Prediction of topological insulating behavior in inverse Heusler compounds from first principles

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ABSTRACT

The topological band structures of the X₂YZ inverse Heusler compounds have been investigated by the first-principle calculations. The results of this study clearly indicate that a large number of inverse Heusler compounds naturally exhibit an inverted band structure. We found that, similar to the half-Heusler family, these inverse Heusler compounds can realize the topological insulating state under hydrostatic or uniaxial lattice expansion. Importantly, most of these compounds possess a negative formation energy, which makes them more suitable in material growth and could easily achieve the topological insulating behavior by alloying or proper strain.

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1. Introduction

Topological insulator (TI) is of a new class of materials, which has a full insulating gap in the bulk but contains topologically protected conducting states on edges or surfaces [1-4]. The surface states are chiral and inherently robust to external perturbations, which could be useful for future technological applications in spintronics and quantum computing as well [5,6]. Since the first two-dimensional (2D) TI with quantum spin-Hall effect was predicted in HgTe [7,8], a large number of three-dimensional (3D) TIs have also been proposed theoretically [9-20]. Very recently, the investigations on TIs have also gained great success experimentally: not only in binary compounds, but also the gapless half-Heusler TIs have been confirmed to possess metallic surface electronic state experimentally [21,22]. Most importantly, it was proposed that in the half-Heusler family the topological insulator allows the incorporation of superconductivity and/or magnetism [23].

In particular, the earlier computations [13,14] have proposed that the band structures near the Fermi level in the half-Heusler compounds exhibit almost the same behavior with those of CdTe and HgTe. They argue that the half-Heusler and zinc-blende lattice possess the same crystal symmetry with space group F43m (No. 216). In full-Heusler compounds, however, there exists a family of materials called inverse Heusler possessing the F43m crystal symmetry as well [24,25]. In Fig. 1, we show a comparison between inverse Heusler and zinc-blende structure. In general, the former can be considered as four interpenetrating face-centered-cubic (fcc) lattices, in which the X atoms occupy A(0,0,0) and B(1/4,1/4,1/4) sites, the Y, Z atoms site C(1/2,1/2,1/2) and D(3/4,3/4,3/4) respectively in Wyckoff coordinates. Similarly, the zinc-blende structure contains two inequivalent atoms, which occupy two nearest sites (C and D). We can make an analogous consideration with the previous studies [13,14], the inverse Heusler structure thus can be considered as two hybridized interpenetrating zinc-blendes (actually, a diamond and a zinc-blende structure), which are X–X and Y–Z. It is therefore very important to study whether the topologically nontrivial phase can be exploited in inverse Heusler compounds with 18 valence electrons.

In this work, by performing a systematic investigation on the band topology of some given inverse Heusler compounds, we will show that a large number of potential TIs are waiting for exploit in this rich inverse Heusler family. Our deduction and theoretical basis are mostly judged by making a comparison with the well studied topologically nontrivial binary compound HgTe [7,8]. Similar to the half-Heusler TI, we also find that the topological insulating behavior in these inverse Heusler compounds is sensitive to the variation of lattice constant and also tetragonal uniaxial strain.

2. Computational details

The band-structure calculations were performed using full-potential linearized augmented plane-wave method [26], implemented in the package WIEN2K [27]. Experimental lattice constants are used when available, and others are obtained by minimizing the total energy using generalized gradient
approximation of Perdew–Burke–Ernzerhof 96 including SOC \[28\]. A converged ground state was obtained using 5000 \( k \) points in the first Brillouin zone and set \( K_{\text{max}} \times R_{\text{MT}} = 8.0 \), where \( R_{\text{MT}} \) represents the muffin-tin radius and \( K_{\text{max}} \) is the maximum size of the reciprocal-lattice vectors. The muffin-tin radius of different elements used in calculations are generated by the system. Moreover, wave functions and potentials inside the atomic sphere are expanded in spherical harmonics up to \( l = 10 \) and 4, respectively.

