

Measurement of the Electronic Thermal Conductance Channels and Heat Capacity of Graphene at Low Temperature

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The ability to transport energy is a fundamental property of the two-dimensional Dirac fermions in graphene. Electronic thermal transport in this system is relatively unexplored and is expected to show unique fundamental properties and to play an important role in future applications of graphene, including optoelectronics, plasmonics, and ultrasensitive bolometry. Here, we present measurements of bipolar thermal conductances due to electron diffusion and electron-phonon coupling and infer the electronic specific heat, with a minimum value of $10k_B$ (10^{-22} J/K) per square micron. We test the validity of the Wiedemann-Franz law and find that the Lorenz number equals $1.32 \times (\pi^2/3)(k_B/e)^2$. The electron-phonon thermal conductance has a temperature power law T^2 at high doping levels, and the coupling parameter is consistent with recent theory, indicating its enhancement by impurity scattering. We demonstrate control of the thermal conductance by electrical gating and by suppressing the diffusion channel using NbTiN superconducting electrodes, which sets the stage for future graphene-based single-microwave photon detection.

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I. INTRODUCTION

Electrical transport in graphene has attracted much attention because of the pseudochiral and relativistic nature of the band structure [1,2]. Since both electrons and holes carry energy as well as charge, the thermal transport of Dirac fermions in two dimensions is expected to be as fascinating as its electrical counterpart. Theorists have suggested a number of intriguing possibilities: The relativistic hydrodynamics of a Coulomb-interacting electron-hole plasma may result in deviations from the Fermi-liquid values of the Mott relation, the Wiedemann-Franz ratio [3,4], and electronic specific heat [5]. Thermal transport measurements may reveal the physics of a neutral mode in the fractional quantum Hall effect [6]. The thermal properties of the electron gas are also critical to graphene-based device applications [7,8], as they impact photodetector performance [9], place fundamental limits on the mobility of charge carriers [10], and set the sensitivity of terahertz and microwave-frequency bolometers [11–13], which promise single-photon resolution due to the expected minute specific heat [11,14].

We present measurements of the bipolar thermal conductance over a temperature range of 300 mK to 100 K, using three different sample configurations

(described below). For temperatures below approximately 1 K, we identify the thermal transport due to electron diffusion G_{WF} , test the Wiedemann-Franz (WF) law, and infer the electronic heat capacity, with a minimum value of 10^{-20} J/K at 300 mK, which is 9 times smaller than the previous record [15]. For higher temperatures, we measure the thermal conductance due to phonon emission G_{ep} while varying the charge density. There has been recent theory [16] that explores the effects of electronic disorder on the electron-phonon (ep) coupling mechanism and predicts a substantial modification in comparison to earlier theory in the clean limit [17–20]; the disordered limit is defined by $\lambda_p \gg l_e$, where $\lambda_p = hs/(k_B T)$ is the dominant thermal phonon wavelength, l_e is the electron mean-free path, s is the sound velocity of graphene acoustic phonons, and k_B and h are Boltzmann's and Planck's constants, respectively. We present measurements that confirm both the effect of the disorder and the nature of the ep coupling (scalar or vector, screened or unscreened).

Previous thermal studies of graphene have been limited to measurements of thermoelectric power [21–23] or to measurements of thermal conductance taken at temperatures above the Bloch-Grüneisen (BG) temperature [24,25], at the charge neutrality point (CNP) [11], or without considering the effects of disorder [26]. Recent measurements of the behavior of Josephson junctions have provided insight into electron-phonon coupling at temperatures below 1 K [27]. Significant discrepancies between the theoretical [17–20] and measured values [26] of both the ep coupling temperature power law and the coupling constant are found in some of these experiments.

