

Electron-phonon instability in graphene revealed by global and local noise probes





Nonequilibrium dynamics in graphene, probed both globally and locally. (A) Device schematic: hBN (hexagonal boron nitride) -encapsulated graphene device on diamond substrate containing NV (Nitrogen-Vacany) centers for nanomagnetometry. (Inset) The optical image of clean hBN-encapsulated device A1 (6 μ m x5.4 μ m) (B) Condition for Cerenkov emission of phonons: when vD>vs, stimulated phonon (ph) emission dominates over absorption (right). (C) Two-probe resistance versus carrier density of device A1 (T = 10 K). (D) Current density as a function of applied electric field (T = 80 K) in clean device A1 (blue) and disordered device B1 (7 μ m by 18 μ m, black). The gray dashed line indicates where vD=vs for the longitudinal acoustic mode. (E) Global electronic noise PSD (averaged over 100 to 300 MHz) as a function of bias



power in devices A1 (blue) and B1 (black). Blue curve satisfies vD>vs for P > 0.12μ W/ μ m2. (F) Local magnetic noise (measured by NV nanomagnetometry) versus applied bias power in clean device C1 on diamond substrate. Error bars represent 95% confidence intervals. Credit: *Science*, doi: 10.1126/science.aaw2104

Understanding nonequilibrium phenomena to effectively control it is an outstanding challenge in science and engineering. In a recent study, Trond. I. Andersen and colleagues at the departments of physics, chemistry, materials science and engineering in the USA, Japan and Canada used electricity to drive ultraclean graphene devices out-ofequilibrium and observe the manifested instability as enhanced current fluctuations and suppressed conductivity at microwave frequencies.

Using the experimental setup, they found that direct current at high drift velocities generated a large increase in the noise at gigahertz frequencies and the noise grew exponentially in the direction of the current. Andersen and co-workers credited the observed emission mechanism, to the amplification of acoustic phonons by the <u>Cerenkov effect</u> (a characteristic blue glow resulting from charged particles passing through an insulator at a speed greater than the speed of light in that medium) and have now published the results on *Science*.

The scientists spatially mapped the nonequilibrium current fluctuations using nanoscale magnetic field sensors to reveal that they grew exponentially along the direction of carrier flow. Andersen et al. credited the observed dependence of the phenomenon on density and temperature, to electron-phonon <u>Cerenkov instability</u> at <u>supersonic drift</u> velocities. Supersonic drift velocities occurred when the population of certain phonons increased with time due to forced Cerenkov emission, when the drift velocity of electron conduction was greater than the



velocity of sound $(V_D > V_S)$ in the medium. The <u>experimental results</u> can offer the opportunity to generate tunable terahertz frequencies and construct active phononic devices on two-dimensional materials.

Nonequilibrium phenomena driven in electronic and <u>optical systems</u> display rich dynamics, which can be harnessed for applications as <u>Gunn</u> diodes and lasers. <u>Two-dimensional materials</u> such as graphene, are an increasingly popular new platform to explore such phenomena. For instance, modern ultraclean graphene devices demonstrate <u>high</u> mobilities and can be driven to high electronic velocities with predicted instabilities to include <u>hydrodynamic instabilities</u> in <u>electronic fluids</u> and <u>Dyakonov-Shur</u> instabilities where the <u>driven electrons can amplify</u> plasmons.





TOP: Measurement circuit. Circuit diagram for the measurement of noise (red box) and AC differential conductivity (yellow box). LEFT: Device fabrication on diamond substrate. (A) Device schematic: Monolayer graphene (grey chain) was graphite contacted and encapsulated with hexagonal boron nitride (hBN). Few-layer graphene (FLG) was used as topgate. (B-H) Micrographs of device



fabrication, with 40 µm scalebar in (B)-(G) and 500 µm in (H). (B) Exfoliated graphene. White dashed line indicates monolayer region. (C) Complete stack on diamond substrate with shallow implanted (40 - 60 nm deep) NV centers. (D) Initial contacts and wire for delivering reference noise (left-most electrode). (E) Device after etch to define geometry. (F) Edge contacts constructed through etching and subsequent thermal evaporation. (G) Device with etch mask for disconnecting topgate from edge contacts. Note that ripples visible in the image are entirely contained in the top gate graphene and are not expected to affect the transport properties of the channel graphene, due to the thick (~ 90 nm) hBN dielectric. (H) Entire (2×2 mm2) single crystal diamond, with wire bonded device. RIGHT: Device fabrication on Si/SiO2 substrate. (A) Device schematic: Monolayer graphene (grey chain) was encapsulated with hexagonal boron nitride (hBN). Silicon substrate was used as a global backgate. (B)-(F) Micrographs of device fabrication, with 20 µm scalebar. (B) Exfoliated graphene. (C) Complete stack on substrate. (D) Initial contacts. (E) Edge contacts constructed through etching and subsequent thermal evaporation. (F) Device after geometry-defining etch. Credit: Science, doi: 10.1126/science.aaw2104

