Bose–Einstein condensation of excitons in bilayer electron systems

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An exciton is the particle-like entity that forms when an electron is bound to a positively charged 'hole'. An ordered electronic state in which excitons condense into a single quantum state was proposed as a theoretical possibility many years ago. We review recent studies of semiconductor bilayer systems that provide clear evidence for this phenomenon and explain why exciton condensation in the quantum Hall regime, where these experiments were performed, is as likely to occur in electron–electron bilayers as in electron–hole bilayers. In current quantum Hall excitonic condensates, disorder induces mobile vortices that flow in response to a supercurrent and limit the extremely large bilayer counterflow conductivity.

In many-particle quantum physics bosons are special. Unlike the ubiquitous electrons and their fermionic cousins, any number of bosons can crowd into the same microscopic state. Indeed, Einstein predicted in 1924 that at low temperatures essentially all of the bosons in a macroscopic system would spontaneously 'condense' into the same low-energy quantum state.

In quantum mechanics, an individual particle is represented by a wave that has both amplitude and phase. The most remarkable consequence of Bose–Einstein condensation (BEC) of a vast number of particles is that macroscopic properties become dependent on a single wavefunction, promoting quantum physics to classical time- and length scales. A BEC is a highly ordered state in which the wavefunction phase is coherent over distances much longer than the separation between individual particles. Long-range quantum phase coherence has many dramatic physical consequences, of which the most spectacular is superfluidity, that is, the ability of matter to flow around obstacles with extremely weak, often immeasurable, dissipation.

Buckets of bosons

All known BECs are formed by particles that are actually composite bosons made up of an even number of fermions. Helium-4 atoms, for example, contain two protons, two neutrons and two electrons, and Bose condense to form a superfluid at just a few degrees above absolute zero. Today there is intense interest in BECs in rarefied atomic vapours. In superconductors electrons form pairs at low temperatures. These 'Cooper pairs' are again composite bosons. In this case, the formation of pairs and their collective Bose condensation usually occur at the same low temperature; the transition temperature is controlled by the underlying fermionic physics, not purely by bosonic quantum mechanics. Correspondingly, the average separation between electrons in a given Cooper pair usually greatly exceeds the mean distance between pairs.

An exciton in a semiconductor consists of an electron bound to a hole. In most cases the electron is in the conduction band of the semiconductor, and the hole, which is nothing more than an unfilled electronic state masquerading as a positively charged object, is in the valence band. Excitons are usually created by shining light on the semiconductor, which creates electrons and holes in equal numbers. Optically generated excitons like these are ephemeral objects, decaying quickly via the emission of light.

Electrons and holes are both fermions, but excitons are bosons. After the 1957 pairing theory of superconductivity was solidly established, physicists began to speculate[1] about BEC of excitons in semiconductors. The short lifetime of optically generated excitons has been a major obstacle to realizing this phenomenon[2,3]. Indirect excitons in bilayer quantum well systems have two key advantages over bulk systems and have thus played an increasingly important role in this field[4,5]. Quantum wells are realized in layered semiconductor structures and allow for the confinement of electrons and holes to two-dimensional (2D) planes. Indirect excitons are bound states of conduction-band electrons in one well and valence-band holes in an adjacent well. The separation between electrons and holes reduces the rate at which they recombine into photons. In addition, the spatial separation between electrons and holes causes the excitons to act like oriented electric dipoles that have repulsive interactions. The repulsion is important because it prevents the electrons and holes from agglomerating into an uninteresting electron–hole plasma. Experimental studies of optically generated indirect excitons have uncovered a number of fascinating many-body effects[6,7]. The enhanced exciton mobility, increased radiative decay rate, and photoluminescence noise that has been reported in these systems at low temperatures and high fields, all suggest collective, possibly coherent, behaviour. Still, the requirements of high density and low temperatures are difficult to achieve in this non-equilibrium system, and there is not yet compelling evidence for an exciton BEC.

Exciton condensation is closely analogous to other common electronic orders in which electron–hole pairs condense. For example, charge-density-wave states are formed by condensation of electron–hole pairs located at different points in a crystal's Brillouin zone and ferromagnetism can be regarded as being due to condensation of pairs formed from electrons and holes with different spin indices. The analogies are never complete, however—the broken translational symmetry in charge-density-wave states introduces a number of qualitative distinctions, for example. Ferromagnetism in systems with nearly uniaxial easy-plane anisotropy is perhaps most closely analogous to exciton condensation. Indeed, it has been argued that some of the phenomena that have been studied in the bilayer electron systems of interest here, including collective condensate transport, can also occur[8] in thin-film metallic ferromagnets.

