Electronic Cooling in Graphene

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Energy transfer to acoustic phonons is the dominant low-temperature cooling channel of electrons in a crystal. For cold neutral graphene we find that the weak cooling power of its acoustic modes relative to their heat capacity leads to a power-law decay of the electronic temperature when far from equilibrium. For heavily doped graphene a high electronic temperature is shown to initially decrease linearly with time at a rate proportional to $n^{3/2}$ with n being the electronic density. The temperature at which cooling via optical phonon emission begins to dominate depends on graphene carrier density.

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Introduction.—Energy exchange between the electrons in nanoscale electronic devices and their environment is a key issue in the design of electronic circuits and will play a role in any future graphene-based electronics [1,2]. The dominant electronic cooling mechanism in nearly any solid state environment is energy transfer to phonons. Energy relaxation in a graphene sheet is dominated by transfer to the acoustic and optical phonon modes of its two-dimensional honeycomb lattice and to the optical phonon modes of its substrate [3,4]. In this Letter we address electronic energy relaxation in graphene with a focus on the intrinsic cooling channel provided by the acoustic phonons.

Optical measurements are a particularly useful probe of energy transfer between electrons and phonons and have been employed in the past in studies of electronic cooling in quantum wells [5]. Similar measurements were recently performed on epitaxial graphene samples [6,7]. In a typical measurement electrons are excited to high energies using an optical pulse. The relaxation process of the hot electrons is then monitored using differential transmission spectroscopy. Although transport measurements do in principle provide an alternate way of studying interactions between electrons and phonons [3], the resistivity contribution from acoustic phonons in typical graphene samples is much smaller than the elastic-scattering residual resistivity contribution [8]. Transport is therefore relatively insensitive to the electron-phonon coupling strength. Even for suspended graphene samples in which transport is nearly ballistic, quantum resistance dominates over the phonon induced resistance [9]. On the contrary, cooling of hot Dirac quasiparticles is entirely due to phonons.

Guided by experiment [6,7] we assume that *e-e* interactions thermalize the system throughout the relaxation process. Given the lattice temperature T_L and the electronic density n, the cooling process is then characterized by a single time dependent function, the electronic temperature $T_e(t)$. The time dependent chemical potential μ depends on T_e and n and is determined by the conservation of the number of particles. As the hot electrons equilibrate, the electronic temperature decreases, approaching its equilibrium value T_L.

A unique situation arises for graphene in the *neutral regime* $T_e \gg \mu$. In a typical semiconductor this nondegenerate regime is reached only at high temperatures. In nearly neutral graphene, on the other hand, this regime is accessed at nearly all temperatures of interest. We find that when $T_L \ll T_e \lesssim 180$ K the electronic temperature satisfies a power-law decay law,

$$T_e(t) = \frac{T_0}{\sqrt{t/\tau_0 + 1}}, \tag{1}$$

with a characteristic time

$$\tau_0 = \frac{424}{D^2 T_0^2}\ \mu\text{s}. \tag{2}$$

Here T_0 is the initial temperature of the electrons and D is the screened deformation potential measured in eV [10]. Hereafter we use $\hbar = 1$ and measure all temperatures in meV. Transport measurements have been able to bound the value of D between 10 and 50 eV; however, more precise limits on this important parameter have remained elusive [9]. We propose that Eq. (1) be used to obtain an accurate value of D from experiment.

This slow low-T_e cooling of neutral-regime Dirac quasiparticles differs markedly from the very fast cooling observed when T_e is high. Recent experimental [6,7,11] and theoretical [12] work has demonstrated that very hot electron plasmas ($T_e \sim$ several hundred meV) cool significantly after several picoseconds. We find that low temperature cooling is slower by more than 3 orders of magnitude.

Theory of temperature dynamics.—The assumption of rapid thermalization implies that

$$\partial_t T_e = \mathcal{Q}/\mathcal{C}, \tag{3}$$

where $\mathcal{C} = \partial_{T_e}\mathcal{E}$ is the electronic heat capacity, $\mathcal{Q} = \partial_t \mathcal{E}$ is the electronic cooling power [13], and $\mathcal{E}$ is the energy density of the system. Using the Boltzmann equation we find that

$$\mathcal{Q} = \partial_t \sum_{\mathbf{k}\alpha} \epsilon_{k\alpha} f^{\alpha}_{\mathbf{k}} = \sum_{\mathbf{k}\alpha} \epsilon_{k\alpha} S_{\rm ph}(f^{\alpha}_{\mathbf{k}}), \tag{4}$$

