The thermopower and Nernst Effect in graphene in a magnetic field

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(Dated: January 12, 2009)

We report measurements of the thermopower $S$ and Nernst signal $S_{yx}$ in graphene in a magnetic field $H$. Both quantities show strong quantum oscillations vs. the gate voltage $V_g$. Our measurements for Landau Levels of index $n \neq 0$ are in quantitative agreement with the edge-current model of Girvin and Jonson (GJ). The inferred off-diagonal thermoelectric conductivity $\sigma_{yx}$ comes close to the quantum of Ampers per Kelvin. At the Dirac point ($n = 0$), however, the width of the peak in $\sigma_{yx}$ is very narrow. We discuss features of the thermoelectric response at the Dirac point including the enhanced Nernst signal.

In graphene, the linear dispersion of the electronic states near the chemical potential $\mu$ is well described by the Dirac Hamiltonian. As shown by Novoselov et al. [1, 2, 3] and by Zhang et al. [4, 5, 6] quantization of the electronic states into Landau Levels leads to the integer quantum Hall Effect (QHE). Because of the linear dispersion, the energy $E_n$ of the Landau Level (LL) of index $n$ varies as $E_n = sgn(n) \sqrt{2 e n h^2 B |n|}$, where $B$ is the magnetic induction, $v_F$ the Fermi velocity, $e$ the electron charge, and $h$ is Planck's constant. The quantized Hall conductivity is given by

$$\sigma_{xy} = \frac{4 e^2}{h} \left( n + \frac{1}{2} \right), \quad (1)$$

where the factor 4 reflects the degeneracy of $g$ each of LL (2 each from spin and valley degrees).

Detailed investigations of the longitudinal resistance $R_{xx}$ and Hall resistance $R_{xy}$ have been reported by several groups [1, 2, 3, 4, 5, 6, 7]. By contrast, the thermoelectric tensor $S_{ij}$ is less investigated. $S_{ij}$ relates the observed electric field $E$ to an applied temperature gradient $-\nabla T$, viz. $E = \tilde{S} \cdot (-\nabla T)$. On the other hand, the charge current density $J$ produced by $-\nabla T$ is expressed by the thermoelectric conductivity tensor $\tilde{\sigma}$, viz. $J = \tilde{\sigma} \cdot (-\nabla T)$. Although $J$ is not measured directly, $\alpha_{ij}$ may be obtained by measurements of both $S_{ij}$ and the resistivity tensor $\rho_{ij} = R_{ij}$. (By convention, $-S_{xx}$ is the thermopower $S$; we refer to $S_{yx} = E_y/|\nabla T|$ as the Nernst signal.)

A most unusual feature of the thermoelectric response of a QHE system (for $n \neq 0$) is that, despite the dominance of the off-diagonal (Hall-like) current response, the thermopower displays a large peak at each LL whereas the Nernst signal is small. In the geometry treated by Girvin and Jonson [4, 5, 6] (Fig. 1a), the 2D sample is of finite width along $\hat{x}$, but is infinite along $\hat{y}$ (with the applied magnetic field $H||\hat{z}$). As we approach either edge, the LL energy $E_n$ rises very steeply (bold curve). At $T = 0$, edge currents $I_y$ exist at the intersections (open circles) of $E_n$ with the chemical potential $\mu$. In a gradient $-\nabla T$, the magnitude of $I_y$ is larger at the warmer edge than at the cooler edge because of increased occupation of states above $\mu$. The difference $|\delta I_y|$ is a maximum when $\mu$ is aligned with $E_n$ in the bulk. The corresponding value of $\alpha_{yx}$ is then a universal quantum ($k_B e / h$) $\ln 2$ with units of Amperes per Kelvin ($k_B$ is Boltzmann's constant). In turn, $\delta I_y$ produces a quantized Hall voltage $V_H = (h/e^2) \delta I_y$ that drops $|\hat{x}|$. Hence, conflating these 2 large off-diagonal effects, the thermopower $S$ becomes very large when $\mu$ aligns with $E_n$ ($n \neq 0$). By contrast, the transverse (Nernst) voltage is small (in the absence of disorder).