Excitons in electron–electron bilayers

Here we describe compelling evidence that exciton condensation has recently been discovered, not in electron–hole systems as expected, but in systems with two parallel layers of conduction-band electrons like those illustrated schematically in Fig. 1. Importantly, the excitons in this system are present in equilibrium, waiting patiently for experimenters to reveal their properties.

The techniques required to grow high-quality single and bilayer electron systems are now well established and have been vital in a great many important physics discoveries, notably the famous fractional quantum Hall effect[9]. At first sight it seems impossible to achieve BEC of excitons in electron–electron bilayers, because all the fermions have repulsive interactions. Quantum well electrons are, however, able to perform remarkable tricks when placed in a strong perpendicular magnetic field.

The Lorentz force bends classical electron trajectories into circles—cyclotron orbits. In a 2D quantum system the kinetic energy of these orbits is quantized into discrete units. The set of orbits with a particular energy, a ‘Landau level’, is, however, highly degenerate because the tiny cyclotron orbits can be positioned all across the 2D plane. It turns out that the number of degenerate states in the lowest-energy Landau level is equal to the number of quanta of magnetic flux that pass through the electron layer. At high magnetic field it is easy to enter a regime in which the total number of electrons is less than the number of states in the lowest Landau level. In Fig. 1, for example, we illustrate the circumstance in which the number of electrons in each layer is one-third of the number of available states; this is referred to as filling factor $\nu = 1/3$.

BEC at strong magnetic fields in electron–electron bilayers is most easily understood by making a particle–hole transformation in one of the two layers; we have chosen the bottom layer for this in Fig. 1. The particle–hole transformation[10,11] is one in which we simply keep track of the empty states in the Landau level rather than the full ones. It is mathematically exact, changes the filling factor of the transformed layer from ν to $1 - \nu$, and changes the sign of the carrier charge from negative to positive. The interaction of holes with electrons in the untransformed layer is thus attractive. Particle–hole transformations also change the sign of the kinetic energy, from negative to positive for valence-band holes at zero magnetic field for example. Because the kinetic energy is the same for all electrons and holes in the lowest Landau level, BEC at strong fields is just as likely to occur in electron–electron bilayers as in electron–hole bilayers.

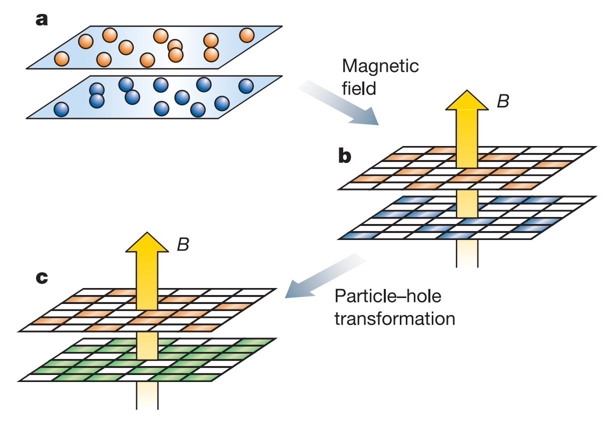

Figure 1 An electron–electron bilayer system in a strong magnetic field is equivalent to an electron–hole bilayer. **a**, Cartoon depiction of two parallel layers of electrons. **b**, In a magnetic field the kinetic energy of 2D electrons is quantized into discrete Landau energy levels. Each such Landau level contains a huge number of degenerate orbitals, here depicted schematically as a checkerboard of sites. If the field is strong enough, all electrons reside in the lowest Landau level, and only occupy a fraction (here one-third) of the available sites. **c**, A particle–hole transformation applied to the lower electron layer places the emphasis on the unoccupied sites—that is, the holes (coloured green) in that layer. This transformation, which is formally exact and completely equivalent to the more familiar transformation used to describe empty valence-band states in a semiconductor as holes, changes the sign of the Coulomb interactions between layers from repulsive to attractive. Exciton BEC occurs when holes in the lower layer bind to electrons in the upper layer. This is most likely to occur when the number of electrons and holes are equal, that is, when each layer is half-filled (this is not the case in this figure).

This last point is a subtle one; indeed, spontaneous coherence[12] and superfluidity[13] were predicted in electron–electron bilayers without explicitly recognizing their equivalence to earlier predictions[14] for electron–hole bilayers. We emphasize, however, that particle–hole transformation of a conduction-band Landau level is completely equivalent to the much more familiar transformation used to map unoccupied valence-band electron states into holes.

