A Phase Field Crystal Method for Multilayer Graphene

Kai Liu

1 College of Education for the Future, Beijing Normal University, Zhuhai, Guangdong Province, China
Email: liuk@bnu.edu.cn

Abstract. Bilayer graphene has been a subject of intense study in recent years. We extend a structural phase field crystal method to include an external potential from adjacent layer(s), which is generated by the corresponding phase field and changes over time. Moreover, multiple layers can be added into the structure. Using the thickness of the boundaries between different stacking variants of the bilayer structure as the key parameter, we quantify the strength of the adjacent layer potential by comparing it with atomistic simulation results. Then, we test the multiple graphene structures, including bilayers, triple layers, and up to 6 layers. We find that in addition to the initial conditions, the manner in which new layers are added into the structure affects the layout of the atomic configuration. Finally, we simulate the case of twisted stacking bilayer graphene under the "magic angle 1.1°", and our results help identify the key physical ingredients to predict what relaxations may occur.

Keywords: graphene, phase field crystal method, stacking fault energy

1. Introduction

Graphene, which is a single layer of carbon atoms tightly bound in a hexagonal honeycomb lattice, is one of the most exciting new two-dimensional materials discovered. Bilayer graphene has attracted a great deal of attention because it can exist with various stacking arrangements and intriguing electronic properties [1, 2, 3, 4]. In 2018, Yuan Cao et al. found that at twist angles of approximately 1.1°, the electronic band structure of twisted bilayer graphene exhibited unconventional superconductivity [5], which draws a great amount of attention to bilayer graphene.

Computational modeling can serve as a route to theoretically understand the difficult-to-measure properties of graphene. On the continuum scale, the phase field crystal (PFC) modeling approach describes the dynamics of phase transformation through an atomically varying order parameter field, which is loosely connected to the atomic density field. The original PFC model was predominantly used to study 2D triangular and three-dimensional (3D) crystal symmetries [6, 7]. It is a promising and widely used approach to model many microstructure phenomena. Recently, PFC has been used to study how anisotropic diffusion of carbon on a surface can form the dendritic graphene structure [8]. By including a rotationally invariant three-point correlation function for the excess free energy, a structural PFC model (XPFC) was set up to address the atomically varying defect and microstructures of graphene and its nucleation and diffusional growth kinetics from a disordered state on a surface [9, 10].

In this paper, we build a new XPFC model for multilayer graphene by extending the XPFC method in [9, 11]. To model the effect of one graphene layer on another in an adjacent structure, we introduce a local...
interaction between the order parameter density and an external potential. The external layer potential that we use in this paper is similar to the first-principles calculations of the generalized stacking fault energy (GSFE) in bilayer graphene from references [11, 12]; however, we use a variant of the phase field density instead. This potential is based on the phase field of the corresponding layer and changes over time, whereas in our previous work, the bottom layer was fixed (as if on a deposition substrate) [11]. Moreover, a multilayer structure can be easily constructed.

In numerical simulations, we first use a case of a long narrow ribbon domain, which consists of 4 parts: a continuous AB and BA region, each of which is nearly 50% of the entire domain, and two narrow transitions between them, to calibrate the contacted-layer potential [11]. Here, the so-called AB and BA stacking has the sublattice atoms of one of the first layers (A or B) directly on top of its opposite sublattice atom (B or A) in the second layer, which is collectively called Bernal stacking [13]. By comparison with atomistic simulations and previous work [11], we quantify the strength of the external layer potential.

Then, we simulate the multilayer structures. We test bilayers, trilayers, and multilayers. We test different initial conditions: well structured, i.e., without 5 or 7 rings, and the hexagons are neatly arranged along the same direction or randomly generated using a constant value with small perturbation of Gaussian noise. We also test various methods of construction: one layer after another, multiple layers after the bottom layer, and all layers simultaneously. Once a base layer is formed, the layers on the top will be affected, and AB (or BA) stacking is more likely formed. Moreover, the defect grain boundaries emerge at similar locations. Finally, we test the case of a twisted bilayer system with a magic angle of 1.1° following the supercell method in [14], and the relaxation process is consistent with our expectation. The simulations are numerically expensive and require a long time (3-7 days to reach steady solidification). To efficiently solve the system in a large domain, we use CUDA C/C++ programming on an Nvidia Quadro GV100, which runs approximately 2 orders of magnitude faster than the normal CPU.

