Excitonic superfluid phase in double bilayer graphene

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A spatially indirect exciton is created when an electron and a hole, confined to separate layers of a double quantum well system, bind to form a composite boson12. Such excitons are long-lived, and in the limit of strong interactions are predicted to undergo a Bose–Einstein condensate–like phase transition into a superfluid ground state13. Here, we report evidence of an exciton condensate in the quantum Hall effect regime of double-layer structures of bilayer graphene. Interlayer correlation is identified by quantized Hall drag at matched layer densities, and the dissipationless nature of the phase is confirmed in the counterflow geometry44–5. A selection rule for the condensate phase is observed involving both the orbital and valley indices of bilayer graphene. Our results establish double bilayer graphene as an ideal system for studying the rich phase diagram of strongly interacting bosonic particles in the solid state.

In bulk semiconductors, an optically excited electron–hole pair interacts through Coulomb attraction to form a bound quasi-particle, referred to as a spatially direct exciton (Fig. 1a). Such excitons are easily generated but recombine on the nanosecond timescale. By confining the electrons and holes to separate, but closely spaced, two-dimensional (2D) quantum wells, strong attraction is maintained but recombination is blocked, leading to long-lived excitons. These so-called spatially indirect excitons are predicted to exhibit a rich phase diagram of correlated behaviours, including a type of superfluid BEC ground state, at temperatures predicted to exhibit a rich phase diagram of correlated behaviours, including a type of superfluid BEC ground state, at temperatures much higher than for similar phenomena in atomic gases14–16.

Reducing the interlayer Coulomb interaction, e2/εd (and therefore the exciton binding energy), whereas reducing the magnetic length increases the intralayer Coulomb energy, e2/εℓB (increasing interaction energy between the excitons). Here ℏ is the reduced Planck constant, e is the elementary charge and ε is the dielectric constant. For GaAs double layers, a minimum interlayer separation of d ≈ 20 nm is required to prevent interlayer tunnelling and maintain sufficiently high mobility, placing a stringent limit on the achievable d/ℓB. Nonetheless, the EC phase in electron-doped GaAs layers is observed to emerge for d/ℓB ≈ 2, with a characteristic energy scale of 800 mK (ref. 15).

Graphene double layers possess several advantages for realizing the EC phase, including wide tunability of carrier density across electrons and holes by field effect gating, single atomic layer thickness allowing interlayer spacing down to a few nanometres without significant tunnelling17, and the possibility of the EC phase transition exceeding cryogenic temperatures18. However, while Coulomb drag measurements of double monolayer graphene (MLG) heterostructures have successfully probed the regime of strong interactions (small d/ℓB), no evidence of the EC phase has been reported19–21. Here, we report measurement of double bilayer graphene (BLG) structures in the QHE regime for interlayer separations spanning d = 2.5 to 5 nm, where d is the thickness of the hexagonal boron nitride (hBN) tunnel barrier. In addition to the potentially more favourable electronic dispersion in comparison with MLG17, the zeroth Landau level (ZLL) of BLG is eigfold degenerate, with the spin and valley isospin degeneracy supplemented by an accidental orbital degeneracy22. This multitude of broken symmetry states further expands the phase diagram of possible superfluid states.

Correlation between the layers in the QHE regime is probed by a combination of Coulomb drag and magnetoresistance measurements in both counterflow and parallel flow geometries (see Methods). At B = 9 T and T = 20 K, the longitudinal drag shows conventional behaviour (Fig. 1d), namely a finite response at partial Landau level (LL) filling that drops to zero when either layer is tuned to a QHE gap. As T decreases and B increases, we observe complete LL symmetry breaking with fully developed QHE gaps for all integer filling fractions. The overall drag signature diminishes at B = 15 T and T = 0.3 K, but apparently remains robust at certain filling fractions, as shown in Fig. 1e. Labelling regions of the plot by the coordinates of the bottom and top layer filling fraction, (νbot, νtop),

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an electron–hole asymmetry is apparent. In the electron–electron (e–e) quadrant, magnetodrag is observed whenever there is partial filling of both the \( \nu = 1 \) and 3 LLs \([1, 1], (1, 3), (3, 1) \) and \((3, 3)\), whereas in the hole–hole (h–h) quadrant it is partially filled \( \nu = 2 \) and 4 LLs \([-2, -2], (-2, -4), (-4, -2) \) and \((-4, -4)\).

