Correlations in a two-dimensional Bose gas with long-range interaction

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received 11 May 2009; accepted in final form 15 July 2009
published online 18 August 2009

PACS 71.35.Lk – Collective effects (Bose effects, phase space filling, and excitonic phase transitions)
PACS 05.30.Jp – Bosonsystems
PACS 73.20.Mf – Collective excitations (including excitons, polarons, plasmons and other charge-density excitations)

Abstract – We study the correlations of two-dimensional dipolar bosons with excitons in semiconductor bilayers as a specific case, in the whole temperature-concentration region. We show that at low concentrations, the Bose degeneracy is accompanied by strong multi-particle correlations and the system behaves as a Bose liquid. At high concentrations the repulsion suppresses quantum coherence and the system behaves as a classical liquid down to a temperature lower than typical for a Bose gas. The interaction energy of the particles is a sensitive tool for a measurement of the correlations. This theory can apply to other systems of bosons with extended interaction.

Investigation of superfluidity of 4He and Bose condensation of alkali atoms stimulated the development of the theory of non-ideal Bose gases. The theory has been developed for contact and hard-sphere interactions that correspond to the hard-core repulsion between helium atoms and between alkali atoms. During the last few decades the search of Bose condensation of excitons in semiconductor heterostructures led to the study of a new type of a Bose system, namely, a system of indirect excitons in coupled quantum wells (sometimes referred to as bilayers) in which electrons and holes are located in different quantum wells [1–3]. Such an exciton system is not only two-dimensional but also the interaction between excitons is essentially a dipole-dipole repulsion that extends to a much larger distance than the exciton radius.

The purpose of the present letter is to demonstrate that the large range of the dipole-dipole repulsion compared to the contact interaction results in a dramatic difference of the system behavior. One of the main new features is that at any small concentration the setting in of the quantum degeneracy of the gas with the reduction of its temperature is accompanied by a setting in of multi-particle correlations. As a result the degenerate system is not a weakly non-ideal Bose gas but rather a Bose liquid. The other feature is that at higher concentration the repulsion between the bosons squeezes their wave functions and significantly reduces their overlap. As a result the temperature of setting of a quantum coherence is reduced compared to what is expected from a Bose gas. While to be specific we discuss dipolar excitons in bilayer semiconductor systems (see the upper insert in fig. 1), these qualitative conclusions remain valid for any Bose system with extended interaction and, in particular, for dipole molecules that are being investigated very actively [4].

The importance of the long-range repulsion on the particle correlations and thermodynamic phases has been recently demonstrated, using numerical methods, by Zimmermann and Schindler [5] for the case of dipolar excitons in bilayers and by Buchler et al for dipolar molecules [4]. In this letter we study the correlation between excitons in the whole temperature-concentration plane. In the most direct way this correlation reveals itself in the average interaction energy of each exciton $E_{int}$ that shows up in experiments as a luminescence blue shift. We evaluate this energy to demonstrate how the correlation affects its magnitude and dependence on relevant parameters. Our main goal is a clarification of the physical picture as well as to single out the main factors that control the
The interaction energy between excitons in coupled quantum wells is

\[
U(r) = \frac{2e^2}{\kappa} \left( \frac{1}{r} - \frac{1}{\sqrt{r^2 + L^2}} \right),
\]

where \(L\) is the separation between the centers of the wells and \(\kappa\) is the dielectric constant. According to refs. [5,6] the role of van der Waals and exchange interaction is small for practically important values of \(L (L > 10 \text{ nm})\) and we neglect them here. We assume that the exciton radius is smaller than the relevant distance between excitons that justifies the neglect of exchange interaction [7] and allows us to consider electrons and holes as point charges.

We assume also that \(nL^2 < 1\), where \(n\) is the exciton concentration. In the opposite case the electron-electron and hole-hole repulsion is stronger than the electron-hole attraction and it is hardly possible to expect a stable exciton phase [8]. For excitons in typical coupled quantum well systems, this means that \(n < 10^{12} \text{ cm}^{-2}\).

