Observation of magnetophonon oscillations in extra-large graphene devices

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Van der Waals materials and their heterostructures offer a versatile platform for studying a variety of quantum transport phenomena due to their unique crystalline properties and the unprecedented ability in tuning their electronic spectrum. However, most experiments are limited to devices that have lateral dimensions of only a few micrometres. Here, we perform magnetotransport measurements in graphene/hexagonal boron-nitride Hall bars of varying widths and show that wider devices reveal additional quantum effects. In devices wider than ten micrometres we observe pronounced magnetophonon oscillations that are caused by resonant scattering of Landau-quantised Dirac electrons by acoustic phonons in graphene. The study allows us to accurately determine graphene’s low energy phonon dispersion curves and shows that transverse acoustic modes are responsible for most of the phonon scattering. Our work highlights the crucial importance of device width when probing quantum effects in van der Waals heterostructures and also demonstrates a precise, spectroscopic method for studying electron-phonon interactions in 2D materials.

Two-dimensional electronic systems exhibit a rich variety of quantum phenomena\textsuperscript{1,2}. The advent of graphene has not only provided a way to study those phenomena for the quasi-relativistic spectrum, but also extended their experimental range\textsuperscript{3,4}, made some observations much clearer\textsuperscript{5,6,7,8} and, of course, revealed many new effects\textsuperscript{9,10,11,12}. These advances are mostly due to graphene’s intrinsically high carrier mobility, that is preserved by state-of-the-art heterostructure engineering in which graphene is encapsulated between hexagonal-boron nitride layers\textsuperscript{13,14} and electrically tuned with atomically smooth metallic gates\textsuperscript{8,15}. Nonetheless, one of the first discoveries in quantum transport, well known for over fifty years\textsuperscript{16,17}, has remained conspicuously absent in graphene - magnetophonon oscillations\textsuperscript{18,19}.

In the presence of an applied magnetic field (B), electrons in pristine crystals become localised in closed orbits and their spectra take the form of quantised Landau levels (LLs) separated by energy gaps. However, an electrical current can still flow in the bulk due to carriers resonantly scattering between neighbouring orbits by the absorption or emission of phonons with energies equal to the LL spacing\textsuperscript{19} (Fig. 1a). In a semi-classical model, the resonant transitions occur between orbits which
just touch in real space and induce “figure of eight” trajectories (Fig. 1b), corresponding quantum mechanically to strong overlap of the tails of their wave functions in the vicinity of the classical turning points. This effect, known as magnetophonon resonance (MPR) causes magnetoresistance oscillations that are periodic in inverse magnetic field\textsuperscript{19,20}. Whereas magnetophonon oscillations have been used extensively for studying carrier-phonon interactions in bulk Si and Ge\textsuperscript{21}, semiconducting alloys\textsuperscript{18} and heterostructures\textsuperscript{22,23,24}, there have been no reported observations in any van der Waals crystal, not even graphene, despite its exceptional electronic quality.

In this Article, we consider a subtle yet crucial aspect for the design of electronic devices based on graphene, namely the lateral size of the conducting channel. It has so far remained small, only a few micrometres in most quantum transport experiments. Our measurements using graphene Hall bars of different widths show that wider samples start exhibiting pronounced magnetophonon oscillations.

**Phonon scattering in wide graphene channels**

Our experiments involved magnetotransport measurements on graphene Hall bars encapsulated by hexagonal-boron nitride, with particular attention paid to ‘wide’ devices with channel widths $W > 10 \mu m$. An optical image of one of our widest devices is shown in Fig. 1c (See Supplementary Section 1 for details of device fabrication). Because the electron-phonon coupling is so weak in graphene\textsuperscript{25}, charge carriers scatter more frequently at the device edges of micron-sized samples rather than with phonons in the bulk, especially at low temperature\textsuperscript{26} ($T$). This is evident when comparing the Drude mean free path ($L_{\text{MFP}}$) for devices of different $W$ and fixed carrier density ($n$) of holes (Fig. 1d). At 5 K, all devices exhibit size-limited mobility ($L_{\text{MFP}}/W$) because carriers propagate ballistically until they collide with the edges of the conducting channel. Even at higher $T$ (50 K), scattering is still dominated by the edges in most of our devices. Only in sufficiently wide samples ($W > 8 \mu m$) do carriers scatter with phonons in the bulk ($L_{\text{MFP}} < W$). In effect, widening the channel makes our measurement more sensitive to bulk phenomena rather than edge effects.

