

Zero-Energy State in Graphene in a High Magnetic Field

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The fate of the charge-neutral Dirac point in graphene in a high magnetic field H has been investigated at low temperatures ($T \sim 0.3$ K). In samples with small gate-voltage offset V_0 , the resistance R_0 at the Dirac point diverges steeply with H , signaling a crossover to a state with a very large R_0 . The approach to this state is highly unusual. Despite the steep divergence in R_0 , the profile of R_0 vs T in fixed H saturates to a T -independent value below 2 K, consistent with gapless charge-carrying excitations.

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The discovery of the quantum Hall effect (QHE) in monolayer graphene crystals provides a new system for investigating relativistic Dirac-like excitations in solids [1–6]. In a magnetic field H , the system forms Landau Levels (indexed by n) that are fourfold degenerate. The Hall conductivity σ_{xy} is accurately quantized as the chemical potential μ is changed from the hole part to electron part of the Dirac spectrum. Considerable attention has focussed on the $n = 0$ Landau Level (LL), especially on the nature of the electronic state at the charge-neutral point ($\mu = 0$) in an intense magnetic field H . Several groups [7–12] have predicted a high-field state with valley polarization. Experiments are actively addressing these issues [13–15]. Jiang *et al.* [14] have inferred that the sublevel gaps at $\nu = 0$ and ± 1 arise from lifting of the spin and sublattice degeneracies, respectively, and inferred a many-body origin for the states. We have found that, in samples with small V_0 (the gate voltage needed to align μ with the Dirac point), the value R_0 of the resistance R_{xx} at the Dirac point diverges steeply with H , i.e., a large H drives the Dirac point to a high-resistance state. Despite the strong H dependence, R_0 saturates to a T -independent value below 2 K, providing evidence for charged, gapless excitations. In samples with large V_0 , this divergence in R_0 is shifted to higher fields.

Following Refs. [1,2,4], we peeled single-layer graphene crystals (3–10 μm in length) from Kish graphite on a Si-SiO₂ wafer. Au/Cr contacts were deposited using e -beam lithography [Fig. 1(a), inset]. We have found that the high-field behavior of R_0 is strongly correlated with V_0 (Table I). All samples (except K22) have μ lying in the electron band (positive V_0). Samples in which $|V_0| < 1$ V (K7 and K22) display a very large R_0 (14) (resistance measured at 14 T and 0.3 K), which arises from the strong divergence mentioned. By contrast, in samples with large $|V_0|$, $R_0(14) \leq 7$ k Ω .

Figure 1(a) shows the variation of R_{xx} in K7 plotted vs the shifted gate voltage $V'_g = V_g - V_0$ with H held at 8, 11, and 14 T (at $T = 0.3$ K). The striking feature here is that the peak corresponding to the $n = 0$ LL increases to >100 k Ω at 14 T, whereas the peaks corresponding to $n = \pm 1$ remain below ~ 7 k Ω . As in Refs. [1–5], the Hall

conductivity σ_{xy} (Panel b) displays plateaus given by [12]

$$\sigma_{xy} = \frac{\nu e^2}{h} = \frac{4e^2}{h} \left(n + \frac{1}{2} \right), \quad (1)$$

