Type II magic angles and interlayer-coupling-dependence of magic angles in commensurately twisted bilayer graphene

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The magic angle, at which the velocity at K point in twisted bilayer graphene becomes zero, is studied theoretically. We study the commensurately-tilted bilayer graphene in the tight-binding model with changing the interlayer distance, which can be tuned by pressure. The almost doubly degeneracy of the bands at small rotating angles ($\lesssim 1^\circ$) and the fourfold degeneracy at the Dirac points are lifted as the rotating angle is large, and the finite gap becomes not negligible at the Dirac K points, although the twofold degeneracies at the Dirac points remain above and below the energy gap. Even at the commensurately-tilted bilayer graphene with moderate rotating angles, the velocity at K points becomes zero due to the merging of the four Dirac points at the critical values of interlayer distance, despite the finite gap at the Dirac points. We call this type of magic angle as Type II magic angle. We find that the crossover from the type II magic angle to the usual magic angle, at which the band gap can be neglected, depends on the choice of a parameter for numerical differentiation to calculate the velocity at K point. The crossover occurs around $\alpha \sim 3.48^\circ$ if we take the parameter to be 0.01, and the crossover angle becomes small as the parameter is small.

I. INTRODUCTION

Single layer graphene has a two-dimensional honeycomb structure with two sites in the unit cell. Two band touch at the corners of the first Brillouin zone (K and K’ points) in single layer graphene. Although a small band gap may open due to the spin-orbit coupling, it is very small and can be neglected. Recently, bilayer graphene with a small rotating angle (twisted bilayer graphene) attracts a lot of interest. The twisted bilayer graphene has a large unit cell as the twist angle becomes small. The velocity at K point is predicted to be zero at the magic angles. It is also shown that the bands at the charge neutral point are exactly flat when the interlayer coupling is finite only between the sites belonging the different sublattice in each layers.

When the band width is small, the interaction between electrons becomes important with respect to the kinetic energy. Indeed, strongly correlated insulator phase and superconductivity in twisted bilayer graphene at the first magic angle ($\sim 1^\circ$) have been observed. The magic angle is predicted to be controlled by pressure, and shown experimentally.

Many theoretical studies have been done, but there exist a lot of mysteries remain to be revealed. Most of the previous studies have been done in the continuous model. In the continuous model the unit cell is infinite. The energy gap at K point in the moiré Brillouin zone is zero. The continuous model can be justified only in the case of small twist angles. On the other hand, the commensurately tilted bilayer graphene has a finite number of sites in the unit cell (see Appendix A). The twisted bilayer graphene in the ambient pressure has the fixed interlayer distance, and the strength of the interlayer coupling with respect to the hopping integrals in the layer is constant. Then the band gap at the Dirac points is negligibly small in small twisted angles. A finite gap, however, exists in general even without the spin-orbit coupling, when the twist angle is finite, as shown in Fig. 1. In this figure the rotating angle is take to be commensurate, i.e., the rotating angle $\alpha$ is given by a pair of integers $(m_1, m_2)$, as shown in Fig. 2. The energy gap at the Dirac points is caused by the coupling between the distant Dirac points in the Brillouin zone for each layer, which is neglected in the continuous model. This situation is similar to the zero modes in the presence of the uniform magnetic field in the single layer graphene with anisotropic hoppings. If the hoppings between the nearest sites are anisotropic in the absence of external

FIG. 1. (color online) Energy gap as a function of the interlayer distance for commensurately rotated angles with $(m_1, m_2) = (2, 1), (3, 2), (4, 3), \cdots , (28, 27)$. The gaps smaller than $\sim 10^{-14}$ are caused by numerical errors and have no serious meanings. Large open black circles, red diamonds, and green squares are the first, second and third magic angles for the commensurately rotated bilayer graphene, respectively. Large blue circles are the type II magic angles with taking $\delta_k = 0.01$. At the ambient pressure $d_z/a = 1.362$. 

FIG. 2. (color online) $\alpha$-dependence of the energy gap at the Dirac K points of the commensurately-tilted bilayer graphene for (19, 18), (27, 26), (28, 27), (29, 28), (30, 29), (31, 30). The gap is negligibly small when the rotating angle is small. 

FIG. 3. (color online) Energy gap as a function of the interlayer distance for different magic angles.
magnetic field, the Dirac points moves from K and K’ points, but the energy gap remains zero at Dirac points unless two Dirac points merge at one of the time reversal invariant momentum, Γ and three M points. There exits the zero mode in a uniform magnetic field in single layer graphene, if the hopping between nearest sites are isotropic. The energy gap becomes finite, however, if the one of the hoppings is larger than the other two hoppings. The opening of the gap in the single-layer graphene in magnetic field with the anisotropic hoppings is shown to be caused by the coupling of two Dirac points.