where

$$S_{\rm ph}(f_{\mathbf{k}\alpha}) = -\sum_{\mathbf{p}\beta}[f^{\alpha}_{\mathbf{k}}(1-f^{\beta}_{\mathbf{p}})W_{\mathbf{k}\alpha\to\mathbf{p}\beta} - \{\mathbf{k}\alpha \to \mathbf{p}\beta\}] \tag{5}$$

is the collision integral. Here $\alpha = v, c$ labels the valence and conduction bands whose energies are $\pm v_g k$ with v_g being graphene's band velocity. The occupation of each band is given by the time dependent Fermi distribution function $f^{\alpha}_{\mathbf{k}} = f(\epsilon_{\mathbf{k}\alpha}, T_e(t), \mu(t))$ and

$$W_{\mathbf{k}\alpha\to\mathbf{p}\beta} = 2\pi \sum_{\mathbf{q}} w^{\alpha\beta}_{q}[(N_q + 1)\delta_{\mathbf{k},\mathbf{p}+\mathbf{q}}\delta(\epsilon^{\alpha\beta}_{\mathbf{kp}} - \omega_q) + N_q \delta_{\mathbf{k},\mathbf{p}-\mathbf{q}}\delta(\epsilon^{\alpha\beta}_{\mathbf{kp}} + \omega_q)] \tag{6}$$

is the transition rate between state $\mathbf{k}\alpha$ and state $\mathbf{p}\beta$. The energy exchanged with the phonon heat bath in the transition is $\epsilon^{\alpha\beta}_{\mathbf{kp}} = \epsilon_{\mathbf{k}\alpha} - \epsilon_{\mathbf{p}\beta}$. In Eq. (6) $N_q = N(\omega_q)$ is the Bose distribution function evaluated at the phonon energy ω_q. For acoustic phonons the transition matrix element is $w^{\alpha\beta}_{q} = D^2 q^2 (1 + s_{\alpha\beta}\cos\theta)/4\rho\omega_q$. Here $s_{\alpha\beta} = 1$ for intraband transitions and $s_{\alpha\beta} = -1$ for the interband ones, $\theta = \theta_k - \theta_p$ is the relative angle between the incoming and outgoing momenta, and ρ is the mass density of graphene. For optical phonons with energy ω_0, $w^{\alpha\beta}_{q} \approx g^2$ where $g \approx 2v_g/a^2\sqrt{2\rho A \omega_0}$ with $a = 1.42$ Å and A being the area of the graphene sheet [14].

We first consider $\mathcal{Q}_a$ the energy transfer to the acoustic phonon bath which is accurately described by the linear energy dispersion $\omega_q = cq$. It is instructive to separate the energy transfer into a loss due to spontaneous emission, $\mathcal{Q}^{\rm spont}$, and a gain due to induced transitions, $\mathcal{Q}^{\rm ind}$. Straightforward manipulations of (4) lead to

$$\mathcal{Q}^{\rm ind} = -\frac{\pi D^2}{2\rho c}\sum_{\mathbf{k}\alpha\mathbf{p}\beta} \epsilon^{\alpha\beta}_{\mathbf{kp}}(1 + s_{\alpha\beta}\cos\theta)(f^{\alpha}_{k} - f^{\beta}_{p}) \times \sum_{\mathbf{q}} q N_q \delta_{\mathbf{k},\mathbf{p}+\mathbf{q}}\delta(\epsilon^{\alpha\beta}_{\mathbf{kp}} - \omega_q). \tag{7}$$

To evaluate $\mathcal{Q}_a$ we exploit the large mismatch between v_g and the sound velocity c and evaluate $\mathcal{Q}_a$ to leading order in $c/v_g \ll 1$. In the limit $c/v_g \to 0$ the scattering is elastic, only intraband transitions are allowed, and there is no energy loss. To leading order in c/v_g interband scattering remains negligible and we can approximate $|\mathbf{p}|$ by $|\mathbf{k}|$ when performing the sum over $\mathbf{q}$ in Eq. (7). In this way we find that to $O(c/v_g)^4$

$$\mathcal{Q}^{\rm ind} = \frac{D^2 T_L}{\rho v_g^2}\int \frac{k^3 dk}{2\pi}[f^{c}_{k} + (1 - f^{v}_{k})]. \tag{8}$$

Similar steps yield $Q^{\rm spont}$ and the total energy loss

$$\mathcal{Q}_a = -\frac{D^2}{\rho v_g^2}(T_e - T_L)\int \frac{k^3 dk}{2\pi}[f^{c}_{k} + (1 - f^{v}_{k})]. \tag{9}$$

As expected, the net energy loss vanishes when the electronic temperature reaches the lattice temperature. Interestingly, to leading order in c/v_g the energy loss is independent of the sound velocity. Because of the absence of interband transitions the cooling power from electrons in the conduction band and from holes in the valence band are simply additive.

