Video Article

Advanced Experimental Methods for Low-temperature Magnetotransport Measurement of Novel Materials

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URL: https://www.jove.com/video/53506
DOI: doi:10.3791/53506

Keywords: Engineering, Issue 107, Nanoelectronics, Nanotechnology, Nano-fabrication, Electron beam lithography, Magnetotransport, Two-dimensional materials, Graphene, Electronic materials

Date Published: 1/21/2016

Citation: Hagmann, J.A., Le, S.T., Richter, C.A., Seiler, D.G. Advanced Experimental Methods for Low-temperature Magnetotransport Measurement of Novel Materials. *J. Vis. Exp.* (107), e53506, doi:10.3791/53506 (2016).

Abstract

Novel electronic materials are often produced for the first time by synthesis processes that yield bulk crystals (in contrast to single crystal thin film synthesis) for the purpose of exploratory materials research. Certain materials pose a challenge wherein the traditional bulk Hall bar device fabrication method is insufficient to produce a measureable device for sample transport measurement, principally because the single crystal size is too small to attach wire leads to the sample in a Hall bar configuration. This can be, for example, because the first batch of a new material synthesized yields very small single crystals or because flakes of samples of one to very few monolayers are desired. In order to enable rapid characterization of materials that may be carried out in parallel with improvements to their growth methodology, a method of device fabrication for very small samples has been devised to permit the characterization of novel materials as soon as a preliminary batch has been produced. A slight variation of this methodology is applicable to producing devices using exfoliated samples of two-dimensional materials such as graphene, hexagonal boron nitride (hBN), and transition metal dichalcogenides (TMDs), as well as multilayer heterostructures of such materials. Here we present detailed protocols for the experimental device fabrication of fragments and flakes of novel materials with micron-sized dimensions onto substrate and subsequent measurement in a commercial superconducting magnet, dry helium close-cycle cryostat magnetotransport system at temperatures down to 0.300 K and magnetic fields up to 12 T.

Video Link

The video component of this article can be found at https://www.jove.com/video/53506/

Introduction

The pursuit of materials platforms for advanced electronics technology demands methods for high-throughput materials synthesis and subsequent characterization. Novel materials of interest in this pursuit may be produced in bulk by direct reaction synthesis[1,2], electrochemical growth[3,4], and other methods[5] in a more rapid fashion than more involved single crystal thin film deposition techniques such as molecular beam epitaxy or chemical vapor deposition. The conventional method to measure the transport properties of bulk crystal samples is to cleave a rectangular prism-shaped fragment with dimensions of approximately 1 mm x 1 mm x 6 mm and attach wire leads to the sample in a Hall bar configuration[6].

Certain materials pose a challenge wherein the traditional bulk Hall bar device fabrication method is insufficient to produce a measureable device for sample transport measurement. This can be because the crystals produced are too small to attach lead wires to, even under a powerful optical microscope, because the desired sample thickness is on the order of one to only a few monolayers, or because one aims to measure a stack of layered two-dimensional materials with near- or sub-nanometer thickness. The first category consists of, for example nanowires and certain preparations of molybdenum oxide bronzes[7]. The second category consists of single to very-few layers of two-dimensional materials such as graphene[8], TMDs (MoS_2, WTe_2, *etc.*), and topological insulators (Bi_2Se_3, $Bi_xSb_{1-x}Te_3$, *etc.*). The third category consists of heterostructures prepared by stacking individual layers of two-dimensional materials by manual assembly via layer transfer, most notably a trilayer stack of hBN-graphene-hBN[9].

Exploratory research of novel electronic materials demands adequate methods for producing devices on difficult-to-measure samples. Often, the first batch of a new material synthesized by direct reaction or electrochemical growth yields very small single crystals with dimensions on the size order of microns. Such samples have historically proven enormously difficult to attach metal contacts to, necessitating improvement of sample growth parameters to achieve larger crystals for easier transport device fabrication, presenting an obstacle in the high-throughput research of novel materials. In order to enable rapid characterization of materials, a method of device fabrication for very small samples has been devised to permit the characterization of novel materials as soon as a preliminary batch has been produced. A slight variation of this methodology is applicable to producing devices using exfoliated samples of two-dimensional materials such as graphene, hBN, and TMDs, as well as multilayer heterostructures of such materials. Devices are adhered and wire-bonded to a package for insertion into a commercial superconducting magnet, dry helium close-cycle cryostat magnetotransport system. Transport measurements are taken at temperatures down to 0.300 K and magnetic fields up to 12 T.

Protocol

1. Preparation of Substrate

1. Obtain 4 inch silicon (Si) wafer composed of heavily-doped p-doped Si covered by approximately 300 nm of SiO_2. This substrate structure will allow the substrate to serve as a back gate.
2. Using drafting/design software, design a 1 cm × 1 cm pattern with evenly spaced features, such as enumerated crosses, in the x and y direction to use as positional locators on the substrate for transferred sample flakes and alignment marks for electron beam lithography (**Figure 1**).
    1. Open a new file in a drafting program such as AutoCAD.
    2. Use polylines to draw the following marks: i) crosses formed of intersecting 2.25 µm × 12.00 µm rectangles labeled with 8.25 µm × 14.25 µm numeric characters spaced 150 µm apart; ii) global positional identifiers formed of corner connected 30 µm squares labeled with 16.50 × 28.50 µm numeric characters; iii) small 15 µm × 15 µm crosses equidistant from any given four positional identifiers spaced 150 µm apart.
    3. Ensure that all positional identifiers fit within 1 cm × 1 cm limits.
    4. Save the file as a .gds file or save as another file (such as .dxf) and convert it to a .gds file.

