Dirac fermions in Fe ultra-thin film

Xiao-Tian Zhang,1,2 Baolong Xu,1,2 Kohji Nakamura,3 and Ryuichi Shindou1,2,*
1International Center for Quantum Materials, Peking University, Beijing, China
2Collaborative Innovation Center of Quantum Matter, Beijing, China
3Department of Physics Engineering, Mie University, Tsu, Mie, 514-8507, Japan

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We show the existence of massive Dirac fermions in electronic band structures of a few Fe atomic layers with perpendicular magnetization. Based on a tight binding model fitted to ab-initio band structure, we observe four distinct massive Dirac fermions near the Fermi level, which result from atomic spin-orbit coupling of Fe and a band inversion between Fe 4s-3d_{x^2-y^2} hybrid orbital band and 3d_{xy} orbital band. These lead to a valence band with finite Chern integer (+2) and chiral edge modes near the Fermi level. When the chemical potential is set inside the Dirac gap by carrier doping, the Hall conductivity exhibits a plateau-like structure with quantized value $2(e^2/h)$, and orbital magnetization shows a prominent increase, latter of which is mostly due to chiral orbital motion of electrons along the edge modes. We discuss the stability of the Dirac fermions in Fe(001) monolayer on MgO(001) substrate and Fe(001) bilayer case.

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I. INTRODUCTION

Since the discovery of graphene1,2 a number of novel two-dimensional electronic phases have been theoretically proposed.3,4 Such efforts include a proposal of so-called ‘Chern insulator’ in graphene with ferromagnetic substrates. Thereby, it was theoretically proposed that magnetic proximity effect from the substrate in combination with atomic spin-orbit interaction of carbon atom could give rise to a finite mass gap in the Dirac fermion of graphene, resulting in the quantized Hall conductance.5,6 On the experiment end, a few atomic layers of 3d-electron ferromagnets themselves such as Fe, Co, Ni and their alloys can be easily grown on various substrates.7 When the thickness of magnetic layers reaches atomic scale, magnetic anisotropy energy associated with surface magnetism dominates over magnetostatic energy, often making magnetic easy axis to be perpendicular to the layer; perpendicular magnetic anisotropy (PMA).8–27 To obtain comprehensive understandings of the PMA phenomena, it is also important to extract common feature in electronic band structures of a few atomic layers of d-electron ferromagnets such as Fe, Co and Ni.

In this paper, we show the existence of massive Dirac fermions near the Fermi level in electronic band structures of bcc Fe(001) a few atomic layers with perpendicular magnetization (Fig. 1). The Dirac fermions are well described by a three-orbital tight-binding model composed of 3d_{xy}, 3d_{x^2-y^2} and 4s orbitals of Fe atom, where an atomic spin-orbit interaction of Fe and a band inversion between 3d_{xy} orbital band and 4s-3d_{x^2-y^2} hybrid-orbital band play essential role for the emergence of the Dirac fermions. From ab-initio band calculations, the mass of the Dirac fermions is estimated to be around 80/60 meV for a Fe monolayer without/with MgO(001) substrate. In a free-standing Fe(001) monolayer, the massive Dirac fermion results in plateau-like feature in Hall conductivity and $dM_{\text{orb}}/d\mu$ near the Fermi level, where $M_{\text{orb}}$ out-of-plane orbital magnetization and $\mu$ the chemical potential. In experiments, the chemical potential can be controlled by an electric gate voltage (out-of-plane electric field). It is shown that the Dirac fermions in Fe monolayers are robust against broken out-of-plane inversion symmetry induced by the electric voltage. A comparison with existing ab-initio band calculations16,28 suggests that the Dirac fermions of the same origin can be also found in electronic band structures of bcc Co(001) and hcp Co(111) monolayers.

FIG. 1: Four massive Dirac fermions in the electronic band structure of a perpendicularly magnetized free-standing Fe monolayer.
II. ELECTRONIC BAND STRUCTURE

Ab-initio calculations are performed using film full-potential linearized augmented plane wave (FLAPW) method based on the local spin density approximation (LSDA) in which the core states are treated fully relativistically and the valence states are treated semi-relativistically. LAPW functions with a cutoff of $|k + G| \leq 3.9$ a.u. and muffin-tin sphere radii of 2.2, 2.2 and 1.4 a.u. for Fe, Mg and O atoms are used, where the angular momentum expansion inside the MT spheres is truncated at $\ell = 8$ (Fe and Mg) and $\ell = 6$ (O) for the wave functions, charge density and potential. The Fe/MgO was modeled by an Fe monolayer on a six-atomic-layer MgO(001) substrate, where Fe atoms locate on top of the O atoms, assuming the in-plane lattice constant matching to the calculated value of bulk MgO while the out-of-plane coordinates of Fe atoms are fully optimized using the atomic-force FLAPW calculations. Note that the structural parameter obtained is fitted near the Fermi level with a tight binding model composed of five 3d orbitals of Fe atom; 4s, 4p_x and 4p_y orbitals of Fe atom;

