Anomalous Bloch oscillation and electrical switching of edge magnetization in bilayer graphene nanoribbon

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Graphene features topological edge bands that connect the pair of Dirac points through either sectors of the 1D Brillouin zone depending on edge configurations (zigzag or bearded). Because of their flat dispersion, spontaneous edge magnetisation can arise from Coulomb interaction in graphene nanoribbons, which has caught remarkable interest. We find an anomalous Bloch oscillation in such edge bands, in which the flat dispersion freezes electron motion along the field direction, while the topological connection of the bands through the bulk leads to electron oscillation in the transverse direction between edges of different configurations on opposite sides/layers of a bilayer ribbon. Our Hubbard-model mean-field calculation shows that this phenomenon can be exploited for electrical switching of edge magnetisation configurations.

Introduction. The existence and behavior of edge states are always attractive in the study of solid physics due to their distinct properties in contrast to bulk states. For the monolayer graphene (MLG) that has zero-gap band structure where the conduction and valance bands touch at the Dirac points [1]6, edge states in MLG ribbon appear as flat bands at the Fermi level, connecting the bulk Dirac points through either sectors of the 1D Brillouin zone depending on edge configurations (zigzag or bearded) [1]2[7][11]. When the bulk gap is opened, these flat-band edge states can be continuously tuned into gapless chiral edge modes through bias control on the edge [7], which have similar origin to the topological domain wall modes in bilayer graphene (BLG) [12][15][16]. With the non-trivial topological properties and relation to the bulk valley transport [7][13], these chiral modes have also been explored in other context such as the laser-induced Floquet system [14] and gapped nanomechanical graphene [17].

The flat edge band, on the other hand, promises the emergence of magnetism when electron interaction is taken into account [18][21]. In zigzag MLG nano-ribbons, the repulsive on-site Coulomb interaction is shown to introduce ground state spin polarization (SP) on the edges, which can be either antiferromagnetic (AFM) or ferromagnetic (FM), i.e., the localized magnetic moments at the opposite edges of the ribbon are antiparallel or parallel [19][21], turning the system into a semiconductor or a conductor (metal), respectively [18][19]. Based on this phenomenon, some interesting applications in spintronics have also been explored in the non-standard-shaped MLG ribbons [21][25]. Compared with MLG ribbon, magnetism in BLG materials and nanostructures is less studied. Most of earlier works focused on the half-metallicity and related magnetic effects in the zigzag BLG nanoribbons [26][29] or bulk BLG system [30]. In this letter, we focus on the motions of electrons correlated with the edge states in bea-zig BLG (top and bottom layer of a BLG ribbon have zigzag and bearded edges respectively, c.f. Fig. [1][c]), which host flat-dispersion edge bands in the entire 1D Brillouin zone [31]. Under interlayer bias, we find that gapless chiral modes appear in the ribbon bulk near the Dirac point, connecting states localised on opposite edges and layers. Bloch oscillation in the edge bands driven by electric field along the ribbon has an unusual form in the real space, where the electrons predominantly oscillate in the transverse direction between opposite edges and layers. Such phenomenon represents an interesting aspect of Bloch oscillation in momentum space, reflected in real space due to the spatial character of topological edge bands. With Hubbard interaction included through a mean-field approach, we show this anomalous Bloch oscillation can be exploited for electric field control of transition between different edge magnetic states of the BLG nano-ribbon, which points to a new possibility of spintronic control.

Anomalous Bloch Oscillation The electronic properties of the biased bea-zig BLG can be described by the tight-binding Hamiltonian [31]

\[ H_{BLG} = H_{bea}^1 + H_{zig}^2 + H_{int} + H_{bias}, \]

where

\[ H_{bea/zig}^l = -t \sum_{\langle i,j \rangle, \sigma} c_{l,i,\sigma}^\dagger c_{j,\sigma} + H.c. \]

represents the tight-binding Hamiltonian of MLG with bearded/zigzag edges and \( l = 1, 2 \) are labels of the bottom and top layers respectively. \( t \) denotes the nearest-neighbor (NN) hopping in MLG with \( c_{i,\sigma}^\dagger c_{i,\sigma} \) being the creation (annihilation) operator of \( \sigma \)-spin electron on site \( i \) in the ribbon, and \( \sum_{\langle i,j \rangle, \sigma} \) only sums over NN pairs. Since there is no Hubbard interaction, i.e., no interaction between different spins, the index \( \sigma \) can be ignored.

