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## Onsager-Casimir frustration from resistance anisotropy in graphene quantum Hall devices


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We report on nonreciprocity observations in several configurations of graphene-based quantum Hall devices. Two distinct measurement configurations were adopted to verify the universality of the observations (i.e., two-terminal arrays and four-terminal devices). Our findings determine the extent to which epitaxial graphene anisotropies contribute to the observed asymmetric Hall responses. The presence of backscattering induces a device-dependent asymmetry rendering the Onsager-Casimir relations limited in their capacity to describe the behavior of such devices, except in the low-field classical regime and the fully quantized Hall state. The improved understanding of this quantum electrical process broadly limits the applicability of the reciprocity principle in the presence of quantum phase transitions and for anisotropic two-dimensional materials.

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### I. INTRODUCTION

It is widely known that graphene exhibits a variety of unique properties [1–4]. In certain forms, including epitaxial graphene (EG) grown on 4H-SiC, this versatile material has been identified as a practical way to develop resistance standards based on a robust quantum Hall effect (QHE) that appears over a useable range of magnetic fields ( $B$  fields), with the key feature being a well-quantized and extended resistance plateau [5–11]. Reported graphene-based standards operate almost exclusively at the filling factor  $\nu = 2$ , although a recent effort has been able to assess the viability of the  $\nu = 6$  plateau [12]. For the  $\nu = 2$  plateau, one expects the resistance value:  $\frac{1}{2} \frac{h}{e^2} = \frac{1}{2} R_K \approx 12906.4037 \Omega$ , where  $h$  is the Planck constant, and  $e$  is the elementary charge.

In the past, the referenced graphene-based standards have been primarily single Hall bar devices, yielding a single operable value of resistance. However, recent advances in fabrication techniques have enabled the assembly of multiple Hall bars in parallel or in series to create resistance values of  $qR_K$ , where  $q$  is a positive rational number [13–19]. Before these forms of standard devices are globally implemented, it is critical to disseminate best practices for characterization of the Hall resistance quantization for  $B$  field and current dependence. The symmetry of electrical conductance for opposite perpendicular directions of  $B$  field is one such criterion, as a

result of the well-known Onsager-Casimir relations (OCRs) [20–22].

In this paper, we investigate the root cause of observed nonreciprocity in three types of large graphene quantum Hall devices: standard Hall bars with a length and width of 2 mm and 400  $\mu\text{m}$ , respectively, arrays of 13 parallel elements with quantized resistance  $R_K/26 \approx 992.8 \Omega$  at the  $\nu = 2$  plateau, and a 6.45 k $\Omega$  array consisting of eight elements in a series-parallel configuration. Electrical characterization of five Hall bars, four 13-element arrays, and one 8-element array yielded very similar results, and the data presented here are representative. All measurements were done in the four-terminal (4-T) measurement configuration, but the arrays are inherently two terminal (2-T) in their design, as required in precise QHE parallel array measurements [15]. Data were obtained by symmetrized lock-in measurements and with a direct current comparator (DCC) resistance bridge to assist in eliminating potential instrumental causes for observing nonreciprocity.

Our analysis determines that the structural anisotropies of EG contribute to the observed asymmetric Hall responses at intermediate magnetic fields. We posit that substrate morphology directly affects electron density variation that reduces the conductivity [23], and by extension, the anisotropic substrate morphology results in electrons experiencing nonuniform pseudomagnetic fields [24]. This results in backscattering, whose presence induces a device-dependent asymmetry, making the reciprocity relations limited in their capacity to describe the behavior of such devices.

### II. EXPERIMENTAL METHODS

#### A. Sample preparation

EG films were grown on 4H-SiC substrates at temperatures near 1900  $^\circ\text{C}$ , with the sublimation of Si atoms allowing excess carbon at the surface to reorganize into a defect-free

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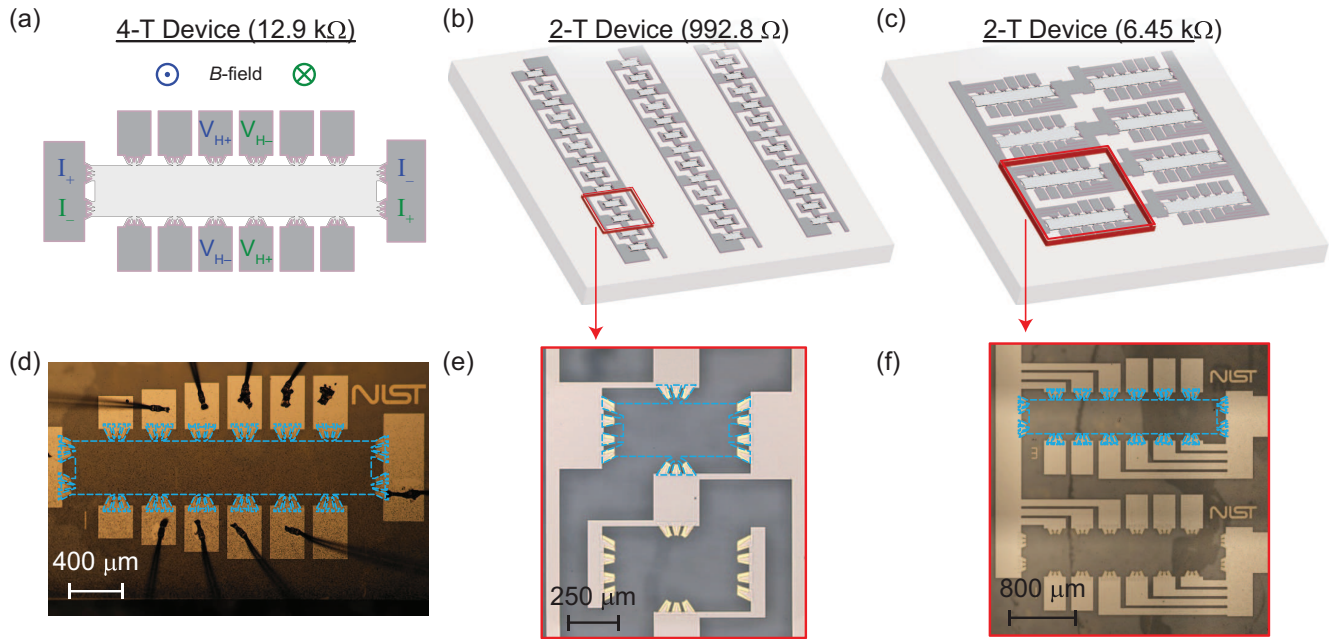


