Topological material discovery with symmetry indicators

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Background and Purpose

Recently, the mathematical concept “Topology” attracts many physicists. There are a lot of researches to find new materials with non-trivial topology.

In order to distinguish topological nature, generally we need to calculate topological invariants, but it is difficult to evaluate it by definition. Though, under the crystalline symmetry, we can easily simulate the value of topological invariant itself or the remainder of integer topological number from the information of the crystalline symmetry. The first example of application is the Fu-Kane formula. This formula enables us to compute the topological invariant of an insulator with time-reversal and inversion symmetry from the set of parity eigenvalues of Bloch function of occupied bands. Recently, this idea is extended to 230 types of space group as the symmetry indicators [1,2] which evaluates the topological nature of materials from irreducible representations of Bloch wavefunctions at high symmetry k-points. Furthermore, we can compute the symmetry indicator of real materials by using irreducible representations obtained from the density-functional-theory (DFT) calculation. This method is useful for data-driven material exploration, which results in the discovery of various kinds of non-magnetic topological materials [3-5].

In this research, we focus on the magnetic topological materials. The magnetic topological materials attract many researchers because of their unique physical phenomena. For example, magnetic topological insulators and magnetic Weyl semimetals exhibit anomalous quantum Hall effect, and skyrmion crystals exhibit topological quantum Hall effect. For the application, energy efficient memory devices can be achieved by controlling the magnetic structure in magnetic topological materials, such as domain wall and skyrmion. These materials should be important as the candidates of various devices.

However, candidates of magnetic topological materials are not enough yet. The purpose of our research is that we theoretically produce the candidates of magnetic topological materials, based on the symmetry indicators for magnetic space groups [2].

Method

There are two steps to determine the topological nature of materials. One is the density functional theory (DFT) calculation by AM, and the other is the calculation of symmetry indicators by SO. Here we explain the details one by one.

1. Computations for the band structure and characters by using DFT calculations

DFT calculation is the widely known as *ab initio* calculation method to evaluate the eigenenergy and wavefunctions of electrons in the material. In this research, we use this DFT calculation to obtain the band structure and characters of Bloch wavefunctions in the band.

We choose Quantum ESPRESSO as the numerical program package of DFT calculation. The data of crystalline and magnetic structure of target materials are downloaded from the database called MAGDATA [6,7] and converted into the form of the input file for QUANTUM ESPRESSO by SeeK-path [8,9]. For efficiency and fewer mistakes, we use the tools to take the calculation of various kinds of materials by QUANTUM ESPRESSO.

We compute the band structure of target materials by original functions of QUANTUM ESPRESSO. Then we see the band dispersion around the Fermi energy, and distinguish the conducting property: insulator, semimetal, or metal. Symmetry indicators are kept invariant unless the band structure is deformed across the Fermi energy. However, for the metal which has a lot of bands around the Fermi energy, the combination of occupied bands from the calculation cannot be so reliable thus we exclude the metal from the list of candidates in advance.

For calculations of symmetry indicators, the characters of Bloch wavefunctions must be computed from QUANTUM ESPRESSO output. The character $\chi_{n\vec{k}}$ is defined as

$$\chi_{n\vec{k}}(g) = \langle \psi_{n\vec{k}} | \, g \, | \psi_{n\vec{k}} \rangle$$

where n is the energy level of the band, $\vec{k}$ is the momentum, g is the symmetry operator, and $\psi_{n\vec{k}}$ is the Bloch wavefunction of the *n*-th band at $\vec{k}$. The data such as Bloch wavefunctions and symmetry operations are converted from QUANTUM ESPRESSO output into the input files of our original program "*qeirreps*" [13] by using qe2respack. The characters of Bloch wavefunctions are produced by *qeirreps* and used for the calculation of symmetry indicators by the Mathematica program.

2. Calculations of symmetry indicators

Symmetry indicator enables us to diagnose the topological nature of materials from the irreducible representations of occupied bands for the little group $G_{\vec{k}}$ of the space group G. The definition is as follows.

At first, the vector b should be defined as the number of each irreducible representation in occupied bands for each high symmetry momentum:

$$b = \left(n_{\vec{k}_1}^{\alpha_1}, n_{\vec{k}_1}^{\alpha_2}, \ldots, n_{\vec{k}_2}^{\beta_1}, n_{\vec{k}_2}^{\beta_2}, \ldots\right)$$

where α and β are the indices for irreducible representations, k is the high symmetry momentum, n is the number of the irreducible representation α at the momentum k. This vector b is invariant under the adiabatic deformation of the band structure so that it enables us to distinguish the phase of matter. We denote the set of the vectors which appear in the band structures under the particular symmetry as {BS}. For the convenience, the vector b are allowed to have minus integer for its elements n. In this setting, {BS} forms the vector space[1].