\[ H_{bias} = \sum_{l,i,\sigma} (-1)^l \frac{U_l}{2} c_{l,i,\sigma}^\dagger c_{l,i,\sigma} + H.c. \]
When \( U_1 = 0 \), topological edge states still exist in the whole \( k_x \) region when crossing the Dirac point. They are connected by a pair of bulk chiral modes, as shown in Fig. 1(c). The degeneracy between edges are lifted due to the broken chiral symmetry, as discussed in [31]. The bulk chiral modes connect states residing in opposite edges of different layers. The connection must be done through the bulk, hence the name. The main difference between the bulk chiral mode we discuss here and those edge chiral modes discussed in previous literatures [7, 12, 14–16] is obvious: bulk chiral modes are obtained by imposing a bias on the bulk, and the wave function of the chiral modes are also distributed in the bulk. While edge chiral modes are obtained via tuning edge on-site energy, whose wave function is localized at the edge. The fact that these connecting chiral modes are bulk states are by themselves interesting in terms of transport. It is often the case the bulk part of the material is insulator so that all electrical transport are dependent on topologically protected edge states. However, in this system we discuss, transport is done by two special pairs of chiral bulk states, one at each valley. These special bulk states at the Fermi level are present only because that there are edge states populating different area of the momentum space, which can not be found in the bulk spectrum of the BLG as shown by the blue bands in Fig. 1(c).

An interesting application of these bulk chiral modes is Bloch oscillation in topological insulators [34, 39]. When applying an electric field \( eE_x = \frac{\partial A(t)}{\partial t} \) along the infinite direction (\( x \) direction in our assumption) of the ribbon, the motion of electrons in the edge states can be approximately described by semiclassical equations of a wave packet. It indicates that the wave vector of the electron will evolve according to \( \dot{k}_x = -eE_x/h \), as shown in Fig. 2(a). Because of the existence of the bulk chiral modes connecting two opposite edges of two different layers, the transition of electrons from left edge of one layer to the right edge of the other layer is possible, giving rise to a Hall-effect-like behaviour of electrons, as illustrated in Fig. 2(a).

The naive conjecture as above may be undermined by the smallness of the gap opened by the bias. Numerically, a bias as large as \( U_1 = 300\text{meV} \) can open a gap of around 5meV between two edge bands in the flat part. Landau-Zener tunneling may cause the electrons to transit between different edge bands, breaking down our conjecture based upon single band picture. However, such a transition is suppressed by the fact that there is no spatial overlap between the wavefunctions of two flat edge bands, i.e. \( \left< \text{lower} | H_{E_x} | \text{upper} \right> \approx 0 \) [40]. Here, we use a wave packet to simulate the motion of electrons in real space. It shows the expected trajectory, which is shown in Fig. 2(b). Notice that in a full cycle, i.e., the momentum \( k_x \) evolves from 0 to \( 2\pi \), a wave packet will move both in \( x \) direction and in \( y \) direction. However, the motion in these two directions are different. Since only the part of edge bands near the Dirac points (bulk chiral modes) has non-vanishing group velocity and the time of wave packet staying in this region is inversely proportional to the field strength, the range of motion of the wave packet in the \( x \) direction is inversely proportional to electric field strength \( E_x \), which can be observed in

FIG. 1. (Color online) The schematic illustration of the atomic structure of biased bea-zig BLG ribbons and related bulk chiral modes. The total number of unit cell in finite direction of the ribbon is chosen as \( N = 60 \), i.e., 120 atoms for each layer for all plots. (a) The atomic structure of biased bea-zig BLG ribbons. The unit vectors are denoted by \( \vec{a}_1 \) and \( \vec{a}_2 \). Ribbons are assumed to be infinite along \( x \). (b) The band structure for the bea-zig BLG ribbon with no bias. Two bands of edge states are highlighted by the solid red lines. The zoomed-in band structure in the region when \( \gamma_1 = 0 \) [40]. Here, we use a wave packet to simulate the motion of electrons in real space. It shows the expected trajectory, which is shown in Fig. 2(b). Notice that in a full cycle, i.e., the momentum \( k_x \) evolves from 0 to \( 2\pi \), a wave packet will move both in \( x \) direction and in \( y \) direction. However, the motion in these two directions are different. Since only the part of edge bands near the Dirac points (bulk chiral modes) has non-vanishing group velocity and the time of wave packet staying in this region is inversely proportional to the field strength, the range of motion of the wave packet in the \( x \) direction is inversely proportional to electric field strength \( E_x \), which can be observed in
The Hubbard interaction $H_{\text{BLG}} + H_U, H_U = U \sum_i n_{i,\uparrow} n_{i,\downarrow}$. (4)

