

Magnetotransport in graphene on silicon side of SiC

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Abstract. We have studied the transport properties of graphene grown on silicon side of SiC. Samples under study have been prepared by two different growth methods in two different laboratories. Magnetoresistance and Hall resistance have been measured at temperatures between 4 and 100 K in resistive magnet in magnetic fields up to 22 T. In spite of differences in sample preparation, the field dependence of resistances measured on both sets of samples exhibits two periods of magneto-oscillations indicating two different parallel conducting channels with different concentrations of carriers. The semi-quantitative agreement with the model calculation allows for conclusion that channels are formed by high-density and low-density Dirac carriers. The coexistence of two different groups of carriers on the silicon side of SiC was not reported before.

1. Introduction

Epitaxial growth of graphene layers on the surface of single-crystalline SiC, reported first in a seminal paper [1], is a well established way to the large scale production of graphene for a future graphene based electronics. Many modifications of this technology have been reported. In addition to standard thermal decomposition of the upper layers of the SiC substrate at temperatures above 1200°C in various atmospheres, chemical vapour deposition (CVD) on the SiC surface can also be used. In the former case graphene arises by rearrangement of carbon atoms left on the SiC surface after Si atoms sublimed out, in the latter carbon atoms have to be added from outside by thermal decomposition of suitable hydrocarbon gas on the surface. Growth details can be expected to influence transport properties of graphene even on SiC substrates of the same crystallographic orientation, due to various types of active defects and/or character of the interface between the graphene and the substrate. In the present paper we report on the experimental study of the and Hall resistances on two series of samples, prepared from wafers epitaxially grown on Si-side of SiC, using either Si sublimation or CVD methods.



2. Experiments

2.1. Graphene growth

Two series of samples have been employed in this study. One of them, denoted as G141, was produced at the University of Linköping, Sweden, on the semi-insulating 6H-SiC(0001) wafer by the Si-sublimation method in an inductively heated furnace. The growth was carried out under highly isothermal conditions at 2000°C and at an ambient argon pressure of 1 bar [2].

Samples in the second series stem from a wafer 699, supplied by ITME Warsaw, Poland. The graphene growth was performed in a commercially available horizontal CVD hot-wall reactor (Epigress V508), inductively heated by an RF generator. Semi-insulating 4H-SiC (0001) substrates were used and propane gas served as the precursor. The substrates were etched in hydrogen and propane mixtures prior to carbon deposition. Precautions had been taken to prevent sublimation of Si atoms in conditions of high temperature ($\approx 1600^\circ\text{C}$) and a low Ar pressure [3].

Raman spectroscopy was used to confirm the presence of graphene.

2.2. Lithography

Samples were patterned by standard photolithographic techniques. Hall bars with a 100 micrometers wide conducting channel, two current contacts and six contacts for measurement of the longitudinal magnetoresistance and the Hall voltages were formed by oxygen plasma etching. The separation of potential leads was 300 micrometers. The aspect ratio of the sample (width/length) was either 1/3 or 1/6, depending on the pair of contacts employed. The macroscopic size indicates that the sample area covers a large number of terraces on the SiC surface and inhomogeneity in the graphene layer cannot be excluded. Ohmic TiAu contacts were prepared by the electron beam assisted deposition and lift-off technique.

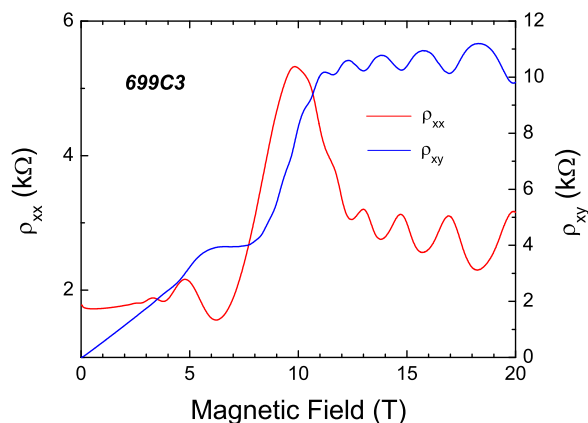


Figure 1. Longitudinal (red curve) and Hall (blue curve) magnetoresistances measured on a sample prepared in ITME Warsaw, Poland. $N_1 = 9.5 \times 10^{11} \text{ cm}^{-2}$, $N_2 = 1.1 \times 10^{13} \text{ cm}^{-2}$.

