Metal—Insulator Transition in Strained Graphene: A Quantum Monte Carlo Study

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Motivated by the possibility of a strain tuning effect on electronic properties of graphene, the semimetal—Mott insulator transition process on the uniaxial honeycomb lattice is numerically studied using the determinant quantum Monte Carlo method. As the simulations are based on the half-filled-repulsive Hubbard model, the system is sign-problem free. Herein, the temperature-dependent DC conductivity to characterize electronic transport properties is used. The data suggest that metal is suppressed in the presence of strain. More interestingly, within the finite-size-scaling study, a novel antiferromagnetic phase arises at around $U-U_c$. Therefore, a phase diagram generated by the competition between interactions, and strain is established, which may help to expand the application of strain effect on graphene.

1. Introduction

Graphene is a 2D sheet of carbon atom-packed hexagonal structure, as shown in Figure 1. Since its discovery,[1] graphene has stimulated a tremendous burst of research activities on fundamental and applied grounds. Graphene shows multiple astonishing performances in several areas, is the toughest 2D material ever measured,[2] and also has extremely high charge carrier mobility at a speed of $c/300$ ($c$ is the speed of light$^{[3,4]}$ that behaves like massless particles. More than this, graphene is impermeable to standard gases$^{[5]}$ and it could be a perfect thermal conductor.$^{[6]}$ Numerous other honeycomb-like systems have attracted a lot of attention, such as silicene,$^{[7]}$ germanene,$^{[8]}$ and phosphorene,$^{[9]}$ which also have excellent properties like graphene. Thus, we expect that these honeycomb-like materials will pave the way toward a new age of materials.

It has been explained in the literature that there is no gap in the band structure of graphene, which exhibits semimetallic properties.$^{[10,11]}$ However, it remains controversial as to whether the energy gap could be modified in real graphene material to realize the metal—insulator transition (MIT)$^{[12–14]}$. If it is possible to achieve the control of the opening and closing of the energy gap in real graphene, the application of the graphene material in the electronic device will be greatly promoted.

Recent theoretical and experimental work shows that hydrogenation$^{[15]}$, application of stress,$^{[16,17]}$ etc. can be used to open the bandgap and achieve MIT in graphene. In this work, we explore different approaches to achieving the insulating state, considering the interactions between electrons. Graphene is clearly a semimetal without any electron—electron interactions, but it will turn into an insulating antiferromagnet (AFM) under very strong interactions.$^{[24]}$ However, it still remains unclear as to what we should expect in real graphene materials. For instance, in the real case, it is common to find stress effect in spite of interaction effects, and it is an amazing thing that the electronic properties could be modified by mechanical forces.$^{[25–31]}$ As for graphene, defects could be introduced naturally or intentionally while growing. The existence of these defects also attracts wide interest.$^{[32,33]}$ In the experiment, the strain could be measured and detected by atomic force microscopy. Strain of 25% on graphene can be achieved$^{[2]}$ so that the system can completely come into the antiferromagnetic region.$^{[2]}$ Theoretically, there are also a lot of works based on the tight-binding model, studying the effect of uniaxial strain on the properties of graphene.$^{[20]}$ It seems that the gap suddenly arises when deformation is beyond 20% without a transition region.$^{[17,20]}$ To illustrate the strain effect in these heavy fermion materials, the electron interactions should also be considered, and it is interesting to find that the quantum critical point (QCP) strain value is suppressed by interactions.$^{[14]}$ The localization effect and mutual effect of these two factors are the key to understanding the mechanism of MIT in strained graphene. In this article, we will focus on the competition between electronic correlations and uniaxial strains, and the phase diagram is also discussed.

