Observation of giant and tunable thermal diffusivity of a Dirac fluid at room temperature

Alexander Block, Alessandro Principi, Niels C. H. Hesp, Aron W. Cummings, Matz Liebel, Kenji Watanabe, Takashi Taniguchi, Stephan Roche, Frank H. L. Koppens, Niek F. van Hulst and Klaas-Jan Tielrooij

Conducting materials typically exhibit either diffusive or ballistic charge transport. When electron-electron interactions dominate, a hydrodynamic regime with viscous charge flow emerges. More stringent conditions eventually yield a quantum-critical Dirac-fluid regime, where electronic heat can flow more efficiently than charge. However, observing and controlling the flow of electronic heat in the hydrodynamic regime at room temperature has so far remained elusive. Here we observe heat transport in graphene in the diffusive and hydrodynamic regimes, and report a controllable transition to the Dirac-fluid regime at room temperature, using carrier temperature and carrier density as control knobs. We introduce the technique of spatiotemporal thermoelectric microscopy with femtosecond temporal and nanometre spatial resolution, which allows for tracking electronic heat spreading. In the diffusive regime, we find a thermal diffusivity of roughly 2,000 cm² s⁻¹, consistent with charge transport. Moreover, within the hydrodynamic time window before momentum relaxation, we observe heat spreading corresponding to a giant diffusivity up to 70,000 cm² s⁻¹, indicative of a Dirac fluid. Our results offer the possibility of further exploration of these interesting physical phenomena and their potential applications in nanoscale thermal management.

During the last few years, signatures of viscous charge flow in the so-called Fermi-liquid hydrodynamic regime were observed in two-dimensional (2D) electron systems, especially graphene, using electrical device measurements and scanning probe microscopy. A second hydrodynamic regime, which has no analogue in classical fluids, can occur very close to the Dirac point. When the Fermi temperature (T_F = E_F/k_B, where E_F is the Fermi energy and k_B is the Boltzmann constant) becomes small compared to the electron temperature T_e, the system becomes a quantum-critical fluid. In this Dirac-fluid regime, the non-relativistic description of the viscous fluid is replaced by its ultra-relativistic counterpart, which accounts for the presence of both particles and holes, as well as for their linear energy dispersion. In line with theoretical predictions in this regime, electrical measurements at cryogenic temperatures indicated a deviation from the Wiedemann–Franz law, and from the Mott relation, and a terahertz-probe study revealed the quantum-critical carrier scattering rate.

Here, we follow electronic heat flow in the diffusive and hydrodynamic regimes at room temperature, and demonstrate a controlled Fermi-liquid to Dirac-fluid crossover, with a strongly enhanced thermal diffusivity close to the Dirac point. These observations are enabled by ultrafast spatiotemporal thermoelectric microscopy, a technique inspired by all-optical spatiotemporal diffusivity measurements, with the crucial difference that the observable is the thermoelectric current, which is directly, and exclusively, sensitive to electronic heat. We use a hexagonal boron nitride (hBN)-encapsulated graphene device that is both a Hall bar for electrical measurements and a split-gate thermoelectric detector (Fig. 1a). Since we use ultrashort laser pulses, with an approximate instrument response time (Δt_{IRF} where IRF means instrument response function) of 200 fs, to generate electronic heat, we are able to examine the system before momentum relaxation occurs, as we measure a momentum relaxation time, τ_m, around 350 fs (Extended Data Fig. 1). In this temporal regime before momentum is relaxed, we enter the hydrodynamic window, because the electron-electron scattering time τ_{ee} is < 100 fs (ref. 27), that is τ_m < Δt_{IRF} < τ_{ee}.

This is a different approach compared to most previous studies, where hydrodynamic effects were observed by using small system dimensions L to eliminate effects of momentum relaxation, that is ν_eτ_e < L < ν_eτ_m (refs. 28,29,30) (ν_e = 10⁶ m s⁻¹ is the Fermi velocity). Our approach furthermore exploits elevated carrier temperatures, which greatly increases the accessibility of the Dirac-fluid regime, as for increasing carrier temperatures the crossover occurs increasingly far away from the Dirac point (Fig. 1b). As we will show, during the hydrodynamic window substantially more efficient heat spreading occurs in the Dirac-fluid regime than in the Fermi-liquid regime and in the diffusive regime (Fig. 1c,d).

