Unusual transport properties in carbon based nanoscaled materials: nanotubes and graphene

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The massless Dirac particle moving at the speed of light has been a fascinating subject in relativistic quantum physics. Nanoscale graphitic materials, such as carbon nanotubes and graphene, now provide us with an opportunity to investigate such exotic effects in low-energy condensed matter systems. The unique electronic band structure of graphene lattice provides a linear dispersion relation where the Fermi velocity replaces the role of the speed of light in the usual Dirac Fermion spectrum. Recent experimental studies reveal that such unconventional electronic structure in graphitic carbon leads to unique electronic transport phenomena in 1-dimensional carbon nanotubes and 2-dimensional graphene. Combined with semiconductor device fabrication techniques and the development of new methods of nanoscaled material synthesis/manipulation enables us to investigate mesoscopic transport phenomena in these materials. The exotic quantum transport behavior discovered in these materials, such as room temperature ballistic transport, unusual half-integer quantum Hall effect, and a non-zero Berry's phase in magneto-oscillations will be discussed in the connection to Dirac Fermion description in graphitic systems.

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1 Introduction

Graphene is a single atomic sheet of graphitic carbon atoms that are arranged into a honeycomb lattice. There are two carbon atoms in one unit cell where each carbon atom sits in two interpenetrated triangular lattices with inversion symmetry between them. This unique topology of hexagonal arrangement of carbon atom renders an unusual energy dispersion relation at the Fermi energy in graphene. Figure 1 displays the real space graphene lattice and the energy band structure of graphene obtained by simple tight binding model. There are two bands: the valence band (\( E < 0 \)) and the conduction band (\( E > 0 \)) touched at six points at the Brillouin zone corners of which two sets of points are inequivalent. These two points in reciprocal lattice (\( \mathbf{K} \) and \( \mathbf{K}' \)) represent different linear superposition of Bloch wave functions in two real space sublattices. The energy dispersion near these points is particularly interesting, where the 2-dimensional (2D) energy spectrum is linear, and thus the electrons always move at a constant speed, the Fermi velocity \( v_F \). This is in perfect analogy to a relativistic massless particle, e.g. photons or massless neutrinos. Therefore the electron dynamics in graphene is effectively ‘relativistic’, where the speed of light is substituted by \( v_F \). Such distinct energy dispersion with unusual Fermi surface topology provides non-trivial Berry’s phase, the effective chirality of the Bloch wave function in two sublattices. In fact, this chiral nature of carriers in graphene has been speculated to have an important implication in the electronic transport in graphitic materials including single walled carbon nanotubes (SWNTs) [1]. In this

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paper we will discuss experimental consequences of the Dirac Fermion spectrum in charge transport, realized in two representative low dimensional graphitic carbon systems: 1-dimensional carbon nanotubes and 2-dimensional graphene.