**Width dependent magnetophonon oscillations**

The main observation of our work is presented in Figure. 1e, which plots the resistance ($R_{xy}$) of a 15 $\mu m$ wide Hall bar (Fig. 1d) as a function of $B$, at two $T$ and fixed $n$. At 5 K we observe two distinct oscillatory features. The first, at relatively low $B < 0.2$ T, are the well-established semiclassical geometrical resonances that occur due to magnetic focussing of carriers between current and voltage probes\textsuperscript{4} (Supplementary Section 2). At higher $B$ (~ 1 T), quantised cyclotron orbits are formed and we observe 1/$B$-periodic SdH oscillations. Their origin is confirmed by noting that the value of $n = 4e/|\hbar \Delta(B^2)|$ extracted from the SdH period ($\Delta(B^2)$) agrees with that determined by Hall effect measurements (Fig. 1e inset). At 50 K, the low-field geometric oscillations remain visible although their relative amplitudes are suppressed due to the reduced carrier mean free path. However, at higher $|B| > 0.2$ T, a new set of oscillations appears with five clear maxima (indicated by red arrows in Fig. 1e). These high-$T$ oscillations are also periodic in 1/$B$ but are distinguished by their markedly slower period. In contrast to $R_{xx}$, the Hall resistance, $R_{xy}$, shows no oscillatory features and has the same value at both $T$ (Fig. 1e inset), confirming that $n$ does not change upon warming the sample.
The observation of the high-$T$ oscillations depends critically on the sample width. This is shown in Fig. 1f which plots the normalised magnetoresistance, $R_{xx}/R_{xx} (B = 0 \text{T})$, for devices with different $W$ at fixed $T$ and $n$. We note that the bulk channels in all our devices are intrinsically clean and free from defects (probed by ballistic transport experiments in Supplementary Section 3). Nonetheless, whereas these oscillations are well developed in the widest devices (resonances marked by purple arrows), they are poorly resolved for devices with $W < 8 \mu m$ and completely absent in the narrowest one ($W = 1.5 \mu m$). As described below, we identify these size dependent, high-$T$ oscillations with MPR.

Figure 1 | Size dependent magnetoresistance oscillations in mesoscopic graphene devices. a, Landau level spectra of graphene. The diagram illustrates a carrier with momentum $k_1$ (black sphere) making a transition between Landau levels (blue and red rings) by resonant absorption of a phonon (brown arrow) with momentum $q = |k_1 - k_2|$ and energy $\hbar \omega_q$. Solid black arrows represent the magnitudes of wavevectors $k_1$, $k_2$ and $q$. b, The motion of a carrier (black sphere) in real space for the resonance condition sketched in a. Red and blue circles with arrows indicate the figure-of-eight trajectories performed on the graphene lattice (grey hexagons). The green arrow indicates direction of the applied electric field ($E$). c, optical image of a graphene device with $W = 15 \mu m$. The edges of the mesa are contoured by the white solid line. d, Open circles plot experimentally determined Drude mean free path $L_{\text{MFP}}$ as a function of $W$ for two $T$ and fixed $n = -2 \times 10^{12} \text{cm}^{-2}$. Black dashed line traces points on the axis where $L_{\text{MFP}} = W$. Solid green line marks the phonon-limited mean free path ($L_{\text{e-ph}}$) at 50 K. Our measurements focussed on the valence band because our wide devices exhibited higher electronic quality for hole doping e, magnetoresistance data $R_{xx} (B)$ for fixed $n = -3.3 \times 10^{12} \text{cm}^{-2}$ measured in our wide device (c) at two different $T$. 50 K curve is off-set vertically for clarity. Inset: Hall resistance $R_{xy} (B)$ measured simultaneously as $R_{xx}$. The solid blue and dashed red lines are data measured at 5 and 50 K respectively. f, $R_{xx}/R_{xx} (B = 0 \text{T})$ measured at fixed $n$ and $T$ in several devices of different $W$. The data close to zero-$B$ is shaded for clarity because it contains semi-classical effects.$^4$

A defining feature of magnetophonon oscillations is their unique non-monotonic temperature dependence, in which their amplitude first increases with $T$ and then decays.$^{24}$ Fig. 2a shows the temperature dependence of $R_{xx} (B)$ for fixed $n$ between 5 and 100 K (5 K steps) for another wide Hall bar device ($W = 15 \mu m$). In this sample, weak magnetophonon oscillations already
appear at 5 K in the field range between the geometric and the SdH oscillations. The resonances are labelled \( p = 1 \) to 5, where the integer \( p \) refers to the number of LL spacings that are crossed during the transition; \( p = 1 \) corresponds to scattering between LLs adjacent in energy (Fig. 1a). With increasing \( T \), the magnetophonon oscillations become more pronounced as more phonons are thermally activated, whilst the SdH oscillations are strongly suppressed. Although both phenomena require carriers that exhibit coherent cyclotron orbits (\( \mu B > 1 \), where \( \mu \) is the carrier mobility), MPR is not obscured by smearing of the Fermi–Dirac distribution across Landau gaps, rather it is enhanced due to an increased number of unoccupied states into which carriers can scatter. Hence magnetophonon oscillations persist to higher \( T \) than SdH oscillations. However, they are eventually damped at high enough \( T \) (Fig. 2b) when LLs become broadened by additional scattering (\( \mu B \sim 1 \)). This non-monotonic behaviour is better visualised in Fig. 2c which plots the oscillatory amplitudes (\( \Delta R_{\text{xx}} \)) as a function of \( T \). Notably, the amplitude of all resonances peak at \( T \) below 60 K, corresponding to a thermal energy of a few meV.

**Figure 2** | Temperature dependence of the magnetophonon effect. a, Magnetoresistance \( R_{\text{xx}}(B) \) for \( T \) between 5 K (blue curve) and 100 K (black curve) in 5 K steps for fixed \( n \) measured in our second hall bar with \( W = 15 \ \mu \text{m} \). b, Extended data set of a showing high \( T \) behaviour (10 K steps). c, Temperature dependence of the amplitude of MPRs, \( \Delta R_{\text{xx}}(T) \), indicated in a by colour coded letters, \( p = 1 - 5 \).

