Anomalous thermopower oscillations in graphene-InAs nanowire vertical heterostructures

Richa Mitra¹, Manas Ranjan Sahu¹, Aditya Sood², Takashi Taniguchi³, Kenji Watanabe³, Hadas Shtrikman⁴, Subroto Mukerjee¹, A.K. Sood¹, Anindya Das¹

¹ Department of Physics, Indian Institute of Science, Bangalore-560012, India.

² Stanford Institute for Materials and Energy Sciences, SLAC National Accelerator Laboratory, Menlo Park, California, 94025, USA.

³ National Institute for Materials Science, Namiki 1-1, Ibaraki 305-0044, Japan.

⁴ Department of Physics, Weizmann Institute of Technology, Israel.

E-mail: anindya@iisc.ac.in

Abstract.

Thermoelectric measurements have the potential to uncover the density of states of low-dimensional materials. Here, we present the anomalous thermoelectric behaviour of mono-layer graphene-nanowire (NW) heterostructures, showing large oscillations as a function of doping concentration. Our devices consist of InAs NW and graphene vertical heterostructures, which are electrically isolated by thin (~ 10 nm) hexagonal boron nitride (hBN) layers. In contrast to conventional thermoelectric measurements, where a heater is placed on one side of a sample, we use the InAs NW (diameter ~ 50 nm) as a local heater placed in the middle of the graphene channel. We measure the thermoelectric voltage induced in graphene due to Joule heating in the NW as a function of temperature (1.5K - 50K) and carrier concentration. The thermoelectric voltage in bilayer graphene (BLG)- NW heterostructures shows sign change around the Dirac point, as predicted by Mott's formula. In contrast, the thermoelectric voltage measured across monolayer graphene (MLG)-NW heterostructures shows anomalous large-amplitude oscillations around the Dirac point, not seen in the Mott response derived from the electrical conductivity measured on the same device. The anomalous oscillations are a signature of the modified density of states in MLG by the electrostatic potential of the NW, which is much weaker in the NW-BLG devices. Thermal calculations of the heterostructure stack show that the temperature gradient is dominant in the graphene region underneath the NW, and thus sensitive to the modified density of states resulting in anomalous oscillations in the thermoelectric voltage. Furthermore, with the application of a magnetic field, we detect modifications in the density of states due to the formation of Landau levels in both MLG and BLG.

1. Introduction

Over the years, dimensionally mismatched two-dimensional (2D) - one-dimensional (1D) heterostructures [1] have demonstrated diverse set of advanced functionalities of the

heterojunctions [2, 3, 4, 5]. Recently, these systems have emerged as a fertile ground for realizing novel phenomena like anomalous Coulomb drag [6], and formation of 1D waveguides in 2D materials [7]. The latter has opened new avenues for applications, where charge carriers in 2D systems are guided through 1D cavities [7], holding potential as a method for transmitting information, analogous to photons in optical fibers. Moreover, such engineering of reduced dimensionality offers a strategy to enhance the thermopower (or Seebeck coefficient) of a material [8, 9, 10, 11]. However, to realize the true potential of these reduced dimensions and manipulate them further, it is important to probe the modulation in the local density of states (DOS), for which non-invasive probes are essential. In this regard, non-invasive thermo-electric measurements are suitable tools [12, 13, 14, 15, 16, 17], which can be employed for mixed-dimensional systems. In comparison to quantum capacitance [18, 19] and scanning microscopy techniques [20, 21, 22, 23, 24, 25, 26, 27, 28], thermopower measurements can be easily implemented in 2D-1D systems.

In conventional thermoelectric measurements [15, 29, 30], a heat source is typically placed a few microns away and electrically isolated from the actual device to create a spatially-uniform temperature gradient. In our devices, an InAs nanowire (diameter ~ 50 nm) placed vertically on the graphene channel (almost in its center) and separated by a thin hexagonal boron nitride spacer (hBN ~ 10 nm) acts like a local nano-heater. Passing a current through the nanowire (NW) generates heat, and its close proximity leads to heat transfer into the graphene channel and hence a finite thermoelectric voltage. By utilizing this unique heating method, we observe unprecedented large oscillations in the thermoelectric voltage measured across monolaver graphene (MLG) at low temperatures (~ 1.5 K to 20K) as a function of the carrier concentration around the Dirac point. Notably, no oscillations are seen in the Seebeck coefficient calculated using Mott's formula based on the measured resistance. The magnitude of oscillations in the MLG devices reduces with increasing temperature and qualitatively follows the trend of Mott's formula at higher temperatures (> 20 K). In contrast, for bilayer graphene (BLG) devices, the thermoelectric voltage does not show oscillations and follows Mott's prediction up to the lowest temperature (1.5K), with an expected sign change around the Dirac point. Notably, our observation differs from the oscillations observed in ref [15] where the Universal conductance fluctuations (UCF) are manifested as oscillations both in resistance as well as in thermopower at low temperatures.

We explain the observed oscillations of the thermopower with density near the Dirac point, as a consequence of changing effective carrier type inside the graphene channel. We propose that this can arise due to the 1D confinement of carriers in graphene, and subsequent formation of sub-bands that modify the local DOS. To support this proposal, we analyze the carrier density profile in the graphene channel underneath the NW and show that the electrostatic potential of the NW creates a cavity for the charge carriers in graphene, resulting in a modulation of local DOS and hence large oscillations in thermoelectric voltage. To understand why the oscillations are seen only in the thermoelectric response and not in the resistance data, we calculate the

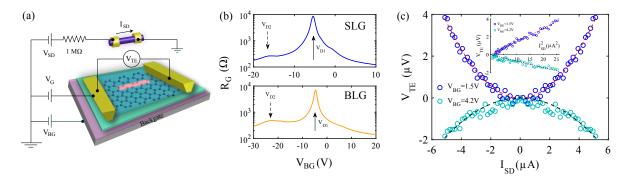


Figure 1: (a) Device schematic for measuring thermoelectric voltage in graphene. The heterostructure consists of an InAs NW on top of a hBN encapsulated graphene stack assembled on Si/SiO₂ substrate. Thermoelectric voltage (V_{TE}) is measured across two probes on graphene while a constant DC current (I_{SD}) is passed through the NW. Passing current through the NW heats the region of graphene below the nanowire, and gives rise to a finite thermoelectric voltage in graphene. In this setup carrier density in graphene (n_G) can be tuned with the backgate voltage (V_{BG}) . Voltage V_G is applied to the graphene to tune the density of the NW. (b) 2-probe resistance of graphene (R_G) versus backgate voltage for MLG (upper panel) and BLG (lower panel). V_{D2} and V_{D1} indicate (shown by arrows) gate voltages corresponding to charge neutrality points for graphene beneath the NW and for rest of the graphene channel, respectively. (c) V_{TE} plotted with I_{SD} at T=1.5K for two different $V_{BG} = 1.5V$ and $V_{BG} = 4.2V$ indicated by dark blue and sky blue open circles, respectively. The red and black dashed lines are parabolic fits to the data. V_{TE} plotted with I_{SD}^2 in the inset. The linear fits (red and black dashed lines) show that V_{TE} has thermoelectric origin.

temperature profile in the NW-graphene heterostructures. These calculations reveal that the temperature gradient exists predominantly in the region of the graphene that is underneath the NW. The resulting sub-bands from the confining potential give rise to large DOS at certain fillings and hence a high thermoelectric voltage. The absence of such thermoelectric oscillations in NW-BLG heterostructures, due to the weaker confinement, confirms the validity of our physical picture. However, the observed aperiodicity in the thermopower oscillations suggests that the dominant electron-hole puddles near the Dirac point in monolayer graphene may have additional contribution to the oscillations. As a step forward, we measure the thermoelectric voltage in the presence of a perpendicular magnetic field and observe periodic oscillations in both the MLG and BLG devices due to the formation of Landau levels (LL). This further confirms our model which suggests that the formation of a confinement potential gives rise to DOS modulation, and subsequent oscillations in thermoelectric voltage in MLG at zero magnetic field.

2. Experimental details

In this section we describe the device structure of the MLG-NW and BLG-NW devices. The hybrid heterostructures are fabricated by transferring an InAs NW on top of hBN encapsulated MLG or BLG assembled on Si/SiO_2 substrate using a dry transfer technique [31, 32]. The NW and the graphene are separated by a thin layer of hBN

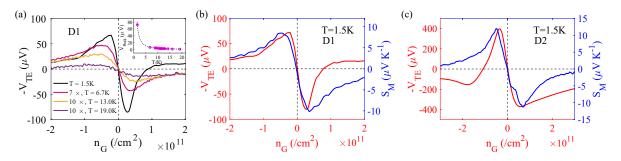


Figure 2: Thermoelectric response of NW-BLG devices: (a) V_{TE} versus n_G plotted for different temperatures. The data at 6.7K, 13K and 19K are magnified by 7x, 10x and 10x, respectively. The horizontal and vertical dashed lines indicate the zero voltage and the zero density levels, respectively. V_{TE} flips sign across the charge neutrality point. (Inset) The magnitude of the dip in V_{TE} on the hole side, is plotted versus temperature. The dashed line is a fit to the eye. (b) and (c) are comparison between V_{TE} and Seebeck coefficient for two devices D1 and D2 respectively. The blue lines represent the Seebeck coefficient (S_M) calculated using the Mott formula (Eqn. 1) at T = 1.5K versus n_G . The red lines are V_{TE} versus n_G at T=1.5K for D1 and D2 devices respectively. For BLG, the shape of the V_{TE} matches well with the Seebeck coefficient S_M .

