

High Thermoelectric Power Factor in Graphene/hBN Devices

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ABSTRACT

Fast and controllable cooling at nanoscales requires a combination of highly efficient passive cooling and active cooling. While passive cooling in graphene-based devices is quite effective due to graphene's extraordinary heat-conduction, active cooling has not been considered feasible due to graphene's low thermoelectric power factor. Here we show that the thermoelectric performance of graphene can be significantly improved by using hBN substrates instead of SiO₂. We find the room temperature efficiency of active cooling, as gauged by the power factor times temperature, reaches values as high as 10.35 Wm⁻¹K⁻¹, corresponding to more than doubling the highest reported room temperature bulk power factors, 5 Wm⁻¹K⁻¹ in YbAl₃, and quadrupling the best 2D power factor, 2.5 Wm⁻¹K⁻¹, in MoS₂. We further show that in these devices the electron-hole puddles region is significantly reduced. This enables fast gate-controlled switching of the Seebeck coefficient polarity for applications in n- and p-type integrated active cooling devices.

KEYWORDS: graphene, Seebeck coefficient, thermoelectric power factor, screened Coulomb scattering, electron-hole puddles

As the size of the electronic components shrinks, larger power densities are generated, resulting in local hot spots. The small size of these hot spots and their inaccessibility make it difficult to maintain a low and safe operating temperature.¹ Solid-state integrated active thermoelectric coolers could solve the long lasting electronic cooling problem.^{2,3} In the normal refrigeration mode of thermoelectric coolers, heat is pumped from the cold side to the hot side. However, there is an increasing need to pump heat from the hot spots generated on the chip to the colder ambient reservoir. In this mode of operation, both passive and active cooling can be used.⁴ In the case of passive cooling, where heat is transported via the phonon channel, the performance is fixed by the thermal conductance. In contrast, active cooling which uses the Peltier effect to pump heat via the electronic channel can be controlled and tuned with applied current. The performance of Peltier cooling is a function of the thermoelectric power factor, $PF = \sigma S^2$, where σ is the electrical conductivity and S is the Seebeck coefficient. In this manuscript, we also use the notation of PFT , referring to PF times temperature T which has a more convenient unit of $\text{Wm}^{-1}\text{K}^{-1}$ (same as thermal conductivity). Although there is no theoretical limit on PF , the interplay between the Seebeck coefficient and the electrical conductivity in highly doped bulk semiconductors, has so far prevented the realization of very large thermoelectric power factors.⁵⁻⁷

Single-layer graphene possesses extraordinary electronic and thermal properties.⁸⁻¹⁰ In particular its higher mobility, which due to the weak electron-phonon interaction persists up to room temperature, can be orders of magnitude higher than in other 2D thermoelectric materials, such as semiconducting transition metal dichalcogenides (TMDs).¹¹⁻¹⁴ Theoretical and experimental studies show that the Seebeck coefficient in graphene could reach values comparable to that in bulk semiconductors by decreasing the carrier density.¹⁵⁻²¹ Both the Seebeck coefficient and the mobility play an important role in active cooling. At the same time graphene's extremely large thermal conductivity also enables efficient passive cooling.²² Furthermore, the ability to control its carrier density by electrostatic gating rather than by chemical doping imparts graphene an important advantage over bulk materials.

As a purely 2D material, the electronic properties of graphene are severely affected by its surroundings. Experiments demonstrate that the most commonly used SiO₂ substrate has many surface charged states and impurities which cause strong Coulomb scattering that limits the mobility and introduces large potential fluctuations in G/SiO₂ samples.²³⁻²⁵ The potential fluctuations induce electron-hole puddles (EHP) in the vicinity of the charge-neutrality point (CNP) and prevent gating for lower carrier density.²⁴ Depositing graphene on hBN substrates, which are relatively inert and free of surface charge traps, produces G/hBN samples with smaller potential fluctuations and higher mobility than G/SiO₂.²⁶⁻²⁸ Here we report on measurements of the thermoelectric properties, S and PF , for G/hBN and G/SiO₂ samples.

