Strong correlation effects in 2D Bose-Einstein condensed dipolar excitons

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By doing quantum Monte Carlo \emph{ab initio} simulations we show that dipolar excitons, which are now under experimental study, actually are strongly correlated systems. Strong correlations manifest in significant deviations of excitation spectra from the Bogoliubov one, large Bose condensate depletion, short-range order in the pair correlation function, and peak(s) in the structure factor.

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Two-dimensional (2D) dipolar excitons (DEs) with spatially separated electrons (\(e\)) and holes (\(h\)) are extremely interesting due to increased lifetime, which permits to achieve different quasi-equilibrium exciton phases predicted for the system, e.g., 2D DE superfluid in extended systems \([1, 2, 3, 4, 5]\) and traps \([6]\), Bose-Einstein condensate \([3, 4, 7]\), crystal phase \([8, 9]\) and supersolid \([10]\).

A number of interesting phenomena, such as Josephson effects \([11]\), light backscattering and other interesting optical properties \([12]\) can also be observed in the system.

DE systems can be created in coupled quantum wells (CQWs) \([1]\) or in a single quantum well (SQW) applying a large external electric field (see theory in \([13]\)). At present, a large experimental progress has been achieved in the study of 2D DE collective properties in CQWs \([14, 15, 16, 17, 18, 19]\) and SQW \([20]\).

The superfluidity and coherent properties of equilibrium \(e-h\) systems (in a sense of "spatially separated semimetal") have been also theoretically studied \([1, 2, 4, 11]\). An important progress was achieved by studying an electron bilayer in high magnetic field with \(1/2 + 1/2\) filling of Landau levels \([21]\). It can be proved that the properties of the system can be presented as a BCS state of a spatially separated (composite fermion) \(e-h\) system at zero magnetic field \([1, 2, 22]\). The predicted \(e-h\) superfluidity and Josephson-like effects in this system were observed later on experimentally \([23]\). It is worth noticing that the 2D dipolar Bose systems under consideration have been recently realized in atomic systems with large dipolar moments (\(e.g.\), for Cr atoms) \([24]\) and polar molecules \([25]\).

The majority of theoretical models describe the excitonic Bose condensate as an ideal or weakly correlated gas. Unfortunately, these approaches have a very limited range of applicability. Indeed, at small densities the repulsive dipole-dipole potential can be described by one parameter \(a\), the \(s\)-wave scattering length, and the properties are expected to be universal, i.e., to be the same for all interaction potentials having the same value of scattering length and, in particular, to be the same as in a system of hard-disks of diameter \(a\). In fact, the latter is known to be weakly correlated only in ultra-rarefied systems \([7]\). So, the model of weakly correlated excitons holds only in ultra-rarefied gases which have extremely low critical temperature. For real experimental excitonic densities such models can provide only a qualitative description. Thus, a more accurate model should be worked out and a more precise study should be done in order to describe 2D DEs in quantum wells (QWs).

This paper is devoted to a detailed microscopic study of 2D DEs by means of the diffusion Monte Carlo (DMC) technique. We prove that excitons are in fact strongly correlated in all the main up-to-now experiments with CQW \([16, 17, 18]\) in which low-temperature collective effects in exciton luminescence have been observed. We have obtained the following results supporting this fact:

(i) The dimensionless compressibility \(\zeta = (m^2/2\pi\hbar^2)/\chi\) and the contribution of dipole-dipole collisions to the chemical potential \(\zeta' = \mu_c/m/2\pi\hbar^2n\) are much larger than unity \(\zeta, \zeta' \gg 1\) \((\chi \text{ and } \mu_c\) are the corresponding dimensional quantities, \(m\) is the exciton mass, and \(n\) is the total exciton density in \(g_{ex}\) spin degrees; \(g_{ex} = 4\) in GaAs). For weakly correlated excitons one has \(\zeta, \zeta' \ll 1\).

(ii) The Bose-condensate density \(n_0\) of excitons at \(T = 0\) is 2-4 times smaller than the total density \(n\) (on the contrary, in the weak-correlation regime one assumes \(n_0 \approx n\)).

(iii) There are prominent peaks in both the pair distribution function and the structure factor providing an evidence of short-range order. Moreover, in the structures studied in Refs. \([16, 17, 18]\) at typical exciton density, \(e.g., n = 5 \cdot 10^{10}\ \text{cm}^{-2}\) two peaks in the structure factor and three bumps of the pair distribution function are clearly visible.

(iv) The excitation spectrum strongly deviates from the weakly-correlated Bogoliubov prediction although, even in the crossover between exciton and \(e-h\) regimes, there is still no roton minimum as found in Refs. \([16, 17, 18]\).

