Topological transition of graphene from a quantum Hall metal to a quantum Hall insulator at $\nu = 0$

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New Journal of Physics 13 (2011) 113008 (10pp)
Received 6 July 2011
Published 3 November 2011
Online at http://www.njp.org/
doi:10.1088/1367-2630/13/11/113008

Abstract. An exciting theme in condensed matter physics is the search for new states of matter. Over the last few years, a new class of topological states has been discovered—the topological insulator—which greatly expands our knowledge about quantum states. One important topic is the transition from the new topological state to other known states, such as the superconductor or normal band insulator. Graphene at filling factor $\nu = 0$ was known to be a topological insulator (called a quantum Hall metal (QH-metal)) protected by the electron–hole symmetry. A recent surprising experiment indicates that graphene can also be a normal band insulator (called a quantum Hall insulator (QH-insulator)) at $\nu = 0$ in a strong magnetic field. Here we show that a transition from a topological insulator to a band insulator can occur in graphene at $\nu = 0$. The topological transition results from the competition between the magnetic field-driven Peierls-type lattice distortion (originating from the Landau level degeneracy) and random bond fluctuations from the intrinsic sheet-buckling. The critical field that separates a QH-metal from a QH-insulator depends on the strength of bond fluctuation. The picture explains well why the field required for observing the QH-insulator is lower for a cleaner sample.

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1. Introduction

The intriguing quantum Hall effect (QHE) in graphene has attracted much attention in recent years [1]. The quantum Hall (QH) plateaux were initially found to be [2, 3] \( \sigma_{xy} = (4e^2/h)(n + 1/2) = ve^2/h, n = 0, \pm 1, \pm 2, \ldots \). The physics of charge transport is the same as that of the usual QHE with graphene’s specificity: the factor of 4 comes from spin and valley (called \( K \) and \( K' \) points) degeneracy, and 1/2, in edge state terminology, is from the interesting splitting of the \( n = 0 \) Landau level (LL) at sample edges due to the particle–hole symmetry [4, 5]. This QHE rule was soon violated in stronger fields, and anomalous plateaux of \( \nu = 0, \pm 1 \) [6] due to the lift of spin and valley degeneracy were discovered, although the detailed symmetry-breaking mechanisms are still under debate [7]. The recent surprise comes from the discovery of the insulating state at the charge-neutrality point (CNP) in a high magnetic field \( B \) by Checkelsky et al [8], because it disagrees with the early appealing theory [4, 5] that explained all experiments then existing well. This discovery was confirmed by a temperature dependence study of magneto-transport near CNP (zero energy) by Zhang et al [9]. Experiments [8, 9] reveal the following universal features. (i) Depending on the sample quality and applied field, graphene at CNP in the QH regime can be either a quantum Hall metal (QH-metal) or a quantum Hall insulator (QH-insulator) (see below). (ii) A magnetic field can drive graphene at CNP from the QH-metal state at a relative low field into the QH-insulator state at a high enough field. The critical field required for such a transition is lower for a higher-quality sample. (iii) The nature of the transition seems to be of Kosterlitz–Thouless (KT) type.

There are two topologically different \( \nu = 0 \) states. One is the QH-metal [4] that occurs when the spin split is larger than the valley split, as illustrated in figure 1(a). The \( n = 0 \) LL splits first into spin-up (blue curves) LL and spin-down (red curves) LL. Further splits of valley degeneracy near the sample edges create two counter-propagating charge- and spin-carrying edge channels, which results in a residual conductance of \( 2e^2/h \). This is a so-called topological insulator state protected by electron–hole symmetry [4]. The other possibility is the QH-insulator when the valley degeneracy lifts first with a smaller spin split, primarily due to the Zeeman effect. As shown in figure 1(b), the \( K-LL \) and \( K'-LL \) move symmetrically to the opposite direction of zero energy (due to electron–hole symmetry). As a result, the topological edge channels are absent at zero energy, and a true insulator appears. The QH-insulator was previously excluded from a possible \( n = 0 \) state, mainly due to the early experimental observation of saturated longitudinal resistance at low temperature [4]. However, the recent observation of an insulating phase near CNP suggests that the QH-insulator is also a possible \( \nu = 0 \) state [8, 9].
Figure 1. Schematic illustration of the position-dependent energy of \( n = 0 \) LL for the QH-metal (a) and QH-insulator (b). The position (in units of magnetic length \( l_c = \sqrt{\hbar/eB} \)) is measured from the sample edge \( (y_0 = 0) \) and \( \epsilon_0 \) represents the energy gap between \( n = 0 \) and \( n = 1 \) LLs. The blue and red lines represent the spin-up and spin-down states, respectively.

