Coulomb Oscillations and Hall Effect in Quasi-2D Graphite Quantum Dots

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ABSTRACT

We perform low-temperature electrical transport measurements on gated, quasi-2D graphite quantum dots. In devices with low contact resistances, we use longitudinal and Hall resistances to extract carrier densities of $9.2-13 \times 10^{12} \text{ cm}^{-2}$ and mobilities of $200-1900 \text{ cm}^2\text{V}^{-1}\text{s}$. In devices with high resistance contacts, we observe Coulomb blockade phenomena and infer the charging energies and capacitive couplings. These experiments demonstrate that electrons in mesoscopic graphite pieces are delocalized over nearly the whole graphite piece down to low temperatures.
VTI cryostat or 100 mK in an Oxford dilution refrigerator. The devices with low resistances at room temperature (open dots) displayed only a small increase in their resistance upon cooling, as seen in Figure 2. In such devices with multiple contacts, we performed longitudinal and Hall resistance measurements to extract the carrier density, sign of the carriers, and resistivity. Figure 3 shows data from a 5 nm tall dot, corresponding to approximately 15 stacked graphene sheets, measured at ~100 mK using standard AC lock-in techniques. Similar results were obtained at 4 and 1.5 K. The Hall resistance is approximately linear, and the longitudinal resistance shows weak fluctuations as a function of magnetic field with little change in its average value.

To analyze these results, we make the simplifying assumption that the entire graphite piece is a uniform conductor with a single density and in-plane mobility. From the standard equation for the Hall resistance \( R_{\text{H}} = B/\mu n e \), the slope of the line in Figure 3 corresponds to a density of \( 9.2 \times 10^{12} \text{ cm}^{-2} \). The longitudinal resistance shows only weak fluctuations as a function of magnetic field with little change in its average value.

After accounting for the geometrical factors, we infer the resistance per square, \( R_\text{s} \), of the entire sample. For the 5 nm thick device at 100 mK, we have \( R_\text{s} = 3.4 \text{ k}\Omega \). Using the equation \( \mu = 1/neR_\text{s} \), we get a mobility of \( \mu = 200 \text{ cm}^2/\text{V}\cdot\text{s} \). A similar analysis for the sample with a thickness of 18 nm shown in Figure 1a and 1c, gives a hole density of \( 1.3 \times 10^{13} \text{ cm}^{-2} \).

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We can use a gate to vary the carrier density in the graphite quantum dot. We assume the capacitance to the gate is that of a parallel plate capacitor, $C_g = e_o A/d$, where $d = 200$ nm is the thickness of the SiO$_2$, $e_o$ is the permittivity of free space, $e$ is the dielectric constant of SiO$_2$, and $A$ is the area of the device. This gives a capacitance per area of $C_g' = 1.8 \times 10^{-8}$ F/cm$^2$, implying that 10 V applied to the back gate results in a change of density of $1 \times 10^{12}$ holes/cm$^2$. This is only a small fraction of the total density in even the thinnest samples studied. Nevertheless, it is consistent with a small decrease in conductance observed in many samples at room temperature; the holes are slightly depleted by the gate. At low temperatures, any such changes are obscured by reproducible fluctuations in the conductance as a function of $V_g$.

Devices with room-temperature two-point resistances greater than 20 k$\Omega$ (closed dots) show Coulomb blockade at low temperatures. Data from a device fabricated using the random electrode method is shown in Figure 4. At $T = 100$ mK, the conductance exhibits well-defined Coulomb blockade oscillations with a period in gate voltage of $\Delta V_g = 1.5$ mV. A plot of $dI/dV_g$ vs $V_g$ and $V_{sd}$ is shown in Figure 4. The maximum voltage that could be applied and still be in the blockade regime is $\Delta V_{sd} = 0.06$ mV.

A device made by the designed electrode method is shown in Figure 5. The thickness of the device is 6 nm, corresponding to 18 sheets. The Coulomb blockade oscillations have a period of $\Delta V_g = 11$ mV and a maximum blockade voltage of $\Delta V_{sd} = 0.3$ mV. A third device fabricated by the random electrode method shown in Figure 1d has a height of 5 nm and shows Coulomb oscillations with a period in gate voltage of $\Delta V_g = 1.3$ mV.

To describe these results, we use the semiclassical theory of the Coulomb blockade. The period of the Coulomb oscillations in gate voltage is given by: $\Delta V_g = eC_g/e_o$ and, using the previous expression for $C_g$, we can approximate the area of the graphite quantum dot. For the device in Figure 5 with $\Delta V_g = 11$ mV, the expected area of dot is $A = 0.08 \mu$m$^2$. The measured total area of the graphite piece shown is $0.12 \mu$m$^2$ while the area between the electrodes is $0.05 \mu$m$^2$. This demonstrates that nearly the entire graphite piece is serving as a single quantum dot and it likely extends beyond the electrodes. For the device shown in Figure 1d, the measured gate voltage period is $\Delta V_g = 1.3$ mV, which corresponds to a quantum dot with $A = 0.70 \mu$m$^2$. The area between the electrodes is $0.45 \mu$m$^2$ again implying that the dot extends into the graphite piece lying under the electrodes.

