Low-temperature quantum transport in CVD-grown single crystal graphene

S. Xiang[1], V. Miseikis[2], L. Planat[1], S. Guiducci[1], S. Roddaro[1], C. Coletti[2], F. Beltram[1,2] and S. Heun[1]

[1] NEST, Istituto Nanoscienze-CNR and Scuola Normale Superiore, Pisa, Italy
[2] Center for Nanotechnology Innovation @ NEST, IIT, Pisa, Italy

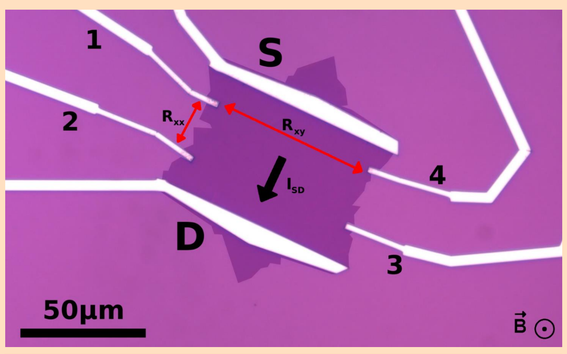
Introduction

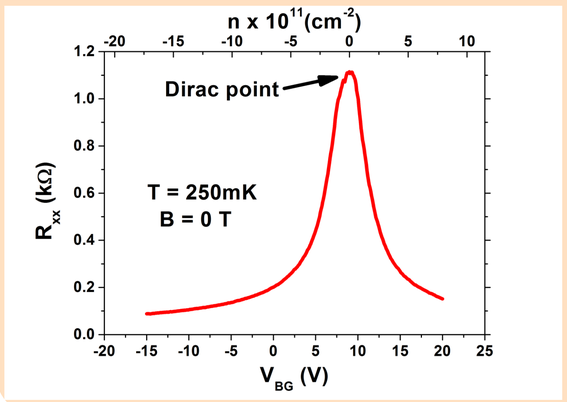
Even if exfoliated graphene remains good for fundamental research (due to its very high quality), its small size, random shape and poor scalability hinder its application in technology. On the other hand, the CVD growth technique produces much larger graphene samples with excellent growth speed and scalability. The drawback is the lower graphene quality, because it is typically polycrystalline and with more defects and contamination. This results in a lower mobility and in an unwanted doping that negatively affect the electrical properties of the CVD graphene. Our approach is to use CVD-grown single crystal graphene to combine the excellent quality of exfoliated graphene and the excellent scalabity of CVD graphene. In our experiment, we fabricated several Hall bars in which we measured electron transport at low temperature to prove the good electronic quality of our CVD single crystal graphene[1].

CVD-grown single crystalline graphene

Our graphene is grown in a commercial reactor on a copper substrate starting from CH_4. In the growth process we used ex-situ oxidized copper foils to dramatically increase the distance between the nucleation sites of the graphene on copper (up to several millimeters); then the growth is stopped before the flakes cross each other in order to avoid grain boundaries. The results are monocrystalline graphene flakes extending up to several millimeters. Afterwards the flakes are detatched from the substrate by a semi-dry technique (electrochemical delamination) in order to reduce defects and contamination of the graphene. The monolayer and the monocrystalline nature of the flakes are verified with Raman, SAED, XPS and LEED[2].

