Novel Magnetic Quantization of Bismuthene

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The generalized tight-binding model, being based on the spin-dependent sublattices, is developed to explore the magnetic quantization of monolayer bismuthene. The $sp^3$ orbital hybridizations, site energies, nearest and next-nearest hopping integrals, spin-orbital interactions and magnetic field ($B_z\hat{z}$) are taken into account simultaneously. There exist three groups of low-lying Landau levels (LLs), in which they are mainly from the $(6p_x, 6p_y, 6p_z)$ orbitals, and only the first group belongs to the unoccupied conduction states. Furthermore, each group is further split into the spin-up- and spin-down-dominated subgroups. The six subgroups present the rich and unique $B_z$-dependent LL energy spectra, covering the specific or arc-shaped $B_z$-dependences, the normal/irregular spin-split energies, and the non-crossing/crossing/anti-crossing behaviors. Specially, the second group of valence LLs near the Fermi level can create the frequent inter-subgroup LL anti-crossings since the main and side modes are comparable. The main features of energy spectra can create the special structures in density of states.

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The monoelemental 2D materials have stirred a lot of experimental and theoretical researches since the first discovery of graphene in 2004 by the mechanical exfoliation [1]. They are very suitable for studying the diverse physical, chemical and material properties. Up to now, the successfully synthesized group-IV and group-V systems cover few-layer/multilayer graphene [1–3], silicene [4, 5], germanene [6, 7], tinene [8], phosphorene [9, 10], antimonene [11, 12], and bismuthene [13–18]. Specifically, layered bismuthene are epitaxially grown on the 3D Bi$_2$Te$_3$(111)/Bi$_2$Se$_3$(111)/Si(111) substrates [13–17]. Also they could be directly obtained from the mechanical exfoliation [18]. There are various Hamiltonians, being sensitive to the planar/buckled structures, lattice symmetries, layer numbers, stacking configurations, single- or multi-orbital hybridizations, site energies, spin-orbital couplings (SOCs), and external electric and magnetic fields. How to solve new Hamiltonians is one of the main-stream topics in physics science. This work is focused on the unique quantization of monolayer bismuthene in a uniform perpendicular magnetic field ($B_z \hat{z}$) by using the generalized tight-binding model.

For few-layer bismuthene, there are some studies on geometric structures [15, 19–22], electronic structures [19, 22] and transport properties [18]. Bi atoms could form 2D honeycomb lattice with a highly buckled structure, as clearly identified from the measurements of scanning tunneling microscopy and high-resolution electron diffraction [15, 16, 21]. The angle-resolved photoemission spectroscopy has confirmed the unusual low-lying energy bands with the cone-like, parabolic, and sombrero-shaped dispersions centered at the Γ point [13, 17]. The similar band structures are revealed in the theoretical calculations using the first-principles method [19, 22] and the tight-binding model [20]. They are deduced to be dominated by the geometric structure, the multi-orbital chemical bondings,
the distinct site energies and the strong SOC. Such critical factors are responsible for a 3D rhombohedral Bi semimetal [23, 24]. The combination with a magnetic field is expected to create the diversified Landau levels (LLs) in terms of the $B_z$-dependent energy spectra and the quantum oscillation modes of the spatial probability distributions. This is worthy of a systematic investigation.

In general, there are two theoretical methods in studying the magneto-electronic properties. The effective-mass model is to make a perturbation expansion about the high-symmetry point and then do the magnetic quantization. It is suitable and reliable for layered systems with simple and monotonous band structures, such as, the rich $B_z$-dependent LL energy spectra in monolayer graphene [25, 26], silicene, germanene [27], MoS$_2$ [28], and few-layer AA- and AB-stacked graphenes [29, 31]. This method will become cumbersome for condensed-matter systems in the presence of complicated interactions and composite/non-uniform external fields. As for the generalized tight-binding model, all the significant interactions and the various external fields are included in the calculations simultaneously. For example, it has predicted three kinds of LLs in sliding bilayer graphene with various stacking configurations [32], and the unusual features of LLs in ABC- and AAB-stacked graphene [33, 34]. On the experimental side, scanning tunneling spectroscopy (STS) is the most powerful instrument in measuring the magneto-electronic energy spectra, since the differential tunneling conductance is roughly proportional to density of states (DOS). Up to now, STS measurements are successful in identifying the magnetically quantized energies in AB-stacked graphite [35, 36] and few-layer AB-stacked graphene [37, 38].

