Quantum Hall drag of exciton condensate in graphene

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Exciton condensate is a Bose-Einstein condensate (BEC) of electron and hole pairs bound by the Coulomb interaction[1,2]. In an electronic double layer (EDL) under strong magnetic fields, filled Landau states in one layer bind with empty states of the other layer to form exciton condensate[3–9]. Here we report exciton condensation in bilayer graphene EDL separated by hexagonal boron nitride (hBN). Driving current in one graphene layer generates a near-quantized Hall voltage in the other layer, signifying coherent exciton transport[4,6]. Owing to the strong Coulomb coupling across the atomically thin dielectric, quantum Hall drag in graphene appears at a temperature ten times higher than previously observed in GaAs EDL. The wide-range tunability of densities and displacement fields enables exploration of a rich phase diagram of BEC across Landau levels with different filling factors and internal quantum degrees of freedom. The observed robust exciton condensation opens up opportunities to investigate various many-body exciton phases.

An exciton BEC is formed when a large fraction of excitons occupy the ground state, establishing macroscopic coherence with weak dipolar repulsion[1,3]. However, optically generated excitons have short lifetimes. They can quickly recombine and release a photon, which leads to the annihilation of excitons. By trapping the released photon in an optical cavity, recent studies have shown the BEC of exciton-polaritons, consisting of a superposition of an exciton and a photon[10–13]. Another way to achieve a large density of long-lived excitons is to place electrons and holes in spatially-separated parallel conducting layers, where excitons can form across the layers. In semiconducting EDLs, such indirect excitons can be formed by optical excitation[14] or electrical doping[15]. One salient feature of the exciton BEC is dissipationless exciton transport, consisting of counter-flowing electrical currents carried by co-traveling electrons and holes[4]. The first experimental observation of this superfluid exciton flow was demonstrated in GaAs EDLs under a strong magnetic field, in which a strong correlation is formed between electron-like and hole-like quasi-particles in quantizing orbits[3–9].

The magnetic-field-induced layer coherence of the EDL can be established in the following way. When a two-dimensional (2D) electron gas of density n is subject to a perpendicular magnetic field B, the kinetic energy of electrons is quantized to discrete Landau levels (LLs). Each LL contains $n_0 = \frac{eB}{h}$ degenerate Landau orbits per unit area, where e is electron charge and h is Planck's constant. If all the orbits in a LL are occupied (i.e., the filling factor $\nu = n/n_0$ is an integer), the 2D electron system forms a quantum Hall state. In the EDL, the filling factor of the individual layer can be specified by $\nu_{top} = n_{top}/n_0$ and $\nu_{bot} = n_{bot}/n_0$, where n_{top} and n_{bot} are the density of top and bottom layer, respectively. If LLs in both layers are partially filled, i.e., ν_{top} and ν_{bot} are non-integer numbers, neither layer can form a quantum Hall state on its own. However, Coulomb repulsion forces the electrons in the two layers to occupy different orbitals in space, leading to spatial anti-correlation between layers. Notably, when the total filling fraction, $\nu_{tot} = \nu_{top} + \nu_{bot}$, becomes an integer, the two layers together can form a coherent state in which each filled state (quasi-electron) in one layer correspond to an empty state (quasi-hole) in the other layer. These bound empty-filled states can be described as excitons in the quantum Hall scenario and yield a strong response in the Coulomb drag experiment: driving current in one layer generates a quantized Hall drag voltage in the other layer. To be specific, the ratio between Hall drag voltage (V_{xy}^{drag}) and drive current (I^{drive}), defined as Hall drag resistance, is then quantized as $R_{xy}^{drag} = V_{xy}^{drag}/I^{drive} = \frac{h}{\nu_{tot}e^2}$. Meanwhile, Hall resistance of the drive layer (R_{xy}^{drive}), obtained from the drive Hall voltage (V_{xy}^{drive}), is also quantized to the same value despite having a partially filled LL, while the longitudinal voltages in both layers vanishes.