$$\hat{H} = \sum_{i,m\uparrow} \epsilon_{i,m\uparrow} \hat{c}_{i,m\uparrow}^{\dagger} \hat{c}_{i,m\uparrow} + \sum_{\langle i,j \rangle; mn} t_{ij;mn\uparrow} \hat{c}_{i,m\uparrow}^{\dagger} \hat{c}_{j,n\uparrow} + \lambda_{so} \sum_{i;mn} \hat{c}_{i,m\uparrow}^{\dagger} [L]_{mn\uparrow\uparrow} [S]_{\uparrow\uparrow} \hat{c}_{i,n\uparrow}$$

(1)

$\hat{c}_{i,m\uparrow}(\hat{c}_{i,m\uparrow}^{\dagger})$ is the electron creation (annihilation) operator on site $i$, for orbital $m$, and spin up (where spin quantization axis is taken along the perpendicular magnetization direction, z-direction). The first term represents an effective atomic energy of each orbital, which includes all the on-site type energies felt by respective orbital, such as exchange splitting energy from majority spin electrons and crystal fields caused by surrounding electrons. The second term represents neighboring hopping term with inter-atomic/intra-atomic transfer integrals $t_{ij;mn\uparrow}$. The angular bracket in $\langle \langle i,j \rangle \rangle$ stands for summation of nearest neighboring and next nearest neighboring sites. The third term is the atomic spin-orbit coupling with coupling strength taken to be $\lambda_{so} = 50$ meV from several literatures. Since an exchange splitting between majority spin bands and minority spin bands estimated from ab-initio band calculations is about 3 eV, being 60 times larger than the atomic spin-orbit coupling strength of Fe, we consider only diagonal part of spin-orbit interactions with respect to spin index; an expected correction due to the off-diagonal parts, $[L_{xz}],[S_{xy}]$, is evaluated to be on the order of 1 meV, being fairly negligible compared to that from the diagonal part. The interatomic transfer integrals $t_{ij;mn\uparrow}$ are given by matrix

FIG. 2: (a) Electronic band structures for minority spin near the Fermi level (without atomic SOI) from LSDA calculation (black) and 8-bands tight-binding model (blue). The Dirac fermion is depicted by a green circle. (b) from the 6-bands tight-binding model (with atomic SOI). The lowest band which acquires the Chern number 2 is drawn with red bold line. The massive Dirac fermions are depicted by a green circle. (c) from LSDA calculation with MgO(001) substrate and without atomic SOI. The orbital characters of bands labeled as 1, 2 are $3d_{xz}$, $3d_{yz}$, while those of 3, 4, 5, 6 are $3d_{x^2-y^2}$, $3d_{xy}$, 4s, 3d_{z^2} respectively.

$\epsilon_{i,m\uparrow} = \sum_{\ell} \langle \ell | t_{ij} | \ell \rangle \hat{c}_{i,m\uparrow}^{\dagger} \hat{c}_{j,n\uparrow}$

$\hat{c}_{i,m\uparrow} = S_{\uparrow}^{\dagger} \hat{c}_{i,m\uparrow} S_{\uparrow}$

$\hat{c}_{i,m\uparrow}$ is the electron creation operator on site $i$, for orbital $m$, and spin up.
elements in the Slater Koster table\textsuperscript{34} such as $V_{sd}$, $V_{sp}$, $V_{ddσ}$, $V_{ddτ}$, $V_{ddδ}$ and so on. These matrix elements in the Slater Koster table along with the effective atomic energies $\epsilon_{nℓτ}$ are used as fitting parameters, whose best fitted values in the absence of spin-orbit interaction are shown in Table I. The table shows reasonable fitting values compared to the Solid State Table\textsuperscript{34} and the lattice constant of the square-lattice Fe monolayer evaluated from the same first principle calculation ($a = 2.95\,\text{Å}$).

Figure 2(a) plots the band structure along high symmetry $k$ points obtained from both our eight-bands tight binding model and LSDA calculations with $λ_{σ0} = 0$. Notably, there exists four linearly dispersive band crossings along the zone boundary connecting $M$ point and $X$ point near the Fermi level. As shown below, these linear dispersions are well described by massless Dirac fermions without the spin-orbit interaction. With the atomic spin-orbit interaction, each massless Dirac fermion acquires same sign of the mass, endowing a valence band with finite Chern number $+2$.

