A phonon irreducible representations calculator

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The irreducible representation of band structure is important for physical properties. Based on phonopy and recently developed SpaceGroupIrep package, we developed a package PhononIrep, which can get the band irreducible representation for arbitrary \( \mathbf{k} \) point at first-principles level. As an application, we for the first time predict the cubic crossing Dirac point can exist in conventional crystal phonon systems \( \text{X}_3\text{Y} \) (\( \text{X} = \text{Ti}, \text{Nb}, \text{Ta}, \text{Y} = \text{Au}, \text{Sb} \)). We hope this package will facilitate phonon research in the future.

Introduction. Group representation theory is a powerful tool to investigate novel properties in theoretical physics. In solid state physics, representation theory can not only greatly simplify the first-principles calculations [1], but also can help us to investigate the connectivity of energy bands in crystals [2], the lattice vibrations [3], the selection rules [4], the effective Hamiltonians [5–9]. Very recently, researchers use group representation theory to search topological insulators and emergent particles [10–22].

At present, there are four packages Irrep, irvsp, qeirrepr and SpaceGroupIrep [23–26] focus on space group irreducible representations. The data in first three packages are compatible with BCS website [27], and in SpaceGroupIrep is compatible with Bradley and Cracknell’s (BC) book [4]. However, those packages are mainly used for electronic systems not for the phonon systems except irvsp can determine the phonon irreducible representations in tight-binding level [25]. In addition, the phonopy package can only give the irreducible representations of \( \Gamma \) point but not for general \( \mathbf{k} \) point [28, 29].

In this work, we make an expansion of SpaceGroupIrep, namely, PhononIrep, which can get the band irreducible representation for arbitrary \( \mathbf{k} \) point in first-principles level. The only necessary inputs of PhononIrep is the force constant from Phonopy and the structure of unit cell. As an example, we for the first time proposed the cubic crossing Dirac point (CCDP) in phonon system. PhononIrep will stimulate further studies on the fascinating properties for phonon system.

Method. In current version of phonopy, the irreducible representations can be obtained for only \( \Gamma \) point, but for general \( \mathbf{k} \) points phonopy can only calculate the character \( \chi_{\mathbf{k}}(Q) \) of \( \mathbf{k} \). Then \( \chi_{\mathbf{k}}(Q) \) can be completely reduced by orthogonality relationships

\[
\chi_{\mathbf{k}}(Q) = \sum_{l} a_l \chi_{l}(Q)
\]

where \( Q = \{R|t\} \) with rotation part \( R \) and translation part \( t \) is an element of the little group of \( \mathbf{k} \), \( l \) is the band index.

The procedures for calculating the irreducible representations of phonon are as follow (Fig.(1)): Start form a unit cell structure. First, use finite displacement method or density functional perturbation theory to get the second-order force constants. Second, use phonopy to calculate the characters of \( Q \). Third, convert the character to BC convention. This step is done by convert the input cell and irreducible representation’s labels into BC conventions [24]. Finally, calculate the irreducible representations by comparing the obtained characters and the character table of the little group of \( \mathbf{k} \).

Comparing to irvsp, PhononIrep have the following advantages:

- Calculate phonon irreducible representations from first-principles results directly, there is no need to convert the force constants into tight-binding Hamiltonian.
- There is no need to standardise the unit cell structure manually by phonopy or spglib before the first-principles calculation.

![Flowchart of calculating the irreducible representations of phonon.](image-url)
The label of irreducible representations is given for both Mulliken [30] and Γ label [4, 31, 32].

The inputs of PhononIrep are clearly and easy to set. In principle, only the structure and force constants information are sufficient to get the irreducible representations. One can