Bright and dark excitons in an atom-pair–filled optical lattice within a cavity

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Abstract – We study electronic excitations of a degenerate gas of atoms trapped in pairs in an optical lattice. Local dipole-dipole interactions produce a long-lived antisymmetric and a short-lived symmetric superposition of individual atomic excitations as the lowest internal on-site excitations. Due to the much larger dipole moment the symmetric states couple efficiently to neighbouring lattice sites and can be well represented by Frenkel excitons, while the antisymmetric dark states stay localized. Within a cavity only symmetric states couple to cavity photons inducing long-range interactions to form polaritons. We calculate their dispersion curves as well as cavity transmission and reflection spectra to observe them. For a lattice with aspherical sites bright and dark states get mixed and their relative excitation energies depend on photon polarizations.

A dilute gas of bosonic atoms near $T=0$ in an optical-lattice has proved an ideal test-system to study important quantum phenomena of solid-state physics with well-controllable parameters [1,2]. A striking example is a reversible quantum phase transition from the superfluid into the Mott insulator phase [3] simply by changing the optical-potential depth. In the Mott insulator case each optical-lattice site is filled with a fixed number of atoms down to one or two atoms per site. For a deep enough lattice the atoms cannot move and form an artificial crystal. Naturally it is interesting to study further complex solid-state phenomena in this system, e.g. by exploiting the internal atomic level structure, which bears a strong analogy to the excitonic dynamics of molecular crystals (Frenkel excitons) as predicted in ref. [4]. By the help of an optical cavity these excitons get strongly coupled over large distances via photons and form polaritons. Recently strong resonant light-matter coupling for a Bose-Einstein condensate in an optical cavity has been achieved [5]. The scattering of excitons and cavity polaritons off vacancies in an optical lattice is studied by the present authors in [6]. The case of three-level atoms in an optical lattice was considered in [7]. In the present paper we investigate a special interesting case of such excitons [8] and cavity polaritons [9] which can appear only for lattice filled with two atoms per site, which is straightforward to prepare in optical lattices by the help of a Mott insulator state with filling factor 2.

Let us start from a degenerate gas of effective two-level atoms trapped in an optical lattice, located within a cavity with a single-cavity mode close to resonance to an internal atomic transition. For simplicity we consider only a single-lattice plane transverse to the cavity [10,11]. The lattice laser is tuned far off resonance to the atomic excitations and results in light shifts of the ground and excited states with periodicity of half laser wavelength, i.e. $a = \lambda / 2$. The ground- and excited-state optical-lattice minima are assumed to be located at the same positions, which can be realized for alkali or alkaline atoms, and at certain magic laser frequencies the excited state even experiences an equal shift as the ground state [2,12]. We believe that more general lattice configurations might imply new physics, but this goes beyond our aim here. At temperatures close to $T = 0$, the atomic center-of-mass motion is confined to the lowest Bloch band and the ground- and excited-state atoms can be mathematically treated as two kinds of bosons leading to a Bose-Hubbard model with two boson components. It possesses a rich phase diagram containing a Mott insulator phase and a superfluid phase [13].

For a deep lattice atomic tunneling is suppressed so that each atom retains his identity with negligible overlap of the electronic wave functions of atoms at different sites.

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Fig. 1: Left: the two on-site atoms. Right: two nearest-neighbor sites.

As the ground-state wave function (Wannier function) at each site is still much larger than the size of an atom $a$, that is $a \gg \bar{a}$, we can for the moment also neglect the short-range part of the molecular potentials and treat their interaction by a pseudo-potential involving only the scattering length. This makes the artificial atomic lattices similar to molecular or noble atom crystals. While the electronic excitations can transfer between atoms due to dipole-dipole interactions, there is no direct electron exchange or particle tunneling.

