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Scientific session of the Physical Sciences Division of the Russian Academy of Sciences dedicated to the 60th anniversary of the Zavoisky Kazan Physical-Technical Institute of the Kazan Scientific Center, Russian Academy of Sciences (9 February 2006)

A scientific session of the Physical Sciences Division of the Russian Academy of Sciences (RAS) dedicated to the 60th anniversary of the Zavoisky Kazan Physical-Technical Institute of the Kazan Scientific Center, Russian Academy of Sciences, was held in the Conference Hall of the Zavoisky Kazan Physical-Technical Institute on 19 December 2005. The following reports were presented at the session:

(1) **Krokhin O N** (Lebedev Physics Institute, RAS, Moscow) "50 years of quantum electronics";

(2) **Gorbunov A V, Timofeev V B** (Institute of Solid State Physics, RAS, Chernogolovka, Moscow region) "Bose condensation of interwell excitons and spatial structure of luminescence in lateral traps";

(3) **Chekalin S V** (Institute of Spectroscopy, RAS, Troitsk, Moscow region) "The unique femtosecond spectrometric complex as an instrument for ultrafast spectroscopy, femto-chemistry, and nanooptics";

(4) **Salikhov K M** (Zavoisky Kazan Physical-Technical Institute of the Kazan Scientific Center, RAS) "Timeresolved EPR spectroscopy of nonequilibrium spin systems produced during spin-dependent photophysical and photochemical processes in condensed media";

(5) **Manenkov A A** (Prokhorov General Physics Institute, RAS, Moscow) "The role of electron paramagnetic resonance in the development of quantum electronics: facts and comments";

(6) **Smirnov A I** (Kapitza Institute for Physical Problems, RAS, Moscow) "Magnetic resonance modes in spin-gap magnets";

(7) **Kochelaev B I** (Kazan State University) "Evolution of antiferromagnetic cuprates in high-temperature superconductors";

(8) Garifullin I A (Zavoisky Kazan Physical-Technical Institute of the Kazan Scientific Center, RAS) "The superconductor/ferromagnet proximity effect and its potential application in spintronics."

The main propositions of O N Krokhin's report were published in *Usp. Phys. Nauk* **174** 1117 (2004) [*Phys. Usp.* **47** 1045 (2004)]. A brief presentation of reports 2-6 and 8 is given below.

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Bose condensation of interwell excitons and spatial structure of luminescence in lateral traps

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We discuss experiments involving direct observations of the collective state in the Bose gas of interacting interwell excitons in double GaAs/AlGaAs quantum wells. In recent years, the possibility of excitonic Bose condensation in quasi-two-dimensional semiconductor systems was actively studied, both experimentally and theoretically [1-16]. These investigations have acquired fundamental significance in connection with impressive achievements in the study of the Bose-Einstein condensation of diluted and deeply cooled atomic gases accumulated in magnetic traps [17]. Among the phenomena studied most thoroughly in experiments are excitons in tunnel-bound double quantum wells in heterostructures [1-3]. In these structures, upon application of a bias voltage in the direction perpendicular to the heterolayers, photoexcited electrons and holes separate to become distributed between the neighboring quantum wells, and are bound together by the Coulomb attraction to make up interwell excitons (Fig. 1). Unlike intrawell excitons, interwell excitons have a dipole moment even in the ground state and cannot combine into exciton molecules or many-particle complexes because of the dipole-dipole repulsion. Interwell excitons are experimentally attractive because they are relatively long-lived: their lifetimes are much longer than the characteristic thermalization times [1, 10]. The Bose condensation of the interwell excitons may be realized only when their free motion is spatially restricted to the planes of the wells, i.e., in lateral traps. In the traps, the interwell excitons are much easier to accumulate at sufficiently low temperatures, and it is thus simpler to realize the critical conditions whereby their Bose condensation may occur [11]. Such traps may be of various origins. For instance, they occur in a natural way in the presence of large-scale fluctuations of the chaotic potential associated with random variations of quantum well widths. The traps for the interwell excitons may be prepared with the aid of inhomogeneous deformations [12] and by producing a strongly

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Figure 1. Diagram of optical transitions in heterostructures with double quantum wells (QWs) under a bias voltage applied in the direction perpendicular to the plane of the quantum wells. Optical transition D corresponds to the radiative recombination of a direct (intrawell) exciton and transition I to an indirect (intrawell) exciton.

nonuniform electric field, for instance, employing the tip of a tunnel microscope [13].