3. Develop or order from a commercial source a photomask with the above 1 cm × 1 cm pattern arrayed across a 4 inch × 4 inch space to allow full coverage of copies of the pattern on a 4 inch Si wafer.
4. Lithographically pattern photoresist mask on Si wafer.
    1. Attach a 4 inch wafer chuck to a photoresist spinner. Ensure it is adequately fastened.
    2. Using clean tweezers, place a Si wafer on the spinner chuck. Ensure that the wafer is centered on the chuck.
    3. Using a plastic pipette, cover the entire wafer in polydimethylglutarimide-based lift-off resist (LOR).
    4. Spin the LOR-coated wafer at 4,000 rpm for 45 sec.
    5. Bake the LOR-coated wafer at 170 °C for 5 min.
    6. Let the LOR-coated wafer cool for 1-2 min.
    7. Using clean tweezers, place the LOR-coated Si wafer on the spinner chuck. Ensure that the wafer is centered on the chuck.
    8. Using a plastic pipette, cover the entire wafer in conventional Novolac positive photoresist designed for a 60 sec bake at 115 °C to produce a 12.3 kÅ coat.
    9. Spin the wafer at 5,000 rpm for 60 sec.
    10. Bake the wafer at 90-110 °C for 60 sec.
    11. Let the wafer cool for 1-2 min.
    12. Prepare for contact lithography by placing the photomask in a mask aligner with the patterned chrome of the mask facing down (towards the wafer) and loading the double-layer-photoresist-coated wafer beneath the mask so that the photoresist faces up (towards the mask).
    13. Align the wafer to the mask so that the entirety of the wafer will be patterned with the arrayed 1 cm × 1 cm patterns.
    14. Expose in UV-broadband (350 nm to 450 nm) light using the I-line (365 nm band-pass filter) at 100 mJ/cm^2, or 20 mW/cm^2 for 4.8 sec.
    15. Following exposure, develop the photoresist coating the wafer by immersing it in a conventional Novolac positive photoresist-compatible developer solution at RT for 40-60 sec with mild and consistent agitation.
    16. Following immersion in the bath of developer solution, rinse the wafer in deionized water.
    17. Using a nitrogen gun, blow-dry the developed photoresist mask-coated wafer.

5. Deposit Cr/Au metal onto the photoresist mask-coated wafer using an electron beam evaporator.
    1. Vent electron beam evaporator chamber.
    2. Place wafer face down on sample-holder plate.
    3. Open electron beam evaporator chamber door.
    4. Place sample-holder plate into substrate holder with wafer facing down.
    5. Check that Cr and Au are among the deposition sources.
    6. Close the electron beam evaporator chamber door and pump the chamber to at least 4×10^{-5} Pa (3×10^{-7} Torr).
    7. Deposit 50 Å of Cr at 0.5 Å/sec.
    8. Deposit 750 Å of Au at 1 Å/sec.
    9. Following deposition, allow chamber to cool for about 20 min.
    10. Vent electron beam evaporator chamber.
    11. Remove sample-holder plate and remove the wafer from the plate.
    12. Close the electron beam evaporator chamber door and pump the chamber.

6. Perform a metal liftoff.
    1. Prepare a bath of acetone or *N*-methyl-2-pyrrolidone-based solvent sufficient to immerse the 4 inch wafer. Heat the solvent bath to 75 °C on a hot plate and hold at this temperature.
    2. Immerse the 4 inch wafer in the solvent bath. Cover the bath so that the solvent does not evaporate excessively.
    3. Let the wafer sit in the solvent bath at 75 °C for 6-24 hr. Be careful not to let all of the solvent evaporate.
    4. Hold sample just below the surface of the solvent using a pair of tweezers and delicately spray acetone from a spray bottle across the surface of the wafer to remove lifted-off metal.
    5. Rinse the wafer in a bath of isopropanol for 1-2 min.
    6. Rinse the wafer in a bath of deionized water for 1-2 min.

    7. Using a nitrogen gun, blow-dry the wafer.

7. Dice the wafer into individual sample pieces using a diamond scribe or a dicing saw. If a dicing saw is used, protect the wafer surface with a PMMA mask.
    1. Attach a 4 inch wafer chuck to a photoresist spinner. Ensure it is adequately fastened.
    2. Using clean tweezers, place the wafer on the spinner chuck. Ensure that the wafer is centered on the chuck.
    3. Using a plastic pipette, cover the entire wafer in polymethyl methacrylate (PMMA).
    4. Spin the PMMA-coated wafer at 5,000 rpm for 120 sec.
    5. Bake the PMMA-coated wafer at 180 °C for 120 sec.
    6. Let the PMMA-coated wafer cool for 1-2 min.
    7. Dice the sample into individual sample pieces approximately 1 cm × 1 cm in size.

