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Article

A simple and efficient criterion for ready screening of potential topological insulators

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ABSTRACT

Topological materials are a new and rapidly expanding class of quantum matter. To date, identification of the topological nature of a given compound material demands specific determination of the appropriate topological invariant through detailed electronic structure calculations. Here we present an efficient criterion that allows ready screening of potential topological materials, using topological insulators as prototypical examples. The criterion is inherently tied to the band inversion induced by spin-orbit coupling, and is uniquely defined by a minimal number of two elemental physical properties of the constituent elements: the atomic number and Pauling electronegativity. The validity and predictive power of the criterion is demonstrated by rationalizing many known topological insulators and potential candidates in the tetradymite and half-Heusler families, and the underlying design principle is naturally also extendable to predictive discoveries of other classes of topological materials.

1. Introduction

Topological materials are a rapidly expanding family of quantum matter, which can be classified into topological insulators (TIs) [1,2], topological crystalline insulators (TCIs) [3], topological Dirac semimetals (TDs) [4,5], topological Weyl semimetals (TWs) [6–8], topological nodal-line semimetals (TDNLs) [9–12], nodal-chain metals [13], nodal-link semimetals [14], triple degenerate nodal point semimetals [15,16] and even beyond [17]. Such materials are attracting intensive attention in condensed matter physics and materials science, due to their intriguing physical properties and promising technological applications. For a given compound system, identification of its topological nature is generally complex, demanding specific determination of the appropriate topological invariant through detailed electronic structure and Berry curvature calculations. For instance, to identify whether a material is a three-dimensional (3D) TI or not, one would need to evaluate the Z_2 invariant [18]. On a microscopic level, the topologically nontrivial nature is tied to the appearance of inverted bands of different symmetries in the electronic structure, as described for the 3D TIs, and also emphasized for other classes of topological materials.

For most topological materials, band inversions have been demonstrated to be induced by delicate synergistic effects of different physical factors, including chemical bonding, crystal field, and, most notably, spin-orbit coupling (SOC). In particular, for the most widely studied topological systems of 3D TIs, SOC has been identified to play the vital role in inducing band inversion. Earlier studies of 3D TIs had been carried out with the calculations of Z_2 on a case-by-case basis [19]. In a more recent study [20], a variational high-throughput descriptor, χ_{TI}, was successfully developed for predicting TIs, given by the ratio of the energy difference between the conduction band and valence band at the special time-reversal-invariant momentum point of the Brillouin zone with over without the SOC effect. Using this descriptor, tens of new candidate TIs have been proposed. Most recently, a numerical multiscale approach, combining continuous $k \cdot p$ methods, tight-binding models, and *ab initio* calculations, was developed to derive the topological invariants of various materials via high-throughput screening [21]. Yet at the implementation level, these approaches of commendable success still have to rely on detailed band structure calculations based on first-principles.

In this work, we propose an efficient criterion that allows ready screening of potential topological materials, also using 3D TIs as prototypical examples. The criterion is inherently rooted in the presence or absence of band inversion induced by SOC, and is uniquely defined by a minimal number of two elemental physical

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properties of the constituent elements: the atomic number and Pauling electronegativity, rather than inputs from detailed band structure calculations within density functional theory (DFT). The validity and predictive power of the criterion is first demonstrated by rationalizing all the known topological or trivial insulators in the tetradymite and half-Heusler families. We further use this criterion to identify over ten new 3D TIs within the tetradymite family, including Sb_2Se_3, which so far had been widely regarded as a trivial insulator. The underlying materials design principles of our present study can also be extended to predictive discoveries of other classes of topological systems.

2. Models and methods

As mentioned above, our proposed criterion for ready screening of 3D TIs is uniquely defined by the atomic number and Pauling electronegativity of the constituent elements, and there are no needs for any inputs from DFT calculations. However, to check the validity and predictive power of the criterion, the band structures of nine binary tetradymites have been calculated by using the projector augmented-wave (PAW) method [22,23] within the framework of DFT. The code is implemented in the Vienna *ab initio* simulation package (VASP) [24–26]. The exchange-correlation energy is in the form of Perdew-Burke-Ernzerhof (PBE) with the generalized gradient approximation (GGA) [27]. The cutoff energy for the plane-wave basis set is 500 eV. A $15 \times 15 \times 15$ Monkhorst-Pack k mesh [28] is used for the Brillouin zone integrations. The atomic positions are fully relaxed until the magnitudes of the force acting on each atom become smaller than 0.01 eV/Å. The van der Waals (vdW) interactions are explicitly included in our calculations by adopting appropriate functionals [29,30], which was often ignored in previous work in this area. The SOC effect is taken into account, which is a very important factor to the TI nature. In addition, the method proposed by Fu and Kane [31–33] is adopted to calculate the Z_2 invariant, which is based on the analysis of parity for systems with inversion symmetry.