(v) In the superfluid phase, the superfluid excitonic density \(n_f\) is close to total density \(n\) \([26]\). In weakly correlated systems, the quantity \(n_f\) is significantly smaller than \(n\) at the superfluid transition.

(vi) The superfluid transition temperature in an infinite system \(T_s\) is only slightly smaller than the degeneration temperature \(T_{deg}\), but the quasi-condensation temperature is 2-2.5 times larger than \(T_{deg}\) \([27]\). According
is the exciton dipole moment, the hole charge, $\varepsilon$ is the dielectric constant of the QW structure ($\varepsilon \approx 12.5$ in GaAs), and $\varepsilon_0 = 1/4\pi$ is the permittivity of vacuum.

Parameters of recent experiments \[14, 15, 16, 17, 18\] are summarized in Table I.

| Structure      | $D$, nm $1/x_0^2$, cm$^{-2}$ | Ref. |
|----------------|-----------------------------|------|
| GaAs/AlGaAs CQWs | 15.5 | $1.56 \cdot 10^{10}$ | \[14\] |
| InGaAs/GaAs CQWs | 10.5 | $1.83 \cdot 10^{11}$ | \[15\] |
| AlAs/GaAs CQWs  | 13.6 | $2.64 \cdot 10^{10}$ | \[16\] |
| GaAs/AlGaAs CQWs | 12.3 | $3.94 \cdot 10^{10}$ | \[17\] |
| GaAs/AlGaAs CQWs | 14.1 | $2.28 \cdot 10^{10}$ | \[18\] |
| GaAs SQW       | $\approx 6$ | $7 \cdot 10^{11}$ | \[20\] |

Table I: Experimental structures and values of their $e$-$h$ separations $D$ and dimensionless densities $n = 1/x_0^2$ corresponding to dimensionless density $\bar{n} = 1$. The exciton mass is $m = 0.14m_0$ for InGaAs/GaAs CQWs and $m = 0.22m_0$ for other CQWs and SQW, with $m_0$ being the free electron mass.

to the theory of weakly correlated excitons these temperatures are logarithmically small compared to $T_{\text{deg}}$.

(vii) The excitonic Bose condensate profile in a 2D large-size harmonic trap at $T = 0$ differs appreciably from the Thomas-Fermi inverted parabola.

For Timofeev’s experiment \[20\] with SQW we find excitons to be intermediate correlated.

We have performed DMC simulations at $T = 0$ of a 2D homogeneous system with $N = 100$ DEs in a square simulation box with periodic boundary conditions and for 12 different dimensionless densities $\bar{n} \equiv nx_0^2 = 2^{1-i}$, where $i$ is an integer number ($1 \leq i \leq 12$), and

$$x_0 = \frac{md^2}{4\pi\varepsilon_0\varepsilon d^2} = \frac{me^2D^2}{4\pi\varepsilon_0\varepsilon d^2} > 0 \quad (1)$$

is the characteristic length scale for the excitonic dipole-dipole interaction $V(r) = \hbar^2 x_0/mr^3$. Here, $d = eD > 0$ is the exciton dipole moment,

$$D = \left| \int_{-\infty}^{\infty} (|\psi_e(z)|^2 - |\psi_h(z)|^2) z dz \right|$$

is the effective $e$-$h$ separation, with $\psi_e(z)$ and $\psi_h(z)$ the wavefunctions of $e$ and $h$ in the $Oz$ direction, $e > 0$ is the hole charge, $\varepsilon$ is the dielectric constant of the QW structure ($\varepsilon \approx 12.5$ in GaAs), and $\varepsilon_0 = 1/4\pi$ is the permittivity of vacuum.

Parameters of recent experiments \[14, 15, 16, 17, 18\] are summarized in Table I.

We note that the excitonic collective properties are experimentally observed in Refs. \[16, 17, 18\] at dimensionless densities $1/4 \leq \bar{n} \leq 4$. At the lowest density $\bar{n} = 1/256$ of our computation the superfluid crossover temperature in the DE system of Ref. \[20\] is rather low, $T_c \approx 0.12$ K. At the highest density we have considered, $\bar{n} = 8$, the dimensional density of DEs in the structure of Ref. \[14\] is rather large, $n \approx 1.2 \cdot 10^{11}$ cm$^{-2}$.

The trial wave function used for importance sampling in DMC is similar to the one used in our previous studies; for details of the calculation we refer the reader to Ref. \[28\].

