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Contactless Magnetoresistance in Large Area Epitaxial Graphene Grown on SiC Substrates

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Abstract: The present contactless measurements of magnetoresistance of large area graphene films grown by chemical vapor deposition method on semi-insulating SiC substrates. For this purpose we propose microwave technique using single post dielectric resonator operating at frequency about 13.5 GHz. Comparison of microwave measurements with classic van der Pauw method shows good agreement and proves usefulness of contactless technique in magnetoresistance studies. Experiments have been performed at 4.2 K in magnetic fields up to 7 T. Significant differences in graphene magnetoresistance depending on the orientation and crystal structure of SiC substrates have been observed.

Key words: Graphene, magnetoresistance, contactless measurement, microwaves.

1. Introduction

Graphene is considered as a very promising material for future electronic applications. Its great potential is confirmed by countless publications mostly concerning few micron-sized films obtained by micromechanical cleaving. Among them, there are studies involving magnetic field like magnetoresistance and Hall effect, which shed much light onto electronic structure of almost ideal small area graphene sheets [1-7]. Unfortunately, industrial applications demand not only high quality but also large area graphene sheets produced in repeatable process. A promising technology, which in the future can simultaneously meet these requirements, is epitaxial growth on semi-insulating SiC substrates [8-10]. Up to now, number of works on transport properties of epitaxial graphene on silicon carbide have been performed [11-14], mostly using invasive methods with contacts. Thus, the development of a method that is both harmless and reliable is of great importance. So far, only room-temperature microwave technique that is suitable for contactless studies of sheet resistance of large area graphene sheets has been proposed [15, 16]. The magnetic and low temperature studies with the contactless approach are however missing. The main aim of our study is bridge this gap by extending room-temperature method and develop new suitable magnetoresistance graphene characterization technique.

2. Experiments

Details of the microwave assembly are shown in Figs. 1a and 1b. In our setup graphene film is placed in copper cavity in front of the home-made single post

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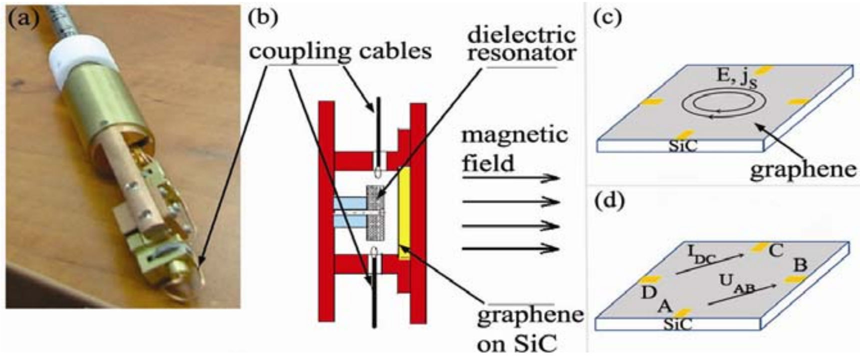

Fig. 1 (a) Photograph of the resonator and nonmagnetic cryo-stick with the maximum diameter of 25 mm. (b) Schematic of 13.5 GHz single post dielectric resonator intended for contactless sheet resistance and magneto-resistance measurements. Draft of the sample for AC (c) and DC (d) experiment. The electric contacts (Ti/Au) are placed at the middle of each edge of the sample. The opposite electrodes are located 8mm from each other. Black arrows show different current path used in both methods.

dielectric resonator (SiPDR). The resonator operates on a quasi TE_{011} mode at frequency about 13.5 GHz. It excites in the sample alternating electric currents that have only azimuthal components, as it is depicted in Fig. 1c. Diameter of the circle-shaped current path reflects dimensions of the RF resonator (about 3.5 mm). Flowing currents in the case of nonzero resistance imply energy dissipation affecting easily measurable quantities characterizing SiPDR like resonant frequency f_{res} and Q-factor. Because the letter one depends on the conductivity in the measured graphene the detailed analysis yields graphene sheet resistance R_s. For this purpose an electromagnetic analysis based on mode-matching and Rayleigh-Ritz techniques have been used (formulas not shown here). For more details see references [15, 16]

