FORMATION OF AN ORDER IN A SYSTEM OF EXCITON CONDENSED PHASE ISLANDS IN QUANTUM WELLS

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A theory of exciton condensed phase creation in a two-dimensional system is presented. The consideration takes into account the mutual influence of exciton condensed phase islands through exciton concentration fields. For the solution of the problem the kinetic and Fokker—Planck equations are applied. The theory is applied to explain the appearance of the periodical fragmentation which was observed last years in luminescence from the ring around a laser spot in a crystal with double quantum wells. The dependence of the radius of condensed phase islands and the distance between islands is obtained as a function of temperature. The influence of fluctuations on the periodical structure is studied.

The electrons and holes of indirect excitons in semiconductor coupled quantum wells are localized in different wells, and, as a result, the excitons have long lifetime. Such a system presents interest for the study of exciton condensation phenomena [1]. Recently, the interesting experiments were fulfilled for indirect excitons in AlGaAs and InGaAs crystals with coupled quantum wells. In works [2—7], the authors have observed a ring structure in the emission of indirect excitons outside a laser spot. The distance between the ring and the excitation spot center grows with increasing the pumping and can reach several hundreds of microns that is much larger than the exciton diffusion length. At a low temperature ($T \leq 2$ K), the external ring is fragmented into a periodical structure over macroscopic lengths. The mechanism of the luminescence ring formation was proposed in [6, 7]. The origin of the fragmentation along the ring is unclear. In the present work, the appearance of this fragmentation is explained by the creation of islands of exciton condensed phases along the ring. In [8, 9], it was shown that a high-density exciton system is unstable in the presence of an attractive interaction between excitons relative to the appearance of a periodical structure. To obtain the spatial dependence of the exciton generation rate, we shall use the suggestion of works [6, 7] that, in the absence of the light irradiation, a quantum well contains electrons, and holes created by light are captured by a quantum well more effectively than electrons. As a result, the charge separation takes place from a positive charge value at the center of a laser spot to a negative one far from the spot. The emission of light occurs from the ring where the product of the electron and hole densities has maximum. For the determination of the density distribution, we have used the system of equations similar to that investigated in [6, 7], but we added the equation for the exciton density. Lets $n_e, n_h, n_{\rm ex}$ designate the well densities of electrons, holes, and excitons, respectively. These values satisfy the following kinetic equations:

$$\frac{\partial n_e}{\partial t} = D_e \triangle_2 n_e + K_e(r) - W n_e n_h - \frac{n_e - n_0}{\tau_e}, \qquad (1)$$

$$\frac{\partial n_h}{\partial t} = D_h \triangle_2 n_h + K_h(r) - W n_e n_h - \frac{n_h}{\tau_h}, \qquad (2)$$

$$\frac{\partial n_{\rm ex}}{\partial t} = D_{\rm ex} \triangle_2 n_{\rm ex} + G - \frac{n_{\rm ex}}{\tau_{\rm ex}}. \qquad (3)$$

Here, D_e, D_h and $D_{\rm ex}$ are the diffusion coefficients, τ_e, τ_h and $\tau_{\rm ex}$ are the lifetimes for electrons, holes, and excitons, respectively, W is the electron-hole recombination rate, $K_e(K_h)$ is the electron (hole) creation rate in the well, and G is the exciton production rate ($G = qW n_e n_h, q \leq 1$).

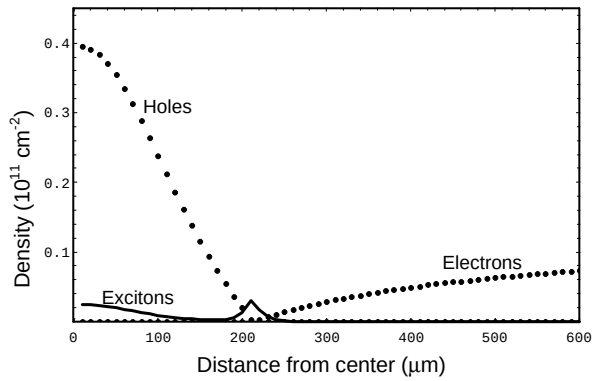

Fig. 1. Dependence of electron, hole, and exciton densities on the distance from the center

