

# Exciton correlations in coupled quantum wells and their luminescence blue shift

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In this paper we present a study of an exciton system where electrons and holes are confined in double quantum well structures. The dominating interaction between excitons in such systems is a dipole-dipole repulsion. We show that the tail of this interaction leads to a strong correlation between excitons and substantially affects the behavior of the system. Making use of qualitative arguments and estimates we develop a picture of the exciton-exciton correlations in the whole region of temperature and concentration where excitons exist. It appears that at low concentration degeneracy of the excitons is accompanied with strong multiparticle correlation so that the system cannot be considered as a gas. At high concentration the dipole-dipole repulsion suppresses the quantum degeneracy. As a result there exists a temperature region where the system behaves a classical liquid; such a region does not exist in case of contact interaction. We calculate the blue shift of the exciton luminescence line which is a sensitive tool to observe the exciton-exciton correlations.

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## I. INTRODUCTION

A very active investigation of excitons in coupled quantum wells for more than two decades was first motivated by the possibility to reach Bose condensation and superfluidity in this system. Further experiments discovered a very large number of related phenomena and quite rich physics of the system (see Refs. 1–3 and references therein). Theory predicts the existence of many phases with different and unusual properties.<sup>4–7</sup> The most expected and searched for is the settling in of coherence in such two-dimensional (2D) exciton systems. A coherence of the exciton Bose condensate has to reveal itself in some coherent properties of the exciton luminescence. Investigation of the luminescence led to discovery of not only its coherence<sup>8,9</sup> but also a number of patterns not completely understood so far.<sup>10–13</sup>

A substantial role in these phenomena is played by the interaction between excitons. Typically, interaction between bosons (<sup>4</sup>He atoms and alkali atoms) is of a short range and the theory of nonideal Bose gas has been developed for contact interaction.<sup>14,15</sup> In two coupled quantum wells where electrons are confined in one well and holes are confined in the other excitons formed by the coulomb binding of spatially separated charges have a dipole moment. Dipole moments of all excitons are directed in the same way and the interaction between the excitons is mainly dipole-dipole repulsion, Fig. 1. Given separation between the centers of the wells,  $d$ , the dependence of this repulsion on the widths of the wells is very weak<sup>16</sup> and with a good accuracy the repulsion is the same as interaction between pairs of point-like positive and negative charges separated by distance  $d$ , Eq. (2.1). Contrary to the contact interaction the dipole-dipole interaction has a significant tail and due to this tail the exciton gas in some respects is dramatically different from Bose gas with contact-like interactions.

The dipole-dipole repulsion increases the exciton energy and leads to a blue shift of its luminescence line. The blue shift is usually evaluated as an average value of the exciton interaction energy with other excitons and can be easily obtained with the help of the mean-field approximation that produces “plate capacitor formula,”<sup>17</sup>

$$E_{\text{int}} = \frac{4\pi n e^2 d}{\kappa}, \quad (1.1)$$

where  $n$  is the exciton 2D concentration,  $d$  is the separation between the centers of the wells, and  $\kappa$  is the dielectric constant. This formula can be understood in the following way. Equal concentration  $n$  of electrons and holes in two wells creates potential difference  $\Delta\phi = 4\pi n e d / \kappa$  between them. Creation of one more indirect exciton in these wells requires transfer of an electron or a hole from one well to the other. In the presence of other excitons this increases the necessary energy by  $e\Delta\phi$  that gives Eq. (1.1). This expression is typically used in experiments for an estimate of the exciton concentration from the measured blue shift of the luminescence.

Recently Zimmermann and Schindler<sup>18</sup> noticed, however, that dipole-dipole repulsion leads to a significant exciton pair correlation. The repulsion prevents excitons to come very close to each other and creates a depleted region around each exciton. The pair correlation leads to a reduction in the coefficient in Eq. (1.1) by about 10 times (depending on the excitons temperature).<sup>18</sup> This means that previous experimental estimates of the exciton concentration based on the capacitor formula underestimated the concentration by up to 10 times.

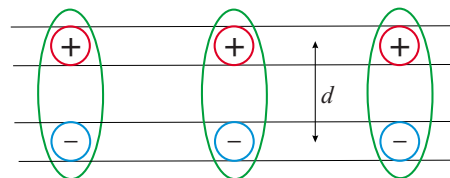


FIG. 1. (Color online) Spatially indirect excitons in coupled quantum wells. Holes are confined in one well while electrons are confined in the other.  $d$  is the separation between the centers of the wells. Given  $d$  the dependence of the exciton-exciton interaction on the well widths (i.e., on the vertical size of the electron and hole wave functions) is very weak. (Ref. 16)  $d$  and the average separation between excitons are assumed to be larger than the exciton radius  $a_X$ .

TABLE I. The main parameters characterizing an exciton gas in coupled quantum wells.

$d$	Separation between electron and hole wells	$a_B$	Bohr radius with a reduced electron—hole mass
$m_e, m_h$	Electron and hole masses	$b$	Bohr radius with an exciton mass
$m$	Reduced electron-hole mass	$a_X$	Exciton radius
$M$	Exciton mass	$r_0$	Classical minimal distance between excitons
$\epsilon_b$	Exciton binding energy	$k_T$	Thermal wave vector of an exciton
$n$	Exciton concentration	$U(r)$	Interaction energy between excitons
$E_{\text{int}}$	$X$ - $X$ interaction energy per $X$	$g(r)$	Exciton pair correlation function

It makes sense to note that the reduction in the pair correlation function to zero at small distance in a Bose gas with repulsion has been noticed long ago and used in the construction of a many particle variational wave function.<sup>19,20</sup> It is well known in exact solutions for one-dimensional Bose gas.<sup>21–24</sup> In the exciton gas with dipole-dipole repulsion this behavior of the pair correlation function was noticed by Astrakharchik *et al.*<sup>7</sup> This behavior was also used by Kash *et al.* to explain a narrowing of the exciton luminescence line at low temperature.<sup>25</sup>

The significant pair correlation between excitons substantially affects our understanding of the behavior of this system. This motivated us to develop a general picture of the interaction and correlation between excitons in the whole temperature-concentration plane. This is the subject of this paper. We show that Eq. (1.1) has a very limited region of applicability and limited accuracy. With a decrease in the temperature and an increase in the concentration the role of the pair correlation becomes crucial for interpretation of such phenomena as the blue shift. It appears also that the degeneracy of the exciton system at any small concentration is accompanied by setting in of not pair but multiparticle correlation characteristic for liquids. In other words, a consistent theory of a quantum coherent state has to include multiexciton correlation at any dilute gas. More than this, at higher concentrations the dipole-dipole repulsion reduces the overlap of the wave functions of different excitons. As a result the exciton system can be considered as classical liquid down to temperature well below than the temperature of quantum degeneracy of a system with contact interaction.

Our main purpose is to develop a qualitative understanding of the structure of the system of interacting bosons. Therefore we do not pursue a high accuracy of the results but instead use simplified models and qualitative arguments. Although the results of such an approach are really accurate only in extreme cases with respect to some large or small parameters, they allow us to make analytical calculations and produce a very clear physical picture of relevant phenomena. Availability of such a picture facilitates precise calculations when they are necessary.

