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Stacking order, interaction and weak surface magnetism in layered graphene sheets

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Recent transport experiments have demonstrated that the rhombohedral stacking trilayer graphene is an insulator with an intrinsic gap of 6meV and the Bernal stacking trilayer one is a metal. We propose a Hubbard model with a moderate U for layered graphene sheets, and show that the model well explains the experiments of the stacking dependent energy gap. The moderate on-site Coulomb repulsion drives the metallic phase of the non-interacting system to a weak surface antiferromagnetic insulator for the rhombohedral stacking layers, while the interaction opened energy gap for the Bernal stacking layers are much smaller.

In the past several years, the rapid development in preparing few layer graphene samples has promoted great theoretical¹⁻¹³ and experimental^{14–22} interests in such novel quasi-two-dimensional electron systems. The few layer graphene may be a platform for many new physics issues and is of potential application in electronics. One peculiar feature of the layered graphene system is the stacking order, which offers a new route to manipulate the electronic properties in graphene layers.

The Bernal (or ABA) stacking and the rhombohedral (or ABC) stacking are two stable stacking orders observed in experiments. As shown in Fig.1, in either ABA or ABC stacking order, the second graphene sheet is shifted by one bond length along the C-C bond direction. The third graphene sheet is shifted back and aligned with the first sheet in the ABA stacking, while is shifted further by one more bond length along the same direction in the rhombohedral stacking. So the ABA stacking order is ABABAB \cdots , and the rhombohedral stacking is ABCABC \cdots . The trilayer graphene system is the minimal structure relevant to the stacking orders.

The electronic structures of the graphene layers strongly depend on their stacking orders¹⁻⁴. In the ABA stacking N-layer system, there are N/2 electron-like and N/2 hole-like parabolic sub-bands touching at $\epsilon = 0$ for even N, and an additional sub-band with a linear dispersion for odd N. The states in all the sub-bands are bulk states extended to all the layers. In the ABC stacking layers, the low energy electronic structure is described by two sub-bands with dispersion $\epsilon \sim k^N$ near the points K and K' in the 2D Brillouin zone. These low energy states are localized on the outermost layers, and are zero modes on the surfaces protected by the topology^{23,24}. In two dimension, the dispersion of $\epsilon(k) \sim k^N$ gives a density of states $D(\epsilon) \sim \epsilon^{-1+2/N}$, which is divergent for $N \geq 3$ at $\epsilon = 0$. This indicates a strong instability toward symmetry broken states^{5,24}.

Trilayer graphene systems are of particular interest for



FIG. 1. (Color online). Schematic diagrams of trilayer graphene sheets. (a): Bernal (ABA) stacking and (b): rhombohedral (ABC) stacking. (c) and (d) are their side views. Blue and pink colors represent carbon atoms on sub-lattices A and B, respectively.

it represents the simplest case for the stacking dependent graphene. Very recently, a stacking dependent intrinsic gap in trilayer graphene has been observed in the transport measurement^{21,22}. In the charge neutral case, namely undoped trilayer samples, the experiments indicate that the ABA stacking trilayer graphene is metallic, whereas the ABC stacking trilayer graphene is insulating with an intrinsic gap about 6 meV. Since the noninteracting electronic structure of both stacking orders are gapless hence metallic, the experimental observation of the gap in ABC stacking trilayer is in sharp contrast with the non-interaction picture and points to the importance of the interaction in these systems.

In this paper, we propose that the observed stackingdependent metallic or insulting states can be explained by a Hubbard model with a moderate on-site Coulomb repulsion U. We use a self-consistent mean field theory to show that the ground state of the ABC stacking trilayer is a weak anti-ferromagnet with opposite ferrimagnetic orderings on the top and bottom layers, due to the divergent density of states in the metallic phase. The magnetic ordering opens a gap ϵ_g^{ABC} , which is in good comparison with the experimental data. For ABA stacking trilayer, the moderate on-site Coulomb interaction will also open a spin density wave (SDW) energy gap, but it

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is much smaller than that in ABC stacking case and is hard to detect in transport measurements. Our theory is extended to study stacking-dependent graphene systems with larger number of layers. We have found that it is a general property that, in ABC stacking layers, moderate on-site Coulomb interaction opens an sizable energy gap at the Fermi level (the maximum is about 20 meV depending on the number of the layer) and leads to a weak surface antiferromagnetic state, while the energy gap of the ABA stacking layers is always smaller than 0.22 meV. These results can be further tested in future experiments.