The study of electronic properties of graphene under extreme nonequilibrium conditions therefore provides a productive testbed to assess and monitor exotic transport phenomena. In addition to the use of high-frequency signal generation, Andersen et al. investigated the underlying non-equilibrium dynamics during electron transport in ultraclean graphene devices containing an extremely high electron drift velocity. Understanding <u>nonequilibrium dynamics</u> is vital for many technical applications of graphene; including <u>high frequency transistors</u>, <u>ultrafast incandescent light sources</u> and <u>flexible transport interconnects</u>. However, it is difficult to realize the electronic stabilities in practice, due to increased <u>phonon</u> scattering at high drift velocities.

In principle, while phonon scattering loss is typically irreversible, longlived phonons can act as a dominant source of instability within the



experimental setup. When the electronic drift velocity (V^D) exceeds the velocity of sound (V_S) , phonon emission becomes greater than phonon absorption, resulting in an exponential growth of the phonon population, known as phonon <u>Cerenkov amplification</u>. The phenomenon was <u>long</u> explored in theory as a technique to produce high-frequency acoustic waves, with accompanying experimental evidence in bulk systems and <u>semiconductor superlattices</u> obtained using acoustic and optical measurements thereafter.





Spatially resolved local noise measurements with NV magnetometry. (A) Fluorescence image of NV centers underneath device C2, with false-colored contacts and borders added. (B) NV spin relaxation from polarized to thermal state (dashed line), when current densities $j = 0 \text{ mA}/\mu\text{m}$ (dark blue) and $j = -0.19 \text{ mA}/\mu\text{m}$ (light blue) are passed through the device. Solid lines are fits. ms, spin quantum number. (C) Local magnetic noise near drain contact as a function of graphene current density (device C1) in electron (e)– and hole (h)–doped regime (blue and red, respectively). (D) Spatial map of the local magnetic noise (device C2) at $j = 0.18 \text{ mA}/\mu\text{m}$ and $n = 0.92 \times 1012 \text{ cm}-2$. The spatial profile is consistent with the exponential growth of phonons due to Cerenkov amplification (cartoon, top). Dashed black curve shows the theoretically predicted excess phonon population (offset to account for background noise). a.u., arbitrary units. (E) The growth direction is reversed by changing the current direction (left) or the charge carrier sign (right). Error bars represent 95% confidence intervals. Credit: *Science*, doi: 10.1126/science.aaw2104

In the present work, Andersen et al. used electrically gated graphene devices fabricated on diamond and silicon/silicon dioxide substrates, encapsulated in hexagonal boron nitride (hBN) at cryogenic temperatures (T= 10 to 80 K) to conduct the proposed experiments. The experimental setup provided low-bias transport properties for the ultraclean graphene system with a mobility ranging from 20 to 40 m²/V.s at a carrier density (2×10^{12} cm⁻²), corresponding to <u>nearly ballistic</u> transport. Due to high mobility, carriers could be accelerated by an electric field to high drift velocities to observe nonlinear current response, while a disordered device contrastingly showed linear ohmic behavior.

To study the nonequilibrium behavior, first, Andersen et al. measured the global noise in the <u>source-drain current</u> with a spectrum analyzer,



while varying the applied bias power (*P*). The results indicated a new source of noise in graphene devices with low disorder, encapsulated in hBN. To gain insight to the observed anomaly, the scientists performed spatially resolved noise measurements by constructing graphene devices on diamond substrates with shallow <u>nitrogen-vacancy</u> color-center impurities <u>of 40 to 60 nm in depth</u>. They measured the atom-like spin qubits using confocal microscopy and probed the nanoscale current noise by <u>measuring the resulting magnetic fields</u>.

Andersen et al. probed the spatial dependence of the anomalous noise by optically observing single NV centers along the device to measure their spin relaxation rate. The noise exhibited clear symmetry with the direction of the current, an unexpected outcome since global noise and transport properties are independent of the direction of the current. Then using the device gate, Andersen et al. demonstrated that the local noise signal depended on the flow direction of momentum and not charge. The scientists also showed that the noise was small at the carrier entry point but grew exponentially as the carrier flowed across the 17- μ m long device.