The paper is organized as follows. In Section 2, we introduce the math model and numerical method. In Section 3, we test various cases of multilayer structures. Finally, in Section 4, we end with a discussion of this work and possible future applications.

2. Modeling and Method

We add an adjacent layer potential into the XPFC model [9, 12]. Let \( \rho_i \), \( i = 1, 2 \ldots, n \) describe the spatial phase density of carbon atoms for \( n \) layers of graphene, respectively. Then a dimensionless density field is defined as \( \psi_i = (\rho_i - \bar{\rho}_i)/\bar{\rho}_i \), where \( \bar{\rho}_i \) is the reference density of a disordered phase around which a functional expansion of the free energy is performed. The free energy of a crystallizing system is

\[
F_{\text{total}, i} = F_{\text{id}}(\psi_i) + F_{\text{ex}, 2}(\psi_i) + F_{\text{ex}, 3}(\psi_i) + \sum_{j \in \mathcal{K}_i} F_{\text{adj}}(\psi_j, \psi_i)
\]

which is given by

\[
F_{\text{id}} = \int dx \left( \frac{\psi_i^2}{2} - \eta \frac{\psi_i^3}{6} + \chi \frac{\psi_i^4}{12} \right)
\]

where \( \eta \) and \( \chi \) are dimensionless parameters and we simply set \( \eta = \chi = 1 \). The two-point correlation is based on hard-sphere-like interactions and governed by [9]

\[
F_{\text{ex}, 2} = -\frac{1}{2} \int \psi_i(x) \int C_2(x - x') \psi_i(x') dx' dx.
\]

Here \( C_2 \) is the two-point correlation function defined as [9]

\[
C_2(x) = -R \pi r_0^2 \text{circ} \left( \frac{r}{r_0} \right),
\]
where $r_0$ sets the cutoff for the repulsive term, $R$ is the magnitude of the repulsion, and

$$\text{circ}(r) = \begin{cases} 1, & r \leq 1 \\ 0, & r > 1 \end{cases}$$

(5)

The three-point density correlation is rotationally invariant and sufficiently robust to capture all crystal structures described through a single bond angle [9]. It is governed by

$$F_{\text{ex,3}} = -\frac{1}{3} \int \psi_i(\mathbf{x}) \int C_3(\mathbf{x} - \mathbf{x}', \mathbf{x} - \mathbf{x}'') \psi_i(\mathbf{x}') \psi_i(\mathbf{x}'') d\mathbf{x}' d\mathbf{x}' d\mathbf{x}.$$  

(6)

Here the three-point correlation function $C_3$ is defined by

$$C_3(\mathbf{x} - \mathbf{x}', \mathbf{x} - \mathbf{x}'') = \sum_i C_s^{(i)}(\mathbf{x} - \mathbf{x}') C_s^{(i)}(\mathbf{x} - \mathbf{x}''),$$

(7)

where $C_s^{(i)}$ in polar coordinates reads as [9]

$$C_s^{(1)}(r, \theta) = C_s(r) \cos(m\theta),$$  

(8)

$$C_s^{(2)}(r, \theta) = C_s(r) \sin(m\theta),$$  

(9)

$$C_s(r) = \frac{X}{2\pi a_0} \delta(r - a_0).$$  

(10)

Here $X$ is a parameter that defines the strength of the interaction, $a_0$ corresponds to the lattice spacing given by $r_0/a_0 = 1.22604$, and $m = 3$ defines the bond order of the crystal phase. For the graphene system, $R = 6$ and $X^{-1} = 0.4$ [9].