Fig. 1a shows a schematic of the apparatus, which allows measuring both electrical and thermal transport properties of the material (see Supporting Information). Fig 1b shows the Seebeck coefficient measured in G/hBN and G/SiO₂ samples. In G/hBN sample, the peak value $S = 182\mu\text{V/K}$ at 290K is significantly higher than the corresponding value $S = 109\mu\text{V/K}$ in the G/SiO₂ sample, and the gate voltage at the peak position, $V_p = -2.2\text{V}$, is closer to the CNP than that in the G/SiO₂ sample, $V_p = -4.5\text{V}$. From the measured values of S and the conductivity we calculate the value of $PFT = S^2 \sigma T$ as a function of carrier density shown in Fig. 1c.²⁹ The PFT first increases with decreasing carrier density when far from CNP, then after reaching a peak value, it drops to zero at the CNP. We find that the room temperature peak value of PFT in G/hBN, $10.35 \text{ Wm}^{-1}\text{K}^{-1}$, is almost twice that in G/SiO₂, $6.16 \text{ Wm}^{-1}\text{K}^{-1}$. This value is larger than the record value in bulk materials at room temperature reported for YbAl₃ ($\sim 5 \text{ Wm}^{-1}\text{K}^{-1}$), and larger than the value at room temperature in 2D materials reported for MoS₂ ($\sim 2.5 \text{ Wm}^{-1}\text{K}^{-1}$) and WSe₂ ($\sim 1.2 \text{ Wm}^{-1}\text{K}^{-1}$).²⁹⁻³² We note that this value of the PFT is in fact underestimated since, due to the two-probe measurement of the conductivity, the contact resistance is included in the conductivity calculation. As we discuss later the PFT value increases with temperature and has not yet saturated at room temperature. Therefore, even larger PFT values are expected at higher temperatures.

We next use the linear Boltzmann equation in the relaxation time approximation to relate the Seebeck coefficient to the experimentally controlled quantities. Within this model the response of the electrical and thermal current densities, j and j_q , to the electric field, E , and temperature gradient, ∇T , are given by:¹⁵

$$j = L^{11}E + L^{12}(-\nabla T) \quad (1)$$

$$j_q = L^{21}E + L^{22}(-\nabla T) \quad (2)$$

where $L^{11} = K^{(0)}$, $L^{12} = -\frac{1}{eT}K^{(1)}$, $L^{21} = -\frac{1}{e}K^{(1)}$, $L^{22} = \frac{K^{(2)}}{e^2T}$, and

$$K^{(m)} = \int_{-\infty}^{+\infty} d\epsilon (\epsilon - \mu)^m \left(-\frac{\partial f^0(\epsilon)}{\partial \epsilon} \right) \sigma(\epsilon) \quad m = 0, 1, 2 \quad (3).$$

Here, $\epsilon(k) = \hbar v_F k$, v_F is the Fermi velocity, μ is the chemical potential, $f^0(\epsilon)$ is the equilibrium Fermi-Dirac distribution function. The differential conductivity is $\sigma(\epsilon) = e^2 v_F^2 \frac{D(\epsilon)\tau(\epsilon)}{2}$, $D(\epsilon) = 2|\epsilon|/(\pi\hbar^2 v_F^2)$ is the density of states including the 4-fold degeneracy of graphene, $v_F = 10^6 \text{ms}^{-1}$ is the Fermi velocity, and $\tau(\epsilon)$ is the relaxation time.³³ The Seebeck coefficient is defined as $S = L^{12}/L^{11}$, the electrical and thermal conductivity are $\sigma = L^{11}$ and $\kappa = L^{22}$ respectively, and the Peltier coefficient is $\Pi = L^{21}/L^{11}$.¹⁵
³⁴ Importantly, we note that the Seebeck coefficient is controlled by the energy dependence of the conductivity.

