Final Report, DOE Grant DE-FG02-99ER45780
"Indirect Excitons in Coupled Quantum Wells"
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University of Pittsburgh
PI: David W. Snoke

Abstract

The is the final technical report for this project, which was funded by the DOE from 1999 to 2012. The project focused on experimental studies of spatially indirect excitons in coupled quantum wells, with the aim of understanding the quantum physics of these particles, including such effects as pattern formation due to electron-hole charge separation, the Mott plasma-insulator transition, luminescence up-conversion through field-assisted tunneling, luminescence line shifts due to many-body renormalization and magnetic field effects on tunneling, and proposed effects such as Bose-Einstein condensation of indirect excitons and phase separation of bright and dark indirect excitons. Significant results are summarized here and the relation to other work is discussed.

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Introduction

This project supported 1 graduate student per year for several years, and 2 students for the last few years, with no funds for equipment, and with a small additional amount for supplies, travel, and other ancillary research expenses. The experiments supported involved designing and fabricating GaAs/AlAs semiconductor structures, cooling them to cryogenic temperatures, and doing time-, space- and energy-resolved optical measurements, as various experimental conditions were varied, including stress, electric field, magnetic field, pump intensity and temperature. Theory collaboration over the years included theorists Oleg Berman, Monique Combescot, Jonathan Keeling, Boris Laikhtman, Peter Littlewood, Yurii Lozovik, Steve Simon, Marzena Szymanska, and Roland Zimmermann. Experimental collaborators included Loren Pfeiffer and Ken West at Lucent (now at Princeton University), Ronen Rapaport of Lucent (now at the Technion University, Israel), and Angelo Mascarenhas and Brian Fluegel at NREL.

One of the main motivations for the work was the goal of observing a convincing demonstration of Bose-Einstein condensation (BEC). The system of indirect excitons in coupled quantum wells seems ideal for this purpose, because the lifetime of the particles can be made very long, tens of microseconds in our experiments, and a competing phase, electron-hole liquid, is expected to be forbidden in this system. However, the indirect exciton system also brings instrinsic complications, in particular a very strong, long-range particle-particle interaction (since the indirect excitons are aligned electric dipoles), which makes weakly interacting Bose gas theory inapplicable, and in addition leads to strong renormalization effects. At high density the Mott transition prevents Bose-Einstein condensation, and at low density the particles are likely to be trapped in local disorder minima.

Despite these complications, we were able to establish several clear results over the years with fascinating effects. Our work also allowed us to have the expertise and resources to evaluate related work by other groups.

Significant Results

One of the first significant results was the observation of a fascinating pattern formation effect in which rings of luminscence formed around the laser excitation spot, in some cases at quite far distances, up to mm [1]. This effect was claimed by Butov and coworkers as the result of spontaneous coherence, i.e., BEC [2], but our work [3,4] showed that the ring formation is due to charge separation of electrons and holes, due to the different rates of hopping over the barriers of the wells. Butov and coworkers eventually agreed that the ring formation involved charge separation, but argued that BEC effects were crucial [5]. Our work, and later theoretical work by the group of Alex Ivanov [6], showed, however, that the formation of the ring and the fracturing of the ring into tiny “beads” can be understood entirely in terms of classical physics. For further discussion of this ring effect and the claims to exciton BEC, see the section on the relation of this project to other work, which follows.

Later work in collaboration with NREL, led by Brian Fluegel, observed luminescence rings also in high-lying states [7]. This was shown to be due to electric-field-assisted tunneling into higher-energy states.

Another significant early result was the observation of a huge sensitivity of the exciton line energy to weak magnetic field [8]. In collaboration with theoris Boris Laikhtman, we showed that this effect is due to field-dependent tunneling rates. This effect may have application in the optical detection of magnetic field.

A series three papers in Physical Review Letters [9-11], led by graduate student Zoltan Vörös, demonstrated by very careful studies that the excitons can reach true spatial and thermal equilibrium in a harmonic trap which we created using stress. Accurate measurements were made of the diffusion constant of the excitons and their interaction strength, parameters which are essential for proper theory of the system. Our experiments made possible a test of the interaction theory which is independent of a calibration of the density, which is notoriously hard to determine with accuracy in this system.

