Observation of hydrodynamic plasmons and energy waves in graphene

https://doi.org/10.1038/s41586-022-05619-8

Received: 30 June 2022

Accepted: 2 December 2022

Published online: 22 February 2023

Check for updates

Wenyu Zhao^{1,8}, Shaoxin Wang^{1,2,8}, Sudi Chen^{3,4}, Zuocheng Zhang¹, Kenji Watanabe⁵, Takashi Taniguchi⁶, Alex Zettl^{1,3,4,7} & Feng Wang^{1,3,4,7}

Thermally excited electrons and holes form a quantum-critical Dirac fluid in ultraclean graphene and their electrodynamic responses are described by a universal hydrodynamic theory. The hydrodynamic Dirac fluid can host intriguing collective excitations distinctively different from those in a Fermi liquid¹⁻⁴. Here we report the observation of the hydrodynamic plasmon and energy wave in ultraclean graphene. We use the on-chip terahertz (THz) spectroscopy technique to measure the THz absorption spectra of a graphene microribbon as well as the propagation of the energy wave in graphene close to charge neutrality. We observe a prominent highfrequency hydrodynamic bipolar-plasmon resonance and a weaker low-frequency energy-wave resonance of the Dirac fluid in ultraclean graphene. The hydrodynamic bipolar plasmon is characterized by the antiphase oscillation of massless electrons and holes in graphene. The hydrodynamic energy wave is an electron-hole sound mode with both charge carriers oscillating in phase and moving together. The spatial-temporal imaging technique shows that the energy wave propagates at a characteristic speed of $V_{\rm E}/\sqrt{2}$ near the charge neutrality²⁻⁴. Our observations open new opportunities to explore collective hydrodynamic excitations in graphene systems.

Ultraclean graphene provides an attractive platform to explore Dirac-fluid physics, in which relativistic electrons and holes form a strongly interacting plasma. The electron-electron scattering can dominate over electron-impurity and electron-phonon scatterings in graphene over a substantial temperature range⁵, and the manybody system can be well described by a relativistic hydrodynamic theory^{1,3,4,6-10}. Electrical-transport signatures of the hydrodynamic Dirac fluid in graphene have been observed experimentally, including the hydrodynamic electron flow¹¹⁻¹⁶, the breakdown of the Wiedemann-Franz law¹⁷ and the quantum-critical conductivity¹⁸. The hydrodynamic Dirac fluid is also predicted to host intriguing collection excitations, such as hydrodynamic bipolar plasmons and energy waves^{1-3,6-9,19-25}. These collective excitations are governed by the strong electronelectron scattering and do not depend on the microscopic details of interactions^{2,26}. The hydrodynamic plasmon is a coupled electromagnetic and electron-hole collective excitation with opposite motion of electrons and holes (see Methods). The bipolar nature of this excitation makes it distinct from the conventional monopolar plasmons with only one type of carriers. The hydrodynamic energy wave, also known as the demon mode, is a sound-like mode in which the relativistic electrons and holes move in the same direction (see Methods). This energy wave is analogous to the cosmic sound in high-energy physics, which gives rise to the large-scale fluctuations observed in the cosmic microwave background radiation, a relic of the Big Bang creating the Universe²⁷. Such cosmic sound is predicted to propagate at a universal speed of c/\sqrt{D} , in which *D* is the dimensionality. For Dirac electrons in graphene, the energy sound speed should have a value of $V_{\rm F}/\sqrt{2}$. Although these hydrodynamic collective excitations have been studied extensively in theory^{3,4,7,8,22,25}, their experimental observation in a solid-state system has so far been challenging.

On-chip THz spectroscopy

Here we first report the observation of the hydrodynamic plasmon and energy wave in ultraclean graphene microribbons using on-chip time-domain THz spectroscopy. Our technique exploits the subwavelength confinement of THz pulses in a coplanar waveguide, which bridges the large size mismatch between the graphene microribbons with width around 2 μ m and the THz photons with wavelength up to 3,000 μ m.

Figure 1a shows the schematic of our on-chip THz spectroscopy configuration to measure the absorption of a single graphene microribbon (see Methods for details). The THz emitter and detector are integrated onto a single chip together with a coplanar waveguide. We use a double-modulation scheme to detect the small changes in the THz transmission owing to the microribbon absorption¹⁸. We modulate the time delay (*t*) between the THz generation and detection laser pulses at frequency f_1 and detect the resultant modulation in current, which yields low-noise measurements of dE/dt. Furthermore, we modulate the back-gate voltage using a square wave (between a finite gate voltage V_G

¹Department of Physics, University of California at Berkeley, Berkeley, CA, USA. ²Graduate Group in Applied Science and Technology, University of California at Berkeley, Berkeley, CA, USA. ³Kavli Energy NanoScience Institute, University of California at Berkeley, Berkeley, CA, USA. ⁴Lawrence Berkeley National Laboratory, Berkeley, CA, USA. ⁵Research Center for Functional Materials, National Institute for Materials Science, Tsukuba, Japan. ⁶International Center for Materials Nanoarchitectonics, National Institute for Materials Science, Tsukuba, Japan. ⁷Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA, USA. ⁸These authors contributed equally: Wenyu Zhao, Shaoxin Wang. ^{Se}-mail: fengwang76@berkeley.edu