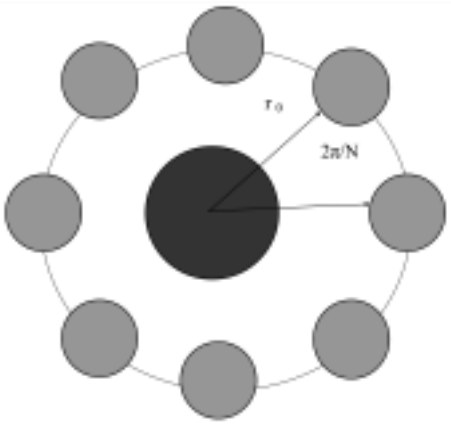

Fig. 2. Position of exciton condensed phase islands around a laser excitation spot

Fig. 1 shows the result of calculations of the electron, hole, and exciton densities for the following values of parameters: $D_e = 200$ cm^2/s, $D_h = 50$ cm^2/s, $D_{\rm ex} = 10$ cm^2/s, $W = 150$ cm^2/s , $q = 0.9$, $\tau_e = \tau_h = 10^{-5}$ s, $\tau_{\rm ex} = 10^{-7}$ s, the spatial distribution of the pumping was approximated by the Gaussian curve with a width of 60 μm and $K_h = 2K_e$. The qualitative picture of the behavior of n_e, n_h as a function of r is similar to results obtained in [6, 7]. The position of the exciton density maximum determines the ring luminescence in experiments. We use the obtained value of exciton density to study the fragmentation of the exciton condensed phase. It is suggested that the condensation occurs due to the exciton-exciton interaction, and it is not the Bose—Einstein condensation in the wave vector space with $\mathbf{k}$=$\mathbf{0}$. We have estimated the van der Waals attraction using the well-known formula of quantum mechanics and showed that the van der Waals interaction between two indirect excitons in coupled quantum wells exceeds the dipole-dipole interactions in the range of distances between excitons of order of several exciton radii (see Appendix). The attractive interaction may cause the existence of the exciton condensed phase. Some models of phase transitions in a system of indirect excitons were studied in [10]. Afterwards, we suggest that the condensed phase exists, and it is described by some parameters which will be determined later.

Due to a finite value of the exciton lifetime, the sizes of exciton condensed phases are restricted. As a result, the condensed phase in the two-dimensional case must exist as a system of islands similar to electron-hole drops in bulk semiconductors. In the studied system, the islands should be localized on the ring where the exciton density has maximum.

Under the following consideration, we shall apply the theory of creation of exciton condensed phase islands in the two-dimensional case [11] to the studied system with nonuniform pumping. Firstly, we study the system of condensed phase islands periodically situated along the ring with the maximum of the exciton density at $r = r_0$ (Fig. 2). $\varphi_m = 2\pi m/N$ is the angle of the m-th island on the ring. Afterwards, we shall show that deviations (fluctuations) from periodicity are small. Let us consider the formation of some island, for example, the island with $m = 0$. We introduce the distribution function f_n which determines a probability for the island with $m = 0$ to have n excitons. The size of disks is determined by four processes: the creation of excitons by pumping, capture of excitons from the environment, escape of excitons from the disk, and the exciton decay. The kinetic equation for the distribution function has the form

$$\frac{\partial f_n}{\partial t} = -j_{n+1} + j_n, \tag{4}$$

where j_n is the probability current,

$$j_n = 2\pi R_{n-1} W_{fi}(R_{n-1}) c(R_{n-1}) f_{n-1} -$$

$$-2\pi R_n W_{if}(R_n) c_i f_n - \pi R_n^2 f_n/\tau_{\rm ex} + \pi R_{n-1}^2 \bar{G}_{n-1} f_{n-1}, \tag{5}$$

R_n is the radius of the disk with n excitons, $W_{fi}(R_n)$ and $W_{if}(R_n)$ are probabilities for the exciton to be captured by the disk and to escape from the disk per unit length of the circle, respectively, $\bar{G}_n = \int G(r) dS_n/\pi R_n^2$ is the mean value of the exciton pumping over island area, $c(R_n)$ and c_i are the concentrations of excitons on the circle of the disk and inside the disk ($c_i = 1/s_o$, s_o is

the area per single electron-hole pair in the island). The exciton concentration $c(r)$ differs from $n_{\rm ex}(r)$ presented in Fig. 1 due to the presence of islands which perturb the exciton concentration field around them. The following condition between the transition probabilities W_{fi} and W_{if} takes place due to the detailed balancing principle:

$$\frac{W_{if}(R)}{W_{fi}(R)} = \frac{W_{if}(\infty)}{W_{fi}(\infty)} \exp\left(\frac{a_2}{R}\right). \quad (6)$$

Here, $W_{fi}(\infty)$ and $W_{if}(\infty)$ are the transition probabilities in the case of a straight line boundary between the condensed and gas phases, $W_{fi}(\infty)/Wif(\infty) = c_i/c_\infty$, c_∞ is the equilibrium concentration of excitons for the straight line boundary between the condensed and gas phases,

$$c_\infty = c_{10} \exp(-\varphi/\kappa T), \quad (7)$$

φ is the condensation energy per exciton, $c_{10} = \gamma(m^*\kappa T/2\pi\hbar^2)$, m^* is the effective exciton mass, γ is the degeneracy of the exciton state, $a_2 = a_l s_o/\kappa T$, a_l is the energy per unit of the disk length.