We utilize the generalized tight-binding model to investigate magneto-electronic properties of monolayer bismuthene. The magnetic Hamiltonian matrix elements, being related
to the sp\(^3\) bondings & site energies, SOC and \(B_z \hat{z}\), are calculated from the tight-binding functions in an enlarged unit cell due to vector potential. The orbital-, spin- and sublattice-decomposed wave functions are delicately evaluated to characterize the dominating oscillation modes and determine the quantum numbers of LLs. The dependences of the LL energies and wave functions on the field strength are explored in detail, especially for the neighboring LL energy spacings, the spin-split energies, and the non-crossing/crossing/anti-crossing behaviors. The LL anti-crossings will be examined from the probability transfer between the main and side modes, as well as the change of spin configurations. The van Hove singularities in creating the special structures of DOS are also discussed.

Bismuthene is composed of buckled hexagonal lattices in which two equivalent A and B sublattices are, respectively, located at two parallel planes with a separation of \(\Delta_z = 1.81\) Å. The primitive unit vectors are indicated by \(\mathbf{a}_1\) and \(\mathbf{a}_2\) with a lattice constant of \(a = 4.33\) Å (Fig. 1(a)), and the buckled structure is associated with the angle between the Bi-Bi bond and the z-axis, \(\theta = 126^\circ\) (Fig. 1(b)). The generalized tight-binding model \([39, 40]\) is utilized to explore the electronic properties under external fields, in which all the critical interactions are taken into account simultaneously. The strong sp\(^3\) orbital hybridizations, the distinct site energies and the significant SOC will dominate the essential properties near the Fermi level. In the bases of \(\{ |6p^A_z\rangle, |6p^A_x\rangle, |6p^A_y\rangle, |6s^A\rangle, |6p^B_z\rangle, |6p^B_x\rangle, |6p^B_y\rangle, |6s^B\rangle \}\otimes \{ \uparrow, \downarrow \}\), the Hamiltonian is expressed as

\[
H = \sum_{i,o,m} E_o C^+_{iom} C_{iom} + \sum_{\langle i,j \rangle, o,o',m} \gamma_{oo'}^{Rij} C^+_{iom} C_{jo'o'm} + \sum_{\langle\langle i,j \rangle\rangle, o,o',m} \gamma_{oo'}^{Rij} C^+_{iom} C_{jo'o'm} \\
+ \sum_{i,p,\alpha,p\beta,m,m'} \frac{\lambda_{\text{SOC}}}{2} C^+_{ip\alpha m} C_{ip\beta m'} (-i\epsilon_{\alpha\beta\gamma} \sigma^\gamma_{mm'}),
\]

(1)
where $C^+_i(C_{i om})$, $i$, $o$, and $m$ stand for the creation (annihilation) operator, lattice site, atomic orbital, and spin, respectively. The first term is the site energy, and $E_o$ of the 6s and 6p orbitals are set to be $-9.643$ eV and $-0.263$ eV, respectively [21]. The second term is the nearest-neighbor hopping integral ($\gamma^{R_{ij}}_{oo'}$) which depends on the type of atomic orbitals, the translation vector of the nearest-neighbor atom ($R_{ij}$), and $\theta$. The various interactions are characterized by the $sp^3$ chemical bondings: $V_{pp\pi} = -0.679$ eV, $V_{pp\sigma} = 2.271$ eV, $V_{sp\sigma} = 1.3$ eV and $V_{ss\sigma} = -0.703$ eV [21], as clearly indicated in Fig. 1(c). For the next-nearest-neighbor atoms, the hopping integral in the third term is independent of $\theta$, since such atoms are located at the same plane. It is related to $V_{pp\pi} = 0.004$ eV, $V_{pp\sigma} = 0.303$ eV, $V_{sp\sigma} = 0.065$ eV and $V_{ss\sigma} = -0.007$ eV. The last term represents the intra-atomic SOC $V\text{soc} = \lambda\text{soc} \hat{L} \cdot \hat{s}$ with $\lambda\text{soc} = 1.5$ eV. $\alpha, \beta$ and $\gamma$ denote the $x$, $y$ or $z$ direction, and $\sigma$ is the Pauli spin matrix. $V\text{soc}$ can also be expressed as $V\text{soc} = \lambda\text{soc} (\frac{L_+ s_\pi + L_- s_{\pi'}}{2} + L_z s_z)$, where $L_\pm$ and $s_\pm$ are the ladder operators for the angular momentum and spin. The SOC is vanishing for the same orbital. It could induce the change of spin configurations between the 6p$_z$ and (6p$_x$,6p$_y$) orbitals & the 6p$_x$ and 6p$_y$ orbitals.