For each density, we compute the ground state energy per exciton $E_0/N$ (see Fig. 1(a)). The DMC results are reproduced within a 0.025% accuracy by the polynomial fit (now we go to $h = m = x_0 = 1$ units, so, $n = \bar{n}$)

$$E_0/N = a_e \exp(b_e \ln n + c_e \ln^2 n + d_e \ln^3 n + e_e \ln^4 n), \quad (2)$$

with $a_e = 9.218$, $b_e = 1.35999$, $c_e = 0.011225$, $d_e = -0.00036$, and $e_e = -0.0000281$.

Using Eq. (2) we calculate the collisional contribution to the chemical potential $\mu_c = dE_0/dN$ and the inverse compressibility $1/\chi = d\mu_c/dn$ in terms of the quantities $\zeta'' = 2E_0/NT_0$, $\zeta' = \mu_c/T_0$, and $\zeta = n/XT_0$ are shown in Fig. 1(b). (Here $T_0 = g_x T_{\text{deg}} = 2\pi n$ is the degeneration temperature of the spin-polarized DEs and $T_{\text{deg}}$ is the degeneration temperature of real (spin-depolarized) DEs.)

We find that at densities $1/4 \leq n \leq 4$ the dimensionless compressibility $\zeta$ is $3 \leq \zeta \leq 8$, i.e., $\zeta \gg 1$. This is an evidence of the presence of strong correlations between 2D CQW DEs whose collective state was investigated in Refs. \[16, 17, 18\].

In Fig. 2(a), we show results for the polar-angle-averaged one-body density matrix

$$\rho_1(r) = \int_{0}^{2\pi} \langle \Psi^+(r) \Psi(0) \rangle d\varphi/2\pi.$$

Here $\Psi(r)$ is the exciton field operator and the brackets $\langle ... \rangle$ denote ground-state averaging.

The asymptotic behavior of $\rho_1(r)$ matches the result derived from a hydrodynamic calculation (see Fig. 2(d))
giving us additional confidence on the accuracy of the DMC simulation.

In Fig. 2(b) we plot DMC results of the Bose-condensate fraction $n_0/n$ and the corresponding polynomial fit on top of the data, also in terms of $\ln n$,

$$n_0/n = a_0^n \exp(b_0^n \ln n + c_0^n \ln^2 n + d_0^n \ln^{3} n),$$

with parameters $a_0^n = 0.3822$, $b_0^n = -0.2342$, $c_0^n = -0.02852$, and $d_0^n = -0.001594$. The results reveal a strong Bose condensate depletion at densities $1/4 \lesssim n \lesssim 4$. This is again a clear manifestation of strong correlations between 2D CQW DEs.

In Fig. 2(c), we show the results of DMC simulations for polar-angle-averaged excitonic pair distribution function

$$\rho_2(r) = \int_0^{2\pi} \langle \hat{\rho}(r)\hat{\rho}(0) \rangle d\varphi / 2\pi$$

($\hat{\rho}(r) = \hat{\Psi}^+(r)\hat{\Psi}(r)$ is the excitonic density operator). The prominent bump in the pair distribution function at densities $1/4 \lesssim n \lesssim 4$ is an evidence of both the short-range order and the strong correlations present in DE systems.

The structure factor $S(p)$ is related to the Fourier transform of the pair distribution function $\rho_2(r)$. We plot the DMC results of $S(p)$ in Fig. 3(a). A sharp peak in $S(p)$ at densities $1/4 \lesssim n \lesssim 4$ is consequence of the short-range order in 2D CQW DE systems.

Using the DMC results of the structure factor $S(p)$ one can predict an upper bound for the excitation spectrum by using the Feynman formula [30]

$$\epsilon_p \leq \frac{p^2}{2S(p)}.$$  

(see Fig. 3(c)). We see that the collective excitation spectrum of the 2D DE system is rather far from the Bogoliubov form (see Fig. 3(d)) in the range $1/4 \lesssim n \lesssim 4$. This fact suggests the relevant role of correlations in the excitonic collective state in CQWs.

We compare two calculations of sound velocity $c_s$ from the low-momentum structure factor slope ($c_s = p/2S(p \to 0)$; see Eq. (4)) and from the compressibility $1/\chi$ ($c_s = \sqrt{\chi}$), where $\chi$ is calculated by differentiation of the energy fit (2). The good agreement between the two calculations (see Fig. 3(b)) is expected from an exact calculation like the present one and supposes a crucial internal consistency check.

In Fig. 4(a), we show results derived from the Landau formula of the temperature dependence of the local (vortex-unrenormalized [31, 32, 33]) superfluid fraction of 2D quasi-condensed [34] DEs $n_{s}(T)/n$ far from the quasi-condensation crossover. Results for the Berezinskii-Kosterlitz-Thouless (BKT) superfluid transition temper-
Here, obtained for both functions are shown in Fig. 5(a).