Fig. 1|THz plasmons in graphene microribbon at 60 K.a, Schematics of the on-chip THz spectroscopy experiment to investigate the absorption of a single graphene microribbon. The THz emitter and detector are trigged by a 532-nm femtosecond laser. A 1.064-µm femtosecond laser is used to pump the microribbon and transiently increase the electron temperature. The gap in the waveguide is narrowed down near the sample area to increase the coupling efficiency between the waveguide and the microribbon. The transmitted pulse is reconstructed by measuring the current collected by the preamplifier as a function of the time delay between the laser pulse trains illuminating the emitter and the detector, **b**. Raw data collected at the delay modulation frequency (dF/dt). **c**. Raw data collected at the sum frequency $(d(\Delta E)/dt)$ at gate voltages from -0.35 V to 0.15 V in steps of 0.025 V. Curves are offset for clarity. d, Measured absorption spectra of the graphene microribbon at various gate voltages. e, Microribbon absorption calculated using FDTD simulations. Red bars below the doping axis in **d** and **e** mark the region in which $|\mu| < T_e$. **f**,**g**, Linecuts of the absorption spectra at selected carrier densities from experiment and simulation, respectively.

and the charge-neutrality voltage $V_{CNP} = -0.1$ V) at f_2 , which allows us to measure the gate-induced THz change $d(\Delta E)/dt$ at the sum frequency $f_1 + f_2$ (ref. ¹⁸).

We first investigate the collective excitations in the graphene microribbon at the base temperature $T_0 = 60$ K. Figure 1b,c show the representative raw data of dE/dt and $d(\Delta E)/dt$ as a function of t at selected gate voltages. The THz absorption spectrum of the microribbon can be calculated as (see Methods for the derivation):

$$A_{V_{\rm G}}(f) - A_{\rm CNP}(f) \approx -2 \operatorname{Re}\left(\frac{\operatorname{FFT}(\mathrm{d}(\Delta E)/\mathrm{d}t)}{\operatorname{FFT}(\mathrm{d}E/\mathrm{d}t)}\right)$$

Here FFT represents fast Fourier transform, $A_{V_G}(f)$ is the absorption spectrum at gate voltage V_G and $A_{CNP}(f)$ is the absorption spectrum at charge neutrality. We measure $A_{V_G}(f) - A_{CNP}(f)$ at different values of V_G . In these data, $A_{CNP}(f)$ is a fixed background that can be obtained by a global fitting of all absorption spectra (see Methods for details). $A_{V_G}(f)$ can then be obtained using the fixed-background subtraction.

Figure 1d shows the absorption spectra $A_{V_G}(f)$ as a function of frequency and gate voltage. The data exhibit strong gate-dependent absorption peaks. At electron temperature $T_e = T_0$, the Dirac-liquid regime characterized by $|\mu| < T_e$ is rather narrow (red bar in Fig. 1d). At $|V_G - V_{CNP}| > 0.03$ V, the sample is already in the Fermi-liquid regime with $|\mu| > T_e$. The THz responses are thus dominated by the plasmon resonance of the Fermi liquid, which shifts to higher energy and gains oscillator strength with increasing carrier density. This behaviour is consistent with the Fermi-liquid plasmon in graphene observed in previous studies^{28,29}. However, we note that the plasmon resonance persists in the Dirac-liquid regime near charge neutrality (Fig. 2f). This provides the first sign of the collective behaviour of the thermally excited electrons and holes.

To understand the absorption data, we perform full-wave electromagnetic simulations using the finite-difference time-domain (FDTD) method (see Supplementary Information). We use the local-conductivity approximation and assume that the graphene conductivity is described by the two-fluid model from ref.¹,

$$\sigma = D_{Z} \pi^{-1} (\tau_{ee}^{-1} + \tau_{d}^{-1} - i2\pi f)^{-1} + D_{F} \pi^{-1} (\tau_{d}^{-1} - i2\pi f)^{-1}.$$

Here D_z and D_F are the Drude weights of the zero-momentum and finite-momentum modes, respectively, and τ_{ee}^{-1} and τ_{d}^{-1} are the electron– electron and disorder-scattering rates³⁰, respectively. This localconductivity model can capture both the Fermi-liquid plasmon and the Dirac-fluid hydrodynamic plasmon, but not the energy waves of the Dirac fluid. Figure 2e,g shows the calculated microribbon absorption spectra. The overall doping dependence in the simulation agrees well with the experimental data, which reaffirms our technique as a reliable approach to characterize the THz response of the graphene microribbon.

Hydrodynamic bipolar plasmon

Next we investigate hydrodynamic plasmons and energy-wave excitations of graphene at elevated electron temperatures. This is achieved by transiently heating the microribbon with a femtosecond pump pulse¹⁸ (Fig. 1a), which keeps the lattice near T_0 and helps avoid the unwanted electron-phonon scattering in an equilibrium heating approach. After pumping, the electron system rapidly (about 150 fs) thermalizes to a high electron temperature and equilibrates with a bath of optical phonons^{31,32}. These phonons decay over 1–2 ps (refs. ^{33,34}), returning the lattice to near equilibrium and lowering T_e to below the optical phonon emission threshold. Following this rapid cooling, a much slower (tens of picoseconds) cooling period ensues^{35–37}. We can thus study the temperature-dependent absorption by timing our probe THz pulse to arrive at the sample at different times during the slow-cooling period³⁵⁻³⁷. By synchronizing the optical heating pulse with the gate modulation (see Methods), we can measure $A(f)_{V_G,T_e} - A(f)_{CNP,T_0}$, in which $A(f)_{V_G,T_e}$ is the graphene absorption spectrum at different gate voltages V_G and electron temperatures T_e and $A(f)_{CNP,T_0}$ is the fixed CNP absorption spectrum at the reference temperature \check{T}_0 . Again, the fixed $A(f)_{CNP,T_0}$ is determined through the global fitting of all THz spectra. After subtracting $A(f)_{CNP.70}$ from the



Fig. 2| Hydrodynamic plasmons in graphene microribbon near charge neutrality. Measured graphene microribbon absorption spectra at 10 ps (a), 20 ps (b), 30 ps (c) and 12.5 ns (d) after the pump pulse. The electron temperature decreases from a to d. Red bars below the doping axis mark regions in which $|\mu| < T_e$. e, Linecuts of the absorption spectra at selected carrier densities from

measured data, we can obtain $A(f)_{V_G,T_e}$. Figure 2a–d shows the gatedependent absorption spectra taken at various delays between the optical heating pulse and the THz probe corresponding to different electron temperatures.