Quantized Hall drag for $\nu_{tot} = 1$ has been observed in the lowest LLs in GaAs EDLs[6]. Coherent tunneling between the layers[5] and perfect current drag measurements[9] further confirmed the presence of the interlayer coherence and the superfluid exciton flow. The BEC realized in semiconducting EDLs, however, turns out to be rather fragile, with a BEC transition temperature T_c in the sub-Kelvin range. This fragility is mainly caused by a relatively large EDL separation *d* required. It is noted that T_c is proportional to the characteristic energy scale $e^2/\varepsilon l$, where ε is the dielectric constant and $\ell = (\hbar/eB)^{1/2}$ is the magnetic length specifying the distance between quasi-particles in a LL[16,17]. Also, the exciton BEC only appears in the strong coupling regime[4], where the d/ℓ ratio is below the critical value of $d/\ell < 2$. Thus reducing *d* substantially below the limit of the semiconducting EDL will likely enhance T_c and increase the exciton binding energy.

Recent progress on 2D van der Waals heterostructures has created a new opportunity to build EDLs using atomically thin materials[18–21]. Owing to electron-hole symmetry and extremely light carrier mass, EDLs consisting of mono- and bilayer graphene have been of particular interest to realize an exciton BEC[22–25]. Furthermore, tunneling currents in graphene-hBN-graphene heterostructures are not appreciable when *d* > 1.5 nm at small biases, due to the large bandgap of hBN and the lattice direction mismatch between graphene layers[18,26]. Initial experiments performed in graphene-hBN-graphene heterostructures demonstrated a strong Coulomb drag effect in the semiclassical regime realized at high temperatures, exhibiting the strong interaction between the two layers in the zero and finite magnetic fields[18–20]. However, experimental evidence of interlayer coherence has yet to be found. In this work, with improved device quality and fabrication technique (see supplementary information (SI)), we demonstrate magnetic-field-induced exciton condensation in graphene EDL.

Our devices are made of two Bernal stacked bilayer graphene (BLG) sheets separated by 3 nm hBN and encapsulated by two thicker hBN layers (20~30 nm) (Fig. 1a, b). The two graphene layers are independently contacted by multiple electrodes (see SI for details). Both graphene layers have mobility $0.5{\sim}1{\times}10^6\mathrm{cm}^2/\mathrm{Vs}$ and exhibit symmetry breaking quantum Hall states at fields as low as 5 T (SI, Fig. S3-4). No appreciable tunneling current is measured above the noise level, providing a lower bound on the tunneling resistance of 1 GΩ. The voltages applied to the top gate (V_{TG}), the bottom gate (V_{TG}) and the interlayer bias between graphene layers (V_{int}) tune the carrier densities of the top and bottom graphene layers (n_{top}, n_{bot}): $n_{top} = C_{TG}V_{TG} - C_{int}V_{int}$; $n_{bot} = C_{BG}V_{BG} + C_{int}V_{int}$. Here C_{TG}, C_{BG}, and C_{int}, are capacitances between the top gate and top layer, the bottom gate and bottom layer, and between the top and bottom graphene layers, respectively. By controlling V_{TG}, V_{BG}, and V_{int}, we can also adjust the average displacement fields; $D_{top} = (C_{TG}V_{TG} + C_{int}V_{int})/2$ and $D_{bot} = (-C_{BG}V_{BG} + C_{int}V_{int})/2$, for the top and bottom layer, respectively. For the drag measurements, we apply current I^{drive} only in the top layer and measure the Hall resistance R_{xy}^{drive} of the current-carrying (drive) layer, and magneto- and Hall drag resistance, R_{xx}^{drag} and R_{xy}^{drag}, of the drag layer under a perpendicular magnetic field *B*. Owing to the Onsager relation, switching the drive and drag layer produces experimentally equivalent drag results (SI, Fig. S5).

Fig. 1c shows measurements of $R_{xy}^{drive}, -R_{xx}^{drag}, R_{xy}^{drag}$ under *B* = 25 T, corresponding to the strong coupling limit (ℓ = 5.1 nm and $d/\ell = 0.58$). In this plot, we adjust V_{TG} and V_{BG} such that the filling fractions of each layer are balanced ($\nu \equiv \nu_{drive} = \nu_{drag}$). We observe that each layer exhibits its own quantum Hall (QH) effect. For $\nu \geq 1$,

