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Permalink

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Journal

Nature Materials

ISSN

1476-1122

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Publication Date

2025-08-25

DOI

10.1038/s41563-025-02316-5

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Peer reviewed

Competition between excitonic insulators and quantum Hall states in correlated electron-hole bilayers

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18

Excitonic insulators represent a unique quantum phase of matter that enables the study of exotic quantum bosonic states. Strongly coupled electron-hole bilayers, which host stable dipolar exciton fluids with an exciton density that can be adjusted electrostatically, offer an ideal platform to investigate correlated excitonic insulators. Based on electron-hole bilayers made of MoSe₂/hBN/WSe₂ heterostructures, here we study the behavior of excitonic insulators in a perpendicular magnetic field. We report the observation of excitonic quantum oscillations in both Coulomb drag signals and electrical resistance at low to medium magnetic fields. Under a strong magnetic field, we identify multiple quantum phase transitions between the excitonic insulator phase and the bilayer quantum Hall insulator phase. These findings underscore the interplay between the electron-hole interactions and Landau level quantization, and enable further exploration of quantum phenomena in composite bosonic insulators.

31
 32 Electron-hole (e-h) bilayers, where a two-dimensional electron gas (2DEG) and a 2D hole gas
 33 (2DHG) are confined to two closely spaced but electrically isolated layers¹⁻⁴, provide a versatile
 34 platform for studying quantum phases of correlated e-h fluids. Strong e-h Coulomb interactions
 35 lead to bound exciton states, while spatial separation suppresses recombination, enabling long-
 36 lived excitons in thermal equilibrium controllable by electrostatic gating. These systems are of
 37 great interest for exploring quantum bosonic and fermionic phases, including excitonic insulators
 38 (EIs)^{1,3,4}, Bose-Einstein condensates^{2,5-9}, and higher-order complexes such as trions and
 39 biexcitons¹⁰⁻¹³.

40

41 Recent advances have enabled the creation of strongly coupled e-h bilayers using transition metal
42 dichalcogenide (TMD) heterostructures^{3,4,12-15}. With large effective mass and reduced dielectric
43 screening in TMD systems, the strong Coulomb attraction across the layers leads to spontaneous
44 formation of interlayer excitons. Compared with earlier system based on III-V semiconductor
45 quantum wells or graphene^{7,16-23}, strong e-h coupling regime is accessible without external
46 magnetic fields. A well-defined EI ground state at zero magnetic field has been reported,
47 characterized by robust exciton binding, a large charge gap (~20 meV)^{3,4} and perfect Coulomb
48 drag^{14,15}. Theoretical studies suggest that applying an out-of-plane magnetic field to such EIs
49 induces quantum oscillations (QOs) via interplay between Coulomb interactions and magnetic
50 cyclotron energy^{24,25}. In the strong field limit, it is also predicted that the magnetic field can
51 periodically destroy exciton binding and turn the EI into two independent quantum Hall insulators
52 (QHIs)²⁴. The appearance of QOs – a defining feature of a metallic system – in correlated
53 insulating phases is intriguing, and has been a topic under decade-long debate²⁶⁻⁴⁰. Experimental
54 evidence for the predicted excitonic QOs and quantum phase transitions have remained elusive.
55

56 In this paper, we report an experimental study of excitonic insulating e-h bilayers under an out-of-
57 plane magnetic field in MoSe₂/hBN/WSe₂ heterostructures. At zero magnetic field, the EI phase
58 is strongly insulating and exhibits perfect Coulomb drag behavior. As a magnetic field is applied,
59 we observe clear QOs of drag currents and electrical resistances. At higher magnetic fields, we
60 further observe multiple EI-QHI phase transitions.

61 **Excitonic insulating electron-hole bilayers**

62

63 Here we fabricate strongly coupled e-h bilayer devices based on MoSe₂/hBN/WSe₂
64 heterostructures, as illustrated in Fig. 1a. The device consists of two TMD monolayers – MoSe₂
65 (electron layer) and WSe₂ (hole layer) – separated by an ultrathin hBN tunneling barrier. They are
66 sandwiched by graphite top gate (TG) and bottom gate (BG) with hBN dielectrics. Dual gates and
67 separate TMD contacts allow independent electrostatic control of electron and hole densities. The
68 MoSe₂ conduction band and WSe₂ valence band form a type-II band gap of ~1.5 eV. This charge
69 gap can be electrically closed by a combination of interlayer bias voltage $V_B \equiv V_h - V_e$ and vertical
70 electric field $V_{BG} - V_{TG}$. We apply a large and fixed electric field, and continuously tune the charge
71 gap with V_B . The symmetric gate voltage $V_G \equiv V_{TG}/2 + V_{BG}/2$ sets the Fermi level and controls
72 the e-h density imbalance.

73 To enable transport measurements, we fabricate two charge reservoir regions with increased
74 interlayer spacing on the two sides of the active area. In these regions, the larger interlayer distance
75 leads to a higher interlayer voltage difference under the same vertical electric field, creating a
76 heavily doped electron-hole plasma (EHP) that serves as efficient exciton contacts^{3,4}. Each
77 reservoir region is equipped with one graphite contact to the MoSe₂ layer, and three platinum (Pt)
78 electrodes for the WSe₂ layer. Fig. 1b shows the optical image of a representative device D1 that
79 has a 2.3 nm (7-layer) hBN tunneling barrier. We focus on this device in the main text, but similar
80 results are obtained in devices D2 and D3 (Extended Data Figs. 1-2). Unless otherwise noted, all
81 measurements are performed at a lattice temperature $T = 0.01$ K.