The elevated electron temperatures lead to pronounced changes in the plasmon excitation and the associated THz absorption in graphene. The most prominent plasmon features match well with the simulation results based on the local two-fluid model described above (Fig. 2g–j). On the basis of the comparison with simulations, we can estimate the electron temperature to be 300 K at 10-ps delay, which then decays to 260 K, 210 K and 105 K at 20-ps, 30-ps and 12.5-ns delay, respectively.

Figure 2e shows THz absorption spectra for several different carrier densities at $T_e = 300$ K. It shows behaviours distinct from those at $T_e = 60$ K (Fig. 1f). At $T_e = 300$ K, graphene is in the Dirac-liquid regime for most of the measured carrier densities (red bar in Fig. 2a). The THz spectra show two features: one strong peak at high frequency and a much weaker shoulder at low frequency. The strong high-frequency resonance corresponds to the hydrodynamic plasmon of the Dirac fluid and it can be captured by the local two-fluid model simulation (Fig. 2g). The weak low-frequency resonance can be attributed to the energy-wave excitation of the Dirac fluid.

We first examine the high-frequency hydrodynamic plasmon of the Dirac liquid at $T_e = 300$ K (Fig. 2a,e). The plasmon resonance frequency barely changes with the charge density in the hydrodynamic Dirac-liquid regime (from $V_G = -0.73$ V to $V_G = 0.53$ V) and it starts to



a. f, Dispersion of hydrodynamic plasmons extracted from experiment results. **g-j**, Simulated microribbon absorption spectra at electron temperatures of 300 K, 260 K, 210 K and 105 K. Red bars below the doping axis mark regions in which $|\mu| < T_e$, **k**, Linecuts of the absorption spectra at selected carrier densities from **g. l**, Dispersion of hydrodynamic plasmons extracted from simulations.

increase at higher charge densities ($V_{\rm G}$ = 0.8 V) when entering the Fermi-liquid regime. This hydrodynamic plasmon behaviour is distinctly different from the monopolar plasmon of the Fermi liquid at 60 K (Fig. 1f), at which the plasmon frequency increases rapidly with doping, especially at the low-density limit.

Figure 2f shows the plasmon resonance frequency as a function of doping at different T_e , in which a smooth crossover between the high-temperature Dirac-liquid hydrodynamic plasmon (that is, weak doping dependence) to the low-temperature Fermi-liquid plasmon behaviour (that is, strong doping dependence) is observed. Notably, the plasmon frequency at finite electron doping ($V_G = 0.8$ V) shows a redshift with increasing T_e and thus increasing total carrier density. This anomalous behaviour—a lower plasmon frequency with a larger total carrier density—is a notable signature of the hydrodynamic plasmon at the Dirac liquid to Fermi liquid crossover^{1,3,24}. The density and temperature dependence of the hydrodynamic plasmon agrees well with the local two-fluid conductivity model simulation (Fig. 2l).

Demon resonance in graphene microribbon

At low frequency, we also notice a weak dispersive feature in the absorption. Figure 3 zooms into the low-frequency region of the data in Fig. 2a–d. The colour bar is changed to increase the contrast of the low-frequency responses. A weak dispersive low-frequency resonance below the hydrodynamic plasmon excitation can be observed in the



Fig. 3 | **Demon mode in graphene microribbon near charge neutrality.** Measured graphene microribbon absorption spectra at $T_e = 300$ K (**a**), $T_e = 260$ K (**b**), $T_e = 210$ K (**c**) and $T_e = 105$ K (**d**). The images are scaled for better visualization of the low-frequency feature.

Dirac-liquid regime at $T_e = 300$ K (Fig. 3a) and $T_e = 260$ K (Fig. 3b). Such an isolated low-frequency resonance is not observed at $T_e = 105$ K (Fig. 3d), at which the Fermi-liquid regime covers most of the measured range and the plasmon resonance appears at a rather low frequency at low carrier concentrations. Similar absorption features are observed in a second device with the same design (see Supplementary Information).

This low-frequency resonance is a signature of the energy wave, also known as the demon mode, of the Dirac liquid at high electron temperatures. The energy wave arises from hydrodynamic responses of the Dirac liquid with the conservation of energy, momentum and charge in the electron system^{1,3,4,22,26}. At charge neutrality, this mode is a collective in-phase oscillation of the electron-hole plasma and has a characteristic linear dispersion and universal speed of $V_F/\sqrt{2}$ independent of other microscopic details². However it has zero dipole moment and

thus does not couple to the electromagnetic field. On the other hand, the energy wave can couple to the charge oscillation at finite doping, which results in a blue shift of the resonance frequency and a finite THz absorption. In the high-electron-density limit (that is, Fermi-liquid regime), this energy-wave mode eventually merges with the plasmon resonance^{6.8}. The charge-coupled energy wave exhibits strong temperature dependence owing to the thermal nature of the resonance.