The adjacent layer potential is

$$F_{j,\ell} = \frac{1}{\lambda} \int d\mathbf{x} [\hat{\psi}_j(\mathbf{x}) \psi_i(\mathbf{x})],$$

(11)

where $j \in \mathcal{K}_j$, $\hat{\psi}_j(\mathbf{x})$ is the corresponding potential field, and $\lambda$ parameterizes the strength of the energy.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure1.png}
\caption{(a) $F_{GSFE}$ from [12], (b) $-F_{GSFE}$, (c) nondimensionalized $\nabla^2 F_{GSFE}$, (d) $\psi_i$, (e) nondimensionalized $\nabla^2 \psi_i$, (f) $-\psi_i$.}
\end{figure}
Then, we define $\tilde{\psi}_j(\mathbf{x})$ as

$$\nabla^2 \tilde{\psi}_j(\mathbf{x}) = D_0 - D_2 \psi_j(\mathbf{x}),$$  \hspace{1cm} (12)

where

$$D_0 = \text{mean}_{\mathbf{x}}(\nabla^2 \tilde{\psi}_j(\mathbf{x}))$$  \hspace{1cm} (13)

as shown in figure 1(f). $\nabla^2 \psi_i$ is not good because the maximums and minimums of the $\nabla^2 \psi_i$ field are not at the locations of the geometric center of the carbon rings or the center of carbon atoms, as shown in figure 1(e).

Here we have compared different definitions of $\nabla^2 \tilde{\psi}_j$, e.g., one that mimics $-F_{GSFE}$ in [12], as shown in figure 1(b), which is better than $\nabla^2 F_{GSFE}$ (used in [11]), as shown in figure 1(c). Meanwhile, the difference between $-F_{GSFE}$ and $-\psi$ is small, and Eq. (12) is more straightforward and stable in the application (see more details in the supplementary materials).

Finally, the evolution of the conserved density $\psi_i$ is governed by

$$\frac{\partial \psi_i}{\partial t} = M_{\psi_i} \nabla^2 \left( \frac{\delta F_{\text{total},i}}{\delta \psi_i} \right),$$  \hspace{1cm} (14)

where $M_{\psi_i}$ is an effective mobility that scales of the diffusional dynamics of $\psi_i$ and we set $M_{\psi_i} = 1$ for convenience, if not specified.

We use a discrete Fourier transform (DFT) method to solve Eq. (14) [9, 11]. A periodic boundary condition is used for each layer, and a semi-implicit method is used to evolve Eq. (14) for numerical stability and computational efficiency. The simulations are numerically expensive in that it usually takes 3-7 days to reach steady solidification. To efficiently solve the system in a large domain, we use a CUDA C/C++, which runs approximately 2 orders of magnitude faster than the normal MATLAB CPU version.

3. Results

3.1. Quantify $\lambda$

Following [11], we use a long narrow ribbon of bilayer graphene to calibrate the parameter $\lambda$.

![Figure 2](image)

**Figure 2.** (a) Bottom layer atoms are divided into 2 groups, one is denoted by lighter green and the other is donated by darker green. (b) Position of atoms for the AB stacking order. (c) Position of atoms for the BA stacking order.

We first generate the AB and BA stacking phase fields, i.e. $\psi_1(\mathbf{x}), \psi_{2,AB}(\mathbf{x})$ and $\psi_{2,BA}(\mathbf{x})$. Using the wellstructured initial condition $\psi_1(\mathbf{x}, t_0)$, where $t_0 = 0$, i.e., ensuring the periodicity for each side of the rectangular domain (for example $L_1: L_2 = 2 \times \sqrt{3}$ and all hexagons are along the same direction), we generate the bottom phase field $\psi_1(\mathbf{x})$ which is consistent with [9]. Next, we use $\psi_1(\mathbf{x}, T)$ to form the adjacent layer potential. $\psi_2(\mathbf{x}, t_0)$ is a constant value (0.3) with a small perturbation of Gaussian noise. Both AB and BA stackings are found, as shown in figure 2. The long bilayer ribbon is constructed by $\psi_2(\mathbf{x})$ and
The long bilayer ribbon is set up as follows. The bottom layer $L_1$ is formed by multiple small patches of $\psi_2(x)$ ($L_1/L_2 = 2/\sqrt{3}$), e.g., 32 patches, connected side to side. $L_2$ is a static setup where there are 4 parallel stripes X-Y-Z-W. X that represent the stacking order AB, e.g., 15.5 small patches of $\psi_{2,AB}(x)$, Z represents the stacking order BA, e.g., 15.5 patches of $\psi_{2,BA}(x)$, and Y, W are disordered, i.e., constant value (0.3) with small perturbation. The X and Z regions grow when the dynamics start, and two interfaces will be created between them.