**Magnetophonon resonance spectroscopy**

For the doping levels and \( B \)-fields at which the oscillations occur, the charge carriers occupy high-index LLs (\( N \sim 20 \) for \( p = 1 \)) separated by small energy gaps (\( \sim 5 \) meV) with a classical cyclotron radius up to \( R_c \sim \hbar k_F/eB \sim 300 \) nm, where \( k_F \) is the Fermi-wave vector. Resonant Inter-LL transitions can therefore only be achieved by scattering with low-energy acoustic phonons that induce figure of eight trajectories (Fig. 1b). This involves phonons of specific momentum \( q = 2k_s \sim 10^9 \) m\(^{-1}\) and energy \( \hbar \omega_q(2k_s) \sim 5 \) meV that scatter carriers between LLs (Fig. 1a). Energy and momentum conservation for such a process requires that \( E_{\text{mp}} - E_N = \hbar \omega_q (2k_s) \), where \( E_N \) is the energy of an electron in the \( N^{\text{th}} \) LL, so that we find resonances occur at \( B \) values given by

\[
B_p = \frac{n \hbar \omega_q}{p e v_F} \tag{1}
\]
Here, \( v_F \) and \( v_s \) are the Fermi-velocity and low-energy acoustic phonon velocity in graphene respectively. This resonant condition is unique to massless Dirac electrons and is strikingly different to the case of massive electrons in a 2DEG \(^{23}\) where \( B_p \) scales with \( n^{0.5} \). On resonance, inelastic scattering between neighbouring orbits (Fig. 1b) gives rise to a finite and dissipative current in the bulk. This resistive behaviour causes maxima in \( \rho_{xx} \) at \( B_p \); the \( 1/B \) periodicity results in oscillations described by \( \Delta \rho_{xx} \sim e^{\gamma/B} \cos(2\pi B F / B) \) where \( B_F \equiv p B_p \) and the factor \( \gamma \) depends on temperature \(^{27}\). Equation (1) predicts that the position of maxima scales linearly with \( n \). With this in mind, Figs. 3a,b plot maps of \( R_{xx} (n, B) \) for one of our 15 \( \mu \)m devices at 5 K (Fig. 3a) and 50 K (Fig. 3b). In addition to the typical Landau fan structure that is dominant at low \( T \) (filling factors, \( \nu \), are marked by blue dashed lines), the maps reveal a broader set of fans at lower \( B \) that are more prominent at 50 K (Fig. 3b). They are caused by MPR (\( p \) values are labelled in red) and demonstrate that their frequency scales linearly with \( n \). Furthermore, the positions of MPR peaks in Fig. 3b can be fitted precisely by equation (1) (red dashed lines) with a constant \( v_s/v_F = 0.0128 \). By studying the temperature dependence of SdH oscillations \(^{28,29}\), we extracted \( v_F = 1.06 \pm 0.05 \times 10^6 \text{ ms}^{-1} \) and, accordingly, determined the acoustic phonon velocity \( v_s = 13.6 \pm 0.7 \text{ kms}^{-1} \). This value is in very good agreement with the speed of transverse acoustic (TA) phonons in graphene calculated by density functional perturbation theory \(^{30,31}\) (\( \sim 13 \text{ kms}^{-1} \)), from which we infer our oscillations arise from inter-LL scattering by low energy and linearly dispersed TA phonons.

![Figure 3](image-url)  
**Figure. 3 Density dependence of magnetophonon oscillations.** a, longitudinal resistance \( R_{xx} \) (grey scale map) as a function of \( n \) and \( B \) measured at 5 K (\( W = 15 \mu \)m). Logarithmic grey scale; White: 1 \( \Omega \) to black: 15 \( \Omega \). The blue dashed lines trace Landau gaps corresponding to high filling factors \( \nu = nh/Be \). b, same as a measured at 50 K. Logarithmic grey scale; White: 5.5 \( \Omega \) to black: 18 \( \Omega \). The red dashed lines plot equation (1) for \( p = 1 \) to 4 which corresponds to electrons scattering between 1 to 4 Landau Levels. Features appearing for \( B < 0.2 \text{ T} \) are the semi-classical geometrical oscillations \(^4\) not relevant in this work (see Supplementary Section 2 for details).
Equation (1) is generic for linearly dispersed phonons in graphene. This motivated us to search for MPR arising from longitudinal acoustic (LA) phonons, which should occur at higher $B$ due to their significantly higher $v_p^{30,31}$. Careful inspection of the data in Fig. 1-2 shows that the last resonance for TA phonons is followed by a weak shoulder-like feature. For further investigation, we studied a dual-gated graphene device that permitted measurements at higher $n \sim 1 \times 10^{13}$ cm$^{-2}$ which, according to equation (1), should separate this feature from the TA resonances. Fig. 4a plots $R_\alpha (B)$ for this device for several $n$. Measurements at these high $n$ reveal the shoulder-like feature developing into a well-defined peak (indicated by coloured arrows). Its position ($B_{p=1}$) is accurately described by equation (1) with $v_p = 21.0 \pm 1.0$ kms$^{-1}$. Given this value is comparable with that calculated for LA phonons in graphene$^{30,31}$, we attribute this feature to inter-LL scattering by LA phonons.