For a simplified model the atomic Hamiltonian then reads [14]

$$H = \sum_{i,\alpha} \hbar \omega_{A,\alpha} B_{i}^{\alpha \dagger} B_{i}^{\alpha} + \sum_{ij,\alpha,\beta} \hbar J_{ij}^{\alpha \beta} B_{i}^{\alpha \dagger} B_{j}^{\beta},$$

(1)

where $B_{i}^{\alpha \dagger}$ and $B_{i}^{\alpha}$ are the creation and annihilation operators of an excitation at atom $(i, \alpha)$, respectively. The summation $i$ runs over the lattice sites, while $\alpha$ labels the two atoms at one site. The first part represents local excitation with a transition frequency $\omega_{A}$, while the second part generates the energy transfer between atoms $(i, \alpha)$ and $(j, \beta)$ with coupling amplitude $J_{ij}^{\alpha \beta}$. In principle $B_{i}^{\alpha}$ are two-level transition operators that forbid two excitations on the same atom. Nevertheless at low excitation density we neglect the possibility of two excitations on the same atom and we can assume the excitations to behave as bosons [15].

As simple example to discuss the basic physics we consider a cubic optical lattice with energy transfer only between atoms on the same site with amplitude $J^{\alpha \beta}(\bar{R}) = -J_{0}$, and between nearest-neighbor sites with strength $J^{\alpha \beta}(\bar{R}) = -J_{1}^{\alpha \beta}$, see fig. 1. $J_{0}$ has to be calculated from the position spread of the atomic Wannier functions, which yields a number that can be estimated (as shown later) from the dipole-dipole interaction in using an interatomic effective distance $\bar{R}$ which obeys $\bar{a} \ll \bar{R} \ll a$. The on-site part of the Hamiltonian in principle can be diagonalized in changing to symmetric and antisymmetric entangled excitations $B_{i}^{1} = \frac{B_{i}^{+} + B_{i}^{\dagger}}{\sqrt{2}}$, and $B_{i}^{2} = \frac{B_{i}^{+} - B_{i}^{\dagger}}{\sqrt{2}}$, to get

$$H = - \sum_{(i,j)} \hbar \left\{ (J_{1} + J_{2}^{\dagger}) B_{i}^{1 \dagger} B_{j}^{2} + (J_{1} - J_{2}^{\dagger}) B_{i}^{2 \dagger} B_{j}^{1} \right\} + \sum_{i} \hbar \left( \omega_{A} B_{i}^{1 \dagger} B_{i}^{2} + \omega_{A} B_{i}^{2 \dagger} B_{i}^{1} \right),$$

(2)

where $\omega_{A} = \omega_{A} - J_{0}$ and $\omega_{A} = \omega_{A} + J_{0}$, which appear in fig. 1. Note that evaluating the local coupling requires integration of dipole-dipole coupling over the local wave functions, but will eventually just give fixed parameters. For these energy transfer parameters we assumed $J_{1}^{2} = J_{2}^{2} = J_{1}$ and $J_{1}^{1} = J_{2}^{1} = J_{1}$. As $J_{1} \approx J_{1}^{*}$, the non-local energy transfer is large only for the symmetric excitations which decay with linewidth of $\hbar \Gamma_{s} \approx 2 \hbar \Gamma_{e}$, which is two times the atomic linewidth [16]. In contrast, the antisymmetric states are shown to be metastable with a long radiative lifetime and negligible next-neighbor coupling.

Using the lattice symmetry a delocalized excitation can be represented in quasi-momentum space by a propagating wave with wave vector $k$. Such a quasi-particle is called Frenkel exciton in molecular crystals [8,14] and our Hamiltonian can be diagonalized by the help of these quasi-particles $\bar{B}^{\nu} = \sqrt{N} \sum_{k} \bar{B}_{k}^{\nu} e^{i k \cdot \bar{R}}$. Here $(\nu = s, a)$, $N$ is the number of lattice sites, and $\bar{r}_{i}$ is the position of site $i$. In a 2D optical lattice $k$ takes the values $k = (k_{x}, k_{y}) = \frac{2 \pi}{\tilde{a}} (n_{x}, n_{y})$, and $n_{x,y} = 0, \pm 1, \cdots, \pm \frac{M}{2}$, with $N = M \times M$, and $M$ is an even number. The Hamiltonian casts into

$$H = \sum_{k} \hbar \omega_{\nu}(k) \bar{B}_{k}^{\nu \dagger} \bar{B}_{k}^{\nu},$$

with the energy dispersion for the symmetric and antisymmetric branches

$$\omega_{\nu,a}(k) = \omega_{A} - 2 (J_{1} + J_{1}^{*}) [\cos(k_{x} a) + \cos(k_{y} a)].$$