2. Model and Numerical Method

The Hubbard model$^{[35]}$ is believed to be a versatile paradigm to study strongly correlated electrons on a lattice. It is an effective model to describe electrons in partially filled energy bands. The parameters of the model, such as hopping and electron—electron interaction, can be considered to be generated by integrating the corresponding degrees of freedom of all energy bands except
partially filled narrow bands. Thus, the model can be used to explain the interaction-driven transition between conducting and insulating behaviors.\textsuperscript{[10]} To shed light on the physical problems involved in the strained correlation fermions, we use the determinant quantum Monte Carlo (DQMC) method to study the half-filled-repulsive Hubbard model on the honeycomb lattice. In the Dirac electronic correlation system, it is already known that there exists a QCP for MIT at around $Uc \sim 3.89t$.\textsuperscript{[37–40]} In this strength of the interaction region, our numerical DQMC method can conduct an accurate simulation. We consider the strained Hubbard Hamiltonian on the honeycomb lattice as

$$H = -\sum_{\text{site}} t_\eta a_\eta a_\eta^{\dagger} + h.c. - \mu \sum_{\text{site}} (n_{\text{air}} + n_{\text{bar}}) + U \sum_{i}(n_{a_i} n_{a_i^{\dagger}} + n_{b_i} n_{b_i^{\dagger}}).$$

(1)

Here $a_\eta^{\dagger}$ ($a_\eta$) are the spin-$\sigma$ electron creation (annihilation) operators at site $i$ on sublattice $a$. $b_\eta^{\dagger}$ ($b_\eta$) are operators acting on sublattice $b$. $U > 0$ is the on-site Coulomb repulsion. $t_\eta$ denotes the hopping integral between two nearest-neighbor sites. The chemical potential $\mu$ determines the average density of the system. $n_{\text{air}} = a_\eta a_\eta^{\dagger}$ and $n_{\text{bar}} = b_\eta b_\eta^{\dagger}$ are the number operators. Strain is introduced through the hopping matrix elements $t_\eta$ along the $x$-direction, which is shown as the red line in Figure 1. $t_{1,1} = t$ and $t_{2} = t - \Delta t$. Here, $\Delta t$ represents a measure of stress strength. We take $t = 1$ as the scale of energy in our system. In this work, we set $\mu = 0$; thus, the system is half filled by electrons. In this case, the Hamiltonian will be particle–hole symmetric even with strain on it, so that the Hamiltonian is sign-problem free to solve in our DQMC simulations.

In the finite-temperature DQMC method, it follows the strategy that takes the partition function as an integral over all possible configurations, which is conducted by random Monte Carlo sampling. To be more specific, we can get static and dynamic observables at any given temperature $T$ through this approach. As the system is sign-problem free due to the particle–hole symmetry, our calculations can still be in high numerical precision at large enough $\beta = 1/T$; thus, we can converge the data to get the ground state. In this work, 4000 sweeps were used to get to the equilibrium state, and 12 000 additional steps were made to measure the system. The periodic boundary conditions were used in the simulation, and all the results are obtained on $2 \times 12^2$ honeycomb lattice, which is large enough considering the size effect. Figure 1 shows the $L = 6$ geometry with strain on it.

We mainly discuss the MIT effect in this article. The temperature-dependent DC conductivity $\sigma_{dc}(T)$ is an intuitive physical observation to help characterize the MIT. So we focus on discussing $\sigma_{dc}(T)$ behavior in our case. According to the fluctuation–dissipation theorem, in the zero-frequency limit, $\sigma_{dc}(T)$ is related to the current–current correlation function. As we could conduct imaginary-time simulations in the DQMC method, the real-frequency quantities can be easily obtained through analytic continuation methods. In our case, we adopted an approximation\textsuperscript{[41]} to define $\sigma_{dc}(T)$, which has been widely used as a benchmark in previous work.\textsuperscript{[42–45]}

The definition of DC conductivity is

$$\sigma_{dc}(T) = \frac{\rho^2}{\pi} \Lambda_{xx}(q = 0, \tau = \beta/2)$$

(2)

In Equation (2), $\Lambda_{xx}$ is the current–current correlation function, and it can be expressed as $\Lambda_{xx}(q, \tau) = \langle j_{\eta}(q, \tau) j_{\eta}(-q, 0) \rangle$, where $j_{\eta}(q, \tau)$ is the Fourier transform of $j_{\eta}(r, \tau)$ (time-dependent current operator) along the $x$-direction. When we focus on studying the system in the temperature lower than the energy scale, at which the density of states has a significant structure, the DC conductivity definition, in Equation (2), can be very practicable.