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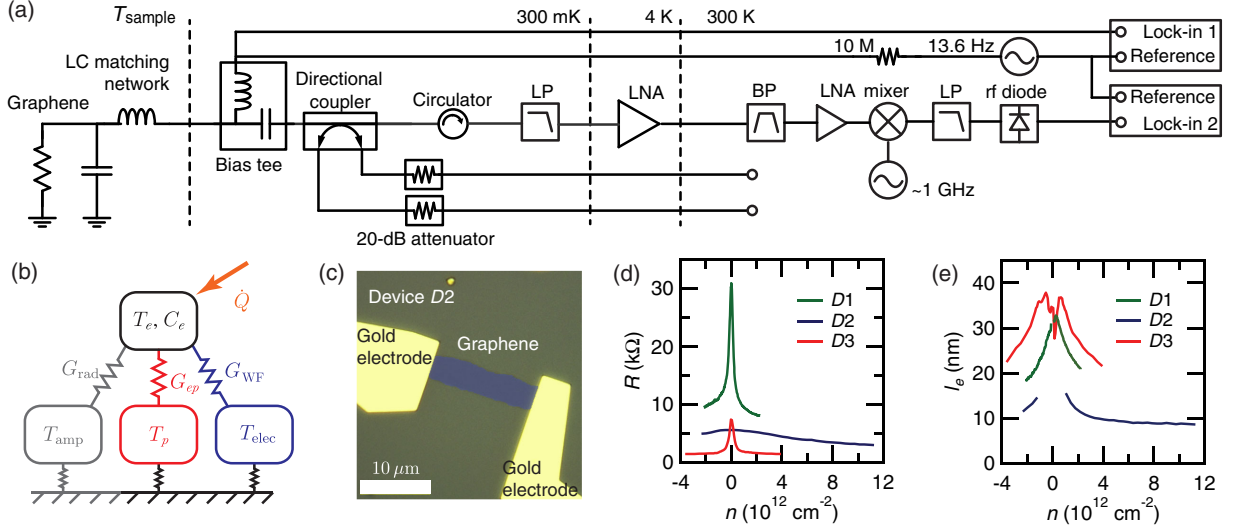


FIG. 1. (a) Experimental setup for simultaneously measuring the thermal and electrical transport of a graphene device: LC, inductor capacitor matching network; LP, low-pass filter; LNA, low-noise amplifier; and BP, band-pass filter. (b) Graphene thermal model. Heat from the graphene electrons can flow out through two different channels: electronic diffusion to the electrodes G_{WF} and ep coupling G_{ep} . (c) Optical micrograph of device $D2$. (d) dc graphene resistances. (e) Electrical mean-free path calculated using $l_e = \sigma E_F / n e^2 v_F$.

II. EXPERIMENTS

We probe thermal transport of the electrons in graphene by applying Joule heating and measuring the electron temperature utilizing ultrasensitive, microwave-frequency Johnson-noise thermometry with a sensitivity of $2 \text{ mK}/\sqrt{\text{Hz}}$ [11,28]. [See Fig. 1(a) and the Supplemental Material (SM) [29] and Refs. [30–33] therein.] Figure 1(b) shows the expected thermal model of the electron gas. With a typical coupling bandwidth of 80 MHz to the graphene [11], one-dimensional thermal transport [34] through blackbody radiation $G_{\text{rad}} (\approx 10^{-15} \text{ pW/K}) \ll (G_{WF}, G_{ep})$ is expected to be negligible in this experiment. We assume both the electrodes and lattice are in thermal equilibrium with the sample stage, as the ep coupling in a normal metal [35] and the boundary thermal conductance of the SiO_2 -graphene interface [36] are large compared to the G_{WF} and G_{ep} thermal channels. Three devices with different electrodes and gating materials (see Table I and the SM) are measured in two cryostats to cover the entire sample temperature range: 0.3–1.5 K and $T > 1.5 \text{ K}$. For all three samples, the device length is much longer than the inelastic scattering length l_e , which avoids any issues of electron shot noise [38–40]. For charge densities that can be reached with our experiment $n = 10^{11}\text{--}10^{13} \text{ cm}^{-2}$, the transition from ep to electron-diffusion cooling is expected to occur at approximately 1 K and should be apparent because of the difference in temperature dependence of the thermal conductance: G_{ep} and G_{WF} are expected to depend on temperature as $T^{\delta-1}$ (with $\delta \geq 3$ typically) and T , respectively.

