Evidence for a Bose-Einstein condensate of excitons

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Abstract – We report compelling evidence for a “gray” condensate of dipolar excitons, electrically polarised in a 25 nm wide GaAs quantum well. The condensate is composed by a macroscopic population of dark excitons coherently coupled to a lower population of bright excitons. To create the exciton condensate we use an all-optical approach in order to produce microscopic traps which confine a dense exciton gas (∼ 10¹⁰ cm⁻²) that yet exhibits an anomalously weak photoemission at sub-kelvin temperatures. This is the first fingerprint for the “gray” condensate. It is then confirmed by the macroscopic spatial coherence and the linear polarization of the weak excitonic photoluminescence emitted from the trap, as theoretically predicted.

The demonstration of Bose-Einstein condensation in atomic gases at micro-kelvin temperatures is a striking landmark [1] while its evidence for semiconductor excitons [2–5] still is a long-awaited milestone. This situation was not foreseen because excitons are light-mass boson-like particles with a condensation expected to occur below a few kelvins [6,7]. An explanation can be found in the underlying fermionic nature of excitons which rules their condensation [8]. Precisely, it was recently predicted that, at accessible experimental conditions, the exciton condensate shall be “gray” with a dominant dark part coherently coupled to a weak bright component through fermion exchanges [9]. This counter-intuitive quantum condensation, since excitons are mostly known for their optical activity, directly follows from the excitons internal structure which has an optically inactive, i.e., dark, ground state [8].

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The Bose-Einstein condensation of semiconductor excitons has received a considerable attention since its theoretical prediction [2–5] in the 1960s. Unlike the most commonly studied bosonic atoms [6], the exciton composite nature plays a key role in their condensation. In semiconductor quantum wells, excitons are made of spin (±1/2) electrons and “spin” (±3/2) holes. These carriers mainly interact through attractive intraband Coulomb processes. Weak interband valence-conduction Coulomb processes also exist, but only for optically active, or “bright”, excitons with total spin (±1) made of (±3/2) holes and (±1/2) electrons. These (repulsive) interband processes bring the energy of bright excitons above the one of “dark” excitons with total spin (±2) made of (±3/2) holes and (±1/2) electrons. As a result, Bose-Einstein condensation of excitons must occur within the low-energy dark states [8,10]. This feature, which makes exciton condensation hard to evidence by conventional optical means, could be the reason why unambiguous signatures of exciton condensates have not been given yet, despite several
decades of active experimental research. By contrast, the excitonic component of a polariton being by construction coupled to light, polariton condensation can be studied through photoluminescence, which recently led to remarkable experiments [11–14].

While in the very dilute regime the exciton condensate must be completely dark, no matter how small the energy difference between dark and bright states is [8], it was recently shown [9] that at sufficiently large density, carrier exchange between bright and dark excitons brings a coherent bright component to the condensate which becomes “gray”. Such a coherence between dark and bright excitons is very similar to what occurs in the well-known phases of superfluid $^3$He (see, e.g., ref. [15]), and in the more recent spinor condensates of ultracold atomic Bose gases [16], where components of these superfluids corresponding to different internal degrees of freedom coexist and are coherent. This coherent coupling allows probing the exciton condensate through the photoluminescence of its bright part. As the bright component is very small, the photoluminescence signal is very weak. Nevertheless, it shall unveil the existence of a dense population of dark excitons, the spatial coherence of the condensate and its internal “spin” structure through the polarisation of the emitted light. In semiconductor quantum wells, the dark nature of exciton Bose-Einstein condensation has been overlooked until very recently [8,10], most probably because the splitting between bright and dark states is small compared to the thermal energy at critical temperature. Here, we present compelling experimental evidence for a “gray” Bose-Einstein condensate of excitons [8,9] through the experimental observation of all its theoretically predicted characteristics.

We study excitons confined in a 25 nm wide GaAs single quantum well embedded in a field-effect device with an electrical polarization, set by the potential $V_g = -4.7$ V applied on a surface electrode, keeping electrons and holes well apart. As the electron and hole wave functions have a small overlap, these dipolar excitons have a long radiative lifetime ($\sim 20$ ns) and a rather large energy splitting between bright and dark states ($\sim 20$ meV, see refs. [17,18]). The electrical polarisation also ensures a repulsive effective exciton-exciton interaction which prevents the formation of biexcitons at the typical density $n_c$ where Bose statistics becomes dominant ($n_c \simeq m_X k_B T/\hbar^2 \sim 10^{10}$ cm$^{-2}$ for 2D excitons with mass $m_X$ at 1 K).