82
83 We first characterize the device at zero magnetic field. Taking advantage of the sensitive doping
84 dependence of optical absorption in monolayer TMDs, the charge doping phase diagram of the e-

86 h bilayer can be determined using optical spectroscopy³. Fig. 1c-d shows the experimentally
87 determined hole and electron densities n_h and n_e as functions of V_G and V_B . The constant vertical
88 electric field ~ 0.45 V/nm reduces the type-II bandgap from 1.5 eV to ~ 0.37 eV. The interlayer
89 bias V_B controls the exciton chemical potential, and hence the effective charge gap $E_g =$
90 0.37 eV $- V_B$. For $V_B < 0.34$ V, the system is a normal insulator (NI) with a finite band gap in
91 which V_G can tune the system from intrinsic to either electron doped or hole doped. With a larger
92 V_B ($0.34 - 0.37$ V), the charge gap becomes smaller than the interlayer exciton binding energy E_b
93 but remains non-zero. The e-h bilayer is known to host an EI state at charge neutrality, which is
94 exciton-compressible but charge-incompressible^{3,4,14,15}. Increasing V_B further destroys the EI
95 through an interaction-driven exciton Mott transition into an EHP phase^{3,4,14}.
96

97 We employ two types of transport measurements to probe exciton behavior. The first is a two-
98 terminal resistance measurement of the WSe₂ layer, with the electron layer open-circuited (Fig.
99 1e). Two separate pairs of Pt electrodes are used for current sourcing and voltage sensing to
100 eliminate the Pt-WSe₂ junction resistance (Methods). Each highly doped reservoir region acts as
101 one terminal for the active region. The measured hole-layer resistance R_h is provided in Fig. 1f.
102 As expected, R_h shares a similar overall shape with the n_h plot in Fig. 1c, being very resistive
103 when $n_h = 0$. It quickly drops to kilohm level when holes are present, with the exception of the
104 EI phase, where it remains very resistive despite finite n_h . This is because in the EI all the holes
105 spontaneously pair with the electrons into charge-neutral exciton, leaving no unpaired mobile
106 charge.
107

108 The second measurement is the closed-circuit Coulomb drag experiment (Fig. 1g), where a drive
109 current I_{drive} is passed through the MoSe₂ layer, and the drag current in the WSe₂ layer I_{drag} is
110 measured. When all electrons and holes pair into excitons, their motion becomes locked, leading
111 to perfect Coulomb drag behavior. The drag ratio in Fig. 1h, defined as $I_{\text{drag}}/I_{\text{drive}}$, reaches unity
112 in the EI phase (see Extended Data Fig. 3 for raw currents). The large R_h and perfect drag are both
113 unambiguous evidence of the EI phase.

114 **Electron-hole fluids in a strong magnetic field**

115 We next examine the behavior of the e-h bilayer under a strong perpendicular magnetic field B .
116 Fig. 2a-b shows the hole resistance and drag ratio at $B = 12$ T. Compared to the zero-field case,
117 the most noticeable difference is the emergence of quasi-periodic structures as the density varies.
118 In the general hole-doped region, R_h exhibits pronounced QOs. Fig. 2c presents a vertical linecut
119 of R_h at $V_G = 0.1$ V (blue dashed line in Fig. 2a), revealing accurately quantized plateaus at
120 h/Ne^2 (N denotes an integer; h and e are the Planck constant and the elementary charge
121 respectively). Meanwhile, there is negligible drag signal. This behavior is consistent with a typical
122 2DHG in the integer quantum Hall (QH) regime⁴¹, where the two-terminal resistance is quantized
123 to the Hall resistance. We assign the quantized plateaus as integer filling of hole Landau levels
124 (LLs) $\nu_h = 1, 2, \dots, 6$, as labeled in Fig. 2a-b. Measuring the longitudinal resistance R_{xx} and the
125 Hall resistance R_{xy} separately confirms periodically vanishing R_{xx} and quantized R_{xy} (Extended
126 Data Fig. 2). Apparently, metallic 2DHG transport is only weakly influenced by the electrons in
127 the adjacent layer.
128

129

130 Although the interlayer coupling effects seem weak for the general $n_e \neq n_h$ case, the behavior
131 near net charge neutrality $n_e = n_h$ displays strikingly different features. The triangular EI region
132 in Fig. 1f and 1h evolves into a series of lobes centered along the charge neutral line (CNL, black
133 dotted line in Fig. 2b). These lobes are characterized by increased R_h and strong drag signals.
134 Neighboring lobes are connected at low V_B but become separated at higher pair densities. Within
135 each lobe, the drag effect is strongest along the central CNL and decreases with finite e-h
136 imbalance, eventually vanishing. Fig. 2d shows a linecut of R_h and drag ratio approximately
137 tracing constant hole filling factor $\nu_h = 3$ (magenta dashed line in Fig. 2a). An insulating peak in
138 R_h and a large drag ratio are observed in the charge neutral region. Away from charge neutrality,
139 the drag signal quickly diminishes while R_h returns to its quantized value $h/3e^2$, indicating a
140 robust hole QH state disrupted only when electron and hole densities match.
141