The four Dirac cones are enveloped by other two dispersive bands composed of $d_{xz}$ and $d_{yz}$, while the out-of-plane mirror symmetry allows us to treat these two bands separately from the other six bands composed of $s, d_{x^2−y^2}$, $d_{xy}$, $p_x$, $p_y$ and $d_z$, because $\langle z\,\text{odd}\,|H|z\,\text{even}\rangle = \langle x\,\text{odd}\,|H|z\,\text{even}\rangle = 0$. The presence of substrate, such as MgO(001), breaks the out-of-plane mirror symmetry, giving rise to a finite mixing between these two groups of bands, while, more importantly, the oxide substrate endows with relatively strong charging energies the out-of-plane $d$ orbitals, such as $d_{xz}$, $d_{yz}$ and $d_z$. Thus, $d_{xz}−d_{yz}$ bands are brought into a higher energy region, while the four Dirac fermions composed by $4s$, $3d_{x^2−y^2}$ and $3d_{xy}$ remain intact (Fig.2(c)). For the sake of clarity, we consider in the following a free standing Fe monolayer and treat these two groups of bands separately, unless dictated otherwise. The case with MgO(001) substrate will be discussed later more carefully.

### III. BAND INVERSION MECHANISM

The emergence of the Dirac fermions and the valence band acquiring the Chern number $+2$ result from (i) a band inversion between $3d_{xy}$ orbital band and $4s$-$3d_{x^2−y^2}$ hybrid-orbital band, and (ii) a complex-valued band mixing between these two bands mediated by the atomic spin orbital interaction. Due to the orbital symmetry, the nearest neighbor intra-orbital transfer integrals make the hybrid orbital band to have positive curvature at the $Γ$ point and negative curvature at the $M$ point, while the $3d_{xy}$ orbital band to have negative curvature at the $Γ$ point and positive curvature at the $M$ point. As a result, the hybrid-orbital band comes lower than $3d_{xy}$ orbital band at the $Γ$ point, while comes higher than $3d_{xy}$ at the $M$ point (‘band inversion’).

In the presence of the atomic spin-orbit interaction with the out-of-plane ferromagnetic moment, $d_{xy}$ orbital is mixed with $d_{x^2−y^2}$ orbital character with pure imaginary coefficient, i.e. $d_{xy} \to d_{xy} + iαd_{x^2−y^2}$, with $α$ proportional to the spin-orbit interaction. Because of this mixing, the hybrid orbital band and $3d_{xy}$ orbital band acquire a complex-valued band mixing, which takes form of $t′\sin k_x\sin k_y + iα(t\cos k_x − \cos k_y)$ in the momentum space; $\sin k_x\sin k_y$ and $\cos k_x − \cos k_y$ are from the $3d_{xy}$ and $3d_{x^2−y^2}$ orbital symmetry respectively. As shown below, the band inversion and the complex-valued band mixing result in the valence band with Chern number $+2$.

The valence band with Chern number $+2$ can be well described by the lowest energy band of a three-bands tight-binding model composed of $4s$, $3d_{xy}$ and $3d_{x^2−y^2}$ orbitals. In the momentum space, the model takes form of,

$$H^{3×3}(k) = \begin{pmatrix}
E_s(k) & -2\sqrt{V_{sd}}s_8s_y & \sqrt{V_{sd}}(c_x − c_y) \\
* & E_{xy}(k) & -2iλ_{σ0} \\
* & * & E_{x^2−y^2}(k)
\end{pmatrix},$$

where $E_i(k) = \xi_i + t_i(c_x + c_y) + 2t'_ic_xc_y$, $c_x,y \equiv \cos k_{x,y}$, $s_{x,y} \equiv \sin k_{x,y}$, subindex $i = s, xy$, $x^2 − y^2$ representing $4s$, $3d_{xy}$, $3d_{x^2−y^2}$ orbitals. $ξ_i$ being their effective atomic energies, $t_i$ and $t'_i$ denotes their nearest and next nearest neighbor intra-orbital transfer integral. The lowest band energy of eq. (2) will be denoted as $E_−(k)$ henceforth.

To see the band topology for the lowest band, let us first derive an effective $2×2$ Hamiltonian out of $H^{3×3}(k)$. To this end, notice first that the lowest band energy is always smaller than $E_s(k)$, $E_{xy}(k)$ and $E_{x^2−y^2}(k)$ for all $k$; $E_− < \min(E_s, E_{xy}, E_{x^2−y^2})$. In such an occasion, we may describe the lowest band, by treating either one of

| $\sigma$ | $\pi$ | $δ$ |
|---|---|---|
| Fe d-Fe d (NN) | -0.363 | 0.261 | -0.061 |
| Fe d-Fe d (NNN) | -0.080 | 0.064 | -0.025 |
| Fe p-Fe d (NN) | -0.392 | 0.157 | — |
| Fe p-Fe d (NNN) | -0.118 | 0.047 | — |
| Fe p-Fe p (NN) | 2.905 | -1.063 | — |
| Fe s-Fe d (NN) | -0.392 | — | — |
| Fe s-Fe d (NNN) | -0.118 | — | — |
| Fe s-Fe s (NN) | -1.419 | — | — |
| Fe s-Fe p (NN) | 2.208 | — | — |

