energy waves that propagate with wave numbers that are good quantum numbers.

III. SUPER-LATTICE EXCITATIONS COUPLED TO PHOTONS

The optical supper-lattice is localized parallel to the nano-photonic waveguide at a distance b, as in figure (2). We consider one-dimensional propagating photons that are represented by the Hamiltonian

$$
H_{ph} = \sum_q \ E_{ph}(q) \ a_q^\dagger a_q, \quad (14)
$$

where $a_q^\dagger$ and a_q are the creation and annihilation operators of a photon of mode q, respectively. The wave number q takes the values $q = 2\pi l/L$, where $l = 0, \pm 1, \pm 2, \cdots, \pm\infty$, and L is the waveguide length. The photon dispersion can be given by

$$
E_{ph}(q) = \frac{\hbar c}{\sqrt{\epsilon}} \sqrt{q_0^2 + q^2}, \quad (15)
$$

where ϵ is an effective dielectric constant, and q_0 is the confinement wave number.
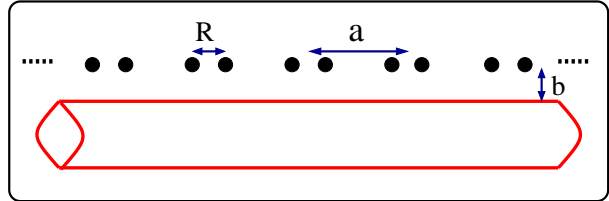

FIG. 2: The optical super-lattice is localized parallel to a nano-photonic wave guide at distance b.

The excitation-photon coupling in the electric dipole approximation is written as $H_I = -\hat{\boldsymbol{\mu}} \cdot \hat{E}$, where the excitation transition dipole operator is given by $\hat{\boldsymbol{\mu}} = \boldsymbol{\mu} \sum_{n,\alpha} \left(S_n^\alpha + S_n^{\alpha\dagger} \right)$, and $\boldsymbol{\mu}$ is the transition dipole.

The photon electric field operator is given by

$$
\hat{E}(r,z) = i \sum_q \sqrt{\frac{E_{ph}(q)}{2\epsilon_0 V}} \mathbf{e} u(r) \left(a_q e^{-iqz} - a_q^\dagger e^{iqz} \right), \quad (16)
$$

where V is the photon effective volume, $\mathbf{e}$ is the photon unit vector polarization, and $u(r)$ is the photon mode function.

The interaction Hamiltonian in the rotating wave approximation, and in the Schrödinger picture, reads

$$
H_I = \sum_{qn,\alpha} \left(f_{qn}^\alpha \ a_q S_n^{\alpha\dagger} + f_{qn}^{\alpha*} \ S_n^\alpha a_q^\dagger \right), \quad (17)
$$

where

$$f_{qn}^\alpha = -i\sqrt{\frac{E_{ph}(q)}{2\epsilon_0 V}} u(b)(\boldsymbol{\mu}\cdot\mathbf{e}) e^{-iqz_n^\alpha}. \qquad (18)$$

The electric field is evaluated at the atom positions with $u(b)$ the mode function at the lattice position. Here z_n^α is the position of atom α at site n, where we write $z_n^1 = z_n - \frac{R}{2}$ and $z_n^2 = z_n + \frac{R}{2}$. In terms of symmetric and antisymmetric operators, we get

$$H_I = \sum_{qn,\nu} \left(f_{qn}^\nu \, a_q S_n^{\nu\dagger} + f_{qn}^{\nu*} \, S_n^\nu a_q^\dagger \right), \qquad (19)$$

with the coupling parameters

$$f_{qn}^s = -i\sqrt{\frac{E_{ph}(q)}{\epsilon_0 V}} u(b)(\boldsymbol{\mu}\cdot\mathbf{e}) \left\{ \frac{e^{-iqz_n^1} + e^{-iqz_n^2}}{2} \right\},$$

$$f_{qn}^a = -i\sqrt{\frac{E_{ph}(q)}{\epsilon_0 V}} u(b)(\boldsymbol{\mu}\cdot\mathbf{e}) \left\{ \frac{e^{-iqz_n^1} - e^{-iqz_n^2}}{2} \right\} \qquad (20)$$

In momentum space we have

$$H_I = \sum_{k,\nu} \left(f_k^\nu \, a_k S_k^{\nu\dagger} + f_k^{\nu*} \, S_k^\nu a_k^\dagger \right), \qquad (21)$$

where

$$f_k^s = -i\sqrt{\frac{E_{ph}(k)N}{\epsilon_0 V}} u(b)(\boldsymbol{\mu}\cdot\mathbf{e}) \cos(kR/2),$$

$$f_k^a = \sqrt{\frac{E_{ph}(k)N}{\epsilon_0 V}} u(b)(\boldsymbol{\mu}\cdot\mathbf{e}) \sin(kR/2). \qquad (22)$$

The interatomic distance R can provide a significant control parameter for the interaction of the photons to the symmetric and antisymmetric excitons. This mechanism can be used to exchange the states from dark into bright and vice versa.

The total Hamiltonian now reads

$$H = \sum_k \left\{ E_{ph}(k) \, a_k^\dagger a_k + E_s(k) \, S_k^{s\dagger} S_k^s + E_a \, S_k^{a\dagger} S_k^a \right.$$

$$\left. + \sum_\nu \left(f_k^\nu \, a_k S_k^{\nu\dagger} + f_k^{\nu*} \, S_k^\nu a_k^\dagger \right) \right\}. \qquad (23)$$

Due to translational symmetry the Hamiltonian is separated in momentum space, and the interaction is between excitons and photons with the same wave number.

In the limit of small k, that is $kR \ll 1$, we have

$$f_k^s \approx -i\sqrt{\frac{E_{ph}(k)}{\epsilon_0 \bar{S} a}} u(b)\mu, \;\; f_k^a \approx \sqrt{\frac{E_{ph}(k)}{\epsilon_0 \bar{S} a}} u(b)\mu \frac{kR}{2}, \quad (24)$$

where $\mu = (\boldsymbol{\mu}\cdot\mathbf{e})$. We used $V = \bar{S}Na$ and $L = Na$, with $\bar{S}$ the photon effective cross section. The coupling

of symmetric excitons to the photons is much larger than the antisymmetric ones.

