Carbon 115 (2017) 229–236

Contents lists available at ScienceDirect

Carbon

journal homepage: www.elsevier.com/locate/carbon

Epitaxial graphene homogeneity and quantum Hall effect in millimeter-scale devices

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ARTICLE INFO

Article history:
Received 8 August 2016
Received in revised form
17 December 2016
Accepted 29 December 2016
Available online 30 December 2016

Keywords:
Epitaxial graphene
Transport mobility
Carrier density
Adsorption-induced molecular doping
Strain
Low-energy electron microscopy
Raman microscopy
Quantized Hall effect

ABSTRACT

Quantized magnetotransport is observed in 5.6 × 5.6 mm^2 epitaxial graphene devices, grown using highly constrained sublimation on the Si-face of SiC(0001) at high temperature (1900 °C). The precise quantized Hall resistance of $R_{xy} = \frac{h}{2e^2}$ is maintained up to record level of critical current $I_{xx} = 0.72$ mA at $T = 3.1$ K and 9 T in a device where Raman microscopy reveals low and homogeneous strain. Adsorption-induced molecular doping in a second device reduced the carrier concentration close to the Dirac point ($n \approx 10^{10}$ cm^{-2}), where mobility of 18760 cm^2/V is measured over an area of 10 mm^2. Atomic force, confocal optical, and Raman microscopies are used to characterize the large-scale devices, and reveal improved SiC terrace topography and the structure of the graphene layer. Our results show that the structural uniformity of epitaxial graphene produced by face-to-graphite processing contributes to millimeter-scale transport homogeneity, and will prove useful for scientific and commercial applications.

1. Introduction

Wafer-scale monolayer graphene [1,2] can be produced by thermal decomposition of certain polytypes of silicon carbide [3] (SiC) or by chemical vapor deposition (CVD) on metal catalyst substrates [2]. While CVD graphene forms randomly oriented domains to match the crystal orientation of the metal catalyst, epitaxial graphene (EG) forms a single domain on monocrystalline wafers of hexagonal SiC(0001) [4] and the insulating SiC substrate is immediately suitable for fabrication of electronic [5], plasmonic [6] and photonic [7] devices. Quantum Hall effect (QHE) standards produced from EG [8,9] can be operated economically at lower magnetic fields and higher temperatures than GaAs-AlGaAs heterostructures [10]; thus EG devices are likely to become the premier source of resistance traceability in practical metrology and their optimization is of great interest to the electrical metrology community.

Efforts to produce nearly defect-free monolayer EG on SiC generally involve control of the high-temperature vapor phase. For example, annealing in atmospheric-pressure Ar gas [11] or in a small confining enclosure [12] helps to raise the partial pressures of sublimated Si, Si_2C and SiC_2 closer to equilibrium at high temperature, and the number of defects in graphene is then reduced and the morphology of vicinal SiC(0001) surfaces is generally improved. However, dissociated carbon atoms may diffuse anisotropically [13], leading to the formation of multiple graphene layers near the edges of the terraces [14]. Furthermore, SiC restructuring [3] and energetically-favorable step-bunching also may produce undesirable terrace facet edges [14,15] that face off-axis by ≈ 30° on vicinal SiC(0001). Atomic-layer-resolved characterization has shown

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http://dx.doi.org/10.1016/j.carbon.2016.12.087
0008-6223/

