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ABSTRACT
Epitaxial graphene on silicon carbide, or epigraphene, provides an excellent platform for Hall sensing devices in terms of both high electrical quality and scalability. However, the challenge in controlling its carrier density has thus far prevented systematic studies of epigraphene Hall sensor performance. In this work, we investigate epigraphene Hall sensors where epigraphene is doped across the Dirac point using molecular doping. Depending on the carrier density, molecular-doped close to the Dirac point could potentially outperform III–V Hall elements in the extended and military temperature ranges. The performance limits of epigraphene Hall sensors doped across the Dirac point. Low carrier densities demand the exploration of other even more suitable materials.1

Hall sensors detect magnetic fields by measuring the Hall voltage $V_{H}$ induced by an external field $B$. High device sensitivity implies a large magnitude of $V_{H}$ response to an external field, for a given bias current $I_B$ or voltage $V_B$. This leads to two important material-related metrics: the current-related sensitivity $S_I = |V_H/(IB)|$ [V/(AT)], which is essentially determined by the Hall coefficient $R_H$ ($\Omega/T$), and the voltage-related sensitivity $S_V = |V_H/(BV_B)|$ [V/(VAT)], which is ultimately limited by the carrier mobility $\mu = R_H/\rho$ [m$^2$/V s], where $\rho$ is the sheet resistance.

Graphene appears to be a natural candidate for highly sensitive Hall elements due to its high mobility and the possibility to tune carrier density $n$ toward charge neutrality (Dirac point). Low carrier density is desirable because it increases the Hall coefficient, $R_H = 1/(\rho n)$. Moreover, since the mobility $\mu = R_H/\rho$ of graphene is inversely proportional to carrier density as $\mu \propto 1/\sqrt{n}$, decreasing $n$ toward neutrality would increase both $S_I$ and $S_V$. In principle, low $n$ leads to an increase in $\rho$, which follows the relation $\rho \propto 1/n$, in the limit where charged impurity scattering dominates (supplementary material S1). Yet, decreasing $n$ can actually lead to a lower magnetic field detection limit, $B_{MIN} = V_N/(I_B R_H) (T/H)$, where $V_N$ is the voltage noise spectral density ($V/\sqrt{Hz}$). If Johnson–Nyquist noise dominates, then $V_N = V_{TH} \propto \sqrt{k_B T \rho}$, with $k_B$ being the Boltzmann constant, $T$ the temperature, and the detection limit scaling as $B_{MIN} \propto V_N/R_H \propto 1/\sqrt{n}$ for a fixed $I_B$. Disorder in real graphene samples prevents it from reaching true charge neutrality, but high-quality graphene can approach low carrier densities $n \sim 10^{10}$ cm$^{-2}$ at cryogenic temperatures.

The highest-quality graphene is obtained by mechanical exfoliation of graphite and encapsulation in hexagonal boron nitride (hBN-G). As a Hall sensor, hBN-G has shown ultra-high device sensitivities and detection limits comparable to those of silicon. However, this approach serves only as a proof-of-principle of the capabilities of graphene Hall sensors since device fabrication cannot be scaled up. Graphene grown using chemical vapor deposition (CVD) is a more...
scalable technology, which can also reach high sensitivities, but reported performance varies greatly,\textsuperscript{15,16} perhaps due to variability in material growth and the need for subsequent transfer to suitable substrates.\textsuperscript{16}

Epitaxial graphene on the SiC substrate (epigraphene) is another attractive scalable technology. The insulating substrate allows for direct mass fabrication of devices over wafer scales,\textsuperscript{17,18} forgoing the need for graphene transfer, thus increasing reproducibility and yield. Epigraphene is also compatible with operation at temperatures exceeding common industrial requirements.\textsuperscript{19,20} Despite these advantages, epigraphene remains relatively unexplored for Hall sensing in the literature, possibly owing to the difficulties in tuning carrier density due to high intrinsic n-doping, pinned by the substrate.\textsuperscript{21–23}

We report the exploration of the performance limits of epigraphene Hall sensors for varying doping levels across the Dirac point. Carrier density control is enabled by a molecular doping method using electron acceptors F4TCNQ assembled on the surface of epigraphene.\textsuperscript{11} Devices doped using this method have already shown excellent electrical properties and low charge disorder, albeit at low temperatures.\textsuperscript{24,25} We investigate Hall sensor figures of merit $B_{\text{MIN}}, S_V, S_I$, and finally thermal stability in ambient conditions from room temperature and just above 200 °C. Furthermore, we establish the limits for optimal operation of epigraphene Hall devices under realistic operational conditions.

Epigraphene was grown on 4H-SiC chips encased in a graphite crucible and heated using RF heating to around 1850 °C in an inert atmosphere of 1 bar argon.\textsuperscript{17} Transmission mode microscopy was performed by taking the power spectral density (PSD) using a voltage amplifier DLPVA-100-F-D from Femto Messtechnik GmbH, with the bandwidth limited to 100 kHz and the measured input noise level of 9 nV/Hz. High-field measurements were performed using a PPMS (Physical Property Measurement System from Quantum Design) cryostat (2–300 K) with a superconducting magnet providing fields up to 14 T. For heating experiments, the sample was mounted using epoxy on a ceramic heater, and temperature was monitored using a Pt100-resistor.