Our technique works by using two ultrafast laser pulses that produce localized spots of electronic heat within tens of femtoseconds. These spots are characterized by an increased carrier temperature T_e > T_p with T_p the lattice temperature (300 K). The degree of spatial spreading of these electronic heat spots as a function of time is governed by the diffusivity D. We control the relative spatial and temporal displacement, Δx and Δt, of the two pulses with sub-100-nm spatial precision and roughly 200 fs temporal resolution. Each laser pulse is incident on opposite sides of a pn junction at a distance Δx/2 from the junction. This pn junction is created by applying opposite voltages ±ΔU with respect to the Dirac point voltages to the two backgates that form a split-gate structure. The two photo-generated electronic heat spots spread out spatially and part of the heat can reach the pn junction after a certain amount of time, generating a thermoelectric current at the junction through the Seebeck gradient. The small region of the pn junction thus

1ICFO (Institut de Ciències Fotòniques), The Barcelona Institute of Science and Technology, Castelldefels, Spain. 2Catalan Institute of Nanoscience and Nanotechnology (ICN2), BIST and CSIC, Bellaterra, Spain. 3School of Physics and Astronomy, University of Manchester, Manchester, UK. 4Research Center for Functional Materials, National Institute for Materials Science, Tsukuba, Japan. 5International Center for Materials Nanoarchitectonics, National Institute for Materials Science, Tsukuba, Japan. 6ICREA - Institució Catalana de Recerca i Estudis Avançats, Barcelona, Spain. ✉e-mail: klaas.tielrooij@icn2.cat

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serves as a local probe of the electron temperature. While each of the heat spots can create thermoelectric current independently, we obtain spatiotemporal information by examining exclusively the signal that corresponds to heat generated by one of the pulses interacting with heat generated by the other pulse—the interacting heat current, $\Delta I_{TE}$. Since the thermoelectric photocurrent scales sublinearly with incident power, we can isolate this interacting heat current $\Delta I_{TE}$ by modulating each laser beam at a different frequency, $f_1$ and $f_2$, and demodulating the thermoelectric current at the difference frequency $f_1 - f_2$. As illustrated in Fig. 1c,f, the higher the diffusivity $D$, the more interacting heat current $\Delta I_{TE}$ remains for increasing $\Delta x$ and $\Delta t$.

Figure 2a shows the measured interacting heat current $\Delta I_{TE}$ as a function of $\Delta x$ and $\Delta t$. As expected, the largest $\Delta I_{TE}$ occurs for the largest spatiotemporal overlap at the pn junction ($\Delta x = \Delta t = 0$). For increasing $|\Delta t|$, we find that the normalized signal extends further spatially, indicating the occurrence of heat spreading (Fig. 2b). This spatial spread is quantified via the second moment $<\Delta x^2>$, which quantifies the width of the profile at different time delays (Methods). Similar to recent all-optical spatiotemporal microscopy\cite{22, 23}, we obtain spatial information beyond the diffraction limit by precise spatial sampling of diffraction-limited profiles. The experimentally obtained spatial spread as a function of $\Delta t$ (Fig. 2c) is very similar to the calculated results (Fig. 2d), obtained by simulating the experiment with a given diffusivity, $D$ (Methods and Supplementary Note 1). The white lines indicate the values of the spatial spread $<\Delta x^2>$ for different $\Delta t$. We also compare the simulated spatial spread $<\Delta x^2>$ versus $\Delta t$ (blue dashed line in Fig. 2e) with the theoretical expectation according to the heat diffusion equation, $<\Delta x^2> = <\Delta x^2>_{\text{focus}} + 2D\Delta t$ (dash-dotted line in Fig. 2e).

Here, $D$ is the same diffusivity that was used as input for the simulation, and $<\Delta x^2>_{\text{focus}}$ is the minimum second moment from the two overlapping pulses (Supplementary Note 2 and Supplementary Figs. 1–4). The initial slope is the same for both the simulated heat spreading and the theoretical spreading following the heat diffusion equation.

We first discuss the experimental results in the diffusive regime, where $|\Delta t| > r_{\text{ms}}$. For three different gate voltage combinations, corresponding to Fermi energies between 75 and 190 meV ($T_F = 900–2200$ K), we extract the spatial spread as a function of time delay (symbols in Fig. 2e), and compare it with the results from simulations (solid lines). For these simulations, we have used the diffusivity values that we obtain directly from electrical measurements of charge mobility $\mu$ on the same device (Extended Data Fig. 1), and the relation between mobility and diffusivity: $D = \mu E_0/2e$ (Methods). We find excellent agreement, if we account for short-lived ultrafast heat spreading around $\Delta t = 0$, which leads to a larger-than-expected initial spread at time zero $<\Delta x^2>_{\text{focus}}$ as we will explain below. The agreement between the measured heat spread for $|\Delta t| > r_{\text{ms}}$ and the one calculated using the measured charge mobilities shows that electronic heat and charge flow together, as expected in the diffusive regime. Furthermore, it confirms that our technique is a reliable method for obtaining thermal diffusivities in a quantitative manner.