As is evident from Eqs. (8) and (9), the energy gain due to the induced transitions is negligible compared to $\mathcal{Q}^{\rm spont}$ at high temperatures when $T_e \gg T_L$. However, as the system cools, the difference between $\mathcal{Q}^{\rm ind}$ and $|\mathcal{Q}^{\rm spont}|$ decreases, vanishing in equilibrium. The vanishing of $\mathcal{Q}$ in equilibrium is assured by the detailed balance condition reflected by the collision integral expression, Eq. (5).

The cooling power due to the intrinsic optical modes is easily estimated when the electron-phonon coupling is approximated by a constant g and phonon dispersion is neglected. Setting the phonon energy to ω_0 it follows from Eqs. (4)–(6) that

$$\mathcal{Q}_o = \frac{g^2\omega_0^4}{(2\pi v_g^2)^2}[N_e(\omega_0) - N_L(\omega_0)]\mathcal{F}(T_e, \mu), \tag{10}$$

where

$$\mathcal{F}(T_e, \mu) = \int_{-\infty}^{\infty} dx |x(x-1)|[f([x-1]\omega_0) - f(x\omega_0)]. \tag{11}$$

Here N_e and N_L are the Bose distribution functions evaluated at the temperatures T_e and T_L, respectively, and the factor $|x(x-1)|$ originates from the electronic joint density of states. For neutral graphene $\mathcal{F} \approx 1/6$ when $T_e \ll \omega_0$. The dominant optical phonon bath in suspended graphene is likely intrinsic. In unsuspended samples scattering by substrate phonons may introduce additional optical modes which are considerably less energetic [3]. We focus below on the energy loss due to acoustic phonons and comment on the role of the intrinsic optical phonon modes only at the end of this Letter.

Neutral limit.—First we consider the neutral regime for which $\mu(t) \ll T_e(t)$. If the equilibrium value of the chemical potential is significantly smaller than the lattice temperature, the system remains in the neutral regime throughout the entire relaxation process; otherwise the system will eventually exit the neutral regime as it approaches equilibrium.

The energy exchanged in a typical transition is T_e, implying a momentum transfer of T_e/v_g. The typical phonon energy is then $T_e c/v_g$, justifying the quasielastic approximation for the scattering by acoustic phonons for *all* values of the electronic temperature. This situation is in marked contrast with the typical scenario in metals in which e-ph scattering becomes highly inelastic below the Bloch-Grüneisen temperature.

In the neutral limit Eq. (9) for $\mathcal{Q}_a$ can be further simplified by setting μ to zero in the integral to obtain a value proportional to T_e^4. Combining this result with Eq. (3) and noting that the energy density per spin and valley in neutral graphene is $\mathcal{E} = 3\zeta(3)T_e^3/2\pi v_g^2$, where ζ is the Riemann zeta function, we find that

$$\partial_t T_e = -\gamma T_e^2(T_e - T_L), \tag{12}$$

where $\gamma = 7\pi^4 D^2/540\zeta(3)\rho v_g^4 = 1.18 \times 10^3 D^2$ $(\mathrm{meV}^2\,\mathrm{s})^{-1}$ with the deformation potential measured in eV. Both the cooling power and the heat capacity decrease as T_e approaches T_L. Because the former decrease is faster, the rate of temperature decrease slows. Far from equilibrium when $T_e \gg T_L$, Eq. (12) is solved by (1). The temperature decays as a power law with a characteristic time of $\tau_0 = \frac{1}{2}\gamma T_0^2$. The simple relationship between T_e and the energy density $\mathcal{E}$ combined with Eq. (1) yields

$$\mathcal{E}(t) \approx \mathcal{E}_0(t/\tau_0 + 1)^{-3/2}, \qquad T_e \gg T_L, \tag{13}$$

where $\mathcal{E}_0$ is the initial energy density of the system. Like the electronic temperature, the energy density decays to equilibrium as a power law when the system is far from equilibrium.

Near equilibrium, when $T_e \gtrsim T_L$, we linearize Eq. (12) with respect to $T_e - T_L$ to find that the electronic temperature decays exponentially to its equilibrium value with a characteristic time given by $\tau_L = 1/\gamma T_L^2 = 848/D^2 T_L^2$ μs.