We use a pump laser ($\lambda_{\text{pump}} = 641.5$ nm) with an energy above the AlGaAs barriers of the quantum well in order to create a dense and well-thermalised exciton gas. For such laser excitation photo-injected electrons and holes are captured by the quantum well with different efficiencies [19,20]: a region richer in holes is formed around the laser excitation, itself surrounded by an electron-rich domain resulting from both the photo-current passing through our device and the modulation doping of the structure (see fig. 1(A)). In this landscape, dipolar excitons are created through the Coulomb interaction between photo-injected electrons and holes, the exciton transport being somewhat complicated by the ambipolar diffusion of excess carriers which screen the external field applied through our top gate electrode.

Figures 1(C) and (D) show the photoluminescence emission 10 ns after the pump pulse at 350 mK and 7 K, respectively. Both reveal a pattern characteristic of the charge separation existing in the quantum well, namely a macroscopic exciton ring formed a few tens of microns away from the pump excitation [21,22]. Figures 1(E) and (F) show the exciton confinement potential together with the profile of the exciton density for these measurements. They are both deduced from a weak probe pulse which injects a very dilute exciton cloud after the pump.
pulse. Note that this probe beam is tuned well below the bandgap of the AlGaAs barriers ($\lambda_{\text{probe}} = 790$ nm) in order to bring essentially no perturbation [23]. Excitons injected by the probe beam emit a photoluminescence at an energy $E_X \simeq (\vec{d} \cdot \vec{F}_{\text{screen}} + u_0 a_X)$. The first term corresponds to the energy increase of a single exciton resulting from the screening field $\vec{F}_{\text{screen}}$ induced by excess carriers, $d \approx e \cdot 15$ nm being the excitonic dipole moment [24]. The second term, where $n_X$ is the exciton density and $u_0$ a parameter given by the geometry of our heterostructure [25], corresponds to the energy increase resulting from repulsive exciton-exciton interactions. By sending the probe pulse 100 ns after extinction of the pump, i.e. when all excitons injected by the pump have recombined, we directly map the excitons confinement as for very small exciton densities $E_X$ reduces to $E_X^{(\text{probe})} = \vec{d} \cdot \vec{F}_{\text{screen}}$. We can estimate the density profile of excitons produced by the pump excitation by sending a probe pulse 10 ns after extinction of the pump as $E_X$ reduces then to $E_X^{(\text{pump})} = (E_X^{(\text{probe})} + u_0 a_X)$. This pump-probe spectroscopy then allows us to extract the whole exciton density injected by the pump pulse, $n_X$, including dark and bright excitons, a crucial ingredient to signal a "gray" condensate.

Figures 1(E), (F) show the profile of the exciton confinement through $E_X^{(\text{probe})}$. It displays a maximum at the pump laser spot, then slowly decreases with the distance to the laser excitation before an abrupt decrease just before the region where the ring is formed. This is physically reasonable because the ring is expected to be located at the interface between the electron-rich and hole-rich domains where the near absence of excess carriers leads to a minimum screening. Most importantly, our measurements reveal at 350 mK that an electrostatic trap, i.e. a local minimum of $E_X^{(\text{probe})}$, is spontaneously formed in the vicinity of the ring (see fig. 1(E)). This potential confines dipolar excitons which are high-field seekers, i.e., attracted by strong field regions. We note that the exciton trap is not homogeneous along the circumference of the ring, i.e., it is not identical on opposite sides (see fig. 1(E)). Its depth can be as large as $\simeq 1.2$ meV at 350 mK, an increase of the bath temperature leading to a reduction or a total suppression of the trap (fig. 1(F)).

We evaluate the exciton density by comparing $E_X^{(\text{pump})}$ to $E_X^{(\text{probe})}$. Figure 1(E) shows at 350 mK that across the emission $E_X^{(\text{pump})}$ is blueshifted compared to $E_X^{(\text{probe})}$. This reveals that the pump pulse creates a dense exciton gas ($n_X \sim 10^{10}$ cm$^{-2}$ for a blueshift $(E_X^{(\text{pump})} - E_X^{(\text{probe})}) \sim 1$ meV). More strikingly, we also note that $E_X^{(\text{pump})}$ is almost constant in the region of the ring (fig. 1(E)), unlike at 7K (fig. 1(F)). This reveals that dipolar excitons completely fill the trap spontaneously formed in the vicinity of the ring. The experimental blueshift (1.2 meV around the position $-25$ μm in fig. 1(E)) leads to an exciton density $\sim 2 \times 10^{10}$ cm$^{-2}$ across the entire trap [25-27]. At the same time, the photoluminescence intensity is reduced by more than ten fold between the positions $(-25)$ and $(-31)$ μm.

