Characteristics of Graphene for Quantized Hall Effect Measurements

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***Abstract* — This paper describes concepts and measurement techniques necessary for characterization of graphene in the development of graphene-based quantized Hall effect (QHE) devices and resistance standards. We briefly contrast the properties of graphene produced by three common processing methods and discuss the conditions necessary for well-developed resistance plateaus to be observed. Methods used to determine the graphene layer thickness are presented. The metrologically relevant characteristics of graphene are correlated with electrical transport measurements in strong magnetic fields.**

***Index Terms* — Electrical resistance measurements, graphene, material properties, measurement standards, quantum Hall effect**

I. INTRODUCTION

The superior electronic properties of two-dimensional (2D) electronic devices made from graphene [1] – [2] hold promise of simplifying and improving the techniques required for maintaining the quantized Hall resistance (QHR) standard. Furthermore, graphene could support the production of QHR arrays of many different resistance values based on novel integrated p-n junction technologies [3], in ways similar to the advances made with Josephson voltage arrays [4]. This paper describes our recent work on the experimental characteristics of graphene's anomalous QHE plateaus and the development of graphene-based resistance standards. The focus of the work is to improve the quality of the graphene layer so that we can produce devices that meet the accepted guidelines for the QHR laboratory standard, and to provide a conceptual basis so that their performance can be well understood.

II. PHYSICAL AND ELECTRICAL PROPERTIES

Graphene is an electrically conducting sheet of sp^2 bonded carbon atoms. Its lattice has planar hexagonal symmetry in devices where the layer rests on a flat, insulating substrate. Thus in graphene, as in some two-dimensional semiconductor structures, a perpendicular magnetic field induces discrete Landau-level states (LL) for electron or hole charge carriers when the thermal energy (temperature) is low and the field strength is high. The gaps between the LL states numbered 0 and ±1 are quite wide for monolayer graphene, making this material particularly favorable for the QHR. This energy gap stabilizes the transverse resistance R_H over a broad range of magnetic flux densities [5] where $R_H(i) = h/ie^2$ with $i = 2$. In addition to the occurrence of anomalous LL energy gaps, the multiple symmetries in the quantum state of monolayer graphene suppress many scattering events [6]. Due to partial symmetry breaking, bilayer graphene has LL energy gaps that are less wide, and similar to those of conventional GaAs-AlGaAs heterostructures and Si-MOSFET QHR devices.

III. DEVICE FABRICATION AND TESTING

A. Graphene Processing and Film Characterization

The graphene samples under investigation are produced by exfoliation, chemical vapor deposition (CVD), and high-temperature annealing of SiC substrates. In the first two methods the graphene film is transferred to a non-conducting substrate for device fabrication. The layer(s) of so-called epitaxial graphene (EG) are grown on semi-insulating SiC wafers. These fabrication methods are described below.

Monolayer samples produced by exfoliation from graphite can produce high-mobility (20 000 cm^2/Vs is typical at 293 K) graphene devices, but with linear dimensions typically limited to about 10 μm, and thus having very small conducting channels and contact area. This limits their durability and current-carrying capability. CVD graphene is grown on metallic substrates and has a grain boundary structure similar to that of the metal. The physical deformation and contamination due to the transfer processes often further reduce the quality of the film. EG is commonly produced on the hexagonal-symmetry faces of 4H-SiC or 6H-SiC polytype wafers, with the underlying substrate having a terraced surface caused by the small miscut angle relative to the atomic planes. Regions of predominantly monolayer graphene can be grown to cover the terraces on Si-face, while small irregular regions of different layer number are produced in few-layer graphene grown on the C-face. The regular Si-face structure in EG is created by the initial growth of a covalently bonded buffer layer oriented to the crystalline structure of the SiC, and the formation of the first true graphene layer occurs by a lifting process as a new buffer layer is produced [7]. Thus, different initiation sites can form in isolation and merge to create large single domains. Due to negative charge transfer from the buffer layer, monolayer Si-face EG has carrier density values near $10^{13}/cm^2$, and the QHR plateaus cannot be observed without external doping or electrical gating of the device.

Characterization of graphene layer thickness is performed for EG in our laboratories using the methods described below. Scanning electron microscopy (SEM) and atomic force microscopy (AFM) are used to give an idea of the surface morphology and graphene coverage. Kelvin probe or electrostatic force microscopy (KPFM/EFM) is more sensitive to the graphene structure and is used with varying surface potential to indicate layer thickness. Raman spectroscopy is capable of determining the presence of graphene and may give an indication of the layer number and quality by line shape analysis. The most definitive analysis is made by low-energy scanning electron microscopy (LEEM).

B. Charge Carrier Density

Recently, we have observed QHR plateaus in large devices made from CVD graphene [8] grown on copper. Indium contacts have been used to minimize processing, with the devices cooled in a ^{4}He cryostat soon after their transfer to SiO_2/Si substrates. This reduces the exposure to air and can produce a very low extrinsic charge doping level such that the Dirac point, where hole and electron densities are balanced, is near 1 V. With this preparation technique, carrier mobilities of around 4000 cm^2/Vs and distinct resistance plateaus (i = 2, 6, and 10) are observed up to at least 77 K. These devices are of cm-scale and have superior QHR properties compared to smaller samples of similar material. However, in these devices the minimum longitudinal resistivity is > 200 Ω, several orders of magnitude larger than desired.

A collaboration of University laboratories in Sweden which included Great Britain's National Physical Laboratory (NPL) has developed a set of Si-face EG devices of width (length) up to 35 μm (160 μm) that produce extremely well-quantized resistance values [5]. With a special polymer electrostatic gating technique [9] developed by the group working at Chalmers University, device carrier densities were reduced by more than an order of magnitude while high carrier mobility was maintained. The method has been used with EG samples produced in our laboratories with similar effects. This polymer is used with UV-light excitation and the effect allows fine tuning of the position of i = 2 plateaus.

C. Hall Bar Dimensions and Contacts

We note that the best graphene QHR devices (at present) have relatively large dimensions, both in the channel and for the electrical contacts. There is some similarity with GaAs devices which tend to improve as the channel width and also length are increased if the channel region is homogeneous. Also it is of interest that the EG devices described in the last section were patterned to lie across many terrace steps and the CVD devices contained many defects. Thus, in analogy with earlier experience for GaAs, large conducting channels of monolayer graphene that contain various small regions of non-planar morphology, different layer numbers, and charged defects may be suitable for QHR devices. If it is not too extreme, the disordered electronic state may provide bulk conducting regions favorable for higher channel currents and broader plateaus that are absent in monolayer samples with very few defects and highly uniform charge density.

IV. Conclusion

In conjunction with the processing of graphene material and fabrication of devices for QHR metrology we are developing characterization techniques that may lead to a better understanding of the conditions necessary for stability of the QHE. These methods will be described and results for several different types and sources of graphene will be presented.

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