R_{xy}^{drive} exhibit QH plateaus at the values $(R_{xy}^{drive})^{-1} = \frac{e^2}{h}, \frac{4}{3}\frac{e^2}{h}, \frac{2e^2}{h}$. In these well-developed integer (ν=1, 2) and fractional (ν=4/3) QH regimes of the individual layers, we find no appreciable drag signal ($R_{xx}^{drag} \approx R_{xy}^{drag} \approx 0$). The vanishing drag signals at low temperatures are expected in the semiclassical picture due to the diminishing scattering phase space[27]. However, the observed drag signals are significantly enhanced when the first LL of both layers are partially filled ($\nu < 1$). In particular, for ν=1/2, where both layers are half-filled and thus $\nu_{tot} = \nu_{drive} + \nu_{drag} = 1$, the Hall drag signal reaches close to the quantization value of $h/e^2 = 25.8$ kΩ, while the magneto-drag (R_{xx}^{drag}) dips to nearly zero. Under the same condition, the Hall resistance in the drive (top) layer, which originally rises beyond h/e^2 as ν_{drive} drops below one (i.e., partially filled LL), re-enters h/e^2 again at ν_{tot}= 1. This re-entrant behavior of R_{xy}^{drive} to the same quantized value of R_{xy}^{drag} indicates that the entire EDL behaves like a single ν=1 quantum Hall system despite that LLs in each layer are only partially filled.

The quantized Hall drag and re-entrant QHE in the drive layer have been observed previously in the GaAs EDLs for ν_{tot}= 1 and are considered as a strong evidence of interlayer coherence and exciton superfluidity[6]. We note, however, the unambiguous demonstration of superfluidity and coherence requires further measurements. A simple physical picture for the observed quantized Hall drag can be built upon a two-fluid picture. In this model, currents in each layer are carried by excitons in the bulk ($I_{ex}^{(i)}$) and quasi-particles flowing on the edge ($I_{qp}^{(i)}$), where the superscript is the layer index. Excitons generate counter flow currents $I_{ex}^{drag} = -I_{ex}^{drive}$; and the zero accelerating electric force requirement on superfluid excitons demands $V_{xy}^{drive} = V_{xy}^{drag} = V_{xy}$. In addition, boundary conditions of the drag and drive layers requires $I_{ex}^{drag} + I_{qp}^{drag} = 0,\ I = I_{ex}^{drive} + I_{qp}^{drive}$. Furthermore, by considering the two layers as a single coherent quantum Hall system at filling fraction υ_{tot}, we have $I_{qp} = I_{qp}^{drag} + I_{qp}^{drive} = \frac{\upsilon_{tot}e^2}{h} V_{xy}$. Summing up, we obtain the experimental observation $R_{xy}^{drive} = R_{xy}^{drag} = h/\upsilon_{tot}e^2$ with vanishing R_{xx}^{drag} and R_{xx}^{drive}.

We found that the observed quantized Hall drag in graphene is much more robust than that of the GaAs EDLs. The signatures of the exciton condensate, i.e., nearly quantized R_{xy}^{drag} and re-entry behavior of R_{xy}^{drive}, persist up to a few Kelvin for $\nu_{tot} = 1$ at B = 25 T (Fig. 1c inset). For quantitative analysis, we compute the counter-flow resistances R_{xy}^{CF} and R_{xx}^{CF} and plot them as a function of 1/T at $\nu_{tot} = 1$ (SI, Fig. S7). While the theoretical expectation for 2D BEC transition is the Kosterlitz-Thouless transition[16,28], we find that the vanishing R_{xy}^{CF} and R_{xx}^{CF} exhibit thermally activating behavior similar to what has been observed in GaAs system[6]. However, the activation gap we obtained ($\Delta \approx 8$ K) is ten times larger than previous reported (SI, Fig. S7b).

The exciton BEC in graphene is also found to be robust against the density imbalance between layers.[29] The signatures of the exciton condensation, i.e., $R_{xy}^{drag} \approx R_{xy}^{drive} \approx h/e^2$ and $R_{xx}^{drag} \approx R_{xx}^{drive} \approx 0$ withstand a range of gate voltages satisfying $\nu_{drag} + \nu_{drive} = 1$, corresponding to the diagonal line specified in Fig. 1d-f. For a more quantitative analysis, we plot R_{xy}^{drag} cut along this diagonal line as an overlay graph in Fig. 1d (white trace). The level of R_{xy}^{drag} quantization indicates that the BEC persists for the density imbalance $\frac{\Delta n}{n_{tot}} = \frac{n_{drag} - n_{drive}}{n_{drag} + n_{drive}}$ up to ~ $\pm 30\%$. Beyond this limit the more stable integer QH states (ν_{drag} or $\nu_{drive} = 0$ or 1) in each layer take over the exciton BEC phase.