The connection between islands occurs due to the dependence of the exciton surface concentration of the considered island $c(R_n)$ in the kinetic equation (4) versus the presence of other islands. We suggest that the distance between disks is larger than the disk radius and the concentration field created by some island slowly changes in the limits of a size of the considered island. As a result, the exciton concentration on the disk with $m = 0$ may by presented as

$$c_{\rm ex}(R) = \bar{n}_{\rm ex}(r_0) + a_0 K_0((R/l) + \sum_{\mu\neq 0} K_0(2r_0 \sin(\varphi_\mu/2)/l), \quad (8)$$

where $K_0(x)$ is the modified Bessel function, l is the diffusion length of a free exciton. The coefficients a_μ are determined by boundary conditions on every disk: the current of excitons to an island should be equal to the difference between the number of excitons captured by the island and the number of excitons which escape from the island:

$$2\pi R D_{\rm ex} \frac{\partial c_{\rm ex}(R)}{\partial R} = 2\pi R(W_{fi} c_{\rm ex}(R) - W_{if} c_i). \quad (9)$$

Due to the system symmetry, all islands have equal parameters: $a_\mu = a_0$ for every μ. Further, we introduce the radius distribution function $f(R) = f_n dn/dR = 2\pi R f_n/s_o$. In the steady case at $n \gg 1$, the solution of Eq. (4) has the form

$$f(\tilde{R}, N) = f_0 \exp(-F(\tilde{R}, N), \quad (10)$$

where

$$F(\tilde{R}, N) = -4\pi \int_0^{\bar{R}} \frac{2\nu\left(\tilde{n}_{\rm ex}(r_0) - \tilde{c}_{10} e^{-\frac{(\varphi-a/\tilde{R})}{T}}\right) - \tilde{R}(1-\tilde{n}_G)\left(1 + \frac{\nu(K_0(\bar{R}/\bar{l})+\sigma(N))}{\bar{l}K_1(\bar{R}/\bar{l})}\right)}{2\nu\left(\tilde{n}_{\rm ex}(r_0) + c_{10} e^{-\frac{(\varphi-a/\tilde{R})}{T}}\right) + \left(\tilde{R}(1+\tilde{n}_G) + 4\tilde{c}_{10} e^{-\frac{\varphi-a/\tilde{R}}{T}}\right)\left(1 + \frac{\nu(K_0(\bar{R}/\bar{l})+\sigma(N))}{\bar{l}K_1(\bar{R}/\bar{l})}\right)} \tilde{R} d\tilde{R}, \quad (11)$$

$$\sigma(N) = \sum_{\mu=1}^{N-1} K_0(2\tilde{r}_0 \sin(\varphi_\mu/2)/\tilde{l}). \quad (12)$$

Here, we have introduced the dimensionless variables $\tilde{R} = R/\sqrt{s_o}$, $c(\tilde{R}) = c(R)/s_o$, $\nu = W_{fi}\tau_{\rm ex}\sqrt{s_o}$, $\bar{n}_G = \bar{G} s_o \tau_{\rm ex}$, φ and a are expressed in (10) and (11) in units of temperature. The most probable radius is determined from the condition

$$\frac{\partial F(\tilde{R}, N)}{\partial \tilde{R}} = 0. \quad (13)$$

The mean radius depends on the number of islands, $\bar{R} = R(N)$. The probability for the system to have N islands with radii $\tilde{R}_1, \tilde{R}_2, ...\tilde{R}_N$ equals

$$W(N, \tilde{R}_1, \tilde{R}_2, ...\tilde{R}_N) \approx \exp(-\sum_i F(\tilde{R}_i, N). \quad (14)$$

By integrating over the radii of the disks, we obtain the probability for the system to have N islands on the ring as

$$W(N) = \exp(-\Phi(N)), \quad (15)$$

where

$$\Phi(N) = -N \ln z(N), z(N) = \int_\infty^0 \exp(-F(\tilde{R}(N), N) d\tilde{R}. \quad (16)$$