Sample



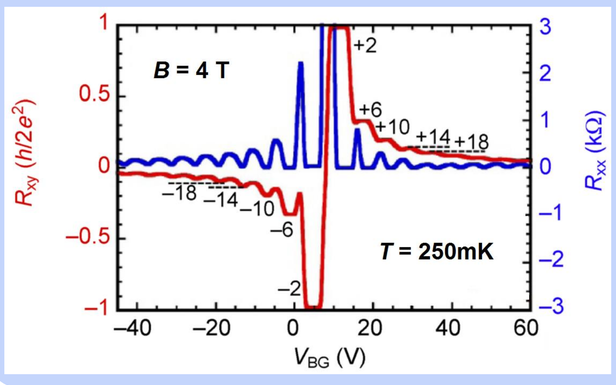


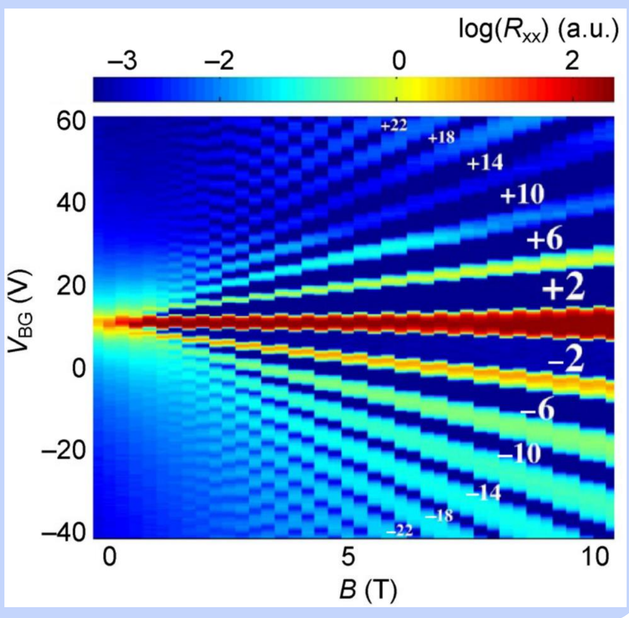
- The graphene flake (dark purple) is then transferred on 300nm of SiO_2 (n++ doped Si substrate used as a backgate).
- Hall bar contacts fabricated with EBL and Cr/Au thermal evaporation.
- Sample not annealed to measure the graphene properties «as is».
- With the four voltage probes, R_{xx} (longitudinal 4-wire resistance) and R_{xy} (transversal Hall resistance) are measured.
- From R_{xx} as a function of backgate voltage (V_{BG}) we extract the mobility (higher than $1.1x10^4 cm^2/Vs$) at 250mK.
- The intrinsic carrier concentration is also extracted: $n_0 = -6.5x10^{11} cm^{-2}$ at V_{BG}=0V (p-doped).

Quantum Hall regime

Already at B = 4 T, on R_{xy} (red), several well developed Quantum Hall (QH) plateaus are clearly visible and they are quantized at the values expected from half-integer QH for monolayer graphene, while R_{xx} shows the oscillations expected at the transition between two QH plateaus.

The color plot below, also called Landau fan, is measured at 250mK: we observe 12 well developed QH plateaus, and this confirms the good quality of our graphene.





Weak localization

Weak localization (WL) is a well estabilished phenomenon due to quantum interference that manifests as a peak in resistance at zero magnetic field. It requires that the elastic relaxation length (l_e) is shorter than the coherence length (l_φ).

Ideally, in graphene, due to the π Berry's phase, we expect weak anti-localization (WAL), but with elastic scattering mechanisms that violate chirality, WL is restored.

We used McCann's model to fit the WL peak[3]:

R_{xx} shifted and normalized with R_0 (resistance at B=0T)

$$\frac{R_{xx}-R_0}{R_0^2} = -\frac{e^2}{\pi h}\left[F\left(\frac{\tau_B^{-1}}{\tau_\varphi^{-1}}\right) - F\left(\frac{\tau_B^{-1}}{\tau_\varphi^{-1}+\tau_{iv}^{-1}}\right) - 2F\left(\frac{\tau_B^{-1}}{\tau_\varphi^{-1}+\tau_*^{-1}}\right)\right]$$