In the outer rim of the ring, the excitonic population then dominantly consists of optically inactive, i.e. dark, excitons. The dense but nearly dark exciton gas, highlighted by the gray area in fig. 1(E), can only be explained by the formation of a condensate which acts as a sink and captures most of the excitons, this condensate being nearly dark, or "gray". Indeed, in the absence of a condensate, i.e. in the classical regime, the dark and bright populations should be very similar, because the dark-bright energy splitting ($\sim 20$ μeV) is smaller than the thermal energy ($\sim 30$ μeV at our lowest bath temperature). This would lead to a much stronger photoluminescence than the one we observe. Such a conclusion is further supported by the spectral profile of the photoluminescence which is essentially identical on the brightest part of the ring and 6 μm outside [24], fully coherent with the fact.
that the total exciton density stays unchanged throughout this region. Finally, fig. 1(F) shows that at higher temperatures we no longer find the spectral signature of a “gray” condensate. We wish to note that the specific position where the condensate is formed certainly is the result of a complex hydrodynamical diffusion, dipolar excitons experiencing a chute from the pump laser spot to the trapping region, while cooling at the same time.

To unambiguously confirm the formation of a “gray” condensate at sub-kelvin bath temperatures, we further study the first-order spatial coherence \(|g^{(1)}|\) of the photoluminescence [24] since the light emitted by the bright part of the condensate reflects its long-range order. The classical regime is distinguished from the quantum regime through its spatial coherence [28,29], the classical coherence length being of the order of the de Broglie wavelength (\(\lambda_{dB} \sim 100 \text{ nm at sub-kelvin temperatures}\)). We experimentally assess the coherence length of bright excitons by using an actively stabilized Mach-Zehnder interferometer. One arm of the interferometer displaces the photoluminescence laterally by \(\delta_x = 1.5 \mu m\) with respect to the second arm; it also tilts it vertically, so that output interference fringes end up aligned horizontally [24,30]. By scanning the phase of the interferometer, we reconstruct point by point the amplitude of the interference contrast. This allows us to draw the map of the emission first-order spatial coherence from which we deduce [24] the exciton coherence length \(\xi\).

Figure 2 shows \(|g^{(1)}|\) in the ring region. At high temperature (\(T_b = 7 \text{ K in fig. 2(D)}\)), the interference contrast does not significantly vary across the emission, staying approximately equal to 10–15% which is the background value of our interference contrast [24]. This leads to a coherence length \(\xi \lesssim 200 \text{ nm}\). At lower temperature (\(T_b = 370 \text{ mK in fig. 2(C)}\), the interference contrast exhibits a pattern correlated with the spatial profile of the photoemission in a way which again reveals a “gray” condensate. Indeed, fig. 3(A) shows that the interference contrast is minimal (~10%) in the brightest part of the ring, while, in the outer region where the photoluminescence intensity is strongly decreased, \(|g^{(1)}|\) can reach \(\approx 40\%\) which is above half the auto-correlation value (70% for \(\delta_x = 0\)). This shows that the observed bright excitons with a coherence length \(\xi \sim 1.5 \mu m\) one order of magnitude larger than the de Broglie wavelength, belong to the condensate. The variation of the excitonic coherence as a function of the bath temperature moreover shows that non-classical correlations become dominant below a critical temperature \(\sim 2 \text{ K}\), the coherence length abruptly increasing from near classical to non-classical values (see fig. 3(C)).

In order to obtain further insight into the internal structure of a gray condensate, we filter the polarization of the photoluminescence. In a “gray” condensate, dark and bright states are coupled by carrier exchanges [9] or other coupling processes as suggested recently [30–33]. Since the lowest energy for degenerate (+1) or (±2) states is obtained for a linear polarisation, due again to carrier exchanges [8], a “gray” condensate must exhibit a linearly polarized photoluminescence. And indeed, fig. 4(B) shows at 370 mK that the photoluminescence is mostly polarized linearly in the outer rim of the ring where macroscopic coherence is also observed (fig. 3(C)) — the degree of circular polarization being not as significant (see fig. 4(C)). In the inner region of the ring, we also observe a linear polarization, but along the orthogonal direction. These observations contrast with recent studies performed in double quantum well heterostructures where dipolar (spatially indirect) excitons exhibit correlated patterns having both linear and circular polarizations [34]. These patterns have been interpreted in terms of coherent exciton transport and spin-orbit coupling [30,32,33]. Our experiments, which do not reveal these patterns, are performed in a single quantum well with a dark-bright energy splitting much larger than in bilayer heterostructures. Since this splitting plays a key role in selecting the specific condensate which is formed, it is reasonable to think that experiments performed in single and double quantum wells probe different regimes. On the other hand, experiments [35] in a double quantum well, performed essentially at the same time as ours, display, when temperature is lowered below a few
Fig. 4: (Color online) Exciton ring at 370 mK (A). (B), (C): pattern of linear (B) and circular (C) polarizations at 370 mK. In (C), the blue and red colors are associated to $\sigma^+$ and $\sigma^-$ polarized light, respectively. In (B), we see that the photoluminescence is mostly linearly polarized in the outer region of the ring where extended spatial coherence is observed. (D) Degree of linear (red) and circular (blue) polarization of the photoluminescence at 370 mK. The solid black line shows the intensity profile of the emission.

kelvins, a marked darkening of the excitonic fluid, which is fully consistent with our results.

