Bose-Einstein condensation of dipolar excitons in quantum wells

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Abstract. The experiments on Bose-Einstein condensation (BEC) of dipolar (spatially-indirect) excitons in the lateral traps in GaAs/AlGaAs Schottky-diode heterostructures with double and single quantum wells are presented. The condensed part of dipolar excitons under detection in the far zone is placed in \( k \)-space in the range which is almost two orders of magnitude less than thermal exciton wave vector. BEC occurs spontaneously in a reservoir of thermalized excitons. Luminescence images of Bose-condensate of dipolar excitons exhibit along perimeter of circular trap axially symmetrical spatial structures of equidistant bright spots which strongly depend on excitation power and temperature. By means of two-beam interference experiments with the use of cw and pulsed photoexcitation it was found that the state of dipolar exciton Bose-condensate is spatially coherent and the whole patterned luminescence configuration in real space is described by a common wave function.

1. Introduction
In the last decade, BEC of excitons in 2D systems based on semiconductor heterostructures has been an object of intensive research [1-22]. In such semiconductor structures 2D exciton polaritons and photoexcited dipolar excitons in spatially separated electron-hole (e-h) layers are in the focus of interests. Recently, a comprehensive set of experiments giving compelling evidence for the BEC of 2D excitonic polaritons in microcavities has been presented in paper [12] (see also [13]). A major difference between the exciton system in semiconductors and the atomic condensate is that photoexcited excitons are ephemeral objects due to the finite lifetime. This time, for instance, in the case of excitonic polaritons in microcavities is very short (a few picoseconds). The system of photoexcited excitons in spatially separated e-h layers (for instance, in double quantum wells (DQW) and in a wide single quantum well (SQW)) under applied electrical bias is very attractive for the search of BEC in many aspects. First of all, because the lifetime of excitons with spatial e-h separation can be controlled by the external bias and is long enough, allowing the excitons to reach kinetic quasi-equilibrium with each other and to be cooled down to the temperature of the lattice where excitons are “immersed”. Particularly, in DQW under electric bias applied in the growth direction and tilting quantized subbands, it is possible to excite excitons whose electron and hole are in different QWs separated by a barrier [2-5]. In the case of DQW such dipolar exciton is called a spatially indirect or interwell exciton (IE), as opposed to a direct (DE) or intrawell exciton in which electron and hole are placed within the same QW. Interwell excitons have a longer lifetime than intrawell ones because of the reduced overlap of electron and hole wave functions in the direction of applied electric field.
Characteristic times of the radiative recombination of dipolar excitons are as large as tens and hundreds of nanoseconds. Therefore, in principle, the gas of interwell excitons can be cooled down to rather low temperatures. Because of broken central symmetry, such excitons have a large dipole moment even in the ground state. In below we consider dipolar excitons which can not be bound into molecules or other complexes due to dipole-dipole repulsion and, consequently, can not condense into a liquid state [20].

BEC in a 2D-exciton gas can occur only under spatial lateral restriction. By collecting 2D-dipolar excitons in a lateral trap, a lower total flux of excitons, i.e. lower pumping power is required. Recently, researchers have been concerned with preparation of controllable traps for dipolar excitons with the use of external impacts like inhomogeneous deformation [9] and no uniform electric field [7, 8,10].

2. Experimental

Here we present set of experiments connected with the Bose-condensation of dipolar excitons in a ring lateral trap in both DQW and SQW GaAs/AlGaAs Schottky-diode heterostructures. Details of the architecture of the used heterostructures one can find in papers [11, 14, 16].