In graphene, this picture needs revision when $\mu$ is at the Dirac point. For the $n = 0$ LL, the nature of the edge currents is the subject of considerable debate [11, 12, 13, 14]. What are the profiles of $S$ and $S_{yx}$? We have measured $S_{ij}$ and $R_{ij}$ to a maximum $H$ of 14 T at 20 and 50 K. Our results reveal that, at 9 T, the thermoelectric response in graphene already falls in the quantum regime at 50 K. The inferred off-diagonal current response $\alpha_{yx}$ is a series of peaks close to the quantum value $(g k_B e / h) \ln 2$ (independent of $n$, $B$ and $T$). We compare our results with the calculations of Girvin and Jonson (GJ), and discuss features specific to the $n = 0$ LL at the Dirac point.

Kim and collaborators [8] have pioneered a lithographic design for measuring the thermopower of carbon nanotubes. We have adopted their approach with minor modifications for graphene. Using electron-beam lithography, we deposited narrow gold lines which serve as a micro-heater to produce $-\nabla T$ and thermometers (Fig 1b). The latter are also used as current leads when $R_{xx}$ and $R_{xy}$ are measured. Above 10 K, the thermometers can resolve $\delta T \sim 10$ mK. The typical $\delta T \sim$ 10 mK between the 2 thermometers. However, the small spacing of the voltage leads ($\sim 2 \mu m$) leads to an uncertainty of $\pm 10\%$ in estimating $\delta T$ between them. A slowly oscillating current at frequency $\omega$ is applied to the heater and the resulting thermoelectric signals are detected at $2 \omega$ and $-90^\circ$ out of phase.

In our geometry with $-\nabla T || \hat{x}$, $H || \hat{z}$, the charge current density $J$ is (summation over repeated indices implied)

$$J_i = \sigma_{ij} E_j + \alpha_{ij} (-\partial_j T) \quad (i = x, y). \quad (2)$$

The (2D) conductivity and thermoelectric conductivity tensors are often written as $\sigma_{ij} = L_{ij}^{(2)} (e^2 / T)$ and $\alpha_{ij} = L_{ij}^{(2)} / T^2$, respectively. Setting $J = 0$, we have for the
FIG. 1: (Color online) (Panel a) The effect of $-\nabla T$ on the edge currents $I_y$ in a QHE system ($n \neq 0$). The energy $E_n$ of a LL (bold curve) increases very steeply at the sample edges, with $\mu$ the chemical potential (dashed line). If $H_z > 0$, $I_y$ is negative (positive) at the left (right) edge, as indicated by open circles. The magnitude $|I_y|$ is larger at the warmer edge. Fermi-Dirac distributions $f(E)$ are sketched at the sides. (Panel b) Curves of thermopower $S = -S_{xx}$ vs. gate voltage $V_g$ in Sample K10 at selected $T$. The curves are antisymmetric about the Dirac Point which occurs at the offset voltage $V_0 = 15.5$ V. The peak value $S_{xx}$ is nominally linear in $T$ from 25 to 300 K (Panel c). Less complete data from sample K59 are also plotted. A photo of Sample K10 (faint polygon) is shown in Panel (d). A micro-heater as well as thermometers (therm) and signal leads are patterned with electron-beam lithography. The black scale bar is 3 $\mu$m.

Observed $E$-fields

$$E_i = -\rho_{ik}\alpha_{kj}(-\partial_j T) = S_{ij}(-\partial_j T),$$

with $\rho_{ij} = R_{ij}$ the 2D resistivity tensor. The thermopower $S = -E_x/|\nabla T|$ equals $\rho_{xx}\alpha_{xx} + \rho_{xy}\alpha_{xy}$ ($S > 0$ for hole doping), while the Nernst signal is given by (with $\rho_{xx} = \rho_{yy}$)

$$S_{yx} = \rho_{xx}\alpha_{xy} - \rho_{xy}\alpha_{xx}. \quad (4)$$

The 2 terms tend to cancel mutually, except at the Dirac point where $\rho_{yx}$ vanishes (see below).

Inverting Eq. (3), we may calculate the tensor $\alpha_{ij}$ from measured quantities. We have

$$\alpha_{xx} = -(\sigma_{xx}E_x + \sigma_{xy}E_y)/|\nabla T|$$

$$\alpha_{xy} = (-\sigma_{xy}E_x + \sigma_{xx}E_y)/|\nabla T|. \quad (5)$$

Under field reversal ($\mathbf{H} \to -\mathbf{H}$), $S$ is symmetric whereas $S_{yx}$ is antisymmetric. For each curve taken in field, we repeat the measurement with $\mathbf{H}$ reversed. All curves of $S$ and $S_{yx}$ reported here have been (anti)symmetrized with respect to $\mathbf{H}$. As for charge-inversion symmetry, we expect the sign of $S$ to change with the shifted gate voltage $V_g' \equiv V_g - V_0$ (i.e. between hole and electron filling), but the sign of $S_{yx}$ stays unchanged ($V_0$ is the offset voltage). However, we have not imposed charge-inversion symmetrization constraints on the curves. Apart from field (anti)symmetrization, all the curves are the raw data.