142 We now focus on the behaviors along the CNL. Fig. 2e displays the CNL linecut of the drag ratio
143 and R_h . With increasing V_B , the drag ratio starts from a plateau at unity, followed by multiple dips
144 at integer fillings, which deepen progressively until exciton binding is lost at $V_B > 0.46$ V. For
145 small integer filling factors ($\nu = 1 - 4$), the drag signal does not completely vanish at the dips,
146 and R_h deviates from the quantized value. Both indicate LL mixing caused by strong e-h attraction.
147 At the $\nu = 5$ dip, the drag ratio drops to zero while $R_h = h/5e^2$ becomes quantized, suggesting
148 complete exciton destruction where electrons and holes form independent QHIs. Between adjacent
149 QHI phases, strong exciton drag appears at partial LL fillings, with peaks near half-integer fillings.
150 The drag current in partially filled LLs is smaller than but comparable to the drive current. A drag
151 ratio below unity can be explained by the edge channels competing with bulk exciton transport. In
152 addition to exciton flow, currents in both layers can also circulates through edge channels due to
153 finite WSe₂ contact resistances (~2.4 kΩ; Extended Data Fig. 4).
154

155 The QOs and phase transitions can be understood most easily in the strong field limit in which LL
156 mixing is weak. This applies at high densities ($V_B > 0.4$ V), where exciton binding is weakened
157 by many-body effects (Extended Data Fig. 5). In this regime, the electron/hole cyclotron energy
158 $\hbar\omega_c$ exceeds E_b . One can always tune V_B to make the energy gap between the active electron and
159 hole LLs larger or smaller than E_b . As illustrated in Fig. 2f, when the LL gap exceeds E_b , exciton
160 binding is destroyed, resulting in two decoupled QHIs at integer fillings. The e-h fluids form
161 single-Slater-determinant states in which all LLs are either full or empty, and the excitonic order
162 vanishes. Conversely, as V_B tunes the LL gap to be smaller than E_b (Fig. 2g), excitons are
163 energetically favorable and spontaneously form. At half-filling, the ground state is known exactly
164 in the limit of small e-h separation and has excitonic order⁴². The physics at strong magnetic fields
165 mirrors the QH excitons previously observed in QH bilayers made of two electron layers^{7,16-18},
166 except that a net charge of zero here gives no quantized Hall resistance. At lower pair densities,
167 where $E_b > \hbar\omega_c$, excitons always form regardless of e-h LL alignment. In this regime LL gap
168 oscillations cannot fully destroy the EI, but will create a periodic modulation of drag ratio and R_h
169 within the EI phase.
170

171 In the strong field limit, theory also predicts EI states when the electron and hole filling factors
172 ν_e, ν_h differ by non-zero integers²⁵. However, our experiments reveal clear EI signatures only near
173 the CNL. Away from it, drag signals drop below 10%. Previous studies suggest that exciton pairing
174 can be sensitive to the spatial structure of the two interacting LLs, favoring those with similar
175 spatial wavefunctions⁴³. In our density range ($\nu \leq 6$), both MoSe₂ electron and WSe₂ hole LLs are

176 fully valley-polarized^{44,45}. The CNL is unique because the electron and hole LLs share the same
177 orbital index. This may explain the lack of excitonic features for $\nu_e \neq \nu_h$, as they may require
178 substantially stronger field, cleaner samples, and/or lower temperature. A full understanding of
179 imbalanced e-h fluids will require further investigation.

181 Fan diagram at net charge neutrality

182 In the strong field limit, e-h bilayers and previously studied electron-electron bilayers should have
183 identical bulk properties. However, at weaker fields, excitonic order rapidly decays in electron-
184 electron systems, but persists in e-h systems. Fig. 3a shows the B dependence of the drag ratio
185 along the CNL. The NI, EI and EHP phases have distinct drag characteristics. In the NI phase at
186 low V_B , the single-particle gap remains stable against magnetic fields – no drive or drag current is
187 detected at any B . At intermediate V_B , the EI phase yields large drag ratios. With increasing
188 magnetic field, the EI region extends to higher V_B due to increased exciton binding energy⁴⁶.
189 Within the EI, QOs emerge when $B \gtrsim 5$ T, forming Landau-fan-like patterns. In the high-density
190 EHP phase, a weak frictional drag oscillates with B . Corresponding behaviors also appear in the
191 R_h data in Fig. 3b: the NI phase remains resistive, the resistance in the EI phase forms Landau-
192 fan-like structures aligned with drag signals, and the EHP phase develops QH resistance plateaus
193 at high field.

194 Fig. 3c shows three linecuts of R_h and drag ratio at fixed V_B values, plotted against $1/B$. They
195 represent different regimes in the EI phase regarding the relative strength between E_b and $\hbar\omega_c$.
196 The cyclotron energy scales with B , reaching ~ 3 meV at 12 T. The exciton binding energy starts
197 near 20 meV at low density and decreases at higher V_B (Extended Data Fig. 5). At $V_B = 0.35$ V
198 (upper panel), where $E_b \gg \hbar\omega_c$, R_h remains above our 1 M Ω measurement range regardless of B ,
199 and the drag ratio remains unity with no noticeable QOs. For $V_B = 0.41$ V (middle panel) the
200 reduced E_b becomes comparable to $\hbar\omega_c$. Clear QOs appear in both drag ratio and R_h , which are
201 periodic in $1/B$. When $E_b < \hbar\omega_c$ at $V_B = 0.43$ V, larger oscillations touch zero at integer LL
202 filling, indicating reentrant EI-QHI transitions in the quantized regime.