Table I: (upper) Tight-binding hopping parameters for minority spin bands, obtained from the fitting to the LSDA calculation. The energy unit is eV. (lower) On-site effective atomic energies for minority-spin $3d$ electrons and $4s$, $4p_x$, $4p_y$ electrons.
these three orbitals as a high energy degree and deriving an effective $2 \times 2$ Hamiltonian for the other two; the lowest band of $H^{2 \times 3}(\mathbf{k})$ is identified with that of the $2 \times 2$ Hamiltonian. As shown in Fig. 2(b) and Fig. 3, the focused valence band has mainly $4s$ and $3d_{xy}$ characters. Thus, we regard $3d_{x^2-y^2}$ orbital as the high energy degree of freedom and treat its couplings with $4s$ and $3d_{xy}$ as perturbations. This leads to the $2 \times 2$ effective Hamiltonian for ‘$4s$ band’ and $3d_{xy}$ band. Quantitatively speaking, ‘$4s$ band’ thus introduced is a $4s-3d_{x^2-y^2}$ hybrid-orbital band rather than purely $4s$ orbital band, because of larger mixing between $4s$ and $3d_{x^2-y^2}$ coming from $V_{sd}$ ($\gg \lambda_{so}$). In fact, the hybrid band has larger $3d_{x^2-y^2}$ orbital character than $4s$ orbital in a certain momentum region (see Fig. 3).

The degenerate perturbation theory gives the $2 \times 2$ Hamiltonian as follows,

\[ \langle i | H_{\text{eff}}^{2 \times 2} | j \rangle = \langle i | H_0 | j \rangle + \frac{\langle i | H_1 | x^2 - y^2 \rangle \langle x^2 - y^2 | H_1 | j \rangle}{E - E_{2s-\gamma^2}(\mathbf{k})} \]  

with $i, j = s, xy$ and

\[ H_0 = \begin{pmatrix} E_x(\mathbf{k}) & -2\sqrt{3}V_{sd}^s s_y & 0 \\ -2\sqrt{3}V_{sd}^s s_y & E_{xy}(\mathbf{k}) & 0 \\ 0 & 0 & E_{2s-\gamma^2}(\mathbf{k}) \end{pmatrix} \]

\[ H_1 = \begin{pmatrix} 0 & 0 & \sqrt{3}V_{sd}(c_x - c_y) \\ 0 & 0 & -2i\lambda_{so} \\ \sqrt{3}V_{sd}(c_x - c_y) & 2i\lambda_{so} & 0 \end{pmatrix} \]  

Equivalently,

\[ H_{\text{eff}}^{2 \times 2} = \begin{pmatrix} \mathcal{E}_s(\mathbf{k}) & \mathcal{E}_{s,xy}(\mathbf{k}) \\ \mathcal{E}_{s,xy}(\mathbf{k}) & \mathcal{E}_{xy}(\mathbf{k}) \end{pmatrix} \]  

with

\[ \mathcal{E}_s = E_s - \frac{3V_{sd}^2(c_x - c_y)^2}{\Delta E}, \quad \mathcal{E}_{xy} = E_{xy} - \frac{4\lambda_{so}^2}{\Delta E}, \]

\[ \mathcal{E}_{s,xy} = -2\sqrt{3}V_{sd}^s s_y s_y - \frac{i2\sqrt{3}\lambda_{so}V_{sd}(c_x - c_y)}{\Delta E} \].

The perturbation treatment is valid as far as $\Delta E \equiv E_{2s-\gamma^2}(\mathbf{k}) - E$ is positive. This condition is satisfied for any $\mathbf{k}$ when $E = E_-(\mathbf{k})$.

The $2 \times 2$ Hamiltonian has two eigenvalues whose smaller one corresponds to the lowest band energy of $H^{2 \times 3}(\mathbf{k})$. Thereby, $E_-(\mathbf{k})$ can be obtained from the following self-consistent equation of $E_-$:

\[ 2E_-(\mathbf{k}) = \mathcal{E}_s(\mathbf{k}) + \mathcal{E}_{xy}(\mathbf{k}) - \sqrt{(\mathcal{E}_s(\mathbf{k}) - \mathcal{E}_{xy}(\mathbf{k}))^2 + 4|\mathcal{E}_{s,xy}(\mathbf{k})|^2}, \]

where the right hand side is given by $E_-(\mathbf{k})$ itself by way of $\Delta E \equiv E_{2s-\gamma^2}(\mathbf{k}) - E_-(\mathbf{k})$. The equation has a solution for $E_-$ which always satisfies $E_- < \min(E_s, E_{xy}, E_{2s-\gamma^2})$ for any $\mathbf{k}$, justifying a posteriori the validity of the perturbative treatment.