Further validation of our model is presented in Fig. 4b, which plots the magnetophonon oscillation frequency for TA phonons ($B_\parallel \equiv pB_{\parallel}$) as a function of $n$ for several different devices (red symbols). It shows a linear dependence (red line) fits the data to equation (1) from all our measured devices over a range of $n$ spanning an order of magnitude. Even the weaker LA resonance was found to occur at the same $B_{p=1} = B_\parallel$ in different devices (blue symbols). Furthermore, the data in Fig. 4b can be transformed directly into phonon dispersion curves (Inset of Fig. 4b) by noting that $q = 2k_F = 2(n\pi)^{0.5}$ and $\hbar\omega_q = (2eB_F v_F \hbar)^{0.5}$. The extended tunability of the carrier density in our dual-gated devices allows measurement of phonon branches up to wave vectors $> 10^9$ m$^{-1}$. Note, the obtained dispersions are significantly more precise than those measured by X-ray scattering experiments in graphite$^{32}$ (purple stars). Studies of magnetophonon oscillations thus enable an all-electrical measurement of the intrinsic phonon dispersion curves in gate-tuneable materials.

Discussion

To understand why magnetophonon oscillations are absent in narrow samples, we first note that figure of eight trajectories (Fig. 1b) have a spatial extent $\sim 4R_c$ which can reach values of several microns for high-order resonances ($p > 3$). If the sample is too narrow, so that $4R_c$ is comparable to $W$, carrier’s trajectories are skewed by elastic scattering at device edges. In this case, they propagate on the edges of the device along skipping orbits$^2$, effectively “short-circuiting” the resistive behaviour of the bulk caused by MPR. However, if $W > 4R_c$, both MPR and skipping orbits contribute to $R_\alpha$. We can estimate the width of the device required to observe MPR by comparing the relative contributions of these two processes. Carriers that diffuse in MPR-induced figure of eight trajectories move a distance $2R_c$ in a characteristic time, $\tau_{e-ph} = L_{e-ph}/v_F$ with a drift velocity $v_{MPR} = 2R_c/\tau_{e-ph}$. This is significantly slower than skipping orbits which can have speeds approaching $v_F$. On the other hand, skipping orbits occupy only a width $\sim R_c$ at each edge, whereas MPR occurs approximately over the full width, $W$, of the bulk. By comparing these two contributions, we deduce that MPR dominates when $Wv_{MPR} \gtrsim 2R_cv_F$. This corresponds to the condition $W \gtrsim L_{e-ph}$ in good agreement with the measured data in Figs. 1d, f.

Our measurements provide an important insight into the intrinsic electron-phonon interaction in graphene: namely, the dominance of carrier scattering by low-energy TA phonons. This is in agreement with recent theoretical work$^{31,33}$ and contrasts with a widely held view that deformation potential scattering by LA phonons prevail over TA phonons$^{34}$. To investigate this point further, we calculated magnetoresistance using the Kubo formula$^{35}$ (Supplementary Section 5). A typical
calculation is shown in Fig. 4c, which plots the contribution \( \Delta \rho_{\alpha \nu} \) of MPR for TA and LA phonon velocities of \( v_s = 13.6 \) and 21.4 kms\(^{-1} \), respectively\(^{21} \) and Fermi velocity\(^{36} \) \( v_F = 1 \times 10^6 \) ms\(^{-1} \). It accurately describes the oscillatory form of the measured data. Such good agreement was only possible if our calculations included the effect of carrier screening\(^{31,33,37,38} \) which significantly reduces the electron-LA phonon deformation potential coupling. Without screening, LA phonons would dominate the observed MPRs (Supplementary Section 5). Our results therefore highlight the importance of carrier screening on electron-phonon interactions and thus helps resolve a longstanding discussion of the relative importance of LA\(^{39,34} \) and TA\(^{38,33} \) phonon scattering in graphene.

**Conclusion**

To conclude, we report the first observation of magnetophonon oscillations in graphene, where the Dirac spectrum strongly modifies the resonant condition with respect to the previously studied electronic systems. Many other two-dimensional crystals can also be expected to exhibit this phenomenon. The oscillations allow direct access to low-energy acoustic-phonon modes that are generally inaccessible by Raman spectroscopy\(^{40,41} \). Our measurements combined with the Kubo calculations provide strong evidence that TA phonons limit temperature-dependent mobility in graphene\(^{31,33} \). Most importantly, graphene’s transport properties are shown to strongly depend on device’s size, even when using conducting channels as wide as several microns. This should motivate further experiments on graphene and related two-dimensional materials in the macroscopic regime, rather than using the current devices that remain essentially mesoscopic.

![Figure 4](image_url)

**Figure. 4 Phonon spectroscopy in graphene by measurement of MPR.** a, Longitudinal resistance \( R_{xx} \) as a function of \( B \) measured for several high \( n \) of holes in our wide \( (W = 13.8 \ \mu \text{m}) \) dual-gated graphene Hall bar. The curves have been off-set for clarity. b, Red symbols plot the fundamental frequency \( B_p \equiv p B_p \) of magnetophonon oscillations as a function of absolute \( n \) for three different devices; open circles correspond to the dual-gated device which allowed high doping. The blue symbols mark the positions \( B_{p c 2} \) of the broad peak which appears clearly at high \( n \) (indicated by
coloured arrows in a). The red solid line represents equation (1), with $v_F = 1.06 \times 10^6 \text{ ms}^{-1}$, and $v_s = 13.6 \text{ kms}^{-1}$. The blue solid line is plotted for the same $v_F$ but with $v_s = 21.0 \text{ kms}^{-1}$. Inset: Data in main panel transformed to phonon dispersion curves. Coloured squares – experimental data points (error bars reflect the measurement error in $v_s$), solid lines plot the equation $\hbar \omega_q = \hbar v_s q$ (same $v_s$ as in main panel), purple stars – data taken from ref. 32. c, calculation of the oscillatory part of the resistivity $\Delta \rho_{xx}(\Omega)$ using the Kubo formula (see Supplementary Section 5 for details).