Demon propagation in graphene

The low-frequency absorption feature in Fig. 3 is rather weak owing to the small oscillator strength of the energy wave. To conclusively establish the existence of the energy wave and examine its physical properties, it is desirable to selectively excite the energy wave and directly

Bias



Fig. 4 | **Spatial-temporal imaging of demon propagation in graphene. a**, Schematic side view of the hBN-encapsulated graphene device with gold waveguide serving as back gates. Gate 2 is set to 1.5 V in all the measurements, which corresponds to a carrier density of $n = 4 \times 10^{11}$ cm⁻² (see Supplementary Information). The energy wave is locally launched by a femtosecond laser away from the waveguide gap. It propagates to the nanogap region and induces a THz electrical signal that can be detected by the on-chip THz detector. By scanning the excitation at different positions and measuring the time delay of the THz signal, the propagation of the energy wave can be determined. **b**, Schematic top view of the on-chip spatial-temporal imaging technique to image the propagation of the demon mode. The waveguide is narrowed down to form a 300-nm-wide gap in the region beneath the graphene to enhance the electrical field and improve the spatial resolution. The induced THz electrical signal propagates along the waveguide and is detected at the GaAs detector. **c**, Optical micrograph of the device close to the sample region. The dashed red rectangle indicates the scanning range of the excitation laser, which is more than 10 µm away from the waveguide gap to avoid unintended background from the direct optical excitation in the gap region. Scale bar, 20 µm.



Fig. 5 | **Demon propagation in graphene at 300 K.** Spatial-temporal imaging data show the demon propagation at carrier densities $n = 0.1 \times 10^{11}$ cm⁻² (**a**), $n = 0.4 \times 10^{11}$ cm⁻² (**b**), $n = 0.8 \times 10^{11}$ cm⁻² (**c**), $n = 1.2 \times 10^{11}$ cm⁻² (**d**) and $n = 1.9 \times 10^{11}$ cm⁻² (**e**). The dashed red line in **a** marks $V_F / \sqrt{2}$ ($V_F = 10^6$ m s⁻¹).

Dispersion (f) and damping (g) of the demon mode at various carrier densities extracted from **a**–**e**. Error bars represent 2σ error from fitting the data in Extended Data Figs. 6 and 7. The dashed blue line marks a linear dispersion with a velocity of $V_{\rm F}/\sqrt{2}$.

image its propagation behaviour. This can be accomplished using a new spatial-temporal imaging technique based on the on-chip THz spectroscopy, as illustrated in Fig. 4a,b. Figure 4a shows a cross-section view of the graphene device configuration. A monolayer graphene sheet $(15 \times 60 \ \mu\text{m})$ was encapsulated between two hexagonal boron nitride (hBN) layers. The hBN (50 nm)/graphene/hBN (80 nm) heterostructure is placed on top of a pair of 50-nm-thick gold waveguide traces, which are laterally separated by a 300-nm-wide nanogap. The waveguide also serves as the back gate to change the carrier density in the graphene.

Figure 4b shows a top view of the whole device and an illustration of the spatial-temporal imaging technique. A 532-nm femtosecond laser is focused to a narrow line with a lateral width of around 1 µm to create an ultrafast local thermal excitation of the electron system in graphene. The thermalization of optically excited electrons happens on a femtosecond timescale^{31,32,38,39} and thus can be considered as almost instantaneous. In the hydrodynamic regime, this transient heat pulse can propagate as energy waves along the graphene channel³ (white arrow in Fig. 4a). In contrast to the THz absorption measurement, the approach here selectively launches the energy wave and avoids the strong background response from the hydrodynamic bipolar plasmon. When the energy wave reaches the waveguide nanogap, it can generate a transient electrical signal (that is, a THz pulse) in the waveguide. Here the nanogap in the waveguide acts as a THz nanoantenna that converts the local charge oscillation associated with the energy wave in weakly doped graphene³ to a strong THz electrical signal (see Methods). This THz signal will propagate along the waveguide and be detected by the GaAs photoconductive detector. By scanning the position of the line excitation and triggering the GaAs detector using a time-delayed pulse, we can measure the THz electrical signal E(t, x) as a function of the pump position (x) and the time delay (t). This spatial-temporal mapping directly examines the propagation behaviour of the energy wave.

Figure 4c shows an optical micrograph of the device close to the graphene region. The dashed red box indicates the laser scan range, which is 10 μ m away from the gap to avoid direct excitation of the waveguide gap by the laser excitation.

Figure 5a–e shows the spatial–temporal evolution of the measured THz signal at different dopings. The graphene temperature is at 300 K in these measurements. A propagating energy wave is clearly observed at all dopings. At extremely low charge density ($n \approx 0.1 \times 10^{11} \text{ cm}^{-2}$), the demon mode has a velocity close to $V_{\rm F}/\sqrt{2}$ (dashed red line in Fig. 5a), in which $V_{\rm F} = 10^6$ m s⁻¹. This propagation speed is a signature of the thermal sound wave in a charge-neutral Dirac liquid. As the doping increases, the demon velocity increases rapidly owing to the coupling with the charge oscillations in graphene. The width of the propagating feature also becomes narrower with increasing doping, suggesting an increasing quality factor.

