

Graphene, universality of the quantum Hall effect and redefinition of the SI system

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New Journal of Physics **13** (2011) 093026 (6pp)

Received 13 June 2011

Published 16 September 2011

Online at <http://www.njp.org/>

doi:10.1088/1367-2630/13/9/093026

Abstract. The *Système Internationale d'unités* (SI) is about to undergo its biggest change in half a century by redefining the units for mass and current in terms of the fundamental constants h and e , respectively. This change crucially relies on the exactness of the relationships that link these constants to measurable quantities. Here we report the first direct comparison of the integer quantum Hall effect (QHE) in epitaxial graphene with that in GaAs/AlGaAs heterostructures. We find no difference in the quantized resistance value within the relative standard uncertainty of our measurement of 8.6×10^{-11} , this being the most stringent test of the universality of the QHE in terms of material independence.

The new quantum *Système Internationale d'unités* (SI) units for mass and current will be based on the fundamental constants of nature: Planck's constant, h , and the electron charge, e . Confidence in the new definition relies mainly on the ability to confirm experimentally the exactness of the relationships that link these constants to measurable quantities. The quantum Hall effect (QHE) defines one such relationship through the theoretical argument that the Hall resistance is quantized in units of h/Ne^2 , where N is an integer. The QHE

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is a fascinating macroscopic quantum effect occurring in two-dimensional (2D) conductors that has become one of the cornerstones of the worldwide reference system for scientific and industrial measurements [1]. However, the hypothesis of resistance quantization units of h/Ne^2 and its independence of material implementation has to be tested experimentally. The appearance of an unusual half-integer variation of the QHE in graphene [2, 3] confirmed the unique electrical properties of this 2D carbon material, where the charge carriers behave as massless Dirac fermions. As well as providing an experimental system for the study of new transport physics, graphene offers the prospect of a more robust implementation of the QHE resistance standard [4].

We report in this paper the result of a highest-precision direct comparison of the quantized resistance, $R = h/2e^2$, realized in an epitaxial graphene QHE sample with the matching $N = 2$ plateau of the QHE in a traditional GaAs/AlGaAs heterostructure device. Demonstrating the equivalence of this resistance in different devices is a vital step in proving the suitability of graphene for metrological use, but is also a useful test of the theory that predicts no corrections to the simple relation $R = h/Ne^2$. The quantum Hall resistance is considered to be a topological invariant, not altered by electron–electron interaction, spin–orbit coupling or hyperfine interaction with nuclei and insensitive to the much more subtle influences of gravity [5]. Recently, a quantum electrodynamical correction to the von Klitzing constant of the order of 10^{-20} was predicted for practical magnetic field values [6]. Because of the fundamental nature of the Hall resistance quantization, experimental tests of its universality are of the utmost importance, especially for improving our knowledge of two fundamental quantities of nature: the electron charge and Planck’s constant. The precision obtained through a universality test as presented here is much greater than what is possible by a comparison to the values of constants h and e [7]. Analysis of the complete set of published results carried out by CODATA [7] showed no deviation from h/e^2 to within 2×10^{-8} , which calls for more accurate measurements.

Soon after the first observations of the QHE in graphene [2, 3], Giesbers *et al* [8] reported an evaluation of the accuracy of the resistance quantization in exfoliated graphene flakes. Unfortunately, the small size of the flakes and electrical contacts, along with the low breakdown current in their devices, made these measurements very difficult. An accuracy of only a few parts per million could be obtained (four orders of magnitude below the state of the art in GaAs and Si) and hence no meaningful conclusions on the universality of the QHE could be drawn. Our own previous work [9] reported the first accurate observation of the QHE in large epitaxial graphene devices. We achieved an accuracy of 3 parts in 10^9 via an indirect method whereby both quantum Hall devices were measured separately against a room temperature standard resistor. Recently, we reported [10] an unusually strong pinning of the $\nu = 2$ quantum Hall state in epitaxial graphene due to charge exchange with the localized states in the substrate, resulting in a very robust resistance quantization, and we demonstrated invariance of the resistance quantization to 0.3 part in 10^9 over a field range of 3.5 T. Importantly for precision metrology, the extraordinarily robust quantum Hall state in these devices sustains very high non-dissipative currents, ensuring a large signal-to-noise ratio.