In neglecting the antisymmetric weak coupling part, the Hamiltonian is written as

$$H = \sum_k \left\{ E_{ph}(k) \, a_k^\dagger a_k + E_s(k) \, S_k^{s\dagger} S_k^s + E_a \, S_k^{a\dagger} S_k^a \right.$$

$$\left. + f_k^s \, a_k S_k^{s\dagger} + f_k^{s*} \, S_k^s a_k^\dagger \right\}. \qquad (25)$$

The antisymmetric excitons are dark and decouple to the photons, while the symmetric excitons are bright and coupled to the photons. Moreover the antisymmetric states, beside their localization, they are metastable with damping rate of $\Gamma_a \approx 0$. The symmetric states form excitons and have finite life-time with damping rate of $\Gamma_s \approx 2\Gamma_A$, two times the single excited atom one [19].

At this point we replace the two-level operators by bosonic ones, that is $S_k^\nu \to B_k^\nu$. In the next section we justify this step and present a bosonization procedure that goes beyond this simple replacement. In the strong coupling regime where the symmetric exciton and photon line widths are smaller than the coupling parameter, the excitons and photons are coherently mixed to form two polariton branches [24]. The Hamiltonian is diagonalized by using the upper and lower polariton operators

$$A_k^\pm = X_k^\pm \, B_k^s + Y_k^\pm \, a_k, \qquad (26)$$

which are a coherent superposition of symmetric excitons and photons. The mixing amplitudes are defined by

$$X_k^\pm = \pm\sqrt{\frac{D_k \mp \delta_k}{2D_k}}, \quad Y_k^\pm = \frac{f_k^s}{\sqrt{2D_k(D_k \mp \delta_k)}}, \quad (27)$$

where $D_k = \sqrt{\delta_k^2 + |f_k^s|^2}$, and the detuning is $\delta_k = \frac{E_{ph}(k) - E_s(k)}{2}$. The diagonal Hamiltonian reads

$$H_{pol} = \sum_{k,r} E_r(k) \, A_k^{r\dagger} A_k^r + \sum_k E_a \, B_k^{a\dagger} B_k^a, \qquad (28)$$

with the polariton dispersions

$$E_\pm(k) = \frac{E_{ph}(k) + E_s(k)}{2} \pm D_k. \qquad (29)$$

The exciton-photon detuning can be changed in order to control the mixing amplitude around small wave number polaritons.

Now we present the results using some typical numbers. For the transition energy we have $E_A = 1.5 \; eV$, for the lattice constant we use $a = 1000 \; \mathring{A}$, and for the interatomic distance we have $R = 100 \; \mathring{A}$. The transition dipole is $\mu = 2.5 \; e\mathring{A}$, the mode function is taken to be $u(b) = 0.25$, and the photon effective area is $S = \pi a^2$, with the effective dielectric constant $\epsilon = 2$.

In figure (3) we plot the polariton energies $E_\pm - E_A$, and the excitation energies $E_s - E_A$ and $E_a - E_A$, as a function of the polarization angle θ at zero wave number

$k = 0$. Here we take zero detuning between the photon and the transition energy, that is $E_{ph} = E_A$. The symmetric states have lower energy than the antisymmetric ones in the range from $\theta = 0^o$ up to the magic angle of $\theta_m = 54.7^o$, and beyond θ_m up to $\theta = 90^o$ the symmetric states have higher energy. At θ_m we have $E_A = E_s = E_a$.

For later use we concentrate in the dark states behavior. For polarization between $\theta = 0^o$ and the angle at which $E_a = E_+$, the dark states have energy higher than the upper polariton branch, and they can intersect the upper polariton dispersion at a finite wave number. For dark states between the angle at which $E_a = E_+$ and the magic angle θ_m they fell in the energy gap between the lower and upper polariton branches. Here the dark states have no intersection with the polariton branches at any wave number. But dark states between the angle θ_m and $\theta = 90^o$ can intersect the lower polariton branch at a finite wave number. The intersection point approaches small k as θ approaches 90°. This regime will be of interest later.
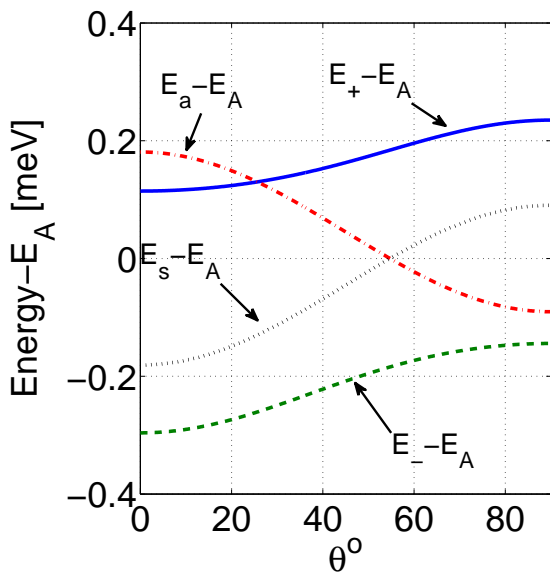

FIG. 4: The energies $E_{\pm} - E_A$, E_{ph}, $E_s - E_A$ and $E_a - E_A$ vs. k at the angle $\theta = 80^o$.

FIG. 3: The energies $E_{\pm} - E_A$, $E_s - E_A$ and $E_a - E_A$ vs. θ at zero wave number $k = 0$.