While the exciton BEC has been discovered only for the half-filled lowest LL in the GaAs EDLs, the gate tunability in graphene EDL devices allows us to explore the phase diagram of possible condensate states other than ν_{tot}= 1. Fig. 2 shows experimental survey for R_{xy}^{drag} as a function of υ_{drive} and υ_{drag}, covering the electron-electron ($\upsilon_{drive}, \upsilon_{drag} > 0$) and hole-hole ($\upsilon_{drive}, \upsilon_{drag} < 0$) regimes. Remarkably, we find at least two additional interlayer correlated states in these regimes: ($\upsilon_{drive}, \upsilon_{drag}$) centered near ~(0.35, 2.65) and

(-1.5, -1.5), corresponding to the drag between ½ - 2½ filled electron LLs ($\nu_{tot} = 3$) and 1½ - 1½ filled hole LLs ($\nu_{tot} = -3$), respectively. Similar to the BEC in $\nu_{tot} = 1$, these states exhibit the near-quantized Hall drag $R_{xy}^{drag} \approx h/\upsilon_{tot}e^2$ and $R_{xx}^{drag} \approx 0$, for a range of ($\upsilon_{drive}, \upsilon_{drag}$) satisfying $\nu_{drive} + \nu_{drag} = \nu_{tot}$ (see Fig 2. b-j). These Hall drag features appear as diagonals in the ($\upsilon_{drive}, \upsilon_{drag}$) plots and are confined to the sectors corresponding to partially filled first (drive) and third (drag) electron LLs (with all symmetries are lifted) and partially filled second (drive and drag) hole LLs. While we cannot rule out other possible scenario compatible with experimental observation, we interpret these interlayer correlated states as exciton BEC for $\nu_{tot} = \pm 3$. Measurements at lower magnetic fields also reveal a signature of developing exciton BEC for (υ_{drive}, υ_{drag})=(2.5, 0.5), the symmetric pair for (0.5, 2.5) discussed above (SI, Fig. S5&8). The relatively weak presence of this symmetric pair is presumably due to the quality difference between the top and bottom graphene layers. We also remark that while $R_{xy}^{drag} \approx h/\upsilon_{tot}e^2$ and $R_{xx}^{drive} \approx R_{xx}^{drag} \approx 0$ are observed in the $\nu_{tot} = \pm 3$ state, we find $|R_{xy}^{drive}| > h/\upsilon_{tot}e^2$ (Fig. 2e and k). Together with less developed quantization of R_{xy}^{drag} in these states, compared to $\nu_{tot} = 1$, we speculate that a dissipative exciton transport of a fragile BEC is responsible for this incomplete re-entrant QHE.

Interestingly, our experimental observations strongly indicate that the apparent electron-hole symmetry of LLs is broken for the exciton BEC. For example, the (0.5, 0.5) BEC exists while (-0.5, -0.5) is absent. The electron-hole asymmetry has been observed in the filling fraction sequences of the fractional quantum Hall effect in bilayer graphene and related with the orbital degeneracy of bilayer graphene LLs[30].

We note that the existence of the exciton BEC at fixed ($\upsilon_{drive}, \upsilon_{drag}$) sensitively depends on the V_{int} (Fig.3 and Fig. S8-10). When the density of each layer is kept close to half-filling by coordinately tuning V_{BG} and V_{TG}, V_{int} changes the displacement field exerted on both layers, $D_{top} = C_{int}V_{int} + n_{top}/2$ and $D_{bot} = C_{int}V_{int} - n_{bot}/2$. Fig. 3c shows R_{xy}^{drag} as a function of D_{top} (top axis of Fig. 3b) and D_{bot} (bottom axis of Fig. 3d) while keeping $\nu_{tot} = 1$ at B=13T, T=1.5K. We observe that R_{xy}^{drag} undergoes multiple distinct transitions between high and low values as V_{int} changes. By comparing the displacement fields of these transitions with those of the integer quantum Hall transitions of each layer (Fig. 3b, d, Fig. S10a), we found a close connection between the Hall drag and the LL character of each layer.