![Figure 3. (a) AB-to-BA transition in the left region; (b) BA-to-AB transition in the right region.](image)

We find four transition types, depending on the angle between the transition region and the shifting direction: $0^\circ, 30^\circ, 60^\circ,$ and $90^\circ$. For example, the transition region is vertical in figure 3 (a) and (b); in figure 3 (a), the angle is $90^\circ$ (the atoms shift horizontally), while in figure 3 (b), the angle is $30^\circ$ (or $-30^\circ$). By comparing with the atomistic simulation results, we quantify the strength of the bottom layer potential by the width of the transition region for each type (angle).

Then, we compute the thickness of the transition region. A nondimensionalized parameter $d_L$, which is the nondimensionalized $x-y$ plane distance between the center of one group of atoms on the bottom layer and the nearest atom centers on the top layer at $x_L$, is defined to measure the distortion between substrate potential $\psi_2(x)$ and graphene field $\psi_2(x)$. For example, the distance in the $x-y$ plane between the center of the lighter green circles and the center of pink balls in figure 3. Thus, $d_L = 1$ for the AB pattern and $d_L = 0$ for the BA pattern. Following [11], the data are fitted by the function

$$d_L = \frac{2}{\pi} \arctan \left( \exp \left( \frac{\pi L}{W} \right) \right),$$

where $W$ is a fitting parameter that represents the thickness of the transition region, and $L$ parameterizes the long side of the ribbon. Eq. (15) gives an excellent fit to the displacements for both atomistic and XPFC simulations [11].

![Figure 4. Relation between the dislocation direction and $W$. Here, the angle between the direction of the transition region and the direction along which the atoms shift is used as the dislocation character.](image)
Compared with previous work [11], we have made two improvements. First, we use a quadratic interpolation to calculate the exact position of the atoms instead of using a specific grid point \((i,j)\). The interpolation is performed separately along the \(x\) and \(y\) directions as follows: suppose \(\psi_{m,n}\) are the \(k\)th local maximums, and the maximum value among \(\{\psi_{i,j} | i \in \{m - k, m - k + 1 \ldots, m \ldots, m + k - 1, m + k\} \} \) (periodic conditions are implemented for points at the domain boundary). Here, \(k\) is related to the size of the atoms. Using the values of \(\psi_{m,n}, \psi_{m \pm 1,n}, \) and \(\psi_{m,n \pm 1}\), we perform a quadratic fitting. The position of the atom is

\[
X_k = m + \frac{(\psi_{m,n+1} - \psi_{m,n-1})}{2(\psi_{m,n+1} + \psi_{m,n-1} - 2\psi_{m,n})}, \tag{16}
\]

\[
Y_k = n + \frac{(\psi_{m+1,n} - \psi_{m-1,n})}{2(\psi_{m+1,n} + \psi_{m-1,n} - 2\psi_{m,n})}. \tag{17}
\]

By this procedure, small change in atoms’ position can be accurately computed even if there is only a slight change in phase field. Moreover, a more accurate and efficient method is adopted to determine \(W(t \to \infty)\).

As discussed in [11], it takes a long time to reach a steady state for the long ribbon. Meanwhile, \(W(t)\) has an exponential tail behavior once \(W(t)\) is close to \(W(\infty)\). We use

\[
W = W_0 + b \exp(-ct), \tag{18}
\]

to fit \(W(t)\). Once \(|W_0 - W(T)|/W_0 < 0.001, W_0\) is accepted.

Using a bisection method, we find that \(\lambda = 15000\) is a proper value for the adjacent layer potential, considering all three cases of \(W_0\) and \(W_{\pm 90}\), as shown in Figure 4. The values of 30 degrees and 60 degrees are not quite accurate, since the direction of the transition region may not be exactly perpendicular to the long side of the ribbon.