3. Results and discussions

We start by first considering topologically trivial and nontrivial insulators within the tetradymite family. In a milestone DFT study [34], by analyzing the presence or absence of band inversion surrounding the energy gap at the high-symmetry Γ point (Δ_Γ), it was identified that three of the four cross combinations of (Sb, Bi) and (Se, Te) are 3D TIs, while the remaining one, Sb_2Se_3, is not. On a detailed level, the gap is largely opened by the local chemical binding of the constituent elements and crystal field splitting, while the SOC tends to pull down the conduction band minimum and push up the valence band maximum in inducing the occurrence of the band inversion with an anti-crossing shape. As an order of magnitude criterion, to induce the band inversion it would be desirable if a TI candidate material has a larger SOC strength λ and a smaller Δ_Γ. The critical or transitional case would require that λ be comparable to Δ_Γ. It is thus reasonable to define a first trial criterion δ_0 as,

$$\delta_0 = \frac{\lambda}{\Delta_\Gamma}, \tag{1}$$

where λ can be expressed by an average value of the atomic SOC strength in a given compound material, and by definition, Δ_Γ is to be calculated without the SOC. According to the above discussions, it is expected that the δ_0 ratio should be larger than 1 for those TI candidates that are likely to exhibit topologically non-trivial nature. In this line of reasoning, we have assumed the systems of interest contain only one or an odd number of such high-symmetry (e.g., the Γ) point in the momentum space, as in the cases studied in the present work.

To verify the existence and validity of such a critical value, we have first double checked nine isostructural tetradymites A_2B_3 (A = As, Sb, Bi; B = S, Se, Te) with the rhombohedral structure. Among them, Bi_2Se_3, Bi_2Te_3 and Sb_2Te_3 have been predicted and/or experimentally observed to be 3D TIs in their ground states [34,35], while As_2Te_3 and Sb_2Se_3 were found to be trivial insulators in their ground states, but become 3D TIs if proper strain is applied [36,37]. Using the present δ_0 criterion based on accurate DFT calculations, we have analyzed their topological properties by calculating the corresponding Z_2 invariants. The results are presented in Fig. 1, plotted as a function of δ_0 defined in Eq. (1). It can be seen that the δ_0 criterion successfully predicts the three experimentally known 3D TIs of Bi_2Se_3, Sb_2Te_3 and Bi_2Te_3, because their δ_0 values are obviously larger than 1 (also see Table 1). More importantly, the δ_0 criterion also predicts two new 3D TIs, As_2Te_3 and Sb_2Se_3, which were believed to be trivial insulators in earlier studies [34,37]. Such apparent discrepancy can be attributed to the lacking of vdW interactions in their DFT calculations. Taking As_2Te_3 as an example, the substantial effect of the vdW interaction results in a smaller Δ_Γ , thereby making δ_0 to be larger than 1. All the details surrounding the vdW interactions are given in the Supplementary Data. Here we also note that, in future experimental realizations of these newly predicted 3D TIs, proper substrates should be invoked to promote the systems to grow into the above-mentioned layered structure.

So far we have demonstrated the successes of the simple δ_0 criterion in correctly predicting the known 3D TIs within the tetradymite family. Nevertheless, similar to the two earlier approaches [20,21], this criterion still depends on electronic structure calculations to obtain Δ_Γ. To conceptually go beyond this standing limitation, we note that, in principle, the SOC strength λ is proportional to the atomic number, while the band gap of a compound is closely related to the electronegativity difference of the constituent atoms [38,39]. In terms of the average atomic number (Z) of the formula unit and the Pauling electronegativity difference ($\Delta\chi$) of the constituent elements, we can modify Eq. (1) as,

$$\delta = \frac{a_0 \alpha^2 \beta Z}{\Delta\chi}, \tag{2}$$

where a_0 is a scaling constant, and we have introduced the fine-structure constant (α) and Rydberg constant (β) to make δ to be dimensionless. For the binary tetradymites A_2B_3, the average atomic number is given by,

$$Z = \frac{2Z_A + 3Z_B}{5}, \tag{3}$$

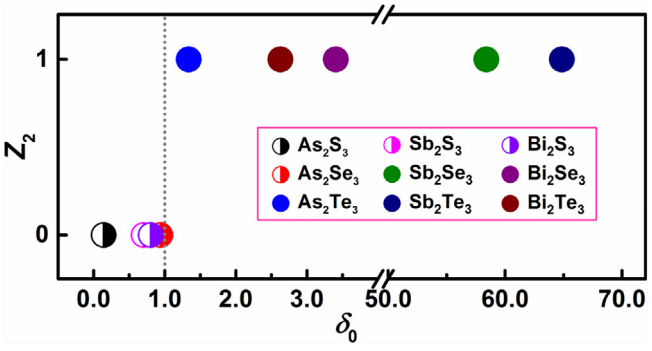

Fig. 1. (Color online) The Z_2 invariant as a function of the δ_0 ratio for nine binary tetradymites. The dashed lines indicate critical value of δ_0.

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