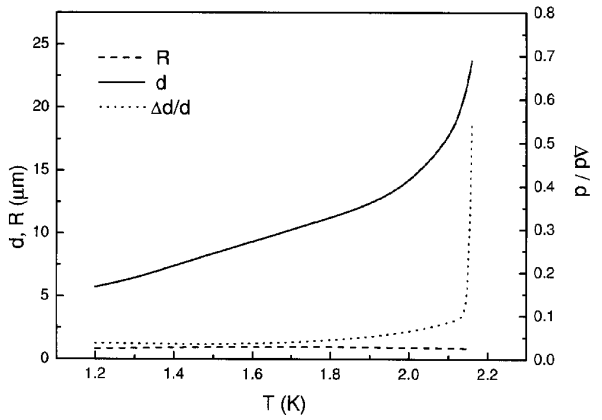

Fig. 3. Dependence of the island radius R, the distance between islands d, and the ratio of the island fluctuation shift to the distance between islands $\Delta d/d$ on temperature

The number of islands is determined by the condition

$$\frac{\partial \Phi}{\partial N} = 0. \tag{17}$$

The formulae (13), (17) determine the mean radius and the number of islands. Since islands of the condensed phases receive excitons from the same source, two islands cannot be situated closely each to other. Moreover, the distance between islands cannot be large, because, in this case, the exciton density between them becomes greater than the critical value and, as a result, a new island may appear. So, there is the specific interaction between the condensed phases through the exciton concentration fields. As a result, the dependence $\Phi(N)$ (17) has minimum at some value $N = N_s$ that determines the number of islands on the ring. We have calculated some parameters of the islands on the ring using Eqs. (10)—(17). The exciton parameters and the parameters of pumping are the same as the parameters in Fig. 1. The following parameters of the condensed phase were chosen: $m^* = 0.37m_0$, $\varphi = 10$ K, $a_2 = 18$ K, $s_o = 10^{-11}$ cm^2. It is seen from Fig. 3 that the distance between islands ($d = 2\pi r_0/N_s$) increases with a rise of temperature, whereas the island radius changes slowly with temperature. For the considered parameters, the condensed phase exists up to 2.16 K.

To estimate the island position fluctuation, we studied the shift of some island versus the angle φ along the ring at a fixed position of all other islands (similar to the Einstein model for crystal oscillations). In this case, the function $\sigma(N)$ in (13) depends on φ ($\sigma(N) \to \sigma(N, \varphi)$) and the function $f(\tilde{R}, N) \to f(\tilde{R}, N, \varphi)$ determines the angle distribution function. Using them, we calculated the mean fluctuation shift of an island along the ring which equals $\Delta d = r_0\sqrt{\overline{\varphi^2}}$. It is seen from Fig. 3 that the relative shift $\Delta d/d$ is small ($\Delta d/d \ll 1$) and increases only in the vicinity of the temperature, at which the condensed phase disappears. By a similar way, the relative fluctuations of the island number ($\Delta N/N_s = \sqrt{\overline{(N - N_s)^2}}/N_s$) were calculated using (15). This value is much less than unity. For example, we have $\Delta N/N_s = 7 \cdot 10^{-3}$ at T=2 K. Thus, the studied system has perfect periodicity. These results explain the periodical fragmentation observed in [2].

We suggested that the exciton gas is nongenerated outside islands. For the considered parameters, the exciton density at the surface of the disks outside them equals $7.2 \cdot 10^8$ cm^{-2} and is significantly lower than the critical Bose—Einstein condensation density. Inside of islands, the exciton density is significantly higher and may be important with regard for the Bose statistics. But the presented theory did not consider the model of condensed phase and needs only some parameters of the condensed phase: the energy and occupied volume per electron-hole pair and the surface energy. Moreover, the condensed phase may present the electron-hole liquid phase.

In the paper, the binding energy per electron-hole pair in the condensed phase was chosen as ($\varphi = 10$ K= 0.8 meV). This value gives the critical temperature of the fragmentation disappearance (2.16 K) close to the experimental value (~2 K).The value of φ is less than the width of the fragment emission line (1.3 meV [2]), and the emission line spectral positions of a free exciton and the fragment coincide practically. Even there is a very small (much less than the width) shift of the fragment emission to the blue side (see Fig. 2,*c* of [2]). But mechanisms that determine the width (scattering by the fluctuation potential, phonons, between excitons, and others) give different contributions to the shapes of the emission spectra (including the shift of lines) for free excitons and the exciton condensed phase. So, if the exciton binding energy to the condensed phase is less than the bandwidth, the physical information from the comparison of the shapes of the emission spectrum of free excitons and of the condensed phase can be obtained only together with the analysis of line broadening mechanisms for these states. For that, the detailed theory of the condensed phase is needed.