In this paper we study the commensurately twisted bilayer graphene in the tight-binding model with changing the interlayer distance. We mainly study the case \((m_1, m_2) = (m_1, m_1 - 1)\). When the tilting angle is smaller than a few degree at ambient pressure, the energy gap in the charge neutral case is negligible, as in the continuous model. The energy gap at the Dirac points becomes large as the twist angle becomes larger or the interlayer distance becomes smaller. Even with the finite energy gap at K point, the velocity at K point in commensurately twisted bilayer graphene becomes zero (i.e. the twist angle is a magic angle) at the critical value of the interlayer distance, which we show depend continuously on the interlayer distance, as shown in Fig. 2.

II. TIGHT-BINDING MODEL IN COMMENSURATELY TWISTED BILAYER GRAPHENE

We study the tight-binding model in the commensurately twisted bilayer graphene. The details are given in Appendix A. The rotating angle \(\alpha\) is given by two integers \(m_1\) and \(m_2\) as

\[
\alpha = \arccos \left( \frac{m_1^2 + 4m_1m_2 + m_2^2}{2(m_1^2 + m_1m_2 + m_2^2)} \right). 
\]

The number of sites in the unit cell is

\[
n_0 = 4(m_1^2 + m_1m_2 + m_2^2), \]

i.e., \(n_0\) A sites and \(n_0\) B sites in the first layer, and \(n_0\) A sites and \(n_0\) B sites in the second layer. We plot \(\alpha\) as a function of \(m_1\) in Fig. 2. The Hamiltonian is given by

\[
\mathcal{H} = - \sum_{i,j} t(r_i, r_j) c_i^\dagger c_j, 
\]

where \(r_i\) and \(r_j\) are the lattice sites in the commensurately twisted bilayer graphene. The hopping matrix elements are given by

\[
t(r_i, r_j) = V_{pp\tau}^0 \exp \left( -\frac{d - d_0}{\delta} \right) \left(1 - \left(\frac{d_z}{d}\right)^2\right) \\
+ V_{pp\sigma}^0 \exp \left( -\frac{d - d_0}{\delta} \right) \left(\frac{d_z}{d}\right)^2,
\]

where \(V_{pp\tau}^0 \approx -0.27eV\) is the transfer integral between the nearest sites in the same layer (the distance between the nearest atoms is \(d_0 = a/\sqrt{3} \approx 0.142nm\)), \(V_{pp\sigma}^0 \approx 0.48eV\) is the transfer integral between the atoms

FIG. 2. (color online). Rotating angle of the commensurately rotated bilayer graphene with \(m_1\) and \(m_2\).

FIG. 3. (color online). First magic angles (black circles), second magic angles (red diamonds), and third magic angles (green squares) in the case of commensurately rotated bilayer graphene with \(m_2 = m_1 - 1\) \((m_1 = 2, 3, 4, \ldots 28)\) as a function of the interlayer distance \(d_z/a\). The blue triangle and orange downward triangle are the magic angle for commensurate angles \(m_2 = m_1 - 2\) \(= 17\) and \(m_1 = 26\) and \(m_2 = 1\) with \(\alpha' = 60^\circ - \alpha\), respectively. These two points are on the smooth line of \((m_1, m_1 - 1)\).
in different layers with the same $x, y$ coordinates at ambient pressure (the distance between the atoms is $d_0 ≈ 0.335\text{nm} ≈ 1.362a$), the decay length of the transfer integrals is $\delta ≈ 0.184a$, and $d = |\mathbf{r}_i - \mathbf{r}_j|$ is the distance between $\mathbf{r}_i$ and $\mathbf{r}_j$. We take the interlayer distance $d_z$ as a variable parameter, which can be changed by pressure. Note that $d = \sqrt{d_x^2 + d_y^2}$, if $\mathbf{r}_i$ and $\mathbf{r}_j$ are in the different layers, where $d_{xy}$ is the distance projected on the plane. Interlayer transfers are large between atoms with $d_{xy} ≪ d_z$, when the interlayer transfers are approximately given by

$$t(\mathbf{r}_i, \mathbf{r}_j) \approx V_{pp\pi}^0 \exp\left(-\frac{d_z - d_0}{\delta}\right). \quad (5)$$

The energy gap at K point is nearly proportional to $\exp(-d_z)$ for small $d_z$ as seen in Fig. 1.

In this paper we take $a$ as a unit of the length, $V_{pp\pi}^0$ as a unit of energy, and $V_{pp\pi}^0\alpha/\hbar$ as a unit of velocity. Therefore, the velocity at the K point in a single layer graphene with only nearest-neighbor hopping, $\sqrt{V_{pp\pi}^0\alpha/\hbar}$, is $\sqrt{2}/a = 0.866$ in the unit of $V_{pp\pi}^0\alpha/\hbar$.