2.1. Electrostatic traps for dipolar excitons

The surface of structures was coated with metallic Au-Cr mask containing circular windows prepared by means of “lift-off” lithography (the diameters of windows were 2, 5 and 10 µm). Through such windows the photoexcitation and simultaneous microscopic detection of photoluminescence with high spectral and spatial resolution (around 1.5 µm) were performed under cw and pulsed laser photoexcitation. It was found that under applied bias and photoexcitation the radial distribution of electrical field inside investigated windows is strongly non-uniform. Scattered field is minimal at the center of window and increases radially to the window edge. Most important is that close to the edge of window electric field behaves in radial direction non-monotone. A rather deep ring-like potential trap for interwell excitons is formed along the perimeter of a window. The appearance of a trap for dipolar excitons follows from the calculations carried out within the framework of electrostatic approximation (see for instance [17]). Experimentally, the existence of such a ring-like potential trap for dipolar excitons in the Schottky-diode heterostructures was controlled by the analysis of spectral shifts of exciton luminescence line under scanning of strongly focused excitation spot in radial direction in the vicinity of window edge. Here we present illustration for a wide SQW which demonstrates accumulation of dipolar excitons in a lateral trap appearing close to the window edge under applied bias. For this aim the enlarged image of a circular window in metallic mask was symmetrically projected onto the entrance slit of spectrometer. The observed luminescence spectrum taken from 5µm-window for SQW is illustrated by figure 1. One can see two bright spots of dipolar exciton luminescence (labeled by I in figure 1) located at the upper and lower edges of window where excitons are just accumulated in the trap. The size of spots in radial direction reflects the trap width.

Figure 1. Photoluminescence spectrum of a wide (250Å) GaAs/AlGaAs single quantum well measured within 5-µm circular aperture on the Schottky gate electrode with high spatial resolution along spectral slit (the x axis). Excitation power \( P_{He-Ne} = 5 \, \mu W \). \( T = 1.7 \, K \).
and amounts to \( \approx 1.5 \mu \text{m} \). The luminescence of direct excitons (D) is seen as well, but at the center of window where scattered electric field is strongly reduced. Besides, one can see in figure 1 weak luminescence traces (the upper and lower traces from the center of pictures) corresponding to the “hot” luminescence of e-h pairs drifting from the window center to the circular ring trap at the window edge. The shape of the traces reflects basically the shape of the trapping potential for the dipolar excitons. The shape of the potential in the radial direction near the bottom of the trap may be described approximately by a parabolic dependence \( V(r) = \alpha r^2 \) where the force constant is \( \alpha \approx 2.2 \text{ meV/} \mu \text{m}^2 \) and the barrier height is \( \Delta V \approx 5 \text{ meV} \) under conditions of figure 1.

2.2. Dipolar exciton Bose-condensation displayed in photoluminescence

Now we turn to the investigations of dipolar exciton luminescence seen directly from the lateral trap (the bright spots in figure 1). The formation and behavior of the narrow line of dipolar excitons, corresponding to exciton condensate is presented in figure 2. This line appears atop the luminescence continuum background connected with localized e-h states [6, 14, and 16]. These experiments were carried out with simultaneous use of two cw lasers, He-Ne and Ti-Sp, to compensate extra charges appearing under investigations (see [14, 16] for details). At the condensation threshold the spontaneous luminescence spectra behave strongly nonlinearly with excitation power increase. The FWHM of the narrow dipolar exciton line drops almost twice. The integrated intensity of the line increases superlinearly in accordance with macroscopic occupation of the lowest state in a trap. Close to the threshold the line shifts to the lower energy side by the amount around \( kT \) in accordance with macroscopic occupation of the lower state in a trap. On the further excitation power increase dipolar exciton line exhibits monotonous high energy shift and slight broadening in accordance with

**Figure 2.** The narrow line of dipolar excitons, corresponding to exciton condensate arises in the spectrum of photoluminescence seen from a 5\( \mu \text{m} \)-window in Schottky-diode SQW heterostructure on excitation power increase. \( P_{\text{He-Ne}} \) rises from 3 to 80 \( \mu \text{W} \) in steps \( \approx 40\% \). \( P_{\text{Ti-Sp}} = 60 \mu \text{W} \). Bias \( U = +1.46 \text{ V} \). \( T = 1.7 \text{ K} \).

**Figure 3.** The far-field (angular) distribution of the spontaneous luminescence of dipolar excitons from a 5\( \mu \text{m} \)-window in Schottky-diode SQW-heterostructure narrows with excitation power increase. \( P_{\text{He-Ne}} \) changes from 3 to 100 \( \mu \text{W} \) in steps \( \approx 45\% \). \( U = +1.46 \text{ V} \). \( T = 1.7 \text{ K} \).
Theoretical prediction [20]. The estimated single-particle thermal de Broglie wavelength nearby the condensation threshold exceeds interparticle distance almost three times and amounts to \(\sim 0.3 \, \mu m\).