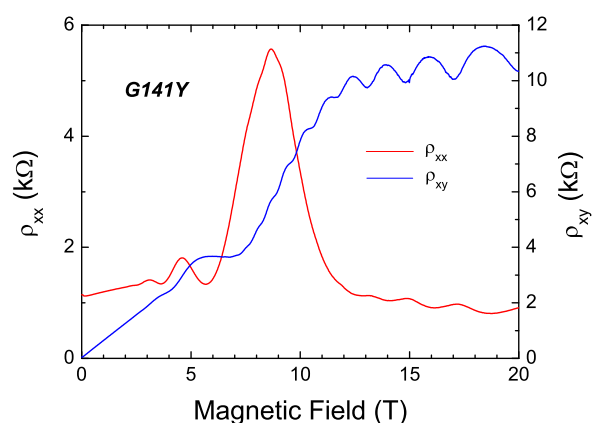


Figure 2. Longitudinal (red curve) and Hall (blue curve) magnetoresistances measured on a sample prepared in University of Linköping, Sweden. $N_1 = 8.9 \times 10^{11} \text{ cm}^{-2}$, $N_2 = 1.4 \times 10^{13} \text{ cm}^{-2}$.

2.3. Magnetotransport measurement

Magnetoresistance $\rho_{xx}(B)$ and Hall resistance $\rho_{xy}(B)$ have been simultaneously measured by a 4-probe low frequency ($f = 10.66$ Hz) AC lock-in method. Excitation current up to 100 nA was taken from the output of the lock-in amplifier using 10 M Ω or 100 M Ω load resistors. Most experiments were performed with samples directly immersed in a bath of liquid helium at 4.2 K. For the investigation of the temperature dependence up to 100 K a VTI has been used with a calibrated Cernox thermometer as a temperature sensor. Transverse magnetic fields up to 22 T were generated by a 10 MW Bitter magnet. Admixtures of ρ_{xx} to ρ_{xy} due to contacts misalignment was removed by symmetrization of all traces measured in positive and negative magnetic fields.

All experiments were realized in LNCMI CNRS Grenoble, France.

3. Results and discussion

The dominant feature observed on the magnetoresistance (MR) curves $\rho_{xx}(B)$ shown in Fig. 1 and Fig. 2 is the presence of two types of oscillations, both periodic in $1/B$ but with different periods. Simultaneously, the Hall resistance $\rho_{xy}(B)$ oscillates at fields above 10 T. All these oscillations disappeared after the samples were rotated into parallel configuration with respect to applied magnetic field, which confirms that the oscillations originate in two-dimensional sheets of charge carriers.

Such a behavior cannot stem from a single conducting sheet and therefore two independent channels have to exist. From the periods of oscillations in longitudinal magnetoresistance $\rho_{xx}(B)$ we can estimate charge carriers concentrations in both channels. Denoting the channels responsible for the „low field” and „high field” oscillations by indices 1 and 2, respectively, we obtained for the sample 699C3 (Fig. 1) the values $N_1 = 9.5 \times 10^{11} \text{ cm}^{-2}$ and $N_2 = 1.1 \times 10^{13} \text{ cm}^{-2}$. The same analysis of the data presented in Fig. 2 gives the concentrations $N_1 = 8.9 \times 10^{11} \text{ cm}^{-2}$ and $N_2 = 1.4 \times 10^{13} \text{ cm}^{-2}$.