In order to get reliability and accuracy of the contactless technique classical four-probe van der Pauw method has been additionally employed. For this method one electrode in the middle of each edge of the sample was fabricated (Fig. 1d). Distances between opposite contacts equal 8mm. Such location allows to use exactly the same samples in two different experiments because no interaction between metal pads and circular currents excited by microwave resonator takes place. Measurements in van der Pauw geometry are performed in following way. Carriers flow straight between two adjacent electrodes and the other two are used for determination of the potential drop. If resistances between given points are defined by the next-mentioned equations:

$$R_{AB,CD} = (V_D - V_C)/I_{AB} \qquad (1)$$

and

$$R_{BC,AB} = (V_B - V_A)/I_{BC} \qquad (2)$$

the graphene sheet resistance R_s can be obtained from the following relation:

$$\exp(-\pi R_{AB,CD}/R_s) + \exp(-\pi R_{BC,AB}/R_s) = 1 \qquad (3)$$

Wafer-scale epitaxial graphene was grown by CVD [10] on three semi-insulating SiC surfaces: 4H-Si, 6H-Si, and 4H-C. Here, symbols 4H and 6H stand for silicon carbide poly-types. The labels Si and C denote polarities (faces) – silicon and carbon, respectively. Substrates samples with dimensions of 10 mm × 10 mm × 0.5 mm used in separated growth runs with intentional changes in growth conditions, which should result in the different graphene quality. Such prepared samples were placed in the microwave assembly, which was mounted at the end of nonmagnetic stick and put into the Oxford Instruments Spectromag cryostat. The temperature was lowered down to 4.2 K. The magnetic field (up to 7 T) was applied in the direction perpendicular to the sample surface using a superconducting magnet. In order to show the details of the measurements with low magnetic field, the relative magnetoresistance was defined as:

$$MR(B) = R(B)/R(0) - 1 \quad (4)$$

3. Results and Discussion

The dependence of the sheet resistance R_s of large area graphene flakes on magnetic field at 4.2 K is presented in Fig. 2. curves (x) stand for results obtained by contactless microwave technique, (y) stand for van der Pauw reference data. For higher magnetic fields (B > 0.5 T) the resistance always shows positive quasi-linear trend, for lower fields (B < 0.5 T) a parabolic behavior and negative relative magnetoresistance can be seen. Most pronounced resistance drop in magnetic field is observed for graphene labeled as 932-4H-C (see Fig. 1a). Relative *MR* assumes value of -5% for B = 0.25 T. Contactless method reproduces this feature, as well as the rest of results on reasonable level. Small positive shift of contactless resistance occurs in whole magnetic field regime and is attributed to systematic error. Maximal MR ratio at the end of linear part of the curve (B = 7 T) equals 35%. Sample denoted as 866-4H-Si (Fig. 2b) exhibits 2% (B = 0.15 T) negative magnetoresistance in van der Pauw experiment but this behavior is not seen in the data from contactless measurements (this issue will be addressed further). For this sample, curves describing the resistance obtained by both techniques follow close each other up to 2.5 T, then start to separate. Maximal magnetoresistance reaches 240% and 310% for contactless and Van der Pauw measurements, respectively. In our opinion there is still good agreement between two sets of results for this graphene flake, despite high divergence in 7 T. As in previous case, similarly for 875-6H-Si graphene sample, small -0.7% negative MR is seen only in data from contact method. Despite this small odd feature, results of both experiments demonstrate linear dependence on magnetic field with maximal change in magnetoresistance reaching 60%. Just like in the case of 932-4H-C sample almost constant shift between curves is attributed to systematic error.

The linear nonsaturating positive magnetoresistance (LMR) for high enough magnetic fields observed in both experiments can be attributed to inhomogeneity in the epitaxially grown graphene films [11]. We treat our epitaxial graphene as inherently disordered because dimensions of the sample are much larger