The periodical Peierls phases, being created by a uniform perpendicular magnetic field, can modulate the hopping integral as $\gamma^{R_{ij}}_{oo'}(B_z) = \gamma^{R_{ij}}_{oo'} \exp \left(i\frac{2\pi}{\Phi_0} \int_{r_j} A(r) \cdot d\mathbf{r}\right)$ and induce an enlarged unit cell. $\Phi_0 (hc/e)$ is the flux quantum. Under the Landau gauge $\mathbf{A} = (0, B_z x, 0)$, a rectangular unit cell covers $4R_B \times 4 \times 25500/B_z$ Bi atoms (Fig. 1(a)), where $R_B$ is the ratio of $\Phi_0$ versus magnetic flux through a hexagon. The area of a reduced Brillouin zone (a small rectangle in Fig. 1(d)) is $4\pi^2/\sqrt{3}a^2R_B$. The magnetic Hamiltonian is built from the space spanned by the $32R_B$ tight-binding functions $\{|A_{om}^i\}; |B_{om}^i\}$, where $i = 1, 2, ..., 2R_B$. By the detailed analytic calculations, this Hermitian matrix could be transferred into the
band-like form to solve LL energies and wave functions more efficiently. When a uniform
electric field is applied along the \( z \)-axis, it can create a Coulomb potential \( V_z/2 \) \((-V_z/2)\) on the site energy of the A (B) sublattice. The generalized tight-binding model could be
further developed to comprehend the magnetic quantization in other layered systems with
complex orbital bondings and spin configurations under composite fields.

Monolayer bismuthene exhibits a feature-rich electronic structure due to the signifi-
cant multi-orbital bondings, site energies and SOC. The low-lying electronic properties are
mainly determined by three energy bands near the \( \Gamma \) point (Fig. 1(d)), as clearly shown
in Fig. 2. Each electronic state is doubly degenerate for the spin degree of freedom. The
first conduction \( (c_1) \) has parabolic energy dispersion centered at the \( \Gamma \) point. The second
valence band presents the valley-like dispersion except a slightly rounded structure near
the \( \Gamma \) point. Specially, the first valence band \( (v_1) \) reveals the sombrero-shaped structure
with the non-monotonic wavevector dependence, in which the extreme points deviate from
the \( \Gamma \) point. There coexist two constant-energy loops within a certain energy range, being
expected to induce the complicated magnetic quantization under their strong competitions.
Apparently, the lowest unoccupied state in the \( c_1 \) band and the highest occupied states in
the \( v_1 \) band (the outer constant-energy loop) have the different wave vectors and thus lead
to an indirect energy gap of 0.293 eV.

Bismuthene exhibits three groups of low-lying LLs with distinct characteristics, as
clearly shown in Figs. 3 and 4. The valence and conduction LLs are asymmetric about the
Fermi level. The first, the second, and the third groups are associated with the magnetic
quantization of the \( c_1, v_1 \) and \( v_2 \) energy bands, respectively. Each LL group is further
split into two spin-dependent LL subgroups because the cooperation of SOC and magnetic
field destroys the spin degeneracy. For any \((k_x, k_y)\) states, these LLs are doubly degenerate owing to the mirror symmetry about the \(z\) axis. For example, at \((k_x = 0, k_y = 0)\), the two degenerate wavefunctions have equivalent spatial distributions, but are localized near the 0 and 1/2 positions of the enlarged unit cell, respectively. The 1/2-localization-center states are chosen to illustrate the main features of LL wavefunctions (Figs. 3(b) and 4(b)). Each LL state is characterized by the spatial probability density on the A and B sublattices with \(sp^3\) orbitals and two spin configurations. This distribution might have a normal zero-point number and present the symmetric/anti-symmetric mode about the localization center, as revealed in a harmonic oscillator (2D electron gas). As a result of the hexagonal lattice, the A and B sublattices possess the same oscillation mode after magnetic quantization.