We can convert the time-domain electrical signal E(t, x) to the frequency domain through Fourier transform,

$$E(t,x) = \int 2\pi [E(f)e^{iq(f)x}e^{-q'(f)x}]e^{-i2\pi ft}df$$

For each frequency *f*, the propagation wave has a corresponding wavevector q(f) and a decay constant q'(f). The value of q(f) and q'(f) can be extracted by fitting the linear phase shift and exponential amplitude decay of the signal as a function of position *x*, respectively (see Methods). This procedure of obtaining frequency-domain information from time-domain data is commonly used in FDTD studies⁴⁰. Figure 5f,g shows the extracted dispersion (*q*) and damping (*q'*) of the energy wave at different carrier densities. Close to charge neutrality, the energy wave has a linear dispersion with a universal speed close to $V_F/\sqrt{2}$. The damping rate of the energy wave is relatively high and it increases quickly with frequency. As the doping increases, the energy wave propagates with a faster speed and a smaller spatial decay rate.

In summary we observed both hydrodynamic plasmons and the hydrodynamic energy waves of Dirac fluid using new on-chip THz spectroscopy techniques. Our study opens new opportunities to explore collective hydrodynamic excitations in low-dimensional materials.

Online content

Any methods, additional references, Nature Portfolio reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41586-022-05619-8.

- Sun, Z., Basov, D. N. & Fogler, M. M. Universal linear and nonlinear electrodynamics of a Dirac fluid. Proc. Natl Acad. Sci. USA 115, 3285–3289 (2018).
- Lucas, A. & Fong, K. C. Hydrodynamics of electrons in graphene. J. Phys. Condens. Matter 30, 053001 (2018).
- Briskot, U. et al. Collision-dominated nonlinear hydrodynamics in graphene. *Phys. Rev. B* 92, 115426 (2015).
- Phan, T. V., Song, J. C. W. & Levitov, L. S. Ballistic heat transfer and energy waves in an electron system. Preprint at https://arxiv.org/abs/1306.4972 (2013).
- Geim, A. K. & Novoselov, K. S. in Nanoscience and Technology: A Collection of Reviews from Nature Journals 11–19 (World Scientific, 2010).
- Levitov, L., Shtyk, A. & Feigelman, M. Electron-electron interactions and plasmon dispersion in graphene. *Phys. Rev. B* 88, 235403 (2013).
- 7. Torre, I. et al. Acoustic plasmons at the crossover between the collisionless and
- hydrodynamic regimes in two-dimensional electron liquids. *Phys. Rev. B* **99**, 144307 (2019). 8. Narozhny, B., Gornyi, I. & Titov, M. Hydrodynamic collective modes in graphene. *Phys.*
- Rev. B 103, 115402 (2021).
 Sun, Z., Basov, D. & Fogler, M. Adiabatic amplification of plasmons and demons in 2D
- systems. *Phys. Rev. Lett.* **117**, 076805 (2016). 10. Moll, P. J., Kushwaha, P., Nandi, N., Schmidt, B. & Mackenzie, A. P. Evidence for
- hydrodynamic electron flow in PdCoO₂. Science **351**, 1061–1064 (2016). 11. Ku, M. J. et al. Imaging viscous flow of the Dirac fluid in graphene. *Nature* **583**, 537–541
- (2020). 12. Bandurin, D. A. et al. Fluidity onset in graphene. *Nat. Commun.* **9**, 4533 (2018).
- Bandurin, D. et al. Negative local resistance caused by viscous electron backflow in graphene. *Science* 351, 1055–1058 (2016).

