Stacking Textures and Singularities in Bilayer Graphene

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We study a family of globally smooth spatially varying two dimensional stacking textures in bilayer graphene. We find that the strain-minimizing stacking patterns connecting inequivalent ground states with local \(AB\) and \(BA\) interlayer registries are two dimensional twisted textures of an interlayer displacement field. We construct and display these topological stacking textures for bilayer graphene, examine their interactions and develop the composition rules that allow us to represent other more complex stacking textures, including globally twisted graphenes and extended one dimensional domain walls.

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Bilayer graphene (BLG) features special functionalities that microscopically derive from various forms of broken sublattice symmetry present when graphene sheets are stacked. These depend on relative lateral translations \([1,3]\), rotations \([4,5]\) and layer symmetry breaking that can occur spontaneously \([6–8]\) or be induced \([9–20]\). There has been important recent progress imaging the stacking order in BLG using dark field transmission electron microscopy \([21,22]\). These experiments reveal rich submicron domain structures with locally registered \(AB\) and \(BA\) regions delineated by dense irregular networks of domain walls, focusing attention on the inevitable competition between intralayer strain and interlayer stacking commensuration energies \([21,23]\).

In this Letter we examine a general family of two dimensional stacking textures in BLG and their defects. We find that the strain minimizing stacking patterns that connect inequivalent ground states are twisted textures of the interlayer displacement field. We construct and display these topological stacking textures in BLG, examine their interactions and develop the composition rules that allow us to represent other observed complex stacking textures, including twisted graphenes and extended one dimensional domain walls.

A relative translation between two graphene layers is represented by an interlayer displacement vector \(\Delta = f_1T_1 + f_2T_2\) where \(T_{1,2} = a \exp(\pm i\pi/3)\) are primitive translation vectors of a graphene lattice with lattice constant \(a\). The lowest energy uniformly translated structures (Fig. 1) align the \(A\) and \(B\) sublattices of the two layers at \((f_1,f_2) = (2/3,1/3)\) (\(AB\) stacking: \(\Delta_\alpha\)) and its complement \((1/3,2/3)\) (\(BA\) stacking: \(\Delta_\beta\)). The interlayer potential is a periodic function \(U(\Delta) = u_0 + u_1 \sum \exp \left(i \left(G_n \Delta + G_n \Delta^* \right) / 2 \right)\) where \(G_n = (4\pi/\sqrt{3}a)\exp(i(2n-1)\pi/6)\) are vectors in the first star of reciprocal lattice vectors. For BLG \(u_1 \simeq 2.1\) meV/atom and \(u_0 = 3u_1\) assigns the zero of energy to the \(\alpha\)- and \(\beta\)-stacked states \([23]\).

The configuration space for \(\Delta\) has the topology of a torus. When \(u_1\) is large the energy minima at \(\alpha\) and \(\beta\) are deep and the shortest trajectories \(\Delta(\vec{r})\) connecting the two inequivalent minima \(\alpha\) and \(\beta\) are three saddle point paths crossing the points labelled \(s_r, s_g\) and \(s_b\) in Fig. 1, indexed by their winding on the two cycles of the torus. Each saddle point trajectory is bisected by a (straight) domain wall which runs along one of the three symmetry-related directions as shown.

We focus on field textures \(\Delta\) that satisfy the boundary conditions \(\Delta(\infty) = \Delta_\alpha\) and \(\Delta(0) = \Delta_\beta\). A simple texture that accomplishes this wraps a stacking domain wall into a loop thereby reversing the stacking order within a confined region, as shown in Fig. 2 for the field \(\Delta_\alpha(z) = (1/2)(\Delta_\alpha + \Delta_\beta - (\Delta_\beta - \Delta_\alpha) \tanh(|z| - R)/\ell)\) where \(z = x + iy\) is the complex coordinate in the plane. This texture connects two ground states through a transition region of width \(\ell\) accumulating lattice strain in an annulus. This passage passes through the value...
algebraically with a logarithmically divergent energy field the exterior solution relaxes to its ground state these objects, with vector charges

defects in “gauge neutral” clusters. On scales large compared to the separation of these objects, with vector charges \( s_i \) located at positions \( z_i \) \( (i = 1, N) \), the exterior displacement pattern is \( \Delta^> = \Delta_\alpha + \sum_{i=1}^N s_i \bar{z}_i / (\bar{z}_i - \bar{z}) \) which eliminates the 1/\( \bar{z} \) tail when \( \sum_{i=1}^N s_i \bar{z}_i = 0 \). An important case is \( N = 3 \) with \( s_1 = s_2 = s_3 \) and \( z_2 = z_3 = z_1 \exp(2\pi i/3) \) and \( z_3 = z_1 \exp(-2\pi i/3) \) which breaks rotational symmetry by the selection of a single direction \( s_1 \) for the triad, but nulls the monopole field by the threefold symmetry of the positions of the defect centers. The left panel of Fig. 4 illustrates the field energy density and critical lines for one such texture. The broken symmetry opens the “red” boundary curve which then links the three defects, while the “green” and “blue” critical lines form closed orbits that are confined around the individual defect centers.

One can iterate this process, uniformly distributing \( N \)
FIG. 3: Two globally smooth stacking textures with \( \Delta(\infty) = \Delta_\alpha \) and \( \Delta(0) = \Delta_\beta \). The density plots give the commensuration (potential) energy densities and the lines are the mapping of the color coded symmetry lines of Fig. 1 onto the textures. The left hand plot represents a texture that is antianalytic everywhere and contains a pole inside its core at \( z = z_0 \) (density plot is cut off at the white disk for clarity). The right hand plot represents a texture that matches an antianalytic function in the exterior region to an analytic function in the interior. The wire frame model of the lattice (background) illustrates the stacking pattern for the right hand texture (lattice constant greatly exaggerated for clarity.)

