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Gap opening in the zeroth Landau level of graphene

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We have measured a strong increase of the low-temperature resistivity ρ_{xx} and a zero-value plateau in the Hall conductivity σ_{xy} at the charge neutrality point in graphene subjected to high magnetic fields up to 30 T. We explain our results by a simple model involving a field dependent splitting of the lowest Landau level of the order of a few Kelvin, as extracted from activated transport measurements. The model reproduces both the increase in ρ_{xx} and the anomalous $\nu=0$ plateau in σ_{xy} in terms of coexisting electrons and holes in the same split zero-energy Landau level.

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In a magnetic field, graphene displays an unconventional Landau-level spectrum of massless chiral Dirac fermions.[1–4] In particular, a Landau level shared equally between electrons and holes of opposite chirality exists at zero-energy around the charge neutrality point (CNP). Due to the coexistence of carriers with opposite charge, graphene behaves as a compensated semimetal at the CNP with a finite resistivity ρ_{xx} and a zero Hall resistivity ρ_{xy}.

Recently, the nature of the CNP in high magnetic fields has attracted considerable theoretical interest (see Ref. 5 and references therein). Experimentally, in the metallic regime, the transport behavior around the CNP can be explained using counter-propagating edge channels.[6] On the other hand, the high-field resistivity at the CNP was shown to diverge strongly, an effect recently analyzed in terms of a Kosterlitz-Thouless-type localization behavior.[7] In high quality graphene samples, made from Kish-graphite, Zhang *et al.*[8,9] observed an additional fine structure of the lowest Landau level in the form of a $\nu=\pm 1$ state. The existence of this state is proposed to be caused by a spontaneous symmetry breaking at the CNP and an interaction-induced splitting of the two levels resulting from this.

In this Rapid Communication we present an experimental study of the transport properties of the zero-energy Landau level in high magnetic fields and at low temperatures. Under these conditions it shows a strongly increasing longitudinal magneto-resistance at the CNP accompanied with a zero crossing of the Hall resistance. This behavior of the resistivities naturally leads by a tensor inversion to a zero minimum in the longitudinal conductivity σ_{xx} and a quantized zero-plateau in the Hall conductivity σ_{xy}. This observation, in stark contrast to the quantum Hall gaps in conventional two-dimensional systems, points to a different type of quantum Hall insulator. The temperature dependence of the σ_{xx} minimum displays an activated behavior. We explain this transition with a simple model involving the opening of a gap (30 K at 30 T) in the zeroth Landau level. We do not observe any indication for a spontaneous symmetry breaking and an interaction-induced splitting at $\nu=\pm 1$ as reported in Refs. 8 and 9 and we tentatively assign this to the relatively larger disorder in our samples made from natural graphite.

The monolayer graphene devices [see top left inset Fig. 1(a)] are deposited on Si/SiO_2 substrate using methods as already reported elsewhere.[10,11] The doped Si acts as a back-gate and allows to adjust the charge-carrier concentration in the graphene film from highly hole doped to highly electron