3. Results and discussions

In Fig. 2, we illustrate the calculated band structures of CdTe and HgTe with those of \( Y_2\text{RuPb} \) and \( Sc_2\text{OsPb} \). For clarity, we will divide into two groups for comparison: CdTe (Fig. 2a) and \( Y_2\text{RuPb} \) (Fig. 2b), HgTe (Fig. 2c) and \( Sc_2\text{OsPb} \) (Fig. 2d), respectively. The band structure of \( Y_2\text{RuPb} \) is very similar to that of CdTe, which exhibits natural band ordering (s-like \( \Gamma_6 \) states (red lines) lie above the p-like \( \Gamma_8 \) states (blue lines)) and opens a direct gap at the \( \Gamma \) point, indicating them just be trivial semiconductors. However, the band structures of HgTe and \( Sc_2\text{OsPb} \) possess an inverted band order, in which the \( \Gamma_6 \) state sits below the \( \Gamma_8 \) state. Meanwhile, the valence and conduction bands away from the \( \Gamma \) point are well separated without crossing each other. Such band inversion only occurs once throughout the Brillouin zone and therefore, HgTe and \( Sc_2\text{OsPb} \) are both topologically nontrivial phases in their ground states.

We next perform a systematic investigation of the band topology of the \( X_2YZ \) (\( X = Sc, Y, La; Y = Ru, Re, Os; Z = Sb, Pb, Bi \))
compounds, which are all proposed to form the inverse Heusler structure. Here we define $\Delta E = E_{T8} - E_{H8}$ as topological band inversion strength, which would be positive for topologically trivial case and negative for topologically nontrivial phase. We have summarized the $\Delta E$ as a function of the lattice constant for these compounds in Fig. 3a. It can be found that most of them exhibit a negative $\Delta E$, indicating the TI candidate. While there are also some cases like Sc$_2$ReSb and La$_2$OsPb that the Fermi level visibly cuts the conduction or the valence bands, we can only call them topological semiconductors with a positive $\Delta E$. For Y$_2$RuPb, Sc$_2$RuPb and La$_2$RuPb, they are just trivial semiconductors with a positive $\Delta E$. As mentioned before, the inverse Heusler structure can be considered as two hybridized interpenetrating zinc-blende sublattices: X–X and Y–Z. It is clear that the inverse Heuslers will possess a positive $\Delta E$ with Y–Z forming the Y–Z zinc-blende, the on the other hand, if Os–Pb, Re–Bi or Re–Sb constitutes the Y–Z zinc-blende, the compounds are prone to be topologically nontrivial. That is to say, the sign of $\Delta E$ is dominated by the Y–Z zinc-blende and X–X one only contributes as a fine tuning of the value. The reason can be explained as follows: the atoms in Y–Z zinc-blende are much heavier than those in X–X and thus introduce a dominant drastic interplay of the SOC, which plays a vital role in the band inversion mechanism [13,14,19].

In order to investigate the experimental synthesis possibilities of the compounds, we have calculated the formation energy using the formula: $E_{fam} = E_{cal} - m \times E_X - n \times E_Y - p \times E_Z$, in which the $E_{cal}$ is the energy of the inverse Heusler compounds under their equilibrium lattice constant, and $E_X$, $E_Y$ and $E_Z$ represent the energy when X/Y/Z crystallized in pure metals, respectively. The coefficients $m$, $n$, $p$ show the proportions of X, Y, Z in the inverse Heusler formula. In Fig. 3b, we show the formation energy of the compounds as a function of the lattice constant. The result exhibits that all the compounds have a negative formation energy, and some are even lower than those of HgTe and CdTe. From the view of the calculated formation energy, we can conclude that the inverse Heusler compounds in our work are promising to be synthetized for experimental characterization.

It is well known that the topological insulating behavior of half-Heusler compounds is not only influenced by interplay of the SOC (mainly produced by heavy atoms), but also by the degree of hybridization (controlled by the lattice) [13,14]. Thus the trivial insulators can be converted to topological nontrivial phases by applying proper hydrostatic expansion. In our inverse Heusler compounds, for example, a 1.8% expansion of the lattice could convert the trivial insulator Y$_2$RuPb (Fig. 4a) into a nontrivial topological insulator (Fig. 4b). Conversely, the Sc$_2$OsPb (Fig. 4c) with inverted band order in its native state will transform into a topological trivial phase (Fig. 4d) under a 2.1% hydrostatic compression. These behaviors are quite similar to those in half-Heusler and chalcopyrite families [13–15,19]. However, we would like to point out that the topological band order in inverse Heuslers appears more sensitive to the variation of lattice constant according to our calculations; nearly all the Heusler compounds studied in our work can make a band order turn under a less than 3% lattice distortion, which may be meaningful in practice TI design and application.