We model N layer graphene systems by using a Hubbard model $H = H_0 + H_U$, where $H_0 = H_{intra} + H_{inter}$ is a tight binding Hamiltonian to describe the kinetic term of the system and H_U describes the on-site Coulomb repulsion. The chemical potential is set to zero, and the average electron per site is one. The intralayer hopping term H_{intra} is the tight-binding Hamiltonian of independent graphene sheets. For simplicity, we only include nearest neighbor hoppings²⁵

$$H_{\text{intra}} = -t \sum_{l\langle ij \rangle \sigma} \{ a_{l\sigma}^{\dagger}(i) b_{l\sigma}(j) + h.c \}$$
(1)

where $a_{l\sigma}(i)$ and $b_{l\sigma}(j)$ are the annihilation operators of an electron on sublattices A and B, respectively. ldenotes the layer index running from 1 to N, and $\langle ij \rangle$ nearest neighbor pairs, and σ the spin. H_{inter} describes the interlayer hopping given by

$$H_{\text{inter}}^{\text{R,B}} = t_{\perp} \sum_{\langle ll' \rangle, \langle ii' \rangle \sigma} \{ a_{l\sigma}^{\dagger}(i) b_{l'\sigma}(i') + h.c \}.$$
(2)

for the rhombohedral or Bernal stacking orders. Here, $\langle ll' \rangle$ is summed over the two adjacent layers, and $\langle ii' \rangle$ is summed over two sites aligned in adjacent layers as shown in Fig.1. The Hubbard term $H_U = U \sum_{li} n_{l\uparrow}(i) n_{l\downarrow}(i)$ will be approximated by a mean field Hamiltonian,

$$H_U^{\rm MF} = U \sum_{l,i\sigma} \langle n_{l\sigma}(i) \rangle n_{l\bar{\sigma}}(i), \qquad (3)$$

where $\bar{\sigma} = -\sigma$. $\langle n_{l\sigma}(i) \rangle$ is determined self-consistently. We consider a spin density wave state and introduce two mean fields on each layer l, one for sublattice A and one for sublattice B, $\langle n_{l\uparrow}^{A,B} \rangle$. The mean fields for spin down are related to the spin-up ones, $\langle n_{l\downarrow}^{A,B} \rangle = 1 - \langle n_{l\uparrow}^{A,B} \rangle$. Note that we have examined possible charge density wave states within the model and found no evidence for that.

We first examine the trilayer graphenes (N = 3). The energy bands for the non-interacting models are shown in Fig.2 (a) for ABC stacking (solid red line) and in Fig.2 (b) for ABA stacking with parameters t = 3.16eV and $t_{\perp} = 0.39$ eV. The non-interacting dispersion in (a) is $\epsilon \sim k^3$ at small k for both conduction and valence bands, which gives rise to a divergent density of states $D(\epsilon) = \epsilon^{-1/3}$ at $\epsilon = 0$, and the wave functions for k near the K or K' points are localized on the outer surfaces. The energy bands in (b) consist of a parabolic and



FIG. 2. (Color online). (a): Low energy bands of ABC stacking trilayer. Solid red line for U = 0 and dashed black line for U = 6.2 eV. (b): Low energy bands of ABA trilayer for U = 0. (c): Mean field energy gaps as functions of U for ABC and ABA trilayer graphene. (d): Logarithmic plots of energy gaps as functions of t/U. The parameters are t = 3.16eV and $t_{\perp} = 0.39$ eV.

a linear dispersions, both of which are not localized on the outer surfaces, and the density of states is a constant at $\epsilon = 0$. The energy gaps associated with SDW orderings are plotted in Fig.2 (c) as functions of U for both the ABC and ABA stacking orders. In the presence of Hubbard U, a SDW energy gap is opened for both ABA and ABC staking trilayer. An interesting phenomenon is that, with the same U, the energy gap of ABC stacking trilayer is always much larger than that of the ABA one [See in Fig.2 (c) and (d)]. Such distinction is attributed to their different density of states near the Fermi level. The divergent density of states of the surface zero modes for the ABC stacking graphene, protected by the momentum topology, actually induces its sensitivity to the interaction.

More quantitatively, there are three distinguished regions in U for the gaps. At $U < 5.5 \,\mathrm{eV}$, both gaps for ABA and ABC stacking are tiny while the gap for ABC stacking is at least one order of magnitude larger than ABA stacking. At 5.5eV < U < 6.4eV, the gap size grows rapidly to be observable (several meV) for the ABC stacking, but remains tiny for the ABA stacking (smaller than 0.2 meV). In this region, the ABC stacking trilayer is insulating with an observable gap while the ABA stacking trilayer remains conduct, considering the temperature in transport measurement is about 1.5 K^{21} . At $U > 6.4 \,\mathrm{eV}$, the gaps for both ABC and ABA stacking orders become observable, and become insulating. Actually the gaps for the two stacking orders become similar at U > 7 eV as we can see from Fig.2 (c). Experimentally, the transport data shows ABC stacking trilayer graphene is an insulator with a gap of 6meV and the ABA stacking trilayer is metallic. In comparison with the experiments,

the mean field calculations of the Hubbard model suggest that the Hubbard U is within the interval of a moderate values 5.5 eV < U < 6.4 eV, i.e. 1.74t < U < 2.03t.