III. ENERGY GAP AND THE VELOCITY AT K POINT

Energy gap at K point in the commensurately rotated bilayer graphene is obtained numerically as $\epsilon_{2n_0+1}(\mathbf{K}) - \epsilon_{2n_0}(\mathbf{K})$, where $\epsilon_{2n_0+1}(\mathbf{K})$ and $\epsilon_{2n_0}(\mathbf{K})$ are the energy at K point in the $(2n_0 + 1)$th band and the $2n_0$th band from the bottom, respectively. We plot them as a function of the interlayer distance $d_z$ in Fig. 1. When the twist angle is small (e.g. $\alpha ≲ 3.481^\circ$, i.e. $(m_1, m_1 - 1)$ with $m_1 ≥ 10$) and $d_z/a$ is near the value at ambient pressure ($d_z/a = 1.362$), the energy gap at K point is negligibly small (smaller than the numerical error $\sim 10^{-14}$) as shown in Fig. 1.

We calculate the velocity of the $(2n_0 + 1)$th band at K point as

$$v = \frac{\epsilon_{2n_0+1}(\mathbf{K}) - \epsilon_{2n_0+1}(\mathbf{K})}{\delta_0 |\mathbf{K}|}, \quad (6)$$

where $\epsilon_{2n_0+1}(\mathbf{K}) = \epsilon_{2n_0+1}(\mathbf{K})$ is the energy of the $(2n_0 + 1)$th band (the band just above the half) at the wave number $(1 + \delta_k)|\mathbf{K}|$ and $\mathbf{K}$ is the wave vector of K point in commensurately-twisted bilayer graphene. Since $1.5\mathbf{K}$ is one of the M points in the extended zone scheme and $|\mathbf{K}| = |\mathbf{K} - \mathbf{G}| = 2|\mathbf{M} - \mathbf{K}|$, we also get by

$$v = \frac{\epsilon_{2n_0+1}(\mathbf{K} + 2\delta(\mathbf{M} - \mathbf{K})) - \epsilon_{2n_0+1}(\mathbf{K})}{2\delta |\mathbf{M} - \mathbf{K}|}. \quad (7)$$

In Fig. 4 we plot the velocities defined by Eq. (6) with $\delta_0 = 0.01$ as functions of $d_z/a$ for $(m_1, m_1 - 1)$ $(m_1 = 2, 3, \cdots, 28)$, $(m_1, m_2) = (19, 17)$, and $(m_1, m_2) = (26, 1)$. In our choice of parameters, the velocity at K point (the Dirac point) in the bilayer graphene is zero when $m_1 = 28$ and $m_2 = 27$ at ambient pressure when $d_z/a = 1.362$, where $a$ is the lattice constant in each layer and $d_z$ is the distance between layers. The result at $m_1 = 28$ and $m_2 = 27$ is consistent with the previous results in the tight-binding approximation[1, 3, 5, 6] and the continuous approximation[1, 3, 5, 6]. We also find that the second and the third magic angles are obtained in the tight-binding model when the interlayer distance becomes small.

As seen in Fig. 4 there is a qualitative difference between the $d_z$-dependences of $v$ near $v = 0$ between those with $m_1 ≥ 10$ ($m_2 = m_1 - 1$) and those with $m_1 ≤ 9$ ($m_2 = m_1 - 1$). The velocity of the upper $(2n_0 + 1)$th band at K point is positive near the critical value of $d_z$ when $m_1 ≥ 10$, (V-shaped cusp), while that is negative.
when $m_1 \leq 9$ (inverted V-shaped cusp). We plot the velocity of the $(2n_0 + 1)$th band (upper band) calculated by Eq. \(6\) and the velocity of the $2n_0$th band (lower band) calculated by

$$v' = \frac{\epsilon_{2n_0}(1 + \delta)|\mathbf{K}| - \epsilon_{2n_0}(\mathbf{K})}{\delta|\mathbf{K}|},$$

\(8\)

for $(m_1, m_2) = (9, 8)$ and $(10, 9)$ in Fig. 5. The velocities of the upper band $v$ and the lower band satisfies $v = -v'$ in these regions. We discuss the $(m_1, m_2) = (9, 8)$ case and $(10, 9)$ case separately in following subsections. The case of $(m_1, m_2) = (2, 1)$ is also discussed.