With this justification in mind, we can readily identify the band topology of the lowest energy band of $H_{\text{eff}}^{2 \times 2}(\mathbf{k})$ as that of $H^{2 \times 3}(\mathbf{k})$. As is clear from Fig. 2(b), the hybrid band comes lower than $3d_{xy}$ band at the $\Gamma$ point; $\mathcal{E}_s < \mathcal{E}_{xy}$ at $\mathbf{k} = \Gamma$, while otherwise at the $M$ point; $\mathcal{E}_s > \mathcal{E}_{xy}$ at $\mathbf{k} = M$. Between $\Gamma$ and $M$ point, these two bands have a mixing due to a finite off-diagonal matrix element $\mathcal{E}_{s,xy}(\mathbf{k})$. Importantly, the matrix element has a complex phase, which acquires $4\pi$ phase, whenever $\mathbf{k}$ goes around the $\Gamma$ point (or $M$ point).

\[ \oint_{\partial S} \nabla \{ \text{arg} \mathcal{E}_{s,xy}(\mathbf{k}) \} \cdot d\mathbf{k} = 4\pi, \]

where $\partial S$ denotes an arbitrary loop which encompasses the $\Gamma$-point (or $M$-point).

The $\pi$ phase winding of the inter-band matrix element and the band inversion between the hybrid band and $3d_{xy}$ band endow the lowest band with the Chern number $+2$. To see this, expand the $2 \times 2$ Hamiltonian in terms of the Pauli matrix, $H_{\text{eff}}^{2 \times 2}(\mathbf{k}) = a_0(\mathbf{k})\sigma_0 + a(\mathbf{k}) \cdot \sigma$, from which the normalized vector $\mathbf{n}(\mathbf{k})$ is introduced by $\mathbf{n} \equiv a/|a|$. According to the projective representation of the Chern invariants $\mathbb{Z}^M$, the Chern number for the lowest band ($\text{Ch}_-$) is given by an integral of solid angle subtended by the unit vector over the first Brillouin zone; $\text{Ch}_- \equiv \frac{1}{4\pi} \int_{BZ} d\mathbf{k} \cdot \nabla \mathbf{n}(\mathbf{k}) \cdot \partial_{\mathbf{k}} \mathbf{n}(\mathbf{k}) \times \partial_{\mathbf{k}} \mathbf{n}(\mathbf{k})$. The integral is quantized to be integer, which counts how many times the unit vector wraps the unit sphere when the momentum $\mathbf{k}$ wraps the first Brillouin zone once. Now that $\mathcal{E}_s < \mathcal{E}_{xy}$ and $\mathcal{E}_{s,xy} = 0$ at the $\Gamma$ point while $\mathcal{E}_s > \mathcal{E}_{xy}$ and $\mathcal{E}_{s,xy} = 0$ at the $M$ point, the unit vector points to the south pole/north pole of the unit sphere when $\mathbf{k}$ at the $\Gamma$/M point respectively. On the other hand, eq. 7 means that the unit vector always winds twice around the pole when $\mathbf{k}$ rotates once around the $\Gamma$ point. This dictates that the Chern integer for the lowest band is $+2$.

The lowest band forms four distinct Dirac fermions along the Brillouin zone boundary, $\{ K_1, K_2, K_3, K_4 \} = \{ (\pi, K), (\pi, -K), (K, \pi), (-K, \pi) \}$. Around each Dirac

FIG. 3: ‘Fat-band’ picture obtained from the 6-bands tight-binding model. Respective orbital character is depicted by the line width (red) in (a) for $4s$, in (b) for $3d_{x^2-y^2}$ and in (c) for $3d_{xy}$. 
point, the effective $3 \times 3$ Hamiltonian conceiving Dirac fermion is linearly expanded in small $q_x$, $q_y$ and $\lambda_{so}$, e.g.

$$H^{3\times3}_{\text{eff}}(k) = M_1(q_x,a)\sigma_x + M_2\lambda_{so}\sigma_y + M_3(q_y,a)\sigma_z + \cdots \quad (8)$$

with $k \equiv q + K_1$, and $M_1 = -0.34$eV, $M_2 = -0.97$, $M_3 = -0.66$eV for the tight-binding parameters in Table I.