where n indexes the fourfold degenerate LL and ν indexes

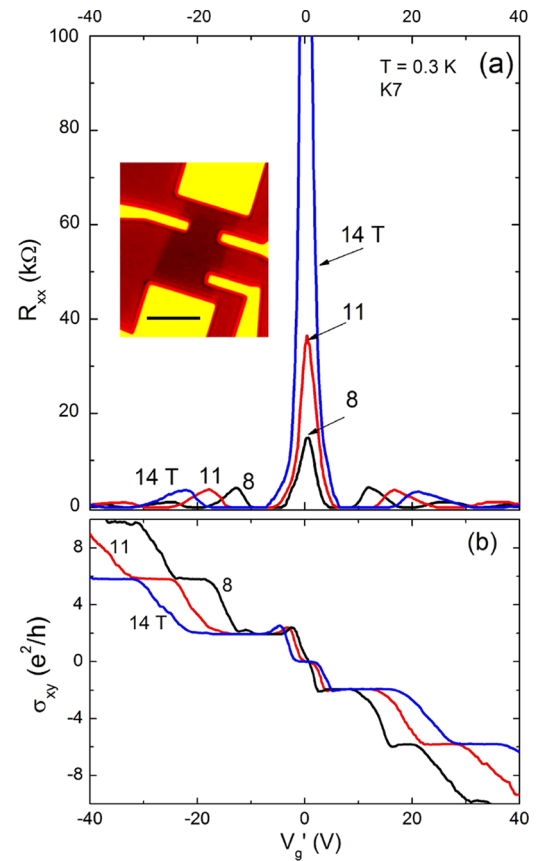


FIG. 1 (color online). The resistance R_{xx} (a) and Hall conductivity σ_{xy} (b) in Sample K7 versus (shifted) gate voltage $V'_g = V_g - V_0$ at 0.3 K with H fixed at 8, 11, and 14 T. Peaks of R_{xx} at finite V'_g correspond to the filling of the $n = 1$ and $n = 2$ LLs. At $V'_g = 0$, the peak in R_{xx} grows to 190 k Ω at 14 T. The inset shows sample K22 in false color (dark red) with Au leads deposited (yellow regions). The bar indicates 5 μm . Panel (b) shows the quantization of σ_{xy} at the values $(4e^2/h)(n + \frac{1}{2})$. At 0.3 K, $\sigma_{xy} = 0$ in a 2-V interval around $V'_g = 0$.

TABLE I. Sample parameters. V_0 is the gate voltage needed to bring μ to 0. $R_0(14)$ is R_0 measured at $H = 14$ T and $T = 0.3$ K. μ_e is the electron mobility at $H = 0$.

Sample	V_0 (V)	$R_0(14)$ (k Ω)	μ_e (1/T)
K5	3	80	0.3
K7	1	190	1.3
K8	12	15	0.6
K18	20	7.5	0.9
K22	-0.6	>280	2.5
K29	22.5	7	0.2

individual sublevels. In K7, the “zero” plateau $\sigma_{xy} \approx 0$ at $V'_g = 0$ is already visible at $H = 8$ T.

Narrowing our focus to the $n = 0$ LL, we examine R_{xx} in the $n = 0$ LL as a function of T , with H fixed at 14 T [Fig. 2(a)]. We see that, from 40 to 0.3 K, R_0 rises steeply from 4 k Ω to 190 k Ω . The curve of the conductivity σ_{xx} plotted vs V'_g reveals a two-peak structure that implies splitting of the fourfold degeneracy by a gap Δ [Fig. 2(b)]. At 100 K, the two peaks are already resolved. With decreasing T , the minimum at $V'_g = 0$ initially deepens rapidly, but saturates below ~ 2 K. The Hall conductivity σ_{xy} at 0.3 K (thin curve) displays a well-defined plateau on which $\sigma_{xy} \approx 0$. Because the next plateau is at $\sigma_{xy} = 2(e^2/h)$, we infer that each of the peaks in σ_{xx} is comprised of 2 unresolved sublevels. The opening of the gap causes σ_{xx} (at $V'_g = 0$) to fall rapidly with decreasing T , until saturation occurs below 2 K.

The behavior of R_0 described here is qualitatively different from earlier reports, for, e.g., Ref. [13]. From studies on several samples (Table I), we find that the offset gate V_0 is a crucial parameter. Figure 2(c) compares the curves of R_{xx} ($n = 0$ LL) in the samples K5, K7, K8, and K29, all measured at 14 T at $T = 0.3$ K. For each sample, we have plotted R_{xx} vs the unshifted gate V_g , so its peak automatically locates V_0 . It is clear that K7 ($V_0 = 1$ V) has the highest peak, followed by K5 ($V_0 = 3$ V), whereas K8 ($V_0 = 12$ V) and K29 (22.5 V) have peaks that are severely suppressed. This pattern is clarified below (when we discuss R_0 vs H).