For a single bilayer graphene, the lowest Landau level (LLL) have eight-fold degeneracy in the single particle picture (between $\nu = -4$ and $\nu = 4$). This SU(8) symmetry space consists of spin degeneracy $|\uparrow\rangle$ and $|\downarrow\rangle$, valley degeneracy K and K' and orbital degeneracy of $N = 0,1$[31–34]. In high quality samples, these symmetries are broken by quantum Hall ferromagnetism (QHFM), and different symmetry-breaking states can be found at different displacement fields for each filling factor[31–34]. The symmetry-breaking state $\nu = 1$ has three transition points at $D = 0$ and $D = \pm D_1$, and $\nu = 0$ has four transition points at $D = \pm D_2, \pm D_3$, which can be identified in Fig. 3b, d. Across these transitions, the fully filled LLs have different spin, valley or orbital indexes. For partially filled LLs, layer polarization and orbital character were identified by a recent study using capacitance measurements[34]. The partially filled LL $0 < \nu < 1$ is found to hold different layer- and orbital-polarized states at different displacement fields (Fig. 3a) (+/- denote the layer polarization, which is equivalent to valley polarization K/K' in the lowest LL, and 0/1 denote the orbital index). The transitions between these states line up with the transitions of $\nu = 1$ and $\nu = 0$ states regarding to displacement field. In our experiment, the $\nu_{tot} = 1$ state is formed between two partially filled bilayer graphene with $0 < \nu_{top}, \nu_{bot} < 1$. We found that Hall drag is suppressed when the partially filled LLs of one bilayer graphene or both bilayer graphene are in the N=1 orbital state (shaded regions in Fig. 3c and Fig. S10).

This also explains the absence of other integer total filling factor states and the broken electron-hole symmetry, as the $-1 < \nu < 0$ ($0 < \nu < 1$) LL is polarized in the N=1 (N=0) orbital state under the same displacement field; the latter is capable of forming an exciton BEC while the former is not. We speculate that the inability of the N=1 orbit to establish an interlayer correlated state is due to its broader spatial wavefunction (Fig. 3a right) and thus weaker interaction. Similarly, the weaker Hall drag signal in the region where $D_{top} > 0$ and $D_{bot} < 0$ can be related to the weaker interaction when the wavefunctions of the two layers are further apart due to the opposite layer polarization.

Methods

Sample fabrication The five single-crystal layers of graphene and hBN (Fig. S1a) are prepared by mechanical exfoliation and van der Waals (vdW) transfer technique[35]. During the process, we choose two strip-shape bilayer graphene and align them into a cross, so we can use the overlapped part as the main channel area while fabricating individual contacts onto the non-overlapped parts (Fig. S1b). The edge contacts and the top gate are fabricated after etching the entire stack into the final device geometry. Usually, the parts of graphene near metal contacts are slightly doped and form unwanted PN junctions due to the metal-graphene work function mismatch. These contact-induced local PN junctions cause contact barriers under high magnetic fields. In order to address this problem, we fabricate local top gates (contact gates) to dope the area of graphene around the contacts. These contact gates are adjusted to highly dope graphene leads next to the metal electrodes with the same carrier type of the channel (Fig. S1c).

Drag measurements In most of our measurements, we use the top layer as the drive layer and the bottom layer as the drag layer (switching the drive and drag layers does not affect our drag experiment results as shown in Fig. S5). We apply $I^{drive} = $ 2nA AC current (17.7Hz) to the drive layer and measure the voltage drop along the longitudinal direction in the drag layer to obtain V_{xx}^{drag}. We also measure the voltage drops along the perpendicular direction in the drag and drive layers to obtain V_{xy}^{drag} and V_{xy}^{drive}, respectively. The magneto and Hall drag resistances are obtained by $R_{xx}^{drag} = V_{xx}^{drag}/I^{drive}$ and $R_{xy}^{drag} = V_{xy}^{drag}/I^{drive}$ from this measurement. One of the contacts in the drag layer is connected to ground through a $1M\Omega$ resistor to allow the gates to tune the density of the drag layer.