For the first group, the 6p\(_x\)- and 6p\(_y\)-decomposed probability distributions present an identical oscillation mode in the A\(_↑\), B\(_↑\), A\(_↓\) and B\(_↓\) sublattices (red and green curves in Fig. 3(b)). Furthermore, they dominate the oscillation modes of the spin-split LLs, in which the number of zero points can serve as a quantum number \((n_{1↑↓})\). The \(n_{1↑}\) and \(n_{1↓}\) LLs, respectively, have the \(↑\)- and \(↓\)-dominated components. The splitting of the \(n_{1↑}\) and \(n_{1↓}\) subgroups are, respectively, shown in Fig. 3(a) by the solid and dashed blue lines. At \(B_z = 30\) T, the first three conduction LLs belong to the \(n_{1↑}\) LLs and then the \(n_{1↑} = n\) and \(n_{1↓} = n + 3\) LLs appear alternatively. The energy spacing between two neighboring LLs in the same subgroup is almost uniform. This directly reflects the magnetic quantization of the parabolic \(c_1\) band (Fig. 2), as observed in 2D electron gas. Furthermore, the spin-split energy between two different subgroups is about 37 meV regardless of the state energy. The third group is very different from the first group in terms of energy spacing, split energy; orbital and spin components. The LL spacing is non-uniform because of the valley-like
energy dispersion. The spin-split energy declines in the increase of state energy, as shown by the dashed and solid red lines in Fig. 3(a). The dominating orbitals cover 6p_z, 6p_x and 6p_y (black, red and green curves in the lower half part of Fig. 3(b)). They have the same quantum mode, while the former and the latter two present the opposite spin configurations. The 6p_z or 6p_x/6p_y component is suitable for serving as the dominating quantum mode \((n^3)\), and the latter is chosen to illustrate the diversified properties among three groups of LLs. For the \(n^3\) \([n^2]\) LLs, there are, respectively, \(n^3\) and \(n^3 + 1\) \([n^3 - 1\) and \(n^3]\) zero points in the 6p_x/6p_y-dependent \((A^\uparrow,B^\uparrow)\) and \((A^\downarrow,B^\downarrow)\) sublattices. The similar zero-point numbers are revealed in the \(n^2\) LLs (Fig. 4(b)).

Specially, the second group of LLs exhibits an abnormal ordering (Fig. 4(a)) and the highly asymmetric probability distributions (Fig. 4(b)). The quantum number \(n^2\) is determined from the 6p_x/6p_y-dependent oscillation mode even if its probability density is lower than that of the 6p_z orbital. The spatial probability density does not present the well-behaved symmetric distribution about the localization center except for the \(n^2 = 0\) and/or 1 LLs. This suggests the superposition of the main and side oscillation modes (the distinct normal modes) in each \(n^2\) LL. Apparently, the LL energy spacing and the spin-split energy do not have the specific relations with state energy, as shown by the dashed \((n^2)\) and solid \((n^2)\) black lines in Fig. 4(a). The unusual LL energy spectrum, being sensitive to the magnetic-field strength, is related to the sombrero-shaped energy dispersion (Fig. 2). At \(B_z = 30\) T, the \(\uparrow\)-dominated (\(\downarrow\)-dominated) LL energies have the ordering of \(E^\uparrow(n^2) > E\downarrow(n^2 - 1)\) for \(n^2 \leq 4\) \((n^2 \leq 5)\), and then the inverse ordering for others. This clearly reflects the small-\(n^2\) LLs arising from the inner valley centered at the \(\Gamma\) point (Fig. 2), the \(n^2\)-dependent energy ordering similar to the wave-vector dependence of energy.
band, and the strong competitions between the inner and outer constant-energy loops.

The low-lying LLs exhibit the rich and unique $B_z$-dependent energy spectra, as clearly illustrated in Figs. 5 and 6(a). The conduction LLs has no intra-subgroup and inter-subgroup crossings/anti-crossings (the dashed and solid blue curves in Fig. 5(a)). Each LL energy exhibits the linear $B_z$-dependence, in which the neighboring LL spacing and the spin-split energy are proportional to the field strength. These are directly reflected from the monotonous wave-vector dependence of a parabolic conduction band. The third- and second-group LLs coexist in the deeper valence energy spectrum (the red and black curves in Fig. 5(b)). They frequently cross each other because of the well-behaved spatial distributions without the same quantum mode. The former roughly have the $\sqrt{B_z}$-dependent energies, especially for the larger $n_{↑↓}^3$ or the stronger $B_z$. This is associated with the quasi-linear valence band.

