Band structure and gaps of triangular graphene superlattices

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General properties of long wavelength triangular graphene superlattice are studied. It is shown that Dirac points with and without gaps can arise at a number of high symmetry points of the Brillouin Zone. The existence of gaps can lead to insulating behavior at commensurate fillings. Strain and magnetic superlattices are also discussed.

I. INTRODUCTION

Graphene is a two dimensional metal when carriers are induced by an electric field. A gap at the Fermi level has been observed by STM measurements (see also). We analyze the gaps induced by a periodic structure, and the possibility that these gaps are generated spontaneously.

Graphene superlattices have been observed in graphene layers grown on transition metals (see also). Superlattices are also found in graphene grown by the decomposition of SiC. In general, graphene superlattices can have interesting properties, such as highly anisotropic transport properties, or Dirac points at finite energies. In general, the study of the properties of graphene superlattices has attracted great interest, due to the many novel features their electronic spectra can show.

In this paper, we study the general properties of triangular graphene superlattices created by a scalar potential, followed by a discussion of strain and magnetic superlattices.

II. BRILLOUIN ZONE OF TRIANGULAR GRAPHENE SUPERLATTICES

We define the lattice vectors of the graphene lattice as:

\[ \tilde{a}_1 \equiv n_x \]
\[ \tilde{a}_2 \equiv \frac{1}{2}n_x + \frac{\sqrt{3}}{2}n_y \]  (1)

A triangular superlattice is described by the unit vectors:

\[ \tilde{b}_1 \equiv n_1 \tilde{a}_1 + n_2 \tilde{a}_2 \]
\[ \tilde{b}_2 \equiv -n_2 \tilde{a}_1 + (n_1 + n_2)\tilde{a}_2 \]  (2)

where \( n_1 \) and \( n_2 \) are arbitrary integers different from zero.

There are three types of high symmetry points in the Brillouin Zone of a triangular lattice, \( \Gamma, M \) and \( K \). There are two inequivalent \( K \) points, \( K \) and \( K' \), at the corners of the hexagonal Brillouin Zone, and three inequivalent \( M \) points, at the centers of the edges. Time reversal exchanges \( K \) and \( K' \), while leaving the \( \Gamma \) point and the three \( M \) points unchanged. The vectors which define these points are such that:

\[ \tilde{\Gamma} \tilde{a}_1 = \tilde{\Gamma} \tilde{a}_2 = 0 \]
\[ \tilde{K} \tilde{a}_1 = \frac{4\pi}{3} \tilde{K} \tilde{a}_2 = \frac{2\pi}{3} \]
\[ \tilde{K}' \tilde{a}_1 = \frac{2\pi}{3} \tilde{K}' \tilde{a}_2 = \frac{4\pi}{3} \]
\[ \tilde{M}_1 \tilde{a}_1 = \pi \tilde{M}_1 \tilde{a}_2 = 0 \]
\[ \tilde{M}_2 \tilde{a}_1 = \pi \tilde{M}_2 \tilde{a}_2 = 0 \]
\[ \tilde{M}_3 \tilde{a}_1 = 0 \tilde{M}_3 \tilde{a}_2 = \pi \]  (3)

The low energy states of graphene lie close to the \( K \) and \( K' \) of the original Brillouin Zone. The positions of
there is a twofold degeneracy at the three

Thus, when $2n_1 + n_2$ is a multiple of three the graphene $K$ and $K'$ points will be mapped onto the $\Gamma_S$ point of the superlattice Brillouin Zone. Otherwise, they will be mapped onto the corners of the Brillouin zone, $K_S$ and $K'_S$. Examples of superlattice Brillouin Zones are given in Fig. 1.

III. DISPERSION NEAR HIGH SYMMETRY POINTS

A. The model

We study superlattices induced by a modulation of the on site energy of the $\pi$ orbitals. We assume that it is a weak perturbation of the graphene Dirac equation, except in cases where degeneracies occur. We consider the Fourier components of the potential with lowest wavevector, of modulus:

$$G = \frac{4\pi}{a\sqrt{3(n_1^2 + n_2^2 + n_1n_2)}}$$

(5)

We write the potential as the sum of symmetric part $V_G$, and an antisymmetric part, $\Delta_G$, with respect to the interchange of sublattices. We neglect for the moment short wavelength components which mix the two inequivalent Dirac points of the unperturbed graphene layer.