Sample self-heating may also obscure the divergence. We find that, below 1 K, self-heating becomes serious when the dissipation exceeds ~ 2 pW. The measurements of R_{xx} vs V_g were repeated at 3 currents ($I = 0.6, 2$ and 15 nA) at $T = 0.3$ K. The results at $I = 0.6$ and 2 nA are virtually identical. However, the curve at 15 nA is 30% smaller near $V'_g = 0$, consistent with heating. Hence, we have kept I at 2 nA to eliminate self-heating as a problem. Heating at the contacts is negligible because of the small contact resistances (~ 1 k Ω) relative to R_0 .

Hereafter, we focus on R_0 , or equivalently, the Dirac-point conductivity $\sigma_{xx}^0 \equiv L/wR_0$ (L and w are the length and width). Curves of the conductivity versus $\log_{10}T$ are shown in Fig. 3(a) at selected fields. In low fields ($H <$

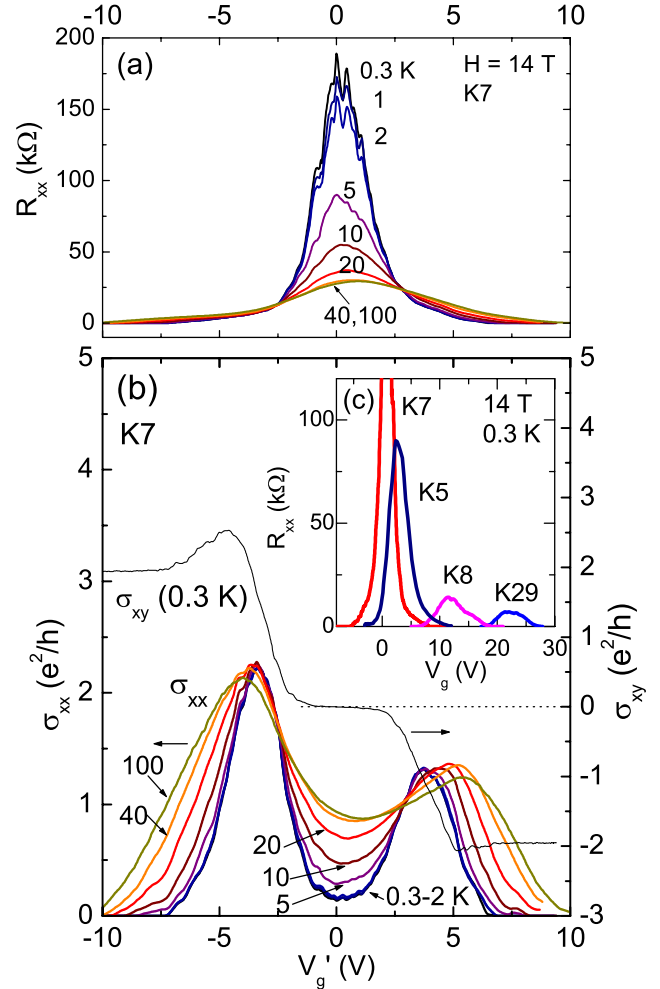


FIG. 2 (color online). The resistance R_{xx} , conductivity σ_{xx} and the Hall conductivity σ_{xy} in K7 vs the shifted gate voltage V'_g , with H fixed at 14 T (we used [5] $\sigma_{xy} = -R_{xy}/[(wR_{xx}/L)^2 + R_{xy}^2]$). As T decreases to 0.3 K, the zero-energy peak in R_{xx} (Panel a) rises steeply to 190 k Ω . Panel (b) shows that, as T decreases, double peaks in σ_{xx} are clearly resolved. Between the peaks, σ_{xx} falls rapidly but saturates below 2 K. The Hall conductivity at 0.3 K (thin curve) displays a clear plateau ($|\sigma_{xy}| < 0.02e^2/h$) in the interval $-1V < V'_g < 1V$. Panel (c) compares R_{xx} (of $n = 0$ LL) vs unshifted gate V_g in the samples K5, K7, K8, and K29 at 0.3 K. In each sample, R_{xx} peaks at V_0 . As V_0 increases, $R_0(14)$ rapidly decreases.