Seven epigraphene Hall sensors [Fig. 1(a)], spread across four chips, were investigated in total. They were designed using symmetric or cross-shaped geometries optimized with respect to $S_V$.\textsuperscript{27,28} Cryogenic measurements on a molecular-doped sensor demonstrates a full transition to the half-integer Quantum Hall regime, with vanishing longitudinal resistance $R_{XX}$ and quantized transverse resistance $R_{XY} = \hbar/(2e^2)$ [Fig. 1(b)]. These measurements verify that the devices are made of high-quality monolayer graphene with uniform doping.

Hall measurements of the transverse resistance $R_{XY} = V_{H}/I_B$ serve as a basis for the evaluation of epigraphene Hall magnetometers. The Hall coefficient, carrier densities, and mobilities are calculated from measurements in low magnetic fields ($B < 0.5$ T) as $R_{H} = dR_{XY}/dB$, $n = 1/(eR_{H})$, and $\mu = R_{H}/\rho$, respectively. For the low-field range, the linearity error of $R_{XY}$ is below 1%, which is determined by the percentage deviation of the raw data from the low-field linear fit [Fig. 2(a)]. The samples were tested up to $B = 13$ T at room temperature. For low doping ($R_{H} = 1284 \Omega/T$), the transversal resistance remains within 5% error in a range of $B = \pm 1.2$ T, but for higher doping ($R_{H} = 949 \Omega/T$), the 5% error range increases to $B = \pm 6$ T. The non-linearity of $R_{XY}$ is approximately $R_{XY} \propto B^2$ and is known to arise from geometrical and material correction effects.\textsuperscript{29} Figure 2 shows a summary of the carrier densities achieved in our experiments. The gap in data near charge neutrality ($n = 0$) indicates the disordered charge-puddle regime, characterized by a highly non-linear low-field $R_{XY}$.\textsuperscript{11} At room temperature, the maximum measured values of $R_{H}$ and $\mu$ are $R_{H} = 1440 \Omega/T$ and $\mu = 2300 \text{cm}^2/(V \text{s})$, respectively. In terms of charge disorder, at room temperature, epigraphene is in the puddle regime for doping levels $|n| < 5 \times 10^{11} \text{cm}^{-2}$, thus setting the maximum $R_{H}$ attainable in our epigraphene samples.

Figure 2(c) shows the linearity of $V_{H}$ at 100 mT up to the bias current of 6 mA, measured for highly and lowly doped devices. We find that for all carrier densities, the current–voltage (I–V) characteristic is linear within 5% error for $I_B < 2.5$ mA. The non-linearity is expected to be

![FIG. 1.](image-url)
ultimately due to self-heating. For instance, the measured Hall voltage may have a longitudinal voltage component, which can change non-linearly with a bias current due to Joule heating (supplementary material S2). For all subsequent measurements, we limit the bias current to below 1.5 mA to ensure a linear I–V behavior within 2% error.

The measurements in magnetic fields are complemented with noise measurements to unveil the minimum detection limit $B_{MIN}$. Figure 3(a) shows the low-bias ($I_B = 10 \mu A$) voltage noise spectral density $V_N$ measured at the Hall voltage terminals for different doping levels. In the low bias regime, the corner frequency of $1/f$ noise is around $\sim 30$ Hz. As doping in epigraphene approaches the Dirac point, the carrier density $n$ and mobilities $\mu$ are extracted from low-field Hall measurements. The Hooge parameter $\alpha_H$, which is an indication of noisiness of the devices, is in the range of $\alpha_H \approx 10^{-3}$ to $10^{-4}$ for $n \approx 10^{11} - 10^{12}$ cm$^{-2}$, lower than that of suspended graphene samples but comparable to that of GaAs. The deviation from ideal linear behavior could be due to joule heating and carrier density excitations. In practical devices, the excess noise can be alleviated by using spinning Hall current measurement techniques.

The measured sensitivities for epigraphene Hall sensors and their dependence on doping, collected across all measured devices, are

![FIG. 2. (a) Hall measurements showing linearity of $R_{XY}$ vs applied magnetic field. The inset shows behavior up to 13 T for different doping. The dotted lines are linear fits to low-field data $B < 0.5$ T. (b) Carrier densities $n$ and mobilities $\mu$ are extracted from low-field Hall measurements. (c) Linearity of Hall voltage measured at a fixed field of 100 mT vs applied bias current for highly ($R_H = 400 \Omega/T; n = 1.6 \times 10^{11}$ cm$^{-2}$) and lowly ($R_H = 1390 \Omega/T; n = 4.5 \times 10^{11}$ cm$^{-2}$) doped devices. The dotted lines are linear fits to low-bias data $I_B < 0.5$ mA. The offset in $V_H$ at zero field can be compensated by orthogonal vdP measurements and spinning current. Typically observed offsets are on the order of 1 mV for a bias current of $I_B = 10 \mu A$ (supplementary material S3).](image)