Our results for the neutral limit are valid to $O(\mu/T_e)^2$ since the particle hole symmetry of the system implies that both $\mathcal{Q}_a$ and $\mathcal{E}$ are even functions of μ/T_e.

Highly doped limit.—We now turn to study the equilibration of hot electrons in the doped regime for which $\mu(t) \gg T_e(t)$. In this regime we use the Sommerfeld expansion to approximate $\mathcal{Q}_a$ and $\mathcal{C}$ and obtain

$$\partial_t T_e = -\gamma_d \frac{T_e - T_L}{T_e}, \tag{14}$$

where $\gamma_d = 3D^2\epsilon_F^3/4\pi^2\rho v_g^4 = 0.133 D^2 n^{3/2}$ meV/ns with n being measured in units of 10^{12} cm^{-2}. Far from equilibrium the high electronic temperature initially decreases linearly with time at an energy rate given by γ_d. Near equilibrium T_e approaches T_L exponentially at a rate of γ_d/T_L.

The doped regime resembles the typical metallic case in that the quasielastic approximation breaks down at low temperatures when the electronic temperature is below the Bloch-Grüneisen temperature $T_{\mathrm{BG}} = 2ck_F$. Therefore our results for the doped regime describe the entire equilibration process for systems in which $T_L > T_{\mathrm{BG}}$. However, for cold doped graphene our results are valid only when $T_e(t) > T_{\mathrm{BG}}$.

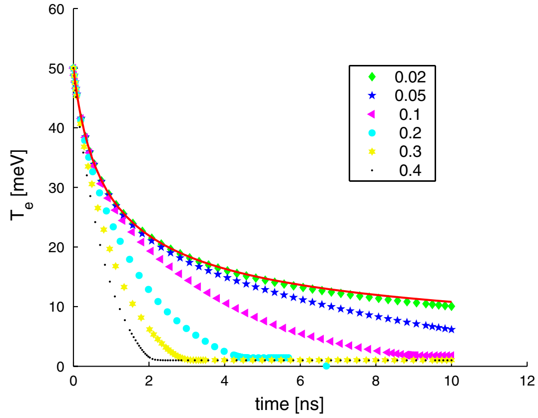

FIG. 1 (color online). Equilibration of the electronic temperature. The evolution of the electronic temperature T_e is plotted for electronic densities of (top to bottom) 0.02, 0.05, 0.1, 0.2, 0.3, and 0.4×10^{12} cm^{-2}. The lattice temperature is 1 meV and the deformation potential is assumed to be 20 eV. The solid line corresponds to the equilibration of T_e in a neutral system.

General solution.—In the general case, when $\mu(t) \sim T_e(t)$, the time evolution of the electronic temperature and chemical potential follow from two coupled differential equations. The first equation

$$0 = -2v_g\partial_t(1/T_e)I_1^{(+)} + \partial_t(\mu/T_e)I_0^{(-)} \tag{15}$$

is obtained from the number equation $\partial_t n = 0$ and expresses the conservation of the number of particles throughout the relaxation process. Here

$$I_n^{(\pm)} \equiv \int \frac{k^n dk}{2\pi}[f_k^c \mp (1 - f_k^v)]. \tag{16}$$

The second differential equation $\partial_t\mathcal{E} = \mathcal{Q}_a$ can be written using Eq. (9) as

$$-3v_g T_e \partial_t(1/T_e)I_2^{(-)} + 2T_e\partial_t(\mu/T_e)I_1^{(+)} = -\frac{D^2}{\rho v_g^2}(T_e - T_L)I_3^{(-)}. \tag{17}$$

We have solved the coupled differential equations (15) and (17) numerically for various values of densities. The evolution of T_e is plotted in Fig. 1 for $T_L = 1$ meV, $T_0 = 50$ meV, and $D = 20$ eV. Clearly the equilibration process is faster for doped systems.

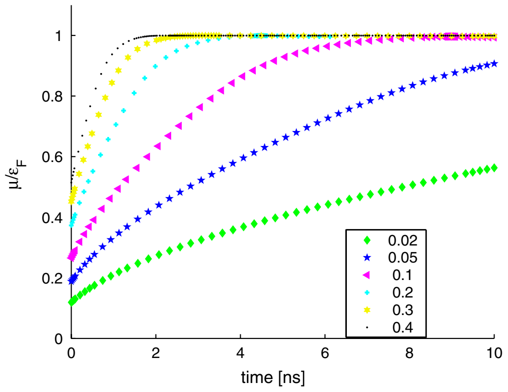

FIG. 2 (color online). Equilibration of the chemical potential. As T_e decreases (see Fig. 1), the chemical potential increases to preserve the electronic density.