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Supplementary Information

1 Device Fabrication

The hexagonal boron nitride (hBN) encapsulated graphene heterostructures were assembled using a dry-peel transfer method. Graphene and hBN flakes were first exfoliated onto O$_2$/Ar plasma cleaned SiO$_2$/Si substrates. The appropriate flakes were then identified by long exposure dark field optical imaging. For the hBN encapsulation layers, we used flakes that were 25 – 100 nm thick. The selected flakes were then assembled using a polypropyl carbonate (PPC) coated Polydimethylsiloxane (PDMS) stamp placed on a glass slide attached to a high precision XYZ micromanipulator$^1$. First, the top hBN encapsulation layer was picked up using the PPC/PDMS stamp. This was done at a fixed substrate temperature $\sim$ 50ºC. The hBN flake attached to the stamp was then used to pick up the graphene flake. During this process, the substrate was heated to 65ºC whilst contacting hBN to graphene and then cooled to 50ºC for peeling the resultant hBN/graphene heterostructure from the substrate. The stack was then placed on the bottom hBN flake (substrate temperature at 65ºC) to fully encapsulate the graphene layer.

Once the heterostructure was prepared, we performed standard electron beam lithography techniques to create the Hall bar geometry. First, we patterned a polymethyl methacrylate (PMMA) mask on the stack to define contact regions leading up to the device channel. The regions unprotected by the mask were etched away using CHF$_3$ + O$_2$ reactive ion etching (RIE), forming narrow trenches. Metal contacts (5 nm Cr/70 nm Au) were then evaporated into the trenches which form high-quality contacts to the graphene edge$^{2,3}$. Next, the same lithography and RIE etching procedures were used to pattern the Hall bar mesa. For one of our devices, we also patterned a metallic top gate above the heterostructure (Fig S1) which allowed us to achieve higher doping levels.

Figure S1; Wide graphene Hall bars. a, Optical image of our wide top-gated device ($W = 13.8$ µm). White scale bar is 10 µm.
2 Magnetic Focussing in wide graphene Hall bars

At low temperature, the charge carriers in our graphene/hBN devices propagate ballistically for several micrometers before scattering. For certain measurement geometries, the 4-probe resistance is governed by direct transmission of charge carriers between current and voltage probes. An example is illustrated in Figure S2a which describes the measurement scheme for a transverse magnetic focussing experiment. In the presence of a perpendicular magnetic field ($B$), ballistic carriers injected from the current source follow curved trajectories and the measured 4-probe resistance ($R_{TMF}$) exhibits maxima for particular values of $B$ when the carriers are focussed directly into the collector voltage probe (coloured arrows in Fig. S2a). This occurs when the cyclotron radius ($R_c \sim \hbar k_F/eB$) becomes commensurate with the distance ($L$) between the current injector and voltage collector ($L$). The resonance condition is given by

$$B = \frac{2\hbar k_F}{eL} s,$$  \hspace{1cm} (S1)

where $k_F$ is the Fermi wave-number and ($s-1$) is an integer number that describes the number of reflections at the device edge. For example, $s = 2$ describes the trajectories traced by the red arrows in Fig. S2a. Figure. S2b shows $R_{TMF}(B)$ at fixed $n$ measured in the geometry illustrated in Fig. S2a. For negative values of $B$, we find a set of maxima that are equally spaced with a period $\Delta B = 2\hbar k_F/eL$. No resonances are observed for positive $B$ because the Lorentz force acts in the opposite direction and bends trajectories away from the voltage collector. Figure. S2c plots maps of $R_{TMF}(B,n)$ for different hole doping. We find the resonances are shifted to higher $B$ for higher $n$, in agreement with Eq. (S1) and the fact that the cyclotron radius is larger for higher $n$. Notably, all the resonances can be described by Eq. (S1) with $L = 7.4$ $\mu$m (dashed lines in Fig. S2c). This value corresponds to the distance between the current injector and voltage collector (labelled in Fig. S2a) and validates the dependence of the oscillatory features given by Eq. (S1).