With Joule heating \dot{Q} applied to the electron gas, the electron temperature T_e is expected to follow the two-dimensional heat transfer differential equation

$$\dot{q} = -\nabla \cdot (\kappa_{WF} \nabla T_e) + \Sigma_{ep}(T_e^\delta - T_p^\delta), \quad (1)$$

where $\dot{q} = E^2/\rho$ is the local Joule heating (such that $\int \dot{q} d^2\mathbf{r} = \dot{Q}$), E is the local electric field, ρ is the electrical resistivity, κ_{WF} is the thermal conductivity due to electronic diffusion, Σ_{ep} is the ep coupling parameter, and T_p is the local phonon temperature. On the right-hand side, the first term describes diffusive cooling through the electron gas, while the second term describes cooling by phonon emission. In a Fermi liquid, the local Joule heating and diffusive cooling are connected through the WF law $\kappa_{WF} = \mathcal{L}_0 T_e / \rho$, where \mathcal{L}_0 is the Lorenz number given by $(\pi^2/3)(k_B/e)^2$. Since the

TABLE I. Sample information and measured quantities. μ is the electronic mobility from fitting [37]. l_e and $k_F l_e$ are quoted for data nearest to $n = 3.5 \times 10^{12} \text{ cm}^{-2}$, corresponding to a Bloch-Grüneisen temperature $T_{BG} = 2(s/v_F)(E_F/k_B) \approx 101 \text{ K}$. The disorder temperature is given by [16] $T_{\text{dis}} = \hbar s/l_e$. At this density, $T_{\text{dis}} < T_{BG}$ and $k_F l_e > 1$.

| Devices | D1 | D2 | D3 |
|---|-----------|-------------|-------------|
| Gate | Local top | Global back | Global back |
| Electrodes | Ti/Au | Ti/Au | Ti/Au/NbTiN |
| Length (μm) | 15 | 15.4 | 4.6 |
| Average width (μm) | 6.8 | 3.6 | 5.4 |
| $\mu (\text{cm}^2 \text{V}^{-1} \text{s}^{-1})$ | 5800 | 200 | 5100 |
| l_e (nm) | 21 | 10 | 22 |
| $k_F l_e$ | 5.5 | 3.3 | 7.5 |
| T_{dis} (K) | 46 | 96 | 42 |
| \mathcal{D} (eV) | 19 | 23 | 51 |

temperature of the sample will not be uniform (the middle will have a higher temperature than the leads) and we measure the average electron-noise temperature, the WF relationship will be modified to $G_{\text{WF}} = \alpha \mathcal{L}_0 T_p / R$, where R is the graphene resistance and $\alpha = 12$ (see the SM for discussion) [41].

By computing the ratio of \dot{Q} to the measured increase in average electron temperature with $\Delta T_e / T_p \ll 1$, we determine the thermal conductance G_{th} . Figure 2(a) shows the results from device *D1* (gold leads, top gated) at various charge-carrier densities. There is a clear transition from a quadratic to a linear temperature dependence at approximately 1 K, which is expected and can be understood as G_{WF} dominating at low temperatures [Fig. 2(b)]. We test the Wiedemann-Franz law for two-dimensional Dirac fermions by plotting G_{WF} versus T/R [Fig. 2(c)] such that the slope is $\alpha \mathcal{L}_0$. We also note that this G_{WF} is not equal to zero at $T/R = 0$, which at this point is not understood. Figure 2(d) shows the measured Lorenz number at different densities. The averaged Lorenz-number values for electron and hole doping are $1.34 \mathcal{L}_0$ and $1.26 \mathcal{L}_0$, respectively.

Our measured Lorenz number is 35% higher than the Fermi-liquid value, 17% higher than the measured value in graphite [42], and comparable to values obtained in other materials [43]. While the electron-electron (e - e) interaction may modify the Lorenz number in a material [3,4,43], other effects such as contact resistance and contributions due to graphene under the contacts could contribute to errors in our calculation of the Lorenz number. Four-point probe measurements of the thermal conductance may solve this problem in the future. We also observe an increase of the Lorenz number near the CNP [Fig. 2(d)]. At the Dirac point and at high temperatures where $E_F \ll k_B T$ (where E_F is the Fermi energy), theory predicts that the system becomes quantum critical and the interaction between massless electrons and holes enhances the Lorenz number [3,4]. The same theory also expects that a deviation from the Fermi-liquid value is possible in the case where $E_F \gg k_B T$, as is true for our impurity-limited samples, but only if the screening is weak [3]. Further experiments in cleaner samples are needed to understand this anomalous behavior in the Lorenz number as well as the offset in Fig. 2(c).

