lek **d I. Mal i** [1,2], **M. Dal i** [1,3], **i -** **Zh** [1], **Zhi W** **g** [1],
le Z ge [1,*]

[1] E. I , f C , B , C 80309,
[2] C f , D n f , f , . B 1048 B , -0316 ,
[3] C C H n , F ABC, 09210-580, A , B

topoloid structures

fi fi

fi

properties in Scheme 1. Given a compilation

their characteristic ACSs, one first establishes t

retical conditions ("design principles") that w

logical property P to exist in such crystal st

one examines if the selected compound and it

structure have the stipulated topological prope

. Finding a positive answer est

candidates for topological properties. This approac

–8] leading to unique transport prop-
erties at lower dimensions. Laboratory examination and future
technological applications of the fascinating predicted properties
of such topological phases naturally require identifying synthe-
sizable and stable, topology-bearing compounds ("topoloids").
Searching for specific materials (characterized by tomic identi-
ties, omposition and tructure, or) that have required tar-
get property P (here, topological properties, but generally also
novel photovoltaic semiconductors, transparent conductors,
thermoelectric compounds or battery materials) generally fol-
lows an "Inverse Design" paradigm [9], illustrated for topological

actually synthesized and proven to be topological. We note that the predicted topoloids were obtained by using just the upper part of the general flow in Scheme 1 denoted as "partial screening flow". The latter screening has been often based on the assumption that the crystal will take up the highest symmetry possible, not considering the possibility of energy-lowering symmetry breaking that obviates topological properties. Unfortunately, both experimental and theoretical databases of structures and properties of compounds sometimes omit information on magnetism, spontaneous defect formation, or how would the structure change when significant doping is required. Furthermore, in the case of polymorphous networks [26] (structures having a distribution of different local motifs), experimental X-ray structure determination often approximates the structure by an artificial, high symmetry primitive unit cell describing the macroscopic average configuration. Such "virtual structures" should not be used as input to electronic structure calculations [26]

(energy lowering) formation of Bi vacancies that condense into ordered vacancy compounds whose low local symmetry defeats the specific degeneracy promoting topology.

(iii) Symmetry lowering by magnetism (spin-polarization) removes the topology-promoting band degeneracy: This is illustrated for the predicted 8-fold band degeneracy of metallic ₂ [17] in assumed nonmagnetic configuration. Once magnetic spin-polarization is allowed, the total energy decreases, and the band degeneracy disappears.

(iv) The predicted topological property requires doping that inherently destabilizes the topological structure: This is illustrated for [18] that requires an upshift of the Fermi energy by ~2 eV to meet the inverting energy bands. This occupation of massively antibonding states leads to a destabilizing increase in total energy relieved by structural transformations that defeat topology.

(v) The topological property is caused by artificial band inversion due to systematic density functional theory (DFT) errors: The method used to assess the band structure—DFT—requires an exchange–correlation energy functional. But current approximated functionals suffer from the self-interaction error that lowers the unoccupied bands and raises the occupied bands, causing, at the extreme, unphysical band inversion. This occurs in and [10–13], well known normal insulators predicted by DFT with current functionals to be topological metals.

Avalible materials databases (e.g., ICSD [31]) contains hundreds of thousands entries. These are first pragmatically narrowed down by theorists [10–13] using filters such as avoid duplications, limit the maximum cell size to be examined, exclude theoretically difficult to calculate atoms (d or f electrons), exclude alloys and other structures with fractional occupation numbers, etc. After this narrowing down, the band structures of the remaining compounds are computed by density functional. For instance, Vergniory et al. [10] calculated 26,938 compounds from which 181 (34) were found to be topological insulators with DFT gap between occupied and unoccupied states of at least 0.001 (0.1) eV. The Swiss TOPOMAT database [13] uses different filters to narrow down the entries ending up with 13,628 directly inspected cases and found 50 (17) topological insulators with DFT band gaps of >0.001 (0.1) eV. We note, however, that the number of compounds and the numerical results in these online databases are regularly changed by the authors.

We have studied only a few compounds from the published lists of predicted topological insulators with finite band gaps. These already illustrate possible failure modalities that can be detected by using simple, additional "search filters" from the same density functional theory used to establish the requisite target band structure. The current list of predicted topological insulators with real band gaps might then be a diminishing domain.

The ᵃ ᵃch

Identifying actual realizations of topological compounds requires theories with a full atomic resolution that recognize site and space symmetries in crystals. The recent rebirth of density functional band theory of solids (and higher-order theories that use

DFT to initialize the problem, such as DFT-DMFT [32], DFT-GW [33,34], DFT-QMC [35]) as a tool for uncovering topological behavior hidden in the spaghetti-like energy band structure, lies in DFT's ability to directly decode the consequences of an assumed ACS on the band structures. This affords a direct mapping of the theory onto the Periodic Table via explicit incorporation of the electron–ion potential $V_{ext} =$

What about metastable compounds:

manipulatable chemical potentials of elements – can be used to

has such a significant energy lowering potential that it might very likely defeat the intended metallic state of the predicted new Fermion.

dominated by anion p states, while cation s orbitals define the conduction band. Indeed, the existing experimental literature on the InN and InAs phases does not reveal any topological prop-

[54] G. Trimarchi, Z. Wang, A. Zunger, Phys. Rev. B 97 (2018) 035107.

[55] B. Khamari, B.R.K. Nanda, Mater. Res. Exp. 6 (2019) 066309.

[56] A. Walsh, A. Zunger, Nat. Mater. 16 (2017) 964.

[57] Q. Liu et al., Phys. Rev. Lett. 121 (2018) 186402.

[58] X. Zhang et al., Adv. Funct. Mater. 27 (2017) 1701266.

[59] F. Hong et al., J. Phys. Chem. C 120 (2016) 6435.

[60] J.-W. Luo, A. Zunger, Phys. Rev. Lett. 105 (2010) 176805.

[61] S. Lany, Phys. Rev. B 87 (2013) 085112.

[62] O. Madelung, Semiconductors, Group IV Elements and III-V Compounds, Springer Science & Business Media, 2012.

[63] Y.-S. Kim et al., Phys. Rev. B 82 (2010) 205212.

[64] J. Klimeš, M. Kaltak, G. Kresse, Phys. Rev. B 90 (2014) 075125.