**A. $(m_1, m_2) = (9, 8)$**

As seen in Fig. 5(a), $v < 0$ at $d_z/a \lesssim 1.173$. This value of the sign-change point, however, is not an intrinsic one, but depends on the choice of $\delta_k$. As shown in Fig. 6, the Dirac point at K is not the degenerated point of four bands. The Dirac point is separated into two Dirac points of upper two bands and lower two bands due to the finite gap at K point. As a result the velocity of the upper band at K point is negative due to the small finite energy gap when $d_z/a = 1.18$, if we take smaller value of $\delta_k$. Therefore, the sign-change point at $d_z/a \sim 1.173$ becomes larger, if we calculate the velocity using a smaller value of $\delta_k$. Even when $d_z/a = 1.20$, there exists a finite gap at K point, as seen in Fig. 6(d). Therefore, the sign-change point at $d_z/a \sim 1.173$ is not intrinsic. In principle, the velocity of K point is negative (or zero) in the limit of $\delta \to 0$, if a gap between $(2n_0)$th band and $(2n_0 + 1)$th band cannot be neglected and there exist massless Dirac points between $(2n_0 + 1)$th band and $(2n_0 + 2)$th band.

On the other hand, the velocity of the upper band depends on the interlayer distance as the reversed V-shaped cusp around $d_z/a \sim 1.166$ in the case of $(m_1, m_2) = (9, 8)$ (see Fig. 5(a)), which is also seen in the case of $m_1 \leq 9$, $m_0 = m_1 - 1$ (see Fig. 3). The reversed V-shaped cusp dependence can be understood as follows. When the interlayer distance is close to the peak $(d_z/a \approx 1.166)$ there exist other Dirac points on the lines of K-K-M in the extended zone scheme of Brillouin zone. When $d_z/a \lesssim 1.166$ or $d_z/a \gtrsim 1.166$, three Dirac points exist near K point, as shown in Fig. 4(a), (b) and (d). The Dirac points move as shown in Fig. 5. At $d_z/a \approx 1.166$
The interlayer distances are taken as 1 \( \text{nm} \). The black arrow indicate the Dirac point in the (2\( m \) + 1)th and (2\( m \) + 2)th bands from the bottom, respectively. The interlayer distances are taken as 1.165a [(a) ab (b)], 1.166a [(c)], and 1.167a [(d)]. Red arrows indicate the Dirac point in the (2\( n \) - 1)th and (2\( n \) - 2)th bands, and black arrow indicate the Dirac point in the (2\( m \) - 1)th and (2\( m \) - 2)th bands.

![FIG. 7. (color online) Energy dispersion in the commensurately rotated bilayer graphene with \( (m_1, m_2) = (9, 8) \). The black, blue, orange, and red lines are the (2\( n \) - 1)th, (2\( n \) + 1)th, and (2\( n \) + 2)th bands from the bottom, respectively. The interlayer distances are taken as 1.165a [(a) ab (b)], 1.166a [(c)], and 1.167a [(d)]. Red arrows indicate the Dirac point in the (2\( m \) + 1)th and (2\( m \) + 2)th bands, and black arrow indicate the Dirac point in the (2\( m \) - 1)th and (2\( m \) - 2)th bands.](image)

![FIG. 8. (color online) The moving Dirac points for the commensurately rotated bilayer graphene with \( (m_1, m_2) = (9, 8) \) as a function of interlayer distance. Four Dirac points merge at K point at \( d_z/a \approx 1.1660 \). Two Dirac points merge and annihilate at M point at \( d_z/a \approx 1.1707 \).](image)

The positions of sign-change and the cusp of velocity are exchanged in \( (m_1, m_2) = (9, 8) \) (Fig. 7(a)) and \( (m_1, m_2) = (10, 9) \) (Fig. 7(b)). The velocity of the (2\( n \) + 1)th band at K point is negative in the narrow region near \( d_z/a \approx 1.185 \) with V-shaped cusp when \( (m_1, m_2) = (10, 9) \), as seen in Fig. 7(a) and Fig. 5(b), where we take \( \delta_k = 0.01 \). The velocity changes sign and become negative at \( d_z/a \approx 1.147 \). The value of the sign-change point \( d_z/a \approx 1.147 \) is not intrinsic, as in the case of \( (m_1, m_2) = (9, 8) \). The V-shaped cusp also depends on \( \delta_k \), as shown in Fig. 5(c). We consider the V-shaped cusp near \( d_z/a \approx 1.185 \). We plot the energy dispersion of the bands near half-filling at \( d_z/a = 1.18 \) and 1.185 for \( (m_1, m_2) = (10, 9) \) in Fig. 8. When we look closer near K point, the small gap between 2\( n \)th and (2\( n \) + 1)th bands is seen and the Dirac point separates into two Dirac points. Therefore, if we take \( \delta_k \) small enough, the velocity of the (2\( n \) + 1)th band at K is negative, although it is positive at \( d_z/a = 1.18 \) with \( \delta_k = 0.01 \) (see Fig. 5(b)). There are other Dirac points beside K (near 0.92K in Fig. 5(b)), and they move to M via K as \( d_z/a \) increases from 1.180 to 1.185. This situation is the same as happened near \( d_z/a = 1.66 \) in \( (m_1, m_2) = (9, 8) \).
Therefore, if we take $\delta_k$ small enough, we expect the reversed V-shaped cusp in the $d_z$-dependence of the velocity of $(2n_0 + 1)$th band at K in the narrow region around $d_z/a \approx 1.185$, where the velocity calculated with $\delta_k = 0.01$ is negative. At the top of the reversed V-shaped cusp, four Dirac points merge and the velocity is zero.

