Quantized non-Abelian, Berry’s flux and higher-order topology of Na\textsubscript{3}Bi

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(Dated: February 12, 2021)

Recent theoretical works on effective, four-band models of three-dimensional, Dirac semimetals suggest the generic planes in momentum space, orthogonal to the direction of nodal separation, and lying between two Dirac points are higher-order topological insulators, supporting gapped, edge-states. Furthermore, the second homotopy classification of four-band models shows the higher-order topological insulators support quantized, non-Abelian Berry’s flux and the Dirac points are monopoles of \textit{SO}(5) Berry’s connections. Due to the lack of suitable computational scheme, such bulk topological properties are yet to be determined from the \textit{ab initio} band structures of Dirac materials. In this work, we report first, comprehensive topological classification of \textit{ab initio} band structures of Na\textsubscript{3}Bi, by computing Wilson loops of non-Abelian, Berry’s connections for several, Kramers-degenerate bands. Our work shows the quantized, non-Abelian, Berry’s flux can be used as a stable, bulk invariant for describing higher-order topology and topological phase transitions.

\textbf{Introduction}: The gauge invariant, spectra of Wilson loops \cite{1} encode physical information about topologically non-trivial configurations of non-Abelian gauge fields \cite{2–5}. The Wilson loops along the periodic imaginary time direction and various non-contractible cycles of torus are known as the Polyakov loops (PL), which are widely used by lattice-gauge theorists for addressing the topological properties and the confinement-deconfinement transitions of gauge fields at finite temperatures \cite{3–5}. Naturally, the PLs of intra-band, non-Abelian Berry’s connections, defined in momentum space, along various high-symmetry axes of Brillouin zone have emerged as powerful tools for describing topology of quasi-particle band-structures \cite{6–9}. The eigenvalues of such PLs, commonly referred to as the Wannier charge centers (WCC) in band-theory literature, allow high-throughput screening of materials database for identifying topological materials, which can support zero-energy, surface states \cite{10}.

However, all topologically non-trivial states do not support gapless surface-states. Consequently, the WCCs cannot identify bulk topology of higher-order topological insulators (HOTI), which exhibit gapped, surface-states, and corner-localized, mid-gap states \cite{11}. In recent years, the concept of HOTIs has also become relevant for understanding bulk topology and surface-states of three-dimensional, topological semimetals (see Fig. 1 ) \cite{12–18}. Therefore, the computational diagnosis of HOTIs from \textit{ab initio} band structures is an important, open problem. In this work, we will solve this problem for two-dimensional HOTIs by computing eigenvalues of planar Wilson loops (PWL) \cite{1, 2}. The PWL defines \textit{SU}(2) Berry’s phase accrued by the wave functions of Kramers-degenerate bands, when they are parallel transported along any closed, non-self-intersecting curve, lying in \(k_x - k_y\) planes of Fig. 1. The eigenvalues of PWLs are gauge invariant, diagnostic tools of quantized, non-Abelian Berry’s flux \cite{19}. We will explicitly demonstrate
the power of this method by performing topological classification of \textit{ab initio} band structures of Dirac semimetal phase of Na$_3$Bi. This manifestly gauge invariant method does not require any explicit knowledge of underlying basis states and is also capable of identifying which bands are topologically non-trivial.

Na$_3$Bi was proposed as the first candidate material for realizing stable DSMs, which arise from linear touching between a pair of two-fold, Kramers-degenerate bands at isolated points of momentum space, along an axis of $n$-fold rotation (say the $\hat{z}$ or c-axis) [20]. The Dirac points are simultaneously protected by the combination of parity and time-reversal symmetries ($P\bar{T}$) and the $n$-fold rotational ($C_n$) symmetry [21, 22]. The qualitative features of DSM phase have been well characterized with the first principles calculations of band structures and various spectroscopic, and transport measurements[20, 23–27]. The low-energy theory of such DSMs is described by the four-band, $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian $H(\mathbf{k}) = c_0(\mathbf{k}) + \sum_{j=1}^{5} d_j(\mathbf{k}) \Gamma_j$, where $\Gamma_j$’s are five, mutually anti-commuting, $4 \times 4$ matrices, and $\mathbb{I}$ is the $4 \times 4$ identity matrix [20]. The topological properties of conduction and valence bands are controlled by the $O(5)$ vector field $d_1 = A k_x$, $d_2 = A k_y$, $d_3 = B k_z (k_x^2 - k_y^2)$, $d_4 = 2 B k_x k_y k_z$, and $d_5 = M_0 - M_1 k_x^2 - M_2 (k_x^2 + k_y^2)$, where $A$, $B$, $M_0$, $M_1$, and $M_2$ are band parameters. For Na$_3$Bi, the parameters $M_0 < 0$, $M_1 < 0$, and $M_2 < 0$ capture band inversion effects, leading to two Dirac points along the six-fold, screw axis at $(0,0,\pm k_D)$, with $k_D = \sqrt{M_0/M_1}$. The particle-hole anisotropy term $c_0(\mathbf{k})$ does not affect band topology.

