Two-Photon Correlations of Luminescence under Bose-Einstein Condensation of Dipolar Excitons

A. V. Gorbunov, V. B. Timofeev, D. A. Demin, A. A. Dremin
Institute of Solid State Physics, Russian Academy of Sciences, Chernogolovka, Moscow region, 142432 Russia

Abstract

Correlations of luminescence intensity (a 2nd order correlator $g^{(2)}(\tau)$, where $\tau$ is the delay time between photons in registered photon pairs) have been studied under Bose-Einstein condensation of dipolar excitons in the temperature range of 0.45–4.2 K. Photoexcited dipolar excitons were collected in a lateral trap in GaAs/AlGaAs Schottky-diode heterostructure with single wide (25 nm) quantum well under electric bias applied between heterolayers. Two-photon correlations were measured with the use of a classical Hanbury Brown-Twiss two-beam intensity interferometer with the time resolution of $\approx 0.4$ ns. Photon “bunching” has been observed near the Bose condensation threshold of dipolar excitons that was determined by the appearance of a narrow luminescence line of exciton condensate at optical pumping increase (FWHM of the narrow line at the threshold $\leq 200$ $\mu$eV). The two-photon correlation function shows super-Poissonian distribution, $g^{(2)}(\tau) > 1$, at time scales of system coherence ($\tau_c \lesssim 1$ ns). No photon bunching was observed with the used time resolution at the excitation pumping appreciably below the condensation threshold. At excitation pumping well above the threshold, when the narrow line of exciton condensate begins to grow in the luminescence spectrum, the photon bunching is decreasing and finally vanishes with further excitation power increase. In this pumping range, the photon correlation distribution becomes Poissonian reflecting the single-quantum-state origin of excitonic Bose condensate. Under the same conditions a first-order spatial correlator, $g^{(1)}(\tau)$, measured by means of the luminescence amplitude interference from spatially separated condensate parts under cw photoexcitation, remains significant on spatial scales around 4 $\mu$m. The discovered effect of photon bunching is rather temperature-sensitive: it drops several times with temperature increase from 0.45 K up to 4.2 K. If we assume that the luminescence of dipolar excitons collected in the lateral trap reflects directly coherent properties of interacting exciton gas, the observed phenomenon of photon bunching near condensation threshold – where exciton density and hence luminescence intensity fluctuations are most essential – manifests phase transition in interacting exciton Bose gas. It can be used as an independent tool for exciton Bose condensation detection.

PACS numbers: 73.21.Fg, 78.67.De

1. Investigations of two-particle spatially-temporal correlations (a 2nd order correlator $g^{(2)}(r, \tau)$) are increasingly popular particularly in connection with the analysis of complex quantum collective phenomena in ensembles of ultracold atoms [1, 2, 3]. The study of radiation intensity correlations, or two-photon correlations, goes back to a classical pioneering work of Hanbury Brown and Twiss [4] and its quantum grounds given later by Glauber [5]. It was demonstrated in these papers that photons emitted by a chaotic light source tend to bunch only under incoherent mixing or under superposition of coherent states (i.e. correlator $g^{(2)}(\tau)$ shows super-Poissonian distribution of two-photon correlations at a mutual coherent time $\tau_c$ of a radiation system). At the same time, in the case of a single-quantum-state source, which is coherent in all orders (i.e. single-mode laser [3], atom laser [3] or Bose condensate of atoms [1]), the correlator $g^{(2)}(r, \tau)$ is exactly of Poissonian type, with no bunching effect. The behavior of a spatially-temporal correlator is defined by quantum statistics of identical particles. Effect of bunching appears only for bosons (photons, magnons, atomsmicrons, etc.), whereas for fermions the two-particle correlations should exhibit antibunching behavior, which was recently observed in experiments for ultracold fermion atoms – $^3$He [2].

Here we present results of our experimental study of luminescence intensity correlations under conditions of Bose-Einstein condensation of dipolar excitons. As far as we know, it is the first experiment of this type for interacting dipolar exciton Bose gas collected in a lateral trap. The importance of luminescence intensity correlations exploration for exciton Bose condensation had been pointed out earlier in the theoretical paper [6]. Recently, behavior of two-photon correlations was investigated experimentally for 2D excitonic polaritons in microcavity heterostructures with quantum wells [7, 8].

