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Supplementary information

Thermodynamics of free and bound magnons in graphene

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Supplementary information for "Thermodynamics of free and bound magnons in graphene"

1. Spin skyrmion model

In order to capture the ν=1 gap suppression by the presence of magnons, we formulate a skyrmion model that takes into account the effects of finite temperature and non-zero charge density on the measured chemical potential $\mu(\nu)$. We assume that both the density of overturned spins and the deviation from ν=1 is small. The energy of an elementary charged excitation at ν=1, $E_s^{e,h}$, is given by

$$E_s^{e,h} = \epsilon_s^{e,h} + \left(s + \frac{1}{2}\right) E_Z,$$

where *e* and *h* denote electron-like and hole-like excitations, respectively, s is the number of excess flipped spins ($s = 0$ for a bare electron or hole) bound to the charge, $\epsilon_s^{e,h}$ is the energy of the charged excitation in the absence of Zeeman coupling (i.e. due to Coulomb interaction alone), and $E_Z = g\mu_B B$ is the Zeeman energy. In the absence of Landau level mixing, particle-hole symmetry implies

$$\epsilon_s^e = \epsilon_s^h.$$

The skyrmion energies ϵ_s^e are taken to be $\epsilon_0^e = 0.6266E_C$, $\epsilon_1^e = 0.5737E_C$, $\epsilon_2^e = 0.5438E_C$, $\epsilon_3^e = 0.5248E_C$, and $\epsilon_{s>3}^e = (0.3133 + \frac{\gamma}{(s+x)^{\frac{1}{2}}})E_C$, where the numbers were taken from finite size calculations by Wójs and Quinn[1] and the expression for *s*>3 is an extrapolation which gives the correct limiting value for infinite *s,* while $\gamma = 0.5320$ and $x = 3.327$ are chosen to fit the values for *s* = 2 and 3. The Coulomb energy E_C is defined as $\frac{e^2}{\varepsilon l_B}$, where ε is the background dielectric constant and l_b is the magnetic length. Here, however, we treat E_C as an adjustable fitting parameter, for which we obtain values corresponding to choices of ε in the range 10.0 to 12.4.

If we assume that energy, charge, and S_Z are conserved, then maximizing the entropy leads to Boltzmann distributions for both species of spin-*s* skyrmion

$$n_s^e = e^{-(E_s^e - s\mu_m - \mu)/T}$$
$$n_s^h = e^{-(E_s^h - s\mu_m + \mu)/T},$$

where n_s^e and n_s^h denote, respectively, the densities of electron-like and hole-like skyrmions with s overturned spins in units of inverse flux quanta, respectively, and we have introduced the electron chemical potential μ and the chemical potential associated with flipped spins—i.e. the magnon chemical potential—μ_m. The total electron and hole densities n_e and n_h are then given by

$$n_e = \sum_s n_s^e, n_h = \sum_s n_s^h,$$

and the total charge density, or equivalently the filling factor ν, is

$$\nu = 1 + n_e - n_h.$$

These formulae show that, for fixed total density, both the population of charge carriers with total spin s and the electron chemical potential—and thus the gap to charged excitations—depend on the temperature and the magnon chemical potential.

The above formulation determines the filling factor ν as a function of μ, μ_m, and T under the assumption that the charged excitations do not interact. However, the experimental $\mu_e(n)$ curves exhibit strong negative compressibility near ν=1, indicating that substantial correlation effects are present. At T=0, for sufficiently low carrier densities, in the absence of impurities, electrons or holes are expected to form a Wigner crystal (or skyrmion crystal if the carriers are skyrmions), whose energy per carrier has been calculated to be[2]

$$E_{WC}(\nu) = E_C(-0.782133|\nu - 1|^{\frac{1}{2}} + 0.2410|\nu - 1|^{\frac{3}{2}} + 0.16|\nu - 1|^{\frac{5}{2}})$$

This would give a contribution to the chemical potential at T=0 of

$$\delta\mu_{WC}(\nu) = \frac{d(|\nu - 1|E_{WC}(\nu))}{d\nu} \approx \frac{3}{2}E_{WC}(\nu)\mathrm{sign}(\nu - 1)$$