Our graphene sample was produced by epitaxial growth on a SiC substrate [9] and shows the properties (such as low contact resistance and negligible longitudinal resistivity) required for accurate metrological use. Its resistance was compared to that of the GaAs device in a null measurement using the standard methods of resistance metrology. (The four-terminal nature of QHE resistors means that some form of bridge circuit is needed, even to compare identical resistors; here a cryogenic current comparator (CCC) [11] was used to establish an exact 1 : 1

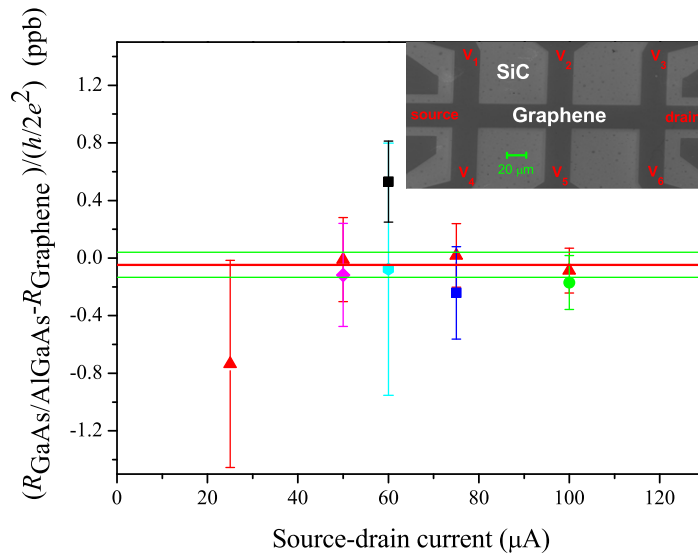


Figure 1. The measured difference between a GaAs/AlGaAs sample and a graphene sample as a function of the source–drain current through the devices for different measurement configurations. Red triangles: GaAs/AlGaAs device 1 in system 1 at 1.5 K—graphene in system 2 at 300 mK; green dot: measured using non-opposite voltage contacts on GaAs/AlGaAs device 1; blue square: measured using non-opposite contacts on a graphene device; pink diamond: GaAs/AlGaAs device 2—graphene. The light blue hexagon was measured in reverse magnetic field for graphene. The black square represents samples exchanged between systems 1 and 2. The red line is the weighted mean of all the data points and the green lines signify ± 1 standard deviation. Inset: SEM picture of the graphene device.

current ratio.) A summary of the results is shown in figure 1 (for details, see the methods section).

The weighted average of all our data is $(R_{\text{GaAs/AlGaAs}} - R_{\text{Graphene}})/(h/2e^2) = (-4.7 \pm 8.6) \times 10^{-11}$. The relative standard uncertainty of 8.6×10^{-11} represents a factor of 35 improvement on our previous result obtained via an indirect measurement [9, 10]. In an indirect measurement, the accuracy is limited by the properties of the resistor used as a transfer standard⁷. Here we directly compare both devices against each other, thereby eliminating many systematic effects. Previously, our knowledge of the universality of the QHE was limited to the level of 2 or 3×10^{-10} for comparison between GaAs and Si or between identical GaAs devices [1]. However, both GaAs and Si are traditional semiconductors with a parabolic bandstructure and governed by the same physics. Graphene is a semimetal with a linear bandstructure and is described by Dirac-type massless charge carriers and so universality in terms of material independence goes well beyond the comparison between two semiconductors. In our universality experiment, the maximum source–drain current that the GaAs device can sustain without dissipation limits the measurement uncertainty, whereas a potentially lower uncertainty can be obtained in a consistency check of two graphene devices.

⁷ Note an important distinction between the precision of the measurement and the accuracy of the result. Precision is used to define the measurement repeatability, whereas accuracy expresses how close the measured value is to the true value (International Vocabulary of Metrology, <http://www.bipm.org/en/publications/guides/vim.html>).

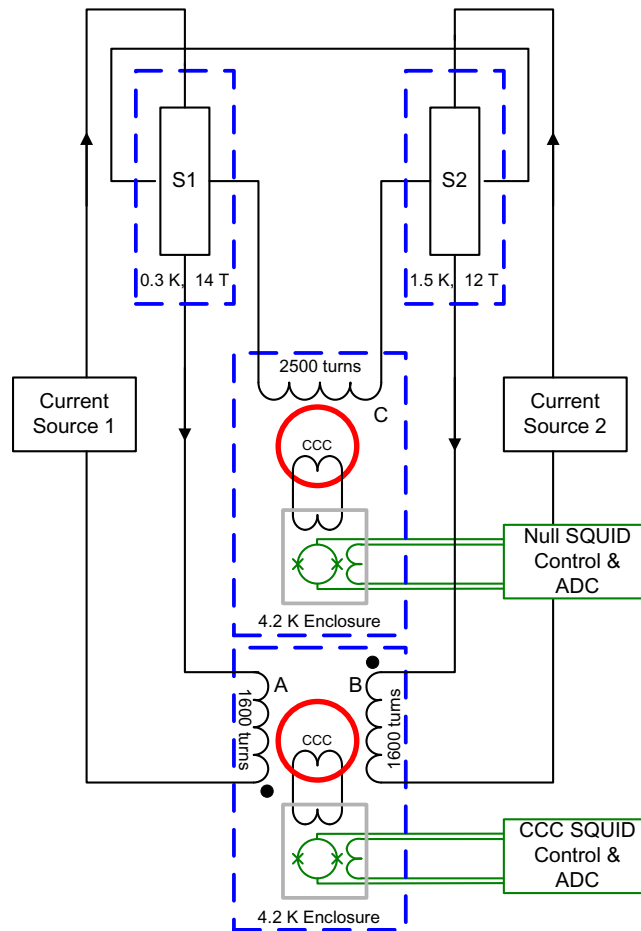


Figure 2. Simplified schematic diagram of the CCC bridge circuit.