(~ 10nm). The lengths of the NW and graphene channels are ~ 0.4-0.6 μ m and 10-12 μ m, respectively, and the width of the graphene is ~ 10-15 μ m. As shown in Fig. 1a, a constant current I_{SD} passes through the NW when a DC voltage is applied across it through a resistor. Joule heating in the NW creates a temperature gradient in graphene from the position of the NW towards the colder graphene probes. We measure the open circuit voltage (V_{TE}) across the graphene channel as shown in Fig. 1a. The carrier concentration in the graphene (n_G) and NW are tuned by the backgate voltage (V_{BG}) and graphene gate voltage (V_G) , respectively. The 2-probe resistance (R_G) of MLG and BLG as a function of V_{BG} are shown in upper and lower panels of Fig. 1b, respectively. The two arrows indicate V_{D2} and V_{D1} , which are the gate voltages corresponding to charge neutrality points for the graphene that is just beneath the NW, and rest of the graphene channel, respectively; this will be discussed in detail later. The higher value of $|V_{D2}|$ indicates that the graphene region below the NW is more n-doped as compared to the rest of the graphene. Fig. 1c shows the measured open circuit voltage, V_{TE} versus I_{SD} plot for two different gate voltages ($V_{BG} = 1.5$ V and 4.2V) for a MLG device. Both the curves show a quadratic dependence of V_{TE} on I_{SD} as indicated by the dashed lines. The inset showing $V_{TE} \propto I_{SD}^2$ confirms that measured V_{TE} across the graphene channel arises from a thermoelectric response. The positive and negative amplitude of V_{TE} refer to the sign change of the thermoelectric voltage with V_{BG} (Fig. 3). Note that in contrast to our previous work [6], where we concentrated on Coulomb drag (part of the signal that flips with current reversal), here we focus mainly on the non-flipping part of the signal. As discussed in section SI 2, in this measurement, the non-flipping part dominates over the flipping part.

Anomalous thermopower oscillations in graphene-In As nanowire vertical heterostructures 5

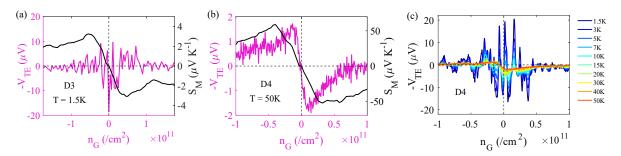


Figure 3: Thermoelectric response of NW-MLG devices: Figure (a) and (b) show comparison between V_{TE} with the Seebeck coefficients for two NW-MLG devices D3 and D4 at T=1.5K and T=50K respectively. The purple lines (left axis) indicate density dependence of V_{TE} for D3 and D4 respectively. The horizontal and vertical dashed lines indicate the zero voltage and Dirac point respectively. On the right axis of (a) and (b) we plot the theoretically estimated Seebeck coefficient S_M for D3 and D4 MLG devices (black) based on the measured electrical resistance and the Mott formula. The overall shape of S_M matches with V_{TE} at higher temperature, although doesn't match with the V_{TE} at the lower temperature. (c) V_{TE} versus n_G plot for device D4 at different temperatures ranging from 1.5K to 50K. In (a) and (c), V_{TE} show rapid oscillations with n_G at lower temperatures (T < 20K). The oscillation amplitude reduces as the temperature increases as well as at higher graphene densities.

3. Results

In this section we will first present the thermoelectric response for two BLG devices D1 and D2 followed by for two MLG devices D3 and D4 respectively. Fig. 2a shows measured V_{TE} with carrier density, n_G , for D1 BLG device at different temperatures ranging from 1.5K to 19K. V_{TE} changes sign across the Dirac point ($n_G = 0$). We see from Fig. 2a that V_{TE} decreases in magnitude with increasing temperature. The inset shows the peak magnitude of V_{TE} (for hole side) as a function of temperature, where the dotted black line is a guide to the eye. In Fig. 2b and 2c we plot the theoretically estimated Seebeck coefficient S_M (blue line) at T =1.5K of the D1 and D2 BLG devices respectively, using the Mott formula:

$$S_M = \frac{\pi^2 k_B^2 T}{3e} \frac{\partial ln\sigma}{\partial n} \frac{\partial n}{\partial \mu} \tag{1}$$

We utilize the measured resistance $(1/\sigma)$ of the BLG device (Fig. 1b lower panel) to evaluate S_M . We compare the experimentally measured V_{TE} with Mott formula of Seebeck coefficient (S_M) as shown in Fig. 2b and 2c by plotting V_{TE} versus n_G (red line) at the left axis and S_M (blue line) at the right axis. We find that the overall shape of the V_{TE} for both the NW-BLG heterostructures follows the trend of the Mott formula.

Fig. 3a, 3b present the comparison of experimental V_{TE} with the Seebeck coefficient for two NW-MLG devices D3 and D4 at T=1.5K and T=50K respectively. In the left axis we plot of V_{TE} with graphene density (purple plot), whereas in the right axis we plot the Seebeck coefficient S_M versus n_G (black line), obtained using Mott's formula for the respective temperatures. Fig. 3c shows the doping dependence of V_{TE} for device D4 at different temperatures ranging from 1.5K to 50K. In Fig. 3a and 3c (for T< 20K), the most striking observation is that the amplitude of V_{TE} changes between positive

and negative values as the carrier density is varied near the Dirac point. Notably, the periodic nature of the oscillation changes with the graphene density; more periodic near $n_G = 0$ and becoming less periodic as we move away from the neutrality point. The oscillations vanish further at higher densities, as well as when the system temperature is increased. For example, at T > 20K, the oscillations disappear and the overall shape of V_{TE} resembles that of the BLG devices (Fig. 2). The comparison in Fig. 3a at T=1.5K shows that S_M calculated from the device gate response doesn't show any oscillations, otherwise present in the density responses of V_{TE} for the MLG devices. However, the overall shapes of V_{TE} and S_M qualitatively agree at T=50K as shown in Fig. 3b. From Fig. 3a and 3c, we find the average period of oscillations in V_{TE} at T=1.5K to be $\delta n \sim$ $1.08 \times 10^{10}/cm^2$ and $0.8 \times 10^{10}/cm^2$ respectively which corresponds to energy scale of ~ 13meV and 10meV ($\delta E_F = \hbar v_F \sqrt{\pi \delta n}$) respectively. In Fig. S6 of Supplementary Info, we plot the standard deviation (SD) of V_{TE} oscillation amplitude as a function of temperature, where the dashed black line is a guide to the eye. The SD is calculated over the density range of $\pm 0.25 \times 10^{11}/cm^2$. We see that the SD approaches zero at ~ 20K, which corresponds to a thermal energy broadening $(3.5k_BT)$ of ~ 7 meV $(k_B$ being Boltzmann constant ~ $1.38 \times 10^{-23} \text{ JK}^{-1}$).

4. Discussion

In this section, we discuss the origin of thermopower in our unique nano-heating geometry and propose possible scenarios which may give rise to the observed oscillations in V_{TE} , otherwise absent in R_G . In a conventional thermoelectric measurement setup, the heater is usually placed asymmetrically on one side of the sample (few μm away) which creates a uniform temperature gradient along the channel length and gives rise to non-zero V_{TE} proportional to the Seebeck coefficient of the material [33, 34]. In contrast, here, InAs NW placed on top of the graphene serves as a local heater. Since it is placed approximately at the center of the channel, passing a current through the NW creates a temperature profile which peaks at the center of the NW and is expected to decay symmetrically on both side of the NW in graphene. To explain the origin of non-zero V_{TE} , we write it in terms of local Seebeck coefficient S(x) and temperature gradient $\frac{\partial T}{\partial x}$ as [12]:

$$V_{TE} = \int_{-L/2}^{+L/2} S(x) \frac{\partial T(x)}{\partial x} dx$$
⁽²⁾

where x is the distance from the center of the channel. If we consider the temperature profile to be Gaussian [35, 12] and centered around the NW, it creates a temperature gradient which is anti-symmetric around the center. For samples with perfect geometrical symmetry i.e. when the NW is placed exactly at the center of the graphene channel, $\frac{\partial T}{\partial x}$ takes equal and opposite magnitudes around the center. If S(x) is uniform or symmetric around the center, we would get zero average V_{TE} from Eqn. (2). It is therefore evident that asymmetry in S(x) or $\frac{\partial T}{\partial x}$ or both can lead to a finite V_{TE} . Asymmetry in temperature profile can arise either due to device geometry if the NW is not placed exactly at the center of the graphene channel, or due to asymmetric Joule heating due to different NW contact resistances at the two ends. For S(x), the symmetry can be broken either by the device skewness or due to non-identical density profiles around the center. Most real devices have intrinsic structural asymmetry as shown in Supplementary Information (section SI 1), which can lead to asymmetry in the temperature profile as well as in the Seebeck coefficient. Thus, the finite V_{TE} observed in our NW-BLG heterostructures is not surprising, and V_{TE} will change its sign only once around the Dirac point when the S(x) changes its sign with charge carrier type as seen in Fig. 2.

