Strong localization in a suspended monolayer graphene by intervalley scattering

Cenk Yanik,1, 2 Vahid Sazegari,1, 2 Abdulkadir Canatar,1, 2 Yaser Vaheb,1, 2 and Ismet I. Kaya1, 2, *

1 Faculty of Engineering and Natural Sciences, Sabanci University, Tuzla, 34956 Istanbul, Turkey
2 Sabanci University Nanotechnology Research and Application Center, Tuzla, 34956 Istanbul, Turkey

(Dated: June 23, 2020)

A gate induced insulating behavior at zero magnetic field is observed in a high mobility suspended monolayer graphene near the charge neutrality point. The graphene device initially cleaned by a current annealing technique was undergone a thermo-pressure cycle to allow short range impurities to be adsorbed directly on the ultra clean graphene surface. The adsorption process generated a strong temperature and electric field dependent behavior on the conductance of the graphene device. The conductance around the neutrality point is observed to be reduced from around \( e^2/h \) at 30 K to \( \sim 0.01 e^2/h \) at 20 mK. A direct transition from insulator to quantum Hall conductor within \( \approx 0.4 T \) accompanied by broken-symmetry-induced \( \nu = 0, \pm 1 \) plateaux confirms the presence of intervalley scatterers.

The nature of the conductivity at Dirac point has been debated since graphene’s first isolation [1]. One of the most important applications of graphene would be in digital electronics if it could be made to have depleteable conductance while maintaining its high mobility. However, in graphene the on/off resistance ratio is hindered by potential fluctuations generally attributed to unintentional doping where minimum conductance is limited by saturation of the average carrier density in the presence of so-called electron-hole puddles. Even ultra-clean high mobility suspended monolayer graphene samples have been observed to have a minimum conductivity [2–6] complying with the theoretical ballistic limit (\( 4e^2/\pi h \)) [7, 8]. On the other hand, insulating behavior around Dirac point has been observed in double-layer graphene heterostructures [9] or in top-gated graphene sheets on hBN substrates [10] by screening the charge puddles.

According to the scaling theory of localization, when the spatial symmetry of a two dimensional system is broken, its conductivity tends to zero. In presence of impurities that have potential range extending much longer than the lattice constant, symmetry is preserved and there is no mixing between K and K’ points in the band structure of graphene. This leads to a positive correction to the conductivity and anti-localization is predicted [11, 12]. On the other hand, by the addition of short range impurities the symmetry is broken and intervalley scattering is allowed. In general, there are two scattering mechanisms for Dirac fermions in graphene, intra-valley and inter-valley scattering. In the presence of long-range disorder potentials, as in the case of graphene on Si substrate, the electrons scatter in each of the two valleys without backscattering [13–15]. However, with short-range or strong long-range disorders [16], e.g., in graphene on hBN or suspended graphene, the dominant scattering is inter-valley scattering which gives rise to backscattering and localization [9–12, 17–22].

Here we report the first time observation of an insulating behavior in a suspended monolayer graphene around its charge neutrality point at zero magnetic field. This peculiar behavior, characterized by highly temperature-dependent strong conductance fluctuations, is mediated by the valley symmetry breaking and attributed to the presence of short-range disorders. The inter-valley scattering length is estimated to be \( l_{iv} \approx 0.1 \mu m \) by gate and temperature dependent measurements as well as the magnetotransport data.

The suspended graphene sample was treated by a two-step procedure that involved removal of long range scatterers followed by deposition of short range scatterers. The sample was first cleaned by a current annealing scheme, through which the graphene sheet and the contact probes were annealed concurrently [23], to the point that a very sharp conductance dip is obtained (Fig. 1). Organics and residues left on graphene are known to generate long range density fluctuations in the form of electron-hole puddles which effectively saturate average carrier density and making the Dirac point unaccessible. The simultaneous annealing of probe and graphene helps to achieve a uniform temperature profile over a graphene sheet at low temperature in a vacuum chamber. This allows a thorough cleaning of graphene from the contamination stuck on it before and during the fabrication process.

In the second step the sample was let undergo a thermal cycle which also caused a brief and mild loosening of the vacuum level in the chamber. An insulating behavior was acquired after the thermo-pressure cycle (TPC) of the high quality ultra-clean suspended graphene sample. In addition to the normal sequence of quantum Hall plateaus for single layer graphene, magnetoresistance measurements reveal emergence of indisputable \( \nu = 0, \pm 1 \) plateaus as a result of broken valley and spin symmetries [5]. This is interpreted to be due to the presence of strong short range scatterers that break the valley symmetry in an ultra-clean graphene sheet.