Although a Wigner crystal is expected to melt at a temperature that is much smaller than E_{WC}, it is expected that the energy of the correlated liquid is not much different from that of the Wigner crystal. Thus, we assume that the contribution of the correlation energy to the chemical potential has the phenomenological form

$$a_{e,h}\frac{3}{2}E_{WC}(\nu)\mathrm{sign}(\nu - 1)$$

where $a_{e,h}$ are parameters we fit to experiment, which we allow to be different in order to reflect the observed asymmetries between electrons and holes in our system. We then calculate the densities of skyrmions at finite temperatures using

$$n_s^e = e^{-(E_s^e - s\mu_m - \mu + a_e\delta\mu_{WC}(\nu))/T}$$

$$n_s^h = e^{-(E_s^h - s\mu_m + \mu + a_h\delta\mu_{WC}(\nu))/T}.$$

With these distributions, the equation $\nu = 1 + \sum_s(n_s^e - n_s^h)$ again constitutes a relation $\mu(\nu)$ for given values of E_C, μ_m, T and $a_{e,h}$, which allows us to extract the thermodynamic parameters as follows. At zero DC bias, there is no magnon pumping and we take $\mu_m = 0$ and T to be the cryostat base temperature at 11 T, $T_{\mathrm{base}} \approx 0.8$ K. Thus, the zero-bias chemical potential $\mu(\nu, V_{\mathrm{DC}} = 0)$ can be fit to obtain estimates of E_C, $a_{e,h}$, and a phenomenological Gaussian density broadening parameter Δν. Extended Data Fig. 2 shows examples of the zero-bias fit results, together with the fit parameters, which are in excellent agreement with the experimental data. These fit parameters are then carried over to compute $\mu(\nu)$ and calculate the ν=1 gap as a function of temperature and magnon chemical potential (Fig. 3a and Extended Data Fig. 6a). An independent estimate of the electron temperature from transport thus allows us to relate the measured gap value under magnon pumping to the corresponding μ_m. Examples of the resulting estimates of μ_m are shown in Figs. 3e and Extended Data Fig. 6b, where the error bar is estimated by allowing a 3% difference between the measured and calculated gap together with the propagated error from the temperature estimates.

Knowledge of the magnon chemical potential at each value of DC bias $\mu_m(V_{\rm DC})$, along with the estimate of the electron temperature, allows us to estimate the remaining thermodynamic quantities of interest. The average number of excess flipped spins per skyrmion $<s>(V_{\rm DC})$ may be calculated straightforwardly from the distributions via

$$< s >= \sum_s s(n_s^e + n_s^h) /(n_e + n_h).$$

The total density of free magnons n_m is given by

$$n_m = (N_B)^{-1} \sum_{|\mathbf{k}|<k_{\max}} n_{\mathbf{k}}^m,$$

with $n_{\boldsymbol{k}}^m$ being the density of magnons at wavevector $\mathbf{k}$ given by the Bose-Einstein distribution

$$n_{\mathbf{k}}^m = \frac{1}{e^{(\epsilon_{\mathbf{k}}^m + E_Z - \mu_m)/T} - 1}.$$

where $\epsilon_{\mathbf{k}}^m = 1.2533 E_{\rm ex}(1 - e^{-\frac{k^2 {l_B}^2}{4}} I_0\left(\frac{k^2 {l_B}^2}{4}\right))$ and I_0 is a modified Bessel function of the first kind, is the energy of a free magnetoexciton, without the Zeeman contribution[3,4]. The cutoff $k_{\max}$ is determined self-consistently by iterating the condition $k_{\max}^2 l_B^2 = \frac{4\epsilon_0^e}{\epsilon_m(k_{\max})}$, where

$$\epsilon_m = \sum_{|\mathbf{k}|<k_{\max}} n_{\mathbf{k}}^m \, (\epsilon_{\mathbf{k}}^m + E_z)$$

is the energy density of the free magnons, until convergence is achieved within 0.01%. The reason for our cutoff choice is as follows. For $k l_B \gg 1$, a magnetoexciton consists of an electron and a hole separated by a large distance, $d = k {l_B}^2$, and its energy will be equal to $2\epsilon_0^e$. In thermal equilibrium at low temperatures, the total number of magnons per flux quantum with $k l_B \gg 1$ will be given by $n_m^> \sim \frac{1}{2} k_{\max}^2 l_B^2 e^{(-2\epsilon_0^e)/T}$, and the associated energy will be $\epsilon_m \sim 2 n_m^> \epsilon_0^e$. On the other hand, if μ is at the center of the energy gap, we should have $n_e = n_h = e^{(-\epsilon_0^e)/T}$. Equating these quantities, we obtain the result for $k_{\max}$ stated above. This relation is consistent with the observation that for a system of linear size L, with just a single magnon present, the requirement $d < rL$, with r a constant of order unity, gives $k_{\max}$ of order $L/{l_B}^2$.