Earlier, we demonstrated [3] that in the operation with double quantum wells in n⁺-i-n⁺-GaAs/AlGaAs heterostructures characterized by large-scale fluctuations in the random potential, a very narrow line emerges in the luminescence spectra of interwell excitons in a threshold manner with an increase in optical pumping. The observations were performed through micrometer-sized round holes in a metal mask. With increasing the temperature, the intensity of this line for a fixed pump power lowers, not in the activation manner, but linearly, to a complete disappearance from the spectrum. Experiments of this kind were performed on different high-quality structures, which somewhat varied in architecture and parameters. Nevertheless, the essence of the effect, which involved the threshold emergence of a narrow interwell-exciton luminescence line with increasing the pump power when observations were made through windows in a metal mask, as well as the disappearance of this line by a close-to-linear power law with increasing the temperature, was always reproducible in general. The investigations performed gave grounds to conclude that this effect is associated with the Bose condensation of the interwell excitons in lateral domains (traps) arising from large-scale chaotic fluctuations.

We now address ourselves to an entirely different problem related to the observation of a spatial luminescence structure under the conditions of the Bose condensation of interwell excitons in lateral traps. Recently, this problem was experimentally solved in Ref. [16], where the luminescence of interwell excitons was investigated with a high spatial resolution, with the luminescence excitation and observation effected through variously sized round windows in a metallic mask. The experiments were performed with $GaAs/Al_xGa_{1-x}As$ heterostructures (x = 0.3) with double GaAs quantum wells 12 nm in width. A four-monolayer AlAs barrier was located between the quantum wells. AlAs barriers of the same thickness were grown at the heteroboundaries between each quantum well and the insulating AlGaAs layers, which were 150 nm thick. The structures were Schottky photodiodes. The role of the inner electrode was played by the two-dimensional electron gas in a broad, 30 nm GaAs quantum well spaced at the distance 150 nm from the double quantum well located deep inside the structure. The two-dimensional electron gas in the broad quantum well appeared due to a silicon-doped delta layer placed near the broad well. A metallic film (a 100 nm thick aluminum layer) sputtered on the heterostructure surface served as an external Schottky gate. A bias voltage was applied between the inner electrode and the Schottky gate.

We first illustrate the quality of the structures with double quantum wells used in our work with the example of the luminescence spectra of interwell and intrawell excitons obtained when the electric voltage across the electrodes was varied (Fig. 1 shows the diagram of optical transitions and Fig. 2 the evolution of the luminescence spectra under variation of the bias voltage). In these experiments, a semitransparent metallic film (the Schottky electrode) covered the entire working area of the structure, and the electric field along the structure surface between the electrodes was rather uniform. Luminescence photoexcitation and observation were effected through a semitransparent metallic electrode. For a bias voltage U > 0.7 V, when a flat-band mode is realized in the structures investigated, the spectrum shows only the intrawell exciton luminescence line D. On application of an electric field, the interwell exciton line I emerges with the width about 1.7 meV. In accordance with the Stark shift of dimensional quantization levels in quantum wells, this line moves towards lower energies as a linear function of the applied bias voltage, while the intrawell exciton line lowers in intensity to eventually disappear from the spectrum. For spectral shifts exceeding the interwell-exciton binding energy by more than an order of magnitude, the intensity of the luminescence line of such excitons remains almost invariable, although their radiative decay time lengthens considerably with the bias voltage. This signifies that nonradiative processes are of little significance in the structures involved and that the structure quality is sufficiently high. Also seen in the luminescence spectra is an intense line with a peak at the energy 1.5143 eV, which corresponds to the direct radiative recombination of two-dimensional electrons in a broad (30 nm) quantum well. This luminescence line lies some distance away from the spectral range of interest and its origin and properties are not discussed here.