Apart from the modulation by the oscillations at higher fields, the Hall curves for both samples clearly exhibit QHE plateaux at $\rho_{xy}(B) = (h/\nu e^2)$. Absence of the plateaux at $\nu = 4$ confirms, that the „low field” oscillations can be attributed to the single layer graphene. The identification of the second conducting layer with substantially higher carrier concentrations N_2

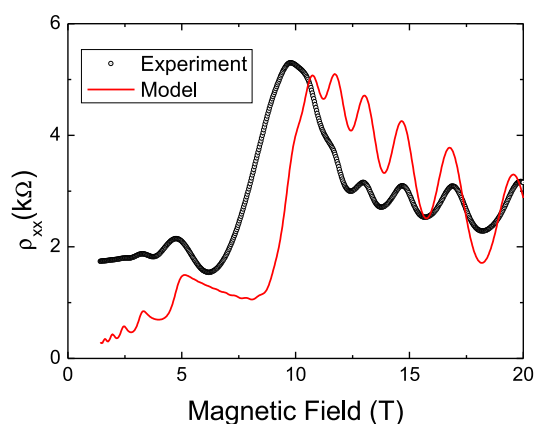


Figure 3. Experimental and model curves of longitudinal magnetoresistance of a sample 699.

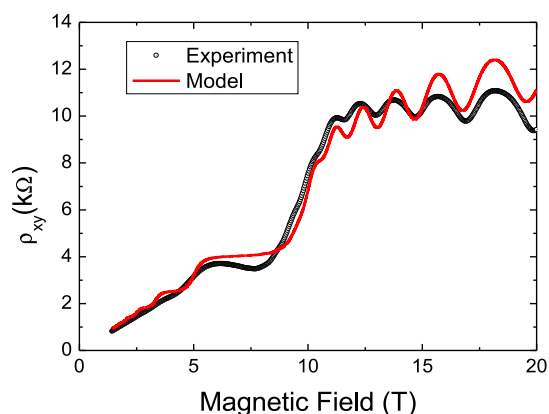


Figure 4. Experimental and model curves of Hall magnetoresistance of a sample 699.

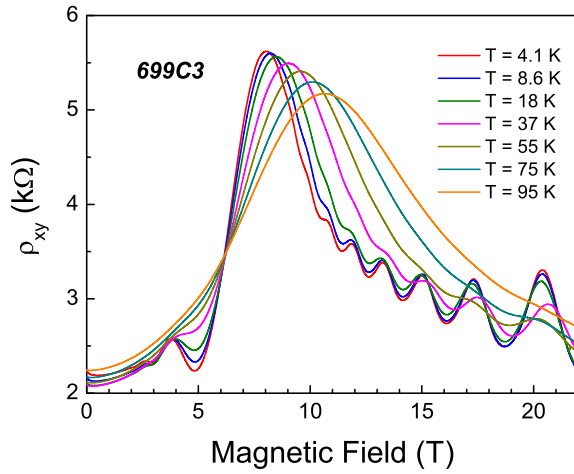


Figure 5. Longitudinal magnetoresistances measured at various temperatures on sample prepared in ITME Warsaw, Poland.

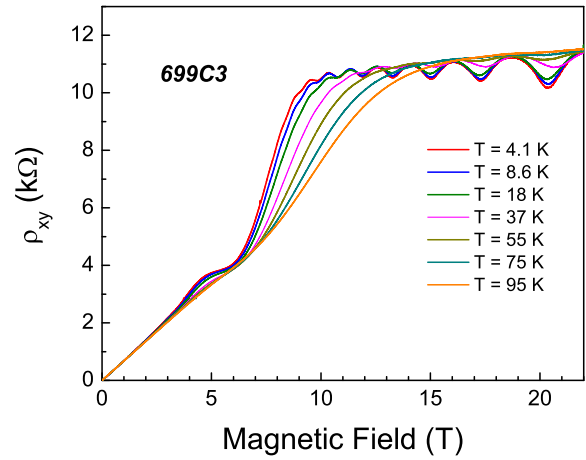


Figure 6. Hall magnetoresistances measured at various temperatures on sample prepared in ITME Warsaw, Poland.

seems to be less straightforward, the Hall plateaux cannot be reached in magnetic fields at our disposal. Assuming the same character of both channels, we could compare experimental data for the sample 699C3 in Fig. 1 with calculations based on the model described in [4], including the appropriate description of the conductivity tensor for graphene [5], and assuming two parallel conductivity channels in the sample.