Based on recent understanding of how the eightfold degeneracy of the ZLL in BLG lifts at large \( B \) (refs 23,24), we can assign a spin, valley, and orbital index to the symmetry broken states of each layer (see Supplementary Information), and observe that strong magnetodrag in Fig. 1e appears only where both layers are in a zero orbital state. Magnetodrag due to momentum or energy coupling\(^{15,22,23}\) is expected to vanish in the zero-temperature limit, whereas the 0.3 K response in Fig. 1e exceeds 1 kΩ in some regions, suggesting a different origin. One possibility is the formation of indirect excitons between the layers that are not yet phase coherent, resembling the EC precursor reported in GaAs double layers\(^{15}\). This interpretation would suggest a selection rule where EC formation is limited to the zero orbital ground states only.

Figure 2a–c shows the longitudinal magnetodrag (\( R_{Dxy}^{\text{long}} \)), Hall drag (\( R_{xy}^{\text{H}} \)), and drive layer Hall conductance (\( \sigma_{xy}^{\text{drive}} \)) for a device with interlayer separation \( d = 3.6 \) nm, measured at \( B = 18 \) T and \( T = 20 \) mK (for simplicity we focus our discussion on the e–e quadrant only, but a complete mapping of the ZLL can be found in the Supplementary Information). A large response is observed in \( R_{Dxy}^{\text{long}} \) and \( R_{xy}^{\text{drive}} \) following a diagonal line corresponding to total filling fraction \( \nu_x = 1, 3 \) and \( 5 \) (\( \nu_y = \nu_{bot} + \nu_x \)). Figure 2d shows \( R_{xy}^{\text{drive}} \) and \( R_{Dxy}^{\text{drive}} \) for varying magnetic field measured along a line of varying drive layer density. \( R_{Dxy}^{\text{drive}} \) shows conventional behaviour with well-defined QHE plateaux observed at \( \nu_{\text{drive}} = 1 \) and 2, while the magnetodrag is near zero at this sample temperature over most of the density range. However, when the drive and drag layer densities sum to \( \nu_x = 1 \), \( R_{Dxy}^{\text{drive}} \) deviates strongly from its single-layer value and exhibits re-entrant behaviour with quantized magnitude \( h/e^2 \).

At the same total filling, \( R_{xy}^{\text{drive}} \) takes on this same quantized value. The amplitude of \( R_{Dxy}^{\text{drive}} \) first rises dramatically in the vicinity of \( \nu_x = 1 \) and then dips rapidly to zero at exact filling. Quantization of both \( R_{Dxy}^{\text{drive}} \) and \( R_{xy}^{\text{drive}} \) at integer total filling, concomitant with a local zero-valued \( R_{Dxy}^{\text{drive}} \), provides strong evidence of the formation of an EC phase\(^{15}\).

Confirmation that a superfluid phase of charge carriers has truly formed is provided by magnetotransport in the counterflow geometry\(^a\), in which charge current is carried through the double-well system by excitons generated (and then annihilated) at the contacts (inset Fig. 2e). Charge-neutral excitons feel no Lorentz force even under very large \( B \) and zero Hall resistance is expected\(^{15}\). Indeed, Fig. 2e shows vanishing counterflow Hall resistance when \( \nu_x = 1 \). The dissipationless nature of the EC is revealed by simultaneous observation of zero longitudinal resistance \( R_{xx}^{CF} \). Figure 2e also plots Hall resistance in the parallel flow configuration, which is a linear combination of drag and counterflow measurements. The Hall resistance in the parallel flow geometry \( R_{xx}^{CF} \) shows a prominent peak at \( \nu_x = 1 \), approaching the quantized value of \( 2h/e^2 \). (This doubling of the quantization is due to the fact that current flows through the double BLG system twice, and \( R_{xx}^{CF} \) is defined as \( V_{xx}/I \) instead of \( V_{xx}/2I \).) The stark difference between \( R_{xx}^{CF} \) and \( R_{xy}^{\text{drive}} \) provides further evidence and confirmation the origin of the \( \nu_x = 1 \) state lies in the strong correlation and interlayer phase coherence between the two BLG layers.