2 Extremely long mean free path in carbon nanotubes

Electron transport in carbon nanotubes are an intense area of interest because of its interesting one-dimensional behaviours. A fundamental quantity associated with transport is the electron mean free path, $L_m$. At low bias, the selection rules that stem from the Berry’s phase discussed in Section 1 make $L_m$ in SWNTs extremely long, provided the electron scattering is governed by atomically smooth scattering potential [1, 2]. Previous transport measurement in the linear response regime suggested the lower bounds for $L_m$ to be $\sim 0.5–1.5 \, \mu m$ at room temperature [3, 4] and $\sim 4 \, \mu m$ at low temperatures [4]. In order to probe $L_m$ directly, we have studied the scaling of resistance ($R$) of individual SWNTs as a function of channel length $L$ [5]. Utilizing ultra-long SWNTs growth [6], we have fabricated multi-terminal devices with channel lengths ranging from 100 nm to 400 $\mu m$ (Fig. 2a). For each pair of electrodes, the linear response conductance was measured at large negative gate voltage ($V_g < -10 \, V$) away from the band gap region of semiconductor or (curvature induced) small gap nanotubes. The measured resistance scales linearly with the channel length if $L_m / L_{loc} \ll L$, where the localization length $L_{loc} \approx 10 \, \mu m$, for most of SWNTs. Figure 2b shows the resistance for an individual SWNT as a function of length. At low temperatures, the resistance is a relatively constant value approaching $h/4e^2$ for small channel length ($L$). This is a signature that the electrons propagate ballistically, indicating $L < L_m$. At longer lengths ($L \sim L_m$), the resistance is linearly dependent on channel length, indicating diffusive behaviour. By fitting the $R(L)$ at different temperatures with the expected resistance scaling [3]: $R(L) = (h/4e^2) (1 + L/L_m)$, we obtained $L_m$ as a function of temperature. Figure 2c displays thus obtained $L_m(T)$. At temperatures lower than $\sim 20 \, K$, the $L_m$ shows a constant value $\sim 10 \, \mu m$, suggesting that static impurities are the main source of scattering, while the decreasing $L_m$ with increasing $T$ indicates the inelastic scattering by phonon might be the major scattering mechanism. Note that even at room temperature, $L_m \sim 0.5 \, \mu m$, implying that the transport in SWNT segments whose length is up to $0.5 \, \mu m$ remains to be ballistic even at room temperature. Such extremely long electron mean free path in the presence of electron–phonon scattering at elevated temperatures is a direct consequence of selection rules associated with the Berry’s phase in underlying graphene band topology that the SWNTs electronic structures are based on [1].
3 Extraction of graphene from bulk single crystals

Recently, graphene has attracted a sudden burst of much attention ever since the development of the efficient experimental method of extracting single atomic layers of graphite via mechanical extractions [7]. This method involves a very simple, yet efficient method for mechanical extractions of mesoscopic graphite pieces. First, the single crystals of graphite is fixed on an adhesive tape and then cleaved to expose a fresh cleaved surface. The next step is to transfer very thin flakes of graphite pieces from the crystals to the surface of silicon wafers by contacting the cleaved surface of the single crystals. Typically, various thicknesses including single layers of graphite with the lateral size 1–100 µm pieces are deposited on the surface of SiO$_2$/Si substrate. In order to measure the thickness of the deposited graphene layers, a cross-correlation study between optical microscope and AFM images was made. As shown in Fig. 3a, there are color contrast changes in the samples with different thicknesses due to interference induced color shifts. These color shifts are sensitive to the number of graphene layers on top of the SiO$_2$/Si substrate. We also note that the thickness of SiO$_2$ layer affects the contrast between different layers. 300 nm thick thermally grown SiO$_2$ layer yields the best contrast for this purpose. Once the optical images are taken, the sample profiles are probed using various AFMs (MultiMode, Digital Instrument and XE100, PSIA). Figure 3b shows an AFM image of the samples in the boxed region in Fig. 3a. The detailed AFM height profile analysis (inset) shows that the apparent height of region (I) and (II) are

![Fig. 3](online colour at: www.pss-b.com) Cross-correlation of optical microscope (a) and AFM images (b) of few layer graphite samples. (c) Optical microscope image of a typical single layer graphene device.
0.8 nm and 1.2 nm, respectively. A step of one atomic layer height (~0.4 nm) is observed between regions (I) and (II), indicating the region (II) has an additional layer of graphene. Since a single sheet of graphene is 0.35 nm thick, the region (I) is comprised of at most two graphene layers. However, considering an additional van der Waals (vdW) distance between graphene and the SiO$_2$ surface (~0.3 nm), we speculate that the region (I) is most likely a single layer, unless the AFM tip to graphene interaction is much stronger than that of the AFM tip to SiO$_2$ interaction. This is highly unlikely, since AFM profile analysis has been successfully used for determining diameter of carbon nanotubes, a material that share much similarity with graphene, where diameters are confirmed by well established cross-correlation between AFM and Raman spectroscopy [8]. Typically, graphene pieces of lateral size ~3–10 µm are chosen for device fabrication. Multiple electrodes arranged in Hall-bar or Van der Paaw geometry, are fabricated on the sample using electron beam lithography followed by Au/Cr (30/3 nm) evaporation and a lift-off process (Fig. 3c). The degenerately doped silicon substrate serves as a gate electrode with 300 nm thermally grown silicon oxide acting as the gate dielectric. By applying a gate bias voltage, $V_g$, the charge density of the sample can be tuned.