Slow dynamics in global electronic measurements. (A) Global noise spectra at n = 2×1012 cm-2. Colored curves: clean device A2 (9.5 µm by 11 µm) at bias ranging from 0 to 0.8 V (bottom to top). Black curve: disordered device B1 at maximum power applied to device A2 (scaled $7\times$). (B) Ac differential conductivity spectra (excitation: -20 dBm) (19) with biases 0 to 0.8 V [top to bottom, colors same as in (A)]. The real (Re) component is suppressed at low frequencies. Gray curve: imaginary (Im) component at 0.8 V. Black curves are fits. (C and D) Features in noise and conductivity spectra shift to higher frequencies in a shorter (6-µm) device (device A1) under similar electric field as maximum in (A) and (B). (E and F) Extracted traversal time from (B) and (D) as a function of drift velocity and device length. Dashed curves correspond to speed of sound in graphene [light gray, transverse acoustic (TA); dark gray, longitudinal acoustic (LA)]. (G) Cartoon of important rates in the driven electronphonon system. During Cerenkov amplification, the correlation time observed in electronic measurements is limited by the phonon traversal time, tT=L/vs. Credit: Science, doi: 10.1126/science.aaw2104

The scientists consistently explained all observations using the electrophonon Cerenkov instability. As a key insight of the study, Andersen et al. showed that when the electronic drift velocity exceeded the speed of sound (supersonic drift velocity), the forward-moving acoustic phonons experienced a faster rate of <u>simulated emission than absorption</u>. Pristine graphene also exhibited long acoustic phonon lifetimes; therefore, an emitted phonon could stimulate the emission of exponential growth in the setup.

When they modelled these effects mathematically, the results agreed well with experimental outcomes, while the anomalous noise further increased with increasing device length. The model predicted that the observed electron-phonon instability would give rise to a <u>conductivity</u> <u>spectrum</u>. The scientists continued to explore the nonequilibrium



dynamics using models of the electron-phonon system.



Dependence on bath temperature and charge density. (A) Global noise PSD as a function of bath temperature at constant drift velocities and $n = 2 \times 10^{12}$ cm–2. (B) Calculated peak phonon emission frequency, which can be tuned via the graphene carrier density (blue: Te = 0 K; red: Te = 320 K). (C) Normalized global current noise as a function of carrier density for different device lengths (j = 0.6 mA/µm). Solid curves show predicted total phonon emission. (D) The charge density at which the noise peaks (npeak) for a wider variety of samples than in (C), with fit (blue). Error bars represent sampling spacing of carrier densities. Credit: *Science*, doi: 10.1126/science.aaw2104.

Since the Cerenkov amplification is sensitive to the phonon lifetime, the scientists expected the effects to intensify at lower temperatures due to <u>slower anharmonic decay</u>. However, as Andersen et al. reduced the temperature from 300 to 10 K, they observed a strong increase in noise – in clear contrast to the decreasing thermal noise observed at low drives (vD \leq vs), suggesting that the amplification process was limited by scattering with thermally occupied modes.



In this way, Andersen et al. <u>extensively detailed</u> how nonequilibrium dynamics stemming from electron-phonon instability could be demonstrated in a 2D material. In the experiments, the driven electron-phonon system showed rich nonequilibrium dynamics that merit further investigations using new techniques to directly characterize the phonon spectrum and gain further insights. Previous theoretical studies had predicted amplified phonons in graphene with frequencies <u>as high as 10</u> <u>THz</u>, substantially higher than those in <u>several other materials</u>.

The experimental system can offer pure electrical generation and phonon amplification in a single micrometer-scale device with wide frequency tunability. Andersen et al. envision applications that will explore coupling to a mechanical cavity to develop a <u>phonon</u> laser, and outcoupling of the amplified sound waves to far-field terahertz radiation for medical imaging and security screening imaging (due to the degree of imaging transparency offered), wireless communications, quality control and process monitoring in manufacturing applications. The results by Andersen et al. represent a promising step towards the development of new-generation active phononic and photonic devices for multidisciplinary applications in future work.

More information: 1. Electron-phonon instability in graphene revealed by global and local noise probes <u>DOI: 10.1126/science.aaw2104</u>, <u>science.sciencemag.org/content/364/6436/154</u> Trond I. Andersen et al. 12 April 2019, *Science*.

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