In Fig. 1 the number of holes in the bottom layer exceeds the number of electrons in the top layer. To create an exciton BEC, these populations should be nearly equal; one should therefore begin with half-filled Landau levels in each layer. Early experiments proved that in this 1/2 + 1/2 situation a bilayer electron system can exhibit a quantized Hall effect[15]. In other words, its Hall resistance, measured with electrical currents flowing in parallel through the two layers, is precisely equal to h/e^2, Planck’s constant divided by the square of the electron charge. This remarkable effect results from a complex interplay of Landau quantization, Coulomb interaction effects, and imperfections in the 2D plane. Although observation of a quantized Hall effect in the bilayer system at this filling factor demonstrates the importance of interlayer Coulomb interactions, it does not on its own suggest the existence of an exciton BEC. The hunt for BEC requires different experimental tools.

Experimental evidence for exciton formation

In a bilayer 2D electron system the two quantum wells are separated by a thin barrier layer. By adjusting the thickness and composition of this barrier, it is possible for electrons in one layer to interact strongly, via the Coulomb interaction, with the electrons in the

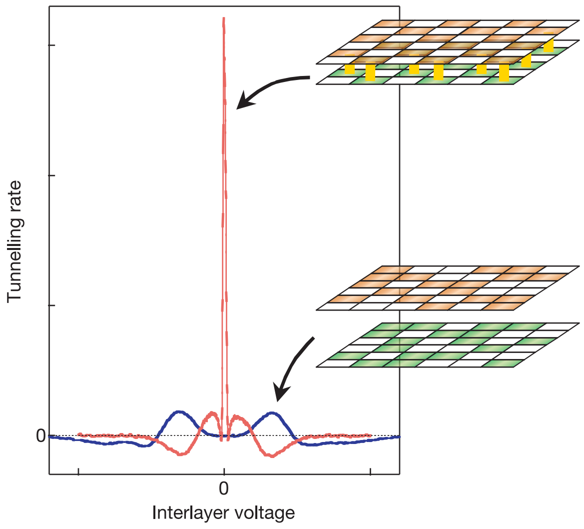

Figure 2 Tunnelling rate versus interlayer voltage in a bilayer electron system. These traces are actual data, and are taken at magnetic fields where exciton condensation is most expected (that is, one half-filling of the lowest Landau level per layer). In the blue trace the layers are relatively far apart, whereas in the red trace they are closer together. The dramatic difference between them is a direct indication that a phase transition in the bilayer system occurs when the layer separation is reduced below a critical value. The huge enhancement of the tunnelling rate at zero energy in the red trace points to interlayer electron–hole correlations: that is, it suggests that every electron is positioned opposite a hole into which it can easily tunnel.

other layer, and yet have an extremely small probability of quantum mechanically tunnelling through the barrier. Achieving this strong-correlation, weak-tunnelling limit is key to the occurrence of excitonic Bose condensation in the bilayer 2D electron system, as we discuss further below.

Figure 2 shows measurements[16] of the rate at which electrons tunnel between the two layers of a bilayer 2D electron system as a function of voltage difference. These data were taken at very low temperatures and with the magnetic field adjusted to produce the half-filling per layer condition. The two traces in the figure differ only in the effective interlayer separation. For the blue trace, the layers are relatively far apart. For voltages near zero, the tunnelling rate is very small. This suppression of the tunnelling is not sensitive to small changes in the magnetic field and grows more severe as the temperature is reduced. Often referred to as a Coulomb gap, the effect is a consequence of strong correlations between electrons—within the individual layers. Tunnelling at low voltages (or, equivalently, low energies) is suppressed because an electron attempting to enter a 2D electron layer at high magnetic field blunders disruptively into the delicate dance being performed by the electrons in that layer as they skilfully avoid one another. This clumsy entry can only produce a highly excited, non-equilibrium state and can occur only at high voltage. Although quite interesting in its own right, the effect obviously does not suggest exciton condensation.

The red trace is Fig. 2 is dramatically different. A slight reduction in the effective layer separation has produced a giant peak in the tunnelling rate. In sharp contrast to the suppression effect in the blue trace, this peak grows rapidly in height as the temperature is reduced and is very sensitive to magnetic field. Small changes in the field away from the value needed to produce a half-filled lowest Landau level rapidly destroy the peak.