![FIG. 3. (a) Noise performance for one Hall sensor measured at different doping levels. The dotted lines are calculated noise levels assuming pure thermal noise of a resistor. (b) Measured voltage noise spectral density vs bias current in another lowly doped device. Noise peaks related to the power line have been partially filtered out digitally with sliding window averaging. Inset: the noise amplitude vs bias current at two different frequencies (black dotted lines).](image)
summarized in Fig. 4(a). The highest $S_I$ is reached for low doping levels, close to the puddle regime $n \sim 5 \times 10^{11}$ cm$^{-2}$. The highest $S_V$ occurs slightly outside the puddle regime, at doping levels $n \sim 6 \times 10^{11}$ cm$^{-2}$. We have performed full noise spectrum characterization [e.g., Fig. 3(b)] for four doping levels to obtain $B_{\text{MIN}} = V_S/(l_B R_{\text{H}})$, which includes not only intrinsic noise of epigraphene (thermal and 1/f noise) but also amplifier noise. Figure 4(b) shows $B_{\text{MIN}}$ as a function of $l_B$, measured at a frequency of 3 kHz for fair comparison to other graphene devices reported in the literature. The best $B_{\text{MIN}} = 47$ nT/H Hz is attained at lowest doping $n \sim 5 \times 10^{11}$ cm$^{-2}$, for $l_B = 400$ μm. At higher frequencies, where the 1/f noise contribution is lower, $B_{\text{MIN}}$ can be naturally lower with $B_{\text{MIN}} = 27$ nT/H Hz, for $n \sim 5 \times 10^{11}$ at 20 kHz [inset Fig. 4(b)].

Finally, Fig. 4(c) shows the thermal stability of the molecular-doped Hall sensor through the temperature coefficient $\Delta T$, defined as the percentage change of $R_{\text{H}}$ from its room temperature value per degree Celsius. Samples doped close to neutrality ($R_{\text{H}} = 1400$ Ω/T) display a temperature coefficient of $\Delta T = -0.6%$/C and undergo irreversible changes in the doping level at $T \approx 80^\circ$C (supplementary material S4). We achieve the highest thermal stability with samples annealed for $\sim 4$ h at $T = 160^\circ$C, after which the room temperature $R_{\text{H}}$ reached a stable value of $R_{\text{H}} \sim 300$ Ω/T due to partial desorption of dopants. After this curing step at 160$^\circ$C, samples showed a fairly low $\Delta T = -0.03%$/C up to $T = 150^\circ$C, while still displaying respectable performance at $T = 150^\circ$C, with $S_V \approx 0.12$ V/(VT), $S_I \approx 300$ V/(AT), and $B_{\text{MIN}} \sim 100$ nT/H Hz.

Table I shows a comparison of our devices with other Hall sensors reported in the literature. The maximum current-related sensitivity in doped epigraphene is found to be on the order of $S_I \approx 1500$ V/(AT) at room temperature. This value is limited by the minimum $n$ attained in our sample ($|n| < 5 \times 10^{11}$ cm$^{-2}$) and is set by the disorder present in the as-grown material, combined with additional contributions from external doping and thermally excited carriers in the dopant layer and the SiC substrate. Decoupling epigraphene and substrate by hydrogen intercalation has led to high $\mu$ at cryogenic temperatures. However, at room temperature, the lowest $\mu$ values reported for H-intercalated epigraphene are all above $1 \times 10^{12}$ cm$^{-2}$ V$^{-1}$ s$^{-1}$, with $\mu \sim 1300$–1700 cm$^2$/V s. These mobilities are lower than the highest reported for epigraphene at room temperature $[\mu = 5500$ cm$^2$/V s] and the ones achieved in this work $[\mu = 2300$ cm$^2$/V s]. Above room temperature, interactions between epigraphene and the substrate via longitudinal-acoustic and remote interfacial phonon scattering further degrade mobility. The stable temperature range ($T < 80^\circ$C) for samples doped close to the Dirac point is determined by our current choice of doping method. A high thermal stability up to $T = 150^\circ$C is achieved after curing the samples at a temperature of 160$^\circ$C for 4 h. The resulting temperature coefficient $\Delta T = -0.03%$/C could then be understood as the intrinsic thermal drift of epigraphene and not due to desorption of dopants. This implies that by using an alternate thermally stable doping scheme, epigraphene could well outperform Hall element-based III–V at high temperatures. Our work paves the way for the development of...
epigraphene Hall sensors for real-world applications, which require durable, controllable, and sensitive devices produced in a scalable way.

See the supplementary material for extra data on sheet resistance vs carrier density, linearity error, offset voltage, and heating ramps.

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DATA AVAILABILITY

The authors declare that the main data supporting the findings of this study are available within this article and supplementary material. Additional data are available from the corresponding author upon request.

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