FIG. 1. (a) An illustration of the 12.9 k $\Omega$  Hall bar devices. Two example four-terminal (4-T) measurements are shown and color-coded with the corresponding in- or out-of-page magnetic field direction. Illustrations of the (b) 992.8  $\Omega$  array and (c) 6.45 k $\Omega$  array devices are provided for clarity. Microphotographs of the respective postfabrication device elements are shown for the (d) 12.9 k $\Omega$  standard Hall bar, (e) 13-element array, and (f) 6.45 k $\Omega$  array.

hexagonal lattice [25]. Chips were first diced from 4H-SiC(0001) wafers with atomically smooth Si-face surface obtained from CREE and chemically cleaned with a 5:1 diluted solution of hydrofluoric acid and deionized water. Before growth, some chips were coated with a very dilute solution of the carbon-based resist AZ 5214E in deionized water to utilize polymer-assisted sublimation growth (PASG) [26]. The silicon-face side of each chip was placed in close proximity ( $<2 \mu\text{m}$ ) with a polished glassy carbon slab (SPI Glas 22) to limit Si escape and improve graphene uniformity. The growth furnace was flushed with argon gas and filled to  $\sim 103 \text{ kPa}$  from a 99.999% liquid argon source. The graphite-lined resistive-element furnace (Materials Research Furnaces Inc.) was held at  $1900^\circ\text{C}$  for 4–5 min. The furnace heating and cooling rates were  $\sim 1.5^\circ\text{C/s}$ .

It should be noted that, after the growth, films were vetted by means of optical and confocal laser scanning microscopy to select those with monolayer coverage  $>99\%$  (and uniform SiC step heights  $<1 \text{ nm}$ ), as described in a previous work [27]. For device fabrication, the EG layer was protected by a 20 nm layer of Pd/Au, followed by a photolithography process that defines the Hall bar and device contact pattern [28,29]. Thus, the Pd/Au layer and some exposed areas of SiC are covered with thicker Au to serve as the contact material with the device. For the 2-T array devices, a 100 nm layer of superconducting NbTiN was applied over the contacts to form device interconnects with superior performance [13]. The separation of the superconductor layer and the EG was  $>80 \text{ nm}$  to prevent undesired quantum effects. Some of the chips were grown without PASG preprocessing, resulting in parallel SiC steps of increased height (1–5 nm) and  $>99\%$  monolayer graphene, enabling us to quantify the influence of the steps themselves.

The final step for fabricating these quantum Hall devices was the functionalization process to regulate the electron density without the need for a top gate. The functional group used was  $\text{Cr}(\text{CO})_3$ , and it has been successfully implemented in a variety of other studies [30,31]. Hexahapto functionalization [ $(\eta^6\text{-graphene})\text{-Cr}(\text{CO})_3$ ] was initiated with a small, nitrogen-filled furnace at  $130^\circ\text{C}$ . The typical electron density of functionalized devices after being stored in air for at least 1 d is of the order of  $10^{10} \text{ cm}^{-2}$ , and its uniformity varies on that same order across the entire chip [31], which can be compared with the typical values of inherent doping in EG of  $10^{13} \text{ cm}^{-2}$  [32]. As a control, some of the devices with the larger step edges were not functionalized to determine whether any sample anisotropies were attributable to the presence of  $\text{Cr}(\text{CO})_3$ .

A set of final device illustrations is shown in Figs. 1(a)–1(c), with corresponding images in Figs. 1(d) and 1(f). The first device type shown in (a) is a 12.9 k $\Omega$  standard Hall bar, suitable for 4-T measurements using distinct source-drain and voltage contacts. The second device type (b) is a 992.8  $\Omega$  array, composed of 13 Hall bars connected in parallel. The third device type (c) is a 6.45 k $\Omega$  array device composed of a  $4 \times 2$  interconnected grid of Hall bars. Both array device types are exclusively 2-T but are measured as 4-T using separate voltage and current leads connected to the superconductor at the source and drain contacts, where we have implemented a multiple-branch design required to optimize the current flow and eliminate the effect of contact resistances [13,15]. The array designs also provide inherent reciprocity for reversal of the magnetic field direction in the QHE regime.

It should be noted that the main difference between the three kinds of devices is the contact configuration, e.g., only the single Hall bar devices could be measured using conventional 4-T magnetoresistance measurements at distinct



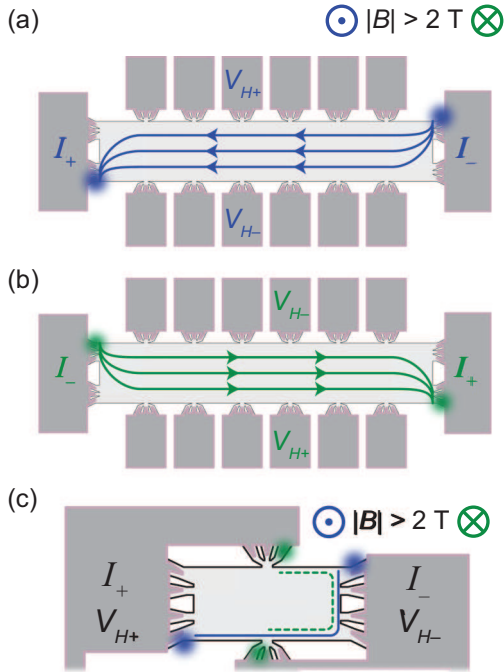


FIG. 2. (a) and (b) Expected current behaviors for the standard Hall bar are illustrated and correspond to a four-terminal configuration for positive and negative  $B$  fields, respectively. (c) Example of an equipotential line in one element of an array device for positive (solid blue) and negative (dotted green)  $B$  field. Blurred circles represent hotspots or approximate points of electron entry or exit where most dissipation occurs in the quantum Hall effect (QHE) regime.

144 contacts. In all array devices, elements share common electrical  
 145 connections formed by the superconductor. For the 992.8  
 146  $\Omega$  array devices, symmetric sets of contacts access each Hall  
 147 bar, whereas for the 6.45 k $\Omega$  array devices, sets of four and  
 148 five contact pads contact each Hall bar element and are inter-  
 149 connected by NbTiN. With these differences, one can confirm  
 150 that the measured anisotropies we will report are not the result  
 151 of a particular contact configuration.