Subsequent work was performed with structures that were coated with an opaque metallic film, which played the role of a Schottky gate. Circular windows of various diameters (from 2 to 20 μ m) were etched in this film using electron-beam lithography; the photoluminescence excitation and recording of interwell and intrawell excitons were effected through these windows. A specifically designed microscopic device enabled observing, through these windows, the spatial luminescence structure with the resolution up to 1 μ m. The magnified image of the window of a sample, through which the photoexcitation was effected, was projected precisely onto the plane of the



Figure 2. Evolution of the photoluminescence spectra of a two-dimensional structure with a double quantum well and a semitransparent electrode deposited atop (a Schottky photodiode) when lowering the voltage across the electrodes from 1.1 V to -1.3 V in 0.05 V decrements. The temperature is T = 1.7 K. The He-Ne laser excitation power is $P = 1 \mu$ W. The size of the luminescence-exciting laser spot on the sample is 30 μ m. Indicated are the luminescence lines of intrawell (D) and interwell (I) excitons. The inset shows the current-voltage characteristic of the structure under investigation; for a voltage above 0.7 V, a flat-band mode is realized in the structure.

photosensitive matrix of a cooled silicon CCD camera. The sample was placed into a helium cryostat, which enabled performing the experiments in the 1.6–25 K temperature range. At a temperature T > 4.2 K, the sample was embedded in He⁴ vapor. With the aid of interference light filters, it was possible to study the spatial structure of luminescence selectively in spectral composition: either for the interwell excitons or for the intrawell excitons under the same experimental conditions. The photoexcitation was effected with an He–Ne laser and the spectra were recorded using a 'Ramanor-U1000' double monochromator. In all experiments conducted with voltage application, the current through the structure did not exceed 100 nA for the pump power 100 μ W.

We first consider the experiments in which the window of a structure was projected onto the entrance spectrometer slit and the slit itself extracted only the central part of the window along the diametric line. At the outset, it was determined that the radial distribution of the electric field inside the structure near the window in the metallic mask was strongly nonuniform. The scattered field was lowest at the center of the window and increased in the radial direction towards the mask boundaries. Most importantly, the electric field behaves in a nonmonotonic manner immediately at the annular edge of the window, such that a circular potential well (trap) for interwell excitons along the window perimeter appears. The very fact that a circular potential well emerges near the edges of the window in the metallic mask is experimentally established from the spectral shifts of the interwell exciton luminescence line in the focused excitation scanning near the

window boundaries. The electric field is uniform under the metallic mask itself, but on the whole the field magnitude is smaller than in the region of the circular potential well. The reasons for such behavior of the electric field near the perimeter of the window in the mask, as well as the shape and depth of the circular potential well in relation to the bias voltage and the photoexcitation conditions, will be discussed and published elsewhere.

In the luminescence spectra for a $5 \mu m$ sized window projected onto the entrance spectrometer slit, observed were bright spots (measuring about $1.5 \mu m$) of interwell exciton luminescence located near the upper and lower edges of the window under investigation, as well as the luminescence of the intrawell excitons (D) at the window center, where the scattered electric field was not high. Figures 3a and 3b show how the narrow line of interwell excitons rises with the pump power against the continuous background, which is associated with localized excitons, when detection is effected under the above conditions. Figures 3c and 3d illustrate how this line vanishes against the continuous background when the temperature is increased from 1.6 to 4.2 K for a fixed pumping. These results are qualitatively equivalent to those published earlier, for instance, in Refs [3, 4].