Our results on material independence are the strongest evidence so far for the hypothesis that the resistance is quantized in units of h/Ne^2 , and thereby support the pending redefinition of the SI units for kilogram and ampere in terms of Planck's constant and the electron charge⁸. Judging from the robustness of the quantization and wide operational parameter space, epitaxial graphene should be the material of choice for quantum resistance metrology.

Methods

The epitaxial graphene sample used in the reported experiment was produced on the Si face of SiC [9]. The graphene Hall bar was encapsulated in a polymer bilayer, a spacer polymer followed by an active polymer able to generate acceptor levels under UV light. More fabrication details can be found elsewhere [12]. The sample had an electron density, n_S , of $4.6 \times 10^{11} \text{ cm}^{-2}$ and mobility, μ , of $7500 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. Note that this mobility is rather low compared with that achieved in exfoliated or suspended graphene and is much lower than that obtained in the best GaAs. Fortunately, in the QHE, disorder is in fact necessary to provide localization of the

⁸ See http://www.bipm.org/en/si/new_si/ for more information.

electron states, and for precision metrology the mobility should not be too high in order to provide a wide quantum Hall plateau. A standard eight-contact Hall bar geometry was patterned on the device with dimensions $160\ \mu\text{m} \times 35\ \mu\text{m}$. The graphene sample was placed in system 1 at 300 mK and 14 T. The two GaAs samples used were traditional GaAs/AlGaAs heterostructures obtained from the PTB (device 1) and LEP (device 2). Device 1 had $n_s = 4.6 \times 10^{11}\ \text{cm}^{-2}$ and $\mu = 4 \times 10^5\ \text{cm}^2\ \text{V}^{-1}\ \text{s}^{-1}$, the size of the chip was $6000\ \mu\text{m} \times 2500\ \mu\text{m}$ and contacts were made from small tin balls at the edge of the chip. Device 2 had $n_s = 5.1 \times 10^{11}\ \text{cm}^{-2}$, $\mu = 5 \times 10^5\ \text{cm}^2\ \text{V}^{-1}\ \text{s}^{-1}$; the chip had an etched Hall-bar geometry of $2200\ \mu\text{m} \times 400\ \mu\text{m}$ and AuNiGe alloyed contacts. Both GaAs devices were placed in system 2 at 1.5 K and either 9.5 T (device 1) or 10.5 T (device 2). Before commencing the high-accuracy measurements all devices were fully characterized according to the guidelines on quantum Hall resistance metrology [1] (i.e. we confirmed that the three-terminal contact resistance measured on the $N = 2$ plateau was of the order of a few ohms for all the contacts used and that the longitudinal resistivity at the measurement current was below $10\ \mu\Omega$). For the graphene device the maximum source–drain current, I_C , at which the device remains in the non-dissipative state was approximately $500\ \mu\text{A}$. For the GaAs devices I_C was $\approx 150\ \mu\text{A}$ for device 1 and $\approx 100\ \mu\text{A}$ for device 2.

The measurements were made with a CCC bridge [11], illustrated in a simplified form in figure 2. Isolated current sources 1 and 2 separately drive the current through samples S1 and S2 and associated windings A and B on the CCC. The current ratio can be set via electronics to a few parts in 10^6 and this ratio is improved to a level of 1 part in 10^{11} by forming a negative feedback loop from the superconducting quantum interference device (SQUID) sensing the net flux in the CCC to one of the current sources. The potential contacts on S1 and S2 are closed in a loop via winding C on a second CCC. This device is configured with just a single winding to measure a current null rather than two windings to establish a current ratio. The data are collected alternately in the forward and reverse current directions so as to eliminate electrical offsets. Measurement uncertainty arises from leakage currents in the connecting cables, residual error in the A/B ratio, accuracy of the negative feedback loop and random noise. The random noise of 8.6 parts per 10^{11} dominates over the other components, estimated to have a combined standard uncertainty of 1.6 parts in 10^{11} .

Acknowledgments

The authors thank Stephen Giblin and Dale Henderson for their assistance. The work was supported by the UK National Measurement Office (NMO) Pathfinder Programme, Swedish Research Council and Foundation for Strategic Research, EU FP7 STREPs ConceptGraphene and SINGLE, EPSRC grant EP/G041954 and the Science & Innovation Award EP/G014787.

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