### B. Quantum Hall transport

153 For quantum Hall transport measurements, a Janis Cryo-  
 154 genics  $^4\text{He}$  cryostat was used. The relevant data were collected  
 155 at magnetic field values between 0 and  $\pm 9$  T to characterize  
 156 the magnetoresistances of the devices. Measurements were  
 157 performed between 1.5 and 10 K with source-drain currents as  
 158 high as 1 mA. Devices were annealed in vacuum, as described  
 159 in Ref. [31], to obtain a desired electron density. The expected  
 160 current behavior at low temperatures and varying magnetic  
 161 fields are shown in Fig. 2. All blurred circles in Fig. 2 repre-  
 162 sent hotspots or areas associated with the majority of electron  
 163 flow to and from the device electrical contacts in the QHE  
 164 regime [33].

165 Regarding the 4-T and 2-T devices, we followed the sym-  
 166 metry relation described in Büttiker's work [20]. Observations  
 167 of strong asymmetry in resistance measurements for the 4-T  
 168 measurements may appear as a result of local current flow  
 169 behavior contributing to the measurement when the  $B$ -field

170 direction is reversed without switching the voltage and current  
 171 electrodes. Rather than measure the potentials purely  
 172 associated with the reservoirs serving as the current source and  
 173 drain, local potentials near the voltage terminals become  
 174 embedded in the response [34]. Such local potentials may  
 175 change when the  $B$ -field direction is reversed. For the 2-T ar-  
 176 ray contact configurations, the same electrodes were used for  
 177 both applying current and measuring voltage differences, and  
 178 the current flow is derived from the normal and QHEs. The  
 179 2-T devices also do not suffer from resistance measurement  
 180 errors due to low impedance lock-in amplifier inputs since any  
 181 current drawn is supplied by the voltage or current source.

### III. OBSERVING NONRECIPROCITY

182 One electrical measurement configuration is equated to a  
 183 second one by means of the OCR [20–22], wherein the current  
 184 terminals are exchanged with the voltage terminals and the  
 185 positive current probe becomes the positive voltage probe, and  
 186 likewise for the negative terminals. Illustrations are shown in  
 187 Fig. 3 for (a) positive  $B$  fields and (b) negative  $B$  fields. For  
 188 this first set of measurements, the focus is on the single-device  
 189 longitudinal resistance (4-T), whose corresponding data are in  
 190 Fig. 3(c). To compare the resistances from the positive and  
 191 negative  $B$ -field cases, the latter is reflected about the vertical  
 192 axis, identical to taking the absolute value of the magnetic  
 193 field reading.

194 Effects of hysteresis due to trapped flux in the super-  
 195 conducting magnet were minimized by the experimental  
 196 procedure. For the 4-T measurements, fixed  $B$ -field values  
 197 were used rather than continuously ramping the field. The  $B$   
 198 field was adjusted to the desired value, always with increasing  
 199 magnitude of  $B$ , followed by resistance measurements using  
 200 a fixed driving current. All first-order thermoelectric effects  
 201 were removed by averaging the measured resistance values  
 202 for positive and negative current directions.

203 Upon first glance, the longitudinal resistances overlap, but  
 204 upon taking the difference of the two curves, as shown in  
 205 Fig. 3(d), a small yet measurable and reproducible change  
 206 is visible (blue curve, left axis). The global minimum of the  
 207 first derivative of the resistance with respect to the positive  $B$ -field  
 208 case (red curve, right axis). In Figs. 3(e) and 3(f), illustrations  
 209 of the 4-T Hall measurements are shown for the positive  
 210 and negative  $B$ -field cases, respectively. The same operations  
 211 were conducted for the corresponding Hall resistances, as  
 212 seen in Fig. 3(g). The resistance difference, defined as  $\Delta R =$   
 213  $R_{B+} - R_{B-}$ , in Fig. 3(h) is more than an order of magnitude  
 214 smaller than the longitudinal case, and although the resistance  
 215 derivative is of similar order, the sign is reversed.

216 The observations of nonreciprocity are not exclusive to  
 217 4-T devices. Using 2-T 992.8  $\Omega$  and 6.45 k $\Omega$  array devices,  
 218 similar differences in the combined (Hall and longitudinal)  
 219 resistances can be seen. In Figs. 4(a) and 4(b), the mixed resis-  
 220 tance (top panel) response maintains a symmetric appearance  
 221 but yields a  $\Delta R$  and first derivative behavior (blue and red  
 222 curves in the bottom panel, respectively) like the 4-T configu-  
 223 ration. In Fig. 4(c), the derivative of the 2-T device resis-  
 224 tance curve (positive  $B$ -field case) and  $\Delta R$  are shown as a function  
 225 of magnetic field for different electron densities. In the ideal  
 226 of magnetic field for different electron densities. In the ideal  
 227