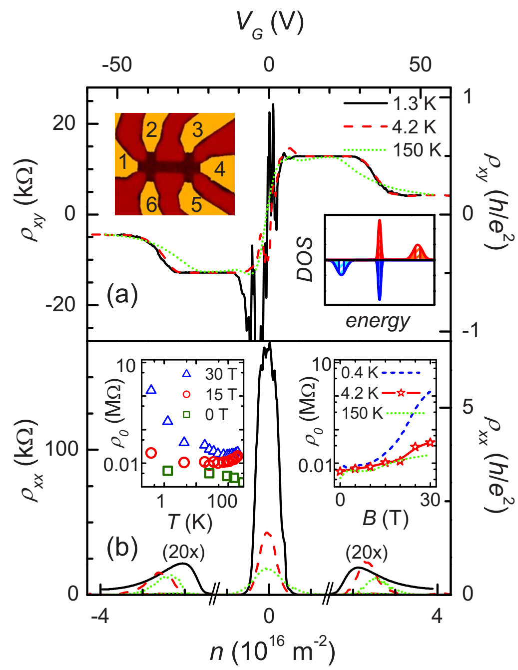

FIG. 1. (Color online) (a) ρ_{xy} as a function of back-gate voltage V_G (top x-axis) and charge carrier concentration n (bottom x axis) measured between contacts 3 and 5 as shown in the scanning electron micrograph in the top left inset at a fixed magnetic field $B=30$ T. (b) ρ_{xx} at $B=30$ T measured at the same temperatures as in (a). Note the x-axis break and the 20 times magnification for high electron/hole concentrations. The left inset in (b) shows the temperature dependence of ρ_{xx} at the charge CNP for different B. The right inset displays the magnetic field dependence of ρ_{xx} at the CNP for different T.

doped. Prior to the experiments the devices were annealed at 390 K during several hours removing most of the surface impurities[12] and increasing the low-temperature mobility from $\mu=0.47\ \mathrm{m^2\,(Vs)^{-1}}$ to $\mu=1.0\ \mathrm{m^2\,(Vs)^{-1}}$. All measurements were preformed with standard ac techniques at low enough currents (1 nA at 0.4 K) to avoid any heating effects in the high-resistance regime.

Charge carriers in graphene behave as massless chiral Dirac fermions with a constant velocity $c\approx 10^6$ m/s and thus a linear dispersion $E=\pm c\hbar|k|$.[1,2,13] The $\pm$ sign refers to electrons with positive energies and holes with negative energies, respectively. In a magnetic field, this linear spectrum splits up into non-equidistant Landau levels at $E_N=\pm c\sqrt{2e\hbar BN}$.[4] Higher Landau levels ($N\geq 1$) are fourfold degenerate and filled with either electrons ($E>0$) or holes ($E<0$). The zeroth Landau level ($N=0$) is half filled with electrons and half filled with holes of opposite chirality.

A consequence of this peculiar Landau-level structure is the half-integer quantum Hall effect.[1,2] Figs. 1(a) and 1(b) visualize this effect by means of magneto-transport experiments in a graphene field-effect transistor (FET) at $B=30$ T. At large negative back-gate voltages ($V_g<-40$ V) two hole levels are completely filled, the Hall resistivity is quantized to $\rho_{xy}=h/6e^2=4.3$ kΩ and the longitudinal resistivity ρ_{xx} develops a zero-minimum. Moving V_g toward zero depopulates the $N=1$ hole level and moves the Hall resistance to the following plateau, $\rho_{xy}=h/2e^2=12.9$ kΩ accompanied by another zero in ρ_{xx}. Sweeping V_g through zero then depopulates the hole states in the zeroth Landau level and, simultaneously, populates electron states in the same level. ρ_{xy} changes smoothly through zero from its negative quantized value on the hole side to a positive quantized value on the electron side whereas ρ_{xx} moves from a zero on the hole side through a maximum at the CNP to another zero minimum on the electron side.

In the following we will concentrate on the electronic behavior of graphene around the CNP in high magnetic fields and at low temperatures. Our experimental findings are summarized in Fig. 1(b). For high magnetic fields ($B>20$ T) ρ_{xx} at the CNP, ρ_0, first starts to decrease with decreasing temperatures (metallic regime[6]) and then strongly increases from a few tens of kilo-ohms to several mega-ohms when the temperatures is lowered down to 0.4 K [left inset in Fig. 1(b)]. The transition temperature to an insulating regime increases with increasing sample quality and increasing magnetic field. Such a field-induced transition can also be seen in the right inset in Fig. 1(b): Whereas the magnetoresistance displays a moderate field dependence for sufficiently high temperatures, it starts to increase strongly for $B>20$ T for the lowest temperature $T=0.4$ K.