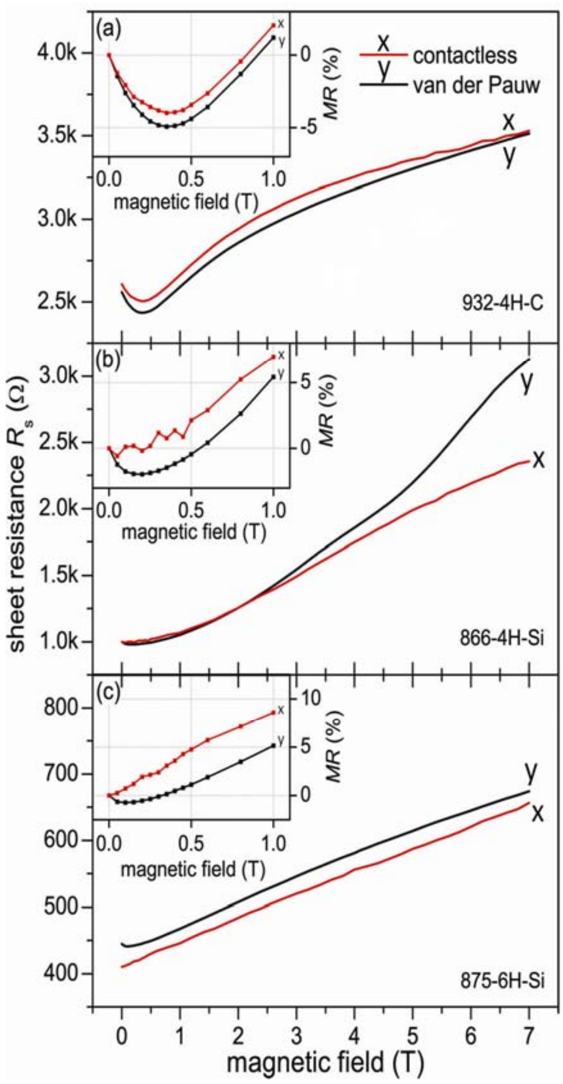

Fig. 2 Resistance versus magnetic field for three large area graphene films grown on semi-insulating SiC with different poly-types and polarities. Curves (x) stand for data obtained by microwave technique, Curves (y) stand for van der Pauw reference data. Insets emphasize negative relative magnetoresistance for low *B*. Temperature equals 4.2 K.

than the domain size of the material. Keeping in mind that area under investigations in two completely different experiments is similar, consistency of the results should not surprise (area examined in microwave experiments has rounded shape with diameter about 3.5 mm, electrodes in van der Pauw method are located at distance of 8 millimeters). As regards of the Shubnikov-de Haas oscillations, we believe that the reason for which we do not see SdH effect is not high enough magnetic field we are able to apply. On the basis of presented data we are not able to distinguish between classical and quantum character of the LMR.

We now turn our attention to the negative magnetoresistance. The MR at low B, clearly seen in the contact measurements, strongly suggests the effect of weak localization (WL) [17, 18]. In generic WL, increase in resistance at zero field is due to an increased backscattering probability. This probability is decreased when magnetic field is applied implying negative MR. In graphene, however, backscattering can occur only in the presence of inter-valley-scattering, caused, e.g., by short range potential. Therefore, resistance peak at $B = 0$ T can be related to the point defects existing in sample. This suggests that the desired quality of large area epitaxial graphene produced by epitaxial technique is still to be improved. CVD graphene quality obtained on Si-terminated surface was higher than that deposited on C-face substrate. The difference between 4H and 6H was also observable, which enables the application of the contactless magnetoresistance technique in graphene growth optimization process.

Another issue clearly seen in the insets of Fig. 2 is related with the fact that unfortunately, resistance drop in magnetic field is not well reproduced by microwave method. There could be two, mutually non-contradictive explanations. First is the accuracy. Results from contactless experiment follow van der Pauw reference data only for the 932-4H-Si sample, which exhibits most pronounced WL feature. For the 866-4H-Si graphene flake experimental points look noisy, for the 875-6H-Si sample no sign of weak localization in contactless curve can be seen. This shows that the accuracy of contactless method (5%) is lower than in classic DC technique [15, 16]. Second explanation could be different currents paths. Because in van der Pauw experiment, current flows from one electrode to other, its path forms straight line, which length is not smaller than distance between considered electrodes. In microwave method, the carriers velocity has only azimuthal component. Moreover, its direction changes with 13.5 GHz frequency. All this can cause the discrepancies in the data coming from two different methods.

At the end, we would like to comment issue related to the carrier concentrations in studied graphene flakes. In presented microwave measurements we did not have the possibility to modulate population of electrons or holes neither by top- nor bottom-gate [19]. This is because both technical solutions would additionally disturb properties of the microwave cavity and interfere with non-invasive approach of the technique. However, the application of gates is in principle possible and one would need to take care of the additional measurement factor introduced by the presence of the metallic layer in the microwave resonator.

4. Conclusions

Summarizing, we performed two different experiments on large area epitaxial graphene on SiC. We compare results obtained by contactless microwave method with reference data from van der Pauw measurements. Good agreement has been achieved, proving usefulness and potential of our novel technique. Small differences related mainly to feature produced by weak localization are supposed to originate from different accuracy and currents paths.

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