We now turn to the description of other experiments in which the spot of luminescence from a window of an appropriate size was projected directly onto the detector (a CCD camera) with the resolution up to 1 μ m, rather than onto a spectral device. In this case, the luminescence of interwell or intrawell excitons was selected with the aid of interference light filters. Under the lowest excitation power,



Figure 3. (a) Photoluminescence spectrum of a narrow interwell exciton line as a function of the pump power for a 5 μ m sized window in a metallic mask (the window is projected onto the entrance spectrometer slit). Values of the excitation power indicated to the right of the curves correspond to the spectra measured at T = 1.7 K. The laser radiation is focused to a spot 30 μ m in size. (b) Intensity of the narrow interwell exciton line as a function of the pump power; the dashed curve shows a quadratic approximation of intensity versus pump power. (c) Intensity variation of the narrow photoluminescence line with the temperature in the range 1.6-4.2 K for the fixed pump power 5 μ W; the temperature values indicated to the right of the curves correspond to the spectra. (d) Temperature dependence of the intensity of the narrow luminescence line; the dashed straight line represents a linear approximation of this dependence.

whereby the average density of interwell excitons does not exceed 10^9 cm^{-2} (this corresponds to a laser pump with the power about $1 \,\mu\text{W}$ in a laser spot measuring 50 μm), the luminescence spot was found to be structureless and the luminescence intensity to be uniformly distributed within the limits of the window [16]. With increasing the pump power, a discrete structure of luminescence spots symmetrically located along the window perimeter appears in a virtually threshold manner: two spots appear initially, then four spots appear with an increase in the pump power, and subsequently six spots appear. The luminescence spots measure about 1.5 – $2 \,\mu\text{m}$. For the pump power about 150 μ W, the number of spots is no fewer than eight, but the structure itself is not clearly resolved. Eventually, for the pump power exceeding $200 \,\mu\text{W}$, the discrete structure of luminescence spots smears, turning into a structureless luminescent ring. In the majority of windows investigated, the intensity distribution over the spots in the structure is not strictly axially symmetric: some of the spots can stand out in intensity.

A clear structure of four equidistant luminescence spots is observed in a window measuring 2 μ m. The luminescence structure in windows measuring 10 μ m is more complex: apart from the discrete axially symmetric spot structure, a radial ring structure is also observed. In windows measuring 20 μ m or more, we failed to observe any structure. We emphasize that the spatial luminescence structure in the case of intrawell exciton luminescence is nonexistent in experiments with windows of any size, i.e., the intensity of intrawell exciton luminescence is, under all conditions realized, uniform and practically constant within the limits of the windows.

By the example of a window 5 μ m in diameter, we investigated the behavior of a temperature-dependent discrete configuration of the hexagonal symmetry made up of six luminescence spots located equidistantly along the perimeter of a circle (Fig. 4). It was found that the effect exhibited a manifestly critical behavior with increasing the temperature for a fixed pumping: specifically, the structure of regularly located luminescence spots began to smear for $T \ge 4$ K, transforming into a single structureless ring at a temperature about 15 K (Fig. 4e).

It is worth noting that the spatial distributions of intrawell exciton luminescence, which showed up in a near-circular form with a fragmentary axial structure along a ring contour, were earlier observed by Butov et al. [5, 6] in double quantum wells and by Snoke et al. [7, 8] in single quantum wells. Such circular luminescence structures appeared under sufficiently intense focused laser excitation, and the dimensions of the circular structures increased to range into the hundreds of microns with increasing the pump power. They owe their origin to the electron depletion and screening of the applied electric field in the domain of laser pumping, as well as to oppositely directed drifts of electrons and holes that developed under these conditions. Therefore, in our view, there is no direct relation between the experiments in Refs [5-8] and the experiments described in the foregoing, the results of which were reported in Ref. [16].