203 The magnetic oscillations in EIs have the same $1/B$ periodicity as conventional magnetic
204 oscillations in metallic states⁴⁷ because both reflect oscillations with period one in LL filling factor,
205 although the mechanism in EIs differs from either Shubnikov-de Haas or de Haas-van Alphen
206 effects. Because QH states are most favored each time the electron and hole filling factors $\frac{nh}{eB}$
207 become an integer, the $1/B$ periodicity is

$$216 \Delta\left(\frac{1}{B}\right) = \frac{e}{nh}.$$

217 The experimentally extracted period agrees with the optically measured carrier densities (Extended
218 Data Fig. 6), supporting our interpretation. In our device geometry the carrier densities in graphite
219 gates are always much higher than n_e and n_h (Extended Data Fig. 7), ruling out the possibility
220 that the observed QOs arise from gate-induced potential modulations⁴⁸, which would produce a
221 very different frequency.

222 Our observation of excitonic QOs and EI-QHI transitions agrees qualitatively with the theoretical
223 predictions^{24,25}. The experimentally observed critical magnetic field, however, is much lower.

220 While gate screening and sample disorder may partly account for this discrepancy, we believe that
221 substantial differences would remain even in ideal conditions. This is likely because of the well-
222 known tendency for mean-field theory to overestimate the robustness of ordered states, particularly
223 in the high-density regime where many-body interactions become important. Improving
224 quantitative agreement between theory and experiment stands as an important challenge.

225 Finite-temperature phase diagram

226

227 Finally, we explore the phase diagram as a function of the pair density and temperature. Fig. 4a
228 shows the temperature dependence of the drag ratio at $B = 12$ T along the CNL. Multiple EI
229 domes emerge at low temperatures, roughly centered around half-integer fillings up to filling factor
230 6. With increased temperature, the drag ratio sharply drops to zero for high-order EI domes, while
231 lower-order ones show smoother transitions (Fig. 4b). The lower exciton melting temperatures at
232 larger filling factors are consistent with reduced binding energy when both electrons and holes are
233 limited to high LLs. The corresponding R_h (Fig. 4c-d) decreases with temperature, confirming the
234 insulating nature of these EI phases.

235

236 Although the QO amplitude decreases monotonically with temperature, it follows two distinct
237 scaling behaviors between the low-temperature EI phase and high-temperature QH liquid phase.
238 In the QH liquid, resistance oscillations in WSe₂ follow the Lifshitz–Kosevich (LK) formula with
239 an effective hole mass of $\sim 0.5 m_0$ (where m_0 denotes the bare electron mass), similar to the
240 behavior of QOs in metallic 2D hole liquids (Extended Data Fig. 8). In contrast, in the low-
241 temperature EI regime the resistance deviates from LK scaling, which is governed by thermal
242 activation of excitons instead. With increasing temperature, more excitons are thermally ionized
243 into highly mobile charges, which reduces the QO amplitude.

244

245 Interestingly, we observe temperature-induced EI-QHI phase transitions at small integer filling
246 factors. As seen in Fig. 4a and Fig. 2e, the drag ratio dips at $\nu = 1 - 4$ does not reach zero at our
247 lowest temperature due to finite LL mixing from Coulomb interactions. Fig. 4e-f shows the
248 temperature dependence of R_h and the drag ratio at $\nu = 3$ and 4. The drag ratio rapidly decreases
249 beyond ~ 5 K for $\nu = 3$ and ~ 2 K for $\nu = 4$. Meanwhile, the deviations of R_h from h/ve^2 also
250 disappear, recovering the quantized values in the QHI. These observations suggest temperature-
251 driven phase transitions, where thermal energy melts the excitons in the EI and restores the QHI
252 phase.

253

254 In summary, we have studied the transport behavior of e-h fluids under magnetic fields and
255 uncovered a rich phase diagram featuring excitonic QOs and reentrant EI-QHI phase transitions.
256 The presence of QOs in our insulating system highlights unconventional insulating phases beyond
257 single-particle band theory. Although QOs in narrow-gap insulators have been reported^{37–40}, their
258 physical origins remain debated in most prior systems^{26–36,48}. With full gate control across different
259 doping phases, optical determination of particle densities in each layer, and separate electrical
260 access to electrons and holes, we conclusively attribute the observed QOs to the EI state,
261 confirming theoretical predictions. These findings establish TMD-based e-h bilayers as a
262 promising platform for further exploration of exciton physics, such as exciton superfluids, vortex
263 lattices⁴⁹, and fractional exciton states.