If any correlation between excitons is neglected then the average number of excitons in an area element \(d^2r\) is \(nd^2r\) and the average interaction energy is described by the “plate capacitor formula”:

\[
E_{\text{int}} = \int U(r) nd^2r = \frac{4\pi ne^2L}{\kappa}.
\]

This result means that the main contribution to \(E_{\text{int}}\) comes from the interaction between excitons at distance of the order of \(L\): \((e^2/\kappa L)(n\pi L^2) = \pi ne^2L/\kappa\). The same expression is obtained from a quantum-mechanical calculation in the mean-field approximation [7,9].

In reality, the correlation between excitons is not small. If one treats the exciton-exciton interaction classically, two excitons with an energy of the relative motion \(E\) can approach each other only to the distance larger than \(r_0(E)\), where \(U(r_0) = E\). Therefore, eq. (2) is valid only if the region of strong correlation is small, i.e., \(r_0 \ll L\). Since the average exciton energy is of the order of the temperature \(T\), the last condition is therefore equivalent to \(T \gg e^2/\kappa L\). This condition can also be written as \(\lambda_T < \pi\sqrt{2bL}\), where \(\lambda_T = \pi\hbar\sqrt{2/MT}\) is the thermal wavelength and \(b = \hbar^2\kappa/Me^2\). The expression for \(b\) differs from the Bohr radius \(a_B = \hbar^2\kappa/me^2\) only by replacement of the electron-hole reduced mass \(m = m_e m_h/(m_e + m_h)\) by the exciton mass \(M = m_e + m_h\). In GaAs/AlGaAs structures the electron effective mass \(m_e = 0.067\) and the heavy hole effective mass in a quantum well \(m_h = 0.14\). This gives \(M \approx 0.21\) and \(b \approx 3\) nm which justifies us to assume in further calculations that \(b \ll L\). Therefore, \(\lambda_T < \pi\sqrt{2bL}\) means also that \(\lambda_T < \pi L\) which justifies a classical description of the exciton-exciton interaction.

Interestingly, the validity of eq. (2) requires \(T \gg e^2/\kappa L\) \(\approx 140\) K for value of \(L \approx 10\) nm. On the other hand the exciton binding energy \(\epsilon_0\) puts an upper limit to the exciton temperature. For \(L \approx 10\) nm, \(\epsilon_0 \approx 4\) meV = 46 K, i.e., if \(T > e^2/\kappa L\) and \(n \approx 10^{10}\) cm\(^{-2}\) more than 90% of excitons under such conditions would be ionized and therefore the validity of eq. (2) is in fact quite limited for such excitonic systems.

When \(T\) decreases and crosses the value of \(e^2/\kappa L\), the minimal distance between excitons \(r_0\) becomes larger than \(L\) and therefore it becomes the characteristic length scale of the exciton-exciton interaction. At this stage eq. (1) can be simplified:

\[
U(r) = \frac{e^2L^2}{\kappa r^3}, \quad r_0(T) = \left(\frac{e^2L^2}{\kappa T}\right)^{1/3}.
\]

At \(r \sim r_0\) the exciton correlation is strong and

\[
E_{\text{int}} = n \int U(r) g(r)d^2r,
\]

where \(g(r)\) is the pair correlation function.

We now introduce two important dimensionless quantities: the first is the “quantum parameter”: \(r_0/\lambda_T\), which is a measure of the quantum character of the interaction, and the second is the gas parameter: \(n\lambda_T^2\), which describes the “diluteness” of the exciton gas. As \(T\) becomes smaller than \(e^2/\kappa L\) correlations become important, but the exciton wavelength is still smaller than \(r_0\), i.e., \(r_0/\lambda_T \ll 1\). At this stage, the interaction between excitons can be considered classically. In this case \(g(r) = e^{-U(r)/T}\) (see, e.g., ref. [10], sect. 32) which results in a \(T\)-dependent interaction energy given by

\[
E_{\text{int}} = \frac{4\pi ne^2L}{\kappa} f_E \left(\frac{e^2}{\kappa LT}\right).
\]