- Berdyugin, A. I. et al. Measuring Hall viscosity of graphene's electron fluid. Science 364, 162–165 (2019).
- Krishna Kumar, R. et al. Superballistic flow of viscous electron fluid through graphene constrictions. *Nat. Phys.* 13, 1182–1185 (2017).
- Jenkins, A. et al. Imaging the breakdown of ohmic transport in graphene. *Phys. Rev. Lett.* 129, 087701 (2022).
- 17. Crossno, J. et al. Observation of the Dirac fluid and the breakdown of the Wiedemann-Franz law in graphene. Science **351**, 1058–1061 (2016).
- Gallagher, P. et al. Quantum-critical conductivity of the Dirac fluid in graphene. Science 364, 158–162 (2019).
- 19. Narozhny, B. N. Electronic hydrodynamics in graphene. Ann. Phys. 411, 167979 (2019).
- Lucas, A. & Sarma, S. D. Electronic sound modes and plasmons in hydrodynamic two-dimensional metals. *Phys. Rev. B* 97, 115449 (2018).
- Kiselev, E. I. & Schmalian, J. Nonlocal hydrodynamic transport and collective excitations in Dirac fluids. *Phys. Rev. B* 102, 245434 (2020).
- 22. Lucas, A. Sound waves and resonances in electron-hole plasma. *Phys. Rev. B* 93, 245153 (2016).
- Dufty, J., Luo, K. & Wrighton, J. Generalized hydrodynamics revisited. Phys. Rev. Res. 2, 023036 (2020).
- Svintsov, D. Hydrodynamic-to-ballistic crossover in Dirac materials. Phys. Rev. B 97, 121405 (2018).
- Fateev, D. & Popov, V. Hydrodynamic terahertz plasmons and electron sound in graphene with spatial dispersion. Semiconductors 54, 941–945 (2020).
- Svintsov, D., Vyurkov, V., Yurchenko, S., Otsuji, T. & Ryzhii, V. Hydrodynamic model for electron-hole plasma in graphene. J. Appl. Phys. 111, 083715 (2012).
- Anderson, L. et al. The clustering of galaxies in the SDSS-III Baryon Oscillation Spectroscopic Survey: baryon acoustic oscillations in the Data Releases 10 and 11 Galaxy samples. Mon. Not. R. Astron. Soc. 441, 24–62 (2014).
- Woessner, A. et al. Highly confined low-loss plasmons in graphene-boron nitride heterostructures. Nat. Mater. 14, 421–425 (2015).
- Ni, G. et al. Fundamental limits to graphene plasmonics. *Nature* 557, 530–533 (2018).
 Müller, M., Fritz, L. & Sachdev, S. Quantum-critical relativistic magnetotransport in
- Mutter, M., Fitz, L. & Sachdev, S. Quantum-critical relativistic magnetotiansport in graphene. *Phys. Rev. B* 78, 115406 (2008).
 Giorz, L. et al. Spacebate of pap aquilibrium Dirac carrier distributions in graphana.
- Gierz, I. et al. Snapshots of non-equilibrium Dirac carrier distributions in graphene. Nat. Mater. 12, 1119–1124 (2013).
- George, P. A. et al. Ultrafast optical-pump terahertz-probe spectroscopy of the carrier relaxation and recombination dynamics in epitaxial graphene. *Nano Lett.* 8, 4248–4251 (2008).
- 33. Yan, H. et al. Time-resolved Raman spectroscopy of optical phonons in graphite: phonon anharmonic coupling and anomalous stiffening. *Phys. Rev. B* **80**, 121403 (2009).
- Kang, K., Abdula, D., Cahill, D. G. & Shim, M. Lifetimes of optical phonons in graphene and graphite by time-resolved incoherent anti-Stokes Raman scattering. *Phys. Rev. B* 81, 165405 (2010).
- Strait, J. H. et al. Very slow cooling dynamics of photoexcited carriers in graphene observed by optical-pump terahertz-probe spectroscopy. Nano Lett. 11, 4902–4906 (2011).
- Graham, M. W., Shi, S.-F., Ralph, D. C., Park, J. & McEuen, P. L. Photocurrent measurements of supercollision cooling in graphene. *Nat. Phys.* 9, 103–108 (2013).
- Song, J. C. & Levitov, L. S. Energy flows in graphene: hot carrier dynamics and cooling. J. Phys. Condens. Matter 27, 164201 (2015).
- Mics, Z. et al. Thermodynamic picture of ultrafast charge transport in graphene. Nature 6, 7655 (2015).
- Tielrooij, K.-J. et al. Photoexcitation cascade and multiple hot-carrier generation in graphene. Nature 9, 248–252 (2013).
- Schneider, J. B. Understanding the finite-difference time-domain method. https://eecs. wsu.edu/-schneidj/ufdtd/ (2010).

Publisher's note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Springer Nature or its licensor (e.g. a society or other partner) holds exclusive rights to this article under a publishing agreement with the author(s) or other rightsholder(s); author self-archiving of the accepted manuscript version of this article is solely governed by the terms of such publishing agreement and applicable law.

© The Author(s), under exclusive licence to Springer Nature Limited 2023

Methods

Fabrication of graphene samples

We used a dry-transfer method with a propylene carbonate (PC) stamp to fabricate the graphene sample. hBN, monolayer graphene and few-layer MoS_2 were first exfoliated onto Si substrates with a 285-nm SiO_2 layer. For the graphene device in Extended Data Fig. 1a, we used the PC stamp to pick up the hBN flake (100 nm), graphene, gate spacer hBN (90 nm) and few-layer MoS_2 in sequence. The heterostructure was subsequently released on a fused-quartz substrate, which has negligible loss in the THz frequency range. For the graphene device in Fig. 4c, we used the PC stamp to pick up the hBN flake (50 nm), graphene, gate spacer hBN (80 nm) and then released the heterostructure onto a prepatterned waveguide.

Fabrication of on-chip devices for THz spectroscopy measurements

The GaAs flakes were transferred onto the same substrates using polydimethylsiloxane stamps. The positions of the emitter and detector were aligned to make sure the waveguide can overlap with the graphene sample. To suppress unintended reflections of the THz pulse from waveguide ends and electrodes, the emitter and detector were placed 1.6 mm apart and far away from both ends of the 7-mm-long waveguide. The waveguide traces consist of a 200-nm gold layer on top of a 3-nm Cr layer, patterned using standard photolithography methods and in good contact with the GaAs flakes. Standard electron-beam lithography was used to make the mask for etching the graphene into narrow microribbons (Extended Data Fig. 1a) or long channels (Fig. 4c). Reactive-ion etching with CHF₃ and O₂ etching gases (40 sccm and 6 sccm) was used to etch away the hBN. The graphene was then etched with O₂ only. Before fabricating the narrow waveguide on the graphene microribbon, we released another hBN flake (50 nm) on the top surface to avoid direct electric contact between the gold waveguide and graphene.

On-chip THz spectroscopy of graphene microribbon

A 532-nm femtosecond laser (150-fs pulse duration) is split into two beams, which are then focused onto the emitter and the detector, respectively (Fig. 1a). Both the emitter and the detector are far away from the waveguide ends and the distance between them is around 1.6 mm. The large separation between different components avoids the overlap between the main pulse and several reflection signals in the time-domain measurements. Only the directly transmitted pulse is collected and analysed. The substrate is made of fused quartz to minimize unwanted absorption in the THz range. The waveguide consists of two parallel gold traces of 20 μ m width separated by a 15- μ m gap (Extended Data Fig. 2). The gap is tapered and narrowed down near the sample to enhance the sample–waveguide coupling.

Extended Data Fig. 1a shows the optical micrograph of the device close to the sample region and Extended Data Fig. 1b shows the sample schematics in the cross-sectional view. An ultraclean graphene microribbon of 2 μ m width and 20 μ m length is encapsulated in hBN layers. The doping level of graphene is controlled by a back gate made of few-layer MoS₂ for its negligible THz absorption. The waveguide is separated from the graphene by 150-nm-thick hBN.