264

265 **Acknowledgements**

266 This work was primarily funded by the U.S. Department of Energy, Office of Science, Basic
 267 Energy Sciences, Materials Sciences and Engineering Division under Contract No. DE-AC02-05-
 268 CH11231 within the van der Waals heterostructure program KCFW16 (device fabrication and
 269 transport measurements). Optical characterization was supported by the AFOSR award FA9550-
 270 23-1-0246. K.W. and T.T. acknowledge support from the Japan Society for the Promotion of
 271 Science (KAKENHI grants 21H05233 and 23H02052) and World Premier International Research
 272 Center Initiative, Ministry of Education, Culture, Sports, Science and Technology, Japan. R.Q.
 273 acknowledges support from Kavli ENSI graduate student fellowship.

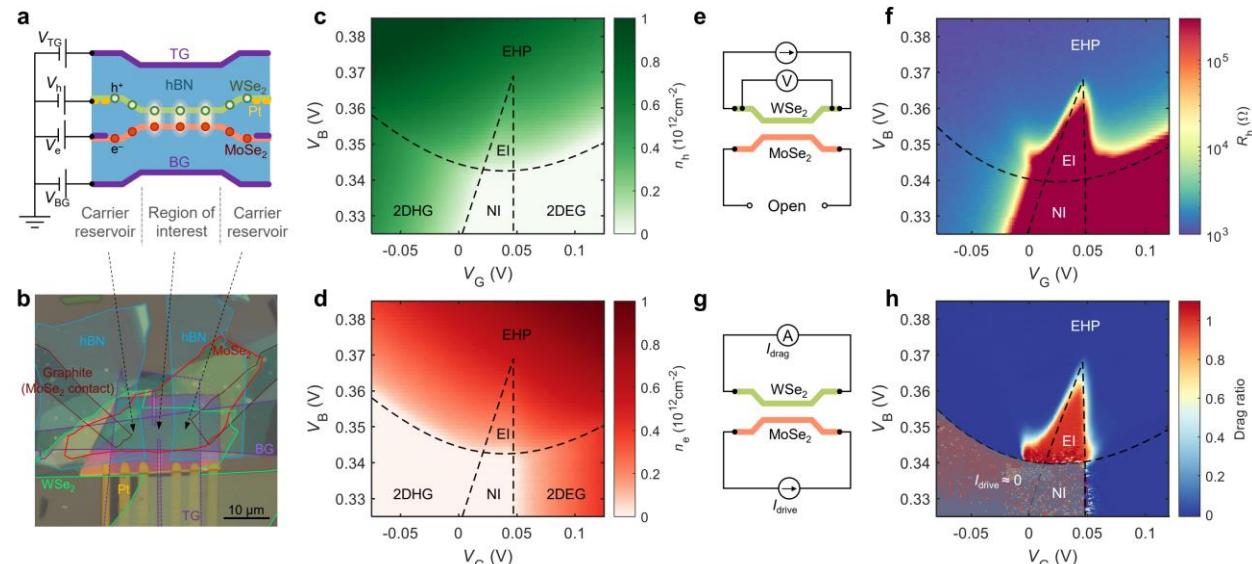
274 **Author contributions**

275 F.W. and R.Q. conceived the research. Q.L., R.Q. and J.N. fabricated the devices with help from
 276 Z.C. and S.C. R.Q. performed the optical measurements. R.Q. and Z.Z. performed the transport
 277 measurements with help from H.K., J.X. and C.S. R.Q., Z.Z. and F.W. analyzed the data with
 278 inputs from M.F.C., B.Z. and A.H.M. K.W. and T.T. grew hBN crystals. All authors discussed the
 279 results and wrote the manuscript.