The non-zero V_{TE} signals in Fig. 2 and 3 are due to the intrinsic asymmetry in the device geometry; this is always present in real samples due to unavoidable uncertainties in device fabrication. Thus, the resultant thermopower *magnitude* is dependent on the inherent asymmetry of the devices, and may vary from device to device. This is reflected as a finite background signal in V_{TE} as shown in Fig. 2b and 2c, where V_{TE} can have positive or negative values at higher n_G which is device specific. However, the oscillations in V_{TE} are unaffected by the asymmetry, and are reproducible across multiple devices.

From the previous discussion, it is clear that to understand the anomalous oscillations of V_{TE} in NW-MLG heterostructures, it is necessary to look beyond Eqn. 2. Since thermopower is directly proportional to $\frac{\partial ln\sigma}{\partial n}$, the changing sign in V_{TE} with the density indicates that the effective carrier type varies as n changes. In order to understand this, we first investigate the local density modulation in graphene. Two different Dirac points in the gate response of R_G (Fig. 1b) indicate a non-uniform density profile along the channel. The 2D colormap in Fig. 4b shows how R_G evolves with the V_{BG} and V_{G} gate voltages. The black and green dashed lines highlight the variation of the main Dirac point (V_{D1}) and the weaker Dirac point (V_{D2}) with V_{BG} , and from their slopes (section SI 3 for the details) we assign V_{D2} and V_{D1} to the graphene part underneath the NW and to the rest of the graphene channel, respectively. The density mismatch can arise from the trapped charge impurities at the interface of NW- hBN-Graphene hybrid. As shown schematically in Fig. 4a, the density mismatch (p - n - p)results in misaligned Fermi energies of the two regions. This can create a cavity for the charge carriers underneath the NW resulting in a modulation of the local DOS as shown by the red line in Fig. 4c (details in the section SI 4). The polarity of S(x) depends on the type of majority charge carrier, and the effective carriers of the cavity will modulate between electron and hole as a function of the Fermi energy shift. Note that $V_{TE} \propto \frac{\partial \sigma}{\partial n}$ leading to a sign change as the Fermi energy passes through the discrete levels of the cavity as shown in Fig. 4c. However, for our geometry, a symmetric case will produce zero thermoelectric voltage (Eqn. 2). Thus, to get the observed oscillations in V_{TE} , the discrete energy levels together with asymmetry in S(x) or $\frac{\partial T}{\partial x}$ are required, the latter being always present in our devices as described in the previous section.

To validate our proposed cavity model, we compare the discrete energy levels responsible for the thermoelectric oscillations with the required cavity dimensions. In the

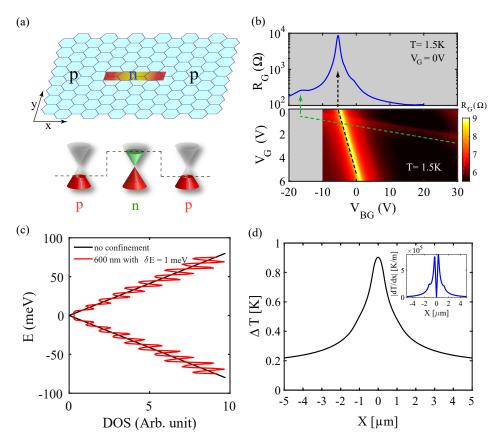


Figure 4: (a) (Upper panel) The hexagonal lattice of graphene is shown in the x-y plane. The redyellow patch indicates the graphene part underneath the NW. The region of graphene below the NW is n type, while the remaining graphene channel is p type, for the gate voltages between V_{D2} and V_{D1} (Fig. 1b). (Lower panel) The schematic of the band-diagram (p - n - p). (b) (Upper panel) Backgate response of R_G for MLG plotted in log-scale at T=1.5K. The two peaks indicated by the vertical arrows indicate non-uniform density across the graphene channel. (Lower Panel) 2D colormap of the R_G in log-scale plotted as a function of V_{BG} and V_G at T=1.5K. The green and black dashed lines highlight the trajectories of the two charge neutrality points with the gate voltages. (c) The red line is the theoretically calculated DOS versus energy for MLG for a cavity with ~ 600nm width with a Gaussian broadening of $\delta E = 1$ meV. The black line corresponds to the case with no confining potential. (d) The total temperature rise ΔT plotted with position along the X direction. (inset) The absolute value of the temperature gradient $|\frac{dT}{dX}|$ plotted with position X. Details about these thermal calculations are described in SI 7.

previous section we have estimated the energy scale responsible for the V_{TE} oscillations $\sim 7 - 10 meV$ corresponding to a cavity size of $0.6 - 0.4 \mu$ m, comparable to the length of the NW (details in section SI 5). Note that the other dimension (diameter $\sim 50 nm$) will produce discrete energy levels with orders of magnitude larger value, not seen in our experiments. From the values of V_{D1} and V_{D2} (Fig. 1b), the estimated strength of the cavity potential is $\sim 115 meV$ for MLG and $\sim 40 meV$ for the BLG device (details in section SI 5), and thus can explain the absence of oscillations in NW-BLG heterostructure. The weaker confinement is expected in BLG due to large DOS at the Dirac point. To explain the density dependence of the magnitude of V_{TE} oscillations

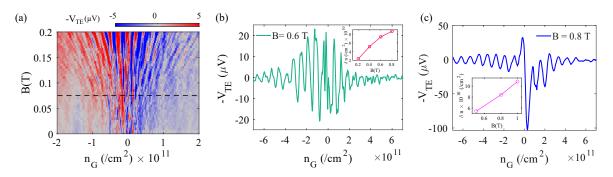


Figure 5: (a) 2D colormap of V_{TE} with graphene density n_G and magnetic field from 0 to 0.2T at T=1.5K for the MLG sample. The black horizontal dashed line indicates the onset of Landau level formation captured by the thermopower. V_{TE} versus n_G at B = 0.6T for MLG (b), and B= 0.8T for BLG (c). Insets in (b) and (c) show the oscillation period as a function of the magnetic field. The solid lines are guide to the eye.

in Fig. 3, we would like to point out the qualitative resemblance between the V_{TE} and Mott's prediction at higher density, where the magnitude of S(x) is expected to decrease with increasing density. Moreover, the effect of screening is also likely to play a role in reducing the strength of the confinement with increasing carrier concentration (SI 5 for the details). Note that the DOS calculation in Fig. 4c does not include the effect of screening, and also we have not considered relativistic effects[36, 37, 38], which are beyond the scope of this work.

We now discuss why the oscillations are seen only in the thermoelectric response but not in Mott's prediction derived from the resistance data. We calculate the temperature profile in the NW-graphene heterostructures using a 3D Fourier heat diffusion model (see section SI 7 for details). The temperature profile and its gradient across the graphene are shown in Fig. 4d for uneven Joule heating due to different NW contact resistances at its two ends. Fig. 4d shows that the temperature gradient is dominant in the region of graphene underneath the NW, and thus contributes to the measured V_{TE} significantly according to Eqn. 1. The contribution to V_{TE} from the remaining part of the graphene channel is small as the temperature gradient is close to zero. In contrast, the resistance measured across the graphene is dominated by the contribution from the rest of the graphene channel (~ $10\mu m$ X $10\mu m$) compared to the very small part of graphene (~ 50nm X 600nm) just underneath the NW.

The NW in this experiment serves a dual purpose, to produce both a highly localized temperature gradient and also a cavity potential in the part of the channel in its immediate vicinity. Fig 5a shows a 2D colormap of thermopower with n_G and magnetic field for the MLG device, where one can observe LLs for $B \ge 0.1T$. Fig. 5b and 5c show the measured periodic oscillations in V_{TE} for MLG and BLG, respectively, at B ~ 0.6T and at ~ 0.8T. As expected, the period of the oscillations increases with increasing magnetic field as shown in the insets of Fig. 5 (SI 6 for details). The period in density $\delta n \sim 0.85 \times 10^{11}/cm^2$ at B =0.8T for BLG corresponds to $\delta E \sim 3.3meV$ ($\hbar^2 \pi \delta n/2m^*$), which qualitatively matches the Landau level spacing of ~ 3.2meV ($\delta E_{LL} \sim \hbar eB/m^*$)

at B = 0.8T, where $m^* \sim 0.03m_e$. For BLG, no oscillations in V_{TE} at B=0 but periodic oscillations at finite magnetic field (section SI 6) further confirm that at zero field there is no modulation in the DOS due to the weaker confinement potential.