Figure 2 shows traces of $S$ vs. $V_g$ at selected $T$. The thermopower $S$ changes sign as $V_g$ crosses the charge-neutral point (Dirac Point), assuming positive (negative) values on the hole (electron) side. The peak value $S_{xx}$ is nominally $T$-linear from ~20 K to 300 K (Fig. 2b).
In sharp contrast to the smooth variation in Fig. 1, the curves of $S$ vs. $V_g$ show pronounced oscillations when $H$ is finite, reflecting Landau quantization of the Dirac states. Figures 2a, b and c display $S$ vs. $V_g$ (bold curves) with $H$ fixed at the values 5, 9 and 14 T, respectively. For comparison, we have also plotted (as thin curves) the conductance $G_{xx}$ vs. $V_g$ (with $H$ fixed at the values 5, 9 and 14 T, respectively). All curves were measured at 20 K. Vertical lines locate the maxima of $G_{xx}$. The sign of $S_{yx}$ was incorrectly assigned in a previous version of this paper [15].

In Fig. 3, we compare the Nernst signal $S_{yx}$ vs. $V_g$ (bold curves) and conductance $G_{xx}$ vs. $V_g$ (thin curves) in J3 at the 3 field values $H = 5$, 9 and 14 T (Panels a, b and c, respectively). All curves were measured at 20 K. Vertical lines locate the maxima of $G_{xx}$. The sign of $S_{yx}$ was incorrectly assigned in a previous version of this paper [15].

In Fig. 4, we compare the Nernst signal $S_{yx}$ vs. $V_g$ (bold curves) and conductance $G_{xx}$ vs. $V_g$ (thin curves) in J3 at the 3 field values $H = 5$, 9 and 14 T (Panels a, b and c, respectively). All curves were measured at 20 K. Vertical lines locate the maxima of $G_{xx}$. The sign of $S_{yx}$ was incorrectly assigned in a previous version of this paper [15].

In the theory of GJ [9], valid for GaAs-based devices, the edge current difference is $\delta I_y = (\mu_e/\hbar)\sum_n \int d\epsilon (\partial \epsilon/\partial k)(\partial f/\partial T)\delta T$, with $f(\epsilon)$ the Fermi-Dirac distribution. The off-diagonal term $\alpha_{xy}$ is given by $(e/\hbar T)\int_{E_n}^\infty d\epsilon (\epsilon - \mu - \epsilon/\hbar T)\delta f/\partial \epsilon$. When $\mu = E_n$, $\alpha_{xy}$ attains a peak value, corresponding to a quantized current per Kelvin, given by ($g=1$)

$$\alpha_{xy}^{max} \approx \frac{k_B e}{\hbar} \ln 2 \sim 2.32 \text{nA/K}.$$  

GJ find that $S$ displays a series of peaks, with the peak profiles of $S$. Moreover, $S_{yx}$ is smaller in magnitude by a factor of 4-5. For the $n = 0$ LL, however, the profiles change character, with $S_{yx}$ displaying a large positive peak. The reason for the enhancement of $S_{yx}$ at the Dirac point is discussed below. The positive sign of $S_{yx}$ at the $n=0$ LL implies that the Nernst $E$-field $E_N$ is parallel to $H \times (-\nabla T)$ [13].
value at LL $n$ given by
\[ S_{\text{peak}}(n) = \frac{k_B}{e} \ln \left( \frac{2}{n + \frac{1}{2}} \right). \tag{7} \]

At low $T$, $S$ is independent of $H$ and $T$.

In Fig. 2 the peak value of $S$ at the $n = -1$ LL increases from 25 at 5 T to 41 $\mu$V/K at both 9 and 14 T. Moreover, as $T$ increases from 20 to 50 K (Panel c), the peak increases only weakly (41 to 48 $\mu$V/K) in sharp contrast with the $T$-linear behavior of $S$ (at the peak) saturates to a value independent of $T$ and $H$ at sufficiently low $T$. This saturation contrasts with the $T$-linear behavior of $S$ in $H=0$ (Fig. 2).