8. Remove organic residue from wafer surface.
    1. Prepare a bath of a sulfuric acid and hydrogen peroxide-based stripper/organic cleaner, a bath of acetone, a bath of isopropanol, and two baths of deionized water.
    2. Let the substrate pieces soak in the acetone bath for 15 min with agitation by high-frequency sonication.
    3. Move the substrate pieces to the isopropanol bath and let it soak for 15 min with agitation by high-frequency sonication.
    4. Move the substrate pieces to a deionized water bath and let it soak for 15 min with agitation by high-frequency sonication.
    5. Move the substrate pieces to the other deionized water bath and let it soak for 15 min with agitation by high-frequency sonication.
    6. Move the substrate pieces to the sulfuric acid and hydrogen peroxide-based stripper/organic cleaner bath and let it soak for 60 min with no agitation.
    7. While the substrate pieces are in the organic cleaner bath, properly dispose the contents of the other baths and clean the glassware. Prepare two new deionized water baths.
    8. Following organic stripping, place the substrate pieces in a deionized water bath and let it soak for 5 min with agitation by high-frequency sonication.
    9. Move the substrate pieces to the other deionized water bath and let it soak for 5 min with agitation by high-frequency sonication.
    10. Using a nitrogen gun, blow-dry the substrate pieces.

2. Transferring Sample Flakes to Substrate

1. Synthesize or obtain high-quality bulk sample from a collaborator or commercial source.
2. Exfoliate sample flakes.
    1. Cut several pieces of standard wafer dicing tape slightly-larger-than 1 cm x 3 cm, leaving the release paper covering the adhesive.
    2. Using a sharp razor, carefully remove a portion of the release paper so that slightly more than 1 cm x 1 cm of adhesive is exposed on each piece of tape.
    3. Firmly press the adhesive portion of one prepared piece of tape against the bulk sample. If bulk sample is composed of very small powder-like pieces, pour a small amount of sample onto a glass slide and press the tape into the amassed sample on the slide.
    4. Peel the tape from the bulk sample ensuring good coverage of sample on the adhesive.
    5. Press the adhesive side of the tape with sample flakes very firmly against the adhesive side of another piece of tape.
    6. Peel the two pieces of tape apart and visually inspect sample coverage on both pieces of tape.
    7. Repeat the process 2.2.5 and 2.2.6 until sample flakes appear nearly transparent.
    8. Firmly press the adhesive side of the tape with sample flakes against a piece of prepared substrate from step 1. Peel the tape away to leave sample flakes adhered to the substrate.

3. Visually search for suitable sample flakes using optical microscope (**Figure 2**) and note their location on substrate utilizing the positional marks patterned in steps 1.2-1.4.
4. Measure sample flake thickness using atomic force microscopy (AFM). Sample flakes should be less than 100 nm in thickness[10].
5. Deposit sputtered silicon dioxide (optional).
   Note: This step is only necessary if the van der Waals force adhering the sample to the substrate is insufficient for adequate adhesion. Doing this disallows the fabrication of the sample into a Hall bar by the procedure outlined in this paper (step 3.3).
    1. Vent sputtering system load lock.
    2. Open load lock door.
    3. Place sample pieces accommodating transferred sample flakes onto sample holder and place sample holder on the transfer arm.
    4. Close the load lock door and pump down the load lock.
    5. Initiate the sequence to transfer the sample holder to the sputtering chamber.
    6. Wait for base vacuum of 2.7×10^{-5} Pa (2×10^{-7}).
    7. Using a DC power supply, sputter 100-200 nm of SiO_2 onto the sample pieces accommodating transferred sample flakes.
    8. Following deposition, initiate the sequence to return the sample holder to the load lock.
    9. Vent the load lock and remove the sample holder.
    10. Remove the pieces from the sample holder.
    11. Pump down the sputtering system load lock.

6. Prepare a stack of flakes of layered materials
   Note: This step is only necessary if the researcher desires to produce a heterostructure composed of multiple flakes exfoliated and identified in steps 2.1-2.4.
    1. Create a transparent mechanical stamp by placing a small drop of Polydimethylsiloxane (PDMS) on a glass slide and cure in vacuum.
    2. Spin Polypropylene carbonate (PPC) on top of the PDMS to serve as the direct contact between the stamp and the layered materials.
    3. Position the mechanical stamp over the first flake of layered material to be used in the heterostructure stack.
    4. Press the stamp down on the sample flake.

    5. Heat the system to approximately 40 °C to increase attraction between PPC and the sample flake.
    6. Slowly lift the stamp up with the sample flake attached to the PPC.
    7. Position the mechanical stamp with the attached sample flake over the next flake of layered material to be used in the heterostructure stack.
    8. Making sure to keep the two sample flakes aligned, slowly lower the stamp so that the flake attached to the PPC comes into contact with the next flake of layered material to be used in the heterostructure stack.
    9. Lightly press the stamp down on the sample flake.
    10. Heat the system to approximately 40 °C to increase attraction between the sample flakes.
    11. Slowly lift the stamp up with the sample flakes attached in a stack.
    12. Repeat steps 2.6.7-2.6.11 until the desired structure is completed.
    13. Transfer the heterostructure stack to a new substrate by gently pressing the stamp containing the stack of layered materials to the substrate piece.
    14. Heat the system to 100 °C.
    15. Slowly lift the stamp up, leaving the stack of flakes of layered materials attached to the substrate piece.