The mechanism of the lift of fourfold degeneracy of \( n = 0 \) LL \([7, 10–12]\) and the existence of a possible insulating state at \( \nu = 0 \) \([13, 14]\) have been discussed extensively. However, how to understand the metal-to-insulator transition at \( \nu = 0 \) is still an open question and no consensus has been reached so far. For example, Yang and Han \([13]\) suggested that graphene will be a QH-metal if the spin splits first, while graphene is a QH-insulator if the valley splits first. However, the study did not answer why the same sample can change from a QH-metal to a QH-insulator. Assuming that there is a field-induced metal-to-insulator transition, Nomura et al \([14]\) tried to identify a possible order-parameter and to argue that the transition is the experimentally observed KT type. In this paper, we uncover the mechanism by which graphene at CNP can change from a QH-metal to a QH-insulator as the magnetic field increases within a single-electron picture. It is shown that bond fluctuations due to intrinsic ripples can resist the \( K–K' \) split in low magnetic fields, resulting in the QH-metal state at CNP. In strong magnetic fields, one should expect a QH-insulator state. This understanding opens the door for experimentally manipulating the electronic structures of graphene at the CNP.

2. Picture

It shall be useful to present first our picture of why a transition from a QH-metal to a QH-insulator is expected in graphene in a strong magnetic field. Firstly, LLs of graphene in a strong magnetic field are highly degenerate. According to the Peierls instability for a solid or the Jahn–Teller effect for a molecule \([15]\), graphene can lower its energy through a lattice distortion \([12]\). The degree of distortion should be proportional to the magnetic field because the LL degeneracy is proportional to the field \([11, 12]\). The distortion destroys \( K–K' \) (or sublattices A and B for \( n = 0 \) LL) degeneracy. According to various estimations \([11, 12, 14, 16]\), the \( K–K' \) split is linear in \( B \)-field for an ordered distortion, similar to the usual Zeeman effect but larger. Although there may be other mechanisms for breaking valley degeneracy, such as electron–electron (e–e) interaction \([7]\), the Peierls instability is robust and universal for highly degenerated systems such as graphene that can also be regarded as a large molecule. Secondly, it is inevitable to get rid of ripples in graphene. The ripples create the intrinsic bond disorders,
and the intrinsic bond fluctuations tend to suppress the $K-K'$ split of $n=0$ LL, because the bond fluctuation restores the inversion symmetry of A- and B-sublattices [17, 18]. As a result, the lattice distortion and the intrinsic bond fluctuations compete with each other. At relative low fields, the fluctuations overtake the distortion effect, and there is no $K-K'$ valley splitting. The graphene at the Dirac point is a QH-metal. However, at high enough fields the Peierls-type lattice distortion dominates and $K-K'$ splitting occurs. The Dirac point becomes a QH-insulator. In the rest of this paper, we shall use the Lanczos method to find out how bulk states of a lattice distorted graphene split in the presence of different bond fluctuations.

3. Model and method

Low-energy excitations of graphene are from $\pi$-electrons that can be modeled by a tight-binding Hamiltonian on a honeycomb lattice:

$$H = \sum_{i} \varepsilon_{\sigma} |i\rangle \langle i| \sigma + \sum_{\langle ij \rangle, \sigma} (t_{ij} + \delta_{t ij}) e^{i \phi_{ij}} |i\rangle \langle j| \sigma + h.c., \quad (1)$$

where $t_{ij}$ is the hopping energy between two nearest-neighbor sites whose value for a pure graphene is $t_0$ = −2.7 eV. $\sigma = \pm$ is spin label. $\varepsilon_{i\sigma}$ is the on-site energy that includes the Zeeman contribution $\pm g \mu_B B / 2$ for spin-up (+) and spin-down (−), leading to a spin-gap $\Delta_s = g \mu_B B = 1.3 \times B[K/T]$. One can also add a random on-site energy to mimic the extrinsic disorders. $\delta_{t ij}$ is for the possible intrinsic random hopping energy described later. The magnetic field is introduced by Peierls’ substitution in the hopping parameter $\phi_{ij} = 2 \pi e / h \int_0^l \vec{A} \cdot d\vec{l}$ [19].