Although the concept of cavity formation leading to thermopower oscillation is highly likely, there are other possibilities which may contribute to the oscillations. The fact that the oscillations are always observed in the vicinity of the Dirac point and decay quickly as the density is tuned away from $n_G = 0$, indicates that the electronhole puddles can play a role in anomalous oscillations in V_{TE} in MLG. Since the device asymmetry is always intrinsic, the overall effective carrier type changes from electron to hole as the Fermi energy is tuned across the Dirac point. The apparent aperiodicity in V_{TE} , especially away from the Dirac point suggests that the charge inhomogeneities near the Dirac point may contribute to the oscillations.

Finally we note that, in this work we compare the measured thermoelectric voltage with the theoretically estimated Seebeck coefficient S_M to emphasize that the oscillations are observed only in V_{TE} , not in R_G . This observation excludes the possibility of UCF [15] as a possible source of oscillations.

5. Conclusions

The thermoelectric response of the NW-MLG devices using InAs NW as a local heater shows anomalous oscillations at low temperatures, which is absent for the NW-BLG devices. The oscillations are only observed in thermopower, not in the electrical resistance. We ascribe them to the changing effective carrier type in the graphene channel with density. By analysing the density profile in graphene, we show that a cavity formed below the nanowire leads to the modification in local density of states which may reflect as alternating sign in the thermoelectric voltage. We also argue that the ubiquitous charge inhomogeneities in graphene near the Dirac point combined with the intrinsic asymmetry may also contribute to the oscillations. Thus, our work will pave the way for designing thermoelectric devices using dimensionally mismatched systems, with the potential to enhance the thermopower in two-dimensional materials. We envisage that decoration of graphene by nanostructures resulting in locally enhanced DOS can lead to a high Seebeck coefficient.

6. Acknowledgement

AD thanks the Department of Science and Technology (DST), India for financial support (DSTO-2051) and acknowledges the Swarnajayanti Fellowship of the DST/SJF/PSA-03/2018-19. AKS thanks DST for financial support under the Nanomission Project and also thanks DST for the support under the Year of Science Professorship. KW and TT acknowledge support from the Elemental Strategy Initiative conducted by the MEXT, Japan and the CREST (JPMJCR15F3), JST. We acknowledge Michael Fourmansky for his professional assistance in NWs MBE growth. HS acknowledges partial funding

Anomalous thermopower oscillations in graphene-InAs nanowire vertical heterostructures11

by Israeli Science Foundation (Grants No. 532/12 and No. 3-6799), BSF Grant No. 2014098 and IMOS-Tashtiot Grant No. 0321-4801. HS is an incumbent of the Henry and Gertrude F. Rothschild Research Fellow Chair.

References

- Jariwala D, Marks TJ, Hersam MC. Mixed-dimensional van der Waals heterostructures. Nature materials. 2017;16(2):170–181.
- [2] Henning A, Sangwan VK, Bergeron H, Balla I, Sun Z, Hersam MC, et al. Charge separation at mixed-dimensional single and multilayer MoS2/silicon nanowire heterojunctions. ACS applied materials & interfaces. 2018;10(19):16760–16767.
- [3] Jeon PJ, Lee YT, Lim JY, Kim JS, Hwang DK, Im S. Black phosphorus-zinc oxide nanomaterial heterojunction for p-n diode and junction field-effect transistor. Nano letters. 2016;16(2):1293– 1298.
- [4] Yang Z, Liu X, Zou X, Wang J, Ma C, Jiang C, et al. Performance Limits of the Self-Aligned Nanowire Top-Gated MoS2 Transistors. Advanced Functional Materials. 2017;27(19):1602250.
- [5] Lee YT, Jeon PJ, Han JH, Ahn J, Lee HS, Lim JY, et al. Mixed-Dimensional 1D ZnO-2D WSe2 van der Waals Heterojunction Device for Photosensors. Advanced Functional Materials. 2017;27(47):1703822.
- [6] Mitra R, Sahu MR, Watanabe K, Taniguchi T, Shtrikman H, Sood A, et al. Anomalous Coulomb Drag between InAs Nanowire and Graphene Heterostructures. Physical Review Letters. 2020;124(11):116803.
- [7] Cheng A, Taniguchi T, Watanabe K, Kim P, Pillet JD. Guiding Dirac fermions in graphene with a carbon nanotube. Physical Review Letters. 2019;123(21):216804.
- [8] Hicks L, Dresselhaus MS. Thermoelectric figure of merit of a one-dimensional conductor. Physical review B. 1993;47(24):16631.
- [9] Hicks L, Dresselhaus MS. Effect of quantum-well structures on the thermoelectric figure of merit. Physical Review B. 1993;47(19):12727.
- [10] Dresselhaus M, Dresselhaus G, Sun X, Zhang Z, Cronin S, Koga T. Low-dimensional thermoelectric materials. Physics of the Solid State. 1999;41(5):679–682.
- [11] Hippalgaonkar K, Huang B, Chen R, Sawyer K, Ercius P, Majumdar A. Fabrication of microdevices with integrated nanowires for investigating low-dimensional phonon transport. Nano letters. 2010;10(11):4341-4348.
- [12] Harzheim A, Evangeli C, Kolosov OV, Gehring P. Direct mapping of local Seebeck coefficient in 2D material nanostructures via scanning thermal gate microscopy. 2D Materials. 2020 aug;7(4):041004. Available from: https://doi.org/10.10882F2053-15832Faba333.
- [13] Cho S, Kang SD, Kim W, Lee ES, Woo SJ, Kong KJ, et al. Thermoelectric imaging of structural disorder in epitaxial graphene. Nature materials. 2013;12(10):913–918.
- [14] Vera-Marun I, Van Den Berg J, Dejene F, Van Wees B. Direct electronic measurement of Peltier cooling and heating in graphene. Nature communications. 2016;7(1):1–6.
- [15] Zuev YM, Chang W, Kim P. Thermoelectric and magnetothermoelectric transport measurements of graphene. Physical review letters. 2009;102(9):096807.
- [16] Park J, He G, Feenstra RM, Li AP. Atomic-scale mapping of thermoelectric power on graphene: Role of defects and boundaries. Nano letters. 2013;13(7):3269–3273.
- [17] Hippalgaonkar K, Wang Y, Ye Y, Zhu H, Wang Y, Moore J, et al. Record high thermoelectric powerfactor in single and few-layer MoS2. arXiv preprint arXiv:150506779. 2015.
- [18] Xia J, Chen F, Li J, Tao N. Measurement of the quantum capacitance of graphene. Nature nanotechnology. 2009;4(8):505.

Anomalous thermopower oscillations in graphene-InAs nanowire vertical heterostructures 12