In the next section we consider in detail the applicability of the mean-field approximation that neglects any correlation between excitons. An exciton gas at lower temperatures where strong pair correlation is important but quantum correlations are still negligible is considered in Sec. III. Further reduction in temperature at low exciton concentrations, when the exciton wavelength becomes larger than the characteristic scale of exciton-exciton interaction, leads to an important

role of quantization in the exciton-exciton scattering while the exciton gas itself is still statistically nondegenerate. This situation is considered in Sec. IV. In Sec. V we consider the situation where multiexciton correlation is important. This happens at low enough temperature in a dilute system and in a more wide range of temperatures in a dense system. It appears that with a temperature decrease in a dilute system degeneracy is accompanied by a set in of a multiexciton correlation. Contrary, in a dense system the classical multiexciton correlations appear to be more important for the blue shift than the quantization of exciton dynamics. In the last section we discuss the obtained results.

The discussion of exciton correlation in different parts of the concentration—temperature plane involves quite a large number of physical parameters. To facilitate reading of the paper we present here the list of main parameters with their physical definitions (Table I). Exact mathematical definitions, if necessary, will be given as soon as the corresponding parameter comes into the discussion.

## II. MEAN-FIELD APPROXIMATION

The interaction energy between excitons in coupled quantum wells is

$$U(r) = \frac{2e^2}{\kappa} \left( \frac{1}{r} - \frac{1}{\sqrt{r^2 + d^2}} \right). \quad (2.1)$$

According to Refs. 4, 16, and 18 the attractive Van der Waals interaction and the exchange interaction are small in practically important values of  $d$ . The simplest way to obtain the average interaction energy is to assume that all excitons are distributed randomly and independently of each other with an average concentration  $n$ , see Fig. 2. Then the average number of excitons in an area element  $d^2\mathbf{r}$  is  $nd^2\mathbf{r}$  and the average interaction energy is

$$E_{\text{int}} = \int U(r) nd^2\mathbf{r} = \frac{4\pi ne^2 d}{\kappa}. \quad (2.2)$$

This result means that the main contribution to  $E_{\text{int}}$  comes from the interaction between excitons at distance of the order of  $d$ :  $(e^2/\kappa d)(n\pi d^2) = \pi ne^2 d/\kappa$ .

Equation (2.2) is valid only under a few assumptions. The first is that the average distance between excitons has to be larger than  $d$ . In other words, the concentration cannot be too large,

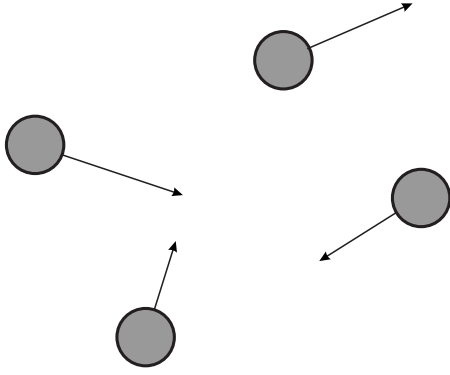


FIG. 2. Uncorrelated motion of excitons. The radius of each circle depicts the characteristic scale of the repulsive potential  $d$ . A random motion takes place only at low concentrations, Eq. (2.3), and rather high temperatures, Eq. (2.5).

$$nd^2 \ll 1. \quad (2.3)$$

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 of the system.<sup>26-29</sup> In all following considerations we assume that inequality Eq. (2.3) is met. Practically, this limitation in coupled well structures with  $d \geq 10$  nm means that the concentration has to be much smaller than  $10^{12}$  cm<sup>-2</sup>.

The other main assumption used in the derivation of Eq. (2.2) is the absence of any correlation between excitons. In reality, if the kinetic energy of the relative motion of any two excitons is  $E$  they can approach each other only to a distance larger than  $r_0(E)$  where  $r_0$  is the root of the equation

$$U(r_0) = E. \quad (2.4)$$

The correlation between excitons can be neglected only if the region where the correlation is important is very small, i.e.,  $r_0 \ll d$ . If the temperature of the exciton gas is  $T$  then  $E \sim T$  and this condition can be written as

$$T \gg e^2/\kappa d. \quad (2.5)$$

Condition (2.5) is equivalent to  $k_T \gg 1/\sqrt{bd}$  where  $k_T = \sqrt{2MT/\hbar}$  is the exciton thermal wave vector and  $b = \hbar^2 \kappa / M e^2$ . The expression for  $b$  differs from the Bohr radius  $a_B = \hbar^2 \kappa / m e^2$  only by replacement of the electron-hole reduced mass  $m = m_e m_h / (m_e + m_h)$  with the exciton mass  $M = m_e + m_h$ . In GaAs/AlGaAs structures the electron effective mass  $m_e = 0.067$  and the hole effective mass at the bottom of the hh1 subband in a quantum well  $m_h = 0.14$ . This gives  $m \approx 0.045$ ,  $M \approx 0.21$ , and therefore  $a_B \approx 14$  nm, and  $b \approx 3$  nm which allows us to assume in further calculations that  $b \ll d$ . Therefore condition (2.5) means also that  $k_T \gg 1/d$ , i.e., the exciton wavelength is much smaller than the characteristic length scale of the potential. This justifies a classical consideration of the interaction between excitons. Also, this inequality in combination with Eq. (2.3) leads to the inequality  $k_T^2 \gg n$  which means that the exciton thermal wavelength is much smaller than the average interparticle distance and therefore the exciton gas is nondegenerate.

It makes sense to note that a quantum mean field approximation also leads to Eq. (2.2).<sup>16,30-32</sup> However, if the exciton radius  $a_X$  is of the order of or larger than  $d$  the exchange interaction also appears to be important.<sup>16</sup> Variational calculations for GaAs/AlGaAs structures with an exciton wave function  $\psi(r) = A e^{-\sqrt{d^2+r^2}/2a_X}$  yields, for layers separation of  $d = 10, 12,$  and  $14$  nm the corresponding exciton radii of  $a_X = 8.7, 9.3,$  and  $9.7$  nm, and exciton binding energies of  $4.7, 4.2,$  and  $3.8$  meV respectively. That is although  $d > a_X$  this inequality is not very strong and there are quantum corrections to the interaction in Eq. (2.1).<sup>16,18</sup>

The exciton binding energy also puts an upper limit to the temperature where the above mean-field description is viable because at temperatures of the order of or larger than the binding energy the majority of the excitons dissociate. According to Eq. (2.5) the low limit for the temperature for  $d = 10, 12,$  and  $14$  nm is  $54$  K ( $4.7$  meV),  $48$  K ( $3.8$  meV), and  $44$  K ( $8.6$  meV), respectively.

The bottom line of these estimates is that Eq. (2.2) is a rather poor estimate: it is really valid only in the temperature range where a significant part of excitons is dissociated.