FIG. 5: The excitation and photon fractions in the upper and lower polariton branches vs. k at zero wave number $\theta = 80^o$. In the upper branch, the full line is for the photon fraction $|Y^+|^2$ and the dashed line for the excitation fraction $|X^+|^2$, and vise versa in the lower branch.
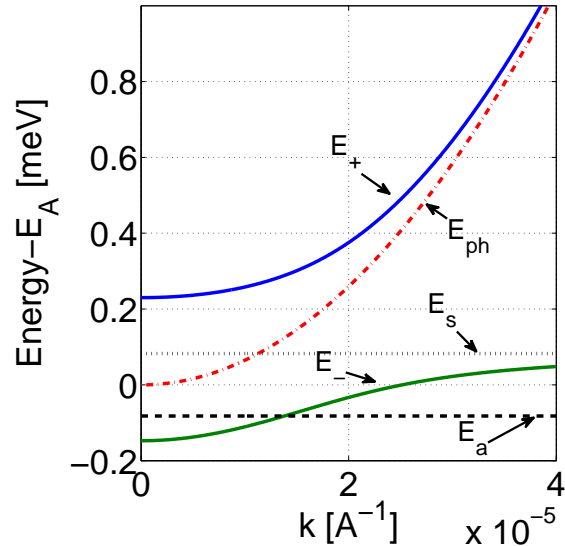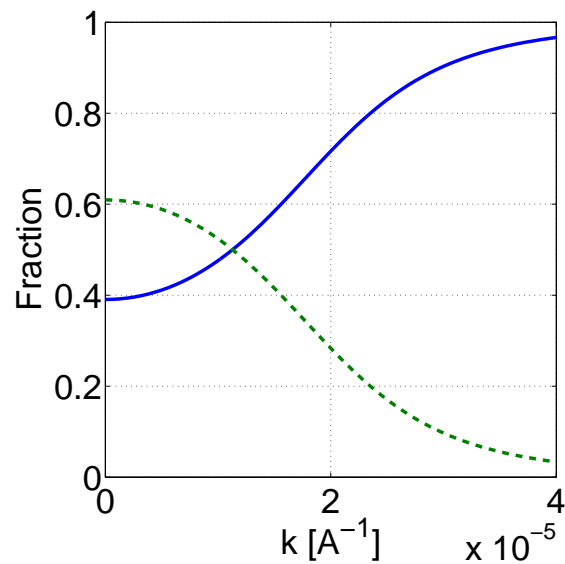
The polariton dispersions are plotted in figure (4) as a function of k. Here we choose $\theta = 80^o$, which is a reasonable angle in real experiments. We plot also the photon dispersion and the symmetric-antisymmetric energies. The dark state now intersects the lower polariton branch at a given wave number. In figure (5) we plot the excitation and photon fractions in the upper and lower polariton branches as a function of k, for $\theta = 80^o$. In the upper branch, the full line is for the photon fraction $|Y^+|^2$ and the dashed line for the excitation fraction $|X^+|^2$, and vise versa in the lower branch.

IV. KINEMATIC INTERACTIONS

Kinematic interactions are intensively studied by us in a previous work, and found to be a significant nonlinear mechanism for collective excitations in optical lattices [20, 21, 25]. In applying a bosonization procedure we transform the excitation spin-half operators into boson ones with additional terms that give rise to interactions

between bosons [22]. In solving the scattering problem we derived an effective potential that we aim to use in the following.

The operators $S_n^{\alpha\dagger}$ and S_n^α are of spin-half. They obey the fermi anti-commutation relation on the same atom, that is

$$S_n^\alpha S_n^{\alpha\dagger} + S_n^{\alpha\dagger} S_n^\alpha = 1 \ , \ S_n^\alpha S_n^\alpha = S_n^{\alpha\dagger} S_n^{\alpha\dagger} = 0, \qquad (30)$$

and the bose commutation relation between different atoms, that is

$$\left[S_n^\alpha, S_m^{\beta\dagger}\right] = \left[S_n^\alpha, S_m^\beta\right] = \left[S_n^{\alpha\dagger}, S_m^{\beta\dagger}\right] = 0, \ (n \neq m, \ \alpha \neq \beta). \qquad (31)$$

Then spin-half operators on a lattice have mixed statistics. They are fermions on-site and bosons among different sites, where they usually termed paulions.

We apply now the bosonization transformation [22]

$$S_n^\alpha \to \left(1 - B_n^{\alpha\dagger} B_n^\alpha\right) B_n^\alpha \ , \ S_n^{\alpha\dagger} \to B_n^{\alpha\dagger} \left(1 - B_n^{\alpha\dagger} B_n^\alpha\right), \qquad (32)$$

where $B_n^{\alpha\dagger}$ and B_n^α are boson operators with the commutation relation $[B_n^\alpha, B_m^{\beta\dagger}] = \delta_{nm}\delta_{\alpha\beta}$. This transformation holds at low density of excitations, where the number of excitations is much smaller than that of lattice sites.

The bosonization yields interacting bosons with $H = H_0 + H_I$, where the free boson part reads

$$H_0 = \sum_k \left\{ E_{ph}(k) \ a_k^\dagger a_k + E_s(k) \ B_k^{s\dagger} B_k^s + E_a \ B_k^{a\dagger} B_k^a \right.$$
$$\left. + \ f_k^s \ a_k B_k^{s\dagger} + f_k^{s*} \ B_k^s a_k^\dagger \right\}, \qquad (33)$$

and the interaction part reads

$$H_I = U \sum_n \left\{ B_n^{1\dagger} B_n^{1\dagger} B_n^1 B_n^1 + B_n^{2\dagger} B_n^{2\dagger} B_n^2 B_n^2 \right\}, \qquad (34)$$

where U is the kinematic effective potential.

In terms of symmetric-antisymmetric operators we have

$$H_I = \frac{U}{2} \sum_n \left\{ B_n^{s\dagger} B_n^{s\dagger} B_n^s B_n^s + B_n^{a\dagger} B_n^{a\dagger} B_n^a B_n^a \right.$$
$$+ \ B_n^{s\dagger} B_n^{s\dagger} B_n^a B_n^a + B_n^{a\dagger} B_n^{a\dagger} B_n^s B_n^s$$
$$\left. + \ 4 \ B_n^{s\dagger} B_n^{a\dagger} B_n^s B_n^a \right\}, \qquad (35)$$

and in momentum space we obtain

$$H_I = \frac{U}{2N} \sum_{kk'\bar{k}} \left\{ B_{k'-\bar{k}}^{s\dagger} B_{k+\bar{k}}^{s\dagger} B_{k'}^s B_k^s + B_{k'-\bar{k}}^{a\dagger} B_{k+\bar{k}}^{a\dagger} B_{k'}^a B_k^a \right.$$
$$+ \ B_{k'-\bar{k}}^{s\dagger} B_{k+\bar{k}}^{s\dagger} B_{k'}^a B_k^a + B_{k'-\bar{k}}^{a\dagger} B_{k+\bar{k}}^{a\dagger} B_{k'}^s B_k^s$$
$$\left. + \ 4 \ B_{k'-\bar{k}}^{s\dagger} B_{k+\bar{k}}^{a\dagger} B_{k'}^s B_k^a \right\}. \qquad (36)$$

We get kinematic interactions between symmetric states and between antisymmetric states, as appear in the first two terms. Interestingly, the last three terms represent interactions among symmetric and antisymmetric states. Here two symmetric states can be scatter into two antisymmetric ones, and vice versa. Moreover the last term represent scattering among symmetric and antisymmetric states, and they will be our main concern in the rest of the paper.