Experimental collaboration with Romen Rapaport and other colleagues at Bell labs led to the development of electric field methods for trapping [12], still used by Rapaport's group and others today.

The final significant result funded by this project was the demonstration and understanding of another fascinating pattern-formation effect, unrelated to the charge-separation effect leading to luminescence rings, discussed above. In this effect, a dark spot appears in the center of the trapped luminescence cloud. Early theory claims by Combescot [13] claimed that this was due to an excitonic condensate of "dark" excitons (excitons in which symmetry forbids radiative recombination). Our work first established that this is not due to charge separation or nonradiative recombination or other more mundane effects, and that there truly is a "dark" population of excitons. We also showed that a simple theory of dark BEC would not work, although we could not rule out that BEC of a dark phase was occurring. In fact, recent followup work by graduate students Nick Sinclair [14] and Jeff Wuenschell gives strong evidence that many-body effects play a significant role.

Overall, this project produced 28 publications, including articles in Nature and Nature Photonics, four Physical Review Letters and two other publications in letters journals, six articles in Physical Review B, six articles in Solid State Communications, a popular press article proposing the idea of excitonic circuits, a book chapter, and six other publications. The work was presented at international conferences too numerous to count, about ten talks per year. The total publication list is presented as an Appendix to this report.

Relation to Other Work

The work in this project in many ways played the role of "fact checker" on high-profile claims by other groups of BEC. While we believe that BEC is allowed in this system, we felt that many claims needed to be stated more carefully. To this day, it is our view that none of the claims to BEC in this system by other groups have been verified adequately.

Much of the history and theory of these various claims has been reviewed in a recent book chapter. Here we summarize some of what is known and what is not known about this system.

• An early claim by Butov and coworkers [15] that evidence for fast diffusion was evidence of BEC has not been substantiated in subsequent studies which directly imaged the diffusion indirect excitons [9]. The important role of the pressure of the excitons on each other due to their strong repulsive interactions was not fully appreciated in the early work by Butov and coworkers.

• An early claim by Butov and coworkers [16] that strong fluctuations of the luminesence were evidence of BEC is now known to be a result of to the exciton Mott transition to conducting plasma at high density, which strongly affects the local electric field felt by the excitons [17]. No claim to strong fluctuations at the BEC point has been made in recent claims of exciton BEC.

• Butov and coworkers [2] originally claimed that the luminescence ring formation and/or the breaking of this ring into beads is a result of "coherence," i.e., BEC. The ring formation including the existence of periodic "beads" is now known to be explained adequately entirely by the electric field effects in a system with two species of charge, namely electrons and holes [3,4,6,18,19]. Butov and coworkers now do not claim that the ring-and-bead effect is evidence of BEC, but claim that it provides the context for BEC at low temperature.

• Recent claims have been made by Butov and coworkers [20,21] and Dubin and coworkers [22] that measurement of coherence of the light emitted by the beads in the ring are evidence of BEC. While these claims have largely been made after our project ended, so that we have not been able to analyze them in detail, we can make several observations. First, the initial claim of coherence by Butov and coworkers [20] was accompanied by a line spectrum with a width of 2 meV, corresponding to a coherence time of 0.3 ps, which is not strongly different from what is expected in a low-temperature exciton gas. Second, later claims do not fully distinguish between the coherence due to many-body effects, i.e. BEC, and the coherence expected normally from ballistic motion of all carriers. The strong density-dependent interaction of the excitons launches them at high velocity from the "beads." At very low temperature, the excitons experience very little phonon scattering, and so travel ballistically for distances of a micron or so. Ballistic motion is also coherent single-particle motion. The observed coherence lengths of the order of a micron are not far from what is expected in low-temperature exciton gases.

• There is presently ambiguity in the theoretical interpretation of the results of coherence luminescence emission. Theorist Monique Combescot has argued [13] that the condensate should always be in a "dark" state, i.e., a non-light-emitting state. In this case, there should be no coherence of the light emission. The above-described experiments, however, claim coherence of the emission as evidence of BEC. Combescot has recently modified her theory to allow for a "gray" condensate [23], in other words, to suggest that at high exciton density, state mixing can occur to make the dark states have a bright component. However, the theory indicated that the density at which this occurs may be above the Mott transtion density.