When the rotating angle $\alpha$ is smaller than $3.481^\circ$ ($m_1 > 10$, $m_2 = m_1 - 1$), we have to take $\delta_k$ much smaller to obtain the negative region of $v$ and reversed V-shaped cusp in $d_z$-dependence of $v$. If we take $\delta_k$ not small enough, we obtain the V-shaped cusps as in Fig. 4 (a).

C. $(m_1, m_2) = (2, 1)$

In this subsection we discuss the twisted bilayer graphene the with $(m_1, m_2) = (2, 1)$ ($\alpha = 21.787^\circ$). The top of the reversed V-shaped cusp obtained by $\delta_k = 0.01$ is positive as seen in Fig. 4 (b). We show that the top of the cusp is zero as we take $\delta_k$ smaller.

In Fig. 10 we plot the energy dispersion for $(m_1, m_2) = (2, 1)$ near half filling at the interlayer distance $d_z/a = 1.0385$ and 1.0395, when the velocity of the $(2n_0 + 1)$th band is small. Although $v$ at $d_z/a = 1.0385$ obtained with $\delta_k = 0.01$ is positive, it should be negative if we take $\delta_k \lesssim 0.001$, as seen in Fig. 10 (b). There exist the moving Dirac points in th K-M line (three moving Dirac points by symmetry), and four Dirac points meet at K when $d_z/a$ is changed. Therefore, the positive value at the top of the reversed V-shaped cusp around $d_z/a \approx 1.039$ becomes zero when we take smaller $\delta_k$.

The moving Dirac point in the case of $(m_1, m_2) = (2, 1)$ moves as $M \rightarrow K \rightarrow \Gamma$, as $d_z/a$ increases around the reversed V-shaped cusp. This motion is reversal to those in the cases of $(m_1, m_2) = (9, 8)$ and $(10, 9)$. Another curious feature in $(m_1, m_2) = (2, 1)$ case is that the velocity of the $2n_0$th band at K does not become small when that of the $(2n_0 + 1)$th band is small, as seen in Fig. 10 (a) and (b). Since the particle-hole symmetry is broken in the commensurately rotated bilayer graphene, this asymmetry may happen, but the real reason is not clear. Despite these curious things, the first magic angle defined by the zero velocity in the $(2n_0 + 1)$th band seems to be a continuous function as a interlayer distance (see Fig. 5).

IV. DISCUSSIONS AND CONCLUSION

In this paper we define the magic angle as the twisted angle at which the velocity of the band just above the half-filling is zero. We showed that the magic angle in the commensurately-twisted bilayer graphene exists as a function of the interlayer distance even when the energy gap at K point is not negligible. The mechanism of causing the magic angle in that case is shown to be the merging of the four Dirac points at K point.
In the continuous approximation, the energy gap at K point is neglected. The energy gap is, however, finite at K point and the degeneracy of the two bands near K point is lifted, if we take into account the coupling between K and K’ points, which is in general finite in the tight binding model. In the tight binding model, the energy gap at K point is very small and can be safely neglected, if the twist angle is small. We obtain the interlayer-distance dependence of the magic angle in the commensurately-twisted bilayer graphene with the 4n_0 cites in the unit cell numerically.

The velocity of the (2n_0 + 1)th band at K as a function of interlayer distance becomes zero as a V-shaped cusp when m_1 ≥ 10 and m_2 = m_1 - 1 (type I magic angle), while it becomes zero as a revered V-shaped cusp when m_1 ≤ 9 and m_2 = m_1 - 1 (type II magic angle), when we calculate the velocity by the numerical differentiation, with taking the limit of δ_k → 0. By studying the cases with (m_1, m_2) = (9, 8) and (10, 9) in detail, we obtain that the magic angle is caused by the merging of four Dirac point at K. At the magic angle one Dirac point with topological number ±1 and three moving Dirac points with topological number ±1 merge at K and K’ points, resulting the topological number ±2. The topological number ±2 means that two band touch quadratically at K and K’ points, resulting the zero velocity.

The crossover from type I magic angle to type II magic angle, however, depends on the parameter for the numerical differentiation, δ_k, and it is not intrinsic. This mechanism of the zero velocity at K point is shown to work in the cases of moderate energy gap between 2n_0th and (2n_0 + 1)th bands at large twist angle (or small interlayer distance). The similar mechanism may work even in the cases of moderate energy gap between 2n_0th and (2n_0 + 1)th bands at large twist angle (or small interlayer distance). The similar mechanism may work even when a rotating angles becomes small. At small rotating angles, the gap between 2n_0th and (2n_0 + 1)th bands is exponentially small, and it can be safely neglected. In that case it is difficult to distinguish type I and type II magic angles. If there is a finite gap, the velocity at K is negative when δ_k → 0 limit is taken. In that sense the magic angle may be always type II.