We see that at densities $1/4 \lesssim n \lesssim 4$ the local superfluid fraction $n_s/n$ at $T \leq T_c$ is close to unity. Moreover, the quasi-condensation temperature $T_q$ at $1/4 \lesssim n \lesssim 4$ is 2-2.5 times higher than the degeneration temperature $T_{deg} = T_0/g_{ex} = 0.25T_0$ and the BKT superfluid transition temperature $T_c \approx 0.22T_0$ is only slightly smaller than $T_{deg}$. All these features are manifestations of the strong correlations between 2D CQW DEs. Besides, our results show that in real, strongly correlated, DE systems quasi-condensate and superfluid phases are much easier to achieve experimentally than if DEs were weakly correlated. (For weakly correlated DEs temperatures $T_c$ and $T_{deg}$ are logarithmically small in comparison with $T_{deg}$ [34, 57]).

Based on our DMC simulations of homogeneous infinite systems (Eqs. (2) and (3)) and applying local density approximation (LDA) we have calculated the total $n(R)$ and Bose-condensate $n_0(R)$ exciton profiles in a harmonic trap of large size ($\langle n(0) L_{TF}^2 \gg 1 \rangle$) at $T = 0$. Here, $L_{TF} = \sqrt{2N/\pi n(0)}$ is Thomas-Fermi (TF) radius and $N \gg 1$ is number of DEs in the trap. The results obtained for both functions are shown in Fig. 5(a).

If the electrostatic contribution is large $\zeta'_e \equiv \mu_e/T_0 = 2e^2D/\epsilon = 10$ [29] (this is its typical value for the experiments of Refs. [16, 17, 18]), the total profile is in very good agreement with the TF inverted parabola (see Fig. 5(a)) [38]. However, the Bose condensate profile at $1/4 \lesssim n \lesssim 4$ differs essentially from the inverted parabola (see Fig. 5(b)). The latter fact shows a failure of the theory of weakly correlated excitons for which the difference between Bose condensate profile and TF inverted parabola is logarithmically small [2].

Correlation effects in a dense 2D DE gas in CQWs can be detected by observation of the exciton luminescence in in-plane magnetic field. Indeed, in absence of magnetic field, the momentum of a recombined exciton $p$ according to the momentum conservation in one-photon exciton recombination is equal to the QW plane projection $\langle |h\omega/c_0| \sin \theta \rangle$ of the momentum of the emitted photon $h\omega/c_0$ [39]. Here $\theta$ is the angle between the normal and emitted photon in free space, $c_0$ is the light velocity in free space, and $\omega$ is the photon frequency. However, if there is in-plane magnetic field $H_{\parallel}$, the dispersion curve $\varepsilon_p$ is shifted by the quantity $p_{H} = eDH_{\parallel}/c_0$ [40]. This results in the following connection between the photon angle $\theta$ and the exciton momentum $p$:

$$\langle |h\omega/c_0| \sin \theta \rangle = |p - p_{H}|.$$  

(5)

If one considers only normal luminescence ($\theta = 0$), then $p = p_{H}$ [5], and thus $\varepsilon_p = \varepsilon_{pH}$. Hence, for spectral-angle luminescence along the normal in the field $H_{\parallel}$ one obtains

$$I_{\theta=0}^{H} = I_{\theta=0}^{H} \delta(\omega - \Omega + \varepsilon_{pH}/h) \quad (p_{H} \gg p_{T}).$$  

(6)
where $H_{\theta=0}^H$ is the spectrally integrated angle luminescence along the normal in the field $H_{\theta}$, $\Omega$ is the exciton resonance frequency for luminescence at $H_{\theta} = \theta = 0$, and $p_T = T/c_s$ is the typical thermal momentum of Bose condensed DEs which is the boundary between thermal $p \ll p_T$ and zero-temperature $p_T \ll p \ll h\sqrt{\hbar}$ regions of quasi-condensate phase fluctuations.

From Eq. (4), we find that the observation of the magnetic-field-induced normal excitonic luminescence line shift enables one to directly measure the excitation spectrum $\varepsilon_p$. In the structure of Ref. [40] ($D = 12.3$ nm) the density of the exciton gas is $n = 2 \cdot 10^{10}$ cm$^{-2}$, the field range $0 \leq H_{\theta} \leq 8$ T corresponds to the momentum range $0 \leq p_H/h \leq 1.5 \cdot 10^6$ cm$^{-1}$. This range covers both long-wave-length (hydrodynamic) and intermediate scales up to short-wave-length (ideal-gas) ones on which $\varepsilon_{ph} \approx p_H^2/2m$. Note that the thermal momentum $p_T$ at $T = 0.2T_0$ is $1 < T < T_0$, $m = 0.22m_0$ and $\zeta = 3.7$ (see above) corresponds to a field $H_{\parallel}^T = (c_0T/eD)^{\sqrt{m}/cT0} \approx 0.2$ T.