Figure 1. The geometry of the $L = 6$ strained honeycomb lattice with 72 sites, where the red and blue colors are labeled the A and B sublattice. The stress is added along the $x$-direction, as shown by red lines, where $t_2 = t - \Delta t$. The dark lines indicate $t_{1,1} = t$. Here, $t$ is the nearest-hopping integrating term and $\Delta t$ represents the strength of strain.
considering such approximation. The applicability has been checked in our recent work of the Hubbard model on the honeycomb lattice.\textsuperscript{44} We can get the MIT critical strength value $U_{c}$ within the formula from the change of low-$T$ behavior of $\sigma_{dc}(T)$. We also investigate the magnetic properties by carrying out measurements of spin–spin correlation functions and the spin structure factor.

$$S_{AF} = \frac{1}{N_{c}} \left\langle \left( \sum_{r \in A} \hat{S}_{z}^{r} - \sum_{r \in B} \hat{S}_{z}^{r} \right)^{2} \right\rangle$$

where $N_{c}$ is the number of sites in the lattice, and $A$ and $B$ represent the sublattice of the honeycomb lattice. Here $\hat{S}_{z}^{r}$ is the $z$-component spin operator. The results are normalized by $\langle \cdots \rangle$ including all the lattice sites. The antiferromagnetic results present in this work are concluded from the constrained-path quantum Monte Carlo (CPQMC) method,\textsuperscript{46–48} which uses the constrained-path approximation in the simulation procedure without a sign problem.

3. Results and Discussion

Our simulations are mainly computed on the $L = 12$ lattice considering the size effect. $N = 2 \times L^{2}$ is the total site number. The quantity of immediate interest in this work is to study the possible MIT of the strained honeycomb lattice, which can be measured by the conductivity and its $T$ dependence behavior. So we show the $\sigma_{dc}(T)$ behavior at low temperature in Figure 2. For some curves, $d\sigma_{dc}(T)/dT < 0$ and $\sigma_{dc}(T)$ diverges as $T$ decreases to the $T \to 0$ limit, which suggests the metallic behavior of the system. In contrast, the system is insulating when $d\sigma_{dc}(T)/dT > 0$ at $T \to 0$ limit. Thus, we can easily tell the MIT point of the strained system.

In Figure 2a and 2b, we describe $\sigma_{dc}(T)$ behaviors at low coupling strengths under several strain strengths. To understand the role of strain on conductivity, we can take a careful analysis of Figure 2a. In spite of $\Delta t$ strength, the conductivity increases as temperature increases until $T \approx 0.25$. However, when $U = 1.0$ and with a strong enough strain strength, $\Delta t = 0.20$, the curve bends down when temperature drops, and $\sigma_{dc}(T)$ approaches zero as $T \to 0$, suggesting the insulating behavior of the system. In contrast, when we take a look at $\Delta t = 0.10$, the red line diverges with the feature of $d\sigma_{dc}(T)/dT < 0$ for $T \leq 0.2$, characteristic of the metallic behavior of the system. Thus, we can clearly obtain the strain-driven MIT critical point from the panels. At $U = 1.0$, the critical $\Delta t$ value can be read as $\Delta t_{c} \approx 0.15 \pm 0.01$. As for Figure 2b, $U = 2.0$. At $\Delta t = 0.12$, $\sigma_{dc}(T)$ curve is concave and $d\sigma_{dc}(T)/dT < 0$ for $T \leq 0.2$.

![Figure 2](https://www.advancedsciencenews.com/)

