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Detecting an exciton crystal by statistical means

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ABSTRACT

We investigate an ensemble of excitons in a coupled quantum well excited via an applied laser field. Using an effective disordered quantum Ising model, we perform a numerical simulation of the experimental procedure and calculate the probability distribution function $P(M)$ to create M excitons as well as their correlation function. It shows clear evidence of the existence of two phases corresponding to a liquid and a crystal phase. We demonstrate that not only the correlation function but also the distribution $P(M)$ is very well suited to monitor this transition.

The exciton is a very fascinating composite particle, which can be generated and investigated in specially designed semiconductor heterostructures – the bilayer systems. In its simplest incarnation it is a bound state of an electron and a hole and is thus of bosonic nature. If the system size of such a compound is small it is natural to ask whether a Bose–Einstein condensation (BEC) of such structures is possible [1,2]. On the other hand, also the possibility of a Cooper-pair-like ground state [the conventional Bardeen–Cooper–Schrieffer (BCS) superconductivity] has been considered in a number of works [3–5]. Recent experimental progress in the field of electronic bilayer systems allowed for a study of all these different fascinating possibilities [6–14].

In most experimental realizations the holes and electrons are spatially separated so that every such indirect exciton has a relatively large dipole moment. It turns out that at some intermediate density, at which the BEC condensation is prohibited, the dipole interactions let the excitons see each other. If the correlations are strong enough even a long-ranged ordering of Wigner crystal type is possible [15–20]. A detection of such kind of crystalline structure is very difficult though. The excitons themselves are usually generated with the help of laser fields and they are rather fragile with respect to irradiation. Thus traditional spectroscopic techniques are very difficult to apply and one needs alternative methods [15]. One such approach is based on the knowledge of the first-order correlation function, the measurement of which was very recently reported in Refs. [13,14].

Here we propose an alternative statistical method of detecting and analyzing the properties of the exciton crystallization phenomenon and discuss its predictive power. A typical experimental cycle would start with the generation of excitons via a laser pulse in a coupled quantum well structure (e.g. GaAs/AlGaAs heterostructure). A perpendicularly applied voltage, which leads to a tilt of the potential landscape, ensures that the dipole moment of all excitons created in this way is parallel. After that the number of excitons M is measured by their recombination. Conducting a large number of cycles one gathers the statistics of M [21,22]. The fact that for a given M a regular arrangement of the excitons on a lattice minimizes their interaction energy should be visible in the probability distribution $P(M)$. Although the correlation function possesses a higher predictive power, we shall show below that the crystallization can even be seen in $P(M)$, which is accessible by much less effort.

We assume that before every measurement cycle the system consists of N valence band electrons, located at random positions $\mathbf{r}_i$. Of course electrons are fermionic while an exciton, being composite particle of electron and hole, can display bosonic properties. In our case the respective statistics are of no importance since we work in the 'frozen gas approximation' where all particles are distinguishable. In our model any electron can be excited to the excitonic state, making it an effective two-level system with the bare electron being the ground state and the exciton being the excited state. Still, to avoid confusion, we would like to introduce the convention to call each effective two-level system a 'particle' regardless of the underlying statistics. A laser field with frequency ω_L excites the system. It is detuned from the actual transition frequency ω_{sp} between the lower bare electron state and the upper excitonic state by Δ, resulting in $\Delta = \omega_{sp} - \omega_L$. By means of the laser intensity the Rabi

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frequency ω^*, which accounts for transitions from the ground to the exciton state, can be changed. In a typical experiment [23] one uses short laser pulses with high intensity, for which the Rabi frequency is of the same order as the exciton binding energy [24]. A circularly polarized light beam (polarization σ) at a suitably chosen frequency can create direct (which are of no importance to our work) and indirect (which are assigned to the excited state of each particle) neutral excitons with well-defined spins σ and $\overline{\sigma} = -\sigma$ by exciting an electron from the valence band of the same or the other semiconductor in the coupled quantum well (layer), respectively (see Fig. 1) [25,22].

In the following we will focus only on the indirect excitons, the particle and hole of which are located in different layers. The formation of direct excitons can be neglected, since either their formation can be suppressed by an appropriate choice of the excitation frequency ω_L or one can simply wait long enough – their recombination time is much shorter. The indirect excitons possess dipole moments d perpendicular to the layers $d = eD$, where D is the interlayer separation, see Fig. 1(a).

Each particle, being a two-level system [26], corresponds to an electron in the ground state and to an exciton in the excited state [27]. We assume the levels to be sharp neglecting effects from the Fermi distribution of the separate bands. We can do so in the limit of a large detuning of the laser from the resonance [24]. The particles interact with the laser light and with each other through the dipole interaction in the excited state. The velocity distribution of excitons in coupled quantum wells [23] can be tuned by efficient cooling [13,14] and the application of a perpendicular magnetic field, which gives rise to a higher effective mass [22,28,29]. With both the laser pulse duration [30] and the experimental measurement taking $\tau < 0.1$ ns and the resulting velocities $v_{Ex} \approx 10$ m/s the displacement of a single particle is $v_{Ex}\tau < 1$ nm. The typical separations of particles in their excited state are of the order 100 nm so that we can assume them to be fixed in space [31]. The size of a particle in its excited state, which is an exciton, is estimated via its Bohr radius $a_B \approx 20$ nm, see e.g. Ref. [32]. Since the electrostatic properties do not depend on the details of the excited (exciton) states we model the system as a randomly arranged interacting ensemble of spin 1/2 sub-systems each representing a single particle, where the up state corresponds to the excitonic state while the down state corresponds to the bare electron. We note that such mapping restricts possible excitations to excitons and does not allow for possible four-particle excitations. Hence, the Hamiltonian reads [33,27]

$$H = -\frac{\Delta}{2}\sum_{i=1}^{N}\sigma_z^{(i)} + \frac{\omega^*}{2}\sum_{i=1}^{N}\sigma_x^{(i)} + \frac{C^*}{4}\sum_{i=2}^{N}\sum_{j=1}^{i-1}\frac{(1+\sigma_z^{(i)})(1+\sigma_z^{(j)})}{|\mathbf{r}_i - \mathbf{r}_j|^3}, \qquad (1)$$