A typical way to drive current in lock-in measurements is applying AC bias voltage V^{drive} on one side of the graphene channel while grounding the other side. However, in Coulomb drag measurements, biasing the drive layer employing this scheme can create spurious drag signals in the drag layer due to drive bias induced AC gating. Because of the finite contact and channel resistances of the drive layer, the direct biasing scheme raises the potential in the middle of the drive layer to ~$V^{drive}/2$ with respect to ground. Since the drag layer is grounded, an AC interlayer bias of ~$V^{drive}/2$ is produced accordingly. In the previous drag experiments performed in GaAs double quantum wells, it has been shown that this AC interlayer bias induces spurious drag signals[36]. In order to prevent the AC interlayer bias, we employ a bridge setup[37] as shown in Fig. S2. In this scheme, the initial 4 Volts AC voltage is reduced down to 4mV through a voltage divider. Then this 4mV AC voltage is fed into the bridge through a 1:1 ground-isolating transformer. By tuning the variable resistor in the bridge, we can adjust the AC electrical potential in the middle of the drive layer to approximate zero. The 4mV is then converted into ~2nA by passing through two 1MOhm resistors in series. This setup also allows us to control the DC interlayer bias V_{int}. To monitor the drive current, we measure the voltage across one of the two 1MOhm resistors.

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.

References

1. Littlewood, P. B. *et al.* Models of coherent exciton condensation. *J. Phys. Condens. Matter* **16,** S3597–S3620 (2004).
2. Snoke, D. *et al.* Spontaneous Bose coherence of excitons and polaritons. *Science* **298,** 1368–72 (2002).
3. Eisenstein, J. & MacDonald, A. Bose–Einstein condensation of excitons in bilayer electron systems. *Nature* 691–694 (2004). at <http://www.nature.com/nature/journal/v432/n7018/abs/nature03081.html>
4. Eisenstein, J. P. Exciton Condensation in Bilayer Quantum Hall Systems. *Annu. Rev. Condens. Matter Phys.* **5,** 159–181 (2014).
5. Spielman, I. B., Eisenstein, J. P., Pfeiffer, L. N. & West, K. W. Resonantly Enhanced Tunneling in a Double Layer Quantum Hall Ferromagnet. *Phys. Rev. Lett.* **84,** 5808–5811 (2000).
6. Kellogg, M., Spielman, I. B., Eisenstein, J. P., Pfeiffer, L. N. & West, K. W. Observation of quantized Hall drag in a strongly correlated bilayer electron system. *Phys. Rev. Lett.* **88,** 126804 (2002).
7. Kellogg, M., Eisenstein, J. P., Pfeiffer, L. N. & West, K. W. Vanishing Hall resistance at high magnetic field in a double-layer two-dimensional electron system. *Phys. Rev. Lett.* **93,** 36801 (2004).
8. Tutuc, E., Shayegan, M. & Huse, D. A. Counterflow measurements in strongly correlated GaAs hole bilayers: Evidence for electron-hole pairing. *Phys. Rev. Lett.* **93,** 36802 (2004).
9. Nandi, D., Finck, a D. K., Eisenstein, J. P., Pfeiffer, L. N. & West, K. W. Exciton condensation and perfect Coulomb drag. *Nature* **488,** 481–4 (2012).
10. Kasprzak, J. *et al.* Bose-Einstein condensation of exciton polaritons. *Nature* **443,** 409–14 (2006).
11. Byrnes, T., Kim, N. Y. & Yamamoto, Y. Exciton–polariton condensates. *Nat. Phys.* **10,** 803–813 (2014).
12. Deng, H., Weihs, G., Santori, C., Bloch, J. & Yamamoto, Y. Condensation of Semiconductor Microcavity Exciton Polaritons. *Science (80-.).* **298,** (2002).
13. Balili, R., Hartwell, V., Snoke, D., Pfeiffer, L. & West, K. Bose-Einstein Condensation of Microcavity Polaritons in a Trap. *Science (80-.).* **316,** (2007).
14. High, A. A. *et al.* Spontaneous coherence in a cold exciton gas. *Nature* **483,** 584–8 (2012).
15. Seamons, J. A., Morath, C. P., Reno, J. L. & Lilly, M. P. Coulomb Drag in the Exciton Regime in Electron-Hole Bilayers. *Phys. Rev. Lett.* **102,** 26804 (2009).
16. Yang, K. *et al.* Quantum ferromagnetism and phase transitions in double-layer quantum Hall systems. *Phys. Rev. Lett.* **72,** 732–735 (1994).
17. Moon, K. *et al.* Spontaneous interlayer coherence in double-layer quantum Hall systems: Charged vortices and Kosterlitz-Thouless phase transitions. *Phys. Rev. B* **51,** 5138–5170 (1995).
18. Gorbachev, R. V. *et al.* Strong Coulomb drag and broken symmetry in double-layer graphene. *Nat. Phys.* **8,** 896–901 (2012).
19. Liu, X. *et al.* Coulomb drag in graphene quantum Hall double-layers. (2016). at <http://arxiv.org/abs/1612.08308>
20. Li, J. I. A. *et al.* Negative Coulomb Drag in Double Bilayer Graphene. *Phys. Rev. Lett.* **117,** 46802 (2016).
21. Lee, K. *et al.* Giant Frictional Drag in Double Bilayer Graphene Heterostructures. *Phys. Rev. Lett.* **117,** 46803 (2016).
22. Min, H., Bistritzer, R., Su, J.-J. & MacDonald, A. Room-temperature superfluidity in graphene bilayers. *Phys. Rev. B* **78,** 121401 (2008).
23. Kharitonov, M. & Efetov, K. Electron screening and excitonic condensation in double-layer graphene systems. *Phys. Rev. B* **78,** 241401 (2008).
24. Perali, A., Neilson, D. & Hamilton, A. R. High-Temperature Superfluidity in Double-Bilayer Graphene. *Phys. Rev. Lett.* **110,** 146803 (2013).
25. Skinner, B. Interlayer excitons with tunable dispersion relation. *Phys. Rev. B - Condens. Matter*