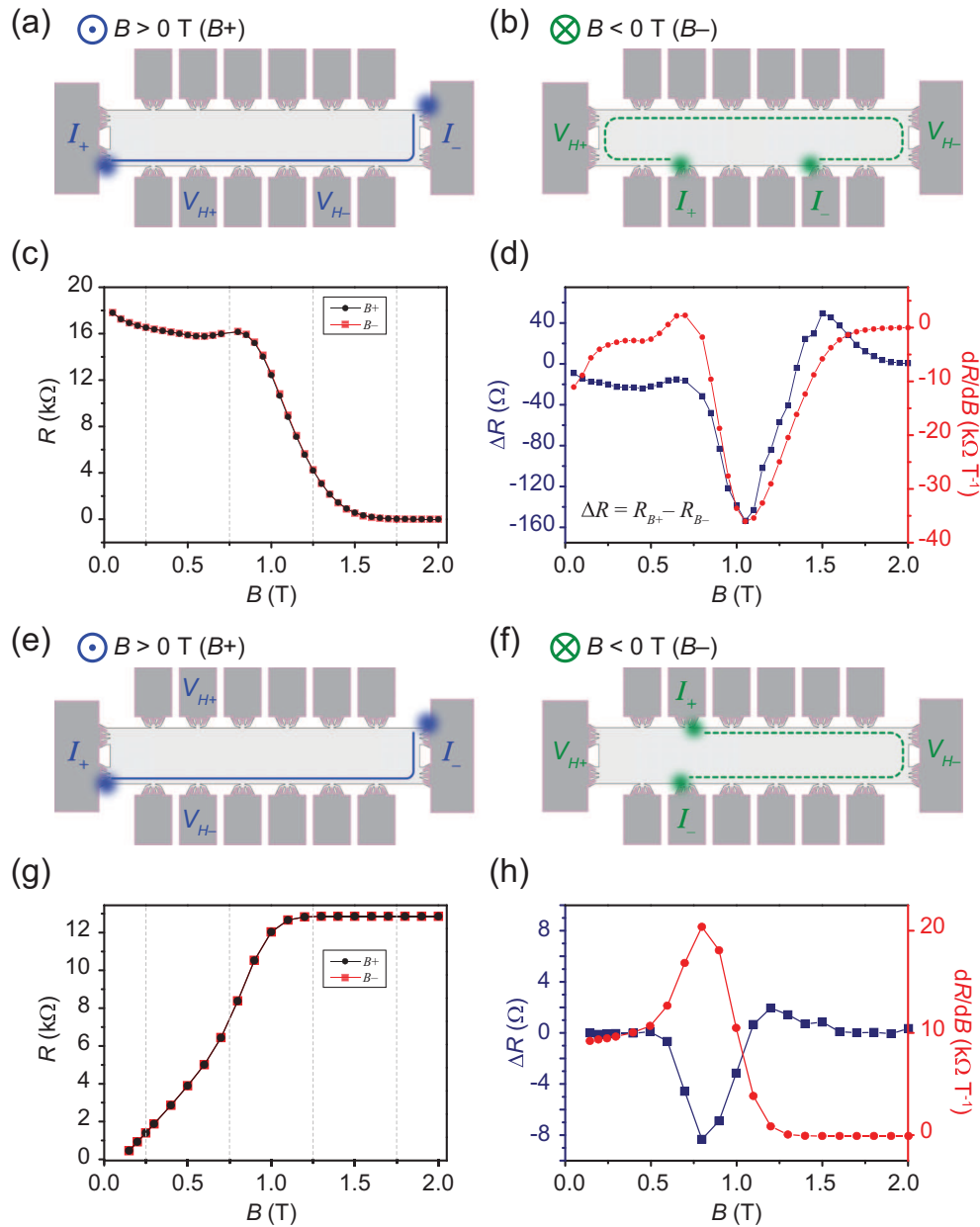


FIG. 3. The differences in the measured longitudinal resistance [four terminal (4-T)] for (a) positive  $B$  field and (b) negative  $B$  field. Both illustrations have an example equipotential line as a reference. The illustrated measurement configurations correspond to the two sets of data in (c). By reflecting the negative  $B$ -field data about the vertical axis, nonreciprocity in the longitudinal resistances may be observed. (d) The difference between the two curves reveals a small yet reproducible effect. For comparison, the first derivative of the resistance with respect to the positive  $B$ -field case is shown in red. (e)–(h) The same analyses were conducted for the corresponding Hall resistances. Note that the epitaxial graphene (EG) on this device was grown via the polymer-assisted sublimation growth (PASG) method, which greatly reduces any inhomogeneity or anisotropy due to the step edges. These devices were functionalized with  $\text{Cr}(\text{CO})_3$ .

228 case, the bottom panel of Fig. 4(c) should yield no differences  
 229 for devices obeying the OCR. The inset of the bottom panel  
 230 shows the peak value of  $\Delta R$  as a function of electron density  
 231 and suggests that the nonreciprocity gradually decreases with  
 232 higher  $n$ .

#### 233 IV. DETERMINING THE CAUSE OF NONRECIPROCITY

##### 234 A. Device inhomogeneities

235 The aforementioned observations of nonreciprocity are  
 236 consistent within all three device types, where multiple de-

vices were measured within each category, prompting a more  
 careful analysis. More data are available in the Supplemental  
 Material [35], including comparisons of the homogeneity of the  
 electron density in all device types as well as the nonreciprocity  
 behavior as a function of injected current. Additionally, consistent  
 nonreciprocity observations for the  $\nu = 6$  plateau are provided.

One immediate consideration to make when seeing any data that  
 do not conform exactly to well-established principles is the quality  
 of the device. Assuming a uniform electron density [35], one may  
 confirm the quality of the device by

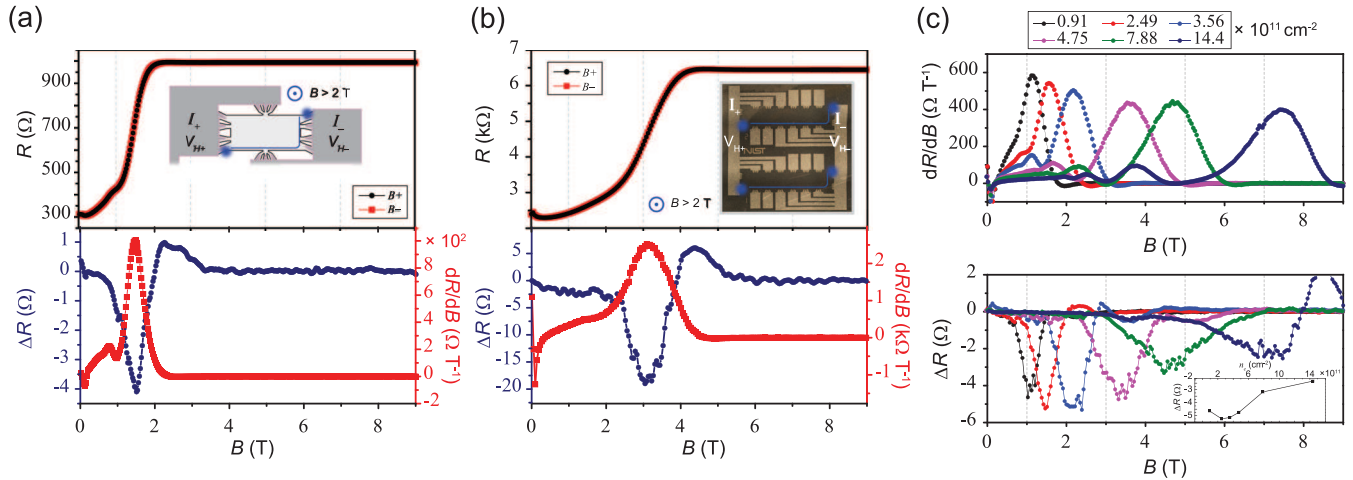


FIG. 4. Measurements using a two-terminal (2-T) device in a four-terminal (4-T) measurement configuration for the (a) 992.8 Ω array and (b) 6.45 kΩ array devices (insets show example equipotential line in solid blue). The top panels show the combined Hall and longitudinal resistance, as well as the corresponding positive  $B$ -field measurement configuration, and the bottom panels show  $\Delta R$  in blue and first derivative of the positive  $B$ -field case in red. (c) Both the derivative of the combined resistance curve for the 992.8 Ω array (positive  $B$ -field case) and  $\Delta R$  between the two conditions are shown as a function of electron density. In the ideal case, the bottom panel should yield no differences for devices obeying the Onsager-Casimir relation (OCR).