Writing the interaction Hamiltonian in terms of polariton operators yields

$$H_I = \frac{U}{2N} \sum_{kk'\bar{k}} \left\{ \sum_{\alpha\beta\gamma\delta} \left(X_{k'-\bar{k}}^\delta X_{k+\bar{k}}^\gamma X_{k'}^{\beta*} X_k^{\alpha*} \right) \right.$$
$$\times \ A_{k'-\bar{k}}^{\delta\dagger} A_{k+\bar{k}}^{\gamma\dagger} A_{k'}^\beta A_k^\alpha + B_{k'-\bar{k}}^{a\dagger} B_{k+\bar{k}}^{a\dagger} B_{k'}^a B_k^a$$
$$+ \ \sum_{\alpha\beta} \left[\left(X_{k'-\bar{k}}^\beta X_{k+\bar{k}}^\alpha \right) A_{k'-\bar{k}}^{\beta\dagger} A_{k+\bar{k}}^{\alpha\dagger} B_{k'}^a B_k^a \right.$$
$$+ \ \left(X_{k'}^{\beta*} X_k^{\alpha*} \right) B_{k'-\bar{k}}^{a\dagger} B_{k+\bar{k}}^{a\dagger} A_{k'}^\beta A_k^\alpha$$
$$\left.\left. + \ 4 \left(X_{k'-\bar{k}}^\beta X_{k'}^{\alpha*} \right) A_{k'-\bar{k}}^{\beta\dagger} B_{k+\bar{k}}^{a\dagger} A_{k'}^\alpha B_k^a \right] \right\}. \qquad (37)$$

We concentrate only in the lower polariton branch, hence we can drop the polariton branch indices. Furthermore, we interest in processes with small k, then one can neglect the dependence of X_k on k and replace it by X_0 at $k = 0$. We obtain

$$H_I = \frac{\Delta}{2} \sum_{kk'\bar{k}} \left\{ X_0^4 \ A_{k'-\bar{k}}^\dagger A_{k+\bar{k}}^\dagger A_{k'} A_k + B_{k'-\bar{k}}^\dagger B_{k+\bar{k}}^\dagger B_{k'} B_k \right.$$
$$+ \ X_0^2 \ A_{k'-\bar{k}}^\dagger A_{k+\bar{k}}^\dagger B_{k'} B_k + X_0^2 \ B_{k'-\bar{k}}^\dagger B_{k+\bar{k}}^\dagger A_{k'} A_k$$
$$\left. + \ 4X_0^2 \ A_{k'-\bar{k}}^\dagger B_{k+\bar{k}}^\dagger A_{k'} B_k \right\}. \qquad (38)$$

Here, in order to simplify the notations, we used A_k for the lower polariton operator, and B_k for the antisymmetric exciton operator. We also defined $\Delta = \frac{U}{N}$, where the effective potential is given by $U = \frac{4\pi\hbar^2}{ma^2}$, and in using $L = Na$, we get $\Delta = \frac{4\pi\hbar^2}{maL}$ [22].

A. Dynamical Interactions

Beside the kinematic interactions that result of quantum statistics, dynamical interactions due to Coulomb forces are also of importance here. The dynamical interactions have the form

$$H_{Dyn} = \sum_{nm} \sum_{\alpha\beta} V_{nm}^{\alpha\beta} \ S_n^{\alpha\dagger} S_m^{\beta\dagger} S_n^\alpha S_m^\beta, \qquad (39)$$

where $\alpha \neq \beta$ for $n = m$. The interaction is among two excited atoms and it is of higher order than the dipole-dipole interaction, e.g., of van der Waals type. As the lattice constant is relatively large, we neglect dynamical interactions among atoms at different unit cells, and we include only interactions among two excited atoms on the same unit cell. Now we have

$$H_{Dyn} = V \sum_n \sum_{\alpha\beta}' S_n^{\alpha\dagger} S_n^{\beta\dagger} S_n^\alpha S_n^\beta, \qquad (40)$$

where the prime indicates that $\alpha \neq \beta$, with $V = V_n^{12}$, and we can write $H_{Dyn} = 2V \sum_n S_n^{1\dagger} S_n^{2\dagger} S_n^1 S_n^2$. The state of two excited atoms in the same unit cell n is defined by $|e_n\rangle = |e_n^1, e_n^2\rangle$ with the energy $E_e = 2E_A + E_B$, and the binding energy $E_B = 2V$.

This result is critical in the present work as it can exclude the possibility of double excitations per site due to conservation of energy, where we have ($E_e \neq 2E_s, 2E_a, 2E_A$). Hence in the next sections we can neglect the scattering of two polaritons into the state $|e_n\rangle$ of two bound on-site excitations. But surely they can be involved in other processes.

V. PARAMETRIC EXCITATION OF THE DARK STATES

It is a big experimental challenge to excite dark antisymmetric states as they are weakly coupled to the light. Usually they are populated through the decay of higher states, or by multi-photon nonlinear processes. Here we exploit the result of the previous section to suggest kinematic interactions as a mechanism for significantly exciting such dark states. The above kinematic Hamiltonian results in mixing interactions that allow scattering among bright polaritons and dark excitons.

We exploit the kinematic scattering of polaritons into dark excitons as a mechanism for populating the dark states. In order to achieve this task in a controllable way we present the following scenario that implies two external sources in pump-probe experiment [26]. An external pump is used to strongly excite polaritons with a fixed wave number, say k. A probe field is applied in order to weakly excite dark states at a different wave number, say $k + q$ or $k - q$, or both. This probe field is needed in order to stimulate scattering of bright polaritons into these dark states. Therefore, for the lower branch we consider only polaritons with a fixed wave number k. The upper polariton branch is excluded due to conservation of energy. As two polaritons of wave number k can scatter into two dark excitons, the conservation of momentum implies two dark excitons with momenta $k + q$ and $k - q$. Due to the fact that dark excitons are dispersion-less and on-site localized, any pair of dark states is allowed in the scattering with wave numbers $k + q$ and $k - q$. Here we consider the pump and probe fields to have the same energy E, where we choose $E = E_{pol}(k) = E_a$. The probe field at wave numbers $k + q$ or $k - q$ are off resonance with polaritons of energy E_a. On the other hand, the pump field can directly and weakly excite dark excitons at k, and in the following we neglect the scattering of these dark excitons into the $k + q$ and $k - q$ states. This configuration is used in order to excite dark excitons with energies separated from the polariton in an efficient and controllable manner, as presented in figure (6).
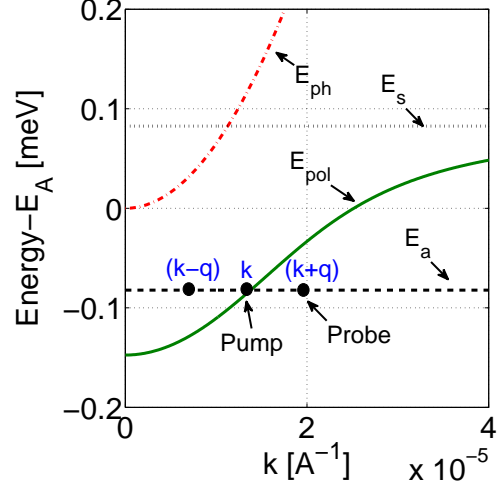
In the light of the above discussion, we consider the