2. Magnon specific heat

The unusual "saturating" behavior of the electron temperature with increasing $V_{\rm DC}$ may in part be explained by the sudden increase of the magnon specific heat which results in the increased magnon chemical potential. Within an ideal Bose gas model, for $T \ll E_C$ the specific heat of the magnon system per unit area is approximately given by

$$C_m/k_B \approx \frac{\sqrt{2}}{\pi^{\frac{3}{2}} E_C l_B^2} (2 k_B T \mathrm{Li}_2(z) + \mu_m \ln(1 - z)),$$

where Li is the polylogarithm function and z is the fugacity $z = e^{\mu_m/T}$ with μ_m being measured from the magnon band edge. On the other hand, at low temperatures, the contribution of the electronic (charged) degrees of freedom to the specific heat is given approximately by

$$\frac{C_e}{k_B} \approx \frac{\pi^2}{3} k_B T \times D(E_F)$$

where $D(E_F)$ is the density of states at the Fermi level. When the electron chemical potential lies deep within the ν=1 gap, the factor $D(E_F)$ becomes very small, and a result the contribution to the specific heat associated with charged excitations is expected to be suppressed as $e^{-\Delta/2k_BT}$.[5] In fact, our measurements of dμ/dn allow us to estimate $D(E_F)$ directly. Taking typical values from e.g. Fig. 1d of a step in chemical potential $\Delta\mu = 20$ meV over a density span of approximately $\delta\nu = 0.02$ (in units of Landau level filling factor), we obtain an estimate for the residual C_e of

$$\frac{C_e}{k_B} \approx \frac{\pi^2}{3} \frac{eB\delta\nu}{h\Delta\mu} k_B T \approx (8.7 \times 10^{12}\ \text{meV}^{-1}) k_B T$$

Fig. S1 depicts the ratio C_m/C_e obtained from the formulas above with $\delta\nu \sim 0.02$ and E_C taken as the phenomenological value of ~20 meV for a range of values of μ_m and T near those realized in the experiment.

We find that, while the magnonic contribution to the specific heat is negligible at base temperature and in the absence of a bias ($\mu_m = 0$), it grows dramatically once magnons are injected into the system, exceeding c_e by as much as a factor of 15 for the largest values of V_{DC}. This is in accordance with the expectation that the electronic contribution is strongly suppressed at integer filling: once magnons are introduced into the system, their contribution to the specific heat immediately dominates that of the electrons. We additionally note that neutral valley excitations, which are not taken into account in the analysis above, will also contribute to the specific heat, resulting in an even larger enhancement as T is raised. To summarize, we conclude that the unusual temperature saturation may be due to the dramatic enhancement of the magnon specific heat which results from the increase of the magnon chemical potential.

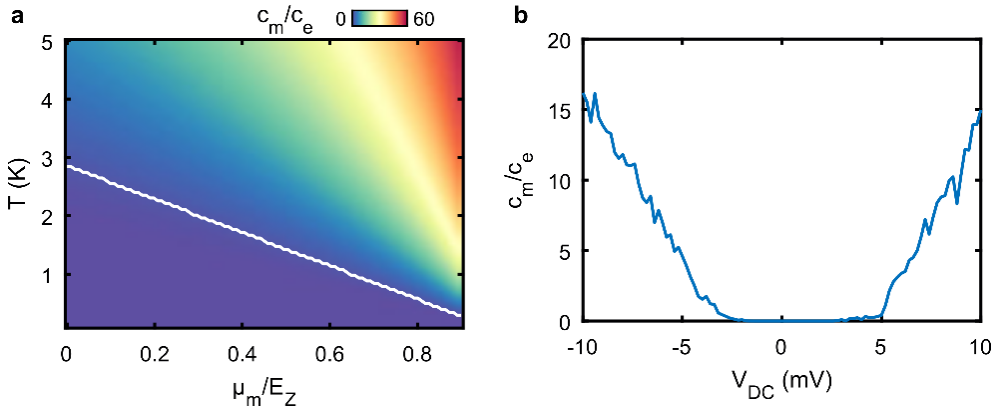

Fig. S1 | Comparison of electronic and magnonic specific heats. a, Plot of the ratio of the specific heats c_m/c_e defined in the formulas above for a range of values of the magnon chemical potential μ_m and temperature T near those observed in the experiment. The white line indicates the contour $c_m=c_e$. **b**, Plot of the ratio c_m/c_e as a function of DC bias V_{DC} obtained by substituting the values of μ_m and T obtained at each DC bias.