Here, since the interaction between different spins is no longer zero, the spin degrees of freedom $\sigma$ should be considered. We show the band structure of $H$ for both spins in Fig. 1(a). The calculating detail and a brief review of the model can be found in the Supplementary 12. Similar bulk chiral modes connecting edge modes appear as those arising in the non-hubbard spinless bea-zig BLG ribbon, with opposite chirality for two spins, as shown in Fig. 1(a). The SP configurations are not limited to two simple types (AFM/FM) as found in the MLG ribbon in Ref. 13, there are 8 inequivalent types of
FIG. 3. (Color online) (a) Band structure of two self-consistent ground states of bea-zig BLG ribbon in Hubbard model, corresponding to Conf. 4 in the Table. Black dashed line is the Fermi level. Black solid lines are bulk bands. Blue (Red) solid lines are edge bands of spin up (down) electrons. (b) Schematic illustration of the Bloch oscillation in the same system. (2) (4) corresponds to the top (bottom) panel of (a). (1) (3) corresponds to the situation where the deviation from the initial state (top panel of (a)) is small (large). The black circle are electrons occupying bands. (c) Top panel: SP configurations corresponding to (a) (top (bottom) panel of (a) corresponds to left (right), respectively), which is defined by the magnetic moment $M$. The blue (orange) solid line represents situation on the top (bottom) layer. The horizontal axis is the site index, increasing along the finite direction of the ribbon. The top layer atoms constitute the first half while the bottom layer atoms constitute the second half. Bottom panel: The schematic illustration of the SP distribution in real space, corresponding to SP configurations shown in the top panel. A transition between these two configurations can be induced by the process illustrated in (b).

SP for BLG cases, as shown in the Table. Here we only demonstrate two types of band structures corresponding to configuration 4 in Table for simplicity in Fig. 4(a).

The essence of the result in the spinless model is that an external field $E_x$ can push electrons through the momentum space, thus causing corresponding motion in the real space. In a spinless model, the occupation of the electrons in the momentum space will not affect the band structure, while the band structure of the Hubbard model is dependent on the momentum space distribution of electrons. Thus, not all configurations in the momentum space is a self-consistent ground state at equilibrium. In fact, as we discussed in last section, there are only 16 (8 × 2) self-consistent solutions of the model. Any unstable configurations should relax to one of them. An electric field $E_x$ will push the system out of equilibrium by shifting the position of the electrons in momentum space. If the shift is small, it is reasonable to expect that the system should relax to the initial condition. However, if the shift is large enough in momentum space, it is possible that the system relax to another ground state when trying to reach equilibrium. The idea of this procedure is schematically illustrated in Fig. 3(b).

Numerically, this is verified by giving different initial

guess to the electron’s momentum space distribution, which would stably converge to different ground state, as illustrated in Fig. 3(a) and (b). Initially, spin up electrons occupy the region $k_x \in [-\frac{2\pi}{3}, \frac{2\pi}{3}]$ while spin down electrons occupy the region $k_x \in [-\pi, -\frac{2\pi}{3}] \cup [\frac{2\pi}{3}, \pi]$. If the occupation is pushed to the positive $k_x$ direction by a small distance, it will stably converge to the initial state, i.e. top panel of Fig. 3(a). If electrons are pushed away more from their initial state, it would stably converge to an equilibrium state that differs from its initial state by an exchange of spin, i.e. bottom panel of Fig. 3(a). The corresponding SP configuration is given in Fig. 3(c). This is an intra-configuration transition between ground states, i.e., Conf. 4 of Table.