Magnetic focussing resonances can also appear in a standard longitudinal resistance measurement ($R_{xx}$) if the voltage probes are located too close to the current contacts (see main text). This occurs if they are closer than the width of the channel and is typically the case in our wider samples (Fig. S2d) because of the limited size of exfoliated flakes and clean areas available for patterning devices. Figure. S2e plots $R_{xx}(B)$ for a fixed $n$ and $T$ in non-quantizing $B$ fields. We find a set of resonances that are periodic in $B$ and occur at higher $B$ for higher $n$ (Fig. S2f). This behaviour resembles that of magnetic focussing although these curves are distinct from a typical measurement (Fig. S2a-c). First, the current injectors are rather wide ($15$ $\mu$m). Second, the oscillations are slightly phase shifted; whereas resonances are equally spaced with a period $\Delta B$, the first (labelled 1 in Fig. S2e) occurs at a value approximately $\Delta B/2$. To understand the origin of those resonances, we used Eq. (S1) to extract $L$ from the oscillation period $\Delta B = 2\hbar k_F/eL$. We carried out this analysis for several different doping levels (Fig. S2f) and found approximately the same $L \approx 5.6$ $\mu$m. This corresponds roughly to the distance between current and voltage probes and suggests that the dominant contribution to the measurement originates from magnetic focussing where carriers are injected from the corners of the device (yellow arrows in Fig. S2d). Further work is required to understand the details of these magnetic focussing resonances.
**Figure. S2.**

**a**, Measurement scheme for a typical transverse magnetic focussing experiment performed in a graphene device with $W = 15$ μm. Yellow and red lines trace trajectories of electrons under the resonance condition (1) for $s = 1$ and 2 respectively. **b**, $R_{\text{TMF}}(B)$ for fixed $n$ and $T$. **c**, Magnetic focussing maps, $R_{\text{TMF}}(B, n)$, for the configuration specified in **a**. Dashed lines are fits to Eq. (S1) with $L = 7.4$ μm and different $s$. **d** Measurement scheme for $R_{xx}$ geometry. **e**, $R_{xx}(B)$ for a fixed $n$ and $T$. **f**, maps $R_{xx}(B, n)$.

### 3 Quasi-ballistic device channels

Figure. 1f of the main text shows that magnetophonon oscillations in graphene appear only in samples in which the channel has a sufficiently large width ($W$). We claim that the size is the important variable because all our devices exhibit similar electronic quality in the bulk; their conducting channels are relatively free from defects/impurities so that low-temperature mobility is limited only by dissipation at the edges$^5$. To prove this, we performed magnetic focussing experiments (described in Supplementary Section 2) in all our devices. The observation of magnetic
focussing resonances requires carriers that propagate ballistically without scattering, thus proving there are no scattering centres along the path between the current injector and voltage collector. Here we present data taken from transverse magnetic focussing experiments performed in our narrowest \((W = 1.5 \, \mu m)\) and widest \((W = 15 \, \mu m)\) samples. The measurement geometries for each device are sketched in Fig. S3a-b. Figure S3c plots \(R_{TMF} (n,B)\) measured in the narrowest device in which injector and collector probes are separated by \(L = 1.5 \, \mu m\). We find pronounced magnetic focussing resonances up to the fourth order \((s = 4)\). We note that similar resonances also appear between any pairs of contacts located in different regions of the device, thus providing further confirmation of the quality and ballistic nature of our channels. Even in our widest samples (Fig. S3b) we detect ballistic electrons focused at voltage probes 20 \(\mu m\) away from current injector (Fig. S3d).

Figure S3. a,b, Measurement scheme for a magnetic focussing experiment performed in our narrowest (a) and widest (b) Hall bar devices (the mesa are contoured in white). The yellow, red and blue curved arrows indicate electron trajectories corresponding to resonances predicted by Eq (S1) with \(s = 1, 2\) and 3 respectively. c,d Transverse magnetic focussing maps, \(R_{TMF} (n, B)\) measured at 5 K in the configurations shown in a and b respectively.
4 Semiclassical model of magnetophonon resonance in monolayer graphene

The magnetoresistivity of the device is given by the approximation

\[ \rho_{yy} \approx \frac{\sigma_{xx}}{\sigma_{yy}} \approx \rho_{xx}, \quad (S2) \]

since the Hall component of the conductance tensor, \( \sigma_{xy} \), is for these experimental conditions.

The longitudinal conductance tensor component, \( \sigma_{xx} \), is determined by the rate of drift of a carrier’s cyclotron orbit centre. This process is illustrated semiclassically in Fig. 1b of the main text which shows the shift caused by an inelastic scattering-induced figure-of-eight transition in \( k \)-space. The energy absorbed (or emitted) by the carrier undergoing an inter-Landau level (LL) transition is given by the energy difference between its initial \( N \) and final \( N' \) states, where \( N \) is the LL index and \( p \) is a positive integer. The quantised energy spectrum of monolayer graphene is given by

\[ E_N = \text{sgn}(N) \sqrt{2|N|} \frac{\hbar v_F}{l_B}, \quad (S3) \]

where \( v_F = 10^6 \text{ ms}^{-1} \) is the Fermi velocity of graphene and \( l_B = \sqrt{\hbar/eB} \) is the quantum magnetic length. For a figure-of-8 transition, an electron in a LL with index \( N \) and orbit radius in \( k \)-space given by \( \kappa_c = \sqrt{2N}/l_B \) makes an inelastic transition to a level with index \( N' = N \pm p \) by absorbing or emitting a phonon with wave vector, \( q \), so that its final radius \( \kappa' = q - \kappa_c \). In real space, the corresponding classical orbits have radii \( R_c = l_B \kappa_c \). The wavevector, \( q \) induces a shift in real space of the orbit centre \( \Delta X = l_B^2 q \) and hence provides a contribution to the current. An excellent fit to the measured period of the magnetophonon oscillations is obtained by considering scattering by linearly dispersed acoustic phonons with energy \( \hbar \omega = \hbar v_s q \), where \( v_s \) refers to velocity of either longitudinal acoustic (LA) or transverse acoustic (TA) phonons. The resonant condition for absorption or emission processes are given by

\[ \hbar \omega = \frac{\hbar v_s}{l_B} \left( \sqrt{N \pm p} + \sqrt{N} \right) = \pm \frac{\hbar v_s}{l_B} \left( \sqrt{N \pm p} - \sqrt{N} \right), \quad (S4) \]

where \( p \) is the change in LL index. Since the magnetophonon oscillations occur at low magnetic fields and high electron densities, \( N \) and \( N' \gg p \); hence Eq. S4 is approximated accurately by the relation

\[ B_p = \frac{nhv_s}{pev_F}. \quad (S5) \]