We now turn to the non-diffusive regime, by exploring the behaviour in the hydrodynamic window, where $|\Delta t| < r_{\text{ms}}$. The experimentally obtained spatial spreads start at a minimum value $<\Delta x^2>_{\text{min}}$ larger than 2 $\mu$m$^2$, rather than starting at an expected $<\Delta x^2>_{\text{focus}} = 0.56\mu$m$^2$. A second device of hBN-encapsulated graphene with similar mobility reproduces this larger-than-expected spatial spread at time zero (Supplementary Note 3 and Extended...
Data Fig. 2). We exclude the possibility of an experimental artefact such as an underestimation of the laser spot size, since we repeated the measurements while scanning through the laser focus, and measured the focus size (Supplementary Figs. 1–4). Furthermore, we observe that the offset depends on the Fermi energy, while keeping all other experimental parameters fixed. Finally, we measured a third device with a lower charge mobility and shorter hydrodynamic time window: \( \tau_{\text{mob}} \approx 100 \text{fs} \), which is smaller than \( \Delta t_{\text{IRF}} \). This device exhibits systematically less heat spreading around time zero (Supplementary Note 4 and Extended Data Fig. 3), consistent with its smaller hydrodynamic time window. We therefore attribute the large experimentally observed minimum \( <\Delta x^2>_{\text{min}} \) in Fig. 2e to ultrafast initial heat spreading that occurs before momentum relaxation takes place, \( \Delta t \lesssim 350 \text{fs} \) (see the schematic illustration of spatiotemporal heat spreading in Fig. 1d). The dynamics of this initial heat spreading are washed out by the finite time resolution \( \Delta t_{\text{IRF}} \) and manifests as a large minimum \( <\Delta x^2>_{\text{min}} \) at time zero. The observed initial spatial spread suggests a thermal diffusivity of \( D = ( <\Delta x^2>_{\text{min}} - <\Delta x^2>_{\text{focus}} >) / 2 \Delta t_{\text{IRF}} \approx 70,000 \text{cm}^2 \text{s}^{-1} \) for the lowest measured \( E_F \) of 75 meV. Simulations of heat spreading with an input diffusivity of 100,000 cm\(^2\) s\(^{-1}\) are indeed consistent with the experimentally observed spread in the hydrodynamic window (the red line in Fig. 2c).

We attribute this observation of highly efficient initial heat spreading to the presence of the quantum-critical electron-hole plasma. We can exclude that the observed initial spreading is the result of ballistic transport, as we calculate that the ballistic contribution to initial heat spreading would give only \( <\Delta x^2>_{\text{ball}} \approx 0.68 \mu\text{m}^2 \) (Supplementary Note 2 and Extended Data Fig. 4). Besides, ballistic transport has a very weak dependence \((<10\%)\) on carrier density in this range, as the Fermi velocity does not change appreciably for the Fermi energies considered here\(^{29}\). The reason for the high diffusivity in the Dirac-fluid regime is that the hot electrons and hot holes that coexist in this regime move in the same direction under a thermal gradient, with inter-particle scattering events conserving total momentum\(^{19}\). We note that typical transport measurements probe...
the sum of the momentum-conserving thermal resistivity (due to carrier–carrier scattering) and the momentum-non-conserving thermal resistivity (due to carrier–impurity and carrier–photon scattering), where the latter dominates at room temperature. The ability of our technique to interrogate the system during the 350 fs before any momentum-non-conserving scattering occurs, means that this contribution to the overall resistivity is negligible. Therefore, we probe exclusively the momentum-conserving
thermal conductivity, which diverges towards the Dirac point and towards infinite electron temperature. To provide further evidence of hydrodynamic heat transport, we demonstrate the ability to control the crossover between the Fermi-liquid and quantum-critical Dirac-fluid regimes via the ratio $T_d/T_F$ by independently varying $T_d$ via the incident laser power and $T_F$ via the applied gate voltage. A larger ratio results in less Coulomb screening and correspondingly stronger hydrodynamic effects due to electron–electron interactions. If $T_d$ is larger than $T_F$, electrons and holes coexist and the Dirac-fluid regime becomes accessible (Fig. 1b). We perform spatial scans in the hydrodynamic window at a temporal delay of $\Delta t = 0$, in a geometry with one laser pulse impinging on the junction, while scanning the other pulse across (x axis) and along (y axis) the junction region. Figure 3a–d shows four representative spatial $\Delta I_{TE}$ maps with varying $T_d/T_F$, yet similar signal magnitudes. Clearly, the signal is broader for larger $T_d/T_F$, indicating faster thermal transport. We repeat these measurements for a range of $T_d$ and $T_F$ values and quantify the initial heat spreading using Gaussian functions, with widths $\sigma_T$ and $\sigma_d$, to describe $\Delta I_{TE}$ at $\Delta t = 0$ as a function of $\Delta x$ or $\Delta y$ (Fig. 3c,f and Supplementary Fig. 5). As expected for a crossover from the diffusive Fermi-liquid regime to the hydrodynamic Dirac-fluid regime, both spatial spreads $\sigma_T$ and $\sigma_d$ increase substantially for increasing ratio $T_d/T_F$. These spreads correspond to a diffusivity up to 40,000 cm$^2$ s$^{-1}$ (Methods), similar to the 70,000 cm$^2$ s$^{-1}$ we found earlier.