In each measurement, the biased GaAs emitter is first triggered by a femtosecond laser pulse and becomes highly conductive for about 1 ps (as determined by the carrier lifetime of GaAs), creating a quasi-transverse electromagnetic THz pulse that travels with minimal dispersion along the waveguide. Then the detector is activated by a time-delayed laser pulse, allowing a current proportional to the instantaneous local electric field (*E*) at the detector to be collected by a current preamplifier.

Double-modulation method for the absorption measurements

We use a double-modulation method to measure the THz absorption of the graphene microribbon¹⁸. The time delay between the two laser

pulse trains for THz generation and detection is modulated at frequency f_1 , whereas the back-gate voltage is modulated using a square wave at frequency f_2 . The detector current is measured at two lock-in frequencies, f_1 and $f_1 + f_2$. These two signals mainly correspond to dE/dt and $d(\Delta E)/dt$, respectively. We then obtain the following quantity after FFT,

$$\frac{\text{FFT}\left(\frac{d(\Delta E)}{dt}\right)}{\text{FFT}\left(\frac{dE}{dt}\right)} = \frac{\text{FFT}\left(\frac{d(E_{V_{G}} - E_{\text{CNP}})}{dt}\right)}{\text{FFT}\left(\frac{d(E_{V_{G}} + E_{\text{CNP}})}{2dt}\right)} = \frac{2i\omega\left(\widetilde{E}_{V_{G}} - \widetilde{E}_{\text{CNP}}\right)}{i\omega\left(\widetilde{E}_{V_{G}} + \widetilde{E}_{\text{CNP}}\right)}.$$
(1)

Here $\tilde{E}_{V_{G}}$ and \tilde{E}_{CNP} are the transmitted THz fields as functions of angular frequency ω at finite gate voltage V_{G} and charge neutrality, respectively. In our experiments, the graphene microribbon only causes a small change to the THz field transmission. In other words, $\tilde{E}_{V_{G}} = \tilde{E}_{0} (1 - \delta_{V_{G}})$ and $\tilde{E}_{CNP} = \tilde{E}_{0} (1 - \delta_{CNP})$, in which \tilde{E}_{0} is the transmitted THz fields without the graphene microribbon, $\delta_{V_{G}} \ll 1$ and $\delta_{CNP} \ll 1$. Expanding equation (1) to the leading order in δ , we have

$$\frac{\mathsf{FFT}\left(\frac{\mathrm{d}(\Delta E)}{\mathrm{d}t}\right)}{\mathsf{FFT}\left(\frac{\mathrm{d}E}{\mathrm{d}t}\right)} \approx \delta_{\mathrm{CNP}} - \delta_{V_{\mathrm{G}}}.$$
(2)

Because the microribbon absorption $A = 1 - |1 - \delta|^2 \approx 2\text{Re}(\delta)$, we arrive at the formula used in the main text,

$$A_{V_{\rm G}} - A_{\rm CNP} \approx 2 \operatorname{Re}(\delta_{V_{\rm G}} - \delta_{\rm CNP}) \approx -2 \operatorname{Re}\left[\frac{\operatorname{FFT}\left(\frac{\mathrm{d}(\Delta E)}{\mathrm{d}t}\right)}{\operatorname{FFT}\left(\frac{\mathrm{d}E}{\mathrm{d}t}\right)}\right].$$
(3)

For measurements at elevated electron temperature, the pump laser transiently heating the sample is modulated using a chopper in synchronization with the back-gate modulation. In this case, we have

$$A_{V_{\rm G},T_{\rm e}} - A_{\rm CNP,T_0} \approx -2 \operatorname{Re}\left[\frac{\operatorname{FFT}\left(\frac{\mathrm{d}(\Delta F)}{\mathrm{d}t}\right)}{\operatorname{FFT}\left(\frac{\mathrm{d}F}{\mathrm{d}t}\right)}\right],\tag{4}$$

in which A_{V_G, T_e} is the microribbon absorption at back-gate voltage V_G and electron temperature T_e and A_{CNP, T_0} is the absorption at charge neutrality and base temperature T_0 .

Determination of the charge-neutrality point

We use a similar double modulation of the optical pump laser to determine the back-gate voltage associated with the charge-neutrality point of graphene. At each fixed gate voltage V_G , we modulate the time delay between THz generation and detection at f_1 and the pump laser at f_2 . We perform lock-in measurements at f_1 and $f_1 + f_2$ and reconstruct $A_{V_G, T_e} - A_{V_G, T_0}$, the microribbon absorption difference between elevated electron temperature T_e and base temperature T_0 at fixed gate voltage V_G . Because $A_{V_G, T_e} - A_{V_G, T_0}$ is symmetric with respect to the charge-neutrality point, the gate voltage of the charge-neutrality point can be accurately determined, as shown in Extended Data Fig. 3.

Graphene microribbon absorption spectra at charge neutrality

The absorption spectra measured in the experiment is proportional to $A_{V_{\rm G}} - A_{{\rm CNP}}$, which contains background from the charge-neutrality point. Because this background has a fixed profile for all the gate voltages, we can extract it by fitting the data to a two-Lorentzian model. The spectra can be decomposed into two Lorentzian components, $A_{V_{\rm G}}$ and $A_{{\rm CNP}}$ (see Extended Data Fig. 4a). The global $A_{{\rm CNP}}$ as shown in Extended Data Fig. 4b is obtained by averaging the fitted $A_{{\rm CNP}}$ over a broad range of gate voltages (0.1–0.8 V). Extended Data Fig. 4c and Extended Data Fig. 4d show the absorption spectra of the graphene microribbon at 60 K before and after the background subtraction, respectively.