We analyze the changes in the Fermi velocity near the Dirac energy induced by the superlattice potential, and the points in the lowest bands of the superlattice where degeneracies persist when $V_G \neq 0$ and $\Delta_G = 0$. As discussed below, this situation gives rise to a new set of Dirac equations at finite energies.

B. Dirac energy at the $\Gamma_S$ point

We consider the case when the $K$ and $K'$ points of the graphene Brillouin Zone are mapped onto the $\Gamma_S$ point of the superlattice Brillouin Zone. Using lowest order perturbation theory, we find, near the $\Gamma_S$ point a renormalization of the Fermi velocity:

$$\delta v_F \approx -\frac{6|V_G|^2}{v_FG^2} + \frac{6|\Delta_G|^2}{v_FG^2}$$

(6)

There is a twofold degeneracy at the three $M_S$ points, if $\Delta_G = 0$. The energy of these states is $v_FG/2$. At finite distances from the $M_S$ points, we can write an effective hamiltonian:

$$H \equiv \left( \frac{v_FG}{\sqrt{3}} + \frac{v_Fk_x}{2} \right) \frac{\Delta_G + 4iv_Gk_x}{G} \frac{v_FG}{2} - v_Fk_x$$

(7)

which gives an anisotropic Dirac equation with a gap:

$$\epsilon_{M_S} \approx v_FG/\sqrt{3} \pm \frac{4v_F^2k^2_x + \Delta_G^2 + 16v_G^2k^2_y}{G^2}$$

(8)

At the $K_S$ and $K'_S$ points, there are three degenerate levels for $V_G = \Delta_G = 0$, with energy $\epsilon_{K_S} = v_FG/\sqrt{3}$. When $V_G \neq 0$ these three levels are split into a doublet, with energy $\epsilon_{K_S}^d = v_FG/\sqrt{3} + V_G/2$, and a singlet, at $\epsilon_{K_S}' = v_FG/\sqrt{3} - V_G$. Expanding around the $K_S$ point, the effective hamiltonian for the doublet is:

$$H \equiv \left( \frac{v_FG}{\sqrt{3}} + \frac{v_Fk_x}{2} + \frac{v_Fk_y}{2} \right) \frac{\Delta_G^2 + 16v_G^2k^2_y}{G^2} + \frac{3\Delta_G}{G}$$

(9)

This is the two dimensional Dirac equation with a mass term. The dispersion relation is:

$$\epsilon_{K_S}^d \approx v_FG\sqrt{3} - V_G + O\left( \frac{\Delta_G^2}{V_G}, \frac{v_Fk_x}{G} \right)$$

$$\epsilon_{K_S}' \approx v_FG\sqrt{3} + \frac{V_G}{2} \pm \frac{v_F^2(k_x^2 + k_y^2)}{4} + \frac{9\Delta_G^2}{16}$$

(10)

There are two sets of degenerate bands, derived from the $K$ and $K'$ points of the Brillouin Zone of graphene. This degeneracy will be broken by short wavelength terms in the superlattice potential.

C. Dirac energy at the $K_S$ and $K'_S$ points

The renormalization of the Fermi velocity near the $K_S$ and $K'_S$ points is the same as in eq. (6). There are doubly degenerate states, even when $V_G \neq 0$, at the six inequivalent points at positions $\vec{L}_S = \vec{K}_S/2$, as shown in Fig. 2. The energy of these points is
\[ \epsilon_{L_{S}} = v_{F} G/2. \] Expanding around these points, we find an effective anisotropic Dirac equation, given by eq.(7).

There are another set of doubly degenerate states at the \( M_{S} \) points. The two states arise from the \( K \) and \( K' \) points of the original graphene Brillouin Zone. The degeneracy persists when \( V_{G} \neq 0 \) and \( \Delta_{G} \neq 0 \), and is only broken by short wavelength components of the superlattice potential. When these components are finite, an effective anisotropic Dirac equation will arise similar to that in eq.(7).