The spatial distribution of the spontaneous luminescence of dipolar excitons detected in the far field exhibited evident angular narrowing on excitation power growth (see figure 3). The observed narrowing of the far-field spatial distribution of dipolar excitons demonstrates that above condensation threshold dipolar excitons are accumulated in \(k\)-space close to \(k \approx 0\), namely, in the range which is more than one order of magnitude smaller than the average thermal wave vector for the Maxwell-Boltzmann distribution at \(T = 1.8 \, K\). This is a direct consequence of Bose-Einstein statistics. The found reciprocal width of luminescence peak \((\Delta k)^{-1} \approx 10^4 \, \text{cm}\) is very close to the radial width of the trap \(\Delta x \leq 2 \, \mu m\) in accordance with uncertainty principle: \(\Delta k \cdot \Delta x \approx 1\). The described spectral behavior on excitation power above threshold is a result of stimulated scattering of dipolar excitons to the lowest state in the trap accompanied by the macroscopic occupation of this state by excitons. The rate of the stimulated exciton scattering \(\gamma_{ex}\) is proportional to the number of excitons in the final state: \(\gamma_{ex} \sim (n_{ex} + 1)\), where \(n_{ex}\) is exciton occupation number at \(k \approx 0\). So, we assume that \(n_{ex} \approx 1\) just at threshold (average exciton concentration at threshold equals to \(5 \cdot 10^9 \, \text{cm}^{-2}\)) and \(n_{ex} > 1\) above threshold. By means of direct measurements of the absolute luminescence intensity and taking into account the setup geometry (i.e. the efficiency of luminescence light collection) and dipolar exciton lifetime it was found that the exciton occupation number at threshold is equal to 1 indeed with an accuracy factor of 2.5. On further increase of excitation power above threshold the width of dipolar exciton line starts to grow and its spectral position moves to the higher energy side reflecting increase of exciton-exciton interaction in qualitative agreement with theoretical predictions published in [20].

### 2.3. Spatial structure of condensed dipolar excitons in a circular trap

Now let us consider experiments on spatially-resolved luminescence when a window of corresponding size was directly projected with high resolution on the CCD-camera avoiding spectrometer [11, 14, 16]. In these experiments the luminescence of dipolar excitons was spectrally selected by means of a narrow-band interference filter. At minimal excitation powers, corresponding to average exciton concentration less than \(10^9 \, \text{cm}^{-2}\), the luminescence intensity within the window is structureless and almost homogeneous. On excitation power increase a discrete luminescence structure consisting of four equidistant spots appears in a threshold manner along the window perimeter. The intensity redistribution between two pairs of spots directed along vertical and horizontal diameters was found to increase with excitation power. The spot sizes are around \(1.5 – 2 \, \mu m\). The geometrical configuration of spots is always aligned along crystallographic direction \(<110>\) in the \{001\} crystallographic plane due to the pinning by random potential fluctuations. Finally, at excitation powers above 200 \(\mu W\) only structureless luminescence of a ring shape remains visible.

**Figure 4.** Spatial structure of dipolar exciton luminescence in the 5-\(\mu m\) window at different temperatures (SQW heterostructure with Schottky-electrode). Excitation power \(P_{\text{He-Ne}} = 10 \, \mu W\). \(U = +1.46 \, V\).

**Figure 5.** Real-space image (a) and optical Fourier-transform (b) for the dipolar exciton luminescence from a 5-\(\mu m\) window, (c) – Fourier-transform for direct exciton emission. \(P_{\text{He-Ne}} = 50 \, \mu W\), \(T = 1.7 \, K\), \(U = +1.46 \, V\).
For the 5 μm window, taken as an example, the behavior of a discrete configuration of four equidistant spots of photoluminescence was studied under temperature variation at a given excitation power. It was established that on temperature increase the discrete luminescence structure starts to wash off at $T > 6$ K. Finally, the whole discrete structure of equidistant spots is merged in a continuous luminescent ring at $T ≥ 10$ K (figure 4).