2. Spatially-indirect or dipolar excitons were investigated in a wide (25 nm) single GaAs quantum well placed in a perpendicular to heterolayers electric field applied between a metal film (Schottky gate) on a surface of AlGaAs/GaAs-heterostructure and a built-in conductive electron layer in a structure [9, 10]. Due to the applied

*e-mail: gorbunov@issp.ac.ru
electric field, dipolar excitons have a big dipole moment in the ground state (more than 100 D). In the system under investigation such excitons do not bind into molecules or other multiparticle complexes due to dipole-dipole repulsion.

Photoexcitation of excitons and luminescence registration was performed through the circular window of ø5 μm size in a metal mask (Schottky gate). Dipolar excitons were collected in a ring lateral trap which arose along the window perimeter because of strongly non-uniform electric field. An increased image of the window was projected on an entrance slit of a spectrometer supplied with a cooled silicon CCD camera. The spectrometer transferred the image from the entrance slit plane to the exit slit plane without aberrations (“imaging spectrometer”), thus permitting to register sample image in a zero order of diffraction grating. The utilized optical system allowed to observe a spatial structure of dipolar exciton luminescence within the window in a metal gate with a resolution down to 1 μm. The sample was mounted in a helium optical cryostat with the working temperature range T from 0.45 K up to 4.2 K. At T < 1.5 K, the sample was immersed directly into liquid 4He, while at T > 1.5 K it remained in cold 3He vapor. A spatial structure of luminescence could be studied with spectral selectivity with the use of optical interference filters. The optical scheme enabled not only to observe a luminescence picture in a 5-μm window with a high spatial resolution, but to carry out also – by means of minor realignments – an optical Fourier-transform of images.

Dipolar excitons were excited by two cw lasers simultaneously: with wavelength λ = 782 nm (photoexcitation with photon energy less than the forbidden gap in AlGaAs barrier – “under-barrier” excitation) and λ = 659 nm (“above-barrier” photoexcitation). By adjusting the power ratio between these lasers, we reached maximal compensation of extra charges in the trap, whereas only the power of under-barrier laser was fixed: P_{782nm} = 10 μW. Temperature is T = 0.46 K. In this case the pumping was carried out by both lasers with λ = 659 nm and λ = 782 nm for maximal compensation of extra charges in a trap, whereas only the power of above-barrier photoexcitation (λ = 659 nm) was varied.

At very weak photoexcitation, an unstructured and asymmetric luminescence band with FWHM about 1.3 meV is visible in a spectrum. The shape of this band does not vary with pumping reduction. The band is inhomogeneously broadened. It originates from excitons localized on fluctuations of random potential because of residual charged impurities and structural defects both in the trap and in its vicinity. With excitation power increase at the violet edge of the band, a narrow spectral line corresponding to the condensed part of dipolar excitons appears above a certain power threshold and grows further in intensity. The linewidth near threshold is about 200 μeV, and its intensity increases in this pumping range superlinearly. The dependence of line intensity on pumping power becomes linear at the further increase of excitation power. At high photoexcitation, the line dominates in spectrum in comparison with an unstructured underlying continuum. The line slightly broadens and moves towards higher energies with pumping. Such a behavior is connected with a repulsive interaction of dipolar excitons on their concentration increase and it was analyzed in detail in [10, 17]. According to our measurements, the spectral line shift and broadening
are comparable. In particular, in the range of photoexcitation corresponding to Fig.1, the ratio of the spectral shift of the line center of gravity (spectral moment $M_1$) to its width (spectral moment $M_2$) is $M_1/M_2 \approx 1.25$. The maximal concentration of excitons in the pumping range of Fig.1 can be estimated using the spectral shift of the line ($\lesssim 300 \ \mu\text{eV}$): it does not exceed $10^{10} \text{ cm}^{-2}$.

An intensity of the luminescence line corresponding to exciton condensate is very sensitive to temperature. At fixed photoexcitation power intensity decreases linearly with temperature, down to its full disappearance in unstructured continuum near the threshold of exciton condensation. The temperature behavior of the narrow line of exciton condensate is illustrated by Fig.2. In the temperature range, $T = 0.45 \div 4.2 \ \text{K}$ the following law for the narrow line intensity $I(T)$ at fixed pumping was established: $I(T) \sim (1 - T/T_c)$, where $T_c$ is a critical temperature and the narrow spectral line disappears above $T_c$.