3. Electron Beam Lithography of Device Structure

1. Using an optical microscope, take well-focused pictures at magnifications of 20X and 100X of the sample flakes/flake stacks that will be used for device patterning. Include at least one positional mark patterned in steps 1.2-1.4 in the image for alignment purposes during electron beam pattern design.
2. Using drafting/design software, prepare design for electron beam lithography.
    1. Open design file produced in step 1.2.
    2. Import 20X image from step 3.1 and adjust the image size accordingly to scale properly with the design.
    3. Align the image to the positional marks in the design corresponding to where the sample flake is located on the patterned substrate piece.
    4. Repeat steps 3.2.2 and 3.2.3 using the 100X image.
    5. Create a new layer in the design program and draw a 6-terminal Hall bar pattern over the sample image such that the exposed region will be etched away, leaving the Hall bar.
    6. Create another new layer in the design program for electrical metal contacts leading from the terminals on the sample to contact pads.
    7. Save the file to a .gds file or save it in another format and convert it to a .gds file.

3. Pattern 6-terminal Hall bar PMMA mask (skip this step if a SiO_2 coating layer was deposited in step 2.5).
    1. Spin a layer of PMMA formulated with a molecular weight of 950,000 onto the sample according to the procedure outlined in steps 1.7.1 to 1.7.6.
    2. Load sample into electron beam lithography system.
    3. Using scanning electron microscopy (SEM), locate alignment marks on the substrate far from the sample[11].
    4. Calibrate the system for proper stage rotation and length scale according to the procedure specific to the electron beam lithography system being used.
    5. Turn off the electron beam to prevent unwanted exposure of the PMMA and center the beam position at the center of the pattern prepared in step 3.2.
    6. Load the .gds file onto the electron beam lithography system computer and program the system to print the Hall pattern layer from step 3.2.5 with the desired resolution according to the procedure specific to the electron beam lithography system being used.
    7. Execute the electron beam system's patterning program to expose the PMMA to the electron beam according to the system's user manual.
    8. Remove the sample from the electron beam system.

4. Etch sample into 6-terminal Hall bar (skip this step if a SiO_2 coating layer was deposited in step 2.5).
    1. Vent reactive ion etching system.
    2. Load sample into etching chamber.
    3. Pump system to approximately 1.3×10^{-3} Pa (1×10^{-5} Torr).
    4. Utilize etch recipe specific to the sample material to etch sample. Note: For hBN/graphene/hBN stacks, a plasma generated from 4 standard cubic centimeters (sccm) O_2 and 40 sccm CHF_3 at 60 W radio frequency (RF) power has an etch rate of about 30 nm/min; a 1-2 min etch is usually sufficient.
    5. Upon completion of the etching process, vent the etching chamber.
    6. Unload sample and pump etching chamber.
    7. Rinse the sample in a bath of acetone for 1-2 min.
    8. Rinse the sample in a bath of isopropanol for 1-2 min.
    9. Rinse the sample in a bath of deionized water for 1-2 min.

5. Pattern PMMA mask for deposition of metal contacts.
    1. Spin a layer of PMMA formulated with a molecular weight of 495,000 onto the sample according to the procedure outlined in steps 1.7.1 to 1.7.6.
    2. Spin a second layer of PMMA formulated with a molecular weight of 950,000 onto the sample according to the procedure outlined in steps 1.7.1 to 1.7.6.
    3. Repeat steps 3.3.2-3.3.8, this time utilizing the contact pattern layer from step 3.2.6.

6. If a SiO_2 coating layer was deposited in step 2.5, etch away SiO_2 in the region exposed by the mask to allow electrical contacts to interface directly with the sample flake.
    1. Hard bake the mask at 180 °C for 5 min.

    2. Let the sample cool for 1-2 min.
    3. Etch the exposed SiO_2 in a 2% HF bath for a sufficient period to expose the sample flake without damaging the PMMA mask — approximately 1-2 min for 100 nm SiO_2, visually checking the progress of the etch every 10-20 sec to make sure the PMMA remains undamaged.
    4. If PMMA mask is damaged, remove the PMMA by rinsing the sample in acetone for 60 sec, then isopropanol for 60 sec, then deionized water for 60 sec, blow-drying the sample using a nitrogen gun, then repeating step 3.5.

7. Deposit Cr/Au metal onto the sample using an electron beam evaporator by repeating step 1.5.
8. Perform a metal liftoff by repeating step 1.6.

4. Perform Magnetotransport Experiment

1. Prepare electrical transport package with fabricated sample by adhering sample to electrical transport system package to be loaded onto transport system probe tip with silver paste and let dry. Use a wire-bonder to connect a thin gold wire from the contact pads of the device to the contact pads of the package.
2. Load the sample into the magnetotransport system.
    1. Attach the package onto the probe tip to be inserted into the magnetotransport system and ensure that it is firmly attached. Connect electrical measurement devices (sourcemeters, lock in amplifiers, *etc.*) to probe and make connections to all three temperature control channels and electrical measurement channels.
    2. Vent the airlock and insert the probe tip into the airlock and lock it into place with a clamp and O-ring.
    Note: Steps 4.2.3 and 4.2.4-4.2.6 and 4.2.8 correspond to steps necessary to undertake transport measurements using a He-3 probe.
    3. Set the minisorb temperature in the insert to 330 K to remove water vapor and open the gas exchange valve and pump the system using a vacuum pump for about 30 min, until the pressure is below 0.67 Pa (5×10^{-3} Torr).
    4. Close the exchange valve and the airlock valve and open the valve separating the airlock space from the measurement space.
    5. Briefly open (for no more than 2 sec) and close the exchange valve to introduce a small amount of He-4 gas into the probe space.
    6. Set the minisorb temperature to 25 K and the mainsorb temperature to 40 K.
    7. Slowly lower the probe into the measurement space until the sample is at field center.
    8. Once the system has reached 2 K, press the 3He condensation sequence button in the controller software to achieve temperatures as low as 0.300 K.