The function \(f_E(x)\) is shown in the lower insert in fig. 1. If \(T \gg e^2/\kappa L\) then \(f_E = 1\) and eq. (5) becomes identical.
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to eq. (2). In the case where $T$ is sufficiently small, i.e., $T \ll e^2/\kappa L$, $E_{\text{int}}$ becomes

$$E_{\text{int}} = 2\pi \Gamma(4/3)n \left( \frac{e^2L^2}{\kappa} \right)^{2/3} T^{1/3} = 2\pi \Gamma(4/3)n r_0^2 T.$$ (6)

This result can be understood in the following way. Around each exciton there is a depletion region of radius $r \sim r_0$. Without repulsion this region would contain $n r_0^2$ excitons with an average energy $T$. The energy necessary to force all of them out of this region is of the order of $n r_0^2 T$.

Further reduction of temperature can violate one of the two conditions of validity of eq. (5). First, $r_0/\lambda_T \propto T^{1/6}$ can become smaller than unity, i.e., the exciton wavelength becomes larger than the potential length scale and the pair correlation has to be described quantum mechanically. Secondly, the gas parameter $nr_0^2 \propto 1/T^{2/3}$ can become larger than unity. This makes multi-particle scattering and multi-particle correlations important, which is characteristic not for a gas but for a liquid. Which violation happens first depends on the concentration, as is discussed in what follows:

a) If $n \ll (b/2L^2)^2$ then with a decrease of the temperature the exciton wavelength $\lambda_T$ becomes of the order of and then larger than $r_0$ while $r_0$ is still much smaller than the average distance between excitons (i.e., $nr_0^2 \ll 1$). In this case the excitons can be still viewed as a correlated gas but the interactions should be considered quantum mechanically. The wave function $\psi(r)$ describing relative motion of two excitons penetrates under the repulsion barrier between them and the effective minimal distance is now smaller than $r_0$. Given the energy of relative motion $E$, the probability to find one exciton at the distance $r$ from the other is $\langle |\psi(r)|^2 \rangle_E$, where $\langle \ldots \rangle_E$ means the average over all directions of the wave vector keeping $E$ constant. For a non-degenerate exciton gas the probability density for an exciton to have energy $E$ is $(1/T)e^{-E/T}$. That is

$$g(r) = \frac{1}{T} \int_0^\infty \langle |\psi(r)|^2 \rangle_E e^{-E/T} dE,$$ (7)

where $\psi(r)$ has to be normalized in such a way that $g(r)|_{r \to \infty} = 1$ (see footnote 1). When $T$ is low enough such that $\lambda_T \gg r_0$ (but still within the classical statistics limit, $\lambda_T \ll n^{1/2}$) $\psi(r)$ falls exponentially at $r < L^2/b$ and is a slow function of $r$ at $r > L^2/b$. The interaction potential falls like $U(r) \propto 1/r^3$ and therefore the main contribution to the integral in eq. (4) comes from the region $r \sim L^2/b$. The exact solution to the Schrödinger equation with potential $\propto 1/r^3$ in this region is

$$\psi(r) = -\frac{2}{\ln(kL^2/b)} K_0 \left( \frac{2L}{\sqrt{br}} \right),$$ (8)

$$k = \sqrt{ME}/\hbar.$$ With a logarithmic accuracy it is possible to put $k \approx 1/\lambda_T$. Then

$$g(r) = \frac{4n}{\ln^2(L^2/\lambda_T k)} K_0^2 \left( \frac{2L}{\sqrt{br}} \right),$$ (9)

and substitution of eq. (9) in eq. (4) results in

$$E_{\text{int}} = \frac{2\pi \hbar^2 n}{M \ln^2(L^2/\lambda_T k)}.$$ (10)

Note that in all cases considered so far $E_{\text{int}}$ is larger than the interaction energy at the average distance between excitons. This means that the main contribution to $E_{\text{int}}$ comes from rare fluctuations of the interaction energy and these fluctuations are of the order or larger than the average value. It is intriguing that while at higher temperatures, where the interaction is classical, there is a clear temperature dependence of $E_{\text{int}}$, as can be seen from eq. (5), at lower temperatures, quantum interactions yield an almost $T$-independent $E_{\text{int}}$, quite similar to the mean-field result, but with a different coefficient.