248 inspecting the transport characteristics. In Fig. 5, three such  
 249 checks are presented. First, the fraction in the top panel  $\beta$  is  
 250 the  $y$ -axis intercept for the lines in the middle panel. A value  
 251 of  $\beta = 0.5$  implies that the behavior of the carriers inside

the graphene are Dirac-fermionlike. It also affirms that the  
 Cr(CO)<sub>3</sub> functionalization did not influence the behavior of  
 those carriers. Furthermore, it supports the notion that the EG  
 is of high quality since the results are like those from other  
 works utilizing high-quality graphene [36]. Note that the error  
 bars are smaller than the data points.

The middle panel of Fig. 5 shows the Landau index  $N$  is  
 plotted against the inverse of the applied  $B$  field for the same  
 set of electron densities as seen in Fig. 4, and the usual linear  
 relationships between these two quantities were verified. Note  
 that the positions of the Landau indices are obtained from the  
 second derivative of the measured resistance [37]. The bottom  
 panel of Fig. 5 shows the second derivative of one of the  
 electron density cases, and its periodicity serves as another  
 confirmation of sample homogeneity (see Supplemental Material  
 [35]).

Additional methods were utilized to evaluate the possible  
 contributions of monolayer EG quality and device contact ar-  
 rangements to nonreciprocity. In Fig. 6(a), the first and second  
 derivatives of 2-T device measurements for both magnetic  
 field polarities are shown for the 992.8 Ω array device in the  
 top and bottom panels, respectively. The similar appearance  
 confirms the symmetry exhibited by the device as it transitions  
 to the quantum Hall regime. Despite this symmetry, the  $\Delta R$   
 behavior still showed peaks aligned with the first derivatives,  
 suggesting that general device quality is not a major contribu-  
 tor in  $\Delta R$ .

As shown in Fig. 6(b), both the zero-field and low-field  $I$ - $V$   
 curves are measured at 1.6 K to verify device linearity, which  
 is another indicator of general quality and homogeneity. The  
 fact that our devices are electrically linear validates the basic  
 requirements for reversed-field reciprocity [21]. The final  
 device quality check was performed with a DCC. These precision  
 measurements [Fig. 6(c)] show, in the high-field limit of 5 to 9 T,  
 that data from both  $B$ -field polarities approach the fully  
 quantized state for currents  $< \sim 700 \mu\text{A}$ . This behavior  
 seen with the DCC confirms that the 992.8 Ω array device

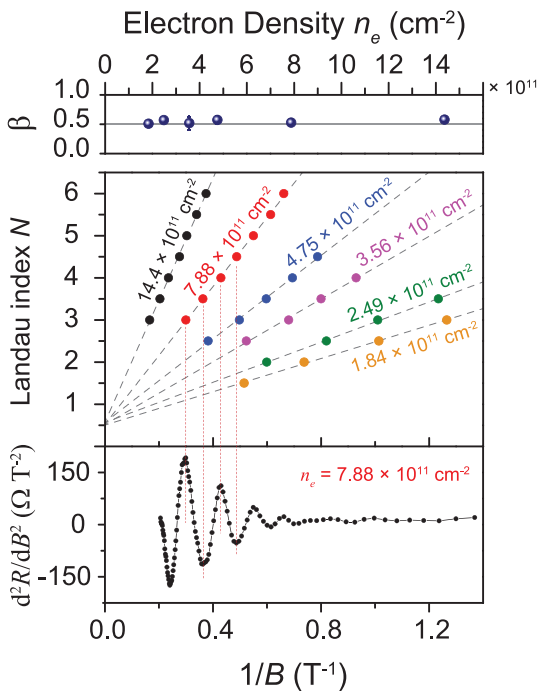


FIG. 5. Data are shown for the 992.8 Ω array. The fraction in the  
 top panel  $\beta$  is the  $y$  intercept for the lines in the middle panel.  
 The middle panel shows the Landau index  $N$  plotted against  $1/B$  for a  
 set of six widely spaced electron densities. The bottom panel shows  
 the second derivative of one of the electron density cases, and its  
 regular periodicity confirms sample homogeneity. This behavior is  
 universal to all tested devices. Error bars indicate a  $1\sigma$  uncertainty  
 of the data collected at the corresponding point.