In Fig. 3a we plot the magnitude of \( R_{xy}^{\text{drive}}, R_{Dxy}^{\text{drive}}, R_{xy}^{\text{drag}} \) and \( R_{Dxy}^{\text{drag}} \) versus \( d/\xi \). For device 37 \( (d = 3.6 \) nm), quantized \( R_{Dxy}^{\text{drive}} \) and \( R_{xy}^{\text{drive}} \) together with zero-valued \( R_{xy}^{\text{drag}} \) persist only over a narrow range, effectively establishing both an upper and lower critical value for \( d/\xi \). The upper bound is understood by the requirement to be in the so-called strongly interacting regime (that is, achieve a minimum effective interlayer interaction). We note that the critical value \( d/\xi \sim 0.6 \) is approximately 30% that was reported for GaAs\(^{14}\). Reducing the interlayer spacing from 3.6 nm to 2.5 nm results in a decrease of the lower critical \( d/\xi \) (Fig. 3a). However, we note that this boundary corresponds to approximately the same absolute magnetic field value of approximately 18 T. This may relate to the minimum magnetic field required to fully lift the ZLL degeneracy (set by sample disorder, which is approximately the same between these two devices). Alternatively this could be signal of a transition to a new, as yet unidentified, phase as \( d/\xi \) tends towards zero.

Figure 3b shows the counterflow Hall resistance \( R_{xx}^{CF} \) plotted as a function of filling fractions \( \nu_{\text{top}} \) and \( \nu_{\text{bot}} \). The EC state, as evidenced by a zero-valued \( R_{xx}^{CF} \), again follows a diagonal line corresponding to \( \nu_x = 1 \). Along this diagonal the state density imbalance, which we parametrize as \( \Delta \nu = \nu_{\text{bot}} - \nu_{\text{top}} \) (\( \Delta \nu = 0 \) only for \( \nu_{\text{top}} = \nu_{\text{bot}} = 1 \)). To understand the effect of this layer imbalance, we examine the temperature dependence of the \( \nu_x = 1 \) state over a large range of \( \Delta \nu \).

The minimum value of the \( R_{xx}^{CF} \) shows activated behaviour with varying temperature (Fig. 3c), allowing us to deduce an associated
Figure 2 | Superfluid exciton condensate. a–c, Magnetodrag (R_{xx}^{\text{drag}}), Hall drag (R_{xy}^{\text{drag}}) and drive layer Hall conductance (σ_{xy}) in the e–e quadrant for device 37 with a tunnel barrier thickness of d = 3.6 nm, measured at B = 18 T, T = 20 mK and V_{bias} = 0 V. d, Line cut of R_{xy}^{\text{drive}}, R_{xy}^{\text{drag}} and the amplitude of R_{xx}^{\text{drag}} near ν_{T} = 1 at different magnetic fields for device 45 with a tunnel barrier thickness of d = 2.5 nm. Inset shows the schematic of the Coulomb drag measurement. e, Line cut of counterflow Hall resistance R_{xx}^{CF}, longitudinal resistance R_{xx}^{σ}, and parallel flow Hall resistance R_{xy}^{Ω} near ν_{T} = 1 measured from the top BLG at B = 18 T. Inset shows the schematic of the counterflow measurement, which enables transport of charge-neutral excitons through the system. The linecuts shown in d and e are taken with non-zero density imbalance between the two BLG layers at ν_{T} = 1, where the condensate phase is fully developed as described in Fig. 3.

Figure 3 | Tunability of the condensate phase. a, R_{xy}^{\text{drive}} and R_{xy}^{\text{drag}} measured at Δν = −0.3 as a function of effective interlayer separation d/λ for device 37 (d = 3.6 nm) and 45 (d = 2.5 nm). Device 45 shows a much smaller lower critical value of d/λ above which full quantization of R_{xy}^{\text{drag}} and R_{xy}^{\text{drive}} are observed. b, R_{xy}^{CF} measured at B = 18 T, T = 20 mK from device 37, as a function of filling factors for the ν_{T} = 1 state. Points along ν_{T} = 1 can be parametrized by the interlayer density imbalance Δν = ν_{bot} − ν_{top}. c, Temperature dependence of R_{xy}^{CF} for device 37 measured at B = 18 T, plotted in an Arrhenius scale, revealing that the energy gap Δ varies with Δν. Inset, the activation gap Δ obtained from c, (open circles) appears symmetric with Δν, and fits well to a parabola.