The gas parameter in the above quantum case is different from the classical $nr_0^2$ one. The exciton-exciton scattering cross-section is $\sigma = \pi/2k \ln^2(kL^2/b)$ (in the 2D case it has units of length). In other words, $\sigma$ is of the order of the wavelength although the potential length scale $L^2/b$ is smaller than the wavelength. Respectively the mean free path of excitons is $l \sim \ln^2(L^2/b\lambda_T)/n\lambda_T$. The gas condition in the quantum case corresponds to the absence of correlation between different scattering events which means that the wavelength has to be much smaller than the mean free path, i.e., $n \ll \ln^2(L^2/b\lambda_T)/\lambda_T^2$. This inequality is identical to $E_{\text{int}}/T \ll 1$ and also with the condition of non-degeneracy with the accuracy of the logarithmic correction. Practically, the logarithm is not large compared to unity and hence the gas state of the exciton system is non-degenerate while the degeneracy is accompanied by strong interactions and multi-particle correlations between the excitons. This is an important outcome of the above analysis.

b) On the other hand, if $n \gg (b/2L^2)^2$ then a decrease of the temperature leads to a violation of the condition $nr_0^2 \ll 1$ while $r_0 \gg \lambda_T$. That is, quantum corrections are negligible and the system behaves as a classical liquid. The dimensional analysis gives

$$E_{\text{int}} = \frac{e^2L^2}{\kappa} n^{3/2} f \left( \frac{e^2L^2}{\kappa T} n^{3/2} \right)$$ (11)

and according to eq. (6) $f(x) = 2\pi \Gamma(4/3)x^{-1/3}$ at $x \ll 1$.

When with growth of the concentration or reduction of the temperature $nr_0^2$ comes close to unity, a free motion of excitons between collisions becomes impossible because each of them is confined in between its neighbors.
This classical picture is valid as long as the size of the confinement region is much larger than the exciton thermal wavelength. In other words the exciton state in a potential well formed by its neighbors is highly excited. The size of the potential well $R \sim n^{-1/2}$ and the motion is semi-classical as long as $\lambda_g \ll R$. The energy at the bottom of the potential well $\sim e^2 L^2 / R^3$ ($z$ is the number of nearest neighbors) is of the same order as the depth of the well. Potential wells for different excitons are different, they are not static and sometimes some excitons overcome or tunnel across the surrounding barrier. But at $nr^2_0 > 1$ these rare occasions do not affect the estimates. In general, this picture is similar to a simple classical liquid and the formation of the potential wells is the starting point of a formation of a short-range order characteristic for liquids [11]. Further reduction of temperature brings particles to lower levels in the potential wells and makes the potential wells more stable. Stronger confinement of the wave functions of each exciton reduces their overlap. At $nr^2_0 > 1$ the energy of the bottom of the well is larger than the temperature and it is the main part of the interaction energy. Eventually at $nr^2_0 \gg 1$ most of the particles are at the ground state in their potential wells and rough estimates can be done assuming that a short-range order has been formed and each exciton is inside a unit cell of a crystal. (We emphasize that we mean formation of short-range order typical in liquids [11] but not crystalization and formation of long-range order [12].) Then the potential energy of an exciton that is displaced by small distance $r$ from the center of the unit cell is

$$U_{cr}(r) \approx \frac{e^2 L^2}{\kappa R^3} \left( C_1 + \frac{C_2}{r^2} \right), \quad (12)$$