For describing low-energy physics of massless Dirac fermions, $d_3$ and $d_4$ terms can be ignored in the renormalization group sense [20, 28, 29]. Such approximate theories predict topologically protected, loci of zero-energy surface-states, also known as the \textit{helical Fermi arcs}, joining the projections of bulk Dirac points on the (100) and the (010) surface- Brillouin zones. Therefore, the spectroscopic detection of \textit{helical Fermi arcs} was often considered to be the smoking gun evidence of bulk topology of DSMs [23, 24]. However, these terms cannot be ignored for addressing topological properties of generic planes and they are responsible for gapping out the helical edge states for all $|k_z| < k_D$ and $k_z \neq 0$ [30–34], and giving rise to higher-order topology [12, 13, 16].

What is the stable, bulk topological invariant of two-dimensional HOTIs? To answer this question, Tyner \textit{et al.} performed detailed \textit{second homotopy} classification of various four-band, tight binding models of DSMs [19]. By considering the topology of coset space $SO(5)/SO(4) = USp(4)/[SU(2) \times SU(2)]$ of Bloch Hamiltonians, they identified the Dirac points as a pair of unit strength, monopole and anti-monopole of non-Abelian, $SO(5)$ Berry’s connections. For all generic, non-trivial $xy$ planes or HOTIs, the conduction and the valence bands were shown to support quantized, non-Abelian flux of magnitude $2\pi$. The magnitude of flux was also shown to be invariant under $SO(5)$ gauge transformations and it was computed from the windings of gauge-invariant, eigenvalues of PWLs of $SU(2) \times SU(2)$ Berry’s connections. We will employ this method for topological classification of Na$_3$Bi, allowing us to address the stability of bulk invariants, when the coset space $USp(4)/[SU(2)^2]$ of four-band models is embedded within a larger coset space $USp(2N)/[SU(2)^N]$ of $N$ number of Kramer-degenerate bands, with $N > 2$.

\textbf{Ab initio band structures:} The crystal structure of Na$_3$Bi, shown in Fig. 2(a), belongs to the space group P6$_3$/mmc and has the lattice constants $a = b = 5.49$ Å, $c = 9.78$ Å. It consists of two non-equivalent Na sites, denoted by Na(1) and Na(2). The honeycomb layers formed by Na(1) and Bi are stacked along the c-axis, with

![Figure 2](image-url)
Na(2) sites located between the layers. The bulk Brillouin zone (BZ) is shown in Fig. 2(b). All first-principles calculations based on the density-functional theory are performed using the Vienna Ab initio simulation package [35, 36], and the exchange-correlation potentials use the Perdew-Burke-Ernzerhof (PBE) parametrization of the generalized gradient approximation [37]. An 11×11×7 grid of \( k \) points and a plane-wave cutoff energy 520 eV are used for self-consistent field calculations. All calculations incorporate the effects of spin-orbit coupling. The calculated band structures within the energy window \([-3 \text{ eV} \rightarrow +2 \text{ eV}\)] are displayed in Fig. 2(c). We have labeled the Kramers-degenerate bands, according to their energy eigenvalues at the \( \Gamma \) point, with \( E_n(0) < E_{n+1}(0) \). The bulk Dirac points arise from linear touching between bands \( n = 6 \) and \( n = 7 \), along the six-fold, screw axis \((A - \Gamma - A \text{ line or the } k_z \text{ axis})\) at \((0,0, \pm k_D)\), with \( k_D \approx 0.29 \times \frac{\pi}{c} \). Their reference energy coincides with the Fermi level.

In order to perform topological analysis of various bands, we have constructed realistic tight-binding models of ab initio band structures, by employing maximally localized Wannier functions. The Wannier functions have been calculated by using the WANNIER90 package [38]. The intra-band, \([SU(2)]^n = SU(2) \times SU(2) \times \ldots \times SU(2)\) connections will be obtained by using the formula \( A_{j,n,s,s'}(k) = -i \psi_{n,s}(k) \partial_j \psi_{n,s'}(k) \), where \( \psi_{n,s}(k) \) are the eigenfunctions of \( n \)-th band, with \( s = \pm 1 \) denoting the Kramers index, and \( \partial_j = \frac{\partial}{\partial k_j} \). We will calculate PWLs of individual \( SU(2) \) Berry’s connections of bands \( n = 1 \) through \( n = 8 \) by utilizing the Z2Pack software package [8, 10].