In this paper we have not concerned the flatness of the band at the magic angle, and we only study the velocity at K point. Indeed, the band is not flat at the magic angle when the interlayer distance is small and the magic angle is not small. However, the zero velocity at K point is the necessary condition for the flat band for small magic angles. Studying the mechanism of magic angles at moderate twist angle and moderate interlayer distance is important to understand the mysterious physics in the twisted bilayer graphene deeper.

FIG. 11. (color online). Commensurately twisted bilayer graphene with (m_1, m_2) = (3, 2). Large (small) open and filled circles are A and B sublattice in the first (second) layer, respectively. The second layer is rotated by π/3 - α. The orange area is the unit cell of the commensurately twisted bilayer graphene.

Appendix A: commensurately twisted bilayer graphene

We take the primitive lattice vectors of the first layer, a_1^{(1)} and a_2^{(1)}, as

\[ a_1^{(1)} = a \left( \frac{\sqrt{3}}{2}, \frac{\sqrt{3}}{2} \right), \]

\[ a_2^{(1)} = a \left( \frac{\sqrt{3}}{2}, \frac{1}{2} \right), \]

where a is the lattice constant a = 0.246nm. The angle between two primitive lattice vectors are π/3;

\[ a_2^{(1)} = R_\theta a_1^{(1)}, \]

where the 2D rotational matrix R_θ is given by

\[ R_\theta = \begin{pmatrix} \cos \theta & -\sin \theta \\ \sin \theta & \cos \theta \end{pmatrix}. \]

Commensurately twisted bilayer graphene is labeled by two integers m_1 and m_2. The primitive lattice vectors L_1 and L_2 of the commensurately twisted bilayer graphene is given by

\[ L_1 = m_1 a_1^{(1)} + m_2 a_2^{(1)}, \]

\[ L_2 = R_\theta L_1^{(1)} = -m_2 a_1^{(1)} + (m_1 + m_2) a_2^{(1)}. \]

The number of sites in the unit cell of the commensurately rotated bilayer graphene is

\[ 4n_0 = 4(m_1^2 + m_1 m_2 + m_2^2). \]
in the supercell, i.e. $n_0$ A sites and B sites in each layer.

The A sites are located at the positions
\[ m_1 a_1^{(1)} + m_2 a_2^{(1)} \]  \hspace{1cm} (A8)
with integer $m_1$ and $m_2$. We define the angle $\alpha$ by the angle between the vectors
\[ \mathbf{L}_1 = m_1 a_1^{(1)} + m_2 a_2^{(1)}, \]  \hspace{1cm} (A9)
and
\[ \mathbf{L}_1' = m_2 a_1^{(1)} + m_1 a_2^{(1)}. \]  \hspace{1cm} (A10)
Two vectors have the same length
\[ |\mathbf{L}_1|^2 = |\mathbf{L}_1'|^2 = a^2 (m_1^2 + m_1 m_2 + m_2^2) = a^2 n_0. \]  \hspace{1cm} (A11)
By using
\[ \mathbf{L}_1 \cdot \mathbf{L}_1' = \frac{a^2}{2} (m_1^2 + 4m_1 m_2 + m_2^2), \]  \hspace{1cm} (A12)
we obtain
\[ \cos \alpha = \frac{m_1^2 + 4m_1 m_2 + m_2^2}{2(m_1^2 + m_1 m_2 + m_2^2)}, \]  \hspace{1cm} (A13)
and
\[ \sin \alpha = \frac{\sqrt{3}(m_1^2 - m_2^2)}{2(m_1^2 + m_1 m_2 + m_2^2)}, \]  \hspace{1cm} (A14)

We can obtain the commensurate twisted bilayer graphene by rotating the second layer either $\alpha$ or $\pi/3 - \alpha$. In order to obtain the Bernal stacking (AB stacking) in the limit $\alpha \to 0$, we study the $\pi/3 - \alpha$ rotation in the AA stacked bilayer graphene ($\alpha$ rotation in the Bernal stacked bilayer graphene) in this paper. Note that
\[ \cos \left( \frac{\pi}{3} - \alpha \right) = \frac{2m_1^2 + 2m_1 m_2 - m_2^2}{2(m_1^2 + m_1 m_2 + m_2^2)}, \]  \hspace{1cm} (A15)
\[ \sin \left( \frac{\pi}{3} - \alpha \right) = \frac{\sqrt{3}m_2 (m_1 + m_2)}{2(m_1^2 + m_1 m_2 + m_2^2)}. \]  \hspace{1cm} (A16)