- [19] Ilani S, Donev LA, Kindermann M, McEuen PL. Measurement of the quantum capacitance of interacting electrons in carbon nanotubes. Nature Physics. 2006;2(10):687–691.
- [20] Zhang Y, Brar VW, Girit C, Zettl A, Crommie MF. Origin of spatial charge inhomogeneity in graphene. Nature Physics. 2009;5(10):722–726.
- [21] Stolyarova E, Rim KT, Ryu S, Maultzsch J, Kim P, Brus LE, et al. High-resolution scanning tunneling microscopy imaging of mesoscopic graphene sheets on an insulating surface. Proceedings of the National Academy of Sciences. 2007;104(22):9209–9212.
- [22] Cui L, Jeong W, Hur S, Matt M, Klöckner JC, Pauly F, et al. Quantized thermal transport in single-atom junctions. Science. 2017;355(6330):1192–1195.
- [23] Cui L, Hur S, Akbar ZA, Klöckner JC, Jeong W, Pauly F, et al. Thermal conductance of singlemolecule junctions. Nature. 2019;572(7771):628–633.
- [24] Martin J, Akerman N, Ulbricht G, Lohmann T, Smet Jv, Von Klitzing K, et al. Observation of electron-hole puddles in graphene using a scanning single-electron transistor. Nature physics. 2008;4(2):144–148.
- [25] Rutter GM, Crain J, Guisinger N, Li T, First P, Stroscio J. Scattering and interference in epitaxial graphene. Science. 2007;317(5835):219–222.
- [26] Li Z, Henriksen EA, Jiang Z, Hao Z, Martin MC, Kim P, et al. Dirac charge dynamics in graphene by infrared spectroscopy. Nature Physics. 2008;4(7):532–535.
- [27] Giannazzo F, Sonde S, Raineri V, Rimini E. Screening length and quantum capacitance in graphene by scanning probe microscopy. Nano letters. 2009;9(1):23–29.
- [28] Berweger S, Weber JC, John J, Velazquez JM, Pieterick A, Sanford NA, et al. Microwave near-field imaging of two-dimensional semiconductors. Nano letters. 2015;15(2):1122–1127.
- [29] Wei P, Bao W, Pu Y, Lau CN, Shi J. Anomalous thermoelectric transport of Dirac particles in graphene. Physical review letters. 2009;102(16):166808.
- [30] Wu PM, Gooth J, Zianni X, Svensson SF, Gluschke JG, Dick KA, et al. Large thermoelectric power factor enhancement observed in InAs nanowires. Nano letters. 2013;13(9):4080–4086.
- [31] Pizzocchero F, Gammelgaard L, Jessen BS, Caridad JM, Wang L, Hone J, et al. The hot pickup technique for batch assembly of van der Waals heterostructures. Nature communications. 2016;7(1):1–10.
- [32] Wang L, Meric I, Huang P, Gao Q, Gao Y, Tran H, et al. One-dimensional electrical contact to a two-dimensional material. Science. 2013;342(6158):614–617.
- [33] Ashcroft N, Mermin N. Solid State Physics (Saunders College, Philadelphia). Google Scholar. 1976:404.
- [34] Goldsmid HJ, et al. Introduction to thermoelectricity. vol. 121. Springer; 2010.
- [35] Hu X, Gong X, Zhang M, Lu H, Xue Z, Mei Y, et al. Enhanced Peltier Effect in Wrinkled Graphene Constriction by Nano-Bubble Engineering. Small. 2020;16(14):1907170.
- [36] Zhao Y, Wyrick J, Natterer FD, Rodriguez-Nieva JF, Lewandowski C, Watanabe K, et al. Creating and probing electron whispering-gallery modes in graphene. Science. 2015;348(6235):672–675.
- [37] Lee J, Wong D, Velasco Jr J, Rodriguez-Nieva JF, Kahn S, Tsai HZ, et al. Imaging electrostatically confined Dirac fermions in graphene quantum dots. Nature Physics. 2016;12(11):1032–1036.
- [38] Matulis A, Peeters F. Quasibound states of quantum dots in single and bilayer graphene. Physical Review B. 2008;77(11):115423.

Supplementary Information: Anomalous thermopower oscillations in graphene-InAs nanowire vertical heterostructures

Richa Mitra¹, Manas Ranjan Sahu¹, Aditya Sood², Takashi Taniguchi³, Kenji Watanabe³, Hadas Shtrikman⁴, Subroto Mukerjee¹, A.K. Sood¹, Anindya Das¹ *

¹Department of Physics, Indian Institute of Science, Bangalore-560012, India.

²Stanford Institute for Materials and Energy Sciences, SLAC National Accelerator Laboratory, Menlo Park, California, 94025, USA.

³National Institute for Materials Science, Namiki 1-1, Ibaraki 305-0044, Japan.

⁴Department of Physics, Weizmann Institute of Technology, Israel.

*anindya@iisc.ac.in

SI 1. Device fabrication

All the devices used in the thermopower measurements are heterostructures of InAs nanowire (NW) and graphene encapsulated by hexagonal Boron Nitride (hBN). In our experiment, we pass current through the NW, which acts as a local heating source, and measure the thermoelectric voltage across the graphene. Graphene flakes are exfoliated from bulk graphite crystals using scotch tape and then transferred onto 285 nm thick SiO₂, thermally grown over p++ doped Si substrate. hBN flakes are separately obtained following a similar exfoliation method. Suitable MLG/BLG flakes are identified under the microscope and encapsulated by hBN flakes using the hot pickup technique [1, 2]. After assembling the graphene stack, we separately pick up the suitable NW from a separate substrate and carefully align it at the center of the hBN/graphene/hBN stack. The thickness of the top hBN, which separates the NW from the graphene, is chosen to be around $\sim 10 - 12$ nm (Fig. S1 c). After assembling the heterostructures, separate probes for graphene and NWs are fabricated (Fig. S1 b). For graphene, we adhere to the conventional one-dimensional edge contact fabrication [3] technique, whereas, for the NWs, ohmic contacts are fabricated following the chemical etching technique [4] where saturated $(NH_4)_2S$ solution is used for removing the native oxide. After contact fabrication, all the samples are characterized at room temperature as well as at T=1.5K. The backgate response of the MLG and BLG has been presented in Fig. 1b of the main manuscript. The typical resistance of the NWs was around $\sim 25 - 50 \ k\Omega$.

As shown in the optical images (Fig. S1 a, b), most of the devices possess intrinsic structural asymmetry where the NW is not placed exactly at the center of the graphene channel, which leads to a non-zero thermopower signal. Non-zero V_{TE} can also result from the unequal contact resistances of the NW probes leading to an asymmetric temperature profile across the center. Fig. S1 c shows the thickness of a top hBN flake to be ~ 10 nm, which separates the graphene channel from the NW.

SI 2. Data extraction

The measurement schematic of our thermopower technique is shown in Fig. S2, where we pass a DC current through the NW (I_{SD}) and measure thermopower voltage V_{TE} across the graphene probe. We apply backgate voltage V_{BG} in the Si/SiO₂ substrate to tune the graphene density n_G and V_G to adjust the NW density. Due to the proximity of the NW to the graphene channel, which is separated by a thin hBN (~ 10nm) layer, passing a current through the NW may generate drag voltage due to Coulomb interaction between the two systems. Here we describe the method used for accounting for the drag effect and extracting the thermopower signal from the raw data. Fig. S3 a and S4 a show 2D colormap of V_{TE} with heater current I_{SD} and n_G for MLG and BLG devices, respectively. The raw signal contains both drag (V_D) as well as the thermopower signal (V_{TE}) . The drag signal (flipping part) flips sign as the direction of the current I_{SD} is reversed whereas the V_{TE} (the thermopower) signal remains unchanged upon reversing the current direction. To separate out the drag

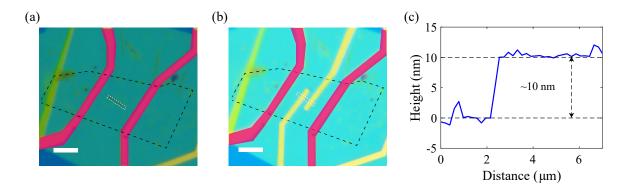


Figure S1 (a), (b) are the optical images of a InAs NW-hBN-Graphene-hBN device. The black and white dashed lines indicate the encapsulated monolayer graphene and the InAs nanowire placed on top of it, respectively. One observes the InAs nanowire been placed asymmetrically between two graphene probes (pink area). The yellow areas in (b) indicate separate metal probes of the InAs nanowire. The scale bar is 5 μ m for both (a) and (b). (c) The AFM data showing thickness of the top hBN to be ~ 10 μ m. This hBN electrically separates the graphene channel from the NW.

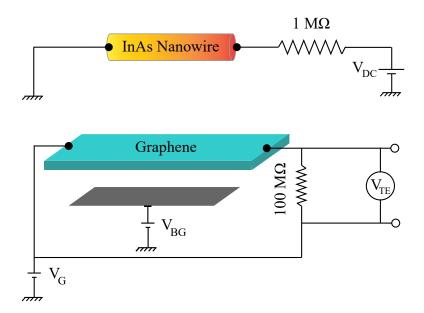


Figure S2 Thermopower measurement schematic. V_{TE} measured in graphene while passing a constant DC current through the NW.

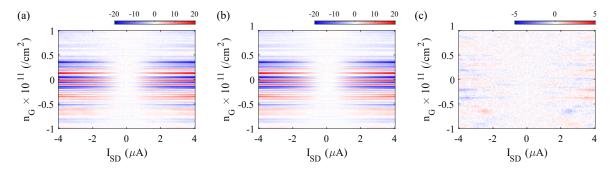


Figure S3 2D colormap of V_{TE} measured in MLG plotted with heating current I_{SD} and with graphene density n_G at T=1.5K and B=0T. Figure (a), (b) and (c) are the raw signal (V_{raw}) and the extracted non-flipping (V_{TE}) and the flipping signals (V_D) respectively.