For \( V_{G} = \Delta_{G} = 0 \) there are six degenerate states at the \( \Gamma_{S} \) point. The long range part of the superlattice potential will hybridize states which are derived from the \( K \) and \( K' \) points of the original graphene Brillouin Zone. We obtain two sets of isotropic Dirac equations, described by eq.(8), and two degenerate states. The short range part of the Dirac equation will break these degeneracies.

\[ \epsilon_{K_{S}} \approx \frac{v_{F} G}{\sqrt{3}} - V_{G} \leq \epsilon_{M_{S}} \approx \frac{v_{F} G}{2} - \Delta_{G} \]
\[ \epsilon_{k_{M_{S}}}^{\pm} \approx \frac{v_{F} G}{2} + \Delta_{G} \leq \epsilon_{K_{S}} \approx \frac{v_{F} G}{\sqrt{3}} + \frac{V_{G}}{2} - \frac{3\Delta_{G}}{4} \quad (11) \]

The results are in reasonable agreement with the analytical description in the previous section. A gap of order \( 2\Delta_{G} \) is induced at the \( M_{S} \) point. In order for this gap to be possible, the following inequalities must be satisfied:

We analyze the bands induced by a \( N \times N \) superlattice. The hopping matrix between \( \pi \) orbitals in neighboring carbon atoms is \( t = 3eV \). The bands for \( V_{G} = 0.3eV \) and \( \Delta_{G} = 0 \) are shown in Fig. 3. The bands show Dirac points at the \( M_{S} \) and \( K_{S} \) points. When \( \Delta_{G} \) is increased to \( \Delta_{G} = 0.1eV \) a gap appears between successive bands, as shown in Fig. 4. The density of states for those two cases is shown in Fig. 5. Note that the potential breaks the electron-hole symmetry of clean graphene, and the gaps are not of the same magnitude for positive and negative energies.

The scaling properties of the Dirac equation imply that, if the dimension of the superlattice is increased, \( G \rightarrow \lambda G \), with \( \lambda < 1 \), a rescaling of the superlattice potential, \( V_{G} \rightarrow \lambda V_{G} \), \( \Delta_{G} \rightarrow \lambda \Delta_{G} \), will lead to the same band structure, with energies scaled as \( E \rightarrow \lambda E \).
### IV. STRAIN SUPERLATTICES

A superlattice can also be produced by inducing strains, which modulate the interatomic hoppings. The corresponding perturbation can be seen as a gauge field, $\hat{A}$, which shifts locally the momentum $\{k\}$. A simple case is when the strains are due to height modulations, $h(\vec{r})$, which can be induced by a substrate. In terms of the Fourier components of the modulation, $h(\vec{G})$, and allowing for the relaxation of the in plane displacements, the effective gauge field can be written as:

$$A_x(\vec{G}) = \frac{(\lambda + \mu)(G_x^2 - G_y^2)}{|\vec{G}|^4(\lambda + 2\mu)} \left[ G_x^2 G_y^2 - (h_{G_y}^G + h_{G_x}^G)G_x G_y + h_{G_y}^G G_x^2 \right]$$

$$A_y(\vec{G}) = \frac{(\lambda + \mu)2G_x G_y}{|\vec{G}|^4(\lambda + 2\mu)} \left[ h_{G_y}^G G_x^2 - (h_{G_y}^G + h_{G_x}^G)G_x G_y + h_{G_y}^G G_x^2 \right]$$

where the tensor $h_{ij}^G$ is the Fourier transform of the function:

$$h_{ij}(\vec{r}) = \frac{\partial h}{\partial x_i} \frac{\partial h}{\partial x_j}$$

and $\lambda$ and $\mu$ are the elastic Lamé coefficients of graphene. The field in eq. 12 has non zero components for all combinations of the type $\vec{G}_k + \vec{G}_l$. When the gauge field, $\hat{A}(\vec{G})$ is parallel to $\vec{G}$, the vector potential can be gauged away, and does not induce an effective magnetic field. This implies that out of the 18 possible values of the vector $\vec{G}_k + \vec{G}_l$, only six contribute to the effective magnetic field. These vectors are given by $\vec{G} \equiv G(3/2, \sqrt{3}/2)$ and the vectors equivalent to it by a symmetry transformation. After some algebra, we obtain for the effective magnetic field:

$$B_{\text{strain}}(\vec{r}) = \frac{\beta \lambda + \mu}{a} \frac{27\sqrt{3}}{8} G^3 h_{G}^2 \left[ 2\cos(\sqrt{3}Gy) + 4\cos \left( \frac{\sqrt{3}Gy}{2} \right) \left( \frac{3Gx}{2} \right) \right]$$

where $\beta = \partial \log(t)/\partial \log(a) \approx 2 - 3$, $t \approx 3\text{eV}$ is the nearest neighbor hopping, and $a \approx 1.4\text{Å}$ is the distance between nearest neighbor carbon atoms. The superlattice defined by the effective magnetic field has a unit vector of length $\sqrt{3}G$, so that the area of its unit cell is smaller than the area of the unit cell of the original superlattice by a factor 1/3.