Mater. Phys. **93,** 2 (2016).

26. Lee, G. H. Electron tunneling through atomically flat and ultrathin hexagonal boron nitride. *Appl. Phys. Lett.* **99,** 243114 (2011).
27. Narozhny, B. N. & Levchenko, A. Coulomb drag. *Rev. Mod. Phys.* **88,** 25003 (2016).
28. Wen, X.-G. & Zee, A. Neutral superfluid modes and '"magnetic"' monopoles in multilayered quantum Hall systems. *Phys. Rev. Lett.* **69,** 1811–1814 (1992).
29. Champagne, A. R., Finck, A. D. K., Eisenstein, J. P., Pfeiffer, L. N. & West, K. W. Charge imbalance and bilayer two-dimensional electron systems at $\nu T = 1$. *Phys. Rev. B* **78,** 205310 (2008).
30. Kou, A. *et al.* Electron-hole asymmetric integer and fractional quantum Hall effect in bilayer graphene. *Science* **345,** 55–7 (2014).
31. Lambert, J. & Côté, R. Quantum Hall ferromagnetic phases in the Landau level $N = 0$ of a graphene bilayer. *Phys. Rev. B* **87,** 115415 (2013).
32. Maher, P. *et al.* Bilayer graphene. Tunable fractional quantum Hall phases in bilayer graphene. *Science* **345,** 61–4 (2014).
33. Lee, K. *et al.* Chemical potential and quantum Hall ferromagnetism in bilayer graphene. *Science* **345,** 58–61 (2014).
34. Hunt, B. M. *et al.* Competing valley, spin, and orbital symmetry breaking in bilayer graphene. (2016). at <http://arxiv.org/abs/1607.06461>
35. Wang, L. *et al.* One-dimensional electrical contact to a two-dimensional material. *Science* **342,** 614–7 (2013).
36. Hill, N. P. R. *et al.* Frictional drag between parallel two-dimensional electron gases in a perpendicular magnetic field. *J. Phys. Condens. Matter* **8,** L557–L562 (1996).
37. Kellogg, M. Thesis: Evidence for excitonic superfluidity in a bilayer two-dimensional electron system. **2005,** (2005).

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Figure legends

Figure 1 | Quantized Hall drag for $\nu_{tot} = 1$ state in bilayer graphene double layers. a, schematic diagram of device and measurement setup. **b**, optical microscope image of the device. Metal leads on the left and right of the image (three on each side) contact the top layer graphene, while others contact the bottom layer graphene. The blue shaded area of graphene is under the top gate; white and green shaded regions are under the contact gate. The contact gates support highly transparent electrical contacts under high magnetic fields, providing reliable data away from $\nu = 0$ insulating state. **c**, $R_{xy}^{drag}, R_{xx}^{drag}, R_{xy}^{drive}$ as a function of filling factors of both layers at B=25T and T=300mK. The exciton BEC can be recognized by near quantized Hall drag ($R_{xy}^{drag} = \frac{h}{e^2}, R_{xx}^{drag} = 0$) with the simultaneous re-entrant quantum Hall in the drive layer. **c** inset, temperature dependence (*T*=0.9, 2.29, 3.13, 4.35, 6, 8 ,10K) of R_{xy}^{drag} at B=25T. **d**, **e**, **f,** $R_{xy}^{drag}, -R_{xx}^{drag}$, R_{xy}^{drive} as a function of filling fractions ν_{top} and ν_{bot}, computed from V_{TG} and V_{BG}. The exciton BEC region appears as a diagonal region satisfying $\nu_{top} + \nu_{bot} = 1$. The white trace in **d** shows the value of R_{xy}^{drag}(axis on the left) along $\nu_{tot} = 1$ line (dashed line in **d**). For more quantitative analysis, we also present several traces of the color plot in SI Fig. S11.