On the other hand, we consider the atomic insulator as the topologically trivial insulator. Atomic insulators are models constructed by putting isolated atoms to the sites with respect to the crystalline symmetry. Atomic insulators are insulating and topologically trivial by definition. We introduce the vector space {AI} as the set of b for atomic insulators. By definition, {AI} is the subset of {BS}. We say the band structure is topological when its vector b does not belong to {AI}.

The symmetry indicator group X_{BS} is defined as the quotient group:

$$X_{\mathrm{BS}} = \{\mathrm{BS}\}/\{\mathrm{AI}\}$$

For all magnetic space groups, the dimension of {BS} and {AI} are known to be same and X_{BS} becomes the finite group $\mathbb{Z}_{n_1} \times \mathbb{Z}_{n_2} \times \cdots \times \mathbb{Z}_{n_N}$[2]. If the vector b of the band structure responds to the trivial element of X_{BS}, that band can be adiabatically connected to the band structure of atomic insulator without gap closing at high symmetry momenta[2]. On the other

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[1] These are not truly vectors. However, we abuse this terminology for simplicity.

[2] When the band can be adiabatically connected to the atomic insulator without gap closing at high symmetry momenta, this situation does not necessary mean the band structure is topologically trivial. For example, in the two-

hand, if the band structure is related to the non-trivial element of X_{BS}, that band has the topological nature. Here, the species of topological phase depend on the symmetry and the element of X_{BS}.

In our research, we determined the irreducible representation, the symmetry indicator, and finally the topological nature of band structures from the result of DFT calculations. This process was done by using our own Mathematica program.

Result and future perspective

We searched the band structure and the topological nature of around 40 kinds of magnetic materials among Mn and Fe compounds. Most of them were metal or topologically trivial insulator/semimetal. We finally found that $CaFe_2As_2$ had the non-trivial topological nature. The physical properties of $CaFe_2As_2$ are shown in following figures: the crystalline and magnetic structure, the Brillouin zone, and the band structure produced by the DFT calculation with spin orbit coupling and Hubbard U=2.

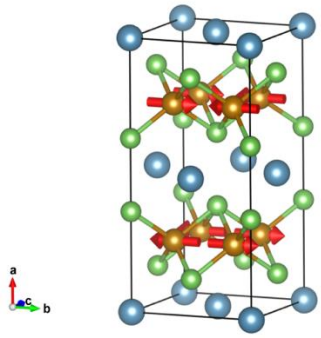

Figure 1 The crystalline and magnetic structure of $CaFe_2As_2$ [6,7]

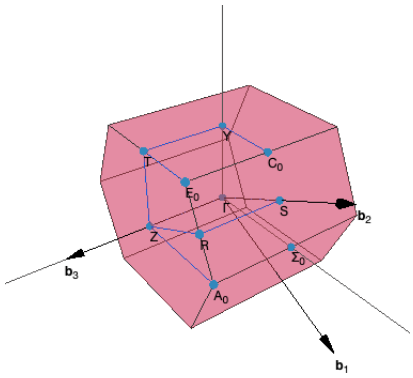

Figure 2 The Brillouin zone of $CaFe_2As_2$ [8,9]

dimensional system with n-fold rotation symmetry without time reversal symmetry, X_{BS} is correspond to the modulo n of the Chern number C. The Chern number $C = n$ is the topologically non-trivial value but related to the trivial element of X_{BS}.