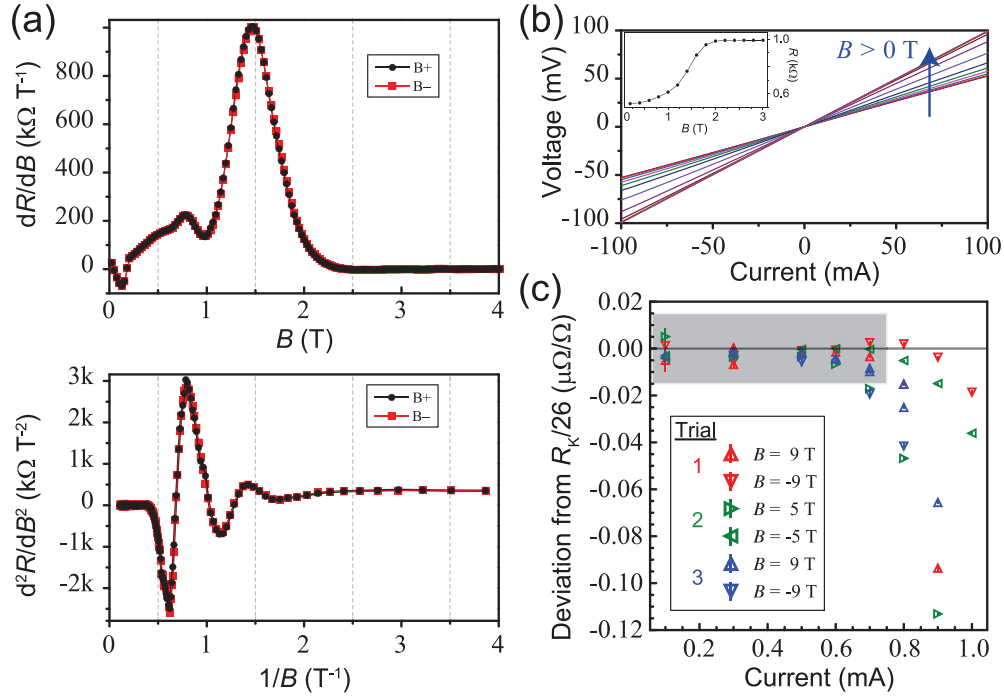


FIG. 6. (a) The first and second derivatives of both resistance measurements for the  $992.8 \Omega$  array device are shown in the top and bottom panels, respectively. The similarity between  $B+$  and  $B-$  confirms that the device inhomogeneity, averaged over the 13 elements, does not determine  $\Delta R$  and suggests that the device quality is not a factor in why those differences appear. (b) Zero-field and low-field  $I$ - $V$  curves are measured at 1.6 K to verify device linearity, another indicator of homogeneity. (c) Direct current comparator (DCC) measurements verify that, in the high-field limit, the resistance for both  $B$ -field directions approaches the value  $\frac{R_K}{26} \approx 992.8 \Omega$  to better than one part in  $10^8$  for currents up to  $700 \mu\text{A}$ , confirming that this quantum Hall effect (QHE) array device utilizes highly homogeneous graphene.

289 was fabricated from the highest quality film growths, as were  
290 all devices.

### 291 B. Anisotropy in EG

292 The data shown in Fig. 4 are consistent with all devices,  
293 namely, that all  $\Delta R$  become small at low  $B$ , in the linear  
294 Hall regime. To explore this behavior more closely, zero-field  
295 measurements were first conducted to confirm whether we can  
296 measure the OCR accurately (to within the noise of the equip-  
297 ment). Without any applied  $B$  fields, the OCR is expected to  
298 hold. Data supporting this zero-field expectation are shown  
299 in Fig. 7. Not only do the devices demonstrate linearity at  
300 high temperatures, but one can also see that  $\Delta R$ , as a function  
301 of temperature, remains at zero within the equipment noise.  
302 This observation supports the notion that device quality is  
303 not a significant contributor to observed asymmetries. We  
304 therefore conclude that the cause of the OCR asymmetries in  
305 our measurements is related to  $B$ -field-induced asymmetry.

306 In micrometer-scale EG-based quantum Hall devices, it has  
307 been reported that  $B$ -field asymmetry mainly resulted from  
308 electron backscattering and was a gate-tunable phenomenon  
309 [38]. Therefore, the OCR may not hold if backscattering takes  
310 place in our system. To further support the notion that  $\Delta R$   
311 results from backscattering, the backscattering strength was  
312 calculated for the injected current in the longitudinal direction  
313 using the following formula:  $\gamma = \frac{R_L}{R_L + R_H}$  [39]. Figure 8(a)  
314 shows the backscattering strength as a function of positive  $B$   
315 field, along with the corresponding  $R_{xx}$  and  $R_{xy}$ . In Fig. 8(b),

the difference of backscattering strength  $\Delta\gamma = \gamma_{B+} - \gamma_{B-}$  for  
316 low fields shows a strong similarity to the measured  $\Delta R$ . 317

318 It should be noted that the backscattering strength shown  
319 in Fig. 8(a) combines all sources of backscattering, including  
320 those from differences in the population of Landau levels as  
321 the  $B$  field changes. This population change has been reported  
322 as being inherently linked to  $B$ -field symmetry [21]. There-  
323 fore, the observed backscattering-related  $B$ -field asymmetry  
324 in our devices must originate by some other cause.

325 In the case of EG, the presence of SiC step edges precludes  
326 the uniform distribution of electrons over the device area. It  
327 can thus be stated that the electron density is directly influ-  
328 enced by local substrate morphologies, which in turn result in  
329 nonuniform  $B$  fields acting on electrons in any deformed areas  
330 [40–42]. The strength of any backscattering depends on the  
331 applied  $B$  field [43,44]. At low  $B$  fields, a diverse population  
332 of states exists in the device due to the transitions between  
333 neighboring Landau levels, electron density fluctuations, and  
334 nonuniform  $B$  field. Thus, electronic states are readily avail-  
335 able for backscattering events. This increased backscattering  
336 results in a greater  $B$ -field asymmetry, thus intensifying the  
337 breakdown of the reliability of measuring the OCR.

338 At high  $B$  fields, this diverse population no longer prop-  
339 agates as such through the device due to the large spacing  
340 between the zeroth and first Landau levels in EG. The Fermi  
341 level, though susceptible to perturbations, still remains within  
342 the Landau gap. Therefore, even if the electron density is  
343 not uniform, wide separation of Landau levels is expected  
344 to suppress any backscattering. This phenomenon is unique



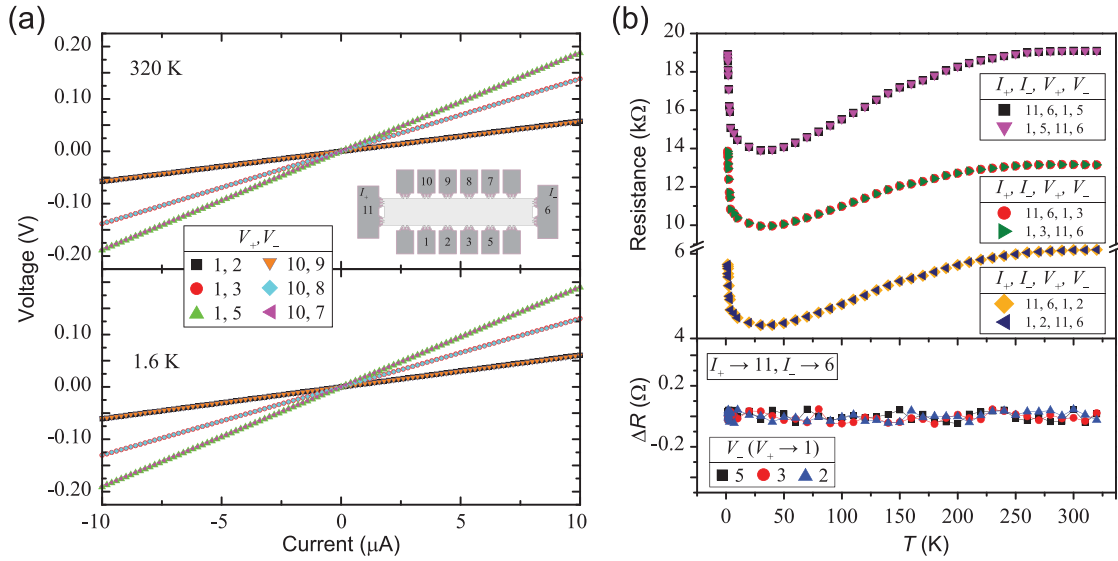


FIG. 7. (a) The  $I$ - $V$  curves at zero field are shown for an example device at 320 and 1.6 K, in the top and bottom panel, respectively. The linearity supports overall device homogeneity. (b) Temperature-dependent measurements of the Onsager-Casimir relation (OCR) at zero field demonstrate that the presence of a  $B$  field gives rise to the observed  $\Delta R$  in other data.

in EG-based systems because the  $\nu = 2$  plateau persists for large  $B$  fields [33]. Note that the backscattering from the step edges is  $B$ -field asymmetric since the current path changes with the direction of the  $B$  field (Fig. 3). The morphology and number of the step edges are different for each current path, thus resulting in different backscattering strengths.