At zero magnetic field the resistivity at the CNP behaves as can be expected, it increases with decreasing temperature toward $h/4e^2$, as carriers are slightly frozen out.

To gain more insight into the peculiar behavior at the CNP we have calculated the longitudinal conductivity σ_{xx} and Hall conductivity σ_{xy} from the experimentally measured ρ_{xx} and ρ_{xy} by tensor inversion. Possible artifacts due to admixtures of ρ_{xx} on ρ_{xy} and vice versa, caused by contact misalignment and inhomogeneities, were removed, by symmetrizing all measured resistances for positive and negative magnetic fields.

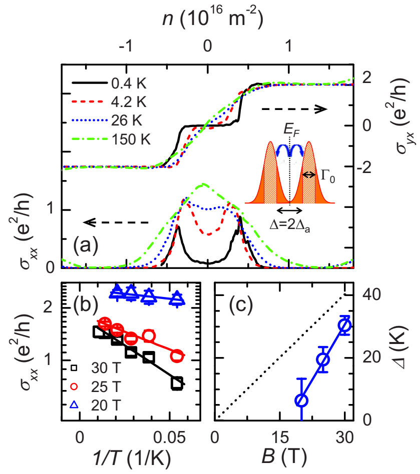

FIG. 2. (Color online) (a) Concentration dependence of σ_{xy} (top panel) and σ_{xx} (bottom panel) at $B=30$ T. The inset sketches the principle of activated transport between two broadened Landau levels. (b) Arrhenius plots of the temperature dependence of σ_{xx} at the CNP in the thermally activated transport regime. (c) Energy gaps Δ as extracted from the slopes of the Arrhenius plots. For comparison, the dotted black line represents the expected gap for a bare Zeeman spin-splitting $\Delta_Z=g\mu_B B$ with a free-electron g-factor $g=2$.

Figure 2(a) shows the two components of the conductivity tensor at $B=30$ T: At high temperatures the longitudinal conductivity σ_{xx} at the CNP shows a maximum and is accompanied by a smooth transition from the $\nu=-2$ hole plateau to the $\nu=2$ electron plateau. This behavior is typical for a quantum Hall state as expected from conventional two-dimensional systems.[14] At low temperatures, however, the longitudinal resistivity ρ_{xx} starts to increase strongly and as a direct consequence the Hall conductivity σ_{xy} develops an additional, clearly pronounced, zero-value quantum Hall plateau around the CNP and the longitudinal conductivity σ_{xx} displays a thermally activated minimum.

Both the minimum in σ_{xx} and the zero-value plateau in σ_{xy} at the CNP disappear at elevated temperatures [Fig. 2(a)] pointing toward an activated transport behavior. Indeed, when plotting σ_{xx} in an Arrhenius plot [Fig. 2(b)],

$$\sigma_{xx}\propto \exp(-\Delta_a/kT), \qquad (1)$$

we can reasonably fit the data with a single Arrhenius exponent, yielding an activation gap Δ_a. For low temperatures, we do not observe a simple activated behavior anymore since other transport mechanisms such as variable range hopping[15,16] start to play an important role. For the lowest temperatures ($T<1$ K), the resistance starts to diverge strongly, an effect which has been suggested to be caused by the abrupt transition to an ordered truly insulating state[7] and comes on top of the simple activated behavior.

We can relate the observed activated behavior to the opening of a gap in the zeroth Landau level [inset in Fig. 2(a)], $\Delta=2\Delta_a$, and, therefore, to a partial lifting of its fourfold degeneracy. The experimentally determined field dependence of this gap is shown in Fig. 2(c). The gap only becomes visible for fields above 15 T because of a finite width Γ_0 of