3. Take transport measurements at a range of temperatures, magnetic fields, gate voltages, *etc.*
    1. For all measurements, simultaneously save, to the data file, the current supplied by the current source, the longitudinal voltage (parallel to the supplied current) measured by the voltmeter/lock-in amplifier dedicated to this measurement, the sample temperature measured by a temperature sensor located near the sample, and the magnetic field generated by the magnet.
    2. If Hall measurements are desired, save the Hall voltage (transverse to the supplied current) to the data file, and if a gate voltage is supplied to tune the carrier density in the channel, save the gate voltage supplied by the corresponding voltage source, as well, for each measurement at the desired sampling rate.
    Note: The sampling rate for the measurements depends on whether the changing experimental parameter (temperature, field, voltage, *etc.*) is being swept (starting at one valuing and ending at a set value with a constant rate of change) or stepped (stabilizing at predetermined values). In the former case, the sampling rate is up to the discretion of the researcher based on the size of the data file they wish to generate. In the latter case, measurements are taken upon stabilization of the changing experimental parameter. Generating and saving data files is handled by data acquisition software.

Representative Results

Figure 3 shows a typical Hall bar device patterned for the purpose of a low temperature magnetotransport experiment. The optical image in the upper figure shows a successfully-fabricated Graphene/hBN Hall bar; the lower image shows the device schematic with the Landauer-Büttiker edge states that arise from the Landau levels (LLs), a transport mechanism that can be used to calculate the values of the quantized Hall resistances, the experimental investigation of which will be discussed as a representative application of the experimental technique detailed in this paper. Often, fabrication of the Hall bar structure constitutes a tremendous challenge in the overall fabrication process. The steps involved in etching the sample into this shape may be skipped and leads may be attached directly to sample flakes as they are following transfer to the substrate piece. However, the imperfect geometry will not permit careful measurement of transport properties, so skipping the steps involved in etching the sample into a Hall bar structure should be constrained to initial measurements.

Experimental parameters include magnetic fields as high as 12 T, temperatures as low as 0.300 K, and gate voltages as high as 30 V. Alternating current may be supplied by the oscillator from a lock-in amplifier with associated lock-in AC voltage measurements, while direct current may be supplied by a sourcemeter with associated DC voltage measurements. Alternating current versus direct current and the magnitude of the current are parameters that must be carefully selected based on the properties, including resistance and breakdown characteristics, of the material being studied. The Hall resistance is defined as the potential difference between, or voltage measured across, leads 6 and 2 in **Figure 3** divided by the applied current. Longitudinal resistance is defined as the potential difference between, or voltage measured across, leads 2 and 3 divided by the applied current. A Graphene Hall bar with the top surface protected by a hBN flake fully encapsulating the Graphene was measured at 1.7 K at magnetic fields ranging from -6 T to +6 T and back gate voltages ranging from -30 V to +30 V. **Figure 4** shows how the Hall resistance changes within this parameter space. The behavior of the Hall resistance measured in the Graphene/hBN Hall bar, specifically the observable plateaus in the Hall resistance corresponding to Landau level filling, is a model example of the quantum Hall effect, a quantum mechanical phenomenon observable only by the application of such advanced magnetotransport technique as described in this paper.

Figure 5 shows a slice of the data presented in **Figure 4** at B = 6 T, with the Hall resistance (R_{xy}) as a function of back gate voltage and the corresponding longitudinal resistance (R_{xx}) as a function of back gate voltage. The measurement clearly shows the that graphene exhibits a quantum Hall effect with quantized Hall resistance values of with integer-value Landau level number *n*, Planck's constant *h*, and electron charge *e*. The quantized Hall resistance plateaus coincide with vanishing longitudinal resistance.