By measuring the slope of the excitation spectrum $\varepsilon_{ph}$ one can obtain sound velocity $c_s \approx \varepsilon_{ph}/p_H$ ($p_H \ll h\sqrt{\hbar}$; see Fig. 3(c)). The dimensionless exciton compressibility is then found as

$$\zeta = m^2c_s^2/2\pi\hbar^2n.$$  

Alternatively, one can determine the dimensionless compressibility $\zeta$ by measuring the magnetic field dependence of spectrally integrated angle luminescence along the normal

$$I_{\theta=0}^H = I_0\frac{q_e^2c_0}{4\pi eD\sqrt{mT_0}}\frac{\sqrt{\zeta}}{H_{\parallel}} \ (p_T \ll eDH_{\parallel}/c_0 \ll h\sqrt{\hbar}). \ (7)$$

Here $q_e = e\omega/c_0 \approx h\Omega/c_0$ is the radiation zone in free space, $I_0 = \hbar(h\Omega/\tau_0)n_0$ is the Bose-condensate luminescence at $T = 0$, $\tau_0$ is the lifetime of a spin-depolarized exciton at $T = 0$ in pure CQW structure, and $\kappa = 1/2$ ($\kappa = 1$) if the luminescence is measured on one (both) sides of CQW plane.

At $n = 2 \cdot 10^{10}$ cm$^{-2}$, $D = 12.3$ nm, $T = 0.1T_0 = 0.5$ K, $m = 0.22m_0$ and $\zeta = 3.7$ the momentum range $p_T \ll eDH_{\parallel}/c_0 \ll h\sqrt{\hbar}$ in (7) corresponds to the field range $(c_0T/eD)^{\sqrt{m}/cT0} \ll H_{\parallel} \ll c_0h\sqrt{\hbar}/eD$, or, $0.1$ T $\ll H_{\parallel} \ll 0.75$ T.

The Bose condensate luminescence at $T = 0$ entering into Eq. (7) can be approximately calculated as thermal-phase-fluctuation quasi-condensate luminescence at low temperatures

$$I_0 \approx \int_0^{p_T} I_{H_{\parallel}=0}^H \frac{2\pi p_H dp_H}{q_e^2} = \kappa\frac{\hbar\Omega}{T_0} \ (T \ll T_0), \ (8)$$

Finally, the measurement of the quantity $I_0$ enables one to determine an important parameter of the exciton interaction, the Bose-condensate density at $T = 0$

$$n_0 = \frac{1}{\kappa}\frac{\tau_0}{\hbar\Omega}I_0. \ (9)$$

In conclusion, Bose-Einstein condensation, superfluidity, and microscopic properties of 2D dipolar excitons in SQW and CQWs have been studied by $ab$ *initio* simulations and analytical calculations. In all the recent experiments on excitonic Bose condensation in CQWs the excitons are strongly correlated. We have shown that in a strongly correlated regime the excitonic collective state is much easier to be achieved than in weakly correlated DEs. Our calculation of the microscopic properties of 2D DEs enables one to investigate quantitatively QW excitonic Bose-Einstein condensation, superfluidity, luminescence and nonlinear optical effects by means of the hydrodynamic method in quantum field theory.

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In the following we will use usual (dimensional) units.

In 2D this property is specific for strong systems and it is not strongly correlated Bose systems.

For spin-depolarized exciton gas in $g_{ee}$ = 4 spin degrees.

See Refs. [11]a and [12].

The chemical potential has two contributions $\mu = \mu_e + \mu_c$. One is of electrostatic nature $\mu_e = 4\pi n e^2 D/\varepsilon$ and associated with the energy of a parallel-sided capacitor forming by charged $e$ and $h$ layers which are separated by the distance $D$. The other contribution $\mu_c$ is due to in-plane dipole-dipole collisions.

Inhomogeneous the large value of the electrostatic contribution $\mu_e$ results in an almost linear density dependence of the total chemical potential. This suppresses the nonlinear density dependence due to in-plane collisions $\mu_c$ and results in an almost linear overall dependence. So, the total density profile in the harmonic trap is close to TF inverted parabola.

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