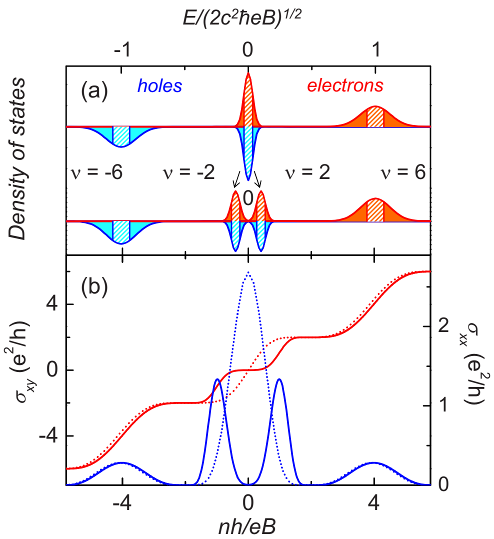

FIG. 3. (Color online) (a) Schematic illustration of the density of states for electrons (red/gray) and holes (blue/dark gray) in graphene in a magnetic field without splitting (top) and including splitting (bottom). The width of the higher Landau levels is in reality much larger than illustrated (see text). (b) σ_{xy} and σ_{xx} as a function of charge carrier concentration for unsplit (dotted lines) and split (solid line) Landau levels calculated for $B=30$ T and $T=1$ K and using $g=2$.

the split Landau level at zero energy; linear extrapolation of the experimental data to zero field yields $\Gamma_0 \approx 30$ K. The linear variation of Δ with magnetic field suggests that the splitting might be spin related. For comparison we have plotted the theoretical Zeeman splitting $\Delta_Z = g\mu_B B$ for a spin-split zeroth Landau level, with a free-electron g-factor $g=2$ (μ_B the Bohr magneton).[8] The slightly higher slope of the experimentally measured gap may be explained by a sharpening of the zeroth Landau level (i.e., a reduction of Γ_0) with increasing field,[17] or, alternatively by a slight exchange enhancement of the splitting. It should be mentioned however, that another origin of the gap than a Zeeman splitting cannot be ruled out experimentally.

The consequences of the opening of a gap can be understood intuitively using a simple model (Fig. 3): The density of states of graphene in a magnetic field consist of a series of Landau levels occupied by electrons or holes. Each Landau level is represented by a Gaussian with a degeneracy of $2eB/h$ for the individual electron and hole state of the zeroth Landau level and $4eB/h$ for the higher levels. The zeroth level is relatively sharp[17] whereas the higher levels are considerably broadened by, among others, random fluctuations of the perpendicular magnetic field caused by the rippled surface of graphene.[18–20] Electrons and holes in the center of the Landau levels are extended (shaded areas) whereas they are localized in the Landau level tails (filled areas).

With this simple model-density-of-states one can straightforwardly calculate the longitudinal conductivity σ_{xx} [Fig. 3(b)] by means of a Kubo-Greenwood formalism[21,22] and the Hall conductivity σ_{xy} [Fig. 3(b)] by summing up all the states below the Fermi energy.[23] The results are shown by the dotted lines in Fig. 3(b) and reproduce all the features of a half-integer quantum Hall effect in graphene.[1,2]

An additional splitting of all the Landau levels is schematically introduced into the model in the lower part of Fig. 3(a). The splitting is only resolved in the lowest Landau level, where the level width is already small enough (20 K). Due to the large broadening of the higher levels (400 K) it remains unresolved here.[17] Fig. 3(b) shows how this splitting nicely reproduces the zero-plateau in σ_{xy} accompanied by a minimum in σ_{xx}. As soon as the splitting exceeds the width of the extended states in the lowest Landau level, the resistivity ρ_{xx} at the CNP starts to increase as a natural consequence of the tensor inversion of the developing minimum in σ_{xx} and the quantized Hall plateau in σ_{xy}, $\rho_{xx}=\sigma_{xx}/(\sigma_{xx}^2+\sigma_{xy}^2) \xrightarrow{\sigma_{xx},\sigma_{xy}\to 0} \infty$. This is indeed in agreement with the measurements shown in Fig. 1. It also illustrates that this quantum Hall insulating state is completely different from traditional quantum Hall states where a minimum in ρ_{xx}, and a finite quantized value of ρ_{xy} are accompanied by a minimum in σ_{xx} and a finite quantized σ_{xy}. It rather resembles a Hall insulator in the quantum limit where ρ_{xx} is diverging, ρ_{xy} is finite (though not zero) and both σ_{xx} and σ_{xy} become zero.[24]