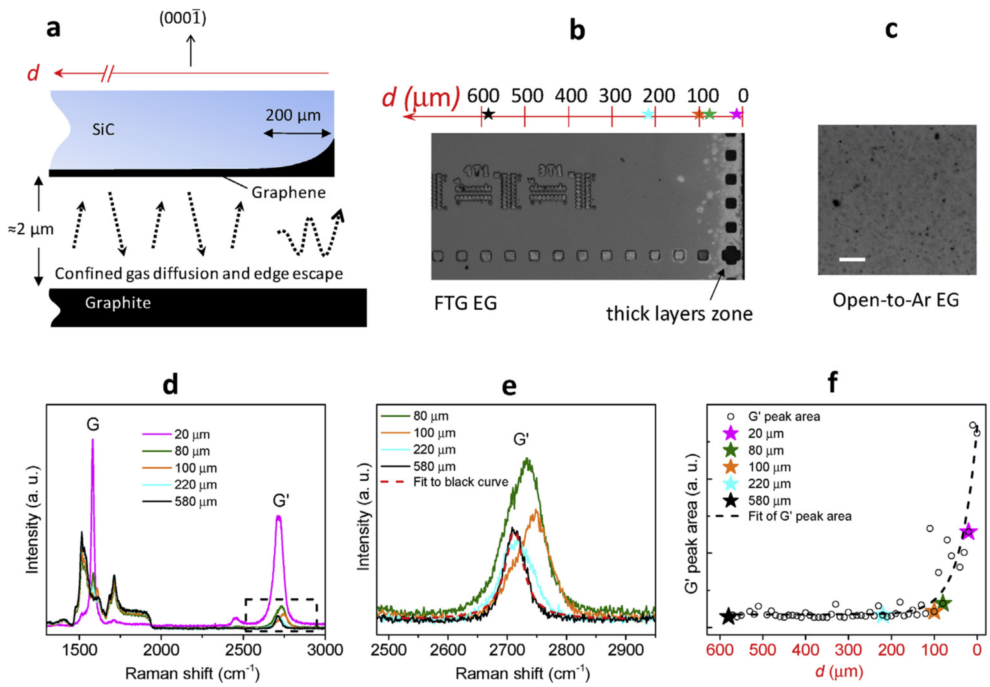

Fig. 1. (a) Diagram showing a cross-section of the SiC sample and polished graphite during FTG processing. (b) Optical image showing the edge region of a 7.6 mm square sample processed at 1950 °C for 1800 s with FTG configuration. The squares and pattern are fiducials etched into the SiC substrate. (c) Optical image of a sample processed with Si-face open to Ar at 1900 °C for 210 s, showing the center region with non-uniform graphene. The white scale bar is 10 µm. (d) Representative Raman spectra vs. distance from the edge of FTG samples, showing G and G′ Raman peaks of EG at positions $d = 20$ µm (magenta), 80 µm (green), 100 µm (orange), 220 µm (cyan), and 580 µm (black). (e) Zoom-in of Raman G′ peak spectra from (b), as indicated by the dashed black box. The G′ peak at $d = 20$ µm is omitted for clarity. The G′ peak at $d = 580$ µm can be fit well with a single Lorentzian curve as shown. (f) Integrated G′ peak area vs. distance from the sample edge. The locations for the data shown in (d)–(f) are marked by colored stars along the axis above the optical image in (b). The data shown in (d)–(f) are marked with the same color scheme.

significant delamination of the carbon buffer layer [15,16] on facet edges that separate adjacent terraces. Here, we report precision measurements of the QHE in millimeter-scale EG devices at high current and correlate the quantized magnetotransport to microscopy data, including structural reorganization of the SiC surface, EG layer number and distribution, and strain as measured by Raman microscopy.

2. Sample preparation

2.1. High temperature EG growth with vapor constraint

To minimize the possible complications due to the substrate, we produce EG using a constraint on vapor diffusion provided by close proximity to polished pyrolytic graphite substrates (SPI Glas 22). The samples were diced from two 76 mm SiC(0001) semi-insulating wafers (Cree, Inc.[3]) of nominal miscut 0.00°, with sample miscut measured to be $\leq 0.10°$ from atomic force microscopy (AFM) images. Samples were rinsed in HF and deionized water before processing, and arranged facing to graphite (FTG) with separation distance limited only by sample and substrate flatness (Fig. 1a). Processing was done in a graphite-lined resistive-element furnace (Materials Research Furnaces Inc.) with heating and cooling rates near 1.5 °C/s. The initial heating occurs in forming gas (96% Ar, 4% H_2) at 100 kPa with at least 30 min cleaning of the substrates at 1050 °C, which may serve to hydrogenate the SiC surface [17,18]. The chamber was then flushed with Ar gas, and filled with 100 kPa Ar derived from 99.999% liquid Ar before annealing at 1900 °C. The annealing process utilized a commercial process controller and a type-C thermocouple located a few cm above the sample.

Figs. 1b and c show the optical images of two samples processed with and without the FTG configuration, respectively. The sample processed with Si-face open to Ar (Fig. 1c) shows inhomogeneous EG coverage over the entire substrate. The FTG sample (Fig. 1b) has uniform EG coverage (grey area) toward its center, while lower vapor pressure near its borders has allowed the formation of thick, graphitic layers (white area). Raman spectra confirm these results and are shown in Figs. 1d–f from five locations along a linear scan of a FTG sample. Both the G band (≈ 1600 cm^{-1}) and the G′ band (≈ 2700 cm^{-1}) from EG evolve with the distance d from the edge and indicate the variation in layer number. Fig. 1e magnifies the region of the G′ band, excluding the peak at $d = 20$ µm, and Fig. 1f shows the exponential decrease of the integrated G′ peak area toward the center of the substrate. At interior distances $d > \approx$ 200 µm, most G′ peaks take on the shape of a single Lorentzian with a full width at half maximum (FWHM) of ≈ 40 cm^{-1}, which is the fingerprint for identifying monolayer EG.

2.2. Device fabrication and molecular doping

In this paper, we will focus on two FTG-grown samples of size 7.6 × 7.6 mm^2. We patterned a semi-octagonal active channel utilizing the center region of the FTG samples, which has an area of 27 mm^2 (see the inset of Fig. 2a). The edge of the active graphene channel is 1 mm from the edge of the substrate so that the periphery region of thick layers is avoided. In fabricating the magnetotransport devices, the active EG surfaces were kept uniformly resist-free by depositing Pd/Au as a thin layer prior to standard photolithography processing, and afterwards removing the Pd/Au

[3] Identification of commercial products or services used in this work does not imply endorsement by the US government, nor does it imply that these products are the best available for the applications described.