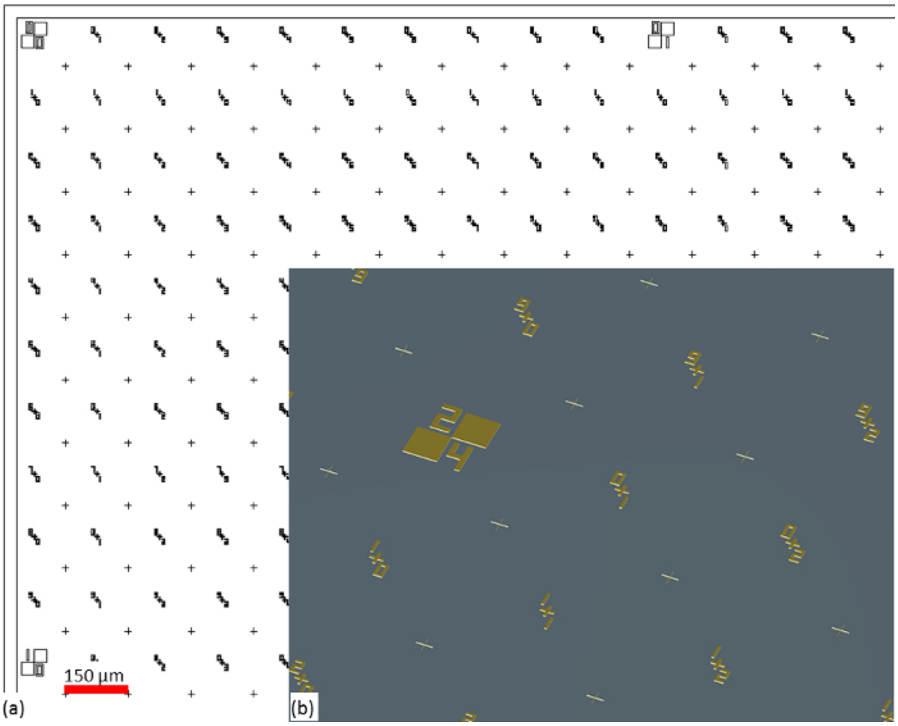

Figure 1. Substrate pattern. (A) a region of the designed configuration for the lithographically-patterned positional/alignment marks for the sample substrate. **(B)** computer rendering of the patterned gold positional/alignment marks. Please click here to view a larger version of this figure.

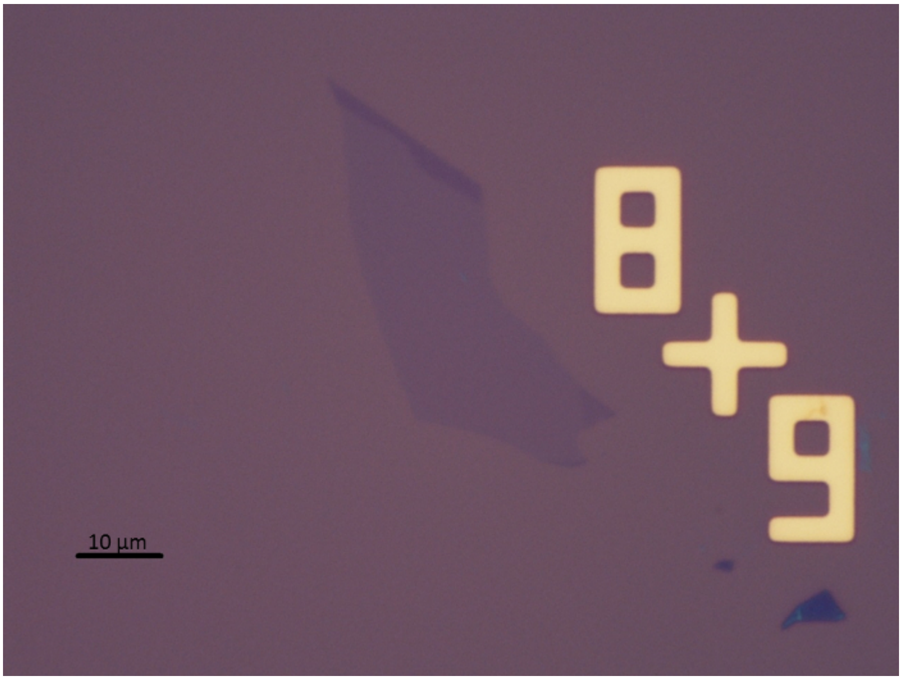

Figure 2. Graphene flake. An image of flake of monolayer graphene adjacent to a Cr/Au positional marker observed through an optical microscope. Please click here to view a larger version of this figure.

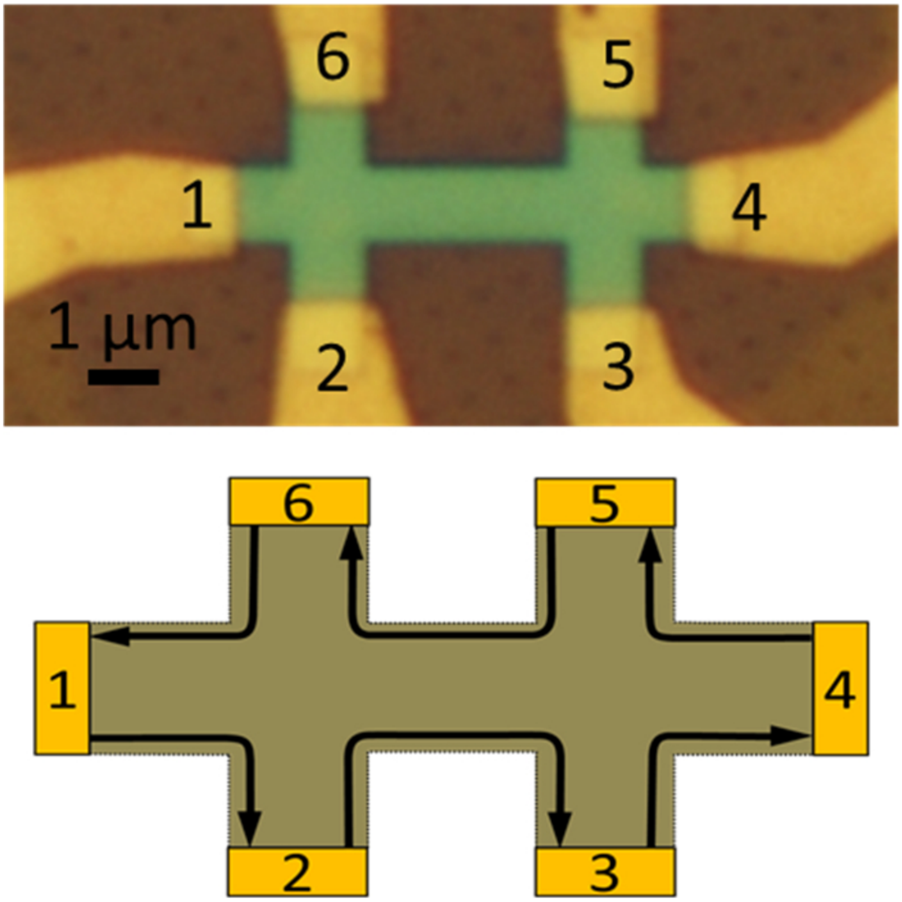

Figure 3. Hall bar device. Upper figure, optical image of Graphene/hBN Hall bar device with metal leads contacted to the terminals. Lower figure, device schematic with contact leads numbered corresponding to numerically labeled terminals in optical image of the device. Arrows demonstrate flow of current arising Landauer-Büttiker edge states. Please click here to view a larger version of this figure.