Because these devices are macroscopic (surface dimensions  $>100 \mu\text{m}$ ), any asymmetric contributions to the resistance from backscattering are averaged over many random current paths. Our data show that anisotropy is inherently a phenomenon observable during the phase transitions of the quantum Hall states. In smaller-scale devices, anisotropies are

expected to have larger impacts [21,26,45–48], with higher temperatures also causing a suppression of backscattering (for temperature suppression data, please see the Supplemental Material [35]). For this paper, since EG was grown with different methods, different surface morphologies were accessible. Furthermore, EG properties varied in that some devices were functionalized, and some were not, and different contact configurations and contact pad compositions were used, while OCR asymmetries were consistently observed.

Due to the complexity and size of the EG system, as well as the subtle, sample-dependent differences in how the step edges form, simulations or comparisons of the absolute values

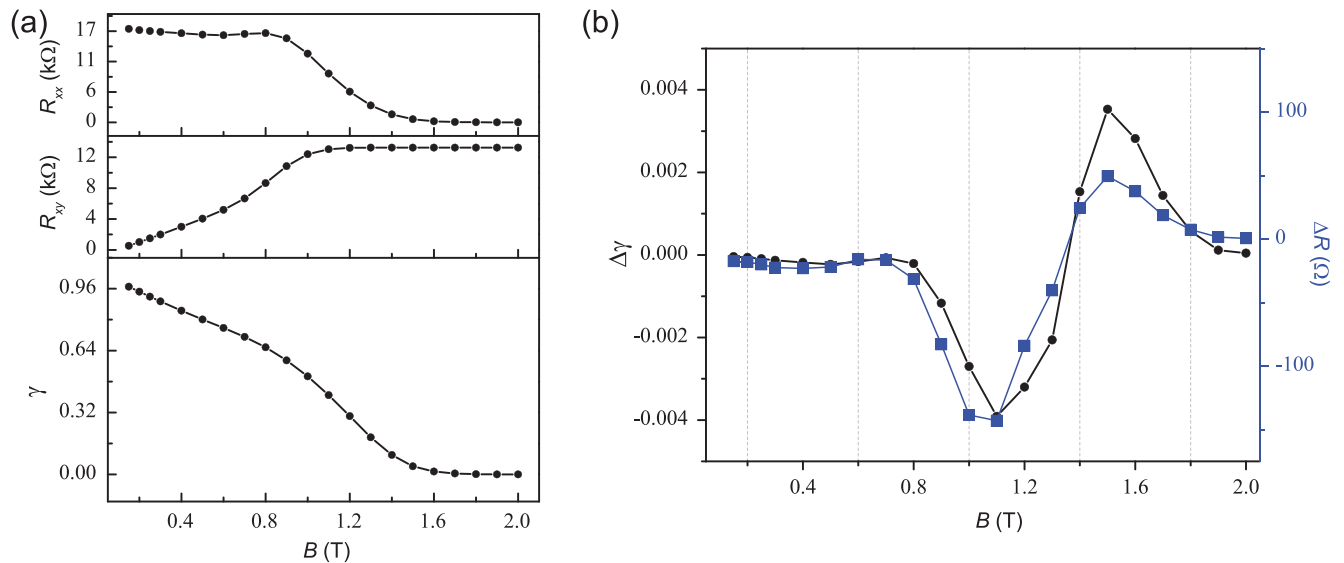


FIG. 8. (a) An example set of longitudinal and Hall resistance measurements (top and middle panel, respectively) are shown to compare with the calculated backscattering strength as a function of  $B$  field (bottom panel) for a 12.9 k $\Omega$  device. (b) The difference of the backscattering strength parameters is shown as a function of  $B$  field (black circles, left vertical axis), with the corresponding observed  $\Delta R$  shown as blue squares (right vertical axis).