9 T), the T dependence of σ_{xx}^0 is quite mild. As H is increased to 14 T, the opening of the gap Δ (between the $n = 0$ sublevels) causes the conductance to decrease sharply below 40 K. However, instead of falling to 0, σ_{xx}^0 saturates below 2 K to a T -independent residual value σ_{res} , as anticipated in the discussion of Fig. 2(b). The existence of this residual σ_{res} , which is highly sensitive to H , is one of our key findings.

The field dependence of σ_{res} is best viewed as a divergent R_0 . Figure 3(b) shows the rising profile of R_0 vs H in sample K7 at selected temperatures. The divergent form at the lowest T (0.3 K) strongly suggests that the system is

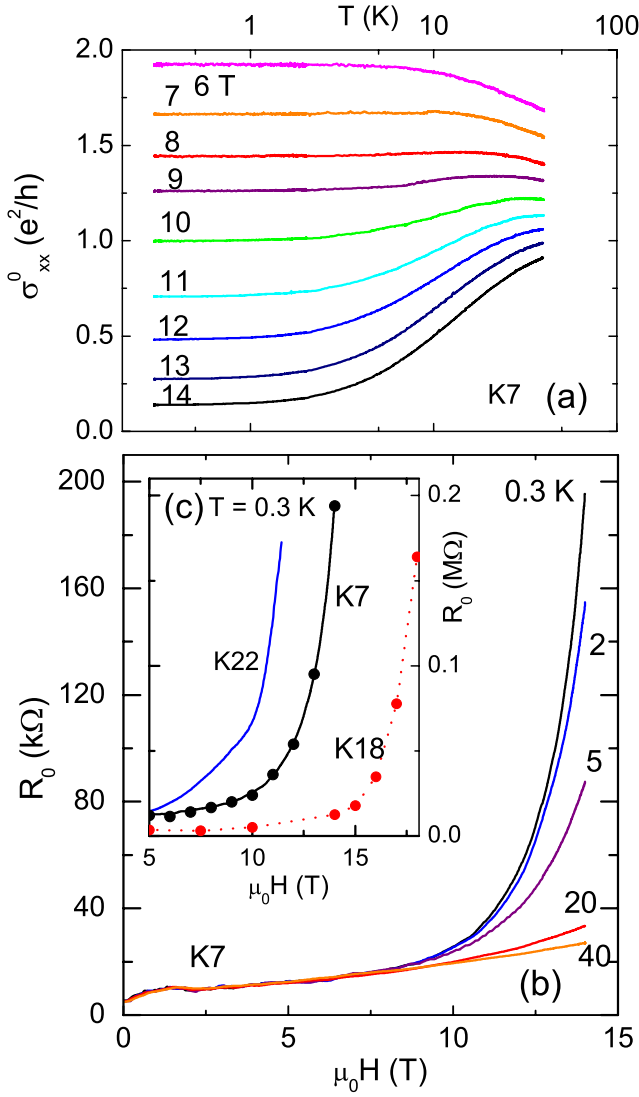


FIG. 3 (color online). The T dependence of σ_{xx}^0 in K7 ($= L/wR_0$) and the H dependence of R_0 at low temperature. Panel (a) shows curves of σ_{xx}^0 vs $\log_{10} T$ with H fixed at 6–14 T. For $H > 8$ T, the gap Δ causes σ_{xx}^0 to decrease markedly until saturation at the residual value σ_{res} occurs below 2 K. Panel (b) displays the steep increase in R_0 vs H in K7 at selected T . At 0.3 K, R_0 appears to diverge at a field near 18 T [see Fig. 4(b)]. Panel (c) compares the $R_0(H)$ profiles in Samples K7, K18, and K22. In sample K18 ($V_0 = 20$ V), the divergence in R_0 becomes apparent only above 14 T, whereas in K22 ($V_0 = -0.6$ V) R_0 starts to diverge at fields lower than in K7. In K7, we have plotted R_0 values measured by sweeping V_g at fixed H (solid symbols) with R_0 measured by sweeping H with V_g fixed at 0 (solid curve), to show consistency.

rapidly approaching a field-induced crossover (or transition) to a state with very large R_0 .