To conclude this section we comment on a usage of the mean-field approximation in explaining phenomena other than the exciton luminescence blue shift. The interaction of one of the particles with all others can be described with the field created at the particle by the environment. In general, this field fluctuates in time and from particle to particle due to different dynamics of particles creating it. If the particle interacts simultaneously with many others and they are not correlated then these fluctuations are cancelled and their resulting amplitude is much smaller than the average value of the field. This is the foundation that makes the mean-field approximation valid. In the exciton gas with dipole-dipole interaction the situation is quite different. The average interaction energy [Eq. (2.2)] is much larger than the interaction between excitons at average distance between them:  $U(n^{-1/2}) \sim e^2 d^2 n^{3/2} / \kappa$  and  $U(n^{-1/2}) / E_{\text{int}} \sim (nd^2)^{1/2} \ll 1$ . This means that the main contribution to  $E_{\text{int}}$  comes from rare pairs of excitons with the distance much smaller than the average one. The large amplitude of the field fluctuations is also confirmed by the calculation of the average of the interaction energy squared,

$$\overline{U^2} = \int U^2(r) n d^2 \mathbf{r} = 2\pi \left( \frac{2e^2 n^{1/2}}{\kappa} \right)^2 \ln \frac{d}{r_0}. \quad (2.6)$$

Discarding the logarithm that comes from the cutoff of the minimal distance between excitons we see that  $E_{\text{int}}^2 / \overline{U^2} \sim nd^2 \ll 1$ . (Under this condition  $\overline{U^2}$  characterizes the luminescence line width, but a discussion of this point is beyond the scope of the paper.)

The message is that while the mean field approximation for the average interaction gives a correct result in the mentioned range of parameters, the calculation of other quantities in this approximation can lead to large errors. It is also important to keep in mind that in spite of the purely classical arguments this statement is true also in the quantum limit.

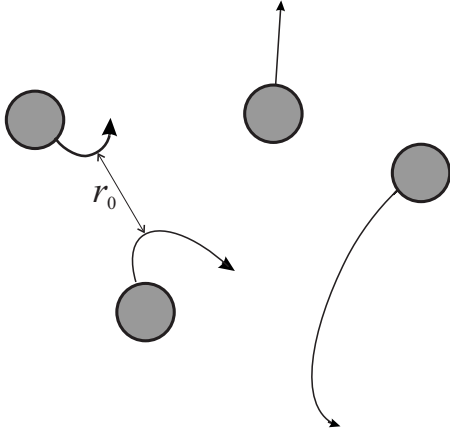


FIG. 3. At low temperatures,  $T < e^2/\kappa d$ , the exciton kinetic energy cannot overcome the repulsion between the excitons. The minimal distance between excitons in average is larger than  $r_0$  defined in Eq. (2.4) where the energy  $E \sim T$ . The correlations between excitons cannot be neglected.

### III. CLASSICAL EXCITON GAS

When the temperature goes down at some point it becomes smaller than the Coulomb interaction at distance  $d$  that violates the condition of the mean-field approximation, Eq. (2.5), and the correlation of excitons cannot be neglected anymore, Fig. 3. At  $T < e^2/\kappa d$  the relevant parameter characterizing the scale of the interaction potential is not  $d$  but the minimal distance between excitons  $r_0$  because  $r_0 > d$ . At the temperature where  $r_0$  crosses the value of  $d$ , condition (2.3) leads to

$$nr_0^2 \ll 1. \quad (3.1)$$

This means that right below the temperature  $T \sim e^2/\kappa d$  there exist a region where the average distance between excitons is larger than  $r_0$  and it is possible to take into account only pair correlations because the probability to find three excitons in mutual proximity is negligible. The condition  $r_0 \sim d$  (i.e.,  $T \sim e^2/\kappa d$ ) means also that  $k_T r_0 \sim k_T d \gg k_T \sqrt{db} \sim 1$  (the last two relations follow from  $d \gg b$  and  $T \sim e^2/\kappa d$ , respectively) and the interaction between excitons can be still considered classically. The same relations mean that  $k_T^2 \gg n$ , i.e., the exciton gas is nondegenerate. A growth of  $r_0$  compared to the exciton radius also makes the exchange corrections to interaction (2.1) less important. As a result at the temperature region right below  $e^2/\kappa d$  it is possible to consider excitons as classical particles.

The blue shift in this temperature and concentration region can be evaluated as

$$E_{\text{int}} = n \int U(r)g(r)d^2r, \quad (3.2)$$

where  $g(r)$  is the pair correlation function. For any given exciton,  $ng(r)d^2r$  is the average number of excitons within area  $d^2r$  at distance  $r$  from it. According to this definition  $g(r)|_{r \rightarrow \infty} = 1$  because at large distance any correlation between excitons disappears. In the leading order in  $nr_0^2$

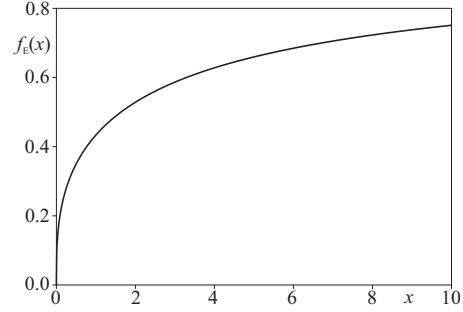


FIG. 4. A plot of dimensionless function  $f_E(x)$  of Eq. (3.5) that defines the blue shift at low exciton concentrations, Eq. (3.1), and moderate temperatures, Eq. (3.14).

$$g(r) = e^{-U(r)/T} \quad (3.3)$$

[see, e.g., Ref. 33, Sec. 32]. Substitution of Eq. (3.3) into Eq. (3.2) gives

$$E_{\text{int}} = n \int U(r)e^{-U(r)/T}d^2r. \quad (3.4)$$

With the interaction Eq. (2.1) this expression is reduced to a function of only one parameter,

$$E_{\text{int}} = \frac{4\pi n e^2 d}{\kappa} f_E\left(\frac{\kappa d T}{e^2}\right). \quad (3.5)$$

Function  $f_E(x)$  is plotted in Fig. 4. The asymptotical behavior of this function is

$$f_E(x) = \begin{cases} 1 + \frac{1}{x} \left( \ln \frac{8}{x} + C - 1 \right), & x \gg 1 \\ \frac{\Gamma(4/3)}{2} x^{1/3}, & x \ll 1, \end{cases} \quad (3.6)$$

where  $C \approx 0.577$  is the Euler constant. At high temperature, Eq. (2.5), the expression for  $E_{\text{int}}$  (3.5) is reduced to the mean-field expression of Eq. (2.2). In the opposite limit, where  $r_0 \gg d$ , the interaction potential is simplified:

$$U(r) = \frac{e^2 d^2}{\kappa r^3}, \quad r \gg d, \quad (3.7)$$

and Eq. (3.4) gives

$$E_{\text{int}} = 2\pi\Gamma(4/3)n \left( \frac{e^2 d^2}{\kappa} \right)^{2/3} T^{1/3} = 2\pi\Gamma(4/3)nr_0^2 T, \quad (3.8)$$

where  $r_0 = (e^2 d^2 / \kappa T)^{1/3}$ . Qualitatively this result can be understood in the following way. Around each exciton there is a depletion region with a radius  $\sim r_0$ . Without repulsion this region would contain  $nr_0^2$  excitons with average energy  $T$ . The energy necessary to force all of them out of the region is of the order of  $nr_0^2 T$ .