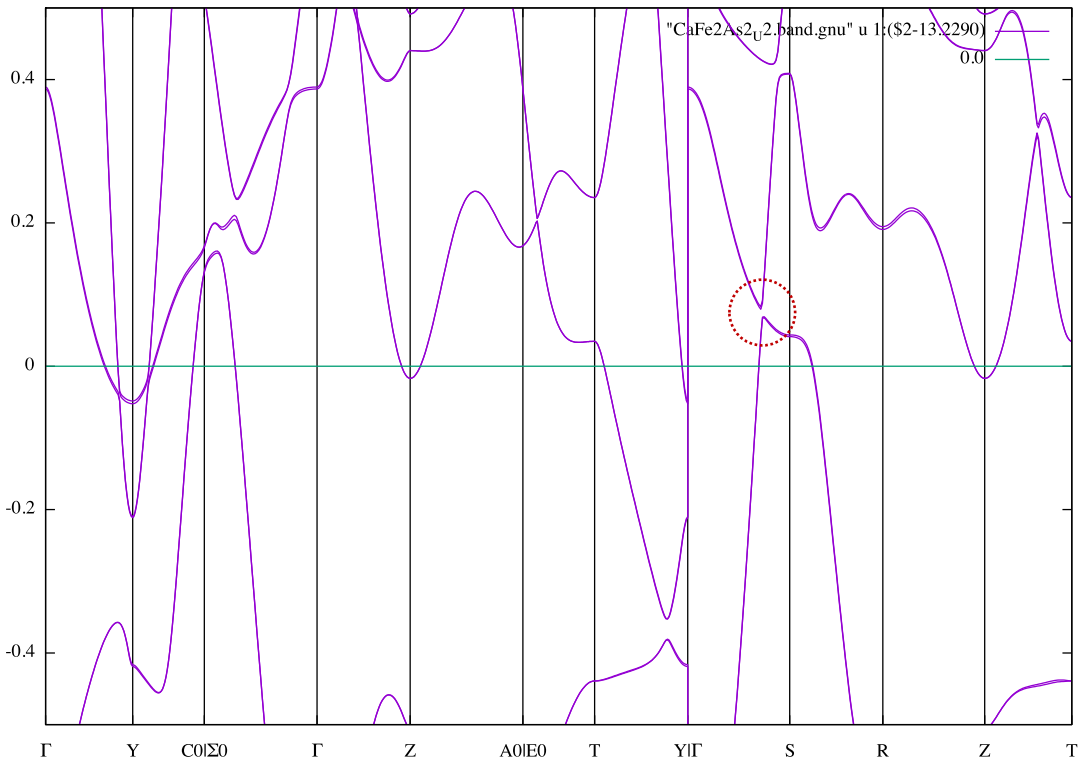

Figure 3 The band structure of $CaFe_2As_2$

The space group symmetry of $CaFe_2As_2$ is C_Amca (MSG 64.480) that contains inversion symmetry. In the system with inversion symmetry, we can diagnose the topological nature from the inversion parities of the occupied bands [2] as

$$\mu = \sum_{\vec{k}\in \mathrm{TRIM}} \frac{n_{\vec{k}}^{+} - n_{\vec{k}}^{-}}{2}.$$

where $n_{\vec{k}}^{\pm}$ is the number of the occupied bands with the eigenvalue ± 1 for the inversion symmetry. For $CaFe_2As_2$, this indicator represents that this material is Weyl semimetal (higher-order topological insulator) if its band structure is gapless (gapful). The bands are two-fold degenerated in the whole Brillouin zone, because of the Kramers degeneracy originated from the inversion and time-reversal symmetry.

There is the linear dispersion around the Fermi energy on the Γ-S line (enclosed by red circle in Figure 3). It seems to be Weyl node which is the feature of the Weyl semimetal. We numerically calculated the monopole charge around this momentum point [10], which resulted in the trivial value for the calculation with spin-orbit coupling. Although, there is the possibility that two Weyl nodes with opposite signs of monopole charge are overlapped by the Kramers degeneracy. Then we also calculated for the case without spin-orbit coupling, which proposed that the spin-up band structure had Weyl node with the non-trivial value of monopole charge.

As a result, in the limit of no spin-orbit coupling, $CaFe_2As_2$ is the Dirac semimetal with the doubly degenerated Weyl nodes on the Γ-S line. These Weyl nodes come from the spin-up and down band structures, respectively. The spin-orbit coupling causes the hybridization of the spin-up and down band structures and makes the small gap on Weyl node. Then finally the material becomes the higher-order topological insulator with the spin-orbit coupling. In fact, our calculation with spin-orbit coupling produces the small band gap at this momentum point.

We expect $SrFe_2As_2$ and $BaFe_2As_2$ have similar band structures with $CaFe_2As_2$. They have the possibility to be more proper candidates of Weyl semimetal or higher-order topological insulator, by the different values of Hubbard-U and spin-orbit coupling. However, these materials have difficulty for numerical calculations because of their strongly correlated effects. Actually, the value of magnetization obtained by our calculation are 2-3 times larger than the experimental results. For these materials, the advanced numerical method itself is the important research topic to evaluate the physical properties accurately [11,12] hence our results based on usual DFT calculation is not reliable anymore.

On the other hand, we release our own Fortran program "*qeirreps*" for computing the character list of Bloch wavefunctions from the output files of QUANTUM ESPRESSO [13]. The original functions of QUANTUM ESPRESSO cannot determine the representations for the system with nonsymmorphic space group symmetries. For the system with nonsymmorphic space groups, "*qeirreps*" can calculate the character list from the output data of QUANTUM ESPRESSO. We can use the result of *qeirreps* to see the symmetry indicator of materials. This program *qeirreps* is an open-source program and everyone can access for the computation of symmetry indicators by using this. Some similar programs have been already released for WIEN2k and VASP [14,15] but these software require paid license. In contrast, QUANTUM ESPRESSO is the open-source program and has more demand for users. Our program *qeirreps* does not respond fully to the magnetic system as in the previous researches [4,14]. In future update, we would like to support the all magnetic materials.

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