In Fig.2 (a), we show the calculated quasi-particle dispersion for the ABC stacking trilayer graphene for a choice of U = 6.2 eV (dashed black line). The corresponding gap is estimated to be $\epsilon_g \approx 5.8 \text{meV}$. Note that, with U = 6.2 eV, the corresponding energy gap of ABA stacking is about 0.18 meV. Our model and the calculations well explain the recent experiments showing the stacking-dependent energy gap in trilayer graphene. The experimentally observed energy gap may be used to estimate the value of U. Our mean field theory suggests that $U \approx 6.2 \text{eV}$. More accurate numerical simulation may improve this estimate.



FIG. 3. (Color online). Schematic illustration of the spin ordering and spin polarization per site with U = 6.2eV: (a) and (b) for ABC stacking trilayer graphene, (c) and (d) for ABA stacking case. The spin polarization as a function of U: (e) for ABC stacking and (f) for ABA stacking.

We now discuss the spin density state and the spin polarization of the ABC stacking layer. From the selfconsistent mean field theory we obtain the site spin polarization on sublattice A or B, defined as $P_s(l,i) = \langle n_{l\uparrow}^{A,B} \rangle - \langle n_{l\downarrow}^{A,B} \rangle$. The calculated spin polarizations are plotted in Fig.3 (b), and the spin structure in the trilayer graphene is schematically illustrated in Fig.3 (a). The spin ordering is antiferromagnetic, where the neighboring spins (intra- or inter- layer) are anti-parallel to each other. However, there is a net spin polarization on the top or bottom layer, so each surface shows ferrimagnetic ordering. The spin polarization is mainly distributed on the two outer surfaces. In each layer, the spin polarizations on sublattices A and B have opposite directions. The net spin polarization is zero in the mid layer, and has opposite sign in the top or bottom layer. There is a symmetry of combined inversion and time reversal: $P_{s}(l = 1, i \in A(B)) = P_{s}(l = 3, i \in B(A))$. Note that the average spin polarization of the whole system is zero. For the parameters given in Fig.3 (b), the site spin polarizations in the top layer are about 6.9×10^{-4} and -9.6×10^{-4} on sublattices A and B, respectively, and the net spin polarization is -2.5×10^{-4} per site in average, which gives a surface magnetization $0.005\mu_B/\text{nm}^2$. The weak surface magnetization on the ABC trilayer graphene is in analogy with the ferromagnetic edge states in graphene $zigzag ribbon^{26}$, in which the density of states of the flat band edge states is divergent, inducing the edge spin polarization in the presence of a weak interaction. The interaction induced gap in graphene zigzag ribbon has been confirmed in a recent STM experiment²⁷. We also show the spin polarization of the ABC stacking trilayer as a function of U in Fig.3 (e).

As a comparison, we discuss the corresponding spin density state for ABA trilayer graphene. The results are shown in Fig.3 (c) and (d). The spin polarization of the ABA stacking trilayer is rather tiny, nearly two order of magnitude smaller than that of the ABC stacking trilayer. The spin structure in ABA stacking case is also quite different, though the spin ordering is still antiferromagnetic. For each layers, there is a nonzero net spin polarization and the largest value appears in the middle layer. We see that the top and bottom layers are equivalent here, because of the mirror symmetry with respect to the middle layer. Note that the net spin polarization of the whole system is still zero. In Fig.3 (f), we give the spin polarization of the ABA stacking trilayer as a function of U.

We now discuss N > 3 graphene layers. For the ABC stacking graphene layers and in the charge neutral case, there is always an interaction U induced gap at the Fermi level with spontaneous surface spin density wave ordering. In Fig.4 (a) and (b), we show the results of N = 24layers as an example. Since the minimum band gap (ϵ_{q1}) is no longer at the K or K' points as we can see from Fig.4 (a), we introduce the second gap ϵ_{g2} for the energy gap at the K or K' points. As shown in Fig.4 (b), the spin polarization is localized near the surfaces. In Fig. 4 (c), we present the energy gaps as functions of N. As N increases, ϵ_{g1} first increases to approach its maximum about 20 meV at N = 9, then decreases to a value of 15meV at N = 24. On the other hand, ϵ_{q2} increases with the layer thickness N and reaches a saturated value about 33meV. The *N*-dependent spin polarization is shown in Fig.4 (d), which increases with the layer thickness, and approaches to a saturated value, which is at least 5 to 6 times of the surface magnetization in the trilayer case. Note that the first principle calculations involving a local spin density approximation has been applied to study 8 layers ABC stacking graphene¹³, and reported a spin density wave ground state. Their result is consistent with the results of the Hubbard model proposed here, while