One can expect that  $E_{\text{int}}$  is of the order of the first virial correction to the chemical potential of the exciton gas. This can be easily checked. The first two terms of the virial expansion give (Ref. 33, Sec. 23)



$$n = \frac{2mT}{\pi\hbar^2} e^{\zeta/T} + \left( \frac{2mT}{\pi\hbar^2} e^{\zeta/T} \right)^2 \int (e^{-U(r)/T} - 1) d^2\mathbf{r}. \quad (3.9)$$

Solution of this equation with respect to the chemical potential  $\zeta$  leads to

$$\zeta = \zeta_0 + \Delta\zeta, \quad \zeta_0 = T \ln \frac{\pi\hbar^2 n}{2MT}, \quad (3.10)$$

and

$$\Delta\zeta = -nT \int [e^{-U(r)/T} - 1] d^2\mathbf{r} = \frac{2\pi n e^2 d}{\kappa} f_\zeta \left( \frac{\kappa d T}{e^2} \right), \quad (3.11a)$$

$$f_\zeta(x) = x \int_0^\infty \left\{ 1 - \exp \left[ -\frac{2}{x} \left( \frac{1}{t} - \frac{1}{\sqrt{t^2 + 1}} \right) \right] \right\} t dt. \quad (3.11b)$$

In extreme cases

$$\Delta\zeta = \begin{cases} \frac{4\pi n e^2 d}{\kappa}, & T \gg \frac{e^2}{\kappa d} \\ \pi\Gamma(1/3) \left( \frac{e^2 d^2}{\kappa} \right)^{2/3} n T^{1/3}, & T \ll \frac{e^2}{\kappa d}. \end{cases} \quad (3.12)$$

That is  $E_{\text{int}}$  and  $\Delta\zeta$  have the same dependence on parameters and differ only by a constant factor. This difference results from the difference of the definitions:  $E_{\text{int}}$  is the correction to the average energy released in an exciton recombination while  $\Delta\zeta$  is the correction to the energy released when an exciton is removed without a violation of the equilibrium in the exciton gas.

Finally, we note that in the case of  $T \ll e^2/\kappa d$  when  $E_{\text{int}}$  is given by Eq. (3.8), the interaction at the average distance  $U(n^{-1/2})$  is still small compared to  $E_{\text{int}}$ :  $U(n^{-1/2})/E_{\text{int}} \sim n^{1/2} r_0 \ll 1$ . The mean-field approximation then gives correct expression only for the average energy because

$$\overline{U^2} = n \int U^2(r) e^{-U(r)/T} d^2\mathbf{r} = \frac{2\pi\Gamma(4/3)}{3} n r_0^2 T^2 \sim E_{\text{int}}^2 / n r_0^2, \quad (3.13)$$

i.e., the fluctuations of the interaction energy are larger than its average value.

The results of this section, Eqs. (3.4) and (3.8), are valid under two conditions: that of small concentration, Eq. (3.1), and

$$k_T r_0 = \left( \frac{2d^2 k_T}{b} \right)^{1/3} \gg 1, \quad (3.14)$$

that validates classical description of the interaction.

The parameter  $n r_0^2$  is the gas parameter which is the ratio of the exciton interaction energy to its kinetic energy. The same parameter indicates the strength of the exciton-exciton scattering. If the impact parameter in a scattering event of two excitons is  $\leq r_0$  then the scattering angle is large. Therefore the scattering cross-section (in 2D case it has units of length) is  $\sim r_0$ . Respectively, the mean-free path is  $l$

$\sim 1/nr_0$ . The scattering is weak, i.e., three and more particle scattering can be neglected if  $l$  is larger than the interparticle distance which means that the gas parameter is small, Eq. (3.1). If this condition is violated the exciton system cannot be considered as a gas, it is a liquid. On the other hand, parameter  $k_T r_0$  characterizes quantum corrections to scattering of excitons. Equation (3.14) is stronger than the nondegeneracy condition because  $k_T r_0 = (k_T/n^{1/2})(n^{1/2}r_0) \ll k_T/n^{1/2}$  because the characteristic scale of the potential  $r_0$  is smaller than the average distance between excitons, Eq. (3.1).

When temperature goes down further both condition of small concentration, Eqs. (3.1), and classical description of exciton-exciton scattering, Eq. (3.14), are at some point violated. Which one is violated first depends on the concentration. If  $n < (b/2d^2)^2$  then quantum effects in the exciton scattering become important when the exciton system can still be considered as a nondegenerate gas. In the opposite case with reduction in the temperature the exciton system becomes a liquid before any quantum corrections to the scattering process become pronounced.

#### IV. EXCITON GAS WITH QUANTUM SCATTERING

If

$$n \ll (b/2d^2)^2 \quad (4.1)$$

and  $k_T r_0 \leq 1$  which is equivalent to

$$k_T \leq b/2d^2, \quad (4.2)$$

then quantum corrections to the exciton-exciton scattering are important but the exciton gas is yet nondegenerate until  $k_T \gg n^{1/2}$ . For the calculation of the interaction energy it is then still possible to use Eq. (3.2) but  $g(r)$  has to be modified to include quantum corrections. This can be done in the following way.

A wave function describing a state of two excitons can be factorized into a wave function of the center of mass and a wave function  $\psi(\mathbf{r})$  describing their relative motion (see the Appendix).  $\psi(\mathbf{r})$  is characterized by a few quantum numbers but in equilibrium the occupation of a state depends only on its energy. This means that, given the energy of the relative motion  $E$  of any two excitons, the probability to find one exciton at the distance  $r$  from the other is  $\langle |\psi(\mathbf{r})|^2 \rangle_E$  where  $\langle \dots \rangle_E$  is the average over all quantum numbers (e.g., the direction of the wave vector) except  $E$ . For a nondegenerate exciton gas the probability density for an exciton to have energy  $E$  is  $(1/T)e^{-E/T}$ . That is,

$$g(r) = \frac{1}{T} \int_0^\infty \langle |\psi(\mathbf{r})|^2 \rangle_E e^{-E/T} dE, \quad (4.3)$$

where  $\psi(\mathbf{r})$  has to be normalized in such a way that  $g(r)|_{r \rightarrow \infty} = 1$  which corresponds to usual normalization for scattering problem.

In a semiclassical approximation, when the exciton wavelength is smaller than the length scale of the interaction potential,  $k_T r_0 \gg 1$ ,

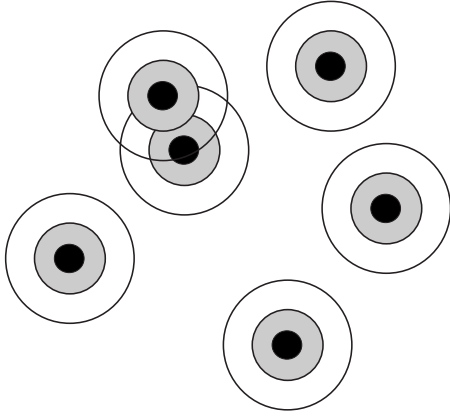


FIG. 5. A sketch of an exciton gas with quantum scattering. The radius of the white circle is of the order of the exciton wavelength,  $\sim 1/k_T$ , the radius of the gray circle is the classical minimal distance between excitons,  $\sim r_0$ , and the radius of the black circle is the quantum minimal distance between excitons,  $\sim d^2/b$ . Here the exciton wavelength is larger than  $r_0$  and the scattering is essentially quantum mechanical. On the other hand the average distance between excitons is larger than their wavelength so that the gas is nondegenerate.

$$\psi(\mathbf{r}) = \frac{A}{[E - U(\mathbf{r})]^{1/4}} e^{iS/\hbar}, \quad (4.4)$$

where  $S$  satisfies the equation

$$\frac{(\nabla S)^2}{M} = E - U(\mathbf{r}). \quad (4.5)$$

Substitution of Eq. (4.4) in Eq. (4.3) results in

$$g(r) = \frac{A^2}{T} \int_{U(r)}^{\infty} e^{-E/T} \frac{dE}{\sqrt{E - U(r)}} = \frac{A^2 \sqrt{\pi}}{\sqrt{T}} e^{-U(r)/T}, \quad (4.6)$$

which is identical to Eq. (3.3) for  $A^2 = \sqrt{T/\pi}$ . Figure 11 of Ref. 18 for  $g(r)$  is related to an intermediate case where  $k_T r_0 \approx 1.7$  and it differs from our classical expression by 20% in the scale of  $r$  which comes from not very large value of  $k_T r_0$  and our simplification of the interaction between excitons.