In light of the importance of V_0 , it is instructive to see how the profile of R_0 vs H varies between samples. Figure 3(c) compares the results in K7, K18 and K22 at $T = 0.3$ K. In K18, where V_0 (20 V) is quite large, the divergence in $R_0(H)$ becomes noticeable only in fields

above 14 T. Conversely, in K22 for which V_0 (-0.6 V) is slightly smaller than in K7, R_0 diverges at field scales smaller than in K7. (Above $H = 12$ T, the curve of R_0 in K22 rises to very large values, reaching 9 M Ω at 33 T. However, for $R_0 > 0.5$ M Ω , it is plagued by breaks in slope and oscillations, suggestive of severe self-heating).

From the trend, it is clear that the divergence in R_0 is shifted to ever higher fields as V_0 increases. Referring back to Fig. 2(c), we now see that the strong suppression of $R_0(14)$ in samples with large V_0 simply reflects the shift of the divergence to larger H . These results underscore the importance of choosing samples with $|V_0| < 1$ V for investigating the intrinsic properties of the Dirac-point. In K7, we also checked that R_0 measured by varying V_g at fixed H or by varying H in fixed V_g agree numerically.

The variation of $R_0(T, H)$ in K7 is conveniently represented in a contour plot in the T - H plane (Fig. 4). Below ~ 2 K, the contour lines are horizontal, which implies that R_0 is unchanged if the sample is cooled in fixed H . This provides evidence that σ_{res} involves gapless excitations. However, if T is fixed, R_0 rises steeply with H , implying proximity to the large- R_0 state (deep-red region). When a system approaches the large-resistance state, its resistivity generally diverges as $T \rightarrow 0$, as a result of either strong localization (variable-range hopping) or the opening of a mobility gap (weak localization is not relevant here because of the intense H). In both cases, decreasing T reduces the conductance because the itinerant states are severely depopulated. Hence, the pattern in Fig. 4(a) is most unusual. The gaplessness of σ_{res} suggests that, below 2 K, these excitations are protected from the effects of changing T . Paradoxically, they are not protected from an increasing H , which reduces the current carried at an exponential rate.

One way to distinguish theories is by how the fourfold degeneracy in the $n = 0$ LL is lifted. In the theory in Refs. [13,16], the exchange energy lifts the spin degeneracy (but nominally not the valley degeneracy) in the bulk. At the edge, lifting of the valley degeneracy creates a pair of counter-propagating edge states. In the sample shown in Ref. [13], the slight increase in R_0 (threefold at 33 T) was explained by increased scattering between the edge states. However, in our data, this mild change with H is actually seen only in samples with $V_0 > 30$ V. Samples with very small $|V_0|$ [K22 and K7 in Fig. 3(c)] are in a radically different category. In large H , the divergence in R_0 (which reaches 9 M Ω at 33 T in K22) is far too large to be explained by counter-propagating edge states. Instead, our results imply a high-field ordered state consistent with the quantum Hall insulator [7–10].

The scenario is suggested by the form of $R_0(H)$, which fits very well to the form $R_0 \sim \xi(h)^2$, where the correlation length ξ has the Kosterlitz-Thouless (KT) dependence

$$\xi_{\text{KT}} \sim \exp(b/\sqrt{1-h}), \quad (h = H/H_0), \quad (2)$$

with H replacing T . Plotting $\ln R_0$ vs $\sqrt{1-h}$, we find that the high-field portion becomes linear [Fig. 4(b)] when H_0