FIG. 6: The energies $E_{pol} - E_A$, E_{ph}, $E_s - E_A$ and $E_a - E_A$ vs. k at the angle $\theta = 80^o$. The cavity pump is at k and the side probe at $k + q$. Two polaritons of wave number k have stimulated scattering into two dark states at $k + q$ and $k - q$.

interaction Hamiltonian with only A_k polaritons, that is

$$
\begin{aligned}
H_I &= \frac{\Delta X_k^4}{2} A_k^\dagger A_k^\dagger A_k A_k + \frac{\Delta}{2} \sum_{kk'\bar{k}} B_{k'-\bar{k}}^\dagger B_{k+\bar{k}}^\dagger B_{k'} B_k \\
&+ \frac{\Delta X_k^2}{2} \sum_{\bar{k}} \left\{ A_k^\dagger A_k^\dagger B_{k+\bar{k}} B_{k-\bar{k}} + B_{k+\bar{k}}^\dagger B_{k-\bar{k}}^\dagger A_k A_k \right. \\
&+ \left. 4 A_k^\dagger B_{\bar{k}}^\dagger A_k B_{\bar{k}} \right\}. \quad (41)
\end{aligned}
$$

The first term represent interactions among polaritons. The second term gives interactions among dark excitons, and as dark states are weakly excited we will neglect this term in the following. The second line includes scattering among polaritons and dark excitons, where the first term describes scattering of two excitons into two polaritons, and vice versa for the second one. The third line represents the scattering of an exciton and a polariton. We concentrate in the interaction Hamiltonian

$$
\begin{aligned}
H_I &= \frac{\Delta X_k^2}{2} \left\{ X_k^2 A_k^\dagger A_k^\dagger A_k A_k + \sum_p \left[A_k^\dagger A_k^\dagger B_{k+p} B_{k-p} \right. \right. \\
&+ \left. \left. B_{k+p}^\dagger B_{k-p}^\dagger A_k A_k + 4 A_k^\dagger B_p^\dagger A_k B_p \right] \right\}. \quad (42)
\end{aligned}
$$

We start by deriving equations of motion for the po-

lariton and exciton operators

$$
i\hbar\frac{d}{dt}A_k = \Big(E_{pol}(k) + \Delta X_k^4\ A_k^\dagger A_k
$$
$$
+\ 2\Delta X_k^2 \sum_p B_p^\dagger B_p\Big) A_k
$$
$$
+\ \Delta X_k^2 \sum_p A_k^\dagger B_{k+p} B_{k-p} + F_k^{pump},
$$

$$
i\hbar\frac{d}{dt}B_{k+q} = \Big(E_a + 2\Delta X_k^2\ A_k^\dagger A_k\Big) B_{k+q}
$$
$$
+\ \Delta X_k^2\ B_{k-q}^\dagger A_k A_k + F_{k+q}^{probe},
$$

$$
i\hbar\frac{d}{dt}B_{k-q} = \Big(E_a + 2\Delta X_k^2\ A_k^\dagger A_k\Big) B_{k-q}
$$
$$
+\ \Delta X_k^2\ B_{k+q}^\dagger A_k A_k + F_{k-q}^{probe}. \tag{43}
$$

We added three external fields, one field pumps the polaritons through the waveguide at wave number k, and the probe fields excite weakly the dark excitons at wave numbers $k+q$ and $k-q$. The pump and probe fields are taken to be of energy $E = \hbar\omega$, where $F_k^{pump} = \tilde{F}_k^{pump}e^{-i\omega t}$, and $F_p^{probe} = \tilde{F}_p^{probe}e^{-i\omega t}$. Hence we transfer into a system that rotate at frequency ω, by using $A_k = \tilde{A}_k e^{-i\omega t}$, and $B_k = \tilde{B}_k e^{-i\omega t}$.

At this point we apply the mean field theory in taking the expectation value of the equations, and by applying the factorization approximation

$$
\langle \tilde{A}_k^\dagger \tilde{A}_k \tilde{A}_k\rangle = \langle \tilde{A}_k^\dagger \tilde{A}_k\rangle\langle \tilde{A}_k\rangle,
$$
$$
\langle \tilde{A}_k^\dagger \tilde{A}_k \tilde{B}_{k+q}\rangle = \langle \tilde{A}_k^\dagger \tilde{A}_k\rangle\langle \tilde{B}_{k+q}\rangle,
$$
$$
\langle \tilde{A}_k^\dagger \tilde{A}_k \tilde{B}_{k-q}\rangle = \langle \tilde{A}_k^\dagger \tilde{A}_k\rangle\langle \tilde{B}_{k-q}\rangle,
$$
$$
\langle \tilde{B}_p^\dagger \tilde{B}_p \tilde{A}_k\rangle = \langle \tilde{B}_p^\dagger \tilde{B}_p\rangle\langle \tilde{A}_k\rangle,
$$
$$
\langle \tilde{B}_{k+q}^\dagger \tilde{A}_k \tilde{A}_k\rangle = \langle \tilde{B}_{k+q}^\dagger\rangle\langle \tilde{A}_k\rangle^2,
$$
$$
\langle \tilde{B}_{k-q}^\dagger \tilde{A}_k \tilde{A}_k\rangle = \langle \tilde{B}_{k-q}^\dagger\rangle\langle \tilde{A}_k\rangle^2,
$$
$$
\langle \tilde{A}_k^\dagger \tilde{B}_{k+p} \tilde{B}_{k-p}\rangle = \langle \tilde{A}_k^\dagger\rangle\langle \tilde{B}_{k+p}\rangle\langle \tilde{B}_{k-p}\rangle, \tag{44}
$$