To plot phase diagram at each fixed temperature in the explored interval $T = 0.45 \div 4.2 \ \text{K}$, the dependence of luminescence spectra on photoexcitation power was investigated. As a result, the threshold power $P_{th}$ was defined when the narrow line of exciton condensate started to appear (disappear) in spectrum. The phase diagram was plotted in coordinates $P - T$, and for its construction the nonlinear range of the narrow line intensity dependence on laser excitation power was used. Fig.2 represents the resultant phase diagram. The phase boundary outlining the area of Bose condensation looks like a linear function of temperature, as it is expected for two-dimensional system.

The previous measurements of such phase diagram were performed for dipolar excitons in a structure with double QW [18]. A large concentration of structural defects in such a structure and, as a consequence, too high exciton mobility edge prevented from correct measurements at low density of free excitons at $T < 1 \ \text{K}$. In the present structure with single wide QW, the density of structural defects is almost ten times as low. In this case one manages to deal with free excitons, i.e. above mobility edge, down to 0.45 K. As a result, the linear extrapolation of phase boundary to the area of even lower temperatures and smaller exciton densities (see Fig.2 directly approaches the origin of coordinates.

4. Simultaneously with the narrow line origination in luminescence spectra of the Bose condensate of dipolar excitons, a spatially-periodic pattern of equidistantly located luminescent spots appears in the luminescence image, which is observed directly from a 5 $\mu\text{m}$-window with spatial resolution about 1 $\mu\text{m}$ and with spectral selection by means of optical interference filter with a bandwidth of 11 $\text{A}$ (details are presented in [16, 14, 15]). The structure of luminescence spots at fixed pumping also turned out temperature-sensitive: the spatially-periodic structure smears into a continuous ring at $T \lesssim 10 \ \text{K}$. In situ optical Fourier transforms of the spatially-periodic patterns which reproduce luminescence intensity distribution in far zone, showed results of both destructive and constructive interference, and also a spatial directivity of the luminescence normal to heterolayers. These result from large-scale coherence of the condensed exciton state in a lateral ring trap. Direct measurements of two-beam interference from pairs of spatially-separated luminescence spots in a ring allowed to estimate the length of spatial coherence, and also the value of the amplitude correlator $g^{(1)}(r) \approx 0.2$ at a distance not less than 4 $\mu\text{m}$. This large scale of spatial coherence means that the experimentally observed periodic luminescent structures are described under the conditions of Bose condensation of dipolar excitons in lateral trap by a single wave function. It should be emphasized that large spatial coherence is exhibited both by Bose condensate of exciton polaritons in microcavity heterostructures with several QWs [19] and by a collective state of spatially indirect excitons in structures with double QWs [20].

5. Now let us turn to the study of two-photon correlations of luminescence intensity under the conditions of exciton Bose condensation. We measured a correlator of intensities:

$$g^{(2)}(\tau) = \frac{<I_1(r, t)I_2(r, t + \tau)>}{<I_1(r, t)><I_2(r, t)>}$$

Here angular brackets mean ensemble averaging, $r$ is a spatial coordinate of emitter, and $\tau$ is a delay time between photons in a registered pair.

Measurements of two-photon correlations were carried out with the use of a two-beam interferometer of inten-

![FIG. 2: The phase diagram of the Bose condensation of dipolar excitons in a ring trap in coordinates “above-barrier photoexcitation power $P_{590nm}$ — temperature $T$. Under-barrier photoexcitation is fixed: $P_{582nm} = 10 \ \mu\text{W}$. Squares correspond to the condensation threshold determined by the appearance of a narrow spectral line. The circle at $T = 0.45 \ \text{K}$ shows the threshold estimated from the maximum position of two-photon correlator vs pumping.](image-url)
FIG. 3: Second-order correlator $g^{(2)}(\tau)$ for the luminescence of dipolar excitons in a ring trap as a function of photoexcitation $P$. (a) Time diagrams $g^{(2)}(\tau)$ at power $P_{659\text{nm}} = 0.01$, 0.02, 0.05, 0.1, 0.3, 2, 6 and 30 $\mu$W from bottom to top, respectively. The graphs are shifted vertically for convenience. (b) The value of $g^{(2)}(0)$ vs power. The arrow shows the Bose condensation threshold in accordance with the phase diagram in Fig. 2 $P_{782\text{nm}} = 10$ $\mu$W. $T = 0.45$ K.

FIG. 4: Second-order correlator $g^{(2)}(\tau)$ for the luminescence of dipolar excitons in a ring trap as a function of temperature $T$. (a) Time diagrams $g^{(2)}(\tau)$. The graphs are shifted vertically for convenience. (b) The value of $g^{(2)}(0)$ vs $T$. $P_{659\text{nm}} = 0.1$ $\mu$W, $P_{782\text{nm}} = 10$ $\mu$W.