The Peierls lattice distortion is either in-plane like the Kekulé bond order wave [11, 14, 16] (see the inset of figure 4(a)) or out-of-plane like the charge density wave (CDW) [12] (see inset of figure 4(d)). The Kekulé bond order wave consists of short and long bonds like in the classical benzene molecule so that the corresponding hopping energy $t_{ij}$ between two nearest-neighbor sites alternates periodically. To introduce the Kekulé distortion into Hamiltonian (1), the hopping energy of the thick bond in the inset of figure 4(a) is set to be $t_{ij} = t_0 + \Delta_{Kekule}/2$. The distortion chosen in such a way results in a valley gap $\Delta_{Kekule} = 2.0 \times B[K/T]$ [11, 14, 16]. The CDW distortion breaks the inversion symmetry of sublattices so that the on-site energy of sublattices changes periodically. The simplest way of introducing the CDW distortion into Hamiltonian (1) is that the on-site energy of sublattice A is set to be $\varepsilon_A = \Delta_{CDW}/2$, whereas the on-site energy of sublattice B is set to be $\varepsilon_B = -\Delta_{CDW}/2$. The distortion chosen in such a way leads to a valley gap $\Delta_{CDW} = 4.2 \times B[K/T]$ [12]. Both types of valley gap are proportional to the strength of magnetic field $B$ and larger than $\Delta_s$.

It is known that ripples (long-ranged corrugation) are intrinsic fluctuations of graphene [20–22]. It is observed that graphene ripples have a height variation of about 0.5–1.0 nm, which occurs on a lateral scale of 8–10 nm. The graphene with random ripples can be simulated by a superposition of a series of plane waves [23]. The out-of-plane displacement is $h(\vec{r}) = C \sum_{\vec{q}_r} C_{\vec{q}_r} \sin(\vec{q}_r \cdot \vec{r} + \delta_\vec{q}_r)$, where $\vec{r} = (x, y)$ is the position of in-plane atoms and $\vec{q}_r$ and $\delta_\vec{q}_r$ are random numbers. Figure 2 is the contour plot of corrugated graphene used in

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5 The wave vectors $\vec{q}_r$ randomly distributed in the range $[2\pi/L, 2\pi/3a]$, where $L$ is the typical length of the system and $a = 1.42 \text{Å}$ is the lattice constant. The phase $\delta_\vec{q}_r$ is randomly distributed in the range $[0, 2\pi]$. $C_{\vec{q}_r} = \sqrt{2}/q^2$ for $q_r > q_{\text{ripple}}$ and $C_{\vec{q}_r} = \sqrt{2}/q_{\text{ripple}}$ otherwise, where $q_{\text{ripple}} = 2\pi/\lambda_{\text{ripple}}$; $\lambda_{\text{ripple}} \approx 8 \text{nm}$ represents the averaged distance of the adjacent peaks of spatial ripples. And the height fluctuation is controlled by the $C$ value. In our calculations, there are $> 100$ random waves to construct the spatial ripples.
Figure 2. Contour plot of a randomly generated landscape of the ripples used in our calculations. The average lateral scale of the ripple is about $\lambda_{\text{ripple}} = 8\text{ nm}$. The height fluctuation is approximately in the range of $[-h_{\text{max}}, h_{\text{max}}]$ and $h_{\text{max}} = 1.0\text{ nm}$.

our calculations. The random shapes and locations of ripples lead to a random change in bond length that in turn causes the hopping energy fluctuations [1]: $\delta t_{ij} = t\alpha \Delta a / a$, where $\Delta a = \sqrt{a^2 + (h(\mathbf{r}) - h(\mathbf{r}'))^2} - a$ and $\alpha = \partial \log t / \partial \log a = -2$ [24].

As was explained in our early discussions, whether graphene is in QH-metal or QH-insulator states depends on how the valley and spin split in the bulk. Thus, one needs to study bulk states in order to determine two topologically different states. To see how $n = 0$ LL splits into spin bands and valley bands in the lattice distorted graphene with intrinsic random hopping, we need to obtain an accurate density of states (DOS) of the model. The Lanczos recursive method [25] on a large lattice (with 1.4 million lattice sites $1200 \times 1200$) is employed. The averaged DOS is computed:

$$\rho(E) = -\frac{1}{\pi} \text{Im} \left( G(E) \right) = -\frac{1}{\pi} \text{Im} \left( \left\langle \psi \left| \frac{1}{E - H + i\eta} \right| \psi \right\rangle \right).$$

(2)

The bar denotes the ensemble average and we use the technology of the random phase state [26] to obtain a good estimate of the average DOS. In the approach, a small artificial cut-off energy ($\eta = 0.1\text{ meV}$) is introduced to simulate an infinitesimal imaginary energy [25]. This artificial broadening may also be viewed as a true broadening accounting for electron–phonon or e–e interactions in reality. This artificial parameter will lead to a small width of LLs in clean graphene. The large lattice samples guarantee that the calculated DOS is free of finite-size errors. In the calculations, the periodic boundary condition has been used.