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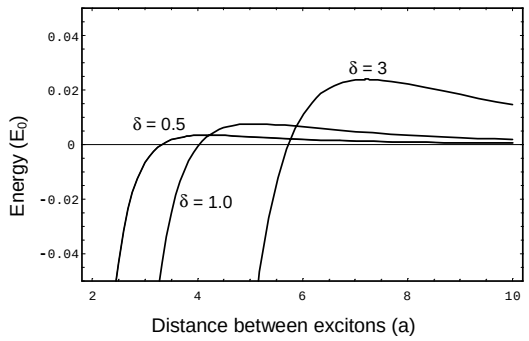

Fig. 4. Dependence of the sum of the van der Waals and dipole-dipole interaction energies between two excitons in coupled quantum wells on the distance between excitons for the different values of the distance between wells δ. E_0 and a are, respectively, the energy and radius of an exciton in the crystal bulk

APPENDIX

To estimate the van der Waals interaction between two excitons, let us use the well-known formula

$$U_{vdw} = -\sum_{i_1\neq 0, i_2\neq 0} \frac{|\langle 0,0 \mid U_{dd} \mid i_1, i_2\rangle|^2}{E_{i_1,i_2} - E_{0,0}}, \qquad (18)$$

where $U_{dd} = \frac{\mathbf{P_1}\cdot\mathbf{P_2}}{\varepsilon R_{12}^3} - \frac{3\mathbf{P_1}\cdot\mathbf{R_{12}}\mathbf{P_2}\cdot\mathbf{R_{12}}}{\varepsilon R_{12}^5}$ is the operator of the dipole-dipole interaction between excitons, R_{12} is the distance between excitons, $|0,0>$ and $|i_1,i_2>$ are the ground and excited states of a system with two excitons. The formula U_{dd} can be used if the distance between excitons is larger than the exciton radius. In calculations, we have used the following model. An electron and a hole are localized in different wells. The distance between wells is equal to δ. For the estimation, we took into account only the ground state and two lowest excited states, the dipole transition matrix element of which from the ground state to the excited one does not equal to zero. The wave functions of such a type have the form

$$|0\rangle = A_0 \exp(-\alpha_0 r), \quad |1a\rangle = A_{1a} r\cos\varphi \exp(-\alpha_{1a} r),$$

$$|1b\rangle = A_{1b} r\sin\varphi \exp(-\alpha_{1b} r), \qquad (19)$$

r is the distance between an electron and a hole in an exciton in the plane of wells, $\alpha_0, \alpha_{1a}, \alpha_{1b}$ are variational parameters. The results of calculations of the dipole-dipole and van der Waals interactions ($\langle 0,0|U_{dd}|0,0\rangle + U_{vdw}$) are presented in Fig. 4 for different values of δ. The energy of a bulk exciton is chosen as a unit of energy, radius, of bulk exciton (the value of order of 50 Å) is the unit of length. At a large distance, the interaction between excitons is dipole-dipole repulsive. But the attractive van der Waals interaction exceeds the dipole-dipole repulsion at distances less than 3—6 exciton radii, and the total interaction is attractive. Taking into account the other higher excited intermediate states in (18) should lead to increasing the presented estimations of the van der Waals interaction. So, the van der Waals interaction is really larger. At small distances between excitons, the exchange interaction becomes important, also the approximation of the dipole-dipole interaction becomes inapplicable. As a result, the total interaction is repulsive at small distances.

It should be noted that the attractive interaction may be significant at low temperatures when the condensed phase is created. At higher temperatures, excitons are distributed uniformly over a crystal, the mean distance between excitons exceeds the region of attraction, and the main contribution to the exciton-exciton interaction is given by the dipole-dipole one.

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ФОРМУВАННЯ ПОРЯДКУ В СИСТЕМІ ОСТРІВЦІВ КОНДЕНСОВАНОЇ ФАЗИ ЕКСИТОНІВ У КВАНТОВИХ ЯМАХ

В.Й. Сугаков

Р е з ю м е

На основі теорії формування конденсованої фази екситонів у двовимірній системі пояснюється утворення періодичних структур, яке було виявлено в останні роки в спектрах люмінесценції напівпровідників з подвійними квантовими ямами. Області конденсованої фази екситонів періодично розміщені у вигляді острівців вздовж кільця, утвореного навколо лазерної плями. Знайдено температурні залежності радіуса острівців, відстані між ними (періоду структури) та флуктуацій відстані.