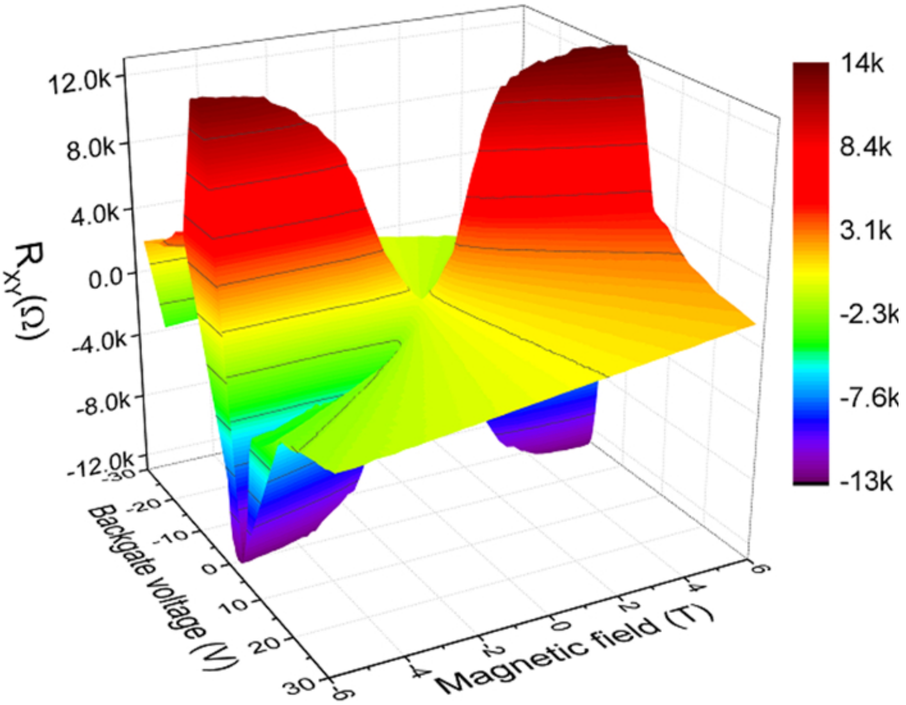

Figure 4. 3D plot of Hall resistance as a function of magnetic field and global backgate. 3D plot of the Hall resistance of Graphene hall bar in the quantum Hall regime as function of magnetic field and global back gate at 1.7 K demonstrating quantum transport behavior. Please click here to view a larger version of this figure.

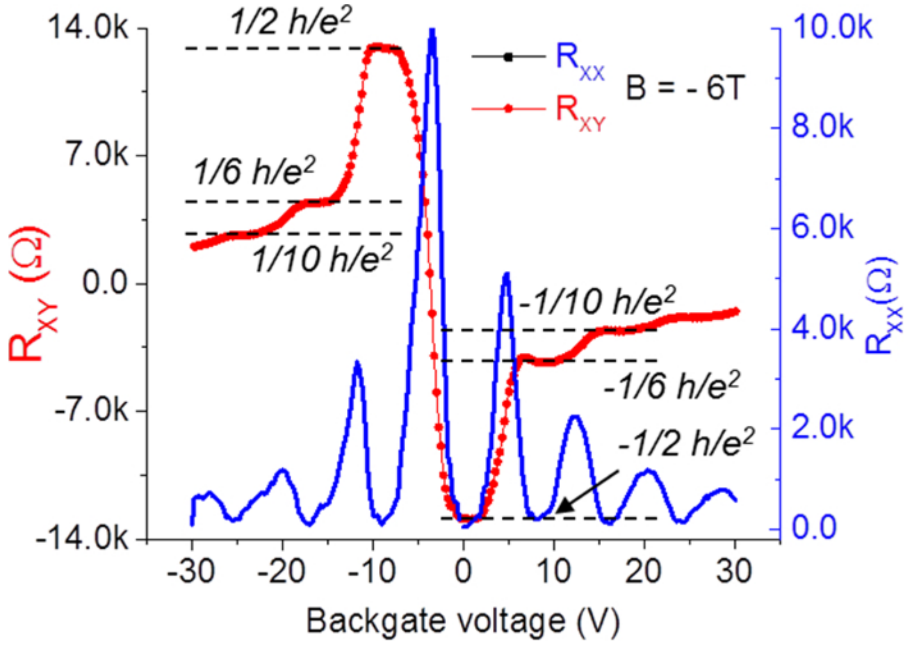

Figure 5. Hall and longitudinal magnetoresistance. Hall (R_{xy}) and longitudinal (R_{xx}) resistance as function of global back gate at fixed magnetic field $|B| = 6$ T demonstrating quantum Hall effect with quantized Hall resistance values coinciding with vanishing longitudinal resistance. Please click here to view a larger version of this figure.