In graphene, however, Berry phase effects lead to a the $\frac{1}{2}$-integer shift in Eq. 4. In evaluating $\sigma_{xy} \sim -\sum_n f(E_n)$, the $\frac{1}{2}$-integer shift implies that $S_{\text{peak}}(n)$ decreases as $k_B \ln 2/(en)$, instead of Eq. 7. The measured values $S_{\text{peak}} = 41 \mu$V/K at $n = -1$ at 9 T already exceeds slightly the predicted value 39.7 $\mu$V/K in Eq. 4. Future experiments on cleaner samples may yield values closer to the predicted value 59.6 $\mu$V/K for Dirac systems. Regardless, for LL with $n \neq 0$, our observations are generally consistent with the GJ theory. In principle, Eq. 7 provides a way to measure $\delta T$ on micron-scales with a resolution approaching voltage measurements.

The most interesting question is the thermolectric response of the $n=0$ LL. This is easier to analyze using the pure thermolectric currents $\alpha_{xx}$ and $\alpha_{xy}$ (obtained using Eqs. 4). In Fig. 4, $\alpha_{xy}$ and $(\alpha_{xx})$ is plotted as bold (thin) curves for $H=9$ T and $T = 20$ K. Panel (b) shows the curves at 14 T. Compared with $S$ vs. $g$, the peaks in $\alpha_{xy}$ are much narrower and clearly separated by intervals in which $\alpha_{xx}$ is nominally zero. Likewise, the purely dispersive profile of $\alpha_{xx}$ is also more apparent. Consistent with the GJ theory, the overall magnitude of $\alpha_{xy}$ (for $n \neq 0$) is larger than that of $\alpha_{xx}$.

A striking feature of $\alpha_{xy}$ is that its peaks are independent of $n$. Their average value $\sim 75$ $\text{nA/K}$ reaches to within 30% of the quantized value of Eq. 6 with $g = 4$ (horizontal dashed line). The largest uncertainty in our measurement is in estimating the gradient between the voltage leads. For the $n = 0$ LL, the shortfall may also reflect incipient splitting of the Landau sublevels (compare 9 and 14 T traces). The uniformity of the peaks in $\alpha_{xy}$ accounts for the observed enhancement of the Nernst peak at the Dirac point. By Eq. 8 $S_{yx}$ is the difference of 2 positive terms. For $n \neq 0$, the 2 terms are matched, and the partial cancellation leads to a dispersive profile. However, for $n = 0$, the vanishing of $\rho_{yx}$ as $V'_{g} \to 0$ strongly suppresses the second term $-\rho_{yx}\alpha_{xx}$. The remaining term $\rho_{xx}\alpha_{xy}$ then dictates the size and profile of the Nernst peak.

We note that the sign of $\alpha_{xy}$ is a direct consequence of the edge-currents 4. When $H_x > 0$, $I_y < 0$ on the warmer edge and $I_y > 0$ on the cooler edge, as depicted in Fig. 1c. As a result, $\alpha_{xy} > 0$ for both hole-and electrondoping.

Our measurements are consistent with the GJ theory for $n \neq 0$. For the $n=0$ LL, however, there is considerable uncertainty about the nature of the edge states in graphene (or whether they exist in large $H$). The simple edge-current picture for understanding the peaks in $\alpha_{xy}$ may need significant revision for $n = 0$, despite the similarity of the peak magnitude. We also note that the peak at $n = 0$ is much narrower (by a factor of 5) than the other peaks. This is also not understood. By going to cleaner samples and higher fields, we hope to exploit this narrow width to resolve splitting of the 4 sublevels at $n = 0$.

We are grateful to P. A. Lee for many discussions. The research is supported by NSF through a MRSEC grant (DMR-0819860).

Note added After we completed these experiments, we learned of 2 thermopower and Nernst experiments on graphene posted recently (Yuri M. Zuev et al., cond-mat arXiv: 0812.1393 and Peng Wei et al., cond-mat arXiv: 0812.1411).

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[15] In a previous posted version of this manuscript (cond-mat arXiv:0812.2866v1), the sign of $S_{yx}$ was incorrectly assigned. After careful checking and further measurements, we have determined that $S_{yx} > 0$ and $\alpha_{xy} > 0$ at $n = 0$ as reported here. The sign of $E_y$ is the same as that for the vortex-Nernst effect in superconductors.