Similar to the half-Heusler compounds, these topologically non-trivial inverse Heusler compounds are not naturally insulating, because the two couples of bands with $\Gamma_8$ symmetry are protected by the cubic symmetry and degenerate together at the $\Gamma$ point. However, the subbands with $\Gamma_8$ symmetry can be opened and form a direct gap by proper strain or doping engineer [11–13,19], which will drive the zero-gap semiconductor into a real TI phase. In order to demonstrate this mechanism in our inverse Heusler TIs, we apply a ±5% uniaxial strain with a constant volume to Sc$_2$OsPb system along the [001] direction. The results are shown in Fig. 5. As a result, the fourfold degeneracy of the $\Gamma_8$ states is broken and forms a gable band gap at Fermi level. Interestingly, the system exhibits different responses upon the uniaxial compression and expansion: when introduced a compression (Fig. 5a), the conduction and valence bands are no longer overlapped and the system becomes an insulator with inverted band order retained, while upon expansion (Fig. 5b) it remains a semimetal as before. The result is well consistent with our previous calculations for half-Heusler LaPtBi [29]. Indeed, a recent search model for possible TIs has suggested that the variational ‘descriptor’, namely ‘strain’ can be associated with the robustness or the feasibility of the TI state [30].
We have shown by the first-principle calculations that a large number of inverse Heusler compounds exhibit an inverted band order naturally and are promising to realize the topological insulating order. We argue that the band structures near the Fermi level are determined by the X–X and Y–Z zinc-blende sublattices jointly, in which the Y–Z dominates the sign of topological band inversion strength \( \Delta E \) and X–X just contributes as a fine tuning.

We also found that the band topology is sensitive to the variation of lattice constant and uniaxial strain. Importantly, all of the inverse Heusler compounds possess a negative formation energy making them more suitable in material growth and could easily achieve the topological insulating behavior by proper strain. With the example of Sc₂OsPb, we show how the gapless system can be driven by uniaxial strain into a topological insulating state.

**Fig. 4.** Band structures of Y₂RuPb and Sc₂OsPb. Y₂RuPb (a) without and (b) with a 1.8% hydrostatic expansion, Sc₂OsPb (c) without and (d) with a 2.1% hydrostatic compression. Here, the red and blue lines represent the subbands with \( \Gamma_6 \) and \( \Gamma_8 \) symmetry, respectively. The application of a hydrostatic expansion in Y₂RuPb causes the \( \Gamma_6 \) states to jump below the \( \Gamma_8 \) states, and leads to a nontrivial topological phase. In contrast, the application of a hydrostatic compression in Sc₂OsPb causes the \( \Gamma_6 \) states to jump above the \( \Gamma_8 \) states, and leads to a trivial topological phase. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

**Fig. 5.** Band structure of Sc₂OsPb under uniaxial strain with constant volume along [001] direction, with a reduction the \( c/a \) ratio by 5% in (a), and an increase in the \( c/a \) ratio by 5% in (b).

### 4. Conclusion

We have shown by the first-principle calculations that a large number of inverse Heusler compounds exhibit an inverted band order naturally and are promising to realize the topological insulating order. We argue that the band structures near the Fermi level are determined by the X–X and Y–Z zinc-blende sublattices jointly, in which the Y–Z dominates the sign of topological band inversion strength \( \Delta E \) and X–X just contributes as a fine tuning. We also found that the band topology is sensitive to the variation of lattice constant and uniaxial strain. Importantly, all of the inverse Heusler compounds possess a negative formation energy making them more suitable in material growth and could easily achieve the topological insulating behavior by proper strain. With the example of Sc₂OsPb, we show how the gapless system can be driven by uniaxial strain into a topological insulating state.
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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.commatsci.2012.12.013.

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