We compare our experimental results to Boltzmann transport calculations following refs. 17,18, including carrier interactions and long-range impurity scattering. We model impurities as Thomas–Fermi screened Coulomb scatterers of density $0.24 \times 10^{12}$ cm$^{-2}$. Figure 3g shows the calculated thermal diffusivity $D$ as a function of $T_d$ and $T_F$ when considering only the hydrodynamic term due to electron–electron interactions, relevant in the hydrodynamic window where $|\Delta t| < t_{sr}$. A higher electron temperature, or lower Fermi temperature, leads to strongly increased diffusivity, signaling a crossover from the Fermi-liquid to the Dirac-fluid regime. This is the same qualitative trend as for the experimental data taken at $\Delta t = 0$ in Fig. 3e–f, where a larger initial width originates from a larger diffusivity, thus supporting our interpretation of a hydrodynamic crossover.

A more quantitative comparison shows that the calculated $D$ in the diffusive regime is around 2,000 cm$^2$ s$^{-1}$ (Fig. 3h), in quantitative agreement with the experiment in the diffusive regime. The obtained thermal diffusivity in the hydrodynamic window close to the Dirac point reaches values above 100,000 cm$^2$ s$^{-1}$, even higher than our experimental estimates of 35,000–70,000 cm$^2$ s$^{-1}$. Using the calculated diffusivities, we estimate the spatial spread at time zero $\sigma_{d,0}$ (Methods), as shown in Fig. 3g. These are similar to the experimentally obtained ones, thus confirming our conclusion of highly efficient heat spreading in the Dirac-fluid regime at room temperature, with a diffusivity that is almost two orders of magnitude larger than in the diffusive regime. We note that the theoretical calculations predict that even higher diffusivities are attainable.

Finally, we discuss the three-dimensional (3D) thermal conductivity, to assess the ability to transport useful amounts of heat. We find roughly 100 W m$^{-1}$ K$^{-1}$ in the diffusive regime (Methods), in agreement with ab initio calculations. In the Dirac-fluid regime, with an electron temperature of roughly 1,000 K, we obtain a thermal conductivity of 18,000–40,000 W m$^{-1}$ K$^{-1}$. This is in agreement with ref. 19, where values up to 100,000 W m$^{-1}$ K$^{-1}$ were predicted theoretically for large $T_d/T_F$. The thermal conductivity we obtain is about three orders of magnitude larger than the one obtained in the Dirac-fluid regime at cryogenic temperatures20. Our results show that in the Dirac-fluid window the electronic contribution to heat transport can be much larger than the phononic contribution with a conductivity of >2,000 W m$^{-1}$ K$^{-1}$ (ref. 20), which is already exceptionally high and can also be enhanced hydrodynamically, as shown recently15. Thus, the Dirac electron-hole plasma can contribute strongly to thermal transport, extracting heat from hot spots much faster than predicted by classical limits.

In conclusion, our results show that the—until recently unachievable—physical phenomena associated with the Dirac fluid do not only offer an exciting playground for interesting physical phenomena, but also hold great promise for applications, for example in thermal management of nanoscale devices. We note that the quantum-critical behaviour can be switched on and off using a modest gate voltage and in systems prepared by standard fabrication techniques. Finally, we believe that the optoelectronic technique we have introduced—with the potential of increased spatial accuracy and temporal resolution—will be a valuable tool to reach a better understanding of the thermal behaviour of a broad range of quantum materials, with great promise for new technological applications.

Online content
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References
1. Polini, M. & Geim, A. K. Viscous electron fluids. Phys. Today 73, 28 (2020).
2. Müller, M., Schmalian, J. & Fritz, L. Graphene: a nearly perfect fluid. Phys. Rev. Lett. 103, 025501 (2009).
3. Foster, M. S. & Aleiner, I. L. Slow imbalance relaxation and thermoelectric transport in graphene. Phys. Rev. B. 79, 085415 (2009).
4. Narozhny, B. N., Gornys, I. V., Titov, M., Schütz, M. & Mirlin, A. D. Hydrodynamics in graphene: linear-response transport. Phys. Rev. B. 91, 035414 (2015).
5. Levitov, L. & Falkovich, G. Electron viscosity, current vortices and negative nonlocal resistance in graphene. Nat. Phys. 12, 672–676 (2016).
6. Zarenia, M., Principe, A. & Vignale, G. Disorder-enabled hydrodynamics of charge and heat transport in monolayer graphene. 2D Mater. 6, 035024 (2019).
7. Moll, P. J. W., Kushwaha, P., Nandi, N., Schmidt, B. & Mackenzie, A. P. Evidence for hydrodynamic electron fluid flow in PdCoO$_2$. Science 351, 1061–1064 (2016).
8. Bandurin, D. A. et al. Negative local resistance caused by viscous electron backflow in graphene. Science 351, 1055–1058 (2016).
9. Krishna Kumar, R. et al. Superballistic flow of viscous electron fluid through graphene constrictions. Nat. Phys. 13, 1182–1185 (2017).
10. Braem, B. A. et al. Scanning gate microscopy in a viscous electron fluid. Phys. Rev. B. 98, 241304 (2018).
11. Goeth, J. et al. Thermal and electrical signatures of a hydrodynamic electron fluid in tungsten diphosphide. Nat. Commun. 9, 4093 (2018).