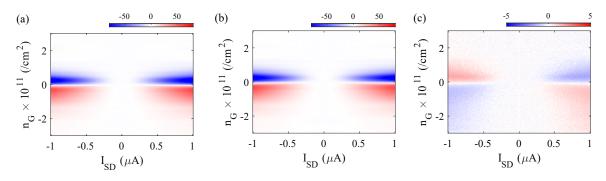


Figure S4 2D colormap of V_{TE} measured in BLG plotted with heating current I_{SD} and with graphene density n_G at T=1.5K and B=0T. Figure (a), (b) and (c) are the raw signal (V_{raw}) and the extracted non-flipping (V_{TE}) and the flipping signals (V_D) respectively.

effect from the raw signal, we adhere to the following method: $V_D \rightarrow -V_D$, as $I_{SD} \rightarrow -I_{SD}$ for drag signals, but the non-flipping part which originates from heating effect ($\propto I_{SD}^2 R$) doesn't change its sign. So, the raw signals can be written as:

$$V^{+}_{raw} = V_D + V_{TE}$$

$$V^{-}_{raw} = -V_D + V_{TE}$$
(1)

where V^+_{raw} and V^-_{raw} are the raw signals when I_{SD} is positive and negative, respectively. Combining these two equations, we get:

$$V_{TE} = \frac{1}{2} (V^+{}_{raw} + V^-{}_{raw}) \tag{2}$$

We extracted the V_{TE} and V_D according to the above equations, which are plotted in Figure S3 b and c for MLG and in S4 b and c for BLG device, respectively. We observe from these plots that $V_{TE} \gg V_D$ for both the cases.

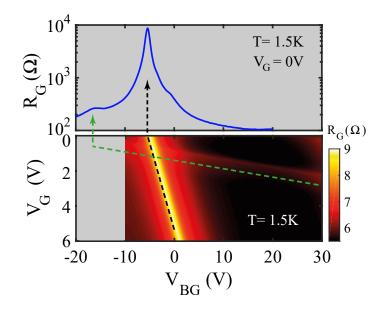


Figure S5 (Upper panel) Graphene 2-probe resistance R_G versus V_{BG} plot at T=1.5K. (Lower panel) 2D colormap of R_G versus V_{BG} and the graphene gate V_G measured at T=1.5K. The black and the green dashed lines indicate the trajectories of the main Dirac point (V_{D1}) and the additional Dirac point (V_{D2}) with gate voltages, respectively. Arrows in the upper panel indicate that the two Dirac points in the 2D plot originate from V_{D1} and V_{D2} .

SI 3. Uneven density profile along the graphene channel

In this section, we discuss how the density profile (p-n-p) in the graphene channel has been interpreted from the experimental data. Figure S5 (upper panel) shows graphene resistance R_G plotted with the V_{BG} where an additional resistive peak appears at V_{D2} along with the main Dirac point at V_{D1} . The second resistance peak appears at a negative gate voltage i.e., $|V_{D2}| > |V_{D1}|$ which indicates that (a) the density profile across the full graphene channel is not uniform, and (b) the region having different density compared to the rest of the graphene channel is more n-type or electron type. This indicates that along the graphene channel length, the effective density profile is p - n - p type.

To analyze the density profile further, we measure R_G as a function of both V_{BG} and V_G at T=1.5K as shown in the lower panel of Fig. S5. Here V_G is the graphene gate voltage, which is applied directly to the graphene to tune the NW density as shown in the measurement schematic in Fig. S2. The black and green dashed lines indicate the trajectories of V_{D1} and V_{D2} with changing gate voltages. The arrows in the upper panel indicate that both V_{D1} and V_{D2} of the two plots are essentially the same. We observe that, as V_G increases, the Dirac point shifts towards positive values of V_{BG} . This is obvious as the graphene channel becomes less electron-doped as we apply + ve V_G and less - ve V_{BG} gate is required to make the graphene charge neutral. From the 2D plot, we find the slope of V_{D1} to be close to unity, which supports the above argument. In our measurement setup,

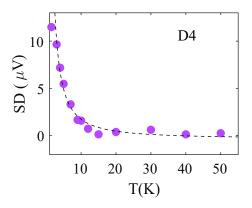


Figure S6 Standard deviation (SD) of oscillations plotted versus temperature (purple solid circles). *SD* has been calculated in the density range of $\pm 0.25 \times 10^{11}/cm^{-2}$ from Fig. 3c of the main manuscript. The black dashed line is a guide to eye.

the graphene and NW are separated by a thin hBN (~ 10nm) flake. So, applying V_G in graphene induces charge carriers in the NW as well as in the graphene region below the NW. Since the electric field generated from the backgate is well-screened by the graphene, NW density is mostly controlled by V_G , whereas V_{BG} controls the density of the rest of the graphene channel. As a result, the graphene portion below NW acquires different doping when graphene gate is applied, i.e., when $V_G \neq 0$. Very interestingly, for all our samples, we observe dual Dirac point even at $V_G = 0$ V (upper panel of Fig. S5), which indicates that the density inhomogeneity of the graphene channel is intrinsically present; most likely due to trap charge impurities at the NW-hBN-Graphene structure. As we observe from the 2D plot, the V_{D2} changes faster than V_{D1} when V_G is applied, which is evident from the above discussion. We also calculate the slope of V_{D2} trajectory and find that the capacitance of the graphene gate $C_G \sim 20 \times C_{BG}$, where C_{BG} is the capacitance of the backgate. From this equation, we estimate the thickness of the top hBN to be $\sim 12 - 15$ nm, which is very close to the thickness of the hBN used (~ 10nm). While calculating the C_G , we have not included the systems' quantum capacitances.

SI 4. Modulation in density of states

According to the algorithm mentioned below, we generated the density of states (DOS) of graphene using a MATLAB code. Let the dimensions of the graphene sheet be (L_X, L_Y) , with Area $(A)=L_X \times L_Y$. Then the spacing of wave vectors (K_X, K_Y) will be, $\Delta K_X = 2\pi/L_X$ and $\Delta K_Y = 2\pi/L_Y$. At first, we construct a 2D K_X - K_Y array, as shown in fig. S7 a. Then, we calculate the energy corresponding to each K-point using the low energy dispersion relation, $E = \hbar v_F \sqrt{(K_X^2 + K_Y^2)}$. Finally, we count the number of K -points $(\delta N(\epsilon))$ for a given energy ϵ , which satisfies $\epsilon - \delta \epsilon < E(K_X, K_Y) < \epsilon + \delta \epsilon$. Once we have all the above information, the DOS(ϵ) is given by,

$$DOS(\epsilon) = 4 \times \frac{\delta N(\epsilon)}{2\delta \epsilon} \times \frac{1}{A}$$

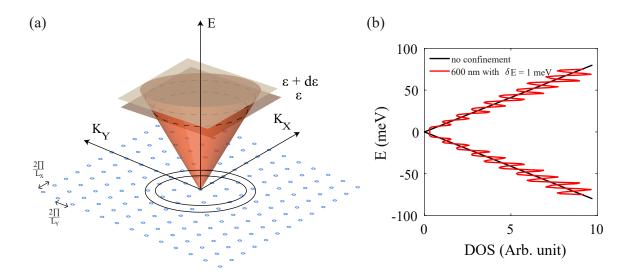


Figure S7 (a) Reciprocal lattice distribution for samples having dimension $L_X \times L_Y$ having equal length and for unequal length. (b) Fermi energy plot with density of states (DOS) for graphene having dimension of $10\mu \text{m} \times 10\mu \text{m}$ (black) and $10\mu \text{m} \times 0.6 \mu \text{m}$ (red) with a Gaussian broadening of $\delta E = 1 \text{ meV}$.

Here, 4 factor in the numerator is the system's degeneracy, i.e., the number of states that can occupy the same K-point.

To incorporate the effect of disorder, we introduced a Gaussian broadening into the system. For this, we average the original DOS with a weightage of Gaussian function, and the resulting effective DOS (DOS') is given by,

$$DOS'(\epsilon) = \int_{-\delta E}^{\delta E} DOS(\epsilon + x)G(x)dx$$

Where, $G(x) = \frac{1}{\sqrt{2\pi\delta E_F^2}} e^{\frac{-x^2}{2\delta E_F^2}}$ is the Gaussian function and δE_F is the Fermi energy broadening of the system. Using the above algorithm, we estimate the DOS for two-dimensional graphene with a dimension of $10\mu m \times 10\mu m$ (black curve) as well as of $10 \ \mu m \times 0.6 \ \mu m$ (red curve) as shows in Fig. S7 b.

SI 5. Cavity strength and Length Scale calculations

Cavity strength: In this section, we discuss how the cavity strength has been calculated. As discussed before, a cavity is created in graphene in the region beneath the NW in the presence of NW's one-dimensional electrostatic potential. From the backgate response of graphene, we observe a second Dirac point V_{D2} appears at negative gate voltages, suggesting more electron-like or 'n' type region in the graphene sheet and implicates

uneven density profile across the graphene channel. As shown in Fig. 4a (lower panel) of the main manuscript, the density mismatch across the graphene sheet leads to misaligned Fermi energies, which creates an energy barrier or confinement for the charge carriers. We have estimated the density mismatch from the backgate voltage difference between V_{D1} and V_{D2} . To estimate the strength of the energy barrier, we use the linear band dispersion of monolayer graphene $E_F = \hbar v_F \sqrt{\pi n_G}$ and parabolic band dispersion $E_F = \frac{\pi \hbar^2 n_G}{2m^*}$ for bilayer graphene with $m^* \sim 0.03m_0$, where v_F , m^* and m_0 are the Fermi velocity and the effective mass of carriers in bilayer graphene and the mass of electron, respectively. For our devices, $\Delta V_{BG} = V_{D2} - V_{D1} \sim 16V$ and $\sim 23V$ for MLG and BLG respectively which is equivalent to energy scale of $\sim 115meV$ and $\sim 45meV$. The R_G versus E_F plots for MLG and BLG devices are shown in Fig. S8 a and b, respectively. The smaller energy barrier in bilayer graphene indicates shallow confinement of charge carriers, which explains the absence of oscillations in BLG devices.