If  $k_T r_0 \ll 1$  the result strongly differs from the classical case. The wave function  $\psi(\mathbf{r})$  penetrates under the repulsion barrier and the minimal distance between excitons is characterized not by  $r_0$  anymore but rather by the distance at which  $\psi(\mathbf{r})$  falls off. The dipole-dipole repulsion  $e^2 d^2 / \kappa r^3$  is trying to push the wave function to larger  $r$  while the kinetic energy  $\sim (\hbar^2 / M r^2)$  is trying to spread it to all available space and in particular to as small values of  $r$  as possible. The distance at which  $\psi(\mathbf{r})$  falls off is characterized by the same order of magnitude of these two tendencies. This gives the distance of  $\sim d^2/b$ , Fig. 5.

When the interaction potential is approximated by interaction between pointlike dipoles, Eq. (3.7), this distance is smaller than  $r_0$ :  $r_0 / (d^2/b) \sim (b/k_T d^2)^{2/3} \gg 1$  because of Eq. (4.2). But due to  $b \ll d$  it is much larger than  $d$  which justifies the approximation of point-like dipoles, Eq. (3.7).

The interaction energy can be estimated as  $U(d^2/b)$  (under the condition  $k_T r_0 \ll 1$  it is larger than  $T$ ) multiplied by the number of excitons in the important region,  $n(d^2/b)^2$ , that gives  $E_{\text{int}} \sim \hbar^2 n/m$ . The exact calculation (Appendix) shows that  $\psi(\mathbf{r})$  falls off exponentially when  $r < d^2/b$  and is a relatively slow function of  $r$  when  $r > d^2/b$ . On the other hand, the potential  $U(r)$  falls off as  $1/r^3$  when  $r > d^2/b$ . As a result, the main contribution to the interaction energy Eq. (3.2) comes from the region  $r \sim d^2/b$ . In this region the wave function is given by Eqs. (A7) and (A12),

$$\psi(r) = -\frac{2}{\ln(kd^2/b)} K_0\left(\frac{2d}{\sqrt{br}}\right), \quad (4.7)$$

where  $k = \sqrt{ME}/\hbar$ . The logarithmic dependence of this wave function on the energy is very weak and with a logarithmic accuracy the substitution of Eq. (4.7) into Eq. (4.3) leads to the following expression for the correlation function:

$$g(r) = \frac{4n}{\ln^2(k_T d^2/b)} K_0^2\left(\frac{2d}{\sqrt{br}}\right), \quad r \ll 1/k_T. \quad (4.8)$$

As a result,

$$E_{\text{int}} = \frac{8\pi n}{\ln^2(k_T d^2/b)} \frac{e^2 d^2}{\kappa} \int_0^{\infty} K_0^2\left(\frac{2d}{\sqrt{br}}\right) \frac{dr}{r^2} = \frac{2\pi \hbar^2 n}{M \ln^2(k_T d^2/b)}, \quad (4.9)$$

where Eq. (6.576.4) of Ref. 34 has been used. Note that in this case again  $E_{\text{int}}$  is larger than the interaction at the average distance between excitons:  $U(n^{-1/2})/E_{\text{int}} \sim (d^2 n^{1/2}/b) \ln^2(k_T d^2/b) \ll 1$  which is due to Eq. (4.1).

The gas parameter in the quantum case is different from classical one,  $n r_0^2$ . Exciton-exciton scattering is strong if two excitons approach each other to a distance equal to their wavelength. In other words, the scattering cross-section is of the order of the wavelength with accuracy of a logarithmic correction (this is the well-known difference between the 2D and the three-dimensional case where at small wave vectors the cross-section goes to a constant, see Ref. 35, problem 7 to Sec. 132, and Appendix) in spite of the fact that the length scale of the potential  $d^2/b$  is smaller than the wavelength. Respectively, the mean-free path of excitons is  $l \sim k_T/n$ . The gas condition in quantum case is the absence of correlations between different scattering events which means that the wavelength has to be much smaller than the mean-free path, i.e.,  $k_T l \gg 1$  or

$$n \ll k_T^2 \ln^2(kd^2/b). \quad (4.10)$$

This inequality is identical with  $E_{\text{int}}/T \ll 1$  and also with the condition of nondegeneracy with an accuracy of the logarithmic correction. Practically the logarithm is not very large and *in the gas state the exciton system is nondegenerate while the degeneracy is accompanied with strong interactions and multiparticle correlations between the excitons, which is characteristic for liquids.*

## V. LIQUID STATE OF THE EXCITON SYSTEM

In this section we consider the temperatures and/or concentrations beyond the limits specified in previous sections.

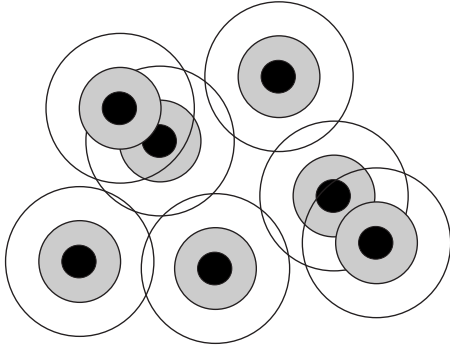


FIG. 6. A rough sketch of quantum exciton liquid. The radius of the white circle is of the order of the exciton wavelength,  $\sim 1/k_T$ , the radius of the gray circle is the classical minimal distance between excitons,  $\sim r_0$ , the radius of the black circle is the quantum minimal distance between excitons,  $\sim d^2/b$ . The average distance between excitons is of the order or smaller than their wavelength and the system is degenerate. But the same condition means that multiparticle correlation is strong and this is a liquid. The picture cannot demonstrate that different excitons are not distinguishable due to an overlap of their wave functions.

In those cases multiexciton correlations are important and the problem is not reduced to a two-particle problem, Fig. 6. Actually all close neighbors are correlated although a long-range correlation may not exist. This situation is characteristic for liquids and therefore we use the term “liquid” for such states of exciton system.

An exact microscopical theory of liquids does not exist and to avoid poorly controllable and rather complicated approximations we make estimates based on reasonable physical arguments. These estimates give not only an approximate value of the blue shift but also its dependence on the concentration and temperature. Also, the arguments and estimates allow us to develop a general understanding of the structure of the exciton system at low temperatures and high concentrations.

The picture is different depending on the exciton concentration compared to  $(b/2d^2)^2$ .

#### A. Low concentration

If the concentration is low according to quantum criterion Eq. (4.1), and the temperature is low enough so that multiparticle correlations take place, i.e., the inequality in Eq. (4.10) is violated, then Eq. (4.9) can be considered as a good estimate that can have only logarithmic corrections. Indeed, the temperature in Eq. (4.9) enters only in the argument of the logarithm and only this argument can change when the temperature goes down. We emphasize once again that violation of condition (4.10) leads not only to degeneracy but also to multiparticle correlation which makes the theory of dilute Bose gas unapplicable.