and we define $\langle \tilde{A}_k\rangle = \tilde{A}_k$, $\langle \tilde{B}_k\rangle = \tilde{B}_k$, $\langle \tilde{F}\rangle = \tilde{F}$, $\langle \tilde{A}_k^\dagger \tilde{A}_k\rangle = \mathcal{N}_k$, and $\langle \tilde{B}_k^\dagger \tilde{B}_k\rangle = \mathcal{I}_k$. For strong pump field we can neglect the contribution of the dark excitons scattering into polaritons, hence we neglect terms that include dark excitons in the first equation. Moreover, we define the renormalized exciton and polariton energies by $\tilde{E}_{pol}(k) = E_{pol}(k) + \Delta X_k^4\ \mathcal{N}_k$, and $\tilde{E}_a = E_a + 2\Delta X_k^2\ \mathcal{N}_k$. The mean field equations of motion read

$$
i\hbar\frac{d}{dt}\tilde{A}_k = \Big(\tilde{E}_{pol}(k) - E\Big)\tilde{A}_k + \tilde{F}_k^{pump},
$$
$$
i\hbar\frac{d}{dt}\tilde{B}_{k+q} = \Big(\tilde{E}_a - E\Big)\tilde{B}_{k+q} + V\ \tilde{B}_{k-q} + \tilde{F}_{k+q}^{probe},
$$
$$
i\hbar\frac{d}{dt}\tilde{B}_{k-q} = \Big(\tilde{E}_a - E\Big)\tilde{B}_{k-q} + V\ \tilde{B}_{k+q} + \tilde{F}_{k-q}^{probe} \tag{45}
$$

where $V = \Delta X_k^2 \mathcal{N}_k$.

At steady state, where $\frac{d}{dt}\tilde{A}_k = \frac{d}{dt}\tilde{B}_k = 0$, we have

$$
\tilde{B}_{k+q} = \frac{\Big(E - \tilde{E}_a\Big)\tilde{\mathcal{F}}_{k+q}^{probe} + V\ \tilde{\mathcal{F}}_{k-q}^{probe}}{\Big(E - \tilde{E}_a\Big)^2 - V^2},
$$
$$
\tilde{B}_{k-q} = \frac{\Big(E - \tilde{E}_a\Big)\tilde{\mathcal{F}}_{k-q}^{probe} + V\ \tilde{\mathcal{F}}_{k+q}^{probe}}{\Big(E - \tilde{E}_a\Big)^2 - V^2}. \tag{46}
$$

The fields have in general two resonances at $E_+ = \tilde{E}_a + V$ and $E_- = \tilde{E}_a - V$. But for the specific case of $\tilde{\mathcal{F}}_{k-q}^{probe} = \tilde{\mathcal{F}}_{k+q}^{probe}$ we get only a single resonance at $E = \tilde{E}_a + V$.

We concentrate here in the case of one probe field at $k+q$, that is $\tilde{\mathcal{F}}_{k-q}^{probe} = 0$, $\tilde{\mathcal{F}}_{k+q}^{probe} = \tilde{\mathcal{F}}^{probe}$, where we get

$$
\tilde{B}_{k-q} = \frac{V\ \tilde{\mathcal{F}}^{probe}}{\Big(E - \tilde{E}_a\Big)^2 - V^2},
$$
$$
\tilde{B}_{k+q} = \frac{\Big(E - \tilde{E}_a\Big)\tilde{\mathcal{F}}^{probe}}{\Big(E - \tilde{E}_a\Big)^2 - V^2}, \tag{47}
$$

with the intensities

$$
\mathcal{I}_{k+q} = \frac{\Big(E - \tilde{E}_a\Big)^2}{\Big[\Big(E - \tilde{E}_a\Big)^2 - \tilde{\Delta}^2\mathcal{N}_k^2\Big]^2}|\tilde{\mathcal{F}}_{k+q}^{probe}|^2,
$$
$$
\mathcal{I}_{k-q} = \frac{\tilde{\Delta}^2\ \mathcal{N}_k^2}{\Big[\Big(E - \tilde{E}_a\Big)^2 - \tilde{\Delta}^2\mathcal{N}_k^2\Big]^2}|\tilde{\mathcal{F}}_{k+q}^{probe}|^2, \tag{48}
$$

where $\tilde{\Delta} = \Delta X_k^2$. It is clear that we interestingly get two resonances at $E_+ = \tilde{E}_a + V$ and at $E_- = \tilde{E}_a - V$.

For the pumped polaritons we have

$$
\tilde{A}_k = \frac{\tilde{\mathcal{F}}_k^{pump}}{\Big(E - \tilde{E}_{pol}(k)\Big)}, \tag{49}
$$

and

$$
\mathcal{N}_k = \frac{|\tilde{\mathcal{F}}_k^{pump}|^2}{\Big(E - \tilde{E}_{pol}(k)\Big)^2}. \tag{50}
$$

In order to examine if such two resonances can be experimentally resolved we include the finite life time for both the exciton and the polariton phenomenologically in changing the energies into complex parameters $\tilde{E}_{pol}(k) \to \tilde{E}_{pol}(k) - i\hbar\Gamma_k^{pol}$, and $\tilde{E}_a \to \tilde{E}_a - i\hbar\Gamma_a$, where Γ_a is the dark exciton damping rate, which is relatively a small number. Γ_k^{pol} is the polariton damping rate, that is defined by $\Gamma_k^{pol} = \frac{1}{2}|X_k|^2\Gamma_s + \frac{1}{2}|Y_k|^2\Gamma_{ph}$, where Γ_s is the symmetric exciton damping rate, and Γ_{ph} is the photon

damping rate. The polariton and dark exciton average numbers are written as

$$\mathcal{N}_k = \frac{|\tilde{\mathcal{F}}_k^{pump}|^2}{\left(E - \tilde{E}_{pol}(k)\right)^2 + \left(\hbar\Gamma_k^{pol}\right)^2}, \qquad (51)$$

and

$$\mathcal{I}_{k+q} = \frac{\left(E - \tilde{E}_a\right)^2 + (\hbar\Gamma_a)^2}{\left(E - \tilde{E}_+\right)^2 \left(E - \tilde{E}_-\right)^2} \mathcal{I}^{probe},$$

$$\mathcal{I}_{k-q} = \frac{\tilde{\Delta}^2 \, \mathcal{N}_k^2}{\left(E - \tilde{E}_+\right)^2 \left(E - \tilde{E}_-\right)^2} \mathcal{I}^{probe}, \qquad (52)$$

where $\mathcal{I}^{probe} = |\tilde{\mathcal{F}}_{k+q}^{probe}|^2$, and $\tilde{E}_\pm = \tilde{E}_a \pm \sqrt{\tilde{\Delta}^2\mathcal{N}_k^2 - \hbar\Gamma_a^2}$.