Discussion

After acquisition of high quality bulk samples, characterized to ensure appropriate composition and structure, samples are patterned into the geometry depicted by exfoliating flakes of sample onto 1 cm × 1 cm pieces of substrate. Substrates composed of heavily p-doped Si covered by approximately 300 nm of SiO_2 are preferred as they increase the experimental parameter space by allowing the application of a back gate. Samples must be sufficiently thin — fewer than 10 nm — to produce a sufficient field effect to tune the chemical potential in the entirety of the conducting channel of the Hall bar device. Sample thickness is controlled by adequately exfoliating flakes from bulk material using standard wafer dicing tape and repeatedly pressing tape with flakes adhered to it to fresh tape until the flakes are of sufficient thickness for the purpose of the planned experiment. Samples transferred to substrate pieces are too small to see with the naked eye, so an optical microscope must be utilized to identify transferred pieces suitable to fabricate into a Hall bar. Sample flake thickness is accurately measured using atomic force microscopy (AFM), however researchers with sufficient experience may be able to identify samples of desired thickness based on the color of the flake.

A notable challenge to the technical procedure described in this manuscript arises when sample flakes do not adequately adhere to substrate pieces by van der Waals forces. In this case, during any number of steps of the fabrication procedure (especially during immersion in solvents) the sample flakes will be knocked or washed off of the substrate piece surface. This is addressed by the novel technique described in this manuscript whereby the sample flake is fixed to the substrate piece by coating the piece in sputtered SiO_2. Once this is done, parts of the sample flake must be exposed to allow the direct adherence of metal contacts. The mask used for the deposition of these contacts may be used to suit this purpose by serving as a mask for the etching of the SiO_2, as this would etch SiO_2 exactly where the metal contacts will be deposited, permitting electrical contact to the sample flake while still keeping the flake secured with sputtered SiO_2 over the majority of its area. Performing this step allows the transport measurement of novel bulk material that has been synthesized into crystals of insufficient size for conventional transport measurements of bulk materials, allowing for the transport study of many new materials as part of the pursuit of exploratory materials science research.

A profound technical advancement offered by the experimental techniques described in this paper come from the ability to stack multiple layered materials into heterostructures. This has numerous advantages. Hexagonal boron nitride (hBN) can be used to sandwich other 2D materials, such as graphene, to product the surface from defects arising from interaction with air, permitting more accurate, defect-free transport measurement of carrier states. Additionally, interesting emergent behaviors may be observable in heterostructures formed from stacks of different materials[12]. Following exfoliation, transfer to substrate piece, and identification of suitable sample flakes, a procedure may be followed to produce a heterostructure stack of multilayer materials involving sample transfer by the careful use of polymers polydimethylsiloxane (PDMA) and polypropylene carbonate (PPC). This method allows for such stacking without introducing globules of these polymers as contaminants between adjacent materials, as stacking occurs by pressing clean surfaces of the layered materials together. A completed heterostructure stack may be transferred to a new substrate piece for device fabrication.

Device fabrication is a rigorous process involving many steps. Once a suitable sample piece has been transferred, identified, and, if desired, stacked into a heterostructure comprised of several individual flakes, the steps of polymer mask application and patterning and several iterations of etching and metal deposition involved in the fabrication process can take several days to produce a single high quality sample. Due to the find-and-probe nature of this methodology, whereby a flake of desired size, thickness, and quality may be found anywhere on the substrate piece and Hall bar dimensions must be determined after the piece has been identified, lithography must be done by electron beam lithography. Electron beam lithography is an advanced lithography technique that allows the direct writing of structures down to nearly 5 nm dimensions via the use of a scanning of a focused beam of electrons. Specific device structures are prepared for each sample. Isotropic etching is done using the plasma generated in a reactive ion etching (RIE) system. For the etching of a hexagonal boron nitride/Graphene/hexagonal boron nitride stack, the gas used for this plasma etch is a mixture of O_2 and CHF_3. Deposited metal contacts consist of a thin layer of Cr, meant to serve as an adhesion layer, and a second layer of 750 nm of Au, chosen for its high electrical conductivity, deposited sequentially in high vacuum in an electron beam evaporation chamber. Fabrication of the device is complete after successful metal lift-off following metal deposition, after which point the device may be bonded to a package and loaded into a magnetotransport cryostat for experimental measurement.

Advancement of the fabrication and experimental techniques described in this manuscript will involve improvements in the procedure by which individual flakes may be stacked into heterostructures. Additionally, exfoliation of individual flakes and stacking of layered materials into heterostructures by the techniques described in this manuscript are limited to materials that are not effected by exposure to air. Additional consideration, such as undertaking much of the procedure in an inert environment, must be taken for materials that are destroyed by oxidation, such as the transition metal dichalcogenides and Bi-chalcogenide topological insulators. Magnetotransport systems will continue to see improvement as stronger magnets and lower temperature cryostats are engineered, leading to more powerful experimental measurement capability.

Disclosures

The authors declare no competing financial interests. Commercial materials, instruments and equipment are identified in this paper to specify the experimental procedure as completely as possible. In no case does such identification imply a recommendation or endorsement by the National Institute of Standards.

Acknowledgements

This work is supported by the National Institute of Standards and Technology of the United States Department of Commerce.

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