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12. Berdyugin, A. I. et al. Measuring hall viscosity of graphene’s electron fluid. Science 364, 162–165 (2019).
13. Sulpizio, J. A. et al. Visualizing Poiseuille flow of hydrodynamic electrons. Nature 576, 75–79 (2019).
14. Sheehy, D. E. & Schmalian, J. Quantum critical scaling in graphene. Phys. Rev. Lett. 99, 226803 (2007).
15. Xie, H. Y. & Foster, M. S. Transport coefficients of graphene: interplay of impurity scattering, Coulomb interaction, and optical phonons. Phys. Rev. B. 93, 195103 (2016).
16. Lucas, A., Crossno, J., Fong, K. C., Kim, P. & Sachdev, S. Transport in inhomogeneous quantum critical fluids and in the Dirac fluid in graphene. Phys. Rev. B. 93, 075426 (2016).
17. Lucas, A. & Fong, K. C. Hydrodynamics of electrons in graphene. J. Phys. Condens. Matter 30, 053001 (2018).
18. Zarenia, M., Smith, T. B., Principi, A. & Vignale, G. Breakdown of the Wiedemann–Franz law in AB-stacked bilayer graphene. Phys. Rev. B 99, 161407 (2019).
19. Crossno, J. et al. Observation of the Dirac fluid and the breakdown of the Wiedemann–Franz law in graphene. Science 351, 1058–1061 (2016).
20. Ghahari, F. et al. Enhanced thermoelectric power in graphene: violation of the Mott relation by inelastic scattering. Phys. Rev. Lett. 116, 136802 (2016).
21. Gallagher, P. et al. Quantum-critical conductivity of the Dirac fluid in graphene. Science 364, 158–162 (2019).
22. Ku, M. J. H. et al. Imaging viscous flow of the Dirac fluid in graphene. Nature 583, 537–541 (2020).
23. Ruzicka, B. A. et al. Hot carrier diffusion in graphene. Phys. Rev. B. 82, 195414 (2010).
24. Zhu, T., Snaider, J. M., Yuan, L. & Huang, L. Ultrafast dynamic microscopy of carrier and exciton transport. Annu. Rev. Phys. Chem. 70, 219–244 (2019).
25. Block, A. et al. Tracking ultrafast hot-electron diffusion in space and time by ultrafast thermomodulation microscopy. Sci. Adv. 5, eaav8965 (2019).
26. Gabor, N. M. et al. Hot carrier-assisted intrinsic photoresponse in graphene. Science 334, 648–652 (2011).
27. Brida, D. et al. Ultrafast collinear scattering and carrier multiplication in graphene. Nat. Commun. 4, 1987 (2013).
28. Staub, T. et al. Interacting electrons in graphene: Fermi velocity renormalization and optical response. Phys. Rev. Lett. 118, 266801 (2017).
29. Kim, T. Y., Park, C. H. & Marzari, N. The electronic thermal conductivity of graphene. Nano Lett. 16, 2439–2443 (2016).
30. Balandin, A. A. et al. Superior thermal conductivity of single-layer graphene. Nano Lett. 8, 902–907 (2008).
31. Machida, Y., Matsumoto, N., Isono, T. & Behnia, K. Phonon hydrodynamics and ultrahigh–room-temperature thermal conductivity in thin graphite. Science 367, 309–312 (2020).

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Methods
Fabrication of split-gate thermoelectric device. The split-gate device with Hall geometry consists of exfoliated, single layer graphene encapsulated by hBN, prepared using standard exfoliation and dry transfer techniques. The hBN-graphene-hBN stack is placed on a predefined split-gate structure made of graphene, grown by chemical vapour deposition, where the gap between the two gates is roughly 100 nm, created via electron-beam lithography and reactive ion etching (RIE). The top hBN and graphene are etched into a Hall bar shape with laser lithography and RIE, keeping the split-gate intact and not etching completely through the bottom hBN. Finally, the Ti/Au side contacts are created by a further step of lithography, RIE and metal evaporation. The fabrication steps are shown in Supplementary Fig. 6.