We now illustrate the results in using the previous numbers. Moreover, we use for the photons $\hbar\Gamma_{ph} = 10^{-10}$ eV, for the symmetric excitons $\hbar\Gamma_s = 10^{-8}$ eV, and for the antisymmetric excitons $\hbar\Gamma_a = 10^{-12}$ eV. The lattice length is taken to be $L = 1$ cm. We fix the polarization angle to $\theta = 80^o$. We pump lower branch polaritons at energy $E_{pol}(k) = E_a$, which is at wave number $k \approx 1.4 \times 10^{-5}$ $\mathring{A}^{-1}$. To calculate the effective interaction parameter we need to evaluate the polariton effective mass, which is of the order of the cavity photon effective mass, hence we have $mc^2 \approx \hbar c q_0 \sqrt{\epsilon}$, and for $E_A = E_{ph}(0)$ we obtain $q_0 = \sqrt{\epsilon}E_A/(\hbar c)$. Here we have $q_0 \approx 10^{-3}$ $\mathring{A}^{-1}$, $mc^2 \approx 3$ eV, and for the interaction parameter we get $\Delta \approx 1.6 \times 10^{-4}$ eV. At this k we get $|X_k|^2 \approx 0.56$ hence we have $\tilde{\Delta} \approx 9.1 \times 10^{-5}$ eV. The pumped polaritons are assumed to be with average number of $\mathcal{N}_k \sim 1$. In figure (7) we plot the scaled intensity $\mathcal{I}_{k-q}/\mathcal{I}^{probe}$ as a function of $E - E_a$. We pump at k and probe at $k + q$ of energy E_a, and hence the dark excitons get excited at $k - q$ with blue shift energy. As dark excitons have small Γ_a, that is much smaller than the interaction parameter $\tilde{\Delta}$ the spectra split into two peaks at $\tilde{E}_+$ and $\tilde{E}_-$. We get parametric amplification of the dark excitons both at $k + q$ and $k - q$ with blue shifts, which can be experimentally resolved.

VI. DARK BOGOLON-EXCITONS

We now aim to get a much deep understanding of the previous section results for the dark states. In order to achieve this target we follow a different direction for treating pair excitations of dark excitons in adopting Bogoliubov mean field theory [23].

We start again from the Hamiltonian (41). Now we assume the strong pump of the polaritons at wave number k to be a classical field, which fits with the previous mean field theory treatment. Hence the first term becomes a number. As before, the dark states are weakly excited
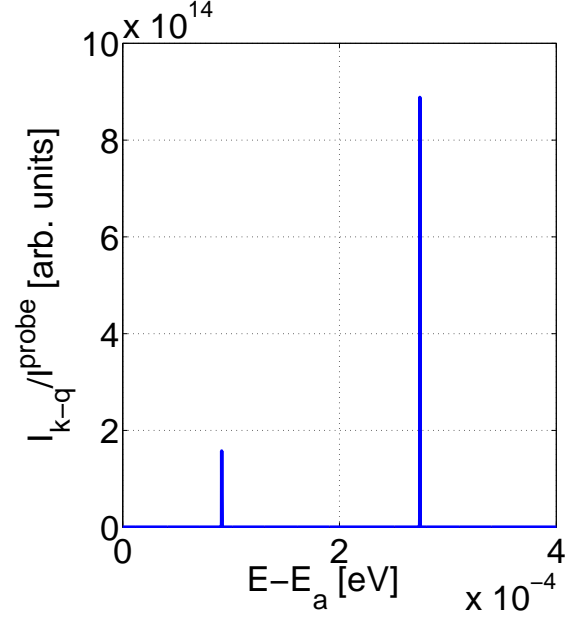

FIG. 7: The scaled intensity $\mathcal{I}_{k-q}/\mathcal{I}^{probe}$ vs. $E - E_a$. The peaks are easily resolved.

then we can neglect the second term of their mutual interactions. These assumptions are in the spirit of Bogoliubov mean field theory [23]. In using $\langle A_k \rangle = \sqrt{\mathcal{N}}e^{-i\omega t}$, the total Hamiltonian reads

$$H = \sum_p \left\{ \frac{1}{2} \left(E_a + 2\Delta\mathcal{N}X_k^2\right) \left(B_{k+p}^\dagger B_{k+p} + B_{k-p}^\dagger B_{k-p}\right) \right.$$
$$\left. + \frac{\Delta\mathcal{N}X_k^2}{2} \left(e^{i2\omega t} B_{k+p}B_{k-p} + e^{-i2\omega t} B_{k+p}^\dagger B_{k-p}^\dagger\right) \right\}. \qquad (53)$$

We transform into rotating frame in applying the transformation $U = e^{-iSt/\hbar}$, hence we have $H \to U^\dagger HU - S$, with $S = E\left(B_{k+p}^\dagger B_{k+p} + B_{k-p}^\dagger B_{k-p}\right)$, to get

$$H = \frac{1}{2}\sum_p \left\{ \left(\tilde{E}_a - E\right)\left(\tilde{B}_{k+p}^\dagger \tilde{B}_{k+p} + \tilde{B}_{k-p}^\dagger \tilde{B}_{k-p}\right) \right.$$
$$\left. + V\left(\tilde{B}_{k+p}\tilde{B}_{k-p} + \tilde{B}_{k+p}^\dagger \tilde{B}_{k-p}^\dagger\right) \right\}, \qquad (54)$$

where $\tilde{E}_a = E_a + 2V$, and $V = \Delta\mathcal{N}X_k^2$.