Therefore, the magnetophonon resonance oscillations are periodic in \( 1/B \) with a frequency \( B_p = pB_{pr} \), as discussed in the main text.
5 Quantum transport calculations

We use the Kubo approach\(^7\) to determine the linear response of the oscillatory longitudinal magnetoresistivity, \(\rho_{xx}\), of monolayer graphene due to the resonant absorption and emission of LA and TA acoustic phonons by the charge carriers in a magnetic field, \(\mathbf{B} = (0,0,-B)\), applied perpendicular to the graphene sheet. The model corresponds to ohmic conditions with the carriers in thermal equilibrium with the lattice vibrations. The electronic spectrum becomes quantised into a series of unevenly spaced Landau levels (LLs) with index \(N\) given by Eq. (S3). It is convenient to use the Landau gauge where \(\mathbf{A} = (0, -Bx, 0)\). The carrier wave function in the \(K^+\) valley and the conduction band is then given by the pseudospinor

\[
\psi_N^{K^+} = \frac{1}{\sqrt{\pi}} \left( -\text{sgn}(N) \phi_N^{|x-X\rangle} \right),
\]

where \(\phi\) are simple harmonic oscillator states along \(x\) and plane waves along \(y\) given by

\[
\phi_N(x) = A_N H_N \left( \frac{x}{\ell_B} \right) \exp \left( \frac{x^2}{\ell_B^2} \right) \exp(ik_y y),
\]

Here \(A_N = \left( \sqrt{\frac{2}{\ell_B^2} 2^N N! \sqrt{\pi}} \right)^{-1}\) is a normalisation constant and \(H_N\) are the Hermite polynomials\(^8,9,10\). A similar relation applies for the valence band and the \(K^-\) valley. In the Kubo approach, the contribution of the TA and LA acoustic phonon scattering to the magnetoconductance \(\sigma_{xx}\) is given by

\[
\Delta \sigma_{xx}^{t,l} = \frac{g_v g_s \pi \hbar^2}{8 e^2} \sum_q \left( \frac{\beta_{qy}}{\ell_B} \right)^2 |C(q)|^2 N_q (N_q + 1) \sum_{N,N'} \sum_{k_y,k_y'} \left[ f(E_i - \hbar \omega_q^{t,l}) - f(E_f) \right] \delta(E_i - \hbar \omega_q^{t,l} - E_N) \left| \tilde{l}_{N,N'}^{t,l}(k_y,k_y',q) \right|^2
\]

Here the subscripts superscripts \(t\) and \(l\) refer to contributions corresponding to the TA or LA phonons respectively, \(g_v = 2\) and \(g_s = 2\) are the valley and spin degeneracies, \(S = L_L\) is the area of the device, \(\ell_B\) is the Boltzmann constant, \(T\) is the lattice temperature, \(|C_{t,l}(q)|^2 = \hbar q / (2 \nu_{t,l})\) are the Fourier components of the scattering potential, \(\rho = 7.6 \times 10^{-8} \text{ g cm}^{-2}\) is the mass density of graphene, \(N_q = (\exp(\hbar \omega_q^{t,l} / k_B T) - 1)^{-1}\) is the Bose-Einstein distribution function for the phonons and \(f(E) = (\exp((E - \mu) / k_B T) + 1)^{-1}\) is the Fermi-Dirac distribution of the carriers with chemical potential, \(\mu\). The matrix elements \(\tilde{l}_{N,N'}^{t,l}(q)\), are given by

\[
\tilde{l}_{N,N'}^{t,l}(k_y,k_y',q) = \int dS \psi_{k_y,N}^* \psi_{k_y',N'} V_q^{t,l} \psi_{k_y',N'},
\]

\(V_q^{t,l}\) describes the charge-carrier phonon coupling for the TA and LA phonons respectively\(^11,12,13,14\) and has the form

\[
V_q^{t,l} = e^{i q r} \begin{pmatrix} 0 & -g_g e^{i2 \varphi} \\ g_g e^{-i2 \varphi} & 0 \end{pmatrix}
\]