The graphene sample used in these experiments was mechanically exfoliated from a natural graphite and then transferred on to a p doped Si substrate covered by 285 nm of SiO\(_x\). Single-layer flakes were identified based
FIG. 1. The conductance at zero field (blue) and at $B = 0.3\, T$ (red) as a function of carrier concentration, measured after current annealing of the suspended graphene but before it is underwent the thermo-pressure cycle. Device has channel length $L = 1\, \mu m$ and width $W = 2\, \mu m$. The lead resistance $R_C = 0.9\, k\Omega$ is estimated from the quantum Hall plateaus and subtracted in the plots. Inset shows the optical microscope image of the measured device. Measurements were performed between the probes labelled as S and D with $I_S = 10\, nA$ applied current at 1.5 K. Dashed lines mark the borders of the suspended graphene. Scale bar is 1 $\mu m$.

on their contrast under the optical microscope and confirmed by Raman spectroscopy. Electron beam lithography is employed to pattern the electrical contacts made from Cr/Au (3/100 nm) followed by a lift-off in acetone. Suspension is achieved by dipping the SiO$_x$ in a buffered oxide etcher (BOE) to remove 185 nm of SiO$_x$ layer. Subsequently the device was transferred into DI water and isopropyl alcohol followed by a gentle nitrogen dry. Electrical measurements were done in a dilution refrigerator with a magnet using standard lock-in techniques. The sample and the metallic leads were annealed at 1.5 Kelvin by passing independently controlled DC currents through them. This technique allowed heating of both graphene and leads independently to sufficiently high temperatures and prevented accumulation of residues to accumulate near the leads. The details of the annealing procedure is provided in Ref. [23]. The annealing is done in repetitive current ramps with increased max current until the resistance peak shifted to near zero gate bias indicating low unintentional doping.

The conductance of the sample after current annealing is displayed as a function of the gate voltage, $V_g$ and the carrier concentration, $n$ in Fig. 1. Parallel-plate capacitor model is used to determine the variation of $n$ with respect to the gate voltage as $n(V_g) = \alpha V_g$ where the value of the coupling factor is determined as $\alpha = 2.7 \times 10^{10}\, V^{-1}\, cm^{-2}$; this value is consistent with the one estimated obtained from the quantum Hall (QH) measurements.

As shown in Fig. 1, the sample was confirmed to be a monolayer graphene via QH measurements where conductance exhibits well developed quantized plateaus at $\nu = \pm 2, \pm 6, \pm 10$ at a magnetic field as small as 0.3 T. Taking the aspect ratio of $W/L = 2$ into consideration, it should be noted that the peak resistivity of the suspended graphene sample after current annealing ($\sim 50\, k\Omega$) is well above the resistance quantum $h/e^2$ which is a hallmark of extremely clean samples with substantially reduced electron-hole puddles [24]. In an ultraclean graphene sample, the conductance can be suppressed well below $e^2/h$ if the average charge density is sufficiently reduced near the neutrality point. In other words, it is the sat-
urated carrier density around the Dirac point due to the presence of electron-hole puddles that determines the minimum of conductance in graphene.

The sample is then taken through an in-situ thermapressure cycle from mK to 200 K and then back to mK along with loosening of vacuum up to $10^{-2}$ mBar after which it adopted strong conductance fluctuations leading to an insulating behavior around the charge neutrality point with mega-ohm resistance peaks. We believe that the ultraclean sample was disordered during the thermapressure cycle by some adsorbents accompanying strong short-range potentials leading to pronounced conductance fluctuations and intervalley backscattering [11, 12]. The conductance exhibits strong fluctuations as the charge density is varied and an insulating behavior at low density regime. In Fig. 2(a), the conductance as a function of gate voltage is plotted at various temperatures up to 30 K. The conductance fluctuations are strongly dependent on temperature especially around the neutrality point and are remarkably suppressed at higher temperatures. A comparison between the gate-dependent conductance before and after the TPC is illustrated in Fig. 2(b). The adsorption of atomic impurities during the TPC caused a suppression of conductance along with strong fluctuations around the Dirac point.

As the temperature is lowered, the transport of electrons becomes coherent and leads to quantum interference corrections to the conductance. In Fig. 3(a), the relative fluctuations of conductance is illustrated in low density regime at different temperatures. It can be seen that as the temperature is decreased the fluctuations in the conductance are strongly pronounced especially around the Dirac point generating the insulating dips seen in Fig. 2(a). The fluctuations are reproducible at different temperatures while intensifying at lower temperatures such that they can diminish the conductance occasionally around the Dirac point and lead to an insulating behavior when the carriers are totally localized in the bulk. Suzuura and Ando [11] showed that in two-dimensional honeycomb lattice the quantum interference correction to the Boltzmann conductivity is given by $\Delta \sigma = \pm (e^2/\pi \hbar) \log(l_\phi/l_e)$, where $l_\phi$ and $l_e$ are the coherence and elastic scattering lengths, respectively. In the case of long-range disorders, the backscattering is forbidden thus the correction is positive whereas in the presence of short-range potentials the intervalley scattering becomes probable and lead to a negative correction to conductivity. The coherence length decreases at higher temperatures, suppress the quantum interference effects and lead to a logarithmic temperature behavior for the conductance correction. The average value of change in the conductance at different temperatures is plotted in Fig. 3(b) which shows a logarithmic suppression of the negative conductance correction for $T \geq 2$ K. Below 2 K, the conductance correction starts to saturate. This is the temperature below which the coherence length exceeds the sample size thus saturates the conductance.