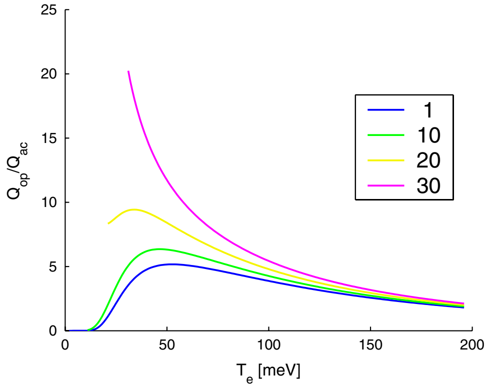

FIG. 3 (color online). The energy loss due to optical phonons divided by the energy loss do to acoustic phonons for neutral suspended graphene is plotted versus the electronic temperature for lattice temperatures of (bottom to top) 1, 10, 20, and 30 meV.

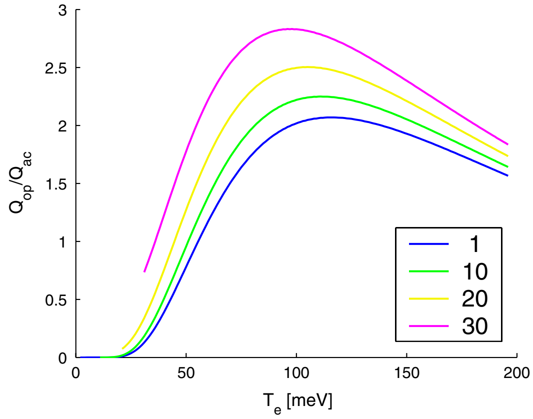

FIG. 4 (color online). The analog of Fig. 3 for a doped system with $n = 10^{13}$ cm^{-2}.

The corresponding evolution of the chemical potential for the different electronic densities is plotted in Fig. 2. The conservation of particle number throughout the equilibration process forces $|\mu|$ to increase with time.

Discussion.—The small low-temperature cooling power of the neutral graphene electronic system is due to both the small joint density of states for electronic transitions and the small energy of acoustic phonons at typical transition momenta. From dimension analysis of $\mathcal{Q}$ and $\mathcal{E}$ and Eq. (3), we find that the instantaneous energy decay rate $\alpha = |\partial_t \ln(\mathcal{E})| = \mathcal{Q}/|\mathcal{E}|$ of a d-dimensional gapless neutral system at zero lattice temperature with conduction and valence band dispersions $\epsilon_k \propto k^s$ satisfies

$$\alpha \sim T_e^{(d-s+1)/s}. \tag{18}$$

Exponential decay of T_e and $\mathcal{E}$ occurs when α is constant, i.e., when $s = d + 1$. In systems like single layer or bilayer graphene with $s < d + 1$, the cooling rate will decrease as a power law of the instantaneous temperature when the system is far from equilibrium.

It is interesting to consider the consequences of this work on the cooling of bilayer graphene. The evolution of T_e of a gapless 2D system with a parabolic dispersion is given by $\partial_t T_e = -\tilde{\gamma}\sqrt{T_e}(T_e - T_L)$. However, we do not expect this relationship to apply precisely in bilayer graphene since the momentum dependence of its energy spectrum cannot be described by a single power [15].

Although the main focus in this work has been energy loss due to acoustic phonons, we emphasize that the energetic optical phonons will play a dominant role in the cooling of graphene at sufficiently high temperatures. To estimate the regime for which acoustic phonons dominate cooling in graphene, we plot $\mathcal{Q}_o/\mathcal{Q}_a$ as a function of T_e. We use a simple model in which $\omega_0 = 196$ meV for both longitudinal and transverse optical branches. In Fig. 3 we plot $\mathcal{Q}_o/\mathcal{Q}_a$ for a neutral graphene sheet for various values of T_L. Surprisingly, the different functional dependencies of $\mathcal{Q}_o$ and $\mathcal{Q}_a$ on T_L and T_e lead to a nonmonotonic dependence of $\mathcal{Q}_o/\mathcal{Q}_a$ on T_e and to the dominance of $\mathcal{Q}_o$ near equilibrium at moderate lattice temperatures. As evident from Fig. 4, the relative cooling power of the acoustic phonons increases with doping, increasing the maximum temperature at which they are dominant.

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