The LL energy spectrum of the second group, as shown in Fig. 6(a), is in sharp contrast with those of the first and third groups (Figs. 5(a) and 5(b)). All the LLs present the arc-like $B_z$-dependence except that the $n_↑^2 = 0$ LL energy monotonously declines with the increasing $B_z$. Their energies agree with that of the $\Gamma$ point ($-0.222$ eV) in the sombrero-shaped band (Fig. 2(a)) when $B_z$ approaches zero. This clearly indicates that the magnetic quantization is initiated from electronic states near the $\Gamma$ point. For very large $n_{↑↓}^2$ (> 30), the LL energies grow quickly as $B_z$ slightly increases from zero. The $n_↑^2$ and $n_↓^2$ LLs reach the maximum energy ($-0.146$ eV), being nearly identical at $B_z \sim 1$ T. This energy corresponds to the highest level of the outer constant-energy loop. With the increase of $B_z$, the spin-dependent two subgroups start to separate, in which the $n_↑^2$ LLs exhibit more drastic changes. As a result, there exist very frequent inter-subgroup crossings.
and anti-crossings, depending on whether the neighboring $n_{↓}^2$ and $n_{↑}^2$ LLs have the same oscillation modes.

The anti-crossings between two spin-dominated subgroups deserve a closer examination. They mainly arise from the $n_{↑}^2 = n$ and $n_{↓}^2 = n + 4$ LLs, as illustrated by the red rectangles in Fig. 6(a). In addition to the major mode with $n$ zero points, these two LLs also possess the side modes with different zero-point numbers. The latter are due to the cooperation of the intrinsic interactions and the magnetic field. Such modes are examined to have $n \pm 3$ zero points by the detailed numerical calculations. For example, the $n_{↑}^2 = 6$ LL strongly anti-crosses with the $n_{↓}^2 = 2$ LL in the range of $60 \, T < B_z < 70 \, T$ (Fig. 6(b)). When $B_z$ increases from 50 T along the higher-energy path (the solid blue arrow), the $n = 6$ main mode in the $A^\uparrow/B^\downarrow$ sublattice declines quickly (Fig. 6(c)), and the $n = 3$ side mode in the same sublattice grows rapidly. Furthermore, the main and side modes, respectively, with 5 and 2 zero points in the $A^\uparrow/B^\downarrow$ sublattice behave similarly. These two modes are comparable near the critical magnetic field ($\sim 65 \, T$). Their roles are interchanged in the further increase of field strength. For example, at $B_z = 80 \, T$, the spatial distribution is dominated by the oscillation mode with 2 zero points in the $A^\uparrow$ and $B^\downarrow$ sublattices (the first row in Fig. 6(c)). That is, the $n_{↑}^2 = 6$ LL is changed into the $n_{↑}^2 = 2$ LL during the variation of $B_z$. The probability transfer between the spin-up and spin-down components is driven by the critical SOC. The similar anti-crossing process is revealed in the inverse transformation along the lower-energy path (the dashed blue arrow).

The van Hove singularities in the energy-wave-vector space can create the special structures in DOS, being sensitive to the effective dimensions. DOS is defined as

$$D(E) = \sum_{n_{↑↓}} \int_{1st \ B_z} \frac{\Gamma'}{[E_{c,v}(n_{↑↓}, k_x, k_y) - E]^2 + \Gamma'^2} dk_x dk_y. \quad (2)$$
Γ′ (=0.1 meV) in the calculations is the broadening parameter. At zero field, the 2D band structure exhibits three shoulder structures (the red dashed circles) and one strong asymmetric peak in the square-root form (the red solid circle), as shown in Figs. 7(a) and 7(b). The first, second and third shoulders situated at $E = 0.148$ eV, $-0.222$ eV and $-0.396$ eV are, respectively due to the band-edge states (the extreme points) in the parabolic conduction band, the rounded inner constant-energy loop centered at the Γ point, and the rounded valley-like valence band (Fig. 2(a)). The latter comes from the outer constant-energy loop with the highest level in the first valence band, since it could be regarded as a 1D parabolic band. Band gap is energy difference between the first shoulder of the conduction states (Fig. 7(a)) and the prominent asymmetric peak. Under the magnetic quantization, the delta-function-like peaks arising from the zero-dimensional LLs come to exist. Their intensities are proportional to the number of LLs. The first group, as shown by the black solid curves in Fig. 7(a) at $B_z = 30$ T, presents a lot of uniform symmetric peaks, being composed of the ↑- and ↓-dominated ones. This further illustrates the absence of crossing and anti-crossing. Specially, the initial three conduction peaks nearest to $E_F$ are associated with the $n_{1\downarrow} = 0 - 2$ LLs (the blue circles), indicating the specific energy spacing between two neighboring LLs. For the higher-energy peaks, they could be utilized to identify the separate contributions of the $n_{1\uparrow\downarrow}$ LLs. As to the valence LLs, they exhibit many double-peak structures and some single peaks (Fig. 7(b)). The former are induced by the frequent crossings and anti-crossings between the $n_{2\uparrow}$ and $n_{2\downarrow}$ LLs, and the similar crossings between the $n_{2\uparrow\downarrow}$ and $n_{3\uparrow\downarrow}$ LLs. The higher-energy valence peaks before the strong asymmetric peak are due to the $n_{2\downarrow}$ LLs. Their energies and numbers could be tuned by the magnetic field strength. The threshold peak energy will, respectively, approach to $-0.148$
eV and −0.222 eV (energies of the asymmetric peak and the second shoulder), when $B_z$ is reduced to 1 T and ~ 0 T.