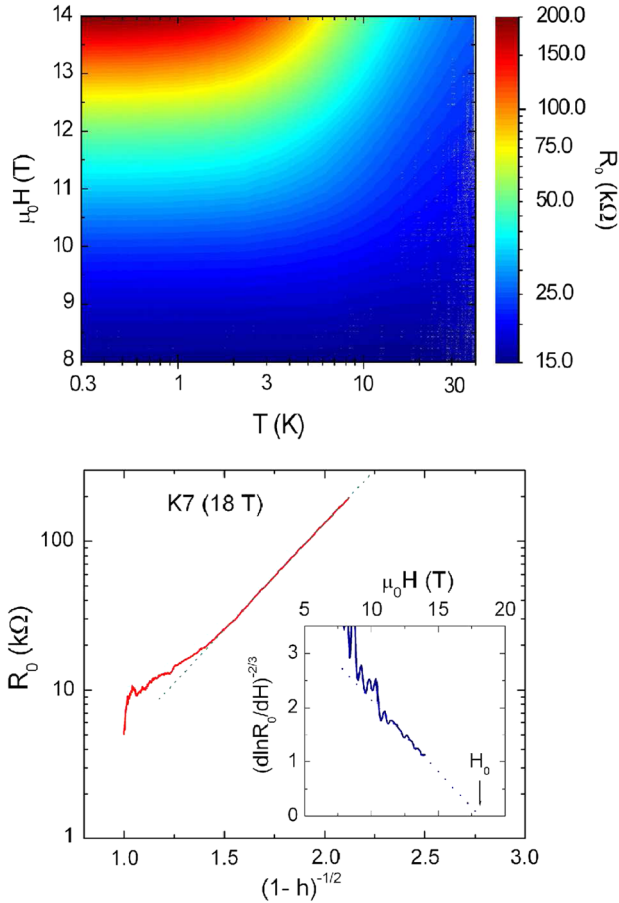


FIG. 4 (color online). (a) The contour plot of $R_0(T, H)$ (K7) in the T - H plane (vertical bar shows values of R_0). The contour lines emphasize the unusual approach to the large- R_0 state. At low T , R_0 is unchanged on a horizontal path (H held fixed), but it rises rapidly on a vertical path (increasing H at fixed T). Panel (b) displays $\log R_0$ vs $1/\sqrt{1-h}$ in K7 at $T = 0.3$ K, with $h = H/H_0$, where $H_0 = 18$ T. The linear segment at large R_0 shows that the divergence is consistent with $R_0(h) \sim \exp[2b/\sqrt{1-h}]$ with $b \sim 0.7$. In Panel (c), the plot of $(d \ln R_0 / dH)^{-2/3}$ vs H shows a high-field linear segment that extrapolates to zero at $\sim H_0$ (18 T).

is adjusted to be 17–18 T. From the slope, we find that the parameter b is ~ 0.7 , consistent with standard KT theory. For self consistency, we may also let the data determine H_0 . By Eq. (2), we have $d \ln R_0 / dH \sim (H_0 - H)^{-3/2}$. Hence a plot of $(d \ln R_0 / dH)^{-2/3}$ vs H should cross the H axis at H_0 . Indeed, this quantity, plotted in Fig. 4(c), becomes linear at large H and extrapolates to zero at 18 ± 0.5 T, in agreement with Fig. 4(b).

We adopt as a working hypothesis that this reflects the approach to a KT transition. The ordered state at large H is destroyed by the spontaneous appearance of defects which increase exponentially in density at H_0 . The steep fall of R_0 below H_0 may reflect the current carried by these defects.

In the quantum Hall insulator [7–10], the exchange energy in large H lifts the valley degeneracy in the $n = 0$ LL to produce a valley polarized state with a bulk gap (counterpropagating edge states are absent). The reduction of the $SU(4)$ symmetry to lower symmetries by the effects of Zeeman energy, disorder or lattice discretization is discussed in Refs. [8,10,12]. If the ordered state indeed has $U(1)$ symmetry, the XY order is susceptible to a KT transition [7]. It would be very interesting to relate the fit of R_0 to Eq. (2) with charged, topological excitations envisioned in the KT transition. Experiments to overcome the self-heating problem at larger R_0 are underway to provide further evidence for a transition to the large-resistance state and to clarify its nature.

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Note added in proof.—A field-induced transition to a CDW gapped state has been proposed by Khveshchenko [17].

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