The primitive lattice vectors of the second layer are given by rotating $\pi/3 - \alpha$:
\[ \mathbf{a}_1^{(2)} = R_{\mathbf{z} \rightarrow -\mathbf{z}} \mathbf{a}_1^{(1)} = \frac{a}{2(m_1^2 + m_1 m_2 + m_2^2)} \left( \sqrt{3}m_1 (m_1 + 2m_2) \right), \]  \hspace{1cm} (A17)
\[ \mathbf{a}_2^{(2)} = R_{\mathbf{z} \rightarrow -\mathbf{z}} \mathbf{a}_2^{(1)} = \frac{a}{2(m_1^2 + m_1 m_2 + m_2^2)} \left( \sqrt{3}m_2 (m_1 + m_2) \right). \]  \hspace{1cm} (A18)

The reciprocal lattice vectors for the first layer ($\mathbf{G}_1^{(1)}$ and $\mathbf{G}_2^{(1)}$) and the second layer ($\mathbf{G}_1^{(2)}$ and $\mathbf{G}_2^{(2)}$) are the vectors in the wave-number space and satisfy the relations,
\[ \mathbf{a}_i^{(\ell)} \cdot \mathbf{G}_j^{(\ell)} = 2\pi \delta_{i,j}, \]  \hspace{1cm} (A19)
and
\[ \mathbf{z} \cdot \mathbf{G}_i^{(\ell)} = 0, \]  \hspace{1cm} (A20)
where $i, j, \ell = 1$ or 2 and $\mathbf{z}$ is the unit vector along the $z$ direction. They are given by
\[ \mathbf{G}_1^{(\ell)} = 2\pi \frac{\mathbf{a}_2^{(\ell)} \times \mathbf{z}}{|\mathbf{a}_1^{(\ell)} \times \mathbf{a}_2^{(\ell)}|} \cdot \mathbf{z}, \]  \hspace{1cm} (A21)
\[ \mathbf{G}_2^{(\ell)} = 2\pi \frac{\mathbf{z} \times \mathbf{a}_1^{(\ell)}}{|\mathbf{a}_1^{(\ell)} \times \mathbf{a}_2^{(\ell)}|} \cdot \mathbf{z}. \]  \hspace{1cm} (A22)
We obtain
\[ \mathbf{G}_1^{(1)} = \frac{2\pi}{a} \left( \frac{\sqrt{3}}{-1} \right) = \frac{4\pi}{3a} (2\mathbf{a}_1 - \mathbf{a}_2), \]  \hspace{1cm} (A23)
\[ \mathbf{G}_2^{(1)} = \frac{2\pi}{a} \left( \frac{\sqrt{3}}{1} \right) = \frac{4\pi}{3a} (-\mathbf{a}_1 + 2\mathbf{a}_2). \]  \hspace{1cm} (A24)
The reciprocal Lattice vectors for the second layer, $\mathbf{G}_{1}^{(2)}$ and $\mathbf{G}_{2}^{(2)}$ are obtained by rotating $\pi/3 - \alpha$,

$$
\mathbf{G}_{1}^{(2)} = R_{z - \alpha} \mathbf{G}_{1}^{(1)} = 2\pi \frac{1}{a} \frac{1}{m_{1}^{2} + m_{1}m_{2} + m_{2}^{2}} \left( \frac{\sqrt{3}}{\sqrt{3}} (m_{1}^{2} + 4m_{1}m_{2} + m_{2}^{2}) \right) - \left( m_{1}^{2} - m_{2}^{2} \right),
$$

and

$$
\mathbf{G}_{2}^{(2)} = R_{z - \alpha} \mathbf{G}_{2}^{(1)} = 2\pi \frac{1}{a} \frac{1}{m_{1}^{2} + m_{1}m_{2} + m_{2}^{2}} \left( \frac{\sqrt{3}}{\sqrt{3}} (m_{1}^{2} - 2m_{1}m_{2} - 2m_{2}^{2}) \right) \frac{1}{m_{1}(m_{1} + 2m_{2})}.
$$

We write the reciprocal lattice vectors of the twisted bilayer graphene as $\mathbf{F}_{1}$ and $\mathbf{F}_{2}$, which are given by

$$
\mathbf{F}_{1} = 2\pi \frac{\mathbf{L}_{2} \times \hat{z}}{(\mathbf{L}_{1} \times \mathbf{L}_{2}) \cdot \hat{z}},
$$

$$
\mathbf{F}_{2} = 2\pi \frac{\hat{z} \times \mathbf{L}_{1}}{(\mathbf{L}_{1} \times \mathbf{L}_{2}) \cdot \hat{z}}.
$$