The stark difference between the two traces in Fig. 2 suggests that a quantum phase transition occurs as a function of effective layer separation. The strong peak in the tunnelling rate seen at small separations is inconsistent with the arguments used to understand the suppression effect at larger separations. Instead of being unaware of the correlations in the layer about to be entered, the tunnelling electron is apparently already participating in the dance. Indeed, the peak suggests that all electrons are strongly correlated with their neighbours in both layers. Moreover, the temperature and magnetic-field dependence of the peak point to the existence of a new and intrinsically bilayer collective state in which electrons in one layer are always positioned opposite holes in the other layer.

Strong electron–hole correlations are necessary but not sufficient for exciton condensation. To make the argument for excitonic BEC in a bilayer 2D electron system more compelling, an experiment demonstrating the transport of electron–hole pairs is needed. But how does one move and detect neutral objects? The key is to note that the uniform flow of excitons is equivalent to ordinary electrical currents flowing in opposite directions in the two layers. Technical tricks have allowed us to make independent electrical connections to the individual layers in bilayer electron samples[17]. With these contacts it is easy to arrange for equal and opposite currents to flow in the two layers and directly test whether or not excitons are available to perform the particle transport.

Figure 3 shows a cartoon version of what is expected from such a counterflow measurement. The two traces represent the Hall voltages expected in the two layers, neglecting all quantum effects except exciton condensation. Because of the Lorentz force on a moving charge, the Hall voltage is, in most cases, simply proportional to the magnetic field. In a bilayer system with counterflowing currents, the Hall voltages in the two layers will be of opposite sign.

If the distance between the two layers is too large, or if the magnetic field is far from the half-filling per layer condition, exciton condensation will not occur. If, however, the layers are closely spaced and the magnetic field is just right, interlayer electron–hole pairs will form and carry the counterflow current. The Hall voltage in each layer should then drop to zero, as suggested in the figure. This prediction can be understood in a variety of ways, most simply by observing that excitons are neutral and thus feel no Lorentz force. Without the Lorentz force the Hall voltage must vanish. This remarkable prediction has recently been confirmed by our own group at Caltech[18] and the effect has been reproduced, in a slightly different bilayer system, by researchers at Princeton[19]. The same experiments do, however, show that the electron–hole transport current flows with a weak but measurable dissipation. The activated temperature dependence of the dissipation is inconsistent with independent exciton transport, because the exciton diffusion constant should then approach a constant as the temperature goes

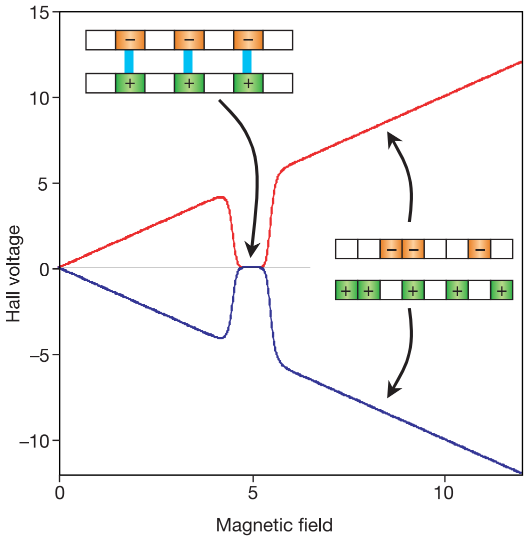

Figure 3 Hall voltage measurements reveal exciton condensation. The two traces schematically indicate the Hall voltages in the two electron layers when the electrical currents flowing in them are oppositely directed. All quantum effects, except exciton condensation, have been ignored. When the currents are carried by independent charged particles in the two layers, non-zero Hall voltages must be present to counteract the Lorentz forces. Because the currents are oppositely directed, these voltages have opposite signs in the two layers. However, if exciton condensation occurs at some magnetic field, the oppositely directed currents in the two layers are carried by a uniform flow of excitons in one direction. Being charge-neutral, these excitons experience no Lorentz force and the Hall voltage is expected to vanish. This remarkable effect has very recently been definitively observed.

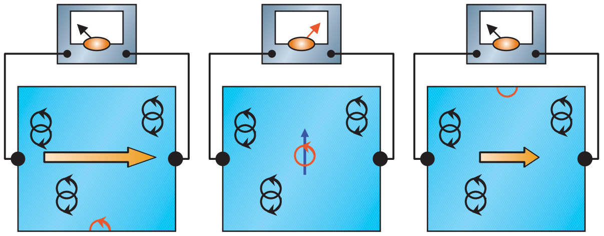

Figure 4 Motion of unpaired vortices leads to dissipation in excitonic superfluids. The elementary excitations in quantum Hall excitonic superfluids are vortices that carry electrical charge and topological charge. In an ideal system at low temperatures vortices occur in bound pairs and do not contribute to the decay of excitonic supercurrents. Disorder, however, is present in all real samples and is capable of producing unpaired vortices. The weak dissipation observed in recent counterflow experiments is associated with the activated transport of such unpaired vortices.

to zero. Instead, these new results can only be explained in the context of collective exciton-condensate transport.