FIGS. 3 and 4 We will consider measurements at $T = 0.45$ K (Fig. 3) in detail. At pumping rate considerably below condensation threshold the distribution of two-photon correlations is poissonian. In this range of photoexcitation only unstructured spectrum of luminescence as wide as 1.3 meV is observed, that corresponds to localized exciton states. Once again we emphasize that the time resolution of the used registration system does not allow us to detect superposition of chaotic coherent sources (in our case these are the localized exciton states) if their times of coherence are much less than 0.4 ns. When approaching condensation threshold, the effect of photon bunching appears and grows with

...sities according to the classical scheme of R. Hanbury Brown and R. Q. Twiss [4]. High-speed avalanche photodiodes (Perkin-Elmer SPCM-AQR-16) with rise time of $\approx 400$ ps were used as single-photon detectors. They were placed symmetrically with respect to non-polarizing beam-splitting cube which equally divides a luminescence light beam coming from the sample. Special screens and diaphragms provided registration only of the useful signal of luminescence and completely excluded effects of diffused light and difficult to control reflections. Photosresponse signal arrived at the start trigger of the electronic "time-amplitude" converter (ORTEC, TAC 567) from one detector and at the stop trigger – from another. Its output was connected to the input of multichannel analyzer (ORTEC, TRUMP-PCI-8K). The technique allowed to detect the intensity correlator $g^{(2)}(\tau) > 1$ under the conditions of superposition of coherent states with mutual coherence time $\leq 0.5$ ns. Correlation measurements of luminescence intensity were carried out within a narrow spectral band containing the line of exciton condensate. This band was cut out from the luminescence spectrum (presented in Fig. 1) by an interference filter with full spectral bandwidth of $\approx 2$ meV. However, this spectral selection could not get rid of the unstructured luminescence background located directly under the narrow line of exciton condensate.

The results are presented in Figs. 3 and 4. We will consider measurements at $T = 0.45$ K (Fig. 3) in detail. At pumping rate considerably below condensation threshold the distribution of two-photon correlations is poissonian. In this range of photoexcitation only unstructured spectrum of luminescence as wide as 1.3 meV is observed, that corresponds to localized exciton states. Once again we emphasize that the time resolution of the used registration system does not allow us to detect superposition of chaotic coherent sources (in our case these are the localized exciton states) if their times of coherence are much less than 0.4 ns. When approaching condensation threshold, the effect of photon bunching appears and grows with...
further increase of optical pumping. Thus, the function of two-photon correlations shows super-poissonian distribution, \( g^{(2)}(\tau) > 1 \), on coherence time scales of the investigated system \( \tau \lesssim 1 \) ns. The measured value of photon bunching is limited by two factors. The former is due to the fact that beside the luminescence of dipolar excitons, an input of the wide spectral continuum caused by localized states is also recorded. The latter is related to the finite time resolution of the used registration system. At pumping appreciably exceeding the threshold, when the narrow line of exciton condensate dominates in luminescence spectra, the effect of bunching decreases and finally, completely vanishes with further increase of optical excitation. The distribution of two-photon correlations becomes poissonian, thus reflecting, as we assume, a single quantum coherent state of exciton Bose condensate. This conclusion proves to be true by direct observations of a large-scale coherence \( (\text{in situ} \text{ Fourier images}) \) and by a 1\textsuperscript{st}-order correlator \( (\text{two-beam interference from spatially separated spots in a luminescence pattern}) \).

Fig. 5 illustrates the behavior of the interference pattern measured by means of the luminescence amplitude interference from two spatially separated \( (\approx 4 \, \mu \text{m}) \) condensate parts under variation of photoexcitation power. In the pumping range, where two-photon correlator approaches unity, \( g^{(2)}(\tau) \rightarrow 1 \), i.e. where the photon bunching disappears completely, the measured 1\textsuperscript{st} order spatial correlator is rather significant, \( g^{(1)}(r) \approx 0.2 \). Only at very high photoexcitation, almost ten times higher than the largest used in the whole set of data presented in Figs. 1-4, the interference pattern in Fig. 5 starts to wash out. In this high photoexcitation range, the exciton Bose condensate is destroyed due to increasing phase decoherence processes (presumably because of strong dipole-dipole repulsive exciton interaction).