The four Dirac points play role of dual magnetic monopoles in a three-dimensional parameter space subtended by $k_x$, $k_y$ and $\lambda_{so}$\textsuperscript{38–41}. The corresponding magnetic field $B_-(k, \lambda_{so})$ is associated with a Bloch wavefunction for the lowest band, $|u_-(k, \lambda_{so})\rangle$ with $H(k, \lambda_{so})|u_-\rangle = E_-|u_-\rangle$. The magnetic field is a rotation of a three-component gauge field $A_-\equiv (\partial_{k_x}, \partial_{k_y}, \partial_{\lambda_{so}})$. $A_-\equiv (k, \lambda_{so})$ are gauge connections of the Bloch wavefunction: $A_- = i\langle u_-|\nabla|u_-\rangle$. Due to the four-fold rotational symmetry, dual magnetic charges at four Dirac points have the same quantized strength $2\pi$, where their sign is same as $-\text{sgn}[M_1M_2M_3] \quad (9)$.

$$\nabla \cdot B_-(k, \lambda_{so}) = 2\pi \sum_{j=1}^{4} \delta(\lambda_{so}) \delta(k - K_j) \quad (9)$$

The Chern integer for the lowest band is the total magnetic flux penetrating through the constant $\lambda_{so}$ plane in the 3D space,

$$\text{Ch}_-(\lambda_{so}) = \int_{\text{BZ}} \frac{dk}{2\pi} \left( \partial_{k_x} A_{-, y}(k, \lambda_{so}) - \partial_{k_y} A_{-, x}(k, \lambda_{so}) \right).$$

When $\lambda_{so}$ goes across the $\lambda_{so} = 0$ plane, the Chern integer changes by $-4$ (Fig. 4).

$$\text{Ch}_-(\lambda_{so} < 0) - \text{Ch}_-(\lambda_{so} > 0) = -4 \quad (10)$$

The time reversal symmetry connects the spinless tight-binding Hamiltonian for $\lambda_{so} > 0$ and that for $\lambda_{so} < 0$ with the relation

$$H^*(k, \lambda_{so}) = H(-k, -\lambda_{so}),$$

which leads to $\text{Ch}_-(\lambda_{so}) = -\text{Ch}_-(-\lambda_{so})$. Combing this with eq. (10), we have $\text{Ch}_-(\lambda_{so} > 0) = +2$.

FIG. 4: Schematic picture of dual magnetic magnetic charges and dual magnetic field in the three-dimensional space subtended by $k_x$, $k_y$ and $\lambda_{so}$. The Chern integer for each $\lambda_{so}$ is given by the surface integral of the field over the first Brillouin zone for the constant $\lambda_{so}$;

FIG. 5: (upper/lower) Hall conductivity/orbital magnetization as a function of the chemical potential for 8-bands tight-binding model for a free-standing Fe monolayer. Owing to the out-of-plane mirror symmetry, the contribution can be decomposed into that from the 6-bands electronic states ($3d_{x^2-y^2}$, $3d_{z^2}$, $4s$, $4p_x, 4p_y$; red color) and that from the 2-bands electronic states ($3d_{xz}, 3d_{yz}$, blue color). The direct band gap region associated with the massive Dirac fermions are specified by a grey-hatched energy window, $[-0.05\text{eV}, 0.03\text{eV}]$.

IV. HALL CONDUCTIVITY AND ORBITAL MAGNETIZATION

The hallmark of the existence of massive Dirac fermions is the transverse conductivity\textsuperscript{38–41}. The Hall conductivity as a function of the chemical potential is calculated for the eight-bands tight binding model for a free standing Fe monolayer (Fig. 5) with;

$$\sigma_{xy} = \frac{e^2}{h} \sum_{n} \int_{\text{BZ}} \frac{dk}{2\pi} \left( \partial_{k_x} A_{n,y} - \partial_{k_y} A_{n,x} \right)$$

where $A_{n, \mu} = i\langle u_n | \partial_{\mu} | u_n \rangle$ with $H(k)|u_n(k)\rangle \equiv \mathcal{E}_n|u_n(k)\rangle$, $n$ the band index. Due to the out-of-plane mirror symmetry, the conductivity can be decomposed
into the 2-bands contribution (from $3d_{xz}$ and $3d_{yz}$ orbitals) and the 6-bands contribution (from $4s$, $3d_{xz} - y^2$, $3d_{xy}$, $3d_{x^2 - y^2}$, $4p_x$, $4p_y$ orbitals). When the chemical potential is inside the Dirac gap ($\mu \approx 0$), the Hall conductivity shows a prominent peak structure with a maximum value around $2e^2/h$. The peak structure is mainly due to a nearly quantized contribution from the 6-bands electronic states. The quantized value is approximately $2e^2/h$ which is a direct consequence of the four massive Dirac fermions near $\mu = 0$. A slight deviation from the quantization is attributed to another small but non-vanishing dual magnetic fields associated with a dispersive band near the $\Gamma$ point.