Exciton superfluidity

Quantum mechanics is invariant under a global change in wavefunction phase, and the spatial phase variation rate determines velocity. In an excitonic BEC the same simple description holds. The velocity of a gas of electron–hole pairs is proportional to the spatial gradient of the condensate phase, and excitonic superfluidity requires the absence of microscopic processes that favour a particular value of the condensate phase. The presence of these processes would invalidate the analogy between the quantum mechanics of the condensate and that of a simple quantum particle. Because any process that transfers electrons between the two bands that form the exciton will be sensitive to the condensate phase, and such processes are not forbidden by symmetry, ideal excitonic superfluidity is—strictly speaking—an impossibility[20]. The dominant condensate-phase-sensitive process in bilayers is interlayer tunnelling, which either creates or destroys an exciton. It is ironic that it was the measurements[16] of this weak process, discussed above, that provided the first compelling evidence for exciton condensation. Experimentally, these processes are expected[21,22] to lead to nonlinearities in the counterflow current–voltage relationship. In our opinion, a full description of the influence of interlayer tunnelling on counterflow transport raises theoretical questions that are fundamentally new and not well understood. The absence of nonlinearities in current experiments suggests that this physics has been masked by dissipation due to the motion of vortices that are nucleated in great numbers in these quantum Hall samples by disorder. We expect that the influence of tunnelling on counterflow transport will become apparent in samples with stronger interlayer tunnelling amplitudes than those studied until now, and in current samples at sufficiently low temperatures.

Because the condensate wavefunction is collective, elementary quantum processes which act on one particle at time cannot effectively reduce the velocity of a BEC. In 2D superfluids, condensate current decay is limited instead by the nucleation and transport of vortices: points around which the condensate phase changes by 2π. The elementary process which leads to phase gradient decay is one in which a vortex moves across the system, as illustrated in Fig. 4. The energy required to create an isolated vortex in the ground state of a ideal 2D BEC is (logarithmically) infinite. At temperatures below the Kosterlitz–Thouless transition temperature, at which the free energy of an isolated vortex vanishes, no unpaired vortices should be present.

Vortices are especially important in bilayer exciton condensates because they tend to be nucleated by disorder. The special role of vortices in these superfluids can be understood by thinking about the Aharonov–Bohm phases associated with the external magnetic field. As explained above, the number of states N in a Landau level is proportional to the number of quanta of magnetic flux that penetrate the 2D layers. Correspondingly, the total phase change of an electron orbital that encircles the system is $2\pi N$. A vortex in the condensate phase therefore either increases or decreases the number of states that can be accommodated in one of the layers by a single unit. Vortices consequently come in four flavours[23] and carry charge equal in magnitude to one-half of the electron charge. In bilayer exciton condensates vortices couple to, and are therefore nucleated by, external disorder potentials. The experimental results discussed above demonstrate that vortex motion in the present excitonic BECs requires only a finite activation energy. In all current samples, free vortices are present down to the lowest temperatures studied.

The road from the early suggestions that there might be an excitonic analogue of superconductivity to these recent discoveries has been marked by unanticipated twists and turns. Excitonic BEC requires strong inter-band interactions, but a very small amplitude for transitions between bands that are due to either one-body potentials or interactions. The possibility of studying the bilayer quantum well systems that satisfy this requirement so beautifully had to wait for the development of the techniques we now have for growing extremely high quality layered semiconductors. The interaction energy gained by excitonic condensation must compete with band energies and energy gains associated with other competing types of order. The understanding that Landau quantization in a strong field could eliminate the band energy cost (even in electron–electron bilayers), and that excitonic condensation could prevail over other orders under some circumstances, grew out of the theory of the fractional quantum Hall effect—something that had not been anticipated at the beginning of this journey. Finally, the question of how collective transport of neutral objects could be detected in an excitonic BEC has always been a thorny issue. The separate contacting techniques that enable counterflow transport measurements, developed with quite different goals in mind, provide a direct solution to this problem. In science as in life, way leads onto way, and destinations are often reached by unanticipated paths. ☐

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