Figure 2 | Exciton BEC in various LL fillings. a, R_{xy}^{drag} as a function of the top and bottom layer filling factors at B=25T, T=300mK and V_{int}= -0.05V. Besides (ν_{drag}, $\nu_{drive}) = (0.5, 0.5)$, a different exciton BEC state is found near $(0.5, 2.5)$. **b, c, d**, zoomed-in plot of R_{xy}^{drag}, R_{xx}^{drag}, R_{xy}^{drive} around $\nu_{tot} = 3$ at a higher field of B=31T. **e**, line-cut of R_{xy}^{drag}, R_{xx}^{drag}, R_{xy}^{drive} along dashed line show in **b, c, d**. **f,** R_{xy}^{drag} as function of filling factors at B=25T, T=300mK and V_{int}= 0.15V. Additional exciton BEC state in the hole-hole regime is identified around $(\nu_{drag}, \nu_{drive}) = (-1.5, -1.5)$. **g, h, i, j,** R_{xy}^{drag}, R_{xx}^{drag}, R_{xy}^{drive}, R_{xx}^{drive} at the same condition as **f**. **k,** line-cut of R_{xy}^{drag}, R_{xx}^{drag}, R_{xy}^{drive}, R_{xx}^{drive} along the dashed line in **g, h, i, j**. The horizontal dashed lines in **e** and **k** indicate $\pm\frac{h}{3e^2}$ where R_{xy}^{drag} and R_{xy}^{drive} are expected to be quantized to.

Figure 3 | Phase transition of $\nu_{tot} = 1$ exciton BEC induced by transverse electric field. a, schematic diagram of Landau levels sequence (left) and wave functions (right) of bilayer graphene QHFM states for the lowest LL corresponding to $-4 < \nu < 4$. In the left diagram, x direction represents displacement field *D* and colored lines denote QHFM Landau levels with different orbital (0/1), layer (+/-) and spin (↑/↓) quantum numbers, as noted next to the lines. Different colors are used for different orbital and layer quantum numbers. As the displacement field changes, the colored lines cross each other, representing QHFM transitions induced by the displacement field. The horizontal black dashed line marks charge neutrality ($\nu = 0$) and the horizontal white dashed line marks Fermi level of half-filled first LL ($\nu = 1/2$). The diagram on the right depicts wave functions of different bilayer graphene QHFM states, with the same color code as the lines in the left diagram. **b (d)**, longitudinal resistance of top (bottom) bilayer graphene as a function of displacement D_{top} (D_{bot}) and density n_{top} (n_{bot}) at B =13T and T=1.5K. The QHFM transitions are marked with arrows. The data in **b** is taken at a fixed $V_{BG} = -5.5$V and the data in **d** is taken at $V_{TG} = -1.1$V. **c**, R_{xy}^{drag} (blue curve) as a function of D_{top} (top axis of **b**) or D_{bot} (bottom axis of **d**) for $\nu_{tot} = 1$ state at B=13T and T=1.5K. Each data point is taken at a different V_{int}, with V_{BG} and V_{TG} tuned around $\nu_{tot} = 1$ to maximize R_{xy}^{drag} (filling factor ν_{top}, ν_{bot} calculated from the gate voltages are marked by the horizontal dashed line in **b** and **d**). The colored bars on the top and bottom of the plot represent orbital and layer character of the half-filled LL ($\nu = 1/2$) of the top and bottom bilayer graphene with the same color code as **a**. The orange and green shaded regions of the main plot which signify N=1 orbital states of the top and bottom layer coincide well with where the Hall drag vanishes, indicating N=1 orbit is incapable of forming exciton BEC phase.