We obtain

$$
\mathbf{F}_{1} = \frac{1}{m_{1}^{2} + m_{1}m_{2} + m_{2}^{2}} \left( m_{1} + m_{2} \right) \mathbf{G}_{1}^{(1)} + m_{2} \mathbf{G}_{2}^{(1)}
$$

$$
= \frac{4\pi}{3a^{2}} \left( 2m_{1} + m_{2} \right) \mathbf{a}_{1} + \left( -m_{1} + m_{2} \right) \mathbf{a}_{2},
$$

and

$$
\mathbf{F}_{2} = \frac{1}{m_{1}^{2} + m_{1}m_{2} + m_{2}^{2}} \left( -m_{2} \mathbf{G}_{1}^{(1)} + m_{1} \mathbf{G}_{2}^{(1)} \right).$$

We can write Eqs. \ref{E29} and \ref{E31} as

$$
\mathbf{G}_{1}^{(1)} = m_{1} \mathbf{F}_{1} - m_{2} \mathbf{F}_{2},
$$

$$
\mathbf{G}_{2}^{(1)} = m_{2} \mathbf{F}_{1} + \left( m_{1} + m_{2} \right) \mathbf{F}_{2}.
$$

We plot Brillouin zone in the twisted bilayer graphene with $m_{1} = 3$ and $m_{2} = 2$ in Fig. [22].

[1] J. M. B. Lopes dos Santos, N. M. R. Peres, and A. H. Castro Neto, “Graphene bilayer with a twist: Electronic structure,” Phys. Rev. Lett. 99, 256802 (2007).
[2] Rafi Bistritzer and Allan H. MacDonald, “Moiré bands in twisted double-layer graphene,” Proceedings of the National Academy of Sciences 108, 12233-12237 (2011).
[3] E. Suárez Morell, J. D. Correa, P. Vargas, M. Pacheco, and Z. Barticevic, “Flat bands in slightly twisted bilayer graphene: Tight-binding calculations,” Phys. Rev. B 82, 121407 (2010).
[4] Hoi Chun Po, Lijun Zou, Ashvin Vishwanath, and T. Senthil, “Origin of mott insulating behavior and superconductivity in twisted bilayer graphene,” Phys. Rev. X 8, 031089 (2018).
[5] Kasra Hejazi, Chunxiao Liu, Hassan Shapourian, Xiao Chen, and Leon Balents, “Multiple topological transitions in twisted bilayer graphene near the first magic angle,” Phys. Rev. B 99, 035111 (2019).
[6] Grigory Tarnopolsky, Alex Jura Kruchkov, and Ashvin Vishwanath, “Origin of magic angles in twisted bilayer graphene,” Phys. Rev. Lett. 122, 106405 (2019).
[7] Yuan Cao, Valla Fatemi, Ahmet Demir, Shiang Fang, Spencer L. Tomarken, Jason Y. Luo, Javier D. Sanchez-Yamagishi, Kenji Watanabe, Takashi Taniguchi, Efthimios Kaxiras, Ray C. Ashoori, and Pablo Jarillo-Herrero, “Correlated insulator behaviour at half-filling in magic-angle graphene superlattices,” Nature 556, 80 (2018).
[8] Yuan Cao, Valla Fatemi, Shiang Fang, Kenji Watanabe, Takashi Taniguchi, Efthimios Kaxiras, and Pablo Jarillo-Herrero, “Unconventional superconductivity in magic-angle graphene superlattices,” Nature 556, 43 (2018).
[9] Matthew Yankowitz, Shaowen Chen, Hryhoriy Polshyn, Yuxuan Zhang, K. Watanabe, T. Taniguchi, David Graf, Andrea F. Young, and Cory R. Dean, “Tuning superconductivity in twisted bilayer graphene,” Science 363, 1059-1064 (2019).
[10] Yasumasa Hasegawa and Mahito Kohmoto, “Quantum Hall effect and the topological number in graphene,” Phys. Rev. B 74, 155415 (2006).
[11] Kenta Esaki, Masatoshi Sato, Mahito Kohmoto, and Bertrand I. Halperin, “Zero modes, energy gap, and edge states of anisotropic honeycomb lattice in a magnetic field,” Phys. Rev. B 80, 125405 (2009).
[12] Yasumasa Hasegawa and Keita Kishigi, “Merging dirac points and topological phase transitions in the tight-binding model on the generalized honeycomb lattice,” Phys. Rev. B 86, 165430 (2012).
[13] Pilkyung Moon and Mikito Koshino, “Energy spectrum and quantum Hall effect in twisted bilayer graphene,” Phys. Rev. B 85, 195458 (2012).
[14] Yasumasa Hasegawa and Mahito Kohmoto, “Periodic landau gauge and quantum Hall effect in twisted bilayer graphene,” Phys. Rev. B 88, 125426 (2013).