280 **Competing interests**

281 The authors declare no competing interests.

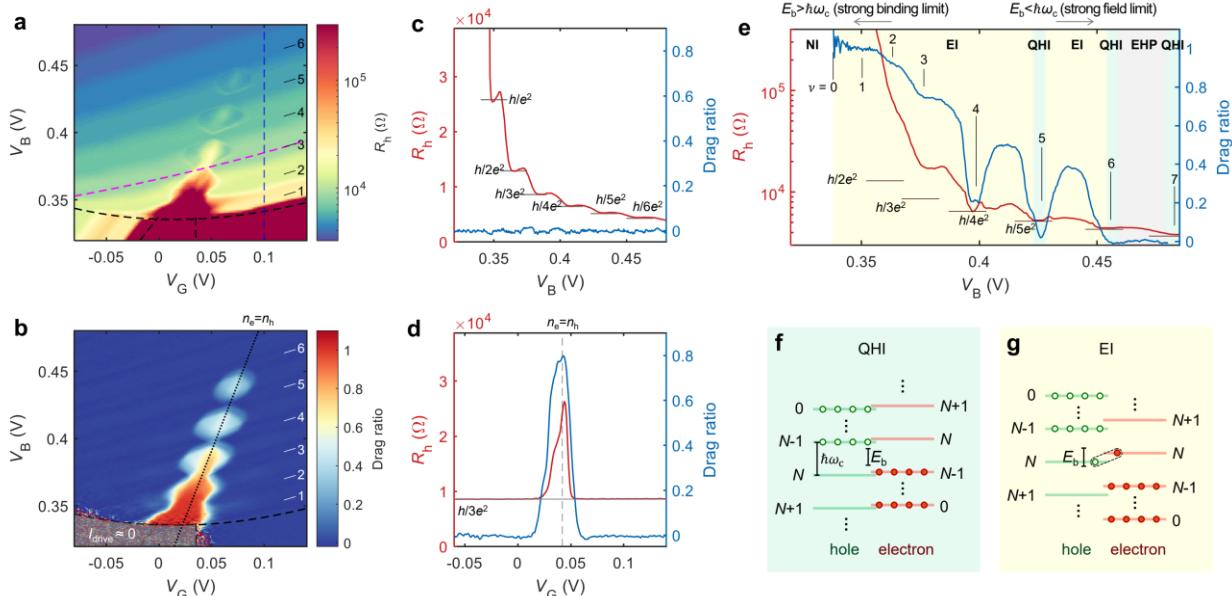
283 **Figures**



285
 286 **Fig. 1 | Strongly coupled electron-hole bilayers.**

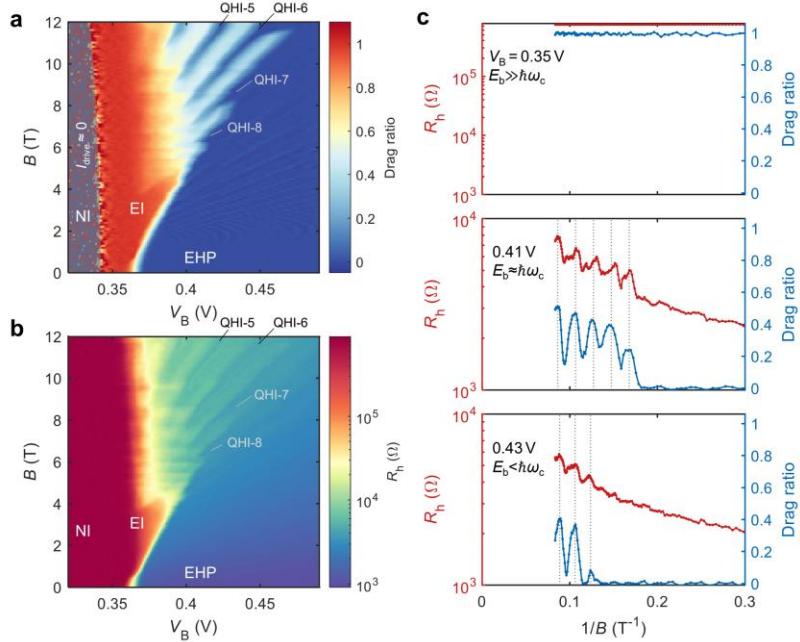
287 **a**, Schematic cross-section of the electron-hole bilayer device based on the MoSe₂/hBN/WSe₂
 288 heterostructure. **b**, An optical microscopy image of device D1 (WSe₂/7-layer hBN/MoSe₂
 289 heterostructure), with flake boundaries outlined. **c-d**, Experimentally determined hole density n_h
 290 in the WSe₂ layer (**c**) and electron density n_e in the MoSe₂ layer (**d**), respectively, measured at
 291 zero magnetic field and a lattice temperature $T = 0.01$ K. The phase diagram is composed of

292 normal insulator (NI), excitonic insulator (EI), two-dimensional electron gas (2DEG), two-
 293 dimensional hole gas (2DHG), and electron-hole plasma (EHP) regions. The vertical electric field
 294 is fixed by antisymmetric gating $V_{BG} - V_{TG} = 7$ V. **e**, Circuit diagram of the two-terminal hole
 295 resistance measurement. The resistance in the WSe₂ layer is measured with the MoSe₂ layer open-
 296 circuited. **f**, Measured hole resistance R_h as a function of V_G and V_B at zero magnetic field. The EI
 297 region exhibits a large R_h despite finite n_h . **g**, Circuit diagram of the closed-circuit Coulomb drag
 298 measurement. A drive current is passed through the MoSe₂ layer, and the closed-circuit drag
 299 current in the WSe₂ layer is measured. **h**, Measured drag ratio $I_{\text{drag}}/I_{\text{drive}}$ as a function of V_G and
 300 V_B at zero magnetic field. Perfect Coulomb drag is observed in the EI region. The bottom-left
 301 region shaded with semitransparent gray color is noisy due to the absence of any drive current
 302 when $n_e = 0$.