The patterned structures of equidistant spots were observed in the case of indirect exciton photoluminescence in DQW as well. The observed structures were also sensitive both to the excitation power and temperature. Luminescence spots in the frames at a given excitation power are merged all together in a continuous ring above 10 K [11, 14].

It is worth to emphasize that spatially-resolved luminescence of direct excitons, measured under the same experimental conditions in the center of a window was always structureless.

2.4. Long-range spatial coherence of dipolar exciton Bose-condensate

As a diagnostics of the Bose-condensation of dipolar excitons confined in a lateral trap it is constructive to know the angular distribution of photoluminescence around direction normal to the 2D-plane. If a peak in the photoluminescence in considered system appeared in the direction normal to the plane it would be a straight evidence of the long-range coherence of condensate, as it was shown recently in [21]. For this aim we performed in situ optical Fourier-transforms of the equidistant-spot patterns (i.e. transformation from real $r$- to $k$-space) [14, 16].

The experimentally found Fourier-transform patterns demonstrate with evidence results of destructive and constructive interference (see figure 5). First of all, it is an evidence of a long-range spatial coherence of the patterned luminescence structure in real ($r$-) space. Secondly, the luminescence of condensate is directed normally to the heterostructure plane in a cone with opening angle $\Delta \phi$ close to $\lambda/D ≈ 0.19$ rad.

2.5. Time-resolved two-beam interference and coherence of exciton Bose-condensate

Recently, we have measured the coherent length independently in a straight way by means of two-beam interference which originates from a pair of luminescence spots seen in luminescent ring (an analogue of the classical experiment on amplitude superposition from two independent light sources) [14, 16]. The interference is easily evidenced with the help of a slit placed in the plane of magnified intermediate image. The slit is positioned symmetrically to choose a couple of bright spots seen at the opposite ends of ring diameter. The interference fringes are observed at a distance adequate for overlapping of light waves emitted from the two sources. As far as distance between the sources is around 4 μm, a conclusion follows about long-range spatial coherence of emitted light [14,16].

![Figure 6](image_url)

**Figure 6.** Interference of light emitted from two spots located at the opposite sides of a 5μm luminescence ring (see figure 4): stationary interference fringes - at the left, and time-resolved pattern – in the main panel. Red (on-line) curve shows laser pulse. Laser power: pulsed – 6 μW, cw – 60 μW.

Now we turn to time-resolved two-beam interference which could give valuable information about
formation and decay dynamics of the coherent collective state arising in a system of dipolar excitons in the ring-shaped lateral trap above excitation threshold. Photoexcitation in time-resolved experiments was realized with pulsed (\( t_p = 100 \) ps, \( f = 10 \) MHz) semiconductor laser (wavelength \( \lambda = 659 \) nm). In addition, a cw laser-diode illumination (\( \lambda = 782 \) nm) was applied: a) to approach neutral charge balance in the structure under study [14] and b) to increase the photoluminescence intensity. A magnified interference pattern was projected onto the entrance slit of high-speed streak-camera with fringes oriented perpendicular to the slit. The setup time resolution was not worse than 200 ps.

The main experimental result is shown in figure 6. Due to the presence of cw photoexcitation the luminescence does not decay to zero between laser pulses: the pulsed signal sits on slowly varying pedestal. But the interference pattern (i.e. horizontal lines in figure 6 corresponding to the interference orders \( m = 0, \pm 1, \pm 2 \)) is not persistent. It appears with some delay (~ 1 nsec) relative to the excitation pulse and it vanishes before the luminescence pulse decayed completely. This fact is confirmed also by the correspondent Fourier analysis. It means that the coherence appears with time delay due to effect of thermalization and “lives” only when a critical exciton density is exceeded.

3. Conclusion
The whole set of presented experiments is a manifestation of collective properties of interacting 2D dipolar excitons and it is a direct consequence of their Bose-statistics. It is an effect of dipolar exciton BEC in a lateral ring-shaped trap. The condensation occurs spontaneously in a reservoir of interacting excitons under thermal quasi-equilibrium. The collective state is coherent and the found length of spatial coherence is close to the perimeter of a trap. We believe that this state is described by a single wave function.

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