gap^4 as a function of the layer imbalance. In the inset of Fig. 3c, we plot the activation gap versus Δν. The data are fitted well by a parabolic dependence\textsuperscript{27} with a minimum of Δ ~ 0.6 K near zero density imbalance. The behaviour of the activation energy suggests that an interlayer density imbalance strengthens the interlayer correlation. Similar observations were previously reported for the
Figure 4 | Interlayer bias. a, \( n_{0p}^{\text{def}} \) measured for device 37, as a function of interlayer bias, \( V_{\text{bias}} \). The \( \nu = 1 \) peak vanishes and reappears as the EC state displays four different transitions with varying \( V_{\text{bias}} \). b, Calculated displacement field \( D \) for the two BLG layers as a function of interlayer bias \( V_{\text{bias}} \) (see Supplementary Information). The dashed and solid lines mark critical \( D \)-field values for transitions between different valley polarizations, \( |K+\rangle \) and \( |K-\rangle \). The grey shaded area corresponds to opposite valley polarization in the double BLG, \( |K+\rangle \rightarrow |K-\rangle \) and \( |K-\rangle \rightarrow |K+\rangle \), whereas the white area indicates the same valley polarization, \( |K+\rangle \rightarrow |K+\rangle \) and \( |K-\rangle \rightarrow |K-\rangle \). White circles in lower panel mark transitions in the relative valley order between the layers.\(^{23,24}\)

\( \nu_1 = 1 \) phase in GaAs double quantum wells\(^{28}\) and may have the same origin. Activated behaviour is observed also for the EC states at \( \nu_1 = 3 \) and 5; however, they exhibit much smaller energy gaps, and are therefore in general less developed compared to the \( \nu_1 = 1 \) state. A description of the features observed at these fillings, as well as the equivalent in the \( e-h \) quadrant, is provided in the Supplementary Information. However, a full analysis of these states is beyond the present manuscript and will be discussed elsewhere.

Finally, we study the stability of the \( \nu_1 = 1 \) state against perpendicular electric field. A voltage bias, \( V_{\text{bias}} \), is applied to one of the BLG layers (the bottom BLG in this case) to induce the displacement field, \( D \). The Hall drag signal shows multiple transitions with varying displacement field (Fig. 4a). The value of the displacement field at each critical point, open circles in Fig. 4b, shows good correspondence with \( D \) values for which we expect a transition of the valley order in at least one of the bilayers\(^{23,24}\). Moreover, it appears that the condensate phase is stabilized (finite drag) when the layers have opposite valley ordering, but suppressed (zero drag) for same ordering (see Supplementary Information). Since valley and layer are approximately equivalent for BLG in the lowest Landau level\(^{29}\), the valley (layer) dependence of the exciton condensate could suggest that interlayer coherence occurs mainly between the two adjacent single-layer graphenes\(^{30}\); however, further work will be necessary to understand the precise role of the valley ordering.

This work marks the beginning of a systematic study of excitonic superfluidity in graphene double-layer heterostructures. The capability of engineering and studying the superfluid state in the quantum Hall regime paves the way for realizing such condensates at higher temperature and possibly zero magnetic field.

Methods

Methods, including statements of data availability and any associated accession codes and references, are available in the online version of this paper.

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**Author contributions**

J.I.A.L., J.H. and C.R.D. designed the experiment. Experimental work and analysis was carried out by J.I.A.L., advised by J.H. and C.R.D. All authors contributed to writing the manuscript.

**Additional information**

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**Competing financial interests**

The authors declare no competing financial interests.
Methods
Our devices are assembled using the van der Waals transfer technique\textsuperscript{31}. The device geometry includes a local graphite bottom gate, an aligned metal top gate and graphite electrical leads, as shown in Fig. 1b and described in ref. 32. The two BLG are separated by a thin layer of hBN. Even for the thinnest hBN used (2.5 nm) the interlayer tunnelling resistance is measured to be larger than $10^9 \Omega$. Correlation between the layers in the QHE regime can be probed by a combination of Coulomb drag\textsuperscript{33} and magnetoresistance measurements in both counterflow and parallel flow geometries\textsuperscript{4,9,15}. In the drag measurement, a current $I_{\text{drive}}$ is sent through the drive BLG layer, while the longitudinal and Hall voltage ($V_{xx}$ and $V_{xy}$) of the drive and drag layers are measured simultaneously. We define the magneto- and Hall drag resistance as $R_{\text{drag}}^{xx} = V_{xx}^{\text{drive}} / I_{\text{drive}}$ and $R_{\text{drag}}^{xy} = V_{xy}^{\text{drive}} / I_{\text{drive}}$. Except where indicated, both BLG layers are grounded with no interlayer bias applied across the hBN tunnelling barrier. In the counterflow (parallel flow) measurement, equal current is sent through both layers, flowing in the opposite (same) direction, while measuring longitudinal and Hall resistance in each layer\textsuperscript{15} (see Supplementary Information for schematics of each configuration).

Data availability. The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

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