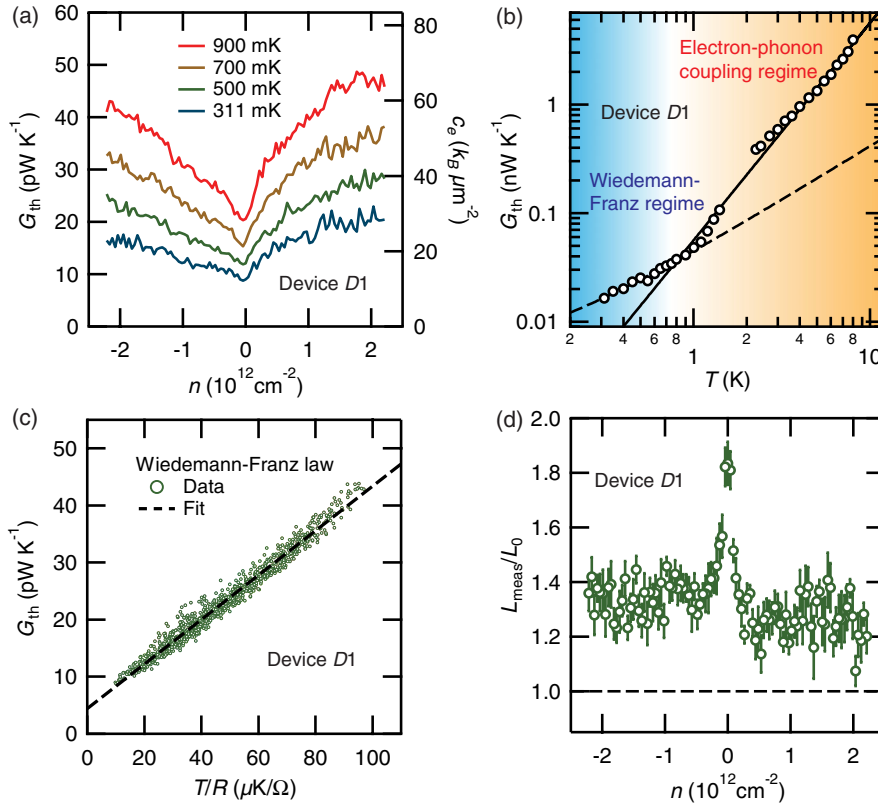


FIG. 2. Data from device *D1* with normal metallic electrodes. (a) The bipolar G_{WF} as a function of charge-carrier density at various sample temperatures. (b) G_{th} data as a function of temperature at $n = -2.2 \times 10^{12} \text{ cm}^{-2}$. The solid line is the power-law fit to the ep thermal conductance above 1.5 K, while the dashed line is the best linear T fit to the WF thermal conductance. The size of the data points represents the measurement error. (c) Wiedemann-Franz law in graphene. Each data point represents a measurement at a different temperature and charge-carrier density. The fitted line is $G_{\text{WF}} = \alpha \mathcal{L}_{\text{meas}} T / R$ with a y offset; $\mathcal{L}_{\text{meas}} = 3.25 \pm 0.02 \times 10^{-8} \text{ W}\Omega\text{K}^{-2}$. (d) The measured Lorenz number $\mathcal{L}_{\text{meas}}$ as a function of density from fitting of the Wiedemann-Franz law. For electrons with $n \leq -0.18 \times 10^{12} \text{ cm}^{-2}$, averaged $\mathcal{L}_{\text{meas}}/\mathcal{L} = 1.34 \pm 0.06$, while for holes with $n \geq +0.18 \times 10^{12} \text{ cm}^{-2}$, averaged $\mathcal{L}_{\text{meas}}/\mathcal{L} = 1.26 \pm 0.07$.

The electronic specific heat capacity c_e in graphene can be determined by applying a two-dimensional kinetic model: $\kappa_{\text{WF}} = (1/2)c_e v_F l_e$, where $C_e = A c_e$ is the total electronic heat capacity, v_F is the Fermi velocity, and A is the device area. Using $l_e = 32$ nm [Fig. 1(e)], we plot C_e on the right-hand side of the y axis in Fig. 2(a). Since $G_{\text{WF}} \propto T$, and l_e does not depend on temperature significantly because of impurity scattering, the measured specific heat is linear in T for all densities. This measured linear behavior agrees with theories for $|E_F| \gg k_B T$, as the minimum $|E_F|$ of our samples is limited by impurity doping. For $|E_F| \ll k_B T$, which is not accessible in this experiment, the specific heat is expected to be proportional to T^2 in the case of massless Dirac fermions [44]. The smallest specific heat attained near the CNP is merely $10k_B/\mu\text{m}^2$ or $1000k_B$ for the whole sample, a factor of 9 smaller than the inferred value in state-of-the-art nanowires used for bolometry [15]. This value is also consistent with our earlier result at 5 K estimated using a bolometric mixing effect [11].