### 3.2. Multilayer Graphene Structure

#### 3.2.1. Bilayer Structure

Next, we simulate the multilayer graphene structures. We first test the bilayers. The first case is that the bottom layer \((L_1)\) is perfectly structured before the top layer \((L_2)\) is added into the system, as shown in Figure 5 (a). When the solidification of \(L_2\) starts, their interaction is turned on. The \(L_2\) layer ends up with a perfect AB/BA stacking layout with \(L_1\), as shown in figure 5 (b), which shows that the bottom layer can determine the layout of the upper layer, especially when it is well structured.

**Figure 5.** (a) Layout of the atomic configuration for the first (bottom) layer, (b) two layers together (top on the bottom), where the blue dots are the atoms on the first layer, and the black dots are the atoms on the top layer.
Figure 6. (a) Layout of the atomic configuration for $L_1$, (b) layout of $L_2$, (c) phase field of $L_1$, and (d) $L_1$ & $L_2$ stacking, where the blue dots are the atoms on $L_1$ and the black dots are the atoms on $L_2$.

The second case is similar to the first one, except that the bottom layer $L_1$ is solidified from a random initial condition, as shown in figure 6 (a). The entire domain can be briefly divided into two regions, with 5/7 carbon rings along the grain boundary. Then, the top layer begins to form with the adjacent layer potential between $L_1$ and $L_2$. As shown in figure 6 (b) and (c), $L_2$ is also highly affected by the bottom layer $L_1$, and $L_2$ can be briefly divided into two regions. The shape and location of the grain boundary, i.e., the 5/7 carbon rings, is close to those on $L_1$. More than half of the entire domain is (close to) the AB/BA stacking, and exceptions are located around the grain boundary, as shown in figure 6 (d) (see more details in the supplementary materials).

Figure 7. (a) Layout of the atomic configuration for $L_1$, (b) layout of $L_2$, (c) $L_1$ and $L_2$ stacking, where the blue dots are the atoms on $L_1$, and the black dots are the atoms on $L_2$, and (d) phase field of $L_2$. 
The last case is that the solidification of the two layers starts at the same time with different initial random conditions. This is an interesting case that helps in understanding the limit of the effect of the adjacent layer potential. The results are shown in figure 7, which shows that the locations of the 5/7 rings of the two layers are not strongly related. Additionally, there are only small regions near the AB/BA stacking order, as shown in figure 7 (c). The layout of the atomic configuration can be divided into several patches by the grain boundaries composed of 5/7 carbon rings and more unstructured than the previous case. Thus, once the solidification is stable, the atomic layout can not be easily changed under the effect of the adjacent layer potential, especially for layouts with defects.

3.2.2. Trilayer Structure. Next, we study the trilayer structures. Again, we start with a well-structured bottom layer (L1); then, the second layer (L2) is added into the system with random initial conditions; finally, the top layer (L3) is added after the solidification of L2. The results are very similar to the first case of bilayer structures, and L3 is almost identical to L1. The L1 and L2 stackings are perfect AB/BA stacking, as are the L2 and L3 stackings.

The second case is that the initial condition of L1 is a constant (0.3) with small perturbations of Gaussian noise. After L1 has been solidified, L2 is added into the system with similar initial conditions; finally, L3 is added. The layouts of the atomic configuration for all three layers are shown in figure 8. The top layer is highly affected by the bottom layer for both L2&L1 and L3&L2. However, L3 is not identical to L1, as shown in figure 8 (f), where only approximately one-third of the atoms on L1 (blue dots) and L3 (red circles) are close to overlapping in the $x - y$ plane.

**Figure 8.** (a) Layout of the atomic configuration for the bottom layer L1, (b) layout of L2, (c) layout of L3, (d) L1 (blue dots) and L2 (black dots) stacking, (e) L2 (black dots) and L3 (blue dots) stacking, and (f) layouts of L1 (blue dots) and L3 (red dots).
We further investigate the details of the differences between the bilayer structure and the trilayer structure. As shown in figure 8 (d) and (e), the L1 & L2 stacking is similar to the bilayer case (in figure 6 (d)), where more than half of the domains are AB/BA stacking. However, the L2&L3 stacking is not the same, since nearly one fourth of the domain around the mid-bottom, i.e., the region circled by a red ellipse is away from the AB/BA stacking. Nevertheless, the atomic layouts for both L2 and L3 in this region are well structured, neatly arranged hexagons along the same direction. This structure can be explained by two reasons. First, the interlayer interactions dominate the process by which L3 evolves to a well-structured layout for the bottom half of the domain. Secondly, the AB/BA stacking between L1 and L2 prevents L2 layer from further evolving to a more neatly arranged hexagonal structure and AB/BA stacking with L3. The stacking order of L2 and L3 in the red ellipse is type of transition state between AB and BA stacking on the left and right.