Figure 2. Temperature dependence of the DC conductivity $\sigma_{dc}$ measured on the $L = 12$ lattice with strain along x-direction. $N = 2 \times L^{2}$ is the total site number. The panels correspond to different couplings: a) $U = 1.0$, b) $U = 2.0$, c) $U = 3.0$, and d) $U = 4.0$. In each figure, lines are guides to the eyes. $\Delta t$ represents the strain strength. Metallic and insulating behaviors are indicated by solid and dashed lines, respectively. The MIT behavior can be easily observed from the low-$T$ behavior of $\sigma_{dc}$, whereas in (d), the insulating behavior is preserved by strain effect.
When $T$ drops to around 0.1, $\sigma_{dc}(T)$ increases rapidly; in other words, the system is metallic. At $\Delta t = 0.14$, in contrast, the conductivity decreases as the temperature decreases, indicating insulating behaviors. Thus, there exist a MIT critical strain strength at around $\Delta t = 0.14 \pm 0.01$ for $U = 2.0$ on the graphene lattice. Then, it moves to the large interaction case, as shown in Figure 2c,d. When $U = 3.0$, the system is still metallic without considering strain effect. So, under small strain strength, it is still possible that graphene can stay in the metallic region. At low $T$ for $T < 0.1$, the sign of $\frac{d\sigma_{dc}(T)}{dT}$ is negative when $\Delta t \leq 0.12$, but it switches to positive when $\Delta t$ is larger than 0.13. Therefore, the MIT critical $\Delta t$ strengths can be read as $\Delta t_c \approx 0.13 \pm 0.01$ for $U = 3.0$. As for $U = 4.0$, which is larger than the critical $U_c = 3.89$ for MIT in the graphene lattice without strain, the system is an insulator. From Figure 2d, we can see that all the $\sigma_{dc}(T)$ curves are bent down when temperature decreases for $T \to 0$. The value of $\sigma_{dc}(T)$ is suppressed when strength of strain increases, that is to say that the insulating behavior is preserved by the strain effect in this case. To describe the metal–insulator phase boundary more precisely, we closely studied the conductivity behavior at around $U_c \approx 3.8$. Thus, according to Figure A1 in Appendix, we make the metal–insulator phase boundary more smooth, as shown in the phase diagram in Figure 3.

In Figure 4, we also investigate the effect of the orientation of the strain. Here, $\alpha$ in each panel is the angle with $x$-axis. Figure 4a is in the metallic regime for $U = 2.0$ and $\Delta t = 0.1$. As $\Delta t$ increases to 0.3, the system turns into the insulating

**Figure 3.** Phase diagram of the strained Hubbard model on the honeycomb lattice at half filling. $\Delta t$ labels the strain strength and $U$ represents the local Coulomb repulsion. The metallic phase boundary is determined by the temperature dependence of the conductivity $\sigma_{dc}$. The metallic region is filled with green color and the (gray) hexagonal mark point is obtained from our DQMC simulation results. The blue region on the left indicates the existence of long-range antiferromagnetic order.

**Figure 4.** Temperature dependence of the DC conductivity $\sigma_{dc}$ measured on the $L = 12$ lattice with strain along different directions. The panels correspond to different couplings and strain strengths: a) $U = 2.0, \Delta t = 0.1$ is in the metallic region and b) $U = 4.0, \Delta t = 0.1$, c) $U = 2.0, \Delta t = 0.3$, and d) $U = 4.0, \Delta t = 0.3$ are in the insulating region. In each figure, lines are guides to the eyes. $\Delta t$ represents for the strain strength and $\alpha$ is the inclined angle between strain and $x$-axis.
regime, as Figure 4c shows. Comparing Figure 4b and Figure 4d, the result at $U = 4.0$ is shown, which stays in the insulating state no matter how strong the strain applied. According to the lines in Figure 4, it seems that the behavior of $\sigma_{dc}$ is pretty similar when $\alpha$ changes. We only consider the nearest hopping in our Hamiltonian, which is a single-shell model. The shell behavior to the angles is very similar to the first shell of the model, including up to the third-nearest-neighbor interactions. The angular dependence is not very important considering atoms in the second and third shells; thus, we get the similar behaviors of $\sigma_{dc}(T)$ here with different orientations of strain. Therefore, we can focus our study on the strength of the strain and ignore its orientation when exploring the strain effect on MIT.