The observed effect of photon bunching proved to be very sensitive to temperature. The measured magnitude of photon bunching decreases several times on temperature increase over the range \( 0.45 \div 4.2 \, \text{K} \) \( (\text{see Fig. 4}) \). This observation may indirectly indicate destruction of an order parameter with temperature. We also emphasize that the regions of maximal photon bunching measured at various temperatures vs photoexcitation power correlate well with the found phase diagram \( (\text{Fig. 2}) \). It means that the maximal two-photon bunching occurs in the area where fluctuations of exciton density are the strongest, i.e. close to the phase boundary. Under the same experimental conditions, no bunching is observed in the spectral range of direct exciton luminescence.

Having assumed that the luminescence of dipolar excitons directly reflects coherent properties of interacting exciton gas, the observed phenomenon of photon bunching nearby condensation threshold, where fluctuations of exciton density and hence luminescence-intensity fluctuations are most significant, manifests phase transition in the interacting exciton Bose gas and it can be used as an independent tool to detect exciton Bose condensation.

The observed effect of photon bunching proved to be very sensitive to temperature. The measured magnitude of photon bunching decreases several times on temperature increase over the range \( 0.45 \div 4.2 \, \text{K} \) \( (\text{see Fig. 4}) \). This observation may indirectly indicate destruction of an order parameter with temperature. We also emphasize that the regions of maximal photon bunching measured at various temperatures vs photoexcitation power correlate well with the found phase diagram \( (\text{Fig. 2}) \). It means that the maximal two-photon bunching occurs in the area where fluctuations of exciton density are the strongest, i.e. close to the phase boundary. Under the same experimental conditions, no bunching is observed in the spectral range of direct exciton luminescence.

Having assumed that the luminescence of dipolar excitons directly reflects coherent properties of interacting exciton gas, the observed phenomenon of photon bunching nearby condensation threshold, where fluctuations of exciton density and hence luminescence-intensity fluctuations are most significant, manifests phase transition in the interacting exciton Bose gas and it can be used as an independent tool to detect exciton Bose condensation.

\[ T. \text{Jeltes, J. M. McNamara, W. Hogervorst et al., \textit{Nature} \textbf{285}, 402 (2007)} \]
\[ A. \text{Öttl, S. Ritter, M. Köhl, and T. Esslinger, \textit{Phys. Rev. Lett.} \textbf{95}, 090404 (2005)} \]
\[ R. \text{Hanbury Brown, R. Q. Twiss, \textit{Nature} \textbf{177}, 27 (1956).} \]
\[ R. \text{J. Glauher, \textit{Phys. Rev. Lett.} \textbf{10}, 84 (1963).} \]
[6] B. Laikhtman, Europhys. Lett. **43**, 53 (1998).
[7] J. Kasprzak, M. Richard, A. Baas et al., Phys. Rev. Lett. **100**, 067402 (2008).
[8] A. P. D. Love, D. N. Krizhanovskii, D. M. Wittaker et al., Phys. Rev. Lett. **101**, 067404 (2008).
[9] A. V. Gorbunov, V. B. Timofeev, JETP Letters **83**, 146 (2006).
[10] A. V. Gorbunov, V. B. Timofeev, JETP Letters **84**, 329 (2006).
[11] V. I. Sugakov, A. A. Chernyuk, JETP Letters **85**, 570 (2007).
[12] L. A. Maksimov, T. V. Khabarova, Doklady Physics **52**, 366 (2007).
[13] V. V. Solov’ev, I. V. Kukushkin, J. Smet et al. JETP Letters **83**, 553 (2006).
[14] V. B. Timofeev, A. V. Gorbunov, J. Appl. Phys. **101**, 081708 (2007).
[15] V. B. Timofeev, A. V. Gorbunov, Phys. Stat. Solidi (c) **5**, 2379 (2008).
[16] C. Schindler, R. Zimmermann, Phys. Rev B **78**, 045313, (2008).
[17] M. Stern, V. Garmider, E. Segre et al., Phys. Rev. Lett. **101**, 257402 (2008).
[18] A. A. Dremin, V. B. Timofeev, A. V. Larionov et al., JETP Letters **76**, 450 (2002).
[19] J. Kasprzak, M. Richard, S. Kundermann et al., Nature **443**, 409 (2006).
[20] S. Yang, A. T. Hammack, M. M. Fogler et al., Phys. Rev. Lett. **97**, 187402 (2006).