The third case is that we start with a well structured L1, and then L2 and L3 are simultaneously added into the system. The final results are identical to those in case 1, where both L1&L2 stacking and L2 & L3 stacking are perfect AB-BA stacking. The fourth case is similar to case 3, except that the initial condition of L1 is of constant value (0.3) with a small perturbation of Gaussian noise. L2 and L3 are added into the system together after the solidification of L1. The results are shown in figure 9. Unlike case 2, here, L1 and L3 are almost identical, as shown in figure 9 (d). The reason is that at the beginning of solidification, the phase field L2 mimics the opposite of L1, i.e., \( \phi_2 \sim -\phi_1 \), and the phase field of L3 mimics the opposite of L2, which is L1, i.e., \( \phi_3 \sim -\phi_2 \sim \phi_1 \). Therefore, the solidification of L3 is highly affected by L1, which is not true for case two that L3 is not directly affected by L1.

The fifth case is similar to case four, except that the effective mobility \( M_\psi \) for L3 is smaller, e.g., \( M_{\psi_3} = \frac{1}{3} M_{\psi_2} \), i.e., the phase field of the top layer L3 diffuses more slowly than L2. The results are shown in figure 10, and there are slight differences between \( L_1 \) and \( L_3 \). In a certain sense, case five is more realistic: once a part of L2 is solidified, carbon rings on L3 may also be solidified onto that region. Certainly, solidification of carbon rings on L3 is slower than on L2.

**Figure 9.** (a) the layout of the atomic configuration for the bottom layer L1, (b) layout L2, (c) layout of L1 (blue dots) and L2 (black dots), (d) layout of L1 (blue dots) and L3 (red dots).
Finally, we run a special case where solidifications of all three layers start simultaneously, each with randomly generated initial conditions with different Gaussian noise. As shown in figure 11, only a small part of the domain for each pair is close to the AB/BA stacking order, as shown in figure 11 (d) and (e). $L1 \& L3$ are far from consistent.

3.2.3. Multiple Layer System. We also simulated a multilayer system, e.g., 4 layers, 5 layers, 6 layers, etc. Here, we present a six-layer graphene structure, where one more layer is added into the system after the
solidification of the previous layer. By doing this, the 4-layer and 5-layer structures are presented. The results are shown in figure 12. The odd layers \( L1, L3, \) and \( L5 \) follow one pattern, and the even layers \( L2, L4, \) and \( L6 \) follow another. As expected, there are small differences between the odd layers, e.g., the locations and shape of the 5/7 carbon rings near the center are different for \( L1, L3, \) and \( L5, \) and the same occurs for the even layers.

### 3.3. Graphene Bilayers Twisted with Magic Angle of 1.1°

Finally, we simulate the case of twisted stacking bilayer graphene under a "magic angle of 1.1°", where unconventional superconductivity was found in so-called magic-angle graphene superlattices [5]. Following [14, 15], we use a supercell method to set up the periodic boundary condition for the twisted layer. Rotation angle \( \theta \) between the two layers is calculated by

\[
\cos \theta = \frac{n^2 + 4nm + m^2}{2(n^2 + nm + m^2)}
\]

where \( n, m \) are the coordinates of the lattice vector \( \mathbf{V}(m,n) \) and \( \mathbf{V}(n,m) \) with respect to the basis \( \mathbf{a}_1(\sqrt{3}/2, -1/2), \mathbf{a}_2(\sqrt{3}/2, 1/2). \)

We set \( n = 59, m = 61 \) when the twisted angle \( \theta = 1.1026°, \) which is a good approximation of the magic angle 1.1° (the combination \( n = 29, m = 30 \) and \( \theta = 1.1213° \) can also be a sufficient approximation and computationally much cheaper). We run the simulation until an equilibrium is reached.