At higher sample temperatures or for large Joule heating, the thermal conductance changes its temperature power-law behavior [Fig. 2(b)] as the dominant cooling

mechanism switches from G_{WF} to G_{ep} ; the crossover temperature is given by $(\alpha \mathcal{L}_0 / \delta R A \Sigma_{ep})^{1/(\delta-2)}$. In this regime, Eq. (1) reduces to [16,19,20]

$$\dot{Q} = A \Sigma_{ep} (T_e^\delta - T_p^\delta). \quad (2)$$

Using a dc current bias, Fig. 3(a) plots the measured T_e versus the applied Joule heating power. For large heating powers, the electron temperature converges to $(\dot{Q}/A \Sigma_{ep})^{1/\delta}$, independent of the initial temperature. The solid lines in Fig. 3(a) are the best fit to $T_e = (\dot{Q}/A \Sigma_{ep} + T_p^3)^{1/3}$, establishing $\delta = 3$. In light of recent theory [16], our experimental data suggest that the ep heat transfer in the disordered limit is primarily due to a weakly screened deformation potential, consistent with recent electrical transport measurements [45]. The simplified physical scenario is that impurity scattering in disordered graphene prolongs the ep interaction time and thus enhances the emission rates. Recent investigations [24,25,46] suggest that the same power law $\delta = 3$ may also govern the disorder-assisted cooling rate of hot electrons for $T > T_{\text{BG}}$, but through a different mechanism.