For \( s_i = s_0 = (\Delta_\beta - \Delta_\alpha)/N \) and \( N \to \infty \) one obtains \( \Delta^\ast = \Delta^\ast_\alpha \) and \( \Delta^\sim = \Delta_\beta \) representing a rigid interlayer translation for \( |z| < R \) and reproducing the annular domain wall pattern of Fig. 1. For general \( N \), one can regard \( \Delta_N \) with \( s_i = s_0 \) as a family of trial solutions where the value of \( N_{\text{min}} \) is selected to minimize the sum of the elastic and commensuration energies \( U_e + U_c \). \( U_e(U_c) \) are increasing/(decreasing) functions of \( N \), with a finite \( N_{\text{min}} \). The domain wall structures experimentally observed in BLG are relatively wide [21, 22] indicating that the system is indeed in the regime dominated by the elastic energy, favoring smooth (smaller \( N \)) over sharp (larger \( N \)) solutions. This is expected since the potential energy landscape has quite broad minima in its low energy configuration space.

The textures given by Eqns. (1,2) realize identical \( \alpha \) stackings at \( |z| = \infty \) and in the near field at \( z = z_0 \) and are stable because of an additional boundary condition that clamps a different state at the origin: \( \Delta(0) = \Delta_\beta \). However, \( \beta \) stacking at the origin occurs for a lattice of possible clamped states, each relatively shifted by discrete lattice translation vectors \( \Delta_{l,m}(0) = \Delta_\beta + lT_1 + mT_2 \). Choices of \( l \) and \( m \) give the winding of the order parameter \( \Delta \) on the two cycles of a torus between \( z = 0 \) and \( z = \infty \) and index topologically distinct solutions \( \Delta_{l,m} \). Fig. 4 displays the field for the case \( l = 1 \) and \( m = 2 \). Remarkably, we find that the interior solution represents a circular domain of uniformly rotated (twisted) graphene continuously matched to an untwisted and minimally strained exterior texture. This illustrates a plausible mechanism for the formation of the observed complex stacking textures in BLG. Isolated domains likely grow with uncorrelated local rotational registries forcing a complex stacking texture in a state of minimum strain when the bilayer becomes continuous.

The textures identified here have well-studied analogs in (at least) two other physical contexts. First, they are similar, though not identical to, the static baby skyrmion defects with charges \( s_i \) on a circle at positions \( Re^{i\phi_i} \), thereby cancelling its higher order multipoles. In this case the texture has an expansion

\[
\Delta_N^\sim = \Delta_\alpha + \sum_{l=1}^{N} s_i \left( 1 - \frac{2e^{-i\phi_i}}{R} \right) ; |z| < R
\]

\[
\Delta_N^\ast = \Delta_\alpha - \sum_{p \geq 1} \left( \frac{R}{z} \right)^p \sum_{l=0}^{N} s_i e^{-ip\phi_i} ; |z| > R
\]

For \( s_i = s_0 = (\Delta_\beta - \Delta_\alpha)/N \) and \( N \to \infty \) one obtains \( \Delta^\ast = \Delta^\ast_\alpha \) and \( \Delta^\sim = \Delta_\beta \) representing a rigid interlayer translation for \( |z| < R \) and reproducing the annular domain wall pattern of Fig. 1. For general \( N \), one can regard \( \Delta_N \) with \( s_i = s_0 \) as a family of trial solutions where the value of \( N_{\text{min}} \) is selected to minimize the sum of the elastic and commensuration energies \( U_e + U_c \). \( U_e(U_c) \) are increasing/(decreasing) functions of \( N \), with a finite \( N_{\text{min}} \). The domain wall structures experimentally observed in BLG are relatively wide [21, 22] indicating that the system is indeed in the regime dominated by the elastic energy, favoring smooth (smaller \( N \)) over sharp (larger \( N \)) solutions. This is expected since the potential energy landscape has quite broad minima in its low energy configuration space.

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solutions of the two dimensional nonlinear sigma model [21, 22]. Normalizing our solution to its maximum value realized on the matching radius, \( \Delta(z)/|\Delta(R)| = \vec{n}_L \) is the projection of a three dimensional unit vector \( \vec{n} \) onto the plane and has a slower far field relaxation of not obtained by a stereographic projection of the sphere.

Baby skyrmion with the same degree instead minimizes \( O(\text{unmeasured}) \) normal component \( n_\perp \). Second, our solutions recognize as the classical field solutions from ordinary 2D electrostatics and magnetostatics where they represent the field profile of a uniformly charged rod or current carrying wire. An interesting difference is that in BLG the field energy density vanishes for two nonzero values of the field (corresponding to degenerate configurations) instead of just one state \((\vec{E}, \vec{B} = 0)\).

Our approach provides a unified treatment of stacking point defects, domain walls and twisted graphene and provides a direction for further investigation of these systems. For example it is possible that the complicated submicron structure observed in these systems can be understood in terms of only a few fundamental stacking motifs and their conformal maps onto spatially varying geometries imposed by pinning centers and irregularities in the sample morphology. More intriguingly, it is possible that a desired BLG stacking texture could be controllably engineered using a combination of choice of substrate, growth face, macroscopic curvature and various forms of submicron templating. Finally, although our model is designed to study static low strain stacking configurations, they may also be important for nonlinear tribological properties of BLG, where defects of the type studied here are generated when an applied mechanical load exceeds a critical yield stress [23].

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