3. Phonon contribution to R_{xx}

Below we present three arguments that phonons do not play an important role in our transport measurements.

First, local thermometry measurements on graphene/hBN devices performed under similar conditions reveal negligible local heating of the phonon temperature[6]: specifically, for a ~5 nW bias applied near ν=2, sub-mK temperature gradients were observed near the edge channel, which are expected to be even smaller in the absence of tip gating. These observations strongly suggest that the phonon temperature remains at the base temperature of the cryostat under an applied bias, and that, as in the case of zero magnetic field, the energy exchanged between the electron and phonon systems is negligible.

Second, while a complete theoretical understanding of the mechanisms controlling R_{xx} is lacking in the field despite four decades of work, an analysis of the electron-phonon coupling in a strong magnetic field within a Debye model finds that the phonon correction to the transverse conductivity σ_{xy} in the lowest Landau level of a parabolic band to be at most[7]

$$\frac{\sigma_{xy}^{\mathrm{e-ph}}}{\sigma_{xy}^{0}} \approx 10^{-2} \times \frac{\omega_c^2}{\omega_D T},$$

where ω_D is the Debye frequency, ω_c is the cyclotron energy and T is the electron temperature. This upper bound on the phonon contribution to σ_{xy}, valid for $\omega_c \gg T$ and $q_D l_B \gg 1$, where q_D is the Debye momentum, is saturated when ν is tuned precisely to the middle of the lowest Landau level and therefore constitutes an overestimate at other fillings. First of all and most importantly, we note that the $1/T$ dependence of this correction implies that relaxation to phonons becomes less important to σ_{xy} as the electron temperature is increased, which we argue below also implies that the phonon correction to R_{xx} becomes less important at higher electron temperatures. Assuming that this relation holds for Dirac fermions in the symmetry-broken lowest Landau level of graphene, we take ω_c to be ~20 meV (the measured ν=1 gap), T to be the ~3 K at which the temperature saturated, and the ω_D of graphene to be 181 meV (corresponding to a very large Debye temperature of 2100 K),[8] which results in an upper bound of $\sigma_{xy}^{\mathrm{e-ph}}/\sigma_{xy}^{0} \approx 9\%$ on the phonon correction under the conditions of our measurements. While this correction to σ_{xy} does not rigorously constrain the measured values of R_{xx}, we note that an empirical law relating R_{xx} and the derivative of R_{xy} has been observed in a variety of experiments and analyzed,[9,10] namely

$$R_{xx} \propto B \frac{dR_{xy}}{dB},$$

which has been found to hold remarkably well under a range of experimental conditions. This empirical law therefore suggests that the estimated ~9% bound on the phonon correction to σ_{xy} in turn corresponds to a ~9% upper bound on the phonon correction to R_{xx}. A 9% uncertainty in R_{xx} is not nearly enough to change any of the conclusions of our manuscript, as changing R_{xx} by 9% would still imply a significant nonzero magnon chemical potential. Therefore, this argument shows first of all that the sudden increase in R_{xx} observed at elevated electron temperatures in our measurement cannot be explained simply by an increase in the rate of phonon relaxation, because the latter should decrease as the temperature increases within the range of interest, and

second of all that the magnitude of the phonon contribution under the conditions of our measurement is too small to significantly change our estimates of the electron temperature. We conclude that phonons are very unlikely to play an important role in our thermometry measurements.

Finally, we note that if R_{xx} were controlled by phonons to a significant degree, it is not clear why the measured electron temperature would exhibit the unusual "two-threshold" behavior shown in Fig. 3d. By contrast, this behavior is naturally explained in terms of the enhancement of the specific heat due to the appearance of a population of free magnons, as explained above.

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