and $R = [n \sin(2\pi/z)]^{-1/2}$ is the radius of the first coordinate circle. The values of the constants can be estimated assuming a crystal order within the first few coordinate circles and a random distribution outside. The dependence of the correlation radius on the order appears quite weak and for hexagonal unit cell $C_1 \approx 10.5$, $C_2 \approx 15$ (compare ref. [13]). If $T \sim (e^2 L^2 / \kappa) n^{3/2}$ (i.e., $nr^2_0 \sim 1$) then $r \sim R$ and there is no short-range order in the system. However, if $T \ll (e^2 L^2 / \kappa) n^{3/2}$ then the short-range order does exist and $r \ll R$. The energy of the ground state in the potential (12) above its bottom is $\sim \sqrt{2C_2(h^2 / ML^2)}(e^2 L^2 / \kappa R^3)$ and quantization is important when the temperature is of the order or smaller than this value, i.e., $T \lesssim (h^2 n / M) \sqrt{2C_2} (L^2 n^{1/2} / b)$. The order of magnitude of $r$ is controlled by the temperature or quantization and in any case, when the short-range order does exist the size of a single-exciton wave function is much smaller than the distance between excitons.

The inequality $r \ll R$ allows us to make two conclusions. First, the bottom of the potential of eq. (12) gives a good estimate for the interaction energy

$$E_{int} \approx 8e^2 L^2 n^{3/2} / \kappa. \quad (13)$$

Second, it is possible to estimate the overlap of the wave functions of adjacent excitons. If $L = 10 \text{nm}$ and $n = 3 \times 10^{11} \text{cm}^{-2}$ the estimate according to the wave function in the harmonic potential of eq. (12) gives for the overlap a value of $\sim 0.2$. The actual value is even smaller because at $r \sim R$ the potential barrier is steeper than harmonic one. Due to the small wave function overlap the temperature at which the spontaneous phase or/and spin coherence [14] in the exciton system is set in is reduced compared to its expected value $\sim h^2 n / M$. This points to a non-monotonic dependence of the quantum coherence onset temperature on the concentration, and it suggests that a lower-density exciton system becomes quantum coherent at higher temperatures than a higher-density system, which is a priori non-intuitive.

All estimates made above open the possibility to make up a general picture that demonstrates the role of particle correlations in a dipolar exciton system at the whole $(n,T)$-plane. This picture is presented in fig. 2. Correlation is not important and the mean-field approximation is applicable only in region I. In region II the exciton system can be considered as a classical gas with strong pair correlations. The difference between region III and II is that in the former exciton-exciton scattering is described by quantum mechanics that changes the pair correlation function. Reduction of the temperature from region III to region IV leads to degeneracy of the exciton system. But simultaneously a strong many-particle correlation is set up. The system cannot be considered as gas, and the mean free path does not exist. Rather surprising is the existence of region V where the system behaves as a classical liquid down to temperatures much lower than the
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degeneracy temperature in the case of contact interaction $\sim \hbar^2 n / M$. The reason is that strong repulsion between excitons squeezes the wave function of each exciton to a size smaller than the average distance between excitons. In this region there is short-range order and with a reduction of temperature the correlation radius grows, i.e., the system may crystallize.

The existence of region V is the result of the tail of the dipole-dipole repulsion. In case of contact interaction an overlap of particle wave functions competes with the repulsion and region V does not exist. This latter case applies, for example, to excitons in one well where an increase of the concentration leads to the Mott transition.

Crossing of one of the lines in fig. 2 by changing temperature or concentration leads to a gradual change of the correlation between excitons, not to a sharp transition. A typical dependence of $E_{int}$ on temperature in the whole temperature region shown in fig. 1.

In conclusion, along with the degeneracy of a 2D Bose system with dipole-dipole interaction a strong multi-particle correlation sets in. At high concentration the repulsion suppresses, by up to a few times, the temperature at which quantum coherence is set in. The interaction energy has different value and different dependence on $T$ or/and $n$ in different regions and is a sensitive tool for measurement of the correlations.

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BL appreciated discussions with M. Stern.

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