We would like to attract an attention to a generally known fact that in a dilute 2D Bose gas the characteristic energy at low temperatures does not depend on the coupling constant, except logarithmic corrections. This comes from the virial theorem, i.e., from comparison of the interaction energy and

kinetic energy and is a generalization of Eq. (4.9) to any interaction between particles. In a dilute gas only two particles can be at the distance where their interaction is important (large quantum uncertainty of the distance  $\sim 1/k_T$  means only that we cannot be sure that they are at this distance). If the interaction between particles is  $U(r)$  then from the virial theorem it follows that  $U(r) \sim \hbar^2/Mr^2$ . The value of  $r$  obtained from this relation is the distance at which the interaction is important. The interaction energy of a particle is the interaction energy between two particles at the distance  $\leq r$  times the probability that two particles come to this distance,  $nr^2$ . This gives  $n\hbar^2/M$ . In a liquid the estimate can have a numerical factor characterizing the number of particles within the interaction radius. This energy gives a temperature scale for both Bose condensation<sup>36,37</sup> and Kosterlitz-Thouless transition.<sup>4,38</sup>

#### B. High concentration

If the concentration is beyond its quantum limitation, i.e., Eq. (4.1) is not satisfied then decrease in the temperature or increase in the concentration leads to violation of the classical gas condition, Eq. (3.1), while the system is still nondegenerate, i.e., Eq. (3.14) holds. In this case the exciton system becomes a classical liquid. As long as Eq. (3.7) is valid the dimensional analysis allows us to express  $E_{\text{int}}$  as a function of only one parameter:

$$E_{\text{int}} = \frac{e^2 d^2}{\kappa} n^{3/2} f\left(\frac{e^2 d^2}{\kappa T} n^{3/2}\right). \quad (5.1)$$

According to Eq. (3.8)  $f(x) = 2\pi\Gamma(4/3)x^{-1/3}$  at  $x \ll 1$ .

When  $(e^2 d^2 / \kappa T) n^{3/2} = r_0^3 n^{3/2}$  grows and becomes of the order of unity a free motion of excitons between collisions becomes impossible because each of them is confined in between its neighbors. In other words an exciton is in a highly excited state in a potential well formed by its neighbors. The size of the well is  $R \sim n^{-1/2}$  and this semiclassical picture is valid as long as the size of the confinement region is much larger than the exciton thermal wavelength, i.e.,  $k_T R \gg 1$ . The energy at the bottom of the potential well is  $\sim z e^2 d^2 / R^3$  (where  $z$  is the number of nearest neighbors) and 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  $nr_0^2 > 1$  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 the formation of a short-range order characteristic for liquids.<sup>39</sup> We emphasize that we mean a formation of a short-range order typical in liquids but not crystallization and a formation of a long-range order,<sup>40</sup> Fig. 7. Further reduction in the temperature brings particles to lower levels in the potential wells and makes the wells more stable. A stronger confinement of the wave functions of each exciton reduces their overlap. When the size of the exciton wave function becomes smaller than  $R$ , the potential for each exciton can be approximated as

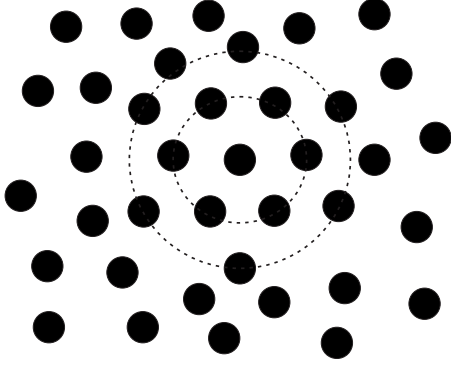


FIG. 7. A rough sketch of a short-range order in a classical liquid. In this example the order extends to two coordinate circles, and at larger distance excitons are not correlated. (Note that this picture is oversimplified: similar order exists around most of the excitons).

$$U_{\text{liq}}(\mathbf{r}) \approx \frac{e^2 d^2}{\kappa R^3} \left( C_1 + C_2 \frac{r^2}{R^2} \right), \quad (5.2)$$

where  $r$  is the distance from the minimum of the well. We estimate the constants  $C_1$  and  $C_2$  assuming a short range order, i.e., there is a crystal structure around an exciton within  $l$  coordinate circles but beyond this region the exciton positions are not correlated. This assumption gives

$$C_1 = \sum_{j=1}^l \frac{z_j}{(R_j/R)^3} + \frac{2\pi}{R_{l+1}/R}, \quad (5.3a)$$

$$C_2 = \frac{9}{4} \sum_{j=1}^l \frac{z_j}{(R_j/R)^5} + \frac{3\pi}{2(R_{l+1}/R)^3}. \quad (5.3b)$$

Here  $R = [n \sin(2\pi/z)]^{-1/2}$  is the lattice constant which is the same as the radius of the first coordinate circle,  $R_j$  is the radius of the  $j$ th coordinate circle, and  $z_j$  is the number of particles at the  $j$ th coordinate circle,  $z_1 = z$ . The resulting values of the constants appear to be very weakly sensitive to the radius of the order, see Table II (compare Ref. 41).

If  $T \sim (e^2 d^2 / \kappa) n^{3/2}$  (i.e.,  $nr_0^2 \sim 1$ ) then  $r \sim R$  and there is no short-range order in the system. However, if  $T \ll (e^2 d^2 / \kappa) n^{3/2}$  then the short-range order does exist, and

TABLE II. Constants which characterize the effective potential well for each exciton in a liquid, Eq. (5.2).  $l=0$  corresponds to very small correlations between the exciton positions and  $l=\infty$  corresponds to a crystal structure.

Coordinate circle	Square lattice, $z=4$		Hexagonal lattice, $z=6$	
$l$	$C_1$	$C_2$	$C_1$	$C_2$
0	6.28	4.71	7.25	5.44
1	8.44	10.67	10.19	14.55
2	8.55	11.18	10.78	15.07
3	8.72	11.29	10.65	15.08
$\infty$	8.84	11.37	10.84	15.13

most of the excitons are at the ground state of their corresponding potential Eq. (5.2). The energy of the ground state above the bottom of the potential well and its radius are

$$\begin{aligned} \hbar\omega_{gs} &\approx \sqrt{2C_2 \frac{\hbar^2}{MR^2} \frac{e^2 d^2}{\kappa R^3}} \\ &= \frac{2C_2 \hbar^2 n}{M} \sqrt{\frac{d^2 n^{1/2}}{b}}, \quad r_{gs} = R \left( \frac{1}{2C_2} \frac{bR}{d^2} \right)^{1/4}. \end{aligned} \quad (5.4)$$

The characteristic size of the exciton wave function  $r$  is controlled by the temperature or the energy of the ground state, whichever is larger, and in any case when  $T \ll (e^2 d^2 / \kappa) n^{3/2}$  the size of a single exciton wave function is much smaller than the distance between the excitons.

The inequality  $r \ll R$  allows us to make two conclusions. First, the bottom of potential  $U_{\text{liq}}(\mathbf{r})$ , Eq. (5.2), gives a good estimate for the interaction energy

$$E_{\text{int}} \approx \frac{10e^2 d^2 n^{3/2}}{\kappa}. \quad (5.5)$$

Comparison of this expression with Eq. (5.1) shows that  $f(x \rightarrow \infty) \approx 10$ . This estimate does not include possible logarithmic corrections.