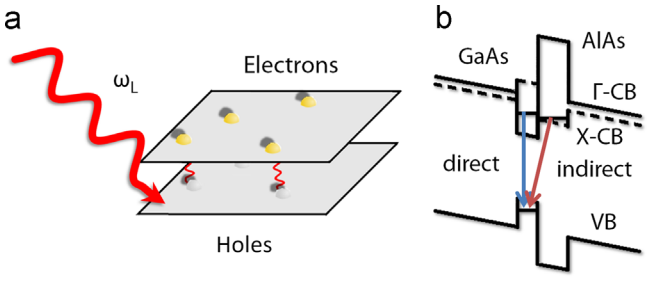

Fig. 1. (a) shows the typical experimental situation: excitons are created in a coupled quantum well via an applied laser field that excites electrons from the valence band into the conduction band and thus forms excitons. Possible exciton states illustrating the exciton binding energy in a typical coupled quantum well such as GaAs/AlAs are shown in (b). These coupled quantum wells have two conduction bands (Γ and X) originating from different points in the Brillouin zone and allow for both the formation of direct and indirect excitons, however, with different binding energies.

where $\sigma_{x,z}^{(i)}$ denote the Pauli matrices. This equation uses the rotating wave approximation when describing the light–matter interaction as in Ref. [34]. It neglects all terms oscillating with frequencies ω_L and higher. From here on we measure all energies in units of Δ entailing $\omega = \omega^*/\Delta$. The interaction strength is therefore measured in units of ΔL^3, leading to the definition $C = C^*/\Delta L^3$. The ground state particle density $n = N/V$, where V is either a one-dimensional interval or a two-dimensional square, is given for all plots.

The model (1) can be interpreted as a (generalized) spin-1/2 Ising model. The Rabi frequency ω^* and the detuning Δ correspond to magnetic fields in x- and z-directions. The third parameter C^* indicates the strength of the effective interaction between the excitons. We should note that a similar model has originally been applied to interactions between Rydberg atoms, where a similar crystal-like phase exists [28,35–37,39]. In the case of Rydberg atoms, induced dipole moments give rise to van-der-Waals interactions. In the case of excitons, in contrast, dipole–dipole interactions between the excitons lead to a stronger dependence of the distance, $\propto |r|^{-3}$.

We are only interested in the case $\Delta > 0$ (corresponding to a laser frequency large enough to overcome the exciton binding energy) because otherwise it is energetically not favorable to produce excitons. However, Δ has to be smaller than the valence-band conductance band separation since otherwise we would produce unpaired electron–hole pairs.

We typically use a large detuning from the transition frequency in accordance with experimental studies [23], so that Δ is larger but still comparable in magnitude to the Rabi frequency. A typical laser field has an excitation frequency of about 1 eV. The excitons have a dipole moment oriented perpendicular to the plane. In this case $C^* = e^2D^2/\epsilon$. For the dielectric spacer between the top and the bottom layer we assume $\epsilon = 12.9\epsilon_0$ being a typical value for GaAs and $D = 11.5$ nm as put forward by Ref. [38]. In the numerical simulations we take the length L of the simulated square in 2D to be $\approx$200–500 nm. For such and larger system dimensions we did not detect any sizeable finite size effects.

The numerical procedure emulates the experimental process by initially generating a random distribution of N electrons [spins in Eq. (1)] in a given one- or two-dimensional volume with open or periodic boundary conditions. Then the corresponding Hamiltonian matrix is set up. For large N the size of this matrix is reduced by the truncation of the Hilbert space, which is done by taking into account only k basis states with the smallest diagonal elements in the Hamiltonian matrix. We systematically checked that all the results do not depend on k. In the next step the eigenvector corresponding to the smallest eigenvalue, the ground state (GS), is calculated by matrix diagonalization. It is given as a linear combination of the previously mentioned basis states and therefore enables an efficient evaluation of different observables. Besides the number of excitons (which can be non-integer since the ground state is generally a superposition) the pair correlation function $g(r)$, which is closely related to the density distribution of excitons, can be computed in the following way: we divide the possible range for distances between electrons in our system into equidistant bins and measure the distance between each pair of electrons to assign it to a certain bin. For each pair the squares of coefficients from the representation of the GS as a linear combination are summed over those states in which the particular pair of electrons is excited. The sum over multiple random arrangements then produces the correlation function.

We start by considering a 1D system with different interaction strengths. Already for weak interactions we see that the correlation function is zero for $r < R_B$, where R_B can be interpreted as the blockade radius, see Fig. 2. This effect is due to a strong dipole field in the vicinity of an exciton which suppresses the excitation of additional ones. As expected R_B increases with increasing interaction strength. Simultaneously an emergence of peaks of