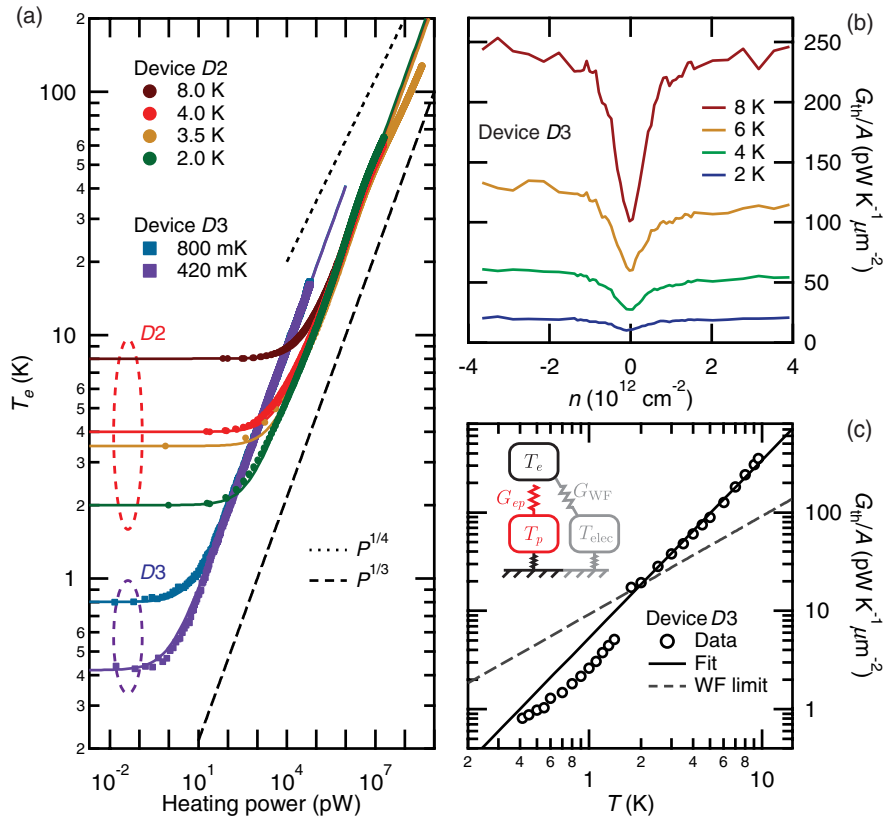


FIG. 3. (a) The measured electron temperature versus dc heating power applied to the graphene devices at different phonon temperatures with $n = 28 \times 10^{12} \text{ cm}^{-2}$ for device D2 and $n = -2.2 \times 10^{12} \text{ cm}^{-2}$ for device D3. The solid lines are fits to Eq. (2). The power law T^3 persists down to 420 mK for device D3 with NbTiN electrodes. (b),(c) Data from device D3 with NbTiN electrodes: (b) the bipolar electron-phonon thermal conductance as a function of charge-carrier density at various temperatures and (c) G_{th} data as a function of temperature at $n = -2.9 \times 10^{12} \text{ cm}^{-2}$. The solid line is the power-law fit to the ep thermal conductance above 1.5 K, while the dashed line is the calculated $G_{\text{WF}} = \alpha \mathcal{L}_0 T/R$ using $R = 1770 \Omega$. The offset between high- and low-temperature data is due to thermal cycling of the device on two cryostats.

For the three devices we fabricate and measure, all show $\delta \approx 3$ except for device *D2* (gold leads, back gated) at temperatures above 50 K and device *D1* near the CNP [see Fig. 4(b)]. In both circumstances, the temperature power law increases from $\delta = 3$ to 4. This behavior of *D1* near the CNP, where transport is expected to be dominated by disorder and charge puddles, is surprisingly consistent with theoretical expectations [17–20] in the clean limit, as reported in Ref. [11]. In this device, as the charge-carrier density decreases to 10^{11} cm^{-2} near the CNP, the screening length grows to 50 nm, which is comparable to the distance to the nearby metallic top gate (100 nm, dielectric constant of 4). We speculate that this screening of the metallic gate may impact the *ep* coupling. Moreover, near the CNP, the impurity scattering is long range in nature and $k_F l_e < 1$, which is outside the regime of validity of the existing *ep* coupling theory for disordered graphene [16]. More experiments and theory are required to understand the nature of the *ep* coupling at low charge-carrier density, which is of particular importance to graphene-based bolometry, as the ultimate sensitivity is expected to

be limited by the *ep* coupling at the lowest carrier densities [11,14].

We can further explore Eq. (2) by measuring the differential thermal conductance at different temperatures and carrier densities using a small ac current bias. If $T_e - T_p \ll T_p$, the *ep* thermal conductance is $\delta A \Sigma_{ep} T_p^{\delta-1}$. Figure 3(b) shows G_{ep} as function of carrier density for device *D3* (superconducting leads, back gated). Figures 2(b) and 3(c) for devices *D1* and *D3*, respectively, show that G_{th} is limited by the *ep* thermal conductance at $T > 1.5$ K with a power law $\delta \approx 3$ for both devices. However, since the electrons in a superconductor have negligible entropy and do not conduct thermally, G_{th} of device *D3* with NbTiN electrodes is not limited by G_{WF} at low temperatures. The dashed line in Fig. 3(c) is the calculated G_{WF} , similar to the dashed line in Fig. 2(b). NbTiN is used because of its higher transition temperature (14 K) to avoid *e-e* interaction that may promote hot electrons over the superconducting band gap [47]. The *ep* data at 0.4 K demonstrate the suppression of heat diffusion by roughly 80%. Recent experiments have also shown Pb to be very effective at suppressing electron diffusion down to temperatures below 100 mK [27].

We can obtain Σ_{ep} and δ by fitting G_{ep} as function of lattice temperature at a constant carrier density. Results are plotted in Fig. 4. For devices *D2* and *D3*, we find δ values of approximately 3.0 and 2.8, respectively. Near the CNP, Σ_{ep} has a minimum but is not vanishing. Furthermore, the fitted Σ_{ep} values vary by an order of magnitude across all three devices. This variation and also the discrete jump in measured G_{th} after thermal cycling in Fig. 3(c) indicate that the underlying *ep* coupling mechanism is strongly modified by disorder. Although it is difficult to compare precisely, our results for Σ_{ep} and δ are in reasonable agreement with the recent measurements of the *ep* coupling using the trapping dynamics of graphene-lead Josephson junctions at temperatures spanning 50 to 700 mK [27].