Moreover, we found that the inter-configuration transition between ground states is possible by following the similar process illustrated in Fig. 3. This is shown in Fig. 4 where Conf. 3 in Table. can be transformed into Conf. 8 through the exchange of spins between the same edge of different layers. The switch of edge-magnetization only happens between two partially filled bands since there are always finite gaps (around 20meV) between these two bands and the other two bands, i.e., one empty edge band and one fully filled edge band, as shown in Fig. 4(a).
the process similar to that shown in Fig. 3(b). The difference (around 20 meV) between these two SP configurations can be induced based on two SP configurations shown in the top panel. A transition illustration of the SP distribution in real space, corresponding to two SP configurations shown in the top panel. A transition between these two SP configurations can be induced based on the process similar to that shown in Fig. 3(b).

Conclusions and discussions. In summary, we study the motions of electrons related with the edge states in bea-zig BLG with and without Hubbard interaction. The chiral modes present in the system without Hubbard interaction is no longer edge chiral states appearing in the spinless MLG ribbons but bulk states. These bulk chiral modes are unconventional, which connect two non-trivial topological phases and can be modulated by a mechanism adjusting property of the bulk instead of adjustments on the edge. When applying an electric field along the infinite direction of the ribbon, the motion of electrons in the edge states can be approximately described by Bloch oscillation based on semiclassical equations of wavepackets. This leads to an anomalous transverse oscillation since the bulk chiral modes connect opposite edges of different layers. For the same system with the Hubbard interaction, the Hall-effect-like behavior persists when the electric field is applied, protected by the spatial character of the topological edge bands. Besides, we also exploit the possibility of electrical switching of edge magnetisation in Hubbard model using this bulk mode. With recent progress in bottom-up approach, synthesis of long/narrow atomically precise graphene ribbon with well-defined edges has become possible. It makes the test of edge-induced magnetism, as well as various properties associated with edge topology, of graphene ribbon possible in the near future. Since all these dynamical effects are related with the bulk chiral states connecting edge states of the system, it is possible for us to generalize our study to other 2D materials with strong edge effect such as transition metal dichalcogenides [51, 52] and materials having Kagome [53] or triangular [55, 56] lattice structure. All of these are potential directions for further study.

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[40] One can also understand this fact from the well-known formula describing Landau-Zener tunneling [41]: $P = e^{-2e\Delta^2/\hbar}$. Where $P$ is the transition probability between two bands, $\Delta$ is approximately the gap between two bands, $\frac{1}{2} \tau$ is approximately the asymptotic slope of the band. Since $\tau$ is infinitely large for our topological flat edge bands, while $\Delta$ is finitely small, then the transition is infinitely suppressed. When near the Dirac point, two edge bands become dispersive and their wavefunctions have appreciable overlap. However, a gap around 30 meV is opened even for zero bias situation, as shown in Fig. 1, which makes the interband transition near the Dirac point still weak. This leads the Bloch oscillation in our system to distinguish from those discussed in the literature, such as Ref. [35] [38]. There the dispersive topological edge bands inevitably need to take Bloch-Zener transition into consideration. It is also distinct from the Bloch oscillation in bulk bilayer graphene. In that condition the bulk bands which are responsible for the Bloch oscillation are highly dispersive, facilitating the transition between nearby bands via Landau-Zener tunneling.

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Difference in the small/large deviation is the magnitude of the electric field. Here, the important time scale are the momentum relaxation time $\tau_p$ and the spin relaxation time $\tau_s$. In graphene nanoribbons with magnetism on the edge, the typical $\tau_p$ is of order of 1 ps [44], and $\tau_s$ can be the order of $10 \sim 100$ ps [45, 46]. If the field is too small and relaxation sets in before the deviation is large enough, it corresponds to condition (1) of Fig. 3(b). On the contrary, if the field is large enough to take electrons far away from the initial configuration in the momentum space within $\tau_p$, then it corresponds to condition (3) of Fig. 3(b), which makes the ground-state exchange, i.e., spin flip possible under the spin relaxation. Therefore, it is easy to give an estimated threshold electric field for the switching described in the text to appear: $E_{\text{threshold}} \sim \pi \hbar a \tau_p \sim 10^{-2} \text{V/\text{nm}}$, where $a = 0.246$ nm is the lattice constant of MLG.

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