Our experiments discussed above were carried out on several dozen windows of diameters 2, 5, 10, and 20 μ m in metallic masks. The spatial configurations of luminescence spots in windows of a fixed size were always reproducible under identical conditions. Therefore, there are no grounds to



Figure 4. Spatial structures of interwell exciton luminescence obtained in measurements with a 5 μ m window in a metallic mask under the pump power 50 μ W at the temperatures 1.7 K (a), 3.0 K (b), 4.25 K (c), 4.33 K (d), and 15 K (e). (f) Intrawell exciton photoluminescence pattern in a 5 μ m window for the pump power 50 μ W and the temperature 1.7 K. The orientation of the sides of each frame corresponds to the directions (110) in the {001} plane of the heterostructure under investigation.

believe that the action of a random potential, which is inevitably present in the samples under investigation, is the prime cause of so regular a formation of the spatially periodic structures of interwell exciton luminescence. In this connection, it is pertinent to note once again that no spatially periodic structures were observed in the case of intrawell exciton luminescence measured under the same experimental conditions.

We now consider other possible causes of the effect under discussion associated with the discovery of spatially periodic, symmetric configurations of interwell exciton luminescence involving luminescence excitation and observation through circular apertures in a metallic mask.

The possibility of the emergence of interwell exciton density waves accompanied by a periodic redistribution of luminescence brightness was predicted in Ref. [18]. However, it is not clear why the period of these waves should change in a stepwise manner with a change in the pump power and why the effect itself should be so critically sensitive to the temperature and the pump on the whole.

Furthermore, in experiments with a focused laser excitation, there always arises a flow of nonequilibrium phonons (a 'phonon wind') from the photoexcitation region, which entrains charge carriers and excitons [19]. Centrally symmetric, spatially extended luminescence patterns, which are reflective of the linear dimensions of the phonon entrainment in the crystal medium, emerge under these conditions; the phonon entrainment magnitude is different for different crystallographic directions [20]. In our case, the pumps are moderate and, most importantly, the temperature criticality of our observed effect is inexplicable on the basis of the phonon wind concept.

The transmission of luminescence radiation through small holes may in principle be affected by surface plasmons of a metallic mask (the polariton effect associated with surface plasmons) [21]. However, with an increase in the window size, this effect is bound to wane and, moreover, the critical behavior under pump and temperature variation should be missing.

Finally, as stated in theoretical paper [15], in the case of attraction between interwell excitons, they can condense into a liquid dielectric phase. By analyzing the nonlinear equation for the exciton density, the author of Ref. [15] arrived at the conclusion that the exciton condensate may have a circular

structure fragmented by equidistantly located droplets of the exciton liquid. However, earlier, in Ref. [9], it was shown that the stability of the interwell-exciton liquid phase under the most optimal conditions is very low. Therefore, the question of stability of the liquid dielectric phase of interwell excitons is still an open question in our opinion.

We believe that the effect discovered has a collective, coherent nature. It occurs in the system of interacting twodimensional interwell excitons in the condensation in a lateral ring-shaped trap. Collective states are characterized by large coherence lengths (over 1 µm) and are destroyed with increasing the temperature due to the destruction of the order parameter. Coherent coupling between the luminescence spots exists and therefore quantum temporal beats should be observable in the conditions of pulsed excitation. The effect has a clearly defined limit on the low-excitationintensity side (low exciton densities) and is not observed when the interwell exciton mobility is below the threshold value defined by the effects of exciton localization in random potential fluctuations. In the strong-pumping domain, the effect also vanishes, which, however, is due to the ionization breakdown of the interwell excitons (the Mott transition). Nevertheless, the pump-power dynamic range within which the effect is observed is rather broad, nearly two orders of magnitude.