In Fig. 1d we show the calculated carrier density dependence of the Seebeck coefficient at 300K in the presence of random potential fluctuations induced by a distribution of charge impurities. The calculation follows the model proposed in Ref. 15 and, for simplicity, considers only the screened Coulomb scattering which is known to be the dominant scattering mechanism in this system (see Supporting Information).^{13, 15}
³⁵⁻³⁷ We note that the monotonic increase of S with decreasing carrier density peaks at the point where the Fermi energy enters the EHP region.^{15, 16} In this region (shadow area in Fig. 1d) both electrons and holes are present, but since they contribute oppositely to S , the value of S drops. Consequently, the smaller the EHP region, the higher the peak value of S . There is, however, a limit to the magnitude of S that is set by

the temperature. When $k_B T$ is comparable to the potential fluctuations energy scale, the peak value of S is controlled by the temperature. The effect of inserting the hBN spacer, typically $d \sim 10\text{nm}$, is to increase the distance from the charge impurities in the SiO_2 substrate which reduces the magnitude of the random potential fluctuations in the graphene plane. This reduces the EHP region and, as a consequence, results in a larger value of S (see Supporting Information). Again, there is a limit to this improvement. In the limit of infinitely large separation, *i.e.* no Coulomb scattering, thermally excited phonons become the dominant mechanism which limits the value of S . In the acoustic phonon-dominated regime, the Seebeck coefficient at room temperature is expected to be smaller than $S = 100\mu\text{V/K}$.¹⁵

As discussed above, the peak position of S marks the boundary of the EHP region, which depends on both the temperature and the extent of the random potential fluctuations. In the high temperature limit this region is dominated by thermal excitations, while at low temperatures it is controlled by the energy scale of the random potential fluctuations. Currently, most measurements of the EHP are carried out by scanning probe microscopy, which are typically performed at low temperatures and over a scanning range much smaller than normal transport devices.^{27, 28, 38} Although the size of the EHP region can be estimated from the gate dependence of the resistivity, the peak position of S provides a more direct measure of the EHP region.²⁶ In Fig. 2a, showing the back-gate dependence of S in the temperature range from 77K to 290K, we note that as temperature decreases so does the peak value of S and its position, V_P , moves closer to the CNP. In the following discussion, we focus on the hole side since the peaks on this side are clearer in the G/ SiO_2 sample. The temperature dependence of V_P , shown in Fig. 2b for both samples, follows an exponential function $V_P(T) = a + b(e^{\alpha T} - 1)$ where a , b and α are fitting parameters. The intercept a at $T=0$ is 0.12V and 0.52V corresponding to density fluctuations of $1.8 \times 10^{10}\text{cm}^{-2}$ and $7.6 \times 10^{10}\text{cm}^{-2}$ for the G/hBN and G/ SiO_2 samples respectively. Both values are comparable to previous results measured by scanning tunneling microscopy at liquid-helium temperature.²⁸ The corresponding energy scale of the random potential fluctuations in the two samples is 21.8meV and 45.4meV, respectively. Seebeck coefficient peak positions extracted from previous studies are also shown.

Unlike the case of the voltage drop in electrical transport, which is insensitive to the sign of the carrier charge, the Seebeck voltage reverses its sign when switching from hole-doping to electron-doping. In the G/hBN sample the polarity of the peak Seebeck coefficient could be reversed with a relatively small gate voltage $\sim 2V_p$. We define the slope of this polarity-switching effect as $\beta = S_p/V_p$, where S_p stands for the peak Seebeck coefficient. In Fig. 2c, β in G/hBN and G/SiO₂ samples at different temperatures are shown together with values extracted from previous studies in G/SiO₂ samples. Clearly, the value of β is strongly enhanced in G/hBN sample.