The 2-bands electronic state also gives a nearly quantized contribution $e^2/h$ to the Hall conductivity near the Fermi level ($\mu \approx -0.1eV$); $3d_{xz}$ and $3d_{yz}$ orbital bands comprise another SOI-induced direct band gap at the $M$ point. The tight-binding Hamiltonian for $3d_{xz}$ and $3d_{yz}$ orbitals is expanded linearly in small $q_x q_y$, $q_x^2 - q_y^2$ and $\lambda_{so}$ with $k = q + (\pi, \pi)$:

$$H(k) = \lambda_{so} \sigma_y + (V_{dd\pi} - V_{dd\sigma})(q_x a)(q_y a) \sigma_x + (V_{dd\pi} - V_{dd\sigma})(q_x a)^2 - (q_y a)^2 \sigma_z + \cdots,$$

where $V_{dd\sigma}^{(t)}$ and $V_{dd\pi}^{(t)}$ denote the Slater-Koster hopping parameters between the (next) nearest neighboring Fe d orbitals. The expansion dictates that the dual magnetic fields for the two bands have $4\pi$ magnetic charge at the $M$ point on the $\lambda_{so} = 0$ plane. This in combination with the symmetry property $H^*(k, \lambda_{so}) = H(-k, -\lambda_{so})$ requires that the integral of the dual magnetic field $\partial_{k_x} A_{n,y} - \partial_{k_y} A_{n,x}$; $n$ is either lower or higher band out of the two bands) near the $M$-point is quantized to be $2\pi$ in the smaller $\lambda_{so}$ limit. When the chemical potential is inside the SOI-induced gap at the $M$ point, one of the two bands is partially filled while the other is empty. Since a fermi surface associated with the filled band is large enough compared to a distribution of the magnetic flux around the $M$ point, the Hall conductivity from the two-band electronic state is nearly quantized to be $e^2/h$ as in Fig. 4.

Emergence of the massive Dirac fermions also results in peculiar chiral modes localized near the boundary of a two-dimensional Fe monolayer. Fig. 5 shows an electronic band structure of the 6-bands tight-binding model with periodic/open boundary condition along the $x/y$-direction of the square-lattice Fe monolayer. When projected onto a surface crystal momentum axis, the four massive Dirac fermions at $k = K_1, K_2, K_3, K_4$ reduce to three distinct valleys with a direct band gap, located at $k_x = \pi, \pm K$ respectively. Now that the gap endows the lower bulk band with the Chern number $+2$ as described above, the bulk-edge correspondence dictates that two localized chiral edge modes appear in the direct band gap of the three valleys (Fig. 6b). In the present case, the direct band gap is also masked by another dispersive bulk band located at $\Gamma$-point, mainly composed of $3d_{x^2 - y^2}$ orbital (Fig. 2). As a result, the chiral edge modes are terminated by the dispersive bulk band around $k_x = 0$ (Fig. 3a,c).

The chiral modes give rise to large out-of-plane orbital magnetization when the chemical potential $\mu$ set inside the Dirac gap. When increasing $\mu$ inside the gap, electrons are added up into the edge modes, which enhances chiral electric currents flowing around the boundary of the two-dimensional system. Irrespective of details of the energy dispersion of the chiral modes, the increase of the current is proportional to the increase of $\mu$. Such chiral edge current contributes to a macroscopic orbital moment $\langle r \times p \rangle_{\partial N/\partial H}$ which results in a linear increase of the magnetic moment with respect to the carrier doping near the Fermi level. To see this situation, we have calculated the orbital magnetization based on the Streda formula $^{51,52}$ which is the derivative of the free energy in the magnetic field $H$, while the total number of electrons $N$ is the derivative in $\mu$. This leads to $\partial N/\partial H = \partial M/\partial \mu$, provided that the free energy is analytic in $\mu$ and $H$. According to Streda $^{53}$ $\partial N/\partial H$ can be
expressed only in terms of the current operators;

\[ \frac{\partial M}{\partial \mu} = \frac{1}{ec} \left\{ \sigma_{xy}(\mu) - \frac{i\hbar}{2} \text{Tr}[J_x G^+(\mu) J_y (\mu - \mathcal{H}) - h.c.] \right\}, \] (11)

where \( J_\nu \) being the current operator (\( \nu = x, y \)) and \( G^\pm(\mu) \equiv 1/[(\mu \pm i\delta) I - \mathcal{H}] \) and \( \mathcal{H} \) being lesser, greater single-particle green functions and Hamiltonian respectively. By an integration over \( \mu \), the orbital magnetization is calculated from the eight-bands tight binding model for a free standing Fe monolayer (Fig. 3). Like the Hall conductivity, the result is decomposed into the 2-bands and the 6-bands contributions. The calculated magnetization exhibits a significant increase as a function of \( \mu \) when \( \mu \) is set inside the Dirac mass gap. The breakdown into the two contributions shows that the increase is mainly due to the 6-bands electronic states, indicating that the orbital moment near \( \mu = 0 \) mainly comes from an orbital motion of electrons along the chiral edge modes. In fact, \( \partial M/\partial \mu \) is nearly quantized in the unit of \( e/\hbar c \), which counts the number of chiral edge modes inside the Dirac gap.