Length Scale calculations: We also estimate the approximate length scale corresponding to the oscillations observed in MLG devices. As shown in Fig. 3a and 3c of main manuscript, the average oscillation period in MLG are $\delta n \sim 1.08 \times 10^{10}/cm^2$ and $0.8 \times 10^{10}/cm^2$ respectively which is equivalent to the energy $\delta E \sim 13meV$ and $\sim 10meV$ respectively. The cavity length for constructive interference is $L = p\lambda$, where λ is the electron wavelength, and p is any positive integer. This equation reduces to $L = \frac{2\pi\hbar v_F}{\delta E}$ for MLG. For our systems $\delta E \sim 7 - 13meV$ which is equivalent to length $0.6 - 0.35\mu$ m. This length scale is very similar to the NW channel length in our devices, which shows that the confinement barrier is created underneath the NW contacts. To estimate the fluctuation period corresponding to the other dimension (diameter) of the NW, we use $L \sim 50nm$ which is equivalent to $\delta n \sim 1.25 - 5 \times 10^{11} cm^{-2}$. Since significant oscillations are not observed among $\pm 0.5 \times 10^{11} cm^{-2}$, we can infer that the confinement or the cavity is formed only along the length of the NW. The cavity is weaker along the diameter, which could be due to shallow confinement arising from the NW's cylindrical shape.

Effect of screening: Now, we will discuss the density dependence of V_{TE} oscillations. It can be seen in Fig. 3a and 3c of the manuscript that the oscillations in V_{TE} get weaker as n_G is tuned away from the Dirac point. The Fermi energy increases along with n_G . However, the DOS oscillations as a function of energy do not show any decrease with increasing E in Fig. 4c of the manuscript. The most probable cause for the decrease in oscillations with increasing n_G is the effect of screening. Note that for MLG, the DOS, in the absence of confinement, goes as |E| and thus increases with increasing n_G . The cavity potential itself is affected by the ability of the layer to screen since it is induced by the NW on top. Thus, as n_G increases, so does the amount of screening effectively, resulting in a decrease in the strength of the confining potential. Since the oscillations are caused due to the confinement, they too decrease in magnitude with increasing n_G . The DOS shown in Fig. 4c of the manuscript, while correctly showing that oscillations in the DOS exist, does not take this effect of screening at large n_G into account since it is difficult to model it accurately. The importance of screening can also be seen from the fact that the estimated cavity potential for MLG is larger than for BLG. In the latter system, the DOS is more or less constant and does not go to zero at the charge neutrality point. Screening in BLG is thus more effective

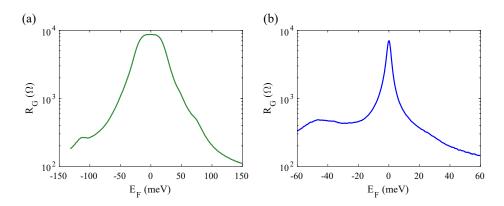


Figure S8 (a) and (b) are plots of R_G vs. Fermi energy E_F for MLG and BLG, respectively. The second peak of R_G appears around 115 meV and 45 meV for MLG and BLG devices.

and the potential induced by the NW much weaker.

SI 6: Thermopower in BLG in presence of magnetic field

Fig. S9 a and b show plots of V_{TE} with n_G for different magnetic fields for the MLG and BLG devices, respectively. For MLG, the oscillation period increases as B is increased, which indicates the formation of Landau levels (LL) for B> 0.1T (see Fig. 5a of the main manuscript). At B=0T, V_{TE} changes sign according to the Seebeck coefficient polarity for the BLG device. However, in the presence of the magnetic field, the thermopower voltage starts to show oscillations, indicating the formation of Landau levels in the BLG. The energy equivalent to the oscillation frequency for BLG (inset Fig. 5c of the main manuscript) matches with the expected energy gap between LLs for BLG.

SI 7: Theoretical Model

To calculate the temperature profile in the graphene layer, we solve the 3D Fourier heat diffusion equation for the multilayer stack. The stack comprises, from top to bottom (thicknesses in parentheses): hBN (10 nm), graphene (0.4 nm), hBN (25 nm), SiO₂ (300 nm), Si (500 μ m). The stack is heated by an InAs NW placed on top of the top hBN layer, represented by a planar rectangular heat source. We consider three components of this heat source: (1) the NW channel (length = 0.6 μ m, width = 50 nm), (2) NW underneath the left contact (length = 0.93 μ m, width = 50 nm), (3) NW underneath the right contact (length = 0.93 μ m, width = 50 nm). The power dissipated in part (1) is P, and in parts (2) and (3) is f_LP and f_RP , respectively. We calculate the 3D temperature profile induced by each of these heat sources separately, and obtain the total temperature profile by superposition.

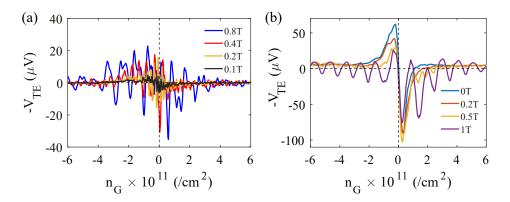


Figure S9 V_{TE} plot with n_G for different magnetic fields at T=1.5K for MLG (a) and BLG (b) respectively.

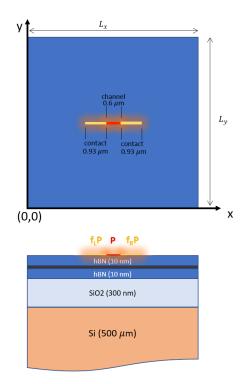


Figure S10 (Upper panel) Top view of the theoretical set up where the NW is represented by the smaller rectangular region on top of the graphene channel (blue region) having dimension of $L_x \times L_y$. Here the yellow and the red portions represents the metal contacts and the channel of the NW respectively. (Lower panel) The side view of the setup showing the multilayer stacking.

Numerical approach

To calculate the temperature profile created a rectangular surface heat source, we use a closed form analytical solution [5] for the steady-state 3D Fourier diffusion equation: $\nabla^2 \theta = 0$, where $\theta(x, y, z) = T(x, y, z) - T_0$ is the temperature rise above ambient. The simulation cell has dimensions L_x and L_y (both taken to be 60 μ m). The heat source, whose center is at coordinates (X_0, Y_0) , has dimensions H_x and H_y along the x and y directions, respectively. We assume adiabatic boundary conditions on the top and lateral surfaces, and isothermal conditions ($\theta = 0$) at the bottom of the Si substrate. Key equations are reproduced below from Bagnall et al. [5] to aid the reader.

The general solution for the temperature rise at a point located at a depth z_j within the *j*th layer is given by:

$$\theta_{j}(x, y, z_{j}) = A_{0j} + B_{0j}z + \sum_{m=1}^{\infty} \cos(\lambda_{m}x) \left[A_{1j}\cosh(\lambda_{m}z_{j}) + B_{1j}\sinh(\lambda_{m}z_{j})\right] \\ + \sum_{n=1}^{\infty} \cos(\delta_{n}y) \left[A_{2j}\cosh(\delta_{n}z_{j}) + B_{2j}\sinh(\delta_{n}z_{j})\right] + \\ \sum_{m=1}^{\infty} \sum_{n=1}^{\infty} \cos(\lambda_{m}x)\cos(\delta_{n}y) \left[A_{3j}\cosh(\beta_{mn}z_{j}) + B_{3j}\sinh(\beta_{mn}z_{j})\right]$$

where $\lambda_m = m\pi/L_x$, $\delta_n = n\pi/L_y$ and $\beta_{mn} = \sqrt{\lambda_m^2 + {\delta_n}^2}$ are the eigenvalues, collectively denoted γ_n . The Fourier coefficients A_{ij} and B_{ij} in the *j*th layer are related by the spreading function, $\phi_j(\gamma_n) = -B_{ij}/A_{ij}$. Assuming a high heat transfer coefficient at the bottommost surface of the Si substrate (i.e. thermally grounded to ambient), and given that its large thickness, $\phi_N(\gamma_n) = 1$. The spreading functions in layers 1 to N-1 are found recursively:

$$\phi_j(\gamma_n) = \frac{\frac{\kappa_j}{\kappa_{j+1}} tanh(\gamma_n t_j) + \phi_{j+1}(\gamma_n) + \frac{\kappa_j}{G_j} \gamma_n \phi_{j+1}(\gamma_n) tanh(\gamma_n t_j)}{\frac{\kappa_j}{\kappa_{j+1}} + \phi_{j+1}(\gamma_n) tanh(\gamma_n t_j) + \frac{\kappa_j}{G_j} \gamma_n \phi_{j+1}(\gamma_n)}$$

where κ_j and t_j denote the thermal conductivity and thickness of layer j respectively, and G_j the thermal boundary conductance between layers j and j+1. The zeroth order Fourier coefficients A_{0j} and B_{0j} are given by:

$$A_{0j} = \frac{Q}{L_x L_y} \left(\sum_{l=j}^{N} \left[\frac{t_l}{\kappa_l} + \frac{1}{G_l} \right] \right)$$
$$B_{0j} = -\frac{Q}{L_x L_y \kappa_j}$$