As a final remark, one might wonder if our essentially two-dimensional geometry would not dramatically affect Bose-Einstein condensation. This is not so because condensation occurs in small regions which stabilizes the condensate [1]. In addition, phase fluctuations, responsible for the main qualitative differences between 2D and 3D systems [1], are then quenched. Hence the condensate should be fairly similar to a 3D condensate.

To summarise we have reported the signatures theoretically predicted for a “gray” Bose-Einstein condensate of dipolar excitons. These were highlighted in microscopic and dark regions where at sub-kelvin temperatures excitons obey a quantum statistical distribution, with a dominant (~ 90%) fraction of dark excitons, and exhibit a macroscopically large spatial coherence. In our experiments the gray condensate is created in a complex parameter space, where most notably we tune both the electric polarisation of our field-effect device and the intensity of the optical excitation. Thus we produce microscopic traps, through the screening of the internal field by optically injected excess charges, where the condensate is spontaneously formed.

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REFERENCES

[1] Pitaevskii L. P. and Stringari S., Bose-Einstein Condensation (Oxford University Press) 2003.
[2] Moskalenko S. A., Fiz. Tverd. Tela (Leningrad), 4 (1962) 276.
[3] Blatt J. M. et al., Phys. Rev., 126 (1962) 1691.
[4] Keldysh L. V. and Kopaev YuV., Sov. Phys. Solid State, 6 (1965) 2219.
[5] Keldysh L. V. and Kozlov A. N., Sov. Phys. JETP, 27 (1968) 521.
[6] Griffin A., Sneke D. W. and Stringari S. (Editors), Bose-Einstein Condensation (Cambridge University Press) 1995.
[7] Moskalenko S. A. and Sneke D. W., Bose-Einstein Condensation of Excitons and Biexcitons (Cambridge University Press) 2000.
[8] Combescot M., Betbeder-Matibet O. and Combescot R., Phys. Rev. Lett., 99 (2007) 176403.
[9] Combescot R. and Combescot M., Phys. Rev. Lett., 109 (2012) 026401.
[10] Combescot M. and Leuenberger M. N., Solid State Commun., 149 (2009) 567.
[11] Deng H. et al., Science, 298 (2002) 199.
[12] Kasprzak J. et al., Nature, 443 (2006) 409.
[13] Ballan R. et al., Science, 316 (2007) 1007.
[14] Wertz E. et al., Nat. Phys., 6 (2010) 860864.
[15] Leggett A. J., Quantum Liquids (Oxford University Press) 2006.
[16] Stamper-Kurn D. M. and Ueda M., Rev. Mod. Phys., 85 (2013) 1191.
[17] Blackwood E. et al., Phys. Rev. B, 50 (1994) 14246.
[18] Gorbunov A. V. and Timofeev V. P., Solid State Commun., 157 (2013) 6.
[19] Butov L. V. et al., Phys. Rev. Lett., 92 (2004) 117404.
[20] Rapaport R. et al., Phys. Rev. Lett., 92 (2004) 117405.
[21] Butov L. V. et al., Nature, 418 (2002) 751.
[22] Sneke D. W. et al., Nature, 418 (2002) 754.
[23] Alloing M., Lemaître A., Gallopin E. and Dubin F., Sci. Rep., 3 (2013) 1578.
[24] Alloing M. et al., arXiv:1304.4101; Alloing M. et al., arXiv:1210.3176.
[25] Ivanov A. L., Muljarov E. A., Mouchliadis L. and Zimmermann R., Phys. Rev. Lett., 104 (2010) 179701.
[26] Schindler C. and Zimmermann R., Phys. Rev. B, 78 (2008) 045313.
[27] Laikhtman B. and Rapaport R., Phys. Rev. B, 80 (2009) 195313.
[28] Naraschewski M. and Glauber R. J., Phys. Rev. A, 59 (1999) 4595.
[29] Bloch I., Haensch T. W. and Esslinger T., Nature, 403 (2000) 166.
[30] High A. A. et al., Nature, 483 (2012) 584.
[31] Ali Can M. and Hakioglu T., Phys. Rev. Lett., 103 (2009) 086404.
[32] Matuszewski M. et al., Phys. Rev. B, 86 (2012) 115321.
[33] Vishnevsky D. V. et al., Phys. Rev. Lett., 110 (2013) 246404.
[34] High A. A. et al., Phys. Rev. Lett., 110 (2013) 246403.
[35] Shilo Y. et al., Nat. Commun., 4 (2013) 2235.