The proposed density of states in Fig. 3(b). not only explains the experimentally observed increase in ρ_{xx} at the CNP but also the behavior of ρ_{xy}. In conventional semiconductors with a gap, ρ_{xy} develops a singularity around around the CNP when either electrons or holes are fully depleted. In our graphene samples, however, ρ_{xy} is moving through zero from the $\nu=-2$ plateau with $\rho_{xy}=-h/2e^2=12.9$ kΩ to the $\nu=+2$ plateau at $h/2e^2$. This observation implies that electrons and holes are coexisting both below and above the CNP and graphene behaves as a compensated semimetal with as many hole states as electron states occupied at the CNP.

It should be mentioned in this context that Zhang *et al.* observed an additional splitting of the lowest Landau level in other samples[8,9] resulting into additional quantum Hall plateaus at $\rho_{xy}=\pm h/e^2=\pm 25.8$ kΩ and $\nu=\pm 1$ minima in ρ_{xx}. To explain these results, the degeneracy of electron and hole states has to be removed by lifting the sublattice degeneracy. In this respect, the absence of $\nu=\pm 1$ state in our and other experiments[6,7,17] points to a larger disorder, confirmed by the lower mobility, which prevents symmetry breaking. Therefore, we deal with a symmetric density of states for electrons and holes with two twofold degenerate split levels shared by electrons and holes simultaneously.

An important consequence of our experiments is that the gaps measured are more than an order of magnitude smaller than interaction induced gaps. Indeed, the characteristic energy of electron-electron correlation $I=e^2/4\pi\epsilon_0\epsilon_r l_B$ (where $l_B=\sqrt{\hbar/eB}$ is the magnetic length and $\epsilon_r \approx 2.5$ is the effective dielectric constant for graphene on silicon dioxide) amounts to about 1400 K at 30 T and exceeds the width of the zero-energy Landau level Γ_0 by nearly two orders of magnitude. According to a simple Stoner-like theory of quantum Hall ferromagnetism[25] this should result in spontaneous breaking of spin and, possibly, valley degeneracy with the formation of quantum Hall ferromagnetism. Since we do not see any evidences of the spontaneous breaking, the effective Stoner parameter is apparently reduced to the value of order of Γ_0.

Probably, as proposed in Fig. 1 of Ref. 25, where a ferromagnetic phase at $\nu=0$ is predicted theoretically, our sample mobility is just not high enough for the stable appearance of such a phase. In the high quality Kish-graphite based samples used in Refs. 8 and 9 the ferromagnetic phase appears to be already stabilized with respect to disorder. A suppression of the Stoner parameter is also what one would naturally expect for narrow-band itinerant-electron ferromagnets due to so called T-matrix renormalization.[26,27] Unfortunately, for the case of quantum-Hall ferromagnetism a consequent theoretical treatment of the effective Stoner criterion with a controllable small parameter is only possible for the limit of high Landau levels[28,29] and, admittedly, numerous other theoretical scenarios may be considered (see, e.g., Ref. 5 and references therein). Therefore, further theoretical efforts are certainly required to clarify this issue.

In conclusion, we have measured the transport properties of the zeroth Landau level in graphene at low temperatures and high magnetic fields. An increasing longitudinal resistance accompanied by a zero crossing in the Hall resistivity at the CNP leads to a flat plateau in the Hall conductivity together with a thermally activated minimum in the longitudinal conductivity. This behavior can be related to the opening of a gap in the density of states of the zeroth Landau level, which reasonably reproduces the features observed experimentally.

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