Furthermore, we also studied the effect of strain on long-range magnetic order. Data shown in Figure 5 and Figure B2 are AF spin-structure factor on lattices up to $L = 15$. For small $U = 2, 3$, there is no long-range order when $U < U_c \approx 3.89$. Even when $\Delta t = 0.30t$, there is no AF order, according to Figure B2a,b. As for small lattices, the correlation strength of antiferromagnetism decreases, whereas strain increases, which indicates that in the weak interaction case, $S_{AF}$ is suppressed by strain. In contrast, in Figure B2c,d, AF correlation strength is enhanced by strain at large $U$. The AF order is protected by strain at strong interaction ($U > U_c$). As Figure 5 shows, it is interesting to note that the AF order could exist even when $U < U_c$. Thus, there is a transition point that exists for long-range order by strain effect. We obtained: a) $U = 3.2$, $\Delta t = 0.30$, b) $U = 3.5$, $\Delta t = 0.25$, c) $U = 3.6$, $\Delta t = 0.25$, and d) $U = 3.8$, $\Delta t = 0.20$ from the finite-size-scaling results. Based on the analysis earlier, the paramagnetic–antiferromagnetic transition boundary could be summarized for nonzero $\Delta t$, as shown in Figure 3.

### 4. Summary

We have carefully studied the electronic and magnetic properties of the strained Hubbard model on the honeycomb lattice. Using the DQMC numerical method, we characterize the conductivity with the $\sigma_{dc}(T)$ behavior at low temperature, thus exploring the mechanism of strain-driven MIT in the honeycomb lattice. According to our simulations, it is clear to see that the conductivity $\sigma_{dc}$ is suppressed by strain effect. When strain is applied, the value of hopping parameter along the strain direction decreases from $t$ to $t - \Delta t$, and thus the critical electron–electron interaction strength $U$ for MIT reduces. The critical strain strength decreases as the local Coulomb repulsion $U$ is enhanced. In other words, under the interaction of strain and Coulomb correlation, the electrons are localized more effectively, which expands the insulating phase and gives us a strain-interaction-driven MIT phase diagram.
As for the magnetic properties, we found that strain could also enhance the AF order around $U_c$ in the clean limit. While strain is affected, we observed the appearance of the antiferromagnetic order phase. It seems that the strain can protect the long antiferromagnetic order when $U$ is strong. In conclusion, we show that strain-interaction-driven graphene could be a promising route to achieve MIT in graphene, and the phase diagram reported in this work could be used as a guidance in the modulation of conductivity in real graphene materials.

5. Appendices

A. Computing the DC Conductivity

In this work, the low-temperature behavior of DC conductivity $\sigma_{dc}$ is used to distinguish metallic or insulating phases. We found that the transition point changes dramatically at around $U \approx 3.80$. So, in Figure A1 in Appendix, we carefully checked the phase boundary by plotting the $\sigma_{dc}(U)$ curves, which indicate a critical transition strength $U_c$ in each panel. While in panel (a),

Figure A1. $\sigma_{dc}$ behavior as the interaction changes at different temperatures. The panels correspond to different strain strengths from $\Delta t = 0.02$ to $\Delta t = 0.12$. The crosspoint in each panel indicates the transition point: a) $\Delta t = 0.02, U \approx 3.80$, b) $\Delta t = 0.04, U \approx 3.80$, c) $\Delta t = 0.06, U \approx 3.75$, d) $\Delta t = 0.08, U \approx 3.70$, e) $\Delta t = 0.10, U \approx 3.65$, and f) $\Delta t = 0.12, U \approx 3.40$. 

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when $U < 3.80$, $\sigma_{dc}$ gradually increases as $\beta$ gets larger, which indicates the metallic behavior. On the other side, when $U > 3.80$, $\sigma_{dc}$ decreases as $\beta$ increases, which acts as an insulator. Thus, the critical transition point for $\Delta t = 0.02$ is b) $\Delta t = 0.04$, $U \approx 3.80$, c) $\Delta t = 0.06$, $U \approx 3.75$, d) $\Delta t = 0.08$, $U \approx 3.70$, e) $\Delta t = 0.10$, $U \approx 3.65$, and f) $\Delta t = 0.12$, $U \approx 3.40$.

B. Existence of the AF Order Phase

To establish the phase diagram, the finite size effect on the AF spin structure factor $S_{AF}$ has been carefully examined in the article. We extrapolated the data to the thermodynamic limit to get the order parameters. As shown in Figure B2, we discussed the strain effect on the honeycomb lattice with various interaction strengths. All the results are summarized in the phase diagram in Figure 3.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

Research data are not shared.

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