In these calculations, concentrations N_1 and N_2 , together with the half-widths $\Gamma_1 = 2.9$ meV and $\Gamma_2 = 6$ meV of Landau levels serve as fitting parameters. The results of the best fit are presented in Figs. 3 and 4. The model describes the data reasonably well, in particular those in Fig. 4 for the Hall resistance. Moreover, the best fit provides concentrations $N_1 = 1.2 \times 10^{12} \text{cm}^{-2}$ and $N_2 = 1.1 \times 10^{13} \text{cm}^{-2}$. These agree quite well with the concentrations derived directly from the periods of both types of MR oscillations observed on the same sample.

In order to further characterize the two conducting channels, we have measured temperature dependence of $\rho_{xx}(B)$ and $\rho_{xy}(B)$ up to about 100 K. The results can be seen in Fig. 3 and Fig. 4. Both types of oscillations persist up to quite high temperatures, which is characteristic property of graphene. Temperature damping of the amplitude of oscillations provides an estimate of the cyclotron effective mass m_c^* of the charge carriers [7]. It implies a reliable subtraction of the monotonous „background” part, which is not easy here, in particular for the „low field” oscillations. Nevertheless, in highest fields, where the resistivity $\rho_{xx}(B)$ from the channel responsible for the „low field” SdH oscillations approaches zero, we got values of m_c^* that agree reasonably well with those derived from the concentrations N_i . The latter stem from the expression $m_c^* = \hbar/v_F$ for the effective mass of the single-layer graphene [7]. Taking $v_F = 1.1 \times 10^6$ m/s [7] and the concentrations N_1 and N_2 given in caption to Fig. 1, we get $m_{c1}^* = 0.018 m_0$ and $m_{c2}^* = 0.061 m_0$, respectively. The curves presented in Figs. 5 and 6 were obtained more than a year later than those shown in Fig. 1 (see [6]). This confirms the stability of the sample properties in time.

We can conclude, that our results for the two wafers produced by widely different technologies can be interpreted as a superposition of oscillations from two parallel two-dimensional conducting channels with substantially different carrier concentrations and apparently also mobilities.

It seems to be surprising, since different type of substrates together with different growth technology can be expected to provide quite different morphology of the epitaxial graphene. Detailed study of the graphene formed on SiC under various environments [8] has shown no apparent differences between results for 6H-SiC and 4H-SiC wafers. Substantial differences can however arise between samples produced by the purely sublimation method and the CVD assisted one. In the former case the existence of an interfacial (buffer) layer is well established and it influences the electronic transport properties of graphene [9], most probably through the concentration and mobility of the charge carriers. But due to the bonding of the buffer layer to the Si atoms in the substrate it loses its graphene-like linear band structure near the Fermi level [9] and cannot thus generate SdH oscillations characteristic for the single layer graphene. In the CVD assisted method, the structure of the interface between graphene and the SiC substrate is less understood, but it probably also influences only the concentration and mobility of the charge carriers in uppermost graphene layers. It is important that both technologies produce graphene, that is far from being flat and homogeneous on the scale larger than a few microns. Graphene seems to nucleate on the edges of the terraces and grow further in islands until they fuse into a layer, which is supported by the diffusion of the carbon atoms along the surface [9]. It can be expected, that not only the morphology and thickness [9], but also the transport properties can be different for graphene at the terraces and their edges [10]. Therefore we speculate, that the surface area of our samples is broken into small interconnected regions with either low or high concentration of carriers, which form a network of parallel and/or series resistors. It may be a combination of the monolayer graphene at the terraces and bilayer graphene at their edges [8], but the electron transport measurement itself can hardly reveal the detailed microstructure of the epitaxial graphene on SiC substrate. Explanation of the origin of dual SdH oscillations observed in our samples thus requires further investigation.

Acknowledgments

The support of the European Science Foundation EPIGRAT project (GRA/10/E006, 670 and 671/N-ESF-EPI/2010/0), GACR No. P204/10/1020 and No. P108/11/0894 projects, programme „Transnational access” contract No. 228043-EuroMagNET II-Integrated Activities, AVCR research program AVOZ10100521, the Academy of Sciences of the Czech Republic project KAN400100652 are acknowledged.

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