**Bulk Topology:** The PWL of \( SU(2) \) connections of \( n \)-th Kramers-degenerate bands, along a closed, non-intersecting curve \( C \), lying in the \( xy \) plane, parametrized by \( k(l) \) is defined as

\[
W_n = P \exp \left[ i \oint \sum_{j=1}^{2} A_{j,n}(k(l)) \frac{dk_j}{dl} \, dl \right],
\]

\[
= \exp \left[ i \theta_n(k_0) \Omega_n(k_0) \cdot \sigma \right],
\]

where \( P \) denotes path ordering and \( k_0 \) corresponds to the size of the loop. The gauge invariant angle \( \theta_n(k_0) \) can be related to the magnitude of non-Abelian, Berry’s flux by employing a non-Abelian generalization of Stokes’ theorem [19, 39–41]. The gauge dependent, three-component, unit vector \( \Omega_n(k_0) \) defining the orientations in \( SU(2) \) color space will not be used for computing any physical properties. When the \( n \)-th Kramers-degenerate bands support quantized, flux of magnitude \( 2\pi \), \( |\Delta \theta_n(k_0)| = |\theta_n(k_0) - \theta_n(0)| \) will interpolate between 0 and \( 2\pi \), as \( k_0 \) is systematically increased from being 0 to a final value \( k_f \), when the area enclosed by the loop becomes equal to the area of two-dimensional, Brillouin zone. This interpolation of \( |\Delta \theta_n(k_0)| \) between 0 and \( 2\pi \) is independent of the shape and the location of \( C \), as long as the final area enclosed by \( C \) equals the area of Brillouin zone. Thus, the geometry and the location/center of \( C \) can be chosen according to the convenience of numerical calculations.

For computational ease, we have followed the rectangular path \( ABCDA \), shown in Fig. 3(a), with \( |AD| = k_0 \) and \( |AB| = \frac{2\pi}{\sqrt{3}} k_0 \). We have varied \( k_0 \) between 0 and \( k_f = k_0 = \frac{4\pi}{\sqrt{3}} \). We first focus on the results for the Dirac band \( n = 7 \), displayed in Fig. 3(b). For all topologically
non-trivial planes with $|k_z| < k_D$, $|\Delta \theta_n(k_0)|$ shows 0 to $2\pi$ interpolation. Therefore, the $SU(2)$ Berry’s connections of band $n = 7$ support a quantized flux of magnitude $2\pi$ for all planes, lying between two Dirac points. For $|k_z| > k_D$, $|\Delta \theta_n(k_0)|$ does not exhibit such interpolation, and the trivial planes support 0 flux. Precisely at the nodal planes with $|k_z| = k_D$, $|\Delta \theta_n(k_0)|$ interpolates between 0 and $\pi$. Therefore, the flux through the nodal plane is equal to $\pi$. This situation is very similar to the half-integer, Hall conductivity (half-integer invariant) of massless Dirac fermions at the topological quantum critical point between Chern insulators and trivial insulators. For all non-trivial (trivial) planes, the Dirac bands $n = 6$ also support $2\pi$ (0) flux, as suggested by Fig. 3(c). These topological properties of Dirac bands are identical to what have been found from the effective, four-band model of $sp$-hybridized DSMs [19].

We have verified that the additional bands ($n \neq 6, 7$) do not support any quantized flux for any generic planes. Only at the $k_z = 0$ mirror plane, the remote bands $n = 3$ and $n = 5$ also possess quantized flux of magnitude $2\pi$ (see Fig. 4(a)). Therefore, the topological distinction between HOTIs and the $k_z = 0$ mirror plane can be demonstrated using PWLs. While the topology of $k_z = 0$ mirror plane can be diagnosed by computing hybrid WCCs of completely occupied bands [10], it does not address which bands are topologically non-trivial. To clearly show that the bulk topological properties of HOTIs are entirely governed by the Dirac bands, we have plotted $|\Delta \theta_n(k_0)|$ for different bands at the nodal (or critical) plane. Only the Dirac bands $n = 6$ and $n = 7$ exhibit $\pi$ flux, controlling the topological phase transitions between HOTIs and trivial insulators (see Fig. 4(b)).

In summary, we have identified the bulk invariants for describing higher-order topology of generic planes of Dirac material Na$_3$Bi. Hence, the PWLs can be used for establishing the topological universality class of DSMs in various compounds such as Cd$_3$As$_2$ [42], Bi$_n$Bi-family [43], Cu$_3$PdN [44], LiGaGe-family [45], PdTe$_2$ [46], $\beta'$-PtO$_2$ [16, 47], VA$_3$ [48], $\beta$-CuI [34], KMGBi [16, 49], FeSn [50]. Our results are insensitive to the number of underlying bands, suggesting the higher-order topology of real materials can be comprehensively addressed with stable, bulk invariants. The proposed method of computing bulk invariants is also insensitive to the underlying details of basis states. Therefore, it can be further developed as a reliable tool for high-throughput screening of HOTIs.

A. C. T., S. S., Q. Z., P. D. and P. G. were supported by the National Science Foundation MRSEC program (DMR-1720139) at the Materials Research Center of Northwestern University. D.P. and J.M.R. acknowledge the Army Research Office under Grant No. W911NF-15-1-0017 for financial support and the DOD-HPCMP for computational resources. Use of the Center for Nanoscale Materials (CNM), an Office of Science user facility, was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-06CH11357.

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