Since the insulating behavior is observed for $|n| \leq 10^{11}$ cm$^{-2}$ and the localization requires a mean free path of the order of the Fermi wavelength, $\lambda_F = (4\pi/n)^{1/2}$, we can estimate the mean free path for intervalley scattering as $l_\text{iv} \sim 0.1$ μm. A similar length scale is also inferred from the magnetotransport data presented in Fig. 4. The field at which the sample transitions to the quantum Hall state ($B \sim 0.4$ $T$) gives a length scale $(\phi_0/B)^{1/2} \approx 0.1$ μm corresponding to a flux quantum $\phi_0 = h/e$ enclosed by cyclotron orbits which sets a minimum on the intervalley scattering length.

The intervalley scattering is also manifested in the quantum Hall regime. Fig. 4 shows the conductance as a function of carrier density at various magnetic fields from 0 to 2 tesla. A direct transition from the insulating behavior around the Dirac point to quantum Hall regime is observed around 0.4 $T$ where a single conduc-
scattering brought the sample into a completely insulating regime near Dirac point. Lifting of the valley symmetry due to strong inter-valley scattering was also reflected in the quantum Hall measurements as $\nu = 0, \pm 1$ plateaus appearing at relatively small fields of $\sim 1 \, \text{T}$. The authors would like to thank İnci Adagideli for his fruitful discussions. This work is funded by Scientific and Technological Research Council of Turkey (TUBITAK) under Project Grant No. 112T990.

1. K. S. Novoselov et al., Science 306, 666 (2004).
2. X. Du, I. Slachko, A. Barker, and E. Y. Andrei, Nat. Nano 3, 491 (2008).
3. K. I. Bolotin et al., Solid State Communications 146, 351 (2008).
4. X. Du, I. Slachko, F. Duerr, A. Luican, and E. Y. Andrei, Nature 462, 192 (2009).
5. K. I. Bolotin et al., Nature 462, 196 (2009).
6. A. S. Mayorov et al., Nano Letters 12, 4629 (2012).
7. M. I. Katsnelson, European Physical Journal B 51, 157 (2006).
8. J. Tworzydo, B. Trauzettel, M. Titov, A. Rycerz, and C. W. J. Beenakker, Phys. Rev. Lett. 96, 246802 (2006).
9. L. A. Ponomarenko et al., Nat. Phys. 7, 958 (2011).
10. F. Amet, J. R. Williams, K. Watanabe, T. Taniguchi, and D. Goldhaber-Gordon, Phys. Rev. Lett. 110, 216601 (2013).
11. H. Suzuura and T. Ando, Physical review letters 89, 266603 (2002).
12. H. Suzuura and T. Ando, Journal of the Physical Society of Japan 72, 69 (2003).
13. P. Ostrovsky, I. Gornyi, and A. Mirlin, Physical review letters 98, 256801 (2007).
14. N. H. Shon and T. Ando, Journal of the Physical Society of Japan 67, 2421 (1998).
15. A. Morpurgo and F. Guinea, Physical review letters 97, 196804 (2006).
16. Y.-Y. Zhang, J. Hu, B. A. Bernevig, X. Wang, X. Xie, and W. Liu, Physical review letters 102, 106401 (2009).
17. B. Kramer and A. MacKinnon, Reports on Progress in Physics 56, 1469 (1993).
18. M. Imada, A. Fujimori, and Y. Tokura, Reviews of modern physics 70, 1039 (1998).
19. F. Evers and A. D. Mirlin, Reviews of Modern Physics 80, 1355 (2008).
20. S. Gattenlöhner, W.-R. Hannes, P. Ostrovsky, I. Gornyi, A. Mirlin, and M. Titov, Physical review letters 112, 026802 (2014).
21. J. Moser, H. Tao, S. Roche, F. Alzina, C. S. Torres, and A. Bachtold, Physical review B 81, 205445 (2010).
22. N. Peres, Reviews of modern physics 82, 2673 (2010).
23. A. Canatar, C. Yanik, V. Sazgari, and I. I. Kaya, To be submitted.
24. S. Das Sarma, E. H. Hwang, and Q. Li, Phys. Rev. B 85, 195451 (2012).
25. S. Das Sarma, S. Adam, E. H. Hwang, and E. Rossi, Rev. Mod. Phys. 83, 407 (2011).