We compare the theory of *ep* coupling in disordered graphene to both the measured electrical and *ep* thermal transport data using [16]

$$\Sigma_{ep} = \frac{2\zeta(3)}{\pi^2} \frac{E_F}{v_F \rho_M} \frac{\mathcal{D}^2 k_B^3}{\hbar^4 l_e s^2}, \quad (3)$$

where \mathcal{D} is the deformation potential and ρ_M is the mass density of the graphene sheet. This theory assumes a short-range scattering impurity potential and $k_F l_e > 1$. The estimated values of \mathcal{D} for devices *D1*, *D2*, and *D3*, at a charge density of $n \approx 3.5 \times 10^{12} \text{ cm}^{-2}$, are 19, 23, and 51 eV, respectively (Table I). The considerable scatter in the inferred values of \mathcal{D} is consistent with the range of values obtained from electrical transport measurements [37,45,48,49]. The inset of Fig. 4(b) shows the inferred deformation potential values in both the thermal and electrical experiments

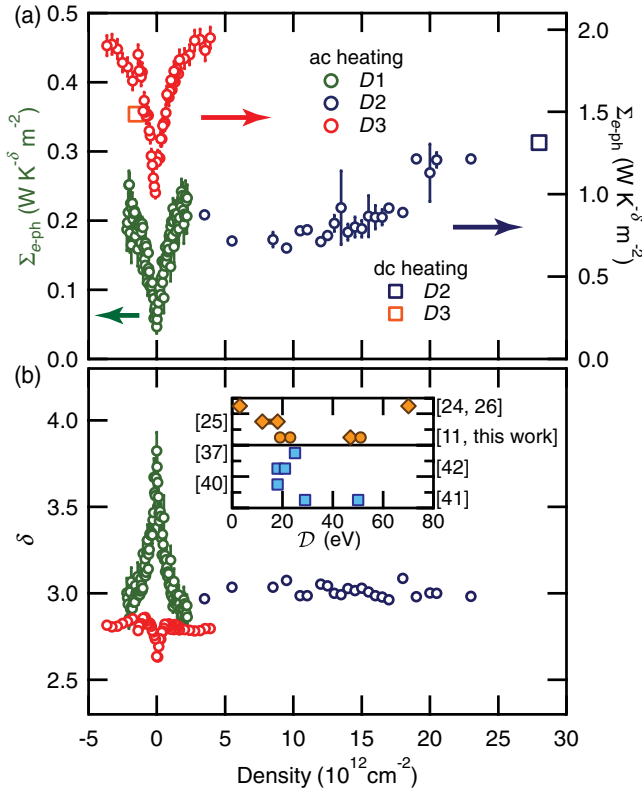


FIG. 4. (a) The electron-phonon coupling parameter Σ_{ep} and (b) the temperature power law δ extracted from the thermal conductance data above 1.5 K as a function of charge-carrier density in three measured devices. Inset: The values of the deformation potential \mathcal{D} in graphene from the literature: Blue squares and orange diamonds represent data based on electrical- and thermal-related measurements, and orange circles represent the data from this report (see also Table I).

reported in the literature [11,24–26,37,45,48,49]. The wide scatter suggests that some factors may not be captured in the ep coupling theory for disordered graphene, such as long-range impurity scattering, substrate-induced effects, or surface-acoustic phonons [50].

III. CONCLUSIONS

In this report, we investigate the bipolar thermal conductance of graphene in both the electron-diffusion and electron-phonon regimes. We find that the ep coupling is strongly modified by electronic disorder and is consistent with scalar coupling in the weak screening limit [16]. ep coupling in the disordered limit is especially relevant for ultrasensitive device applications at low temperatures, since even the cleanest samples [37,49] yet reported ($l_e > 1 \mu\text{m}$) would cross into the disordered limit for temperatures below 1 K. This experiment has validated the WF law for two-dimensional Dirac fermions. It may be possible to study many-body physics in this system through more precise measurements of the Lorenz ratio and with cleaner samples near the CNP. The electronic specific heat inferred through the electron-diffusion measurement is merely $10k_B/\mu\text{m}^2$. Our estimates suggest that a single terahertz photon should be detectable at 300 mK using a $1\text{-}\mu\text{m}^2$ -size sample and a SQUID amplifier [11,14,51]. We have also demonstrated the control of heat flow in graphene by both using the field effect and employing superconducting NbTiN to suppress the thermal diffusion channel. These findings point the way to future experiments to probe both the fundamental and practical electronic thermal properties of this unique atomically thin material.

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