Using eq. 13 and $G = 2\pi/L$, where $L$ is the length of the unit vector of the superlattice, we can write the magnetic length associated to the maximum effective field in the system as:

$$\frac{1}{\ell_B^{\text{max}}} = \frac{\lambda + \mu}{\lambda + 2\mu} \frac{3h_{G}^2}{2L^3}$$

where $h_{G}^{\text{max}} = 6h_{G}$ is the maximum value of $h(\vec{G})$, assuming $h_{G} > 0$. For values $h_{G}^{\text{max}} \approx 1\text{nm}$, and $L \approx 40\text{nm}$, we find $l_B \approx 14\text{nm}$, so that the effective field is such that

### V. MAGNETIC SUPERLATTICES

A superlattice can also be induced by a spatially modulated magnetic field. A combination of a modulated magnetic field and a scalar potential opens a gap at the Dirac energy, and the resulting insulator is a Quantum Hall system, with chiral currents at the boundaries. Here we obtain this effect using second order perturbation theory, instead of the arguments used in. As in the previous sections, we assume the simplest periodicity...
compatible with the superlattice hexagonal symmetry

\[ V(\vec{r}) = V_G \sum_{l=1,\ldots,6} e^{i\vec{G}_{l}\vec{r}} \]

\[ B(\vec{r}) = B_G \sum_{l=1,\ldots,6} e^{i\vec{G}_{l}\vec{r}} \] (16)

The eigenstates of the unperturbed hamiltonian at the \( K \) and \( K' \) points of the Brillouin Zone can be written as \( |0\rangle_A = (1,0) \) and \( |0\rangle_B = (0,1) \), which each component of the spinor corresponds to one sublattice. These states are hybridized with states \( |\vec{G}_\pm\rangle_K = (1, \pm e^{-iG_y}) \) and \( |\vec{G}_\pm\rangle_K' = (1, \mp e^{-iG_y}) \), with energies \( \epsilon_{\vec{G}} = \pm v_F|\vec{G}| \), and \( e^{iG_y} = (G_x + iG_y)/|\vec{G}| \).

The energy of states \( |0\rangle_A \) and \( |0\rangle_B \) are modified in different ways by virtual hoppings into states \( |\vec{G}_\pm\rangle_K \) and \( |\vec{G}_\pm\rangle_K' \), leading to gaps in both valleys. Moreover, the gaps have different signs, showing that time reversal symmetry in the system is broken, and that a Quantum Hall phase has been induced. The gap can be written as

\[ \Delta = \pm \sum_{l=1,\ldots,6} 2v_F \text{Re} \left\{ A_x(\vec{G}_l) + iA_y(\vec{G}_l) \right\} e^{-i\epsilon_{\vec{G}_l}} \]

where \( \vec{A}_G \) is the vector potential, which we define as:

\[ A_x(\vec{G}) = \frac{iG_y B_0}{|\vec{G}|^2 \Phi_0} \]

\[ A_y(\vec{G}) = -iG_x B_0 \]

where \( \Phi_0 = \hbar c/e \) is the quantum unit of flux. Using this expression, we finally obtain:

\[ \Delta = 12 B_G V_G \left( \frac{\Phi_0}{B_0^2} \right) \] (19)

in agreement with\(^24\).

VI. SELF CONSISTENT OPENING OF A GAP

The previous analysis shows that gap can open at finite energies in graphene in the presence of a superlattice potential with a staggered component. When the number of carriers is such that only a small number of subbands are completely filled and the rest are completely empty the electronic energy will be lowered in the presence of the gap. A lattice distortion which leads to the appropriate potential will be energetically favorable if the gain in electronic energy exceeds the formation energy of the distortion, as in the Peierls instability in one dimension.