Conclusions

It is our opinion that this project was prematurely ended, because the field continues to have claims of BEC which are not being adequately fact checked. The project was hampered by the lack of major equipment funds, for example to obtain a dilution refrigerator or a cryostat with strong magnetic field. Nevertheless, important progress was made on understanding the basic physics of this system, and novel effects were seen such as nonequilibrium pattern formation, strong magnetoresistance, and luminescence up-conversion.

The overall picture which emerges is that the interactions between the excitons are so strong that they dominate the physics. This means, for example, that small traps will not be able to contain a condensate, because the condensate will renormalize the potential energy, washing out the trapping potential. Thus, for example, the "beads" seen in the luminescence rings of Butov and coworkers are not trapped condensate, but "anti-traps," in which excitons are ejected at high velocity. This makes the theoretical analysis of the system quite difficult.

The most likely scenario for BEC in this system is a trapped system at very high density and low temperature, in which case the system will more resemble a BCS coherent state, which would also be of great interest. The strong interactions likely deplete the condensate strongly, as in the case of liquid helium. Therefore, the best tests of BEC may not be from luminescence spectroscopy, but from transport studies, similar to those used by Eisenstein [24], Shayegan [25], and coworkers for bilayer condensates in a different system. That work has had wide confirmation and has been studied with theoretical rigor.

Appendix
Publications recognizing funding from this project

• D.W. Snoke, "Dipole excitons in coupled quantum wells: toward an equilibrium exciton condensate," in *Quantum Gases: Finite Temperature and Non-Equilibrium Dynamics* (Vol. 1, Cold Atoms Series), N.P. Proukakis, S.A. Gardiner, M.J. Davis, and M.H. Szymanska, eds. (Imperial College Press, London, 2013).

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• B. Fluegel, K. Alberi, L. Bhusal, A. Mascarenhas, D.W. Snoke, G. Karunasiri, L.

N. Pfeiffer, and K. West, “Exciton pattern generation in GaAs/AlGaAs multiple quantum wells,” Physical Review B **83**, 195320 (2011).

• Z. Vörös, D.W. Snoke, L. Pfeiffer, and K. West, “Direct measurement of exciton-exciton interaction energy,” Physical Review Letters **103**, 016403 (2009) (cover article).

• D.W. Snoke, “Predicting the Ionization Threshold for Carriers in Excited Semiconductors,” Solid State Communications **146**, 73 (2008).

• Z. Vörös and D.W. Snoke, “Quantum Well Excitons at Low Density,” Modern Physics Letters B **22**, 701 (2008).

• B. Fluegel, A. Mascarenhas, D. W. Snoke, L. N. Pfeiffer, and K. West, “Plasmonic All-Optical Tunable Wavelength Shifter,” Nature Photonics **1**, 701 (2007).

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• David W. Snoke, “Excitonic Circuits: New Tools for Manipulating Photons,” Photonics Spectra **40**, 108 (January 2006).

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• G. Chen, R. Rapaport, L.N. Pfeiffer, K. West, P.M. Platzman, S. Simon, Z. Vörös, and D. Snoke, “Artificial Trapping of a Stable High-Density Dipolar Exciton Fluid,” Physical Review B **74**, 045309 (2006).

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• Z. Vörös, R. Balili, D.W. Snoke, L. Pfeiffer, and K. West, “Long-Distance Diffusion of Excitons in Double Quantum Well Structures,” Physical Review Letters **94**, 226401 (2005).

• S. Denev, S. H. Simon, and D.W. Snoke, “Luminescence Ring Formation in Quantum Wells: a Model with Coulomb Interaction,” Solid State Communications **134**, 59 (2005).

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• D.W. Snoke, Y. Liu, Z. Vörös, L. Pfeiffer, and K. West, "Trapping Long Lifetime Excitons in a Two-Dimensional Harmonic Potential," Solid State Communications **134**, 37 (2005).

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