Spatiotemporal thermoelectric current microscopy setup. Our setup enables us to follow electronic heat spreading in space and time, because we use the thermoelectric signal generated by electronic heat interacting at a fixed location (the pn junction), while we vary the spatial displacement of our two laser pulses with respect to this junction and vary the temporal delay between the two ultrashort pulses. This means that we are following in space and time the diffusion of light-induced electronic heat from the location of light incidence to the pn junction. It is the thermoelectric effect at the pn junction, governed by the Seebeck coefficient, that generates our observable signal, the thermoelectric current. We note that although the value of the Seebeck coefficient itself changes when changing the pn junction, which is what we are following with our spatiotemporal technique. Nevertheless, that generates our observable signal, the thermoelectric current. We collect the thermoelectric (TE) photocurrent between the source and drain with respect to the beams is controlled with a piezo scanning stage. The beams with optical pulses incident at a distance Δx on each side of the pn-junction (Figs. 1 and 2). For each Δt of the datasets ΔI_{TE}(Δx, Δt) we calculate the width of the signal via the second moment, which for an ideal Gaussian profile is equal to the squared Gaussian width σ. The second moment is calculated from the pixels Δx, (i=1,..,N) via

<Δx^2> > (Δt) = \sum_{i=1}^{N} Δx_i^2 \Delta I_{TE}(Δx_i, Δt),

with the mean

Δx = \frac{\sum_{i=1}^{N} Δx_i \Delta I_{TE}(Δx_i, Δt)}{\sum_{i=1}^{N} \Delta I_{TE}(Δx_i, Δt)}.

We note that the minimum second moment at the focus <Δx^2>_{focus} of 0.56 μm^2 comes from simulating the symmetric experiment, using as input the measured Gaussian beam width at the focus σ_{focus} = 0.14 μm (Supplementary Note 2). For the ‘asymmetric experiment’ with one optical pulse always incident on the pn junction (data for Fig. 3), we always consider the spatial profile only at time zero. Here we find that Gaussian fits with a background give the most reliable results. The entire set of data is shown in Supplementary Fig. 5. For each dataset ΔI_{TE}(Δx, Δt) or ΔI_{TE}(Δy) taken at Δt = 0, we perform Gaussian fits using the function I(Δx) = a \exp \left(-\frac{Δx^2}{2 σ^2}\right) + b, where the Gaussian squared width σ indicates the thermal spreading. Here, the minimum simulated Gaussian widths are (σ_{gwidth}) = 0.34 μm and (σ_{gwidth}) = 0.44 μm (Supplementary Note 2).

Methods. We characterize our device electrically with four-probe measurements (Extended Data Fig. 1), finding a charge mobility μ of 30,000–50,000 cm^2 V\textsuperscript{-1} s\textsuperscript{-1}, depending on carrier density. The measured mobilities correspond to a momentum relaxation time τ_\text{rel} of 300–500 fs. These relaxation times are longer than the temporal resolution (the IRF) of our measurement technique, Δt = 200 fs, thus allowing us to probe our system before and after momentum relaxation occurs, that is in the non-diffusive and diffusive regime. We use the measured charge mobilities to calculate the expected thermal diffusivity via the Einstein relationμ = \frac{h_v^2}{2 \pi c_e e}, via κ = \frac{\mu_\text{rel}}{\tau_\text{rel}}, leads to the simple relation: D = \frac{\mu_\text{rel}}{\tau_\text{rel}}. We note that we obtain the identical result by calculating D from the ratio of the 2D thermal conductivity k_{\text{2D}} and the electronic heat capacity C_e, using the Wiedemann–Franz law: k_{\text{2D}} = \frac{\pi^2}{3} \frac{k_B T}{\sigma_e} C_e, where T = the Fermi energy is the electron/hole carrier density. For highly doped graphene (E_F > k_B T), the carrier density expression n_{ph} = \frac{|e|}{2 \pi \hbar (E_F^2 - E_F^2)^{1/2}} leads to the simple relation: D = \frac{\mu_\text{rel}}{\tau_\text{rel}}. Given the measured mobilities, we expect thermal diffusivities around 2,000 cm^2 s\textsuperscript{-1} for our sample.