4. Results and discussions

Let us start with the effects of ripples on the DOS of LLs in the absence of symmetry-breaking terms. Figure 3 shows the DOS of LLs with three representative ripple parameters $h_{\text{max}} = 0.35$, 0.7 and 1.0 nm. The comparison with pure graphene (dotted line), the ripples have very different
Figure 3. Averaged DOS of Landau subband for the ripple parameters $h_{\text{max}} = 0.35\,\text{nm}$ (red), $0.7\,\text{nm}$ (blue), and $1.0\,\text{nm}$ (black). For comparison, DOS of a clean graphene is also shown (dotted line). The bond randomness is measured by the height-fluctuation $h_{\text{max}}$ of ripples. The magnetic field is set to be $B = 22\,\text{T}$ and the lateral scale of ripples is $\lambda_{\text{ripple}} = 8\,\text{nm}$.

Effects on the $n \neq 0$ LLs and $n = 0$ LL. For $n \neq 0$ LLs, LLs are broadened to the Landau subbands and the broadened width of Landau subbands depends on the disorder strength. At the same time, the energy level repulsion effect results in the shift of $n \neq 0$ Landau subbands toward zero energy. The behaviors of $n \neq 0$ Landau subbands under the ripple effect are similar to those under the random on-site disorder [25]. However, the ripple effect on $n = 0$ LL is significantly different from the case of on-site disorder or short-ranged hopping disorder [27]. Zero energy should still be a good eigenenergy since the bond fluctuations do not destroy the chiral symmetry. Indeed, it is found that the $n = 0$ LL remains unchanged in the presence of the ripple effect and the $n = 0$ LL peak is anomalously sharp. This is also consistent with earlier publications that used different models and methods [17]. In the presence of symmetry-breaking terms, we show that the fourfold degeneracy of $n = 0$ LL can be resolved.

Figures 4(a)–(c) are the DOS of $n = 0$ LL for in-plane distorted graphene of the Kekulé bond order wave (shown in the inset of figure 4(a)) in the presence of ripples shown in figure 2. At a relatively low magnetic field $B = 9.4\,\text{T}$ (figure 4(a)), the distortion-induced valley gap cannot compete with the hopping fluctuation so that the bond order wave is statistically destroyed. As a result, $K - K'$ degeneracy is statistically restored, and the $n = 0$ LL splits into spin-up (blue solid) and spin-down (red dashed) bands due to the Zeeman energy, as shown in figure 4(a). This is the QH-metal state discussed earlier because the spin splitting dominates the bulk gap of $n = 0$ LL. As the magnetic field is above the critical field $B_c$ (between $9.4\,\text{T}$ and $13.2\,\text{T}$ in our case, as indicated in figures 4(a) and (b)), the valley gap due to the bond order wave appears and $n = 0$ LL splits into four bands. As shown in figure 4(b), two peaks on the left (right) sides of zero energy are for $K$ ($K'$) valleys and the corresponding edge states do no cross each other. The graphene is in the QH-insulator state at CNP. With further increase of the field, the valley splits become even bigger, as shown in figure 4(c) for $21.9\,\text{T}$. To shown that above result is very robust against different types of distortion, we perform similar calculations for the CDW lattice distortion (shown in the inset of figure 4(d)) [12], and the same results are obtained. As shown in figures 4(d)–(f), the valley degeneracy is preserved at $6.6\,\text{T}$ (d) and the
Figure 4. Density of state of \( n = 0 \) LL of lattice distorted graphene in the presence of intrinsic random hopping for various magnetic fields. Panels (a–c) are for in-plane distortion of the Kekulé bond wave with \( B = 9.4 \) T (a), 13.2 T (b) and 21.9 T (c). Panels (d–f) are for out-plane CDW with \( B = 6.6 \) T (d), 11 T (e) and 21.9 T (f). The blue solid (red dashed) curves are for the spin-up (spin-down) states.

The zero-energy state is a QH-metal. The valley degeneracy is broken at 11.0 T (e), resulting in a QH-insulator at CNP. With further increase of field to 21.9 T (f) in this case, the two \( K \) (\( K' \)) bands are clearly located on the same side of the zero-energy point.