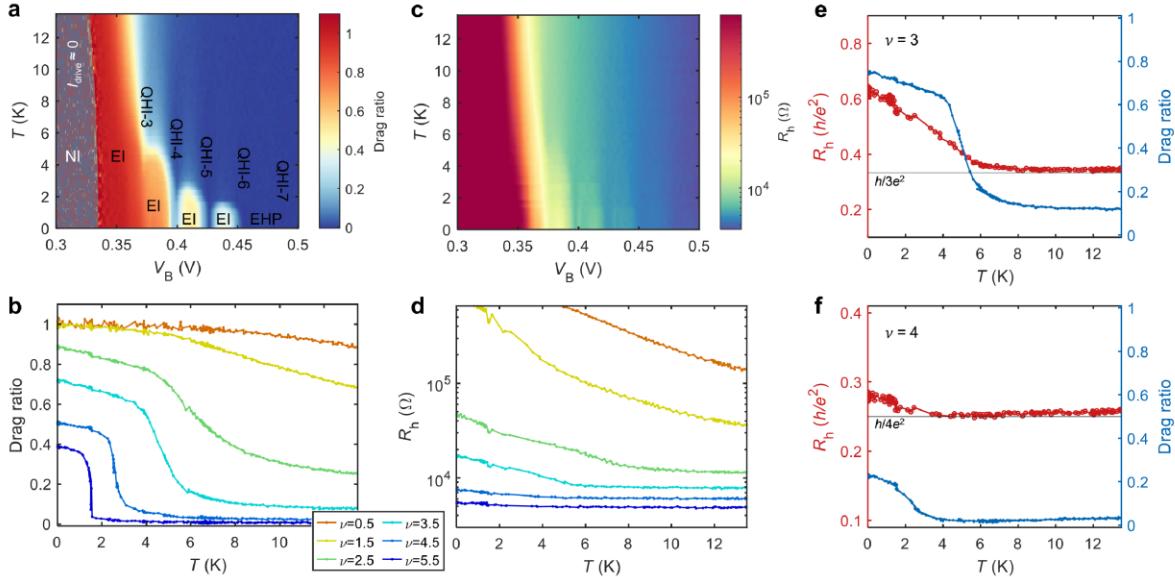
303 **Fig. 2 | Electron-hole fluids in a strong magnetic field.**
 304 **a**, R_h as a function of V_G and V_B at $B = 12$ T. Landau quantization of holes leads to quasi-periodic
 305 structures following n_h contour lines outside the EI region. The EI region develops into a series of
 306 lobes centered near the CNL and exhibits increased R_h values. **b**, Closed-circuit drag ratio
 307 $I_{\text{drag}}/I_{\text{drive}}$ at $B = 12$ T. Strong Coulomb drag is observed in the EI region near the CNL, which
 308 matches the EI region in **a**. Other regions show negligible drag signal. **c**, V_B dependence of R_h (left
 309 axis) and the drag ratio (right axis) at fixed gating $V_G = 0.10$ V (blue dashed line in **a**), where the
 310 electron and hole densities are not equal. At integer hole filling factors, R_h quantizes at h/Ne^2 ,
 311 indicating QH states of holes. **d**, Line cut of R_h (left axis) and the drag ratio (right axis) along fixed
 312 hole LL filling $\nu_h = 3$ (magenta dashed line in **a**). R_h deviates from $h/3e^2$ only near the net
 313 charge neutrality, accompanied by a simultaneous increase in the drag signal. **e**, Line cut of R_h
 314 (left axis) and the drag ratio (right axis) along the net CNL $n_e = n_h$ (black dotted line in **b**). Clear
 315 QOs can be observed in both R_h and drag signals. Multiple EI-QHI phase transitions happen at
 316 higher V_B , where the exciton binding energy and the cyclotron energy are comparable and compete
 317 with each other. The EI regions are characterized by a large drag signal and enhanced R_h , while
 318 the QHI regions exhibit quantized R_h values and very small drag signal. The EI, QHI, and EHP
 319

320 regions are highlighted with yellow, green, and grey shade, respectively. **f-g**, Schematic LL
 321 structures in the bilayer QHI phase (**f**) and the EI phase (**g**) in the strong field limit, $\hbar\omega_c > E_b$.
 322 Depending on the relative size of the LL gap and E_b , excitons can be fully destroyed (QHI phase
 323 in **f**) or spontaneously created (EI phase in **g**).



324
 325 **Fig. 3 | Quantum oscillations at net charge neutrality.**

326 **a**, Closed-circuit drag ratio as a function of perpendicular magnetic field B and interlayer bias
 327 voltage V_B . QOs in the EI phase start to develop at $B \gtrsim 5$ T, and evolve into EI-QHI phase
 328 transitions at higher B . **b**, R_h as a function of B and V_B . The Landau-fan-like structures match with
 329 corresponding features in **a**. **c**, Magnetic field dependence of R_h (left axis) and the drag ratio (right
 330 axis) at three typical V_B values. At small V_B (top panel), no QO is observed due to the large exciton
 331 binding energy at low density. At higher V_B (middle and bottom panels), clear QOs with
 332 $1/B$ periodicity show up in both R_h and drag signals.



333

334 **Fig. 4 | Finite temperature phase diagram at net charge neutrality.**

335 **a**, Drag ratio along the CNL at $B = 12$ T for different V_B and temperatures. Multiple EI domes are
336 centered around half-integer fillings. High-order EI regions are separated by QHI states, while the
337 lower ones are connected. The exciton ionization temperature decreases with the exciton density
338 (i.e. higher V_B). **b**, Linecuts of the drag ratio at different half-integer LL fillings as a function of
339 temperature. **c**, R_h along the CNL at $B = 12$ T for different V_B and temperatures. R_h decreases
340 with increasing temperature in the EI phases, confirming the insulating behavior. **d**, Linecuts of
341 R_h at half-integer LL fillings as a function of temperature. **e-f**, R_h (left axis) and the drag ratio
342 (right axis) at integer LL filling $\nu_e = \nu_h = 3$ (**e**) and 4 (**f**) as a function of temperature. The strong
343 decrease of drag signals and the recovery of R_h quantization at elevated temperature suggests
344 thermally driven EI-QHI phase transitions, in which thermal energy assists the cyclotron energy
345 to dissociate exciton pairing.