(S10)
\[ V_q^i = i e^{i q \cdot r} \begin{pmatrix} g_d(q) & g_g e^{i 2 \varphi} \\ g_g e^{-i 2 \varphi} & g_d(q) \end{pmatrix}, \] (S11)

where \( \varphi \) is the angle between the phonon wave vector and the \( x \) axis, which in our model is defined to be along the zigzag edge of the graphene layer. Here, \( g_g \) and \( g_d(q) \) are the carrier phonon coupling matrix elements corresponding to the gauge and deformation distortions of the graphene lattice. The off-diagonal matrix elements involving \( g_g \) arise from pure shear-like distortions of the lattice which give rise to a “synthetic” gauge field in the Dirac equation. This is unaffected by screening and has been estimated using density functional theory to have a value in the range \( g_g = 1.5 - 4.5 \) eV\(^{11,12,13,14,15} \). We obtain a good fit to the data with \( g_g = 4 \) eV. The on-diagonal terms correspond to strain-induced distortions of the unit cells that change their areas, resulting in local redistributions in the carrier density, \( n \), which screen the deformation potential terms \( g_d(q) \). Therefore we write

\[ g_d(q) = \frac{\tilde{g}_d}{\epsilon(q)}, \] (S12)

where \( \tilde{g}_d \) is the unscreened “bare” deformation potential coefficient and \( \epsilon(q) \) is the phonon wave vector-dependent dielectric function. We use the Thomas-Fermi approximation for \( \epsilon(q) \) which gives

\[ \epsilon(q) = \epsilon_r \left(1 + \frac{q_{tf}}{q}\right), \] (S13)

where \( q_{tf} = 4 e^2 \sqrt{n_i n} / (4 \pi \hbar v_F \epsilon_r \epsilon_0) \) is the inverse Thomas-Fermi screening radius. This takes into account screening by the dielectric environment of the graphene layer, with the dielectric constant \( \epsilon_r \) and by the free carriers in the graphene layer\(^{15,16} \). For free-standing graphene and \( \epsilon_r = 1 \) then \( q_{tf} \approx 8 k_F \). For the case of magnetophonon resonance at high LL index, \( q \sim 2 k_F \) and \( \epsilon(q) \sim 5 \) and thus the deformation potential is strongly suppressed\(^{15} \). In our experiments, the graphene layer is fully encapsulated by hBN and therefore we set \( \epsilon_r = 3.5 \) so that \( \epsilon(q) \sim 7.5 \) and \( g_d(q) \) is reduced further. However, we find that the resistivity is not very sensitive to \( \epsilon_r \) since the deformation potential is very effectively screened by the carriers in the graphene layer. We set \( \tilde{g}_d \) to be 25 eV. We find that in the range from \( \tilde{g}_d = 20 - 30 \) eV the resistivity is not sensitive to this parameter due to the strong screening effect.

To model LL broadening, we replace the delta function in Eq. S8 by

\[ \delta(E) \to \frac{1}{\Gamma \sqrt{2\pi}} \exp \left(-\frac{E^2}{2\Gamma^2}\right), \] (S14)

where \( E = E_N - E_{N'} - qh v_{t,i} \). We use a Gaussian function to aid convergence of our calculation at high LL indices. In the case of short-range scattering, for example by charged impurities, the broadening of the LLs depends on the square root of magnetic field\(^{6,17,18,19} \). Therefore, we set the broadening parameter \( \Gamma = \gamma \sqrt{B} \). We obtain a good fit to the data with \( \gamma = 0.5 \) meV (see Fig. 4b of the main text).

In the Hall regime, the longitudinal magnetoconductivity is given by \( \rho_{yy} = \sigma_{xx} / (\sigma_{xx} \sigma_{yy} + \sigma_{xy}^2) \), where the component \( \sigma_{xy} = ne / B \). Since \( \sigma_{xx} = \sigma_{yy} \ll \sigma_{xy} \) the oscillatory part of \( \rho_{xx} \) due to TA and LA phonon scattering is given by
to a good approximation. Figure S4 shows $\Delta \rho_{xx}$ calculated for three representative carrier densities, 6, 7.5 and $9 \times 10^{12}$ cm$^{-2}$. We find that $\Delta \rho_{xx}(B)$ has a form and amplitude which agrees well with oscillations observed in the measurements shown in the main text. The magnetic field values of the position of the peaks correspond closely to the resonance condition in Eq. 1 of the main text and Eq. S5. In particular, they are periodic in $1/B$ with a frequency, $B_r$, that has a linear dependence on $n$ (Eq. S5). The plot shows that the contribution from the LA phonons to the total resistivity is relatively small and appears only as the $p = 1$ peak in $\Delta \rho_{xx}$ (labelled by the blue arrow in Fig. S7). This is due to two main factors: first, the suppression of the deformation part of the electron-phonon coupling matrix due to free carrier screening (see Eq. S11); second, the energy of the LA phonon is larger than that of the TA phonon when the condition for magnetophonon resonance is satisfied. Therefore, at a given temperature, there is a lower population of LA phonons than TA phonons for carriers to absorb and similarly there are fewer carriers at high enough energy to emit an LA phonon compared to those that can emit a TA phonon. If the deformation part of the electron-phonon were not screened, $\rho_{xx}(B)$ would be dominated by the contribution from the LA phonons, highlighting the importance of the strong screening of LA phonon scattering in graphene.

Figure S4. Calculated $\Delta \rho_{xx}(B)$ for a three different $n$ and $T = 70$ K, using $v_t = 13.6$ kms$^{-1}$ and $v_\parallel = 21.4$ kms$^{-1}$ taken from ref. 14 and the parameters specified in the text. The integers $p$ correspond to resonant inter-LL scattering around the Fermi energy, with $p = |N - N'|$. 

$$\Delta \rho_{xx} = \left( \frac{B}{n e \sigma} \right)^2 \left( \sigma_{xx}^l + \sigma_{xx}^t \right).$$  

(S15)
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