Figure 4 shows clearly that the intrinsic bond fluctuations due to ripples can suppress lattice distortion-induced valley splitting, regardless of whether the distortion is in-plane (Kekulé bond wave) or out-of-plane (CDW), demonstrating the robustness and universality of the picture. As shown in our calculations, the valley degeneracy is lifted and a valley gap appears when the bond fluctuation is not strong enough to restore inversion symmetry of A and B sublattices statistically. It is known that the ripple will introduce a random gauge field that will cause extra scattering and limit charge-carrier mobility in the graphene sample [28]. A cleaner sample should have a weak bond disorder, and the corresponding valley degeneracy can be broken by a smaller lattice distortion. In other words, the critical field \( B_c \) that induces the transition from the QH-metal state to the QH-insulator state depends on the degree of bond fluctuation. The cleaner a sample, the smaller the critical field required to drive the graphene from the QH-metal into the QH-insulator. In our model the bond randomness is measured by the height fluctuation \( h_{\text{max}} \) of ripples. Figure 5 is the \( h_{\text{max}} \) dependence of the critical field \( B_c \) for both Kekulé-type and CDW-type lattice distortions. It is found that the \( B_c \) value for CDW type is smaller than that for Kekulé type because the valley gap in CDW distortion is larger. The inset of figure 5 is the phase diagram in the field-disorder plane. The diagram is consistent with experimental findings [8, 9]. This also explains that the failure of the discovery of the zero-energy insulating state at high fields in experiments before Checkelsky et al [8] is probably due to the sample quality.

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We would like to make a few remarks before ending this paper. (i) Fixing the applied magnetic field, the random fluctuation drives graphene from a $\nu = 0$ insulator to a $\nu = 0$ metal. This is opposite to the role that randomness usually plays in a metal-to-insulator transition. (ii) There are two types of disorder in graphene: the intrinsic bond disorder due to spontaneous ripple formation and extrinsic disorder from the impurity states in the substrate and/or ad-atoms [1], causing on-site-energy randomness in the tight-binding model. Our numerical studies show that the on-site disorders will not suppress the $K-K'$ split, but merely broaden $K$- and $K'$-subbands. Recently, we also found that charge puddle-induced long-ranged on-site disorder has a similar effect [29]. Thus, we believe that the random on-site disorder will not change our picture qualitatively. However, we also point out that the Landau subband broadening can cause inter-Landau-subband mixing [30] that may enhance the critical magnetic field $B_c$ quantitatively. (iii) Our QH-metal is in fact a topological insulator because it is a bulk insulator with conducting edge channels [4], while the QH-insulator is a conventional band insulator. This paper reveals the origin of the magnetic-field-induced topological transition rather than the nature of the transition [14]. (iv) e–e interaction is not included for the following reasons. QH experiments at the integer filling factor do not show effects from e–e interaction [31, 32]. For instance, the observed energy gap at $\nu = 1$ ($\sim 100$ K) at $B = 45$ T is much smaller than the expected value ($\sim 1100$ K) from the e–e interaction [6]. Moreover, the observed energy gap at $\nu = 0$ is proportional to the magnetic field $B$ [32] expected in the current picture instead of $\sqrt{B}$ from the e–e interaction [4]. Furthermore, the present theory seems to be able to explain the observed field-induced transition at $\nu = 0$ without including the e–e interaction. Of course, this does not mean that the e–e interaction will not change the value of the critical field. In any case, it should be interesting to investigate how the interaction modifies the results. (v) Here we propose an experiment to test our picture. It is known that the corrugation effect in suspended graphene [20] is more significant than that in substrate-supported graphene [21]. Our theory can be tested experimentally. According to our theory, the critical field is very sensitive to the number of ripples and the height of ripples. The signature of this theory is the increase of the critical field if one can increase either the number of ripples and/or the heights of the ripples.

6 We also calculate the effects of on-site disorder on the DOS of $n = 0$ LL for in-plane Kekulé bond order and out-of-plane CDW order, respectively. Our results show that on-site disorder can broaden both spin- and valley-resolved $n = 0$ Landau subbands [25], but cannot prevent valley splitting.
Furthermore, if there are more ripples in suspended graphene than that on a substrate, then a higher critical field $B_c$ in suspended graphene is expected.

5. Conclusions

We present a theory for the field-driven topological transition of graphene from the QH-metal state to the QH-insulator state at $\nu = 0$. The transition results from the competition between Peierls-type lattice distortion and random bond fluctuation. Our theory provides not only a clear explanation of the existence of both the QH-metal state at low fields and the QH-insulator state at high fields near the CNP, but also why the critical field for a cleaner sample is lower.

Acknowledgments

This work was mainly supported by Hong Kong RGC grants (numbers 604109, HKU10/CRF/08-HKUST17/CRF/08 and RPC11SC05). Partial support from the NNSF of China (no. 10974187); the NKBRP of China (numbers 2011CB921403 and 2012CB922003); and the KIP of the Chinese Academy of Sciences (no. KJCX2-YW-W22) is also acknowledged.

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