Second, it is possible to estimate the overlap of the wave functions of adjacent excitons. If  $d=12$  nm and  $n=2 \times 10^{11}$  cm $^{-2}$  the estimate according to the wave function in the harmonic potential of Eq. (5.2) gives for the overlap a value of  $\sim 0.14$ . The actual value is even smaller because at  $r \sim R$  the potential barrier is steeper than the harmonic one. Due to the small wave function overlap the temperature at which the phase or/and spin coherence<sup>5</sup> in the exciton system is set in is reduced compared to its the expected value  $\sim \hbar^2 n / M$ . This points to a possible nonmonotonic dependence of the quantum coherence onset temperature on the concentration, and it suggests that a lower-density exciton system may become quantum coherent at higher temperatures than a higher-density system, which is *a priori* nonintuitive. In other words, a long-range interaction suppresses quantum degeneracy.

At low temperature the physics of the transition between quantum liquid and classical liquid with growth of the exciton concentration is the following. At low concentration  $n \ll (b/d^2)^2$ , Eq. (4.1), according to the exciton wave function Eq. (4.7) around each exciton there is a circle with radius  $\sim d^2/b$  inside which the wave function of any other exciton is exponentially small. However, the radius of this circle is much smaller than the average distance between excitons,  $n^{-1/2}$ , and any exciton wave function can easily spread at the area that contains many other excitons avoiding their depleted circular regions. That is wave functions of different excitons overlap forming a quantum liquid (not a gas because of strong exciton-exciton scattering).<sup>42</sup> With growth of the concentration the average distance between excitons decreases that makes spreading of each exciton wave function to a wide area more difficult. This reduces the overlap of the wave functions of different excitons. Finally, when the average distance becomes smaller than the radius of the circular depleted region,  $n^{-1/2} \lesssim b/d^2$ , the wave function of nearly



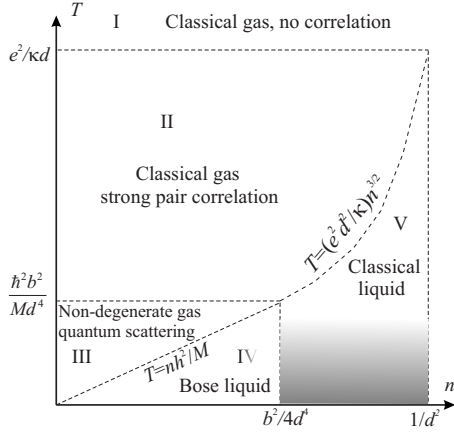


FIG. 8. Different role of the exciton-exciton correlations in an exciton system. The shaded area shows the part of the region V where quantum correlations set in.

each exciton appears to be confined in between its nearest neighbors. The overlap of the wave functions of adjacent excitons is very weak and the system becomes a classical liquid.

## VI. DISCUSSION

Estimates made in the previous sections open the possibility to develop a general picture that demonstrates a role of correlations in the exciton system at the whole  $n-T$  plane. This picture is presented in Fig. 8. Correlations are 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. In region III, contrary to region II, the exciton-exciton scattering is described by quantum mechanics. In other respects this region is similar to region II. Reduction in the temperature from region III to IV leads to degeneracy of the exciton system. But simultaneously a strong multiparticle correlation is set up. The system cannot be considered as a dilute 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 well below  $\hbar^2 n/M$  (compare Ref. 43). The reason is that strong repulsion between excitons squeezes the wave function of each exciton to an area smaller than the average area per one exciton. In this region a short-range order appears and with further reduction in temperature its correlation radius grows. However, contrary to regular classical liquids, the attractive part of the exciton-exciton interaction is negligible<sup>4,18</sup> and it is likely that a long-range order is settled not as a result of a phase transition but as gradual growth of the correlation radius.

A comparison of the values of the exciton binding energy  $\epsilon_b$  and the Coulomb interaction at distance  $d$ ,  $e^2/kd$ , in Table III leads to the conclusion that the correlations between excitons can be neglected only when a significant part of them is dissociated. However, in this case the concentration  $n$  that controls the blue shift in Eq. (1.1) is not the exciton concentration but the sum of the exciton concentration and concentration of electrons or holes. The existence of region V is the

TABLE III. Estimates for the parameters of Fig. 8 for GaAs/AlGaAs quantum well structures with different values of  $L$ .

$d$ (nm)	$\epsilon_b$ (K)	$e^2/kd$ (K)	$\hbar^2 b^2 / Md^4$ (K)	$b^2 / 4d^4$ (cm <sup>-2</sup> )	$1/d^2$ (cm <sup>-2</sup> )
10	54	139	3.8	$2.2 \times 10^{10}$	$1.0 \times 10^{12}$
12	49	116	1.8	$1.1 \times 10^{10}$	$0.69 \times 10^{12}$
14	44	99	1.0	$0.58 \times 10^{10}$	$0.51 \times 10^{12}$

result of the tail of the exciton-exciton repulsion potential. In case of a short range potential (e.g., hard circles) an overlap of the particle wave functions competes with the repulsion and the region of classical liquid behavior does not exist. This happens to excitons in one well where there is no dipole-dipole repulsion and an increase in the concentration leads to the Mott transition but not to a classical liquid.

Two of the lines in Fig. 8, between regions III and IV and between regions II and V, actually comprise one line at which the gas parameter condition is violated. The gas parameter is the product of the concentration and the scattering cross-section squared. Between regions II and V this cross section is classical,  $\sim r_0$ , while between regions III and IV it is quantum,  $\sim 1/k_T$ . The other two lines, between regions II and III and between regions IV and V, separate classical and quantum interactions between excitons. Also, it is necessary to note that at the bottom of region V some quantum coherent phenomena are possible.

It is important to note that the lines separating different regions in Fig. 8 do not correspond to sharp transitions. Crossing of one of the lines by changing the temperature or the concentration leads to a gradual change in the correlation between excitons. Figure 8 demonstrates only the role of correlation but not phases of the system.

Finally, we have to notice that importance of pair correlation in the electron-hole system in coupled quantum wells was studied in a number of papers with the help of numerical simulations. It would be helpful to compare these results with ours. Unfortunately, possibility of the comparison is quite limited because in these papers only electron-electron, hole-hole, and electron-hole correlation have been studied but none of them considered exciton-exciton correlation. So we can make only qualitative comparison of correlation diagram in Fig. 8 with the results numerical simulations.

In Refs. 44–47 the susceptibility of the bilayer was calculated at zero temperature. When the distance between the layers decreases at some specific value of the wave vector  $q$  the susceptibility diverged that corresponded to an instability of a uniform state. The conclusion concerning the non-uniform phase, Wigner crystal or charge-density wave, was based on the value of the wave vector at which the instability took place. According to Fig. 8 the instability is likely to be formation of the exciton crystal. Formation of excitons is confirmed by interlayer and intralayer correlation functions calculated in Refs. 44–47. The value of the interlayer correlation function at zero in-plane distance significantly grows with reduction in the interlayer distance while the intralayer correlation function at zero distance remains small.