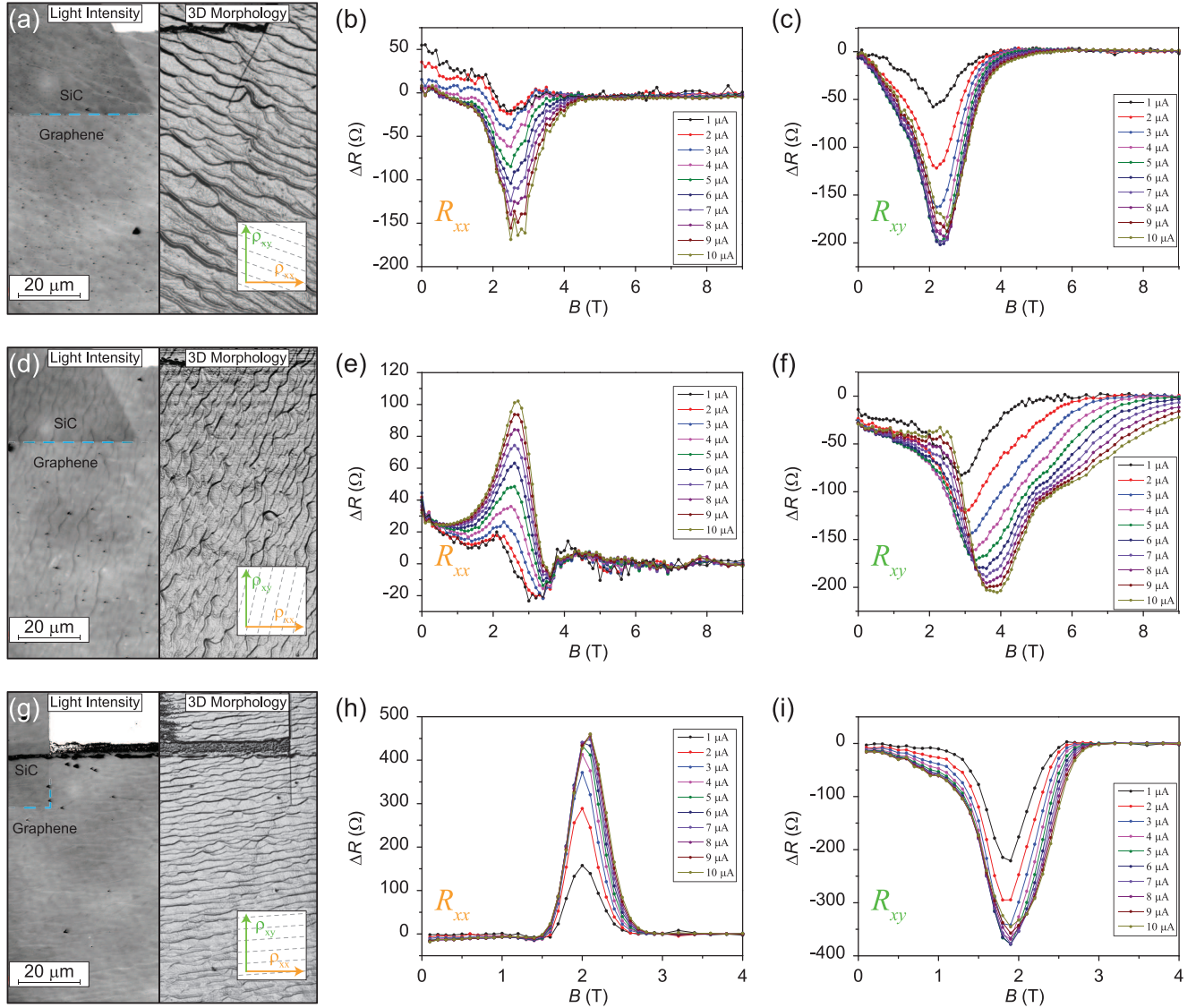


FIG. 9. (a), (d), and (g) Confocal laser scanning microscopy images of three orientations of step edge on three single epitaxial graphene (EG) devices are shown, with an approximate artistic rendering of the directionality of the step edges shown in the lower right inset. The left and right halves of each panel show the light intensity and three-dimensional morphology images, respectively. (b), (e), (h) Differences of the longitudinal resistances are shown as a function of perpendicular magnetic field at different current levels, with variable magnitudes and sign for the step orientations depicted to their left. (c), (f), (i) Similar data are shown, but instead for the Hall resistances.  $\Delta R$  increases in magnitude in all cases as the injection current is increased; however, this effect saturates at higher current for some cases, as shown in (c), (f), (h), and (i).

369 of resistances may not be feasible approaches for assessing  
 370 anisotropies. However, because one may assume that most  
 371 backscattering takes place at the SiC steps, one can expect  
 372 an influence from the step edge orientation on the mea-  
 373 sured anisotropy. Three single Hall bar devices are shown in  
 374 Figs. 9(a), 9(d), and 9(g), with different step orientations (with  
 375 orientations illustrated in the lower right insets). Differences  
 376 for the longitudinal resistances are shown in Figs. 9(b), 9(e),  
 377 and 9(h) as a function of  $B$  field for varying current levels,  
 378 with the corresponding Hall  $\Delta R$  shown in Figs. 9(c), 9(f), and  
 379 9(i).

380 Backscattering anisotropy appears to increase as Hall  
 381 quantization develops, implying that the direction of the cur-  
 382 rent (and its angle to the step edges) plays a significant role  
 383 in the measured OCR asymmetries. For instance, by looking

384 at the step edges in Fig. 9(a) (nearly  $45^\circ$ ),  $\Delta R$  for  $R_{xx}$  and  $R_{xy}$   
 385 have a similar response with  $B$  field (the incident angle for  $B+$   
 386 and  $B-$  is nearly identical). Furthermore, when different sets  
 387 of contact pads were used, there was a small impact on this  
 388 relationship, and the reciprocity differences for  $R_{xx}$  and  $R_{xy}$   
 389 were nearly identical (within 10% of one another).

390 In Figs. 9(d) and 9(g), the step edges are nearly perpendicu-  
 391 lar or parallel, respectively, to the long axis of the devices, and  
 392  $\Delta R$  for  $R_{xx}$  and  $R_{xy}$  have completely different characteristics  
 393 when compared with one another. The most obvious diver-  
 394 gence is in the signs, where the perpendicular step edge case  
 395 has both maxima and minima in the field dependence. Since  
 396 the backscattering process is not the same when the electrons  
 397 sample other regions, comparing data for other contact pads  
 398 was not as fruitful as for the first case. Regardless, it remains

399 evident that  $R_{xx}$  and  $R_{xy}$  are sensitive to the step edge orienta- 419  
 400 tion [40]. Furthermore,  $\Delta R$  undergoes drastic increases with 420  
 401 rising current, particularly for  $R_{xy}$ , but this begins to saturate 421  
 402 at relatively low current level, as seen in other related work 422  
 403 [45]. 423

404 Given that our observations and parametric tests on these 424  
 405 devices are yielding consistent results with other work, our 425  
 406 results suggest that the use of the OCR is not always a reliable 426  
 407 guide to the quality of electrically linear systems and EG 427  
 408 devices in particular. For devices that have inherent surface 428  
 409 morphology differences or anisotropic electrical properties, 429  
 410 additional or alternative tests are warranted. 430

411 **V. CONCLUSIONS**

412 In this paper, we determined a probable cause of OCR 431  
 413 nonreciprocity in three types of quantum Hall devices. After 432  
 414 verifying the functionality of each device and eliminating 433  
 415 many possible sources of asymmetry, it is confirmed that 434  
 416 substrate-induced morphology directly affects the current flow 435  
 417 by inducing electron density variation and, by extension, re- 436  
 418 sults in electrons experiencing nonuniform magnetic fields. 437

This leads to backscattering, whose presence ultimately in- 419  
 duces a device-dependent asymmetry in the quantum Hall 420  
 transitions. This asymmetry renders the OCR limited in their 421  
 capacity to accurately characterize the Hall and longitudinal 422  
 resistances of these devices. Therefore, these observations 423  
 may be useful in any experiment relying on the broader On- 424  
 sager relations because careful assessment of the current flow 425  
 is required. 426

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