Thermal diffusivity and conductivity of the Dirac fluid. We estimate the enhanced thermal diffusivity of the Dirac fluid by comparing the measured width at time zero <Δx^2>_{focus} to the expected width <Δx^2>_\text{theory} explained above, via Δ = (\bar{α}Δx^2)_{calc} = (\bar{α}Δx^2)_{focus}/(\sigma^2). We find values up to 74,000 cm^2 s\textsuperscript{-1} for the symmetric scan (Fig. 2), and 29,000 and 39,000 cm^2 s\textsuperscript{-1} for the x and y directions of the asymmetric scan (Fig. 3), respectively, where <Δx^2>_{focus} is replaced by (σ_e^2) and (σ_h^2). The same calculation for a second device (Supplementary Note 3 and Extended Data Fig. 2) gives a diffusivity of 10,000 cm^2 s\textsuperscript{-1}. The 3D thermal conductivity of Dirac fluid is calculated from the diffusivity D and the electronic heat capacity C_e via κ_{\text{2D}} = DC_e/d, where d is the thickness of graphene, 0.3 nm. For the Dirac fluid, we have τ_\text{rel} > τ_\text{rel} and therefore use the ‘undoped’ electronic heat capacity κ_e = \frac{\hbar v^2}{m_e d} \frac{C_e}{d}, where v (3) = 1.202. With the above estimate D = 35,000–70,000 cm^2 s\textsuperscript{-1} and τ_\text{rel} = 1,000 K, we obtain the 3D thermal conductivity κ_{\text{2D}} = 18,000–40,000 W m\textsuperscript{-1} K\textsuperscript{-1}. This corresponds to a 2D κ_{\text{2D}} = 5 μW K\textsuperscript{-1}. This value is orders of magnitude larger than the value found in ref. 18. The reason for this is that our electron temperature is more than ten times higher, and therefore the electronic heat capacity is 100× higher. Furthermore, we reach a 2D κ_{\text{2D}} > 3, while their maximum is τ/T = 2, which means that we are further in the Dirac-fluid regime with its diverging thermal diffusivity.

Dirac-fluid crossover temperature. Following the treatment in ref. 18, we find the crossover temperature from Fermi liquid to Dirac fluid, as a function of Fermi temperature as

T_{\text{cross}} (T_F) = T_F \left(1 + \ln \left(\frac{T_F}{T_F^c}\right)\right),

with only one pulse at a time, analogous to the experimental difference-frequency demodulation.

Quantifying the spatial spread. The following analysis is conducted both on the experimental and the simulated data of ΔI_{TE}(Δx, Δt) for ‘symmetric experiments’ with optical pulses incident at a distance Δx on each side of the pn-junction (Figs. 1 and 2). For each Δt of the datasets ΔI_{TE}(Δx, Δt), we calculate the width of the signal via the second moment, which for an ideal Gaussian profile is equal to the squared Gaussian width σ. The second moment is calculated from the pixels Δx, (i=1,..,N) via

<Δx^2> > (Δt) = \sum_{i=1}^{N} Δx_i^2 (ΔI_{TE}(Δx_i, Δt)),

with the mean

Δx = \frac{\sum_{i=1}^{N} Δx_i (ΔI_{TE}(Δx_i, Δt))}{\sum_{i=1}^{N} (ΔI_{TE}(Δx_i, Δt))}.

We note that the minimum second moment at the focus <Δx^2>_{focus} of 0.56 μm^2 comes from simulating the symmetric experiment, using as input the measured Gaussian beam width at the focus σ_{focus} = 0.14 μm (Supplementary Note 2). For the ‘asymmetric experiments’ with one optical pulse always incident on the pn junction (data for Fig. 3), we always consider the spatial profile only at time zero. Here we find that Gaussian fits with a background give the most reliable results. The entire set of data is shown in Supplementary Fig. 5. For each dataset ΔI_{TE}(Δx, Δt) or ΔI_{TE}(Δy) taken at Δt = 0, we perform Gaussian fits using the function (Δx) = a \exp \left(-\frac{Δx^2}{2 σ^2}\right) + b, where the Gaussian squared width σ indicates the thermal spreading. Here, the minimum simulated Gaussian widths are (σ_{gwidth}) = 0.34 μm and (σ_{gwidth}) = 0.44 μm (Supplementary Note 2). The axially obtained widths from this dataset as function of gate voltage and optical power are also shown in Supplementary Fig. 5, showing an increase with power, that is a larger T, and an increase towards the Dirac point, that is a smaller T. We estimate the theoretical Gaussian widths in Fig. 3a using (σ_{calc}) = (σ_{gwidth}) = 2DΔx_0, where D are the calculated diffusivities.
where $\lambda = \frac{e^2}{16\varepsilon_0\varepsilon_r v_F h} \approx 0.55/\varepsilon_r$ for graphene with the dielectric environment $\varepsilon_r \approx 3.56$ for hBN. The temperature $T_0 = \frac{2\hbar v_F}{\lambda e^2} \approx 8.4 \times 10^4$ K, with the inter-atomic distance $a_0 = 1.42 \times 10^{-10}$ m. The resulting crossover temperature is shown in Fig. 1b. We note that the relatively high refractive index of the hBN encapsulant makes the Dirac fluid more easily accessible, as it lowers the crossover temperature compared to vacuum, by a factor of about two for the range of $T_F$ values studied here.

**Data availability**

The data that support the findings of this study are available from the corresponding author on reasonable request.

**References**

32. Tielrooij, K. J. et al. Out-of-plane heat transfer in van der Waals stacks through electron-hyperbolic phonon coupling. *Nat. Nanotechnol.* **13**, 41–46 (2018).

33. Zebrev, G. I. in *Physics and Applications of Graphene—Theory* (ed. Mikhailov, S.) 475–498 (IntechOpen, 2011).

34. Rengel, R. & Martin, M. J. Diffusion coefficient, correlation function, and power spectral density of velocity fluctuations in monolayer graphene. *J. Appl. Phys.* **114**, 143702 (2013).