FIG. 4. (Color online). ABC stacking N-layer graphene: (a) energy band and (b) spin polarization for N = 24; (c) the gaps ϵ_{g1} and ϵ_{g2} and (d) surface spin polarization as functions of N. ABA stacking N-layer graphene: (e) energy gap as function of N, (f) spin polarization for N = 9. U = 6.2eV and the hopping parameters are the same as in Fig. 2.

our results are more general and distinguishes different stacking orders.

We have also applied the mean field theory to study the N-layer graphene with ABA stacking. The results are shown in Fig.4 (e) and (f). We see that the energy gaps of the ABA stacking N-layer are much smaller than the ABC stacking cases, and the maximum value is smaller than 0.22 meV. Note that, for N-layer graphene with ABA stacking, there are two kinds of band structure depending on even or odd N. So we can observe an evenodd dependence of the energy gap in Fig.4 (e). However, the energy difference is smaller than 0.02 meV which is quite hard to detect in experiment. We also calculate the corresponding spin polarizations. The spin ordering is still antiferromagnetic. Each layer has a net spin polarization, and the spin polarizations of the neighboring layers have opposite signs. In Fig.4 (f), we show the spin polarization of N = 9 as an example. We should emphasize that, due to the small values of the energy gap and spin polarization, the most possible case is that the ABA stacking N-layer graphene is always metallic in experiment, which is in sharp contrast with the case of ABC stacking N-layer graphene.

We argue that the mean field theory should give a qualitatively or semi-quantitatively correct physics on the stacking dependent instability, or the insulating or metallic states in layered graphene, while more accurate calculations may refine the estimate of the value of U. We remark that the proposed Hubbard model with a moderate U should capture the most important physics for the stacking dependent ground states in layered graphene. We think that the remote hopping can not influence our picture about the experiments, since the on-site interaction U is much larger than the remote hopping terms in moderate U region. The picture in small U region, where the interaction U is comparable with the remote hopping terms, may be affected⁹. It is an interesting issue but unrealistic, and out of the scope of this paper. The intersite Coulomb repulsion has tendency to drive the metallic phase to a charge density wave state, which is not compatible with the on-site U studied in the present work. Since the intersite repulsion is relatively weaker than the on-site Coulomb repulsion U, we may argue that that term may not be relevant. More exotic states such as quantum spin Hall state and anomalous Hall state have been proposed in models with spin-orbit coupling or intersite interaction on honeycomb lattice^{28,29}. The possible realization of these exotic phases in layered graphene will be highly interesting. In view of the very weak spinorbit coupling in graphene³⁰, more detailed study will be needed to explore the possibility. Note that, though unrelated to the stacking order, there have been various proposals for the interaction-driven correlated ground state of bilayer graphene³¹.

In summary, we have proposed a Hubbard model with a moderate U to describe N layer graphene, and applied a mean field theory to study the ground state and the excited energy gap of the charge neutral systems. The metallic state of the ABC stacking layer is found to be unstable against any repulsion U due to the divergent density of states at zero energy. Its ground state is surface antiferromagnetic state with opposite ferrimagnetism on top or bottom surfaces, which opens a gap. The energy gap is estimated to be 5.8 meV for U = 6.2 eV for N = 3. The metallic ground state of the ABA stacking layer is also unstable against the on-site Coulomb repulsion, while the energy gap is at least one order of magnitude smaller than that for ABC stacking which is too small to detect even at moderate U. Our model and calculations well explain the recent transport experiments, showing that the ABC stacking trilayer graphene is an insulator with a gap about 6 meV, and ABA stacking trilayer graphene remains to be metallic. The spin polarization in the spin ordered state is found to be weak, but should be measurable. We also apply our model to study the layered graphene systems with large layer number (N > 3). We found that the ABC stacking graphene multilayer has an sizable interaction induced energy gap at the Fermi level while the interaction opened energy gap for the ABA stacking ones is always tiny. This prediction can be tested in future experiments.

Note added. After posting this paper on arXiv, we learnt about the works done by J. Jung and A. H. MacDonald³², by Haiwen Liu *et al.*³³, by M. M. Scherer *et al.*³⁴ and by V. Cvetkovic and O. Vafek³⁵, in which the ground states of the graphene trilayer have been discussed from different points of view.

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