Efficient THz generation by the energy wave at the nanogap

Our experiment takes advantage of the large field enhancement of the nanogap between the waveguide, similar to near-field-spectroscopy experiments⁴¹. The moving charges associated with the energy wave in weakly doped graphene³ will induce a negligible THz signal in the waveguide when they are above the metal but will generate a strong THz signal when they reach the nanogap.

To demonstrate this effect more quantitatively, we simulate the induced THz field in the waveguide generated by a dipole source at different locations. Extended Data Fig. 5a,b shows the top and side views of the model used in the FDTD simulations. A dipole source is placed 80 nm (hBN thickness) above the waveguide and a field monitor is used to collect the field in the waveguide 60 µm away from the excitation. We check the collected field as a function of the dipole source position and frequency, as plotted in Extended Data Fig. 5c. Extended Data Fig. 5d shows the linecut of the field amplitude for different excitation positions at 0.1 THz. It is obvious that the THz generation becomes strong only when the dipole source is at the nanogap.

Propagation constant of the demon mode in graphene

The frequency-domain response of the wave propagation can be extracted through Fourier transform of the time-domain electrical signal F(t, x). Extended Data Figs. 6 and 7 show the extraction of propagation constant q(f) and q'(f) by fitting the linear phase shift and exponential amplitude decay as a function of position x, respectively.

Data availability

The data that support the findings of this study are available from the corresponding author on request. Source data are provided with this paper.

 Bao, W. et al. Mapping local charge recombination heterogeneity by multidimensional nanospectroscopic imaging. Science 338, 1317–1321 (2012).

Acknowledgements The on-chip terahertz spectroscopy and theoretical analysis of the work was supported by the US Department of Energy, Office of Science, Office of Basic Energy Sciences, Materials Sciences and Engineering Division under contract number DE-AC02-05CH11231 within the vdW heterostructure Program (KCWF16). The device fabrication is supported by the Army Research Office award W911NF2110176. S.C. acknowledges support from Kavli ENSI Heising-Simons Junior Fellowship. K.W. and T.T. acknowledge support from the Element Strategy Initiative conducted by the MEXT, Japan and the CREST (JPMJCR15F3), JST.

Author contributions F.W. conceived the research. W.Z. carried out the on-chip THz measurements. S.W. and Z.Z. fabricated the graphene devices. W.Z., S.W., S.C. and F.W. performed the data analysis. K.W. and T.T. grew the hBN crystals. F.W., W.Z., S.C. and A.Z. wrote the manuscript.

Competing interests The authors declare no competing interests.

Additional information

Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41586-022-05619-8.

Correspondence and requests for materials should be addressed to Feng Wang. **Peer review information** *Nature* thanks Dmitry Svintsov and the other, anonymous, reviewer(s) for their contribution to the peer review of this work.

Reprints and permissions information is available at http://www.nature.com/reprints.



Extended Data Fig. 1 | **On-chip THz spectroscopy on graphene microribbon. a**, Optical micrograph of the device close to the sample region. **b**, Cross-sectional schematics of the sample under study. The graphene microribbon of 2 μm width and 20 μm length is encapsulated by hBN flakes. The doping level of the microribbon is controlled through a few-layer MoS_2 back gate for its minimal absorption in the THz frequency range. **c**, Illustration of the collective motion of electrons and holes in hydrodynamic bipolar plasmon and demon.



 $\label{eq:stended} Extended Data Fig. 2 | Optical micrograph of the graphene microribbon device for hydrodynamic plasmon and demon measurements. The waveguide consists of two parallel gold traces of 20 \,\mu m width separated by a 15- \mu m gap. The gap is tapered and narrowed down near the sample to enhance the sample-waveguide coupling.$



Extended Data Fig. 3 | **Temperature difference of the absorption spectra**, $A_{V_G,T_e} - A_{V_G,T_0}$, **obtained by the double-modulation measurement**. The absorption shows a symmetric profile with respect to $V_G = -0.1$ V, the back-gate voltage associated with the charge-neutrality point.





Extended Data Fig. 4 | **Determination of** A_{CNP} **and background subtraction for the absorption spectra. a**, The measured spectrum (solid curve) is fitted to a two-Lorentzian model, in which L_1 and L_2 represent the fitted absorption spectra at the charge-neutrality point and V_G , respectively. **b**, The global

background absorption spectra obtained by averaging the fitted $A_{\rm CNP}$ over a broad range of gate voltages from 0.1 V to 0.8 V. c, Measured $A_{V\rm G}-A_{\rm CNP}$ at various gate voltages. d, $A_{V\rm G}$ obtained by subtracting the global absorption background associated with $A_{\rm CNP}$.



Extended Data Fig. 5 | **Field response of the nanogap.** Schematic top view (**a**) and side view (**b**) of the model used in the FDTD simulations. **c**, Field amplitude collected at the monitor position. **d**, Linecut of the field amplitude at 0.1 THz.



Extended Data Fig. 6 | **Linear fit of the phase term along the propagation direction for different carrier densities.** From top to bottom, the frequency changes from 0.03 THz to 0.1 THz. *q*(*f*) corresponds to the phase-accumulation slope.



Extended Data Fig. 7 | **Exponential fit of the amplitude decay**, *q*'(*f*), along the propagation direction at different carrier densities. The *y* axis is plotted in logarithmic scale.