### 4 Unusual quantum Hall effects in graphene

In addition to the AFM profile analysis discussed above, there is evidence that the region (I) and (II) in Fig. 3 corresponds to a single and double layer graphene, respectively. We measured magnetoresistance ($R_{xx}$) and Hall resistance ($R_{xy}$) of the samples that have the same color shift corresponding to region (I) and (II) as a function of gate voltage. The quantum Hall effects (QHE) are evident in both types of samples as we observe the quantized Hall resistance to $R_{xy} = h/(e^2\nu)$, where $\nu$ is integer filling factor, and vanishing $R_{xx}$ in certain region of $V_g$. Typical devices made from material from region (II), the ‘double’ layer samples, exhibit a ‘conventional’ QHE, where the quantization condition is described by $\nu = g_s n$ with integer $n$ and the degeneracy of Landau level $g_s = 4$, counting spin degeneracy and sublattice symmetry (Fig. 4a). While the QHE in double layer graphene shows a similar quantization rules as integer QHE in conventional 2D electron gas in semiconductor hetero junctions, the QHE observed in ‘single’ layer graphene is distinctly different [9–11]. Figure 4b displays magnetoresistance and Hall resistance as a function of $V_g$ in a single layer graphene device. Rather than integer multiple filling factors observed in double layers, quantized Hall plateaus shows their filling factors described by half integer multiples: $\nu = g_s (n + 1/2)$.

The sequence of half-integer multiples of quantum Hall plateaus has been predicted by several theories which combine ‘relativistic’ Landau levels (LL) with the particle-hole symmetry of graphene [13]. This can be easily understood from the calculated LL spectrum, $E_n = \text{sgn} (n) \sqrt{2eBn^2|n|}$, where $n$ is

![Fig. 4](online colour at: www.pss-b.com) Magnetoresistance ($R_{xx}$) and Hall resistance ($R_{xy}$) measured as a function of gate voltage $V_g$ for (a) double layer graphene and (b) single layer graphene samples (reproduced from Ref. [10]). The data was taken at temperature 1.7 K with the magnetic field 9 T.
Landau level index for electron \((n > 0)\) and hole \((n < 0)\). Here we plot the density of states (DOS) of the \(g_s\)-fold degenerate (spin and sublattice) LLs and the corresponding Hall conductance \(\sigma_{xy}\) in the quantum Hall regime as a function of energy. Since \(\sigma_{xy}\) is related to the measured \(R_{xx}\) and \(R_{xy}\) by:
\[
\sigma_{xy} = -\frac{R_{xy}}{R_{xy}^2 + W^2 R_{xx}^2 L^2},
\]
where \(W\) and \(L\) is the width and length of the samples, respectively, \(\sigma_{xy}\) exhibits QHE plateaus when \(E_F\) (tuned by \(V_g\)) falls between LLs, and jumps by an amount of \(g_s e^2/h\) when \(E_F\) crosses a LL. Electron–hole symmetry imposed in the graphene system requires \(\sigma_{xy}\) to be asymmetric in energy across the Dirac point. On the other hand, the \(n = 0\) LL is firmly fixed at the Dirac point, regardless of the magnetic field. Thus, the first plateau of \(R_{xy}\) for electron and hole are situated exactly at \(g_s e^2/2h\). As \(E_F\) crosses the next LL, \(R_{xy}\) changes by an amount of \(g_s e^2/2h\), which yields the quantization condition in observed in this experiment.

This unusual quantization condition is a result of the topologically exceptional electronic structure of graphene. A consequence of the combination of time reversal symmetry with the novel Dirac point structure can be viewed in terms of Berry’s phase arising from the band degeneracy point [1]. A direct implication of Berry’s phase in graphene is discussed in the context of the quantum phase of a spin-1/2 pseudo-spinor that describes the sublattice symmetry [2]. This phase is already implicit in the half-integer shifted quantization rules of the QHE. It can further be probed in the magnetic field regime where a semi-classical magneto-oscillation description holds [10, 11].

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**References**