In figure 13 (a), we plot \( d_c \) at \( T = 0. \) Here, \( d_c \) is slightly different from the previous definition. It is the
horizontal distance from the center of the hole on the bottom layer to the nearest center of the hole on the top layer. Therefore, when \( d_c = 0 \), it is AA stacking (or BB stacking), while \( d_c = 1 \) is AB stacking or BA stacking. The advantage of this parameter \( d_c \) is that it is easier to tell the effect of the relaxation between figure 13 (a), the initial setup, and figure 13 (c) when \( T \to \infty \). One can also easily tell the detailed differences between figure 13 (b) and (d), that the relaxation is to reduce the region of the AA stacking, while the AB and BA stacking regions are significantly increased. Taking \( d_c \approx 0.5 \) as the boundary, the area of the AA stacking region is reduced to a quarter of its original value (radius approximately decreased from 28 \( a_e \) to 14 \( a_e \)). The average value of \( d_c \) when \( T = 0 \) is 0.601, comparing with 0.728 when \( T \to \infty \), which helps to quantify the shift.

To distinguish the AB stacking region from the BA region, we defined a new parameter \( \bar{d}_c \) as follows. We first calculate distance \( \bar{d}_c \), which is the horizontal distance from the center of the holes on the bottom layer to the nearest group A atoms on the top layer. Second, we calculate distance \( \bar{d}_c \), which is the horizontal distance from the center of the holes on the bottom layer to the nearest group B atoms on the top layer. Then, we define \( \bar{d}_c = d_1 - d_2 \). Therefore, \( \bar{d}_c = -1 \) for AB stacking while \( \bar{d}_c = 1 \) for BA stacking. In figure 13 (e) and (f), we plot \( \bar{d}_c \) at \( T = 0 \) and \( T = 1.5 \times 10^4 \). In figure 13 (g) and (h), we plot the details of the AB stacking region and BA stacking region when \( T \to \infty \). The results are consistent with our expectations.

**Figure 13.** Initial setup compared with the final equilibrium state. (a) \( d_c(x, y) \) when \( T = 0 \); (b) \( d_c(x, y) \) when \( T \to \infty \); (c) the initial state of the two layers around the AA center when \( T = 0 \); (d) initial state of the two layers around the AA center when \( T \to \infty \); (e) \( \bar{d}_c(x, y) \) when \( T = 0 \); (f) \( \bar{d}_c(x, y) \) when \( T \to \infty \); (g) equilibrium state of the two layers around the BA center when \( T \to \infty \); (h) equilibrium state of the two layers around the AB center when \( T \to \infty \).

4. Discussion

In this paper, we build a new XPFC model for multilayer graphene by extending the XPFC method. The adjacent layer potential that we use in this paper is similar to the generalized stacking fault energy, but we use the corresponding phase field instead. Thus, the adjacent layer potential changes over time, and systems of multiple layers can be easily built up.

We used the width of the AB-BA transition region of a long strip to determine the exact strength of the adjacent layer potential, and the results agree with atomistic simulations. Then, we simulate the multilayer graphene structures. We test bilayers, trilayers, and multiple-layers. We have tried different initial conditions and various orders of construction. Once a base layer has been formed, layers on the top will be
affected, and AB/BA stacking is more likely to be formed. Moreover, the defect grain boundaries will emerge around similar locations. For the well-structured bottom layer, the layers onto it are very likely to solidify into well-structured layouts. In contrast, if there is not a well-structured base layer and multiple layers solidify simultaneously, adjacent layers will not be strongly correlated, and the grain boundaries composed of 5/7 carbon rings are significantly different from one another.

A natural next step is the rotational bilayer structure, for example, the “1.1°” magic degree rotation. Following the supercell method, it is not difficult to construct the initial phase field for all layers. Although several setups of the simulations appear to be unphysical, we hope that our numerical experiments help understand the effect of the adjacent layer potential and the formation of multiple layer graphene structures. We believe that those "imaginary" cases are also as valuable as the "real" ones, since they can not be easily tested by experiments.

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