$F(z) = \ln(z) + \psi(0.5 + z^{-1})$ $\quad \psi(x) =$ Digamma function

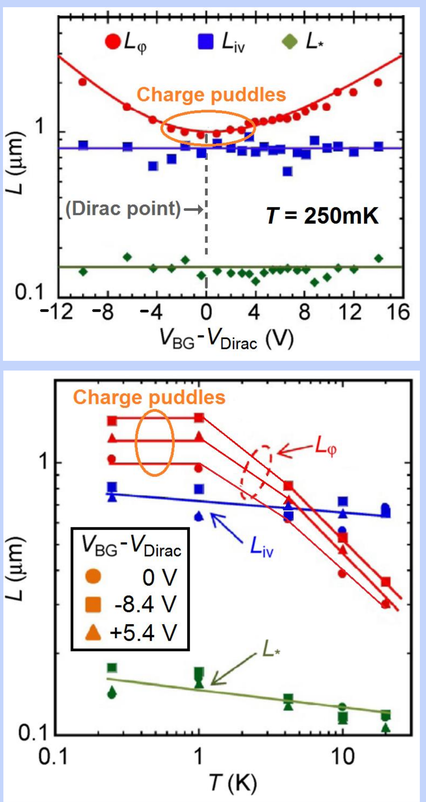
$\tau_B = 4DeB/\hbar$ $\quad D =$ Diffusion coefficient

τ_φ = dephasing time (inelastic)
τ_{iv} = intervalley scattering time (elastic)
τ_* = intravalley scattering time (elastic)

Then the characteristic lengths are calculated from the times extracted from the fit.

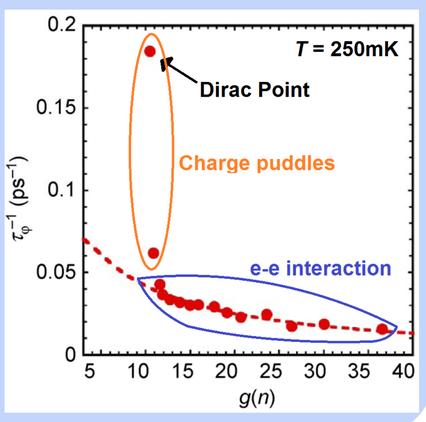
L_{iv} and L_* show no dependence on V_{BG} and only a weak dependence on temperature; on the contrary, L_φ has a strong dependence on both.

It is well known that close to the Dirac point there is a formation of charge puddles in the graphene, and the size of these puddles is the smallest at the Dirac point. These puddles limit the coherence length, because it is like reducing the effective size of the sample. This explains the minimum in L_φ at the Dirac point and also why L_φ has a V_{BG}-dependent saturation at low temperature.



The Nyquist formula: $\frac{1}{\tau_\varphi} \propto k_B T \frac{\ln(g)}{\hbar g}$

relates τ_φ to the conductance g (in units of e^2/h) only by using the e-e interaction as an inelastic scattering mechanism. Far from the Dirac point, where the fit is in excellent agreement with the data, the e-e interaction is the main inelastic scattering mechanism. On the contrary, at the Dirac point, the charge puddles limit the coherence as confirmed also by the poor agreement between the data and the Nyquist formula. Finally, the prominence of the e-e interaction is further confirmed also at higher T by the $\tau_\varphi \propto T^{-1}$ behaviour (data not shown).



Conclusions

We studied CVD-grown single crystal graphene on SiO_2 in electron transport at low temperature. We measured high mobility and low intrinsic carrier concentration (with respect to polycrystalline CVD graphene). In the Quantum Hall regime we observed 12 well developed plateaus.

We used weak localization to measure the coherence length which is found to be always above 1µm at 250mK. We also found that far from the Dirac point, the e-e interaction is the main inelastic scattering mechanism.

All these results show that the quality of our graphene is comparable to what is observed with exfoliated graphene, but with the advantages of the excellent scalability typical for CVD graphene.

References

[1] S. Xiang, V. Miseikis, L. Planat, S. Guiducci, S. Roddaro, C. Coletti, F. Beltram, S. Heun, Nano Research 2016, 9 (6): 1823-1830.
[2] V. Miseikis et al. 2D Mater. 2 (2015) 014006.
[3] E. McCann et al., Phys. Rev. Lett. **97**, 146805 (2006).