The ambipolar nature of graphene, which allows smooth gating between electron and hole doped sectors, together with the large values of β which facilitate switching the polarity of S , extend a distinct advantage in applications where p-type and n-type devices are integrated. This can be seen in the proposed thermoelectric active cooler design shown in Fig. 2d, which can pump heat from the hot end (T_H) to the cold end (T_L) in a controlled and fast manner using combined active and passive cooling. In this G/hBN based device, the p-n legs are arranged thermally in parallel and electrically in series to maximize the active cooling.⁴ Its structure, which is readily realized with lithographically patterned gates is significantly simpler than that of bulk devices that require different materials or different doping for the p and n legs. At the optimal value of applied current, the active cooling power is $P_{active} = PFT_H \cdot T_H/2$.⁴ On the other hand, the passive cooling power is $P_{passive} = \kappa\Delta T$ where $\kappa \sim 600 \text{ Wm}^{-1}\text{K}^{-1}$ is the thermal conductivity of graphene supported on a substrate at room temperature.²² For $T_H = 330\text{K}$ and $\Delta T = 30\text{K}$, active cooling contributes an additional 10% over the passive cooling. At higher temperatures, as PFT increases and thermal conductivity decreases, the contribution of active cooling increases further.

The temperature dependence of the Seebeck coefficient at a fixed back gate voltage for both samples is shown in Fig. 3a. The corresponding carrier density in G/hBN and G/SiO₂ is $2.0 \times 10^{12} \text{ cm}^{-2}$ and $3.0 \times 10^{12} \text{ cm}^{-2}$, respectively. Peak Seebeck coefficient values extracted from previous studies are also presented and show similar values as in our G/SiO₂ sample. The Seebeck coefficient values measured in both devices show a nonlinear temperature dependence, which is contrary to the linear dependence predicted by Mott's

formula $S = -\frac{\pi}{3e} k_B^2 T \frac{1}{\sigma} \frac{\partial \sigma}{\partial \epsilon}$.³⁹ This deviation may not be surprising in the low carrier density regime, where Mott's formula does not hold, but nonlinear behavior is observed even at higher density (see Supporting Information). The deviation from linearity indicates that the scattering processes in this system are not captured by a constant relaxation-time approximation. Indeed in the case of screened Coulomb scattering, the temperature dependence of the Seebeck coefficient is quadratic rather than linear.¹⁵ Using this model, we calculate the temperature dependence of the Seebeck coefficient shown in Fig. 3a. The temperature dependence of the measured and calculated *PFT* is also shown in Fig. 3b together with a comparison with values extracted from previous studies. The calculation overlaps with experimental results quite well. At high temperatures and high carrier density, the violation of Mott's formula in graphene was recently attributed to inelastic electron-optical phonon scattering.⁴⁰ For the carrier density range of the measurements reported here, the agreement between the experimental results and the screened Coulomb scattering model suggests that electron-phonon interactions can be neglected.

In summary, the conductivity and Seebeck coefficient are measured in G/hBN and G/SiO₂ samples in the temperature range from 77K to 290K. At room temperature, the peak Seebeck coefficient in G/hBN reaches twice the value measured in G/SiO₂ and the peak *PFT* value reaches 10.35 Wm⁻¹K⁻¹, which significantly exceeds previously reported records in both 2D and 3D thermoelectric materials. In G/hBN we find that the density fluctuations due to the substrate induced random potential fluctuations, $1.8 \times 10^{10} \text{cm}^{-2}$, represents a four-fold reduction compared to the value in G/SiO₂ sample $7.6 \times 10^{10} \text{cm}^{-2}$. Our findings show that the fast and low-power bipolar switching make it possible to integrate all-in-one graphene p-type and n-type devices. The study demonstrates the potential of graphene in thermoelectric applications especially in electronic cooling where large thermal conductivity (passive cooling) and large thermoelectric power factor (active cooling) are needed simultaneously.

ASSOCIATED CONTENT

Supporting Information

Device fabrication, Seebeck measurement, summary of recently reported *PFT* in 2D materials, Seebeck coefficient calculated from conductivity, nonlinear dependence on temperature, results from other G/hBN samples, and calculation details.

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Author contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Notes

The authors declare no competing financial interest.

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Figure captions

Figure 1. Thermoelectric measurement of Graphene at room temperature. (a) Optical micrograph of the graphene on hBN (G/hBN) device. (b) Measured Seebeck coefficient in G/hBN and G/SiO₂ devices as a function of back gate at 290K. Inset: measured resistance in both devices at 290K. (c) Measured *PFT* in both samples as a function of back gate at 290K. (d) Simulation of carrier density dependence of the Seebeck coefficient at 300K using the screened Coulomb scattering model for two values of the hBN thickness, d , and random potential fluctuations, E_{RP} , induced by charge impurities (See Supporting Information). The rectangular shadow corresponds to the EHP region in a sample with $d=10\text{nm}$, and $E_{RP}=40\text{meV}$.