V. SUBSTRATE, ELECTRIC GATE VOLTAGE AND MULTIPLE-LAYER EFFECTS, AND HCP CO(111) MONOLAYER CASE

The Dirac fermions found in a free-standing Fe monolayer are robust against various perturbations such as the oxide substrate, out-of-plane applied electric field (e.g. electric gate voltage applied perpendicular to the layer) and multiple-layer effects. Firstly, being a doubly degenerate point in an electronic energy band structure, the dual magnetic monopole (charge) discussed above is a stable point defect in the 3-dimensional parameter space subtended by \( k_x, k_y \) and \( \lambda_{so} \); they cannot disappear by themselves. The symmetry property \( H^+(k, \lambda_{so}) = H(-k, -\lambda_{so}) \) further requires these defects to be in the \( \lambda_{so} = 0 \) plane, which guarantees the existence of massive Dirac fermions even for small \( \lambda_{so} \). To annihilate these Dirac fermions, one generally needs to either re-invert the band inversion between \( 3d_{xy} \) band and \( 4s-3d_{x^2-y^2} \) hybrid band or reduce completely the inter-layer couplings among \( 4s \) orbital, \( 3d_{xy} \) and \( 3d_{x^2-y^2} \) orbitals. Unlike out-of-plane \( 3d \) orbitals, however, these in-plane \( 3d \) orbitals has little influences from the substrate and out-of-plane electric field. As a result, we can readily find the massive Dirac fermions of the same origin even in the presence of various perturbations.

Fig. 7 shows an electronic band structure for Fe (001) monolayer with MgO(001) substrate, where every Fe atom locates right above the oxygen of the MgO substrate. Due to crystal fields from these oxygens, three out-of-plane \( 3d \) orbital bands are brought up into a higher energy region. Due to the charge neutrality, the Dirac fermions formed by \( d_{xy} \) orbital and \( 4s-3d_{x^2-y^2} \) hybrid orbitals come lower than the Fermi level (\( E \approx -0.6eV \)). The size of the SOC-induced Dirac gap is estimated around 60 meV from the \textit{ab-initio} band calculation. The applied out-of-plane electric field has little effects on these Dirac fermions either. Even under a very large out-of-plane electric field (\( \pm 1 \) V/Å), four Dirac fermions are barely affected.

Fig. 7 shows an electronic band structure obtained from a tight-binding model for a free-standing Fe bilayer, where the number of massive Dirac fermions are doubled. Due to interlayer hoppings, a Dirac fermion from one layer and that from the other repel with each other in energy. When the chemical potential is around these Dirac gaps, the transverse conductivity shows a peak structure with its maximum value around 2\( e^2/h \). The out-of-plane orbital magnetization increases as a function of the chemical potential inside the gaps. These features are essentially same as in the free-standing Fe monolayer case.

A comparison between an existing \textit{ab-initio} band calculation\cite{17} and tight-binding analysis indicates that the massive Dirac fermions of the same kinds are also induced by the atomic spin-orbit interaction in minority-spin band in a hcp Co(111) monolayer with perpendicular magnetization. Thereby, \( d_{xz} \) and \( d_{yz} \) orbital bands comprise two massive Dirac fermions with positive mass at \( K \) and \( K' \) point respectively, which correspond to \( 4\pi \) magnetic charge at the \( M \)-point in the Fe(001) monolayer case. Meanwhile \( 4s \), \( d_{x^2-y^2} \) and \( d_{xy} \) orbitals form six massive Dirac fermions with positive mass along the high symmetric \( k \) lines connecting \( \Gamma \) and \( K(K') \) and 2 massive Dirac fermions with negative mass at \( K \) and \( K' \).
VI. CONCLUSION

Massive Dirac fermions are discovered near the Fermi level of an electronic band structure of Fe ultra-thin film. The Dirac gap is induced by atomic spin-orbit coupling on the order of 50 meV. The topological gap opening results from a band inversion between 3d_{xy} and 4s-3d_{x^2-y^2} hybrid orbital band, giving rise to a finite Chern number in a valence band. Inside the gap, the Hall conductivity (v.s. chemical potential) exhibits plateau-like structure with nearly quantized values, while orbital magnetization (v.s. chemical potential) increases rapidly due to the macroscopic orbital moment induced by topological chiral edge modes. The magnitude of the calculated orbital magnetization due to the chiral edge current is on the same order of experimental literature value being hardly negligible in general. Massive Dirac fermions in Fe ultra-thin film are shown to be robust against perpendicular inversion symmetry breaking (such as substrates or electric gate voltages) as well as multi-layer effect. More importantly, we found in Fe(001)/MgO(001) that the massive Dirac fermions are nicely separated from other dispersive bulk bands in energy, which may give a useful hint to explore possible ‘Chern insulator’ in transition metal ferromagnetic thin film. Considering richness of the transition metals with various substrates, we anticipate that Dirac fermion physics and Chern insulators may be observed experimentally in transition metal thin films in future.

* corresponding author; rshindon@pku.edu.cn

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