The charging energy for the dot is determined by its total capacitance $C$ and is equal to the maximum blockade voltage observed: $eC = \Delta V_{sd}$. Notably, for our graphite quantum dots, the ratio of the charging energy to the gate voltage period is small: $\alpha = (e/C)/\Delta V_g = C_g/(C_s + C_d + C_g) \ll 1$, where $C_s$ and $C_d$ are the capacitances to the source and drain electrodes. The devices shown in Figure 4 and Figure 5 have $\alpha = 1/25$ and $1/40$, respectively. Such small values imply

**Figure 4.** (a) Current as a function of gate voltage with $V_{sd} = 10 \mu$V at $T = 100$ mK for a device fabricated by the random electrode method. Coulomb oscillations are observed with a period in gate voltage of $\Delta V_g = 1.5$ mV. (b) The differential conductance $dI/dV_{sd}$ plotted as a color scale versus $V_{sd}$ and $V_g$. Blue signifies low conductance and red high conductance. The charging energy of the quantum dot is equal to the maximum height of the diamonds: $\Delta V_{sd} = 0.06$ mV. The center-to-center spacing between the diamonds is the Coulomb oscillation period $\Delta V_g = 1.5$ mV.

**Figure 5.** (a) The differential conductance $dI/dV_{sd}$ plotted as a color scale versus $V_{sd}$ and $V_g$ for the device shown in (d) at $T = 100$ mK. The charging energy is $\Delta V_{sd} = 0.3$ mV and the gate voltage period is $\Delta V_g = 11$ mV. (b) Current as a function of gate voltage with $V_{sd} = 10 \mu$V at $T = 100$ mK for the device shown in (d). Coulomb blockade oscillations are seen with a period of $\Delta V_g = 11$ mV. (c) Current as a function of gate voltage versus magnetic field with $V_{sd} = 10 \mu$V at $T = 100$ mK for the device shown in (d). (d) AFM image of a device fabricated using the designed electrode method. The white rectangular outlines show the position and size of the electrodes that were evaporated on the device. The total area of the graphite piece is $0.12 \mu$m$^2$ and the area under the source and drain electrodes is $0.013 \mu$m$^2$ and $0.015 \mu$m$^2$, respectively. The area of the graphite piece between the source and drain electrodes is $0.05 \mu$m$^2$. (inset) Line trace showing the 6 nm height of the graphite.
that the graphite pieces have a much greater capacitive coupling to the source and drain electrodes than to the gate.

We can estimate the source and drain capacitance per unit area $C_{s,d}$ using the measured charging energies and the area of the electrodes over the graphite. For the device shown in Figure 5, this yields a capacitance per unit area of $C_{s,d} = 2 \times 10^{-6}$ F/cm$^2$. Using the parallel plate capacitor equation, $C = \varepsilon_0 \varepsilon_r / d$ yields $d \varepsilon_r = 0.5$ nm. This is consistent with a very thin tunnel barrier between the electrode and the graphite. The origin and nature of this barrier is unknown. Since it is only present in the few samples that show Coulomb blockade oscillations (closed dots), it is likely the result of a contamination layer between the metal and the graphite.

We also examined the magnetic field dependence of the Coulomb blockade oscillations (Figure 5c). The closed dot at $B = 0$ T has well-defined oscillations. As the magnetic field is increased, the peaks evolve in a complicated fashion. Most notably, the oscillations no longer go to zero, suggesting that the dot becomes more open. The open dot shows complex fluctuations in the peak positions as a function of magnetic field. Other devices showed similar transitions from closed to more open dots along with changes in the peak positions as a function of field. The origin of these effects is under investigation.

In conclusion, we fabricated and measured quasi 2-D graphite quantum dots. In devices with good contacts (open dots), we performed resistivity measurements and extracted carrier densities of $9.2 - 13 \times 10^{12}$ cm$^{-2}$ and mobilities of 200–1900 cm$^2$/V·s. In the case of tunnel contacts (closed dots), we observed Coulomb charging phenomena and inferred the gate and source-drain capacitances. Future studies will investigate the nature and role of interlayer coupling between the sheets and explore the single-particle energy level spectrum and the effects of a magnetic field. Studies on devices with a variety of thicknesses and improved control over the contacts will help address these issues.

**Note Added:** During preparation of this manuscript, we became aware of three other groups$^{16-18}$ reporting transport measurements of thin graphite layers that are related to our measurements of open dots (Figure 3).

**References**