Figure 2. Temperature dependence of Seebeck coefficient and EHP region. (a) Measured Seebeck coefficient in the G/hBN device as a function of back gate and temperature. (b) Temperature dependence of peak positions of the Seebeck coefficient (V_p) on the hole side for G/hBN (solid squares) and G/SiO₂ (open squares) devices are shown together with the exponential fit discussed in the text (solid lines). (c) Slope of polarity-switching effect from both our devices (solid squares for G/hBN and open squares for G/SiO₂). Values of V_p and slope in G/SiO₂ samples (open triangles) extracted from previous studies are also shown. (d) Sketch of proposed active cooler with integrated n-type and p-type legs.

Figure 3. Temperature dependence of Seebeck coefficient and *PFT* at fixed carrier density. (a) Measured Seebeck coefficient in G/hBN (solid squares) and G/SiO₂ (open squares) devices are plotted together with the theoretical values (solid lines) calculated by using the screened Coulomb scattering model discussed in the text. Dashed lines serve as guides to emphasize the nonlinear behavior. (b) Measured *PFT* (solid and open squares) from both devices are compared with theoretical values (solid lines). Peak Seebeck coefficient and *PFT* values extracted from previous studies in G/SiO₂ samples (open triangles) are also presented.

Figure 1

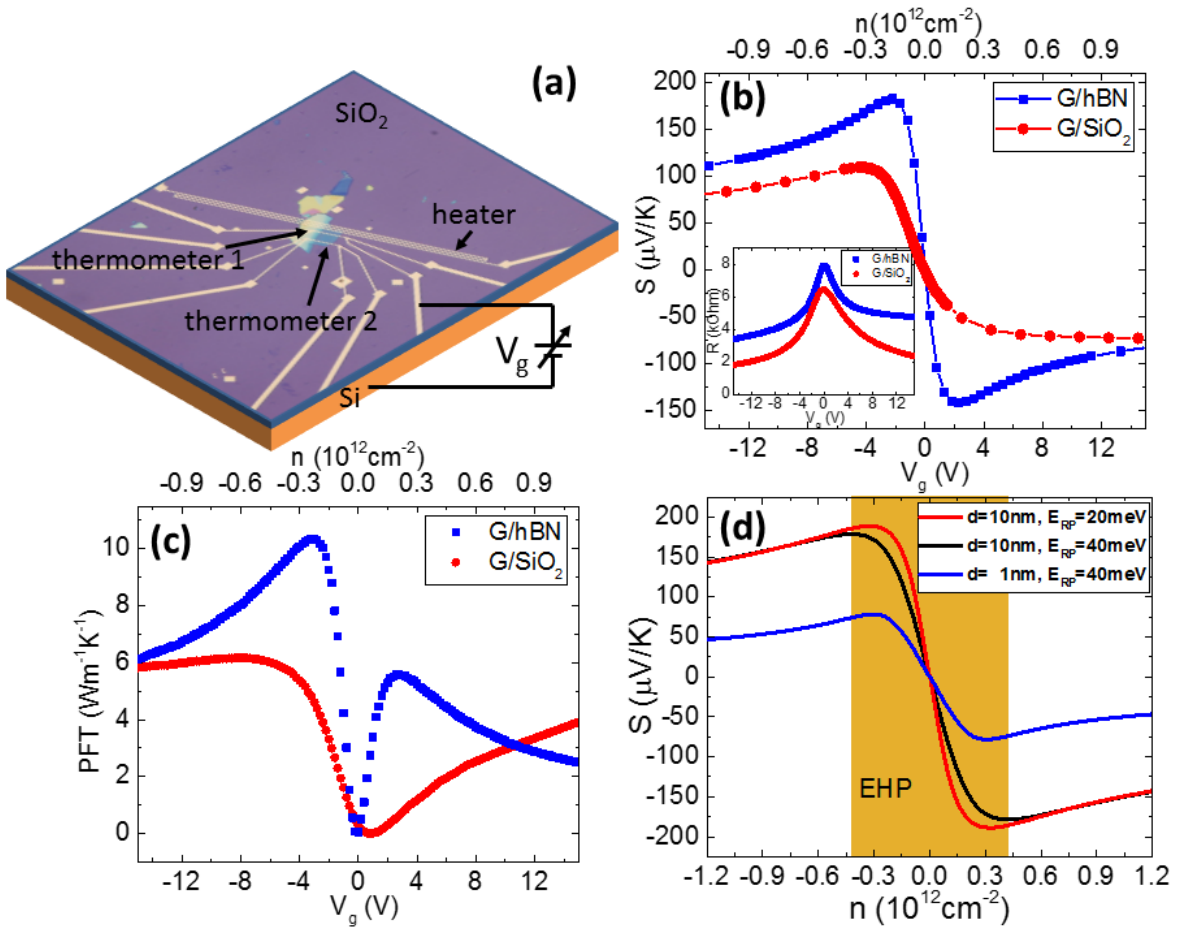


Figure 2

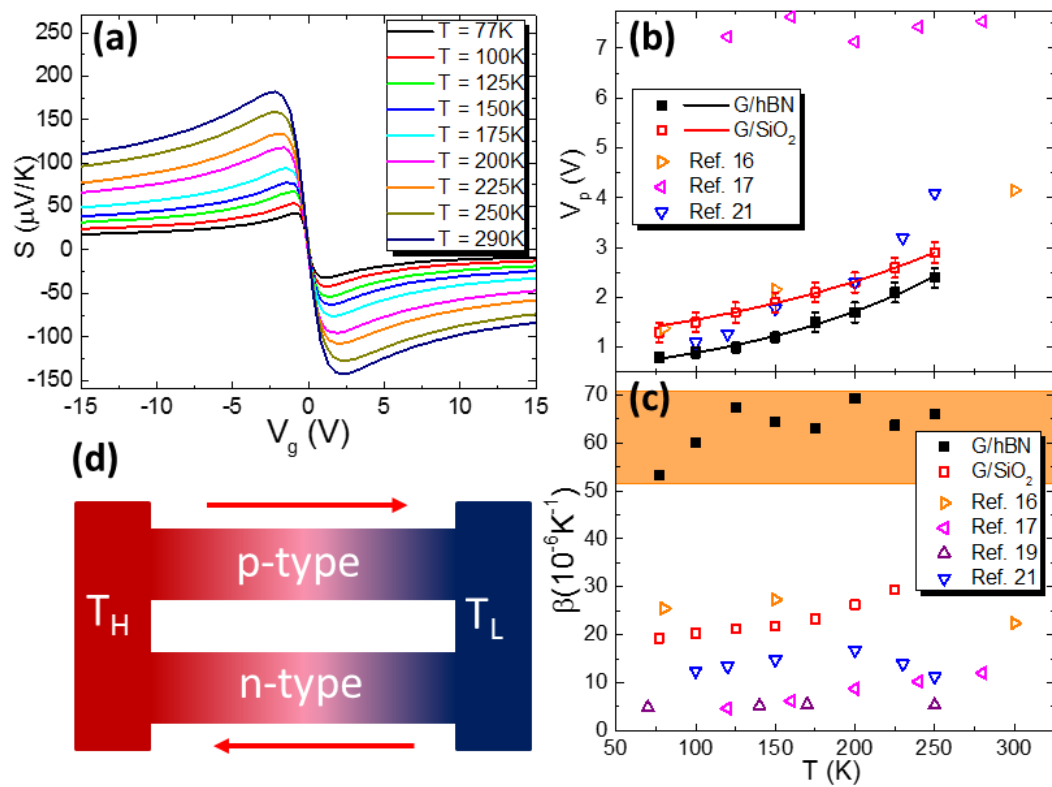


Figure 3