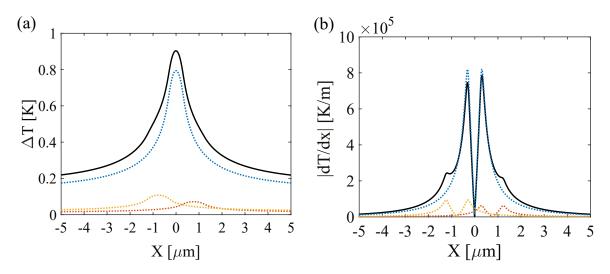


Figure S11 (a) The temperature rise ΔT plotted with position along the X direction. (b) The absolute value of the temperature gradient $\left| \frac{dT}{dX} \right|$ plotted with position X. For both the plots, the dashed blue, yellow, and red curves correspond to the individual heating contributions from the NW channel, the left, and right contacts, respectively. The total temperature rise and the temperature gradient are shown by the black curves respectively.

Higher order Fourier coefficients in the top layer, $A_{i1}(i = 1, 2, 3)$ are given by:

$$A_{11} = \frac{4P \cos(\lambda_m X_0) \sin(\frac{1}{2}\lambda_m H_x)}{L_x L_y H_x \kappa_1 \lambda_m^2 \phi_1(\lambda_m)}$$
$$A_{21} = \frac{4P \cos(\delta_n Y_0) \sin(\frac{1}{2}\delta_n H_y)}{L_x L_y H_y \kappa_1 \delta_n^2 \phi_1(\delta_n)}$$
$$A_{31} = \frac{16P \cos(\lambda_m X_0) \sin(\frac{1}{2}\lambda_m H_x) \cos(\delta_n Y_0) \sin(\frac{1}{2}\delta_n H_y)}{L_x L_y H_x H_y \kappa_1 \lambda_m \delta_n \beta_{mn} \phi_1(\beta_{mn})}$$

Finally, Fourier coefficients in the lower layers are defined using the recursive relation:

$$A_{ij} = A_{i,j-1} \left[\frac{\cosh(\gamma_n t_{j-1}) - \phi_{j-1}(\gamma_n) \sinh(\gamma_n t_{j-1})}{\frac{\kappa_j}{G_j} \gamma_n \phi_j(\gamma_n) + 1} \right]$$

Since some of the layers in the stack are thermally anisotropic, we account for this by defining effective isotropic values for thickness and thermal conductivity: $\kappa_{j,iso} = \sqrt{(\kappa_{j,z}\kappa_{j,xy})}$ and $t_{j,iso} = t_j/\sqrt{\kappa_{j,z}/\kappa_{j,xy}}$, where 'xy' and 'z' refer to in-plane and cross-plane components.

Thermal parameters

The inputs to this model are the thickness (t) and thermal conductivity (κ) of each layer, and the thermal boundary conductance (G) between adjacent layers. Below we describe how these properties are estimated at a base temperature of ~ 5 to 10 K, based on prior experiments and calculations. We emphasize that our goal is not to calculate the precise temperature profile, but to obtain a first order estimate of the temperature gradients.

For κ_{xy} of hBN, we refer to previous low T measurements [6] of heat transport in crystallites of diameter ~ 1µm and thickness ~ 50-100 nm. At low T, it is reasonable to assume that phonon boundary scattering is specular, and therefore κ_{xy} is thickness-independent. Since the lateral dimensions will limit phonon transport, we must scale κ_{xy} by 10× to account for the distance between the metal contacts in our devices, viz. 10 µm; this gives $\kappa_{xy} \approx 30$ Wm⁻¹K⁻¹ at 5 K. We assume that at low T, intrinsic cross-plane transport in hBN is not limiting, but rather governed by interfaces (see below), and take a sufficiently large κ_z so that it does not matter.

For graphene, we first estimate the electronic contribution to κ_{xy} using the Wiedemann Franz law, which was previously shown [7] to be valid below 20 K. Based on the measured electrical conductivity, we estimate this to be at most $\approx 2 \text{ Wm}^{-1}\text{K}^{-1}$ at highest doping. To estimate the lattice contribution, we refer to first principles calculations [8] for polycrystalline graphene of grain size $\sim 10 \ \mu\text{m}$, which gave $\sim 100 \ \text{Wm}^{-1}\text{K}^{-1}$ at 5 K. However, since these calculations were for suspended graphene, we must account for phonon scattering with the substrate. To first order, we assume the same suppression factor as at room temperature [9] and estimate the lattice contribution to be $\approx 20 \ \text{Wm}^{-1}\text{K}^{-1}$. For SiO₂ and doped Si, we refer to previous low T measurements and extrapolate where necessary [10, 11]; $\kappa_{SiO2} \approx 0.1Wm^{-1}K^{-1}$, and $\kappa_{Si} \approx 1W \ m^{-1}K^{-1}$.

Interfaces are known to limit heat transport in vdW heterostructures. For graphene/hBN, we refer to calculations based on non-equilibrium Green's function [12]; $G_{hBN/graphene} \approx 1 \text{ M}$ Wm⁻² K^{-1} at 5 K. The same thermal conductance is assumed for the hBN/SiO₂ and SiO₂/Si interfaces - they do not significantly affect the temperature profile in graphene.

For the calculations shown in the main text, we take $P = 1 \mu W$, and impose an asymmetry in heating at the contacts by assuming $f_L = 0.15$, $f_R = 0.1$. The gradient of the temperature profile is non-zero largely within $\sim \pm 1 \mu m$ of the center as show in Fig. S11. Fig. S11 a shows the plot of calculated temperature rise ΔT versus position X whereas Fig. S11 b shows the temperature gradient plot with X. Both the plots shows individual contribution from the NW channel, left and right contacts towards the mentioned parameters.

References

- Novoselov, K. S. *et al.* Electric field effect in atomically thin carbon films. *science* 306, 666–669 (2004).
- [2] Purdie, D. *et al.* Cleaning interfaces in layered materials heterostructures. *Nature communications* **9**, 5387 (2018).
- [3] Wang, L. et al. One-dimensional electrical contact to a two-dimensional material. Science 342, 614–617 (2013).
- [4] Suyatin, D., Thelander, C., Björk, M., Maximov, I. & Samuelson, L. Sulfur passivation for ohmic contact formation to inas nanowires. *Nanotechnology* 18, 105307 (2007).
- [5] Bagnall, K. R., Muzychka, Y. S. & Wang, E. N. Analytical solution for temperature rise in complex multilayer structures with discrete heat sources. *IEEE Transactions on Components, Packaging and Manufacturing Technology* 4, 817–830 (2014).
- [6] Sichel, E., Miller, R., Abrahams, M. & Buiocchi, C. Heat capacity and thermal conductivity of hexagonal pyrolytic boron nitride. *Physical review B* 13, 4607 (1976).
- [7] Crossno, J. *et al.* Observation of the dirac fluid and the breakdown of the wiedemannfranz law in graphene. *Science* **351**, 1058–1061 (2016).
- [8] Fugallo, G. *et al.* Thermal conductivity of graphene and graphite: collective excitations and mean free paths. *Nano letters* **14**, 6109–6114 (2014).
- [9] Yang, J. et al. Thermal conductance imaging of graphene contacts. Journal of Applied Physics 116, 023515 (2014).
- [10] Dutta, M. & Jackson, H. E. Low-temperature thermal conductivity of amorphous silica. *Physical Review B* 24, 2139 (1981).
- [11] Asheghi, M., Kurabayashi, K., Kasnavi, R. & Goodson, K. Thermal conduction in doped single-crystal silicon films. *Journal of applied physics* **91**, 5079–5088 (2002).
- [12] Liu, Y. et al. Thermal conductance of the 2d mos 2/h-bn and graphene/h-bn interfaces. Scientific reports 7, 43886 (2017).