In a recently published theoretical paper, Keeling et al. [14] showed that the vortex nature of the condensate produced in the interwell-exciton Bose condensation in a lateral trap manifests itself in the properties of the angular luminescence intensity distribution due to a destructive interference. We emphasize that the expected real-space vortex configurations presented in Ref. [14] are practically identical to our observed patterns of bright luminescence spots equidistantly spaced along the perimeter of a 5 µm window. By way of illustration, we show the far-field photoluminescence patterns obtained from 5 µm windows in a metallic mask, which correspond to the optical Fourier transform of the real spatial structure observed through the window (Figs 5a-5d). The patterns given in Figs 5a and 5b and the patterns given in Figs 5c and 5d pertain to two different windows. The patterns in Fig. 5a and Fig. 5c were obtained for a low pump power (for a below-threshold mobility of the interwell excitons), when the real spatial luminescence intensity distribution in the window is struc-



Figure 5. Far-field photoluminescence intensity distribution patterns obtained through windows in a metallic mask. The patterns correspond to the optical Fourier transform of a real spatial luminescence structure (T = 1.7 K). The pairs of images in Figs 5a and 5b and in Figs 5c and 5d correspond to two different 5 μ m windows in a metallic mask. The patterns in Figs 5a and 5c were obtained for a low pump power, when the spatial intensity distribution in the window is structureless; the patterns in Figs 5b and 5d were obtained for a substantially higher pump power (about 90 μ W), when a spatial luminescence structure with the hexagonal symmetry appears in the window, which is similar to the structure shown in Figs 4a-4d.

tureless. In this case, the far-field intensity distribution is also uniform. We emphasize that the concentric interference fringes observed in the far-field patterns are due to diffraction by the exit pupil of the optical system and bear no direct relation to the subject under discussion. The situation is significantly different in the operation with a higher-intensity pump (about 90 μ W in power), when a spatially periodic luminescence structure with the hexagonal symmetry similar to that shown in Figs 4a-4d is observed in the window. Under these conditions, the far-field patterns depicted in Figs 5b and 5d clearly exhibit the effect of destructive interference (the intensity dip at the center of the far-field pattern), which is qualitatively consistent with the theoretical predictions in Ref. [14]. The above experimental observations are one of the manifestations of the coherence of the collective interwell exciton state in the conditions of Bose condensation in a lateral trap. Furthermore, the discovery of the destructive interference in the far-field patterns is an indication that the luminescence intensity of the condensed exciton phase has a manifest angular directivity relative to the normal to the structure surface, which is a salient feature of superradiance [22]

At the same time, the question of the extent to which the system of interwell excitons is electrically neutral remains open. An attempt to answer this question invites investigations of interwell exciton condensation under the resonance photoexcitation conditions.

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The unique femtosecond spectrometric complex as an instrument for ultrafast spectroscopy, femtochemistry, and nanooptics

S V Chekalin

1. Introduction. Retrospective excursus

Nearly the last forty years of the fifty-year-long history of quantum electronics have been related to ultrashort laser pulses (USPs) [1]. These light 'bullets' occupy a special place in the history of research in ultrafast process dynamics, because they have facilitated the swiftest progress in the measurement of short time intervals over the last several centuries (Fig. 1). The use of USPs enabled real-time investigations of the dynamics of ultrafast processes that had previously been considered 'experimentally unobservable.' Furthermore, the record-high USP power, which allows realizing light fields that are significantly stronger than intraatomic ones, makes the pursuance of unique experiments feasible [1]. The making of the first USP lasers in the USSR was fostered by the prospect of obtaining record-high powers for the solution of the laser fusion problem. Employing a laser facility [2] developed in the Quantum Radiophysics Laboratory in the Lebedev Physics Institute, RAS (FIAN), the first-ever successful experiment on the initiation of a thermonuclear reaction by high-power USP irradiation of a lithium deuteride target was implemented in 1968 [3]. The pulse energy amounted to 20 J for the pulse duration 20 ps. This 25 m long facility, which was the world's highest-power facility at that time, delivered no more than one pulse in