35. Lui, C. H., Mak, K. F., Shan, J. & Heinz, T. F. Ultrafast photoluminescence from graphene. *Phys. Rev. Lett.* **105**, 127404 (2010).

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**Author contributions**

K.-J.T. conceived and supervised the project. A.B. developed the experimental setup under supervision of M.L. and N.F.v.H. and input from K.-J.T. N.C.H.H. performed sample fabrication, with material input from K.W. and T.T., under the supervision of F.H.L.K. and K.-J.T. A.B. performed the measurements under the supervision of K.-J.T. A.P. performed the theoretical hydrodynamic transport calculations. A.B. and K.-J.T. interpreted and analysed the data, with input from M.L., A.W.C. and N.F.v.H. A.B. developed the model that simulated the experiment with input from K.-J.T. A.W.C. and S.R. developed the ballistic transport model. A.B. and K.-J.T. wrote the paper, with input from all authors.

**Competing interests**

The authors declare no competing interests.

**Additional information**

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**Supplementary information**

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**Correspondence and requests for materials**

Correspondence and requests for materials should be addressed to K.-J.T.

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Extended Data Fig. 1 | Electrical mobility measurement. Momentum relaxation time from four-probe measurements and corresponding (calculated) heat diffusivity (solid line). Four-probe measurements were performed by applying 1 V to a MΩ series resistor, such that a current of 1 μA flows between the two outer contacts of the device (see inset). We then measure the voltage drop between two lateral contacts of the graphene device. This yields the sheet conductance $\sigma$ as a function of gate voltage. We then use $\sigma = ne\mu$, in order to extract the mobility $\mu$ and use $\tau_{mr} = \frac{m*}{e^2}$ to obtain the momentum relaxation time. Here, $n$ is the carrier density, $E_F$ is the Fermi energy, $v_F \approx 10^6$ m/s is the Fermi velocity, and $e$ is the elementary charge. We measured up to $E_F = 150$ meV, and extrapolated the data to higher Fermi energies. The three symbols indicate the Fermi energy and predicted diffusivity corresponding to the diffusivity measurements in Fig. 2e.
Extended Data Fig. 2 | See next page for caption.
Extended Data Fig. 2 | Spatiotemporal results from a second device. a, Asymmetric spatiotemporal $\Delta I_{TE}$ maps for three different gate voltages. b, Extracted width as a function of $\Delta t$. A lower Fermi level leads to a higher time-zero width, in accordance with hydrodynamic transport, as presented for the main device in the manuscript. (c) $\Delta I_{TE}$ maps, taken at $\Delta t = 0$, as a function of beam offset ($\Delta x$, $\Delta y$), as well as sample height ($z$). d, extracted line profiles for the two dimensions. e, Resulting signal width $\sigma^2$ for both dimensions as extracted from Gaussian fits at each z-position. The same measurements are presented in Supplementary Fig. 4 for the main device. These experiments were performed with two beams of wavelength 443 nm and 886 nm, respectively.
Extended Data Fig. 3 | Spatiotemporal results from a third device. a, Microscope image of the device. This sample has split-gates made from graphite, with a gap that is 200 nm. It then has a 30 nm thick layer of SiO$_2$, and then we transferred a graphene flake grown by chemical vapour deposition (CVD) on top of the split-gate structure using an hBN flake. b, Gate-dependent current measurement, which gives an estimated mobility of this half-encapsulated CVD graphene sample of around 8,500 cm$^2$/Vs (solid lines). c–d, Asymmetric spatiotemporal $\Delta I_{\text{TE}}$ maps at time zero for different gate voltages and laser powers without (c) and with normalization (d). e–f, Comparison between first and third device. Time zero Gaussian widths for spatial scans with one pulse on the junction and the second one scanning across (e) and along (f) the graphene pn-junction, as a function of power and gate voltage, for both the first device (hBN-encapsulated with high mobility, presented in the main text, blue-purple colours, ‘hBN’) and the third device (on SiO$_2$ with low mobility, yellow-red colors, ‘SiO$_2$’). The low-mobility sample with shorter hydrodynamic time window shows systematically less heat spreading around time zero, in agreement with our picture of hydrodynamic heat spreading during the hydrodynamic time window.
Extended Data Fig. 4 | Ballistic spreading simulation. a, Time-dependent output distributions for quantum mechanical calculations for a single electron (top row) and an ensemble of independent electrons (bottom row). b, Resulting width $\sigma_{ball}^2$ for ballistic transport for the Monte Carlo method with varying Fermi velocities, as well as the quantum calculation for an ensemble of independent electrons. Both calculations essentially agree and the spread within 0.25 ps leads to final widths of below 0.25 $\mu$m$^2$ for realistic values of the Fermi velocity.