In graphene on top of a metal or other substrate with a large dielectric constant, such out of plane displacements lead to changes in the on site energies of the π orbitals. An electron in a given carbon atom experiences the image potential due to the screening. A change of position of \( \Delta z \) leads to a change of the image potential of order \( ee^*\Delta z/(4d^2) \), where \( e^* = e(e_0 - 1)/(e_0 + 1) \) is the image charge, \( e_0 \) is the dielectric constant of the substrate, and \( d \) is the distance to the substrate. A vertical displacement of \( \Delta z \sim 1\AA \) when the graphene layer is at a distance \( d \sim 3\AA \) of the substrate can lead to shifts of the onsite energies of order 0.1eV. The electronic gain of energy due to the existence of a gap, per unit cell, is then \( E_{\text{elec}} \approx \Delta G \approx ee^*\Delta cd^2 \).

The elastic energy per unit cell required to create a staggered distortion of amplitude \( \Delta z \) is of order \( E_{\text{elas}} \approx \kappa \Delta z^2a^{-2} \), where \( \kappa \approx 1\text{eV} \) is the bending rigidity of graphene.

A gap will exist above a threshold for the superlattice potential, \( V_G \sim \Delta G \gtrsim v_F G(1/\sqrt{3} - 1/2) \). The area of the Brillouin Zone of the supercell, \( \sqrt{3}G^2/2 \) should be close to the area within the Fermi surface of the unperturbed graphene, \( \pi k_F^2 \). Hence, a spontaneous staggered distortion is favored if:

\[ E_{\text{elec}} + E_{\text{elas}} \approx -\frac{ee^*\Delta z}{a^2} + \kappa \left( \frac{\Delta z}{a} \right)^2 < 0 \]

\[ V_G \approx \frac{ee^*\Delta z}{a^2} > v_F G \left( \frac{1}{\sqrt{3} - 1/2} \right) \]

\[ \frac{\sqrt{3}G^2}{2} \approx \pi k_F^2 \] (20)

The last equation in (20) implies that \( G \propto k_F \), and from the first two we obtain that \( v_F G \lesssim (ee^*)^2a^2d^{-4}\kappa^{-1} \).

Hence, a spontaneous distortion is energetically favored for carrier densities such that:

\[ \rho = \pi k_F^2 \lesssim \left( \frac{ee^*}{v_F^2k_F^2a^2d^8} \right)^4 \] (21)

and the staggered distortion, with inverse wavelength \( G \sim k_F \) of the order:

\[ \Delta z \sim \frac{v_Fk_Fd^2}{ee^*} \] (22)

For a metal, we have \( e_0 = \infty \) and \( e^* = e \). For \( a \approx 2\AA, \ d \approx 3\AA \) and \( \kappa \approx 1\text{eV} \), we find that a staggered corrugation is energetically favored for carrier densities \( \rho \lesssim 10^{13}\text{cm}^{-2} \), leading to maximum deformations of 0.2 Å. For SiO\(_2\), where \( e_0 = 3.9 \) and \( e^* \approx 0.6e \), the corrugations takes place for carrier densities \( \rho \lesssim 3 \times 10^{12}\text{cm}^{-2} \), and maximum deformations of 0.15 Å.

The formation of these long wavelength modulations is only possible in systems with a high degree of order, as the superlattice features will be reduced by disorder. For instance, weak scatterers with concentration \( n_{\text{imp}} \), which change the onsite potential by an amount of order \( \Delta G \), lead to a mean free path \( l_{\text{imp}} \sim v_F^2/(\Delta G^2 k_F a^3 n_{\text{imp}}) \).

A large concentration of these defects, \( n_{\text{imp}} \sim a^{-2} \) will suppress the formation of a superlattice, while affecting little the conductivity, \( \sigma \sim (e^2/h) \times (k_F l_{\text{el}}) \sim (e^2/h) \times (v_F^2a^{-2}\Delta G^2) \).
VII. CONCLUSIONS

We have analyzed the formation of Dirac points and gaps at high energy points of triangular graphene superlattices. We have shown that in some cases a gap can be formed over the entire Fermi surface, making graphene insulating. We have discussed instabilities which might give rise to the spontaneous formation of gaps of this type. General properties of strain and magnetic superlattices are also discussed.

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