In Refs. 48 and 49 the phase diagram of the classical electron-hole bilayer system was studied. Considering the

behavior of the interlayer and intralayer correlation function the authors showed that at small separation between the electron and hole layers excitons are formed. They also concluded that at large values of the gas parameter, i.e., large the potential energy compared to the kinetic energy, the system is solid (two correlated Wigner crystals) while at small values of it the system is liquid. This argument goes back to Wigner<sup>50</sup> and we also used it in the development of the correlation diagram Fig. 8. The important point here is that in the case of Coulomb interaction the potential energy dominates at low concentration while for dipole-dipole interaction which we consider it dominates at high concentration.

In Refs. 27 and 29 the phase diagram at the plane  $(d, n)$  was investigated at zero temperature. Two conclusions of this paper support our diagram Fig. 8. The first one is that the system is in the excitonic phase at small enough value of  $nd^2$  and is two component plasma (or two correlated Wigner crystals) at large values of this parameter. The other conclusion is the same as in Refs. 48 and 49: at large concentration the potential energy dominates the kinetic energy and excitons form a crystal.

**VII. CONCLUSIONS**

We studied an exciton system in coupled quantum wells where electrons and holes are confined in different wells and the main interaction between excitons is a dipole-dipole repulsion. We found that in the most part of the temperature-concentration plane the system is characterized by a strong exciton-exciton correlation. At some parts of this plane the system behaves as a gas with a strong pair correlation. In other parts where the gas parameter condition is violated, i.e., where the probability to find more than two excitons close to each other becomes of the order of unity, the correlation is multi- exciton and the system has to be considered as a liquid. In particular, *at low concentrations degeneracy of the system is accompanied by a setting in of multiexciton correlations.* The system is a Bose liquid and no theory of weakly nonideal or rare Bose gas can describe its behavior. At high concentration the strong confinement of each exciton wave function due to repulsion between excitons suppresses quantum correlations. *In some temperature region the system is a classical liquid; such a region does not exist in case of contact interaction.* We developed a correlation diagram, Fig. 8 that characterizes some phases of the system: gas, quantum liquid, classical liquid, and solid. The blue shift of the exciton luminescence has different value and a different dependence on the exciton temperature and concentration depending on how close excitons can come to each other. Therefore it is a sensitive tool for measuring of the exciton-exciton correlations.

**ACKNOWLEDGMENTS**

B.L. appreciates discussions with M. Stern and A. L. Efros.

**APPENDIX: EXCITON-EXCITON SCATTERING**

Here the problem of the exciton-exciton scattering is considered under the condition of small energy of relative motion,

$$E = \frac{\hbar^2 k^2}{M} \ll \frac{e^2}{\kappa d}, \tag{A1a}$$

and large wavelength,

$$k \ll b/2d^2. \tag{A1b}$$

For two excitons the center of mass momentum and the momentum of relative motion are defined as

$$\mathbf{K} = \mathbf{k}_1 + \mathbf{k}_2, \quad \mathbf{k} = \frac{\mathbf{k}_1 - \mathbf{k}_2}{2}. \tag{A2}$$

The two-exciton wave function is factorized

$$\Psi(\mathbf{r}_1, \mathbf{r}_2) = \frac{1}{\sqrt{S}} e^{i\mathbf{K}(\mathbf{r}_1 + \mathbf{r}_2)/2} \psi(\mathbf{r}_1 - \mathbf{r}_2), \tag{A3}$$

and the Schrödinger equation for the wave function describing their relative motion is

$$-\frac{\hbar^2}{M} \nabla^2 \psi(\mathbf{r}) + U(r) \psi(\mathbf{r}) = E \psi(\mathbf{r}). \tag{A4}$$

Under the condition Eq. (A1a) the minimal distance between excitons is much larger than  $d$  (i.e., at  $r \sim d$  the wave function is negligibly small) and the interaction potential can be approximated with Eq. (3.7). The condition of a very long exciton wavelength, Eq. (A1b), makes it possible to simplify Eq. (A4) in two regions. In the region where the distance between excitons is much smaller than the wavelength of their relative motion,  $kr \ll 1$ , the coordinate dependence of the wave function comes only from the potential energy and the characteristic scale  $r \sim d^2/b$ . At this scale the kinetic energy can be neglected and Eq. (A4) is reduced to

$$\nabla^2 \psi(\mathbf{r}) - \frac{d^2}{br^3} \psi(\mathbf{r}) = 0, \quad kr \ll 1. \tag{A5}$$

When the distance between the excitons is much larger than  $r_0 = (e^2 d^2 / \kappa E)^{1/3} = (2d^2 / k^2 b)^{1/3}$  the interaction energy is small compared to the kinetic energy and

$$\nabla^2 \psi(\mathbf{r}) + k^2 \psi(\mathbf{r}) = 0, \quad r \gg r_0. \tag{A6}$$

Due to  $kr_0 = (kd^2/b)^{1/3} \ll 1$  the two regions overlap at  $r_0 \ll r \ll 1/k$ .

Solutions to both Eqs. (A5) and (A6) are expressed in Bessel functions. When  $kr_0 \ll 1$  only S scattering is important and it is enough to find angular independent solution of Eq. (A5). The solution that goes to zero at  $r \rightarrow 0$  is

$$\psi(\mathbf{r}) = A_1 K_0 \left( \frac{2d}{\sqrt{br}} \right) = A_1 K_0 \left( 2kr_0 \sqrt{\frac{r_0}{r}} \right), \quad kr \ll 1. \tag{A7}$$

The solution to Eq. (A6) describing scattering is

$$\psi(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}} + A_2 H_0^{(1)}(kr). \tag{A8}$$

Making use of asymptotes<sup>34</sup>

$$K_0(z) = -\ln \frac{z}{2} - C + O(z^2 \ln z), \quad |z| \ll 1, \tag{A9a}$$

$$H_0^{(1)}(z) = 1 + \frac{2i}{\pi} \left( \ln \frac{z}{2} + C \right) + O(z^2 \ln z), \quad |z| \ll 1, \quad (\text{A9b})$$

where  $C=0.577$  is the Euler constant it is easy to match the solutions in the intermediate region  $r_0 \ll r \ll 1/k$ ,

$$-A_1 \left( \ln \frac{kr_0^{3/2}}{r^{1/2}} + C \right) = 1 + A_2 \left[ 1 + \frac{2i}{\pi} \left( \ln \frac{kr}{2} + C \right) \right]. \quad (\text{A10})$$

This gives

$$A_1 = -\frac{2}{\ln(kd^2/b) + 3C - \ln 2 - i\pi/2},$$

$$A_2 = \frac{i\pi/2}{\ln(kd^2/b) + 3C - \ln 2 - i\pi/2}. \quad (\text{A11})$$

According to Eq. (A1b) the argument of the logarithm is small and with the logarithmic accuracy

$$A_1 = -\frac{2}{\ln(kd^2/b)}, \quad A_2 = \frac{i\pi/2}{\ln(kd^2/b)}. \quad (\text{A12})$$

From the asymptote<sup>34</sup>

$$H_0^{(1)}(z) = \sqrt{\frac{2}{\pi z}} e^{i(z-\pi/4)}, \quad |z| \gg 1, \quad (\text{A13})$$

it follows

$$\psi(r) = e^{ikr} + \frac{e^{i\pi/4}}{\ln(kd^2/b)} \sqrt{\frac{\pi}{2kr}} e^{ikr}, \quad kr \gg 1. \quad (\text{A14})$$

That is the scattering cross section (in 2D case it has units of length) is

$$\sigma = \frac{\pi^2}{k \ln^2(kd^2/b)}. \quad (\text{A15})$$

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