346

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464

465 **Methods**

466

467 **Device fabrication.** We use a dry-transfer method based on polyethylene terephthalate glycol
468 (PETG) stamps to fabricate the heterostructures. Monolayer MoSe₂, monolayer WSe₂, few-layer

469 graphene and hBN flakes are mechanically exfoliated from bulk crystals onto SiO_2/Si substrates.
470 We use 6-10 nm hBN as the gate dielectric and 2-3 nm thin hBN as the interlayer spacer. Prior to
471 the stacking process, metal electrodes for the hole layer (7 nm Pt with 3 nm Cr adhesion layer) are
472 defined using photolithography (Durham Magneto Optics, MicroWriter) and electron beam
473 evaporation (Angstrom Engineering) onto a high resistivity SiO_2/Si substrate. The graphite top
474 gate is cut using an atomic force microscope tip⁵⁰ to eliminate undesired current paths in the
475 unmatched WSe_2 monolayer regions.

476 A 0.5 mm thick clear PETG stamp is employed to pick up the flakes at 65-75 °C. A >100 nm thick
477 hBN is first picked up by the stamp to protect the following layers. Then the graphite top gate, the
478 top dielectric hBN, the monolayer WSe_2 , the thin hBN spacer, the two thicker hBN spacers in the
479 reservoir regions, the two graphite contacts, the monolayer MoSe_2 , the bottom dielectric hBN, and
480 the bottom graphite gate are sequentially picked up. The whole stack is released onto the
481 prepatterned Pt electrodes at 100 °C, followed by dissolving the PETG in chloroform for one day.
482 Finally, metal electrodes (5 nm Cr/60 nm Au) are defined using photolithography and electron
483 beam evaporation.

484 **Optical measurements.** All the optical and transport measurements are performed in a dilution
485 refrigerator (Bluefors LD250) with a base lattice temperature of 10 mK. The signal wires are
486 filtered at the mixing chamber flange (QDevil) before reaching the sample. The d.c. voltages on
487 the device are applied with Keithley 2400/2450 source meters or Keithley 2502 picoammeters.
488 The reflection spectroscopy is performed with a supercontinuum laser (YSL Photonics) as the light
489 source. The laser is focused on the sample by a 10× objective (Olympus PLN 10X). The reflected
490 light is collected after a spectrometer by a CCD camera (Andor Shamrock 303i) with 1000 ms
491 exposure time. We use reflection spectroscopy to determine the electron and hole densities shown
492 in Fig. 1c-d and Extended Data Figs. 5-6. A detailed description of the methodology can be found
493 in ref.³.

494 **WSe_2 hole resistance measurements.** The detailed circuit diagram for the two-terminal WSe_2
495 hole resistance measurements is shown in Extended Data Fig. 9a. Good electrical contacts are
496 crucial for accurate two-terminal resistance measurements of QH devices. In our device geometry,
497 the contact resistance includes two parts: the resistance at Pt to WSe_2 junction, and resistance at
498 the reservoir region to region of interest junction. Since we have multiple Pt electrodes in each
499 reservoir region, we exclude the Pt- WSe_2 junction resistance using two separate pairs of Pt
500 electrode for sourcing the current and measuring the voltage. A current excitation at 17 Hz is
501 applied between two WSe_2 electrodes, and the voltage drop between another two electrodes is used
502 to determine the hole layer resistance. However, note that multiple contacts are connected to the
503 same highly doped reservoir region. It does not exclude the contact resistance from the reservoirs
504 to the region of interest. Therefore, the measured R_h should be interpreted as two-terminal
505 resistance in the QH regime⁴¹, where each highly doped reservoir region acts as one terminal.

506 The Pt- WSe_2 contact resistance can be determined by comparing the resistance using two different
507 measurement configurations shown in Extended Data Fig. 4a. In configuration I, all three Pt
508 electrodes in each reservoir region are linked together to source currents and measure the voltage
509 drop. The resulting two-terminal resistance includes the Pt- WSe_2 contact resistance. In
510 configuration II (the configuration used in the main text), different pairs of Pt electrodes are used
511 for current excitation and voltage measurement, excluding the Pt- WSe_2 contact resistance.

512 Extended Data Fig. 4b shows the measured resistance of the two configurations. Their difference
513 is around 2 k Ω and depends weakly on the magnetic field.

514 **Coulomb drag measurements.** Extended Data Fig. 9b shows the detailed circuit diagram of the
515 Coulomb drag measurements. Closed-circuit drag measurements require good electrical contacts
516 for the drag layer. The graphite-MoSe₂ contact resistance is 2-3 M Ω , much larger than the Pt-WSe₂
517 contact. We therefore choose the MoSe₂ layer as the drive layer and the WSe₂ layer as the drag
518 layer. A 5 mV_{rms} a.c. voltage excitation at 17 Hz is applied between the two MoSe₂ contacts. A 10
519 k Ω potentiometer is used to distribute the a.c. voltage between the two contacts to minimize
520 interlayer capacitive coupling. The drag current is measured between the left three WSe₂ electrodes
521 and the right three WSe₂ electrodes.

522 The interlayer tunnelling current is at picoampere level (Extended Data Fig. 10), providing a lower
523 bound of the tunnelling resistance $> 10^{10} \Omega$. This is many orders of magnitude larger than the in-
524 plane resistance, and therefore its contribution to the measured drive and drag currents is negligible.

525

526 **Data availability**

527 All data that support the findings of this study are available from the corresponding author upon
528 request. Source data are provided with this paper.

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531 **Methods-only References**

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