The form of the Hamiltonian appeals to the Bogoliubov transformation [23]

$$\alpha_p = u \, \tilde{B}_{k+p} + v \, \tilde{B}_{k-p}^\dagger,$$
$$\beta_p = u \, \tilde{B}_{k-p} + v \, \tilde{B}_{k+p}^\dagger, \qquad (55)$$

with

$$[\alpha_p, \alpha_p^\dagger] = [\beta_p, \beta_p^\dagger] = 1, \qquad (56)$$

and

$$[\alpha_p, \beta_p^\dagger] = [\beta_p, \alpha_p^\dagger] = [\alpha_p, \beta_p] = [\alpha_p^\dagger, \beta_p^\dagger] = 0, \quad (57)$$

where $u^2 - v^2 = 1$. The inverse transformation is

$$\tilde{B}_{k+p} = u\,\alpha_p - v\,\beta_p^\dagger,$$
$$\tilde{B}_{k-p} = u\,\beta_p - v\,\alpha_p^\dagger. \quad (58)$$

In terms of the new operators the Hamiltonian is reads

$$H = \sum_p \Bigg\{ \left(\tilde{E}_a - E\right) v^2 - V\,uv$$
$$+ \left[\frac{1}{2}\left(\tilde{E}_a - E\right)(u^2 + v^2) - V\,uv\right]\left(\alpha_p^\dagger \alpha_p + \beta_p^\dagger \beta_p\right)$$
$$+ \left[\frac{V}{2}(u^2 + v^2) - \left(\tilde{E}_a - E\right)uv\right]\left(\alpha_p \beta_p + \alpha_p^\dagger \beta_p^\dagger\right) \Bigg\}. \quad (59)$$

We need the second line to vanish, then we can choose u and v such that

$$\frac{V}{2}(u^2 + v^2) = \left(\tilde{E}_a - E\right)uv. \quad (60)$$

We take u and v to be real, and we choose $u > 0$, hence from $u^2 - v^2 = 1$, we use $u = \cosh t$ and $v = \sinh t$, and the above relation gives $\tanh 2t = V/\left(\tilde{E}_a - E\right)$, and we get the solution

$$u^2 = \frac{1}{2}\left(\frac{\tilde{E}_a - E}{2\tilde{E}_0} + 1\right), \; v^2 = \frac{1}{2}\left(\frac{\tilde{E}_a - E}{2\tilde{E}_0} - 1\right), \quad (61)$$

where

$$\tilde{E}_0 = \frac{1}{2}\sqrt{\left(\tilde{E}_a - E\right)^2 - V^2}. \quad (62)$$

Using this solution, the Hamiltonian is given by

$$H = \sum_p \left\{ \tilde{E}_0 - \frac{\tilde{E}_a - E}{2} + \tilde{E}_0\left(\alpha_p^\dagger \alpha_p + \beta_p^\dagger \beta_p\right) \right\}. \quad (63)$$

As $\alpha_p = \beta_{-p}$ and $\beta_p = \alpha_{-p}$, we define the operators

$$C_p = u\,\tilde{B}_{k+p} + v\,\tilde{B}_{k-p}^\dagger,$$
$$C_{-p} = u\,\tilde{B}_{k-p} + v\,\tilde{B}_{k+p}^\dagger, \quad (64)$$

then we can write

$$H = \sum_p \left\{ \frac{1}{2}\left(\bar{E}_0 - \tilde{E}_a + E\right) + \bar{E}_0\,C_p^\dagger C_p \right\}, \quad (65)$$

where here $\bar{E}_0 = 2\tilde{E}_0$. The diagonal Hamiltonian represents dark state bogolons, in which two dark states of momenta $k+p$ and $k-p$ are strongly correlated. These elementary excitations are termed here bogolon-excitons and found to be dispersion-less with energy $\bar{E}_0$.

We use a probe field that weakly excites the dark excitons at $k+q$, then the Bogoliubov mean field equations of motion in rotating frame for the new operators are

$$i\hbar\frac{d}{dt}\mathcal{C}_{k+q} = \bar{E}_0\,\mathcal{C}_{k+q} + u\,\tilde{\mathcal{F}},$$
$$i\hbar\frac{d}{dt}\mathcal{C}_{k-q} = \bar{E}_0\,\mathcal{C}_{k-q} - v\,\tilde{\mathcal{F}}. \quad (66)$$

At steady state

$$\mathcal{C}_{k+q} = -\frac{u\,\tilde{\mathcal{F}}}{\bar{E}_0}, \; \mathcal{C}_{k-q} = \frac{v\,\tilde{\mathcal{F}}}{\bar{E}_0}. \quad (67)$$

The results exactly confirm the previous section ones, which can be obtained in using the inverse of the (64) transformation.

We conclude that in the case of a single probe field at $k+q$, the two dark excitons, which appear through the scattering of two k polaritons, are spontaneously correlated and form two coherent states that are separated by the interaction energy V in the optical spectra.

VII. CONCLUSIONS

In the present paper we showed how coherent states of electronic excitations are spontaneously formed only through resonant dipole-dipole interactions and in exploiting the lattice symmetry, without the aid of additional external fields. The study is presented for an optical super-lattice with two atoms per unit cell in which dark and bright excitons appear. In localizing the one dimensional atomic super-lattice parallel to a nanophotonic waveguide the bright excitons are strongly coupled to the waveguide photons to form polaritons as the system natural collective excitations.

Excitons behave as bosons at low intensity of excitations, but as the atomic transitions are of two-level systems a mechanism is required to forbid two excitations from being localized on the same atom. A bosonization procedure from two-level systems into bosons was suggested by us in other work that excludes double excitation at the same atom by including kinematic interactions among excitons. In adopting this procedure for the present atomic super-lattice the kinematic interactions interestingly couple dark and bright excitons.

We used the kinematic interactions in order to excite dark excitons in an efficient and controllable way. We suggested a pump-probe experiment in which two pumped polaritons of a given momentum k can be scattered into two dark excitons of momenta $k+p$ and $k-p$, where the process is induced by a probe field. The two dark excitons are found to be strongly correlated and can be represented as dark bogolon-excitons.

Bogolon-excitons can be implemented as qubits for quantum information processing, and their long life-time allow them to serve as memory states. The above parametric amplification scenario can be used to write into

these states, and the addition of asymmetry into the system can be used to read out. Furthermore, achieving a fixed population of dark bogolon-excitons in a controllable way is an important step toward achieving the long awaited BEC of excitons.

The consideration in the present paper event hough presented for atomic super-lattice can be also realized for a one dimensional organic crystal with two molecules per unit cell. Here one get an anisotropic crystal and the orientation of the molecules relative to the axis gives rise to two Davydov exciton branches, in which both can be bright or one bright and the other dark. This fact can enrich the present physical properties as the polarization direction provides an extra degree of freedom.

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