The diverse magnetic quantization phenomena of monolayer bismuthene are explored in detail using the generalized tight-binding model. The main features of LLs are determined by the multi-orbital chemical bondings, the distinct site energies, the nearest and next-nearest hopping integrals, the significant spin-orbital couplings and the magnetic field. The theoretical model could be further developed to solve new Hamiltonians in emerging 2D materials under the uniform/non-uniform external fields, e.g., the magnetic Hamiltonians with various interactions in few-layer bismuthene, antimonene [11, 12], phosphorene [9, 10] and arsenene [20, 41, 42]. Moreover, the generalized tight-binding model could combine with the single- and many-particle theories to study the other essential physical properties, such as magneto-optical spectra [43], magnetoplasmons [44], and quantum transports [45].

The low-energy electronic structure covers the parabolic conduction band, the sombrero-shaped valence band (the inner and outer constant-energy loops) and the rounded valley-like valence band, with an indirect gap of 0.293 eV. Such energy bands are closely related to three groups of $(6p_x, 6p_y, 6p_z)$-created LLs, in which each group is split into the $\uparrow$- and $\downarrow$-dominated subgroups. The dominating oscillation mode of the $6p_x/6p_y$-projected probability distribution could provide a good quantum number. The first and third groups possess the well-behaved wave functions, while the second group might have the main and side modes in the $n_{\uparrow\downarrow}^2$ LLs. The former two do not present the anti-crossing behavior in the $B_z$-dependent energy spectra. However, the frequent anti-crossings occur between the $n_{\uparrow}^2 = n$ and $n_{\downarrow}^2 = n + 4$ LLs. The LL energies of the first, second and third groups, respectively, exhibit the linear, arc-shaped and square-root-form $B_z$-dependences. Furthermore,
the normal spin-split energies are only revealed in the first group. The van Hove singularities in parabolic bands, the outer constant-energy loop and LLs, respectively, lead to shoulders, a prominent asymmetric peak and many delta-function-like peaks. The predicted electronic energy spectra could be examined by STS measurements.

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Figure captions

Figure 1: (a) The $x - y$ projections of monolayer bismuthene with an enlarged rectangular unit cell in $B_z \hat{z}$, (b) the buckled structure, (c) the various orbital hybridizations; (d) the Brillouin zone of the hexagonal lattice with the high-symmetry points and the reduced first Brillouin zone. $a_1$ and $a_2$ in (a) are lattice vectors, and the subscript of $A_i$ corresponds to the $i$-th Bi atom.

Figure 2: Energy bands of bismuth along the high-symmetry points. Also shown are the 3D band structures near the $\Gamma$ point.

Figure 3: (a) The spin-dependent LL energies for the first and the third groups at $B_z = 30$ T; (b) the orbital-projected probability distributions in the $A^\uparrow$, $B^\uparrow$, $A^\downarrow$ and $B^\downarrow$ sublattices.

Figure 4: Similar plot as Fig. 3, but shown for the second group of LLs.

Figure 5: The $B_z$-dependent LL energy spectra for the first, second and third groups (the blue, black and red curves) in the absence/presence of crossings.

Figure 6: (a) The crossing and anti-crossing energy spectra due to the spin-up- and spin-down-dominated second group of LLs, (b) the $n^\downarrow_2 = 6$ and $n^\uparrow_0 = 2$ LL anti-crossing within a certain range of $B_z$; the variations of probability distributions along the (c) higher- and (d) lower-energy paths.

Figure 7: The low-energy density of states of monolayer bismuthene under $B_z = 0$ and 30 T for (a) conduction and (b) valence states.
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