(3)

The antisymmetric branch is almost dispersionless, and can be considered as a localized state at each site with energy $\omega_{s,a} \approx \omega_{A} + h J_{0}$. Only symmetric states propagate in the lattice. The symmetric-branch bandwidth is $8 h J_{1}$, between $k = 0$ and the boundary of the Brillioun zone at $k = \pi/a$. For small wave vectors, $k \ll 1$, where $k = |k|$, the symmetric-branch dispersion reads $\omega_{s}(k) = \omega_{A} - h J_{0} - 8 J_{1} + \frac{2 k^{2}}{2m \Gamma_{e}}$, with the effective mass $m_{e,f} = h^{2}/(4 J_{1} a^{2})$. At zero wave vector, $k = 0$, the symmetric excitation has a shift of $\omega_{s} - h J_{0} - h J_{1}$ relative to free atoms, which can be easily observed. Also such a shift has influence on optical-lattice clock states [17].

As the symmetric coherent exciton states decay approximately with rate $\Gamma_{s}$, in order to observe exciton effects the excitation linewidth needs to be smaller than the exciton bandwidth, i.e. $\Gamma_{s} < 8 h J_{1}$. Let us calculate this for alkali atoms of the transition $2 S_{1/2} \leftrightarrow 2 P_{3/2}$, where we have $\omega_{A} = 1.5-2.5$ eV and a linewidth $\Gamma_{e} = 1-5 \times 10^{-8}$ eV. The energy transfer $h J(\bar{R})$ is calculated from the dipole-dipole interaction between dipoles $\bar{\mu}_{1}$ and $\bar{\mu}_{2}$ separated by a distance $\bar{R} = R \bar{R}$, i.e.

$$h J(\bar{R}) = \sum_{ij} \frac{\mu_{i}^{\dagger} \mu_{j}^{\dagger}}{4 \pi \epsilon_{0} \bar{R}^{2}} \left\{ - (\delta_{ij} - \bar{R}_{i} \bar{R}_{j}) q^{2} \bar{R}^{2} \cos q \bar{R} + (\delta_{ij} - 3 \bar{R}_{i} \bar{R}_{j}) (\cos q \bar{R} + q \sin q \bar{R}) \right\},$$

(4)

here $(i,j = x,y,z)$, with $\omega_{A} = c q$. For linear polarization $\bar{R} = R \bar{R}$ in the $x$-direction $\bar{\mu} = \bar{\mu}_{2} = \mu \hat{x}$, we get $h J(\bar{R}) = \frac{-\mu^{2}}{2 \epsilon_{0} \bar{R}^{2}} [\cos q \bar{R} + q \sin q \bar{R}]$ along $x$. The distance between two atoms at neighbouring sites equals the
where perpendicular direction nance to the atomic transitions so they will not generate the cavity photons energies here are much closer to resonant, the lattice lasers are treated as classical fields, we consider the coupling parameter \( f_{k,i} \). The opticallattice is located by the electric dipole interaction between two on-site atoms we can neglect the oscillation terms, to get for the previous case \( J_0(R) = -\mu^2 / (2\pi\epsilon_0 R^3) \). For \( R = 80 \) Å we get \( J_0 = 0.001 \) eV. The next-nearest-neighbor terms turn out to be smaller than the excited-state linewidth and unimportant for exciton formation.

Let us now add a planar cavity made of two parallel perfect mirrors to study coherent excitation transfer mediated by the cavity photons. The optical lattice is located in the middle between the cavity mirrors as in fig. 2. While the lattice lasers are treated as classical fields, we consider a quantized cavity field. In contrast to previous work [18] the cavity photons energies here are much closer to resonance to the atomic transitions so they will not generate extra periodic forces.

The electromagnetic field is free in the cavity plane with the in-plane wave vector \( \vec{k} \) confined only in the perpendicular direction \( z \) with wave vector \( k_z = m\pi/L \), and \( m = 0, 1, 2, \ldots \). Here we consider only the longitudinal mode \( m \) closest to resonance to the atom excitation and with a fixed polarization. The cavity photon Hamiltonian is given by \( H_c = \sum_{\vec{k}} \hbar \omega_c(k) \hat{a}^\dagger_{\vec{k}} \hat{a}_{\vec{k}} \) where \( \hat{a}^\dagger_{\vec{k}} , \hat{a}_{\vec{k}} \) are the creation and annihilation operators of a cavity photon with in-plane wave vector \( \vec{k} \), respectively. The dispersion is \( \omega_c(k) = \frac{\hbar}{\sqrt{\epsilon_0\mu_0 k^2 + \left(\frac{m\pi}{L}\right)^2}} \), with \( L \) the distance between the mirrors and \( \epsilon = 1 \). Note that as the optical lattice is located in the middle, between the cavity mirrors, \( m \) needs to be an odd number to get maximum electric field at the lattice position.

The atomic excitations are coupled to the cavity photons by the electric dipole interaction \( V = -\hat{\mu} \cdot \vec{E} \), where \( \hat{\mu} = \sum_{\alpha} (B^{\alpha\dagger} \hat{B}^\alpha) \) is the material dipole operator, and \( \vec{E} \) is the quantized cavity electric field. The transition dipoles \( \hat{\mu} \) here are taken to be equal for all the atoms. The interaction Hamiltonian, in the rotating-wave approximation, reads \( V = \sum_{\vec{k},i} \sum_{\alpha} \left( f^\alpha_{k,i} \hat{B}^{\alpha\dagger} \hat{a}_\vec{k} + f^\alpha_{k,i} \hat{a}^\dagger_\vec{k} \hat{B}^\alpha \right) \). The coupling parameter \( f^\alpha_{k,i} \) between atom \( \alpha \) at site \( i \) and a photon with wave vector \( \vec{k} \), is \( f^\alpha_{k,i} = i \sqrt{\frac{\hbar \omega_c(k)}{2\pi\epsilon_0 L}} (\hat{\mu} \cdot \hat{e}_\vec{k}) e^{i \vec{k} \cdot \vec{r}_i^a} \), where \( S \) is the cavity mirror area, and \( \vec{r}_i^a \) is the atom position. In terms of symmetric and antisymmetric operators we get \( V = \sum_{\vec{k},i,\alpha} \left( f^\alpha_{k,i} \hat{B}^{\alpha\dagger} \hat{a}_\vec{k} + f^\alpha_{k,i} \hat{a}^\dagger_\vec{k} \hat{B}^\alpha \right) \), where we defined the symmetric and antisymmetric coupling parameters \( f^\alpha_{k,i} = \frac{f^\alpha_{k,i} + f^\alpha_{-k,i}}{\sqrt{2}} \) and \( f^\alpha_{k,i} - \frac{f^\alpha_{k,i} + f^\alpha_{-k,i}}{\sqrt{2}} \). As the two atoms are on the same site, we have approximately \( f^\alpha_{k,i} \approx f^\alpha_{k,i} \) and the antisymmetric excitations (dark states) almost decouple from the cavity photons. Only the symmetric excitations can get strongly coupled to the cavity photons. The position of atom \( \alpha \) at site \( i \) is \( \vec{r}_i^\alpha = \vec{r}_i + \vec{d}_\alpha \). We assumed that each one of the two atoms has the same average on-site position, \( \vec{d}_\alpha \). For the coupling we can write \( f^\alpha_{k,i} \approx -i \sqrt{\hbar \omega_c(k) \pi \epsilon_0 / 4\lambda^2} (1 \pm e^{i\phi} |d_i - d_j|) \), where \( |d_i - d_j| \) is the average deviation with a fixed direction. The coupling of the antisymmetric states to the cavity photons, even though weak, can play an important role in coupling these states with the external world.

We get mainly strong-coupling cavity photons of transverse wavelength much longer than the distance between the two atoms and excitons of about the same wave vector. Hence we can assume \( |d_i - d_j| \ll 1 \) and thus we get \( f^\alpha_{k,i} \approx -i \sqrt{\hbar \omega_c(k) \pi \epsilon_0 / 4\lambda^2} \), and \( f^\alpha_{k,i} \approx 0 \). In the \( k \) space we have

\[
H = \sum_{\vec{k}} \left\{ \hbar \omega_c(k) \hat{B}_{\vec{k}}^\dagger \hat{B}_{\vec{k}} + \hbar \omega_c(k) \hat{a}_{\vec{k}}^\dagger \hat{a}_{\vec{k}} \right\} + \hbar f_{k} \hat{B}_{\vec{k}}^\dagger \hat{a}_{\vec{k}} + \hbar f_{k} \hat{a}_{\vec{k}}^\dagger \hat{B}_{\vec{k}} + \sum_{\vec{k}} \hbar \omega_a \hat{B}_{\vec{k}}^\dagger \hat{B}_{\vec{k}}. \tag{5}
\]

In the strong-coupling regime where the exciton and photon linewidths are smaller than the coupling strength, the exciton and the photon are mixed to form the new system excitations, called polaritons [9,14] diagonalizing the first part of the above Hamiltonian. As the exciton effective mass is much larger than the photon ones, the exciton dispersion around the exciton-photon coupling can be neglected and one can simply use \( \omega_x \approx \omega_A - J_0 - \delta J \). The diagonalized Hamiltonian is

\[
H = \sum_{\vec{k},\alpha} \hbar \Omega_{\vec{k},\alpha} \hat{A}_{\vec{k},\alpha}^\dagger \hat{A}_{\vec{k},\alpha} + \sum \hbar \omega_a \hat{B}_{\vec{k}}^\dagger \hat{B}_{\vec{k}}. \tag{6}
\]

We have two polariton branches with dispersions \( \Omega_{\vec{k},\alpha} = \pm \sqrt{\omega_x(k)^2 + \Delta \epsilon(k)} \), where \( \Delta \epsilon(k) = \left( \omega_x(k) - \omega_A \right) / 2 \), and the exciton-photon detuning is \( \delta \epsilon(k) = \left( \omega_x(k) - \omega_a \right) / 2 \). The splitting at the exciton-photon intersection point, where \( \delta \epsilon = 0 \), is \( 2 f_{k,i} \), which is called the Rabi splitting. The polaritons are coherent superpositions of symmetric excitons and photons, with the diagonal operators \( \hat{A}_{\vec{k},\alpha} = \hat{X}_{\vec{k}}^\dagger \hat{B}_{\vec{k}} + Y_{\vec{k}}^\dagger \hat{a}_{\vec{k}} \) with exciton and photon amplitudes \( X_{\vec{k}} = \pm \sqrt{\frac{\Delta \epsilon(k)}{2 \Delta \epsilon(k)}} \) and \( Y_{\vec{k}} = \frac{f_{k}}{\sqrt{2 \Delta \epsilon(k)}} \). In fig. 3(a) we illustrate this plotting the upper and lower polariton energy branches. As example the exciton energy around small in-plane wave vector is taken to be \( \hbar \omega_x = 1.999 \) eV, and we have \( \hbar \omega_a = 2.001 \) eV. The distance between the cavity mirrors is \( L/m = 3102 \) Å, which is chosen to give zero detuning between the symmetric exciton and the cavity photon dispersions at zero in-plane.
wave vector. For $m = 3$ we get $L \approx 1 \mu m$. The transition dipole is $\mu = 4 e \AA$, and the optical-lattice constant is $a = 2000 \AA$. The exciton-photon coupling energy, by using $S = Na^2$, is $|hf| = 0.00015 eV$, where we neglected the $k$-dependence for small in-plane wave vectors. At zero in-plane wave vector the Rabi splitting energy of 0.0003 eV is clear. At large wave vectors the upper polariton branch tends to the cavity photon dispersion, and the lower branch tends to the symmetric exciton dispersion. In fig. 3(b) we plot the exciton and photon weights in the lower and upper polariton branches. $|X^\pm|^2$ and $|Y^\pm|^2$. At zero in-plane wave vector the polariton is half exciton and half photon. For large in-plane wave vectors the lower polariton becomes much more excitonic than photonic, and vice versa for the upper polariton.

So far the situation is closely analogous to a lattice with occupancy equal to one with rescaled coupling. However, in order to exhibit and use the extra degrees of freedom here, we consider the case of an optical lattice with asymmetric sites, where one of the orthogonal pairs of counterpropagating lasers has a different intensity and thus the on-site potential is elongated in one direction, e.g. $x$. Hence the two on-site atoms will have an average distance $\bar{R} = |d_2 \pm d_1|$ in the $x$-direction, even after considering the local atom wave functions. Thus, we have $\bar{a} \ll R \ll a$ and then $J_0 \gg J_1$, which holds for the lowest Bloch bands. Hence, the atoms do not immediately form molecules, and the long-range part of the potential of the two atoms is the dipole-dipole interaction. The atomic transition dipole is induced by the cavity photon, and is oriented along the electric field of the cavity photon. We treat the case of in-plane transition dipoles, namely the general dipole is $\vec{\mu} = \mu(\cos \theta, \sin \theta)$, which depends on $\theta$, the angle between the dipole $\vec{\mu}$ and the $x$-axis, see fig. 4. The resonance dipole-dipole interaction between the two on-site atoms is $\hbar J_0(\theta) = \frac{\mu^2}{4\pi\epsilon_0 R^3}(1 - 3 \cos^2 \theta)$. We have $\hbar J_0(\theta = 0) = -2\mu^2/(4\pi\epsilon_0 R^3)$, and $\hbar J_0(\theta = 90) = \mu^2/(4\pi\epsilon_0 R^3)$, with $\hbar J_0(\theta \approx 54.74^\circ) = 0$. The detuning energy between symmetric and antisymmetric states can change sign and also for a fixed polarization can be vanishing.

Using the above results, in fig. 5(a) we plot the symmetric and antisymmetric states, and also the two polariton branches, as a function of $\theta$, for the case of zero in-plane wave vector, that is $k = 0$. It is clear that the symmetric-antisymmetric states change sign at $\theta \approx 54.74^\circ$. At this angle the two states are degenerate. We chose zero detuning between the cavity photon and the symmetric state at $\theta = 0^\circ$. The maximum detuning between the symmetric state and the cavity photon appears at $\theta = 90^\circ$. In fig. 5(b) we plot the excitonic and photonic weights in the lower and upper polariton branches as a function of the angle $\theta$. The symmetric-antisymmetric splitting and the Rabi splitting are easily controlled by changing the field polarization direction.

In order to observe these system eigenmodes we couple the internal cavity modes to the external world, i.e. the external radiation field in and out coupled through the nonperfect cavity mirrors, and calculate the cavity input and output fields in a standard quantum-optical approach. Similarly we will include atomic spontaneous emission via an effective exciton damping. In fig. 6 we plot the corresponding transmission and reflection spectra [4] for
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an incident field with zero in-plane wave vector, \( k = 0 \), where the electric field is parallel to the mirrors. We choose the following numbers for the linewidths: the symmetric exciton linewidth is \( \hbar \Gamma_s = 10^{-7} \) eV, the mirror linewidth is \( \hbar \gamma = 10^{-5} \) eV. In fig. 6(a) we plot the transmission, and in fig. 6(b) the reflection, for different polarization angles. The peaks of the transmission, and the dips of the reflection correspond to the two polariton branches, which are the real eigenmodes of the system. Large transmission and reflection in regions where the polariton is much more photonic than excitonic is obtained.

In summary, excitons and cavity polaritons involving resonant excitations of ultracold atoms in an optical lattice acquire intriguing new properties for the case of two atoms trapped at each site. Antisymmetric local excitations with a long lifetime and weak neighbor coupling point to applications for long-time memory in optoelectronics and quantum information. Symmetric excitations strongly couple to neighboring atoms and cavity photons and those form polaritons mediating controlled long-range interactions. Controlled excitations can be facilitated via decay of higher-energy states by nonlinear optical processes. Similarly one could exploit the weak coupling of the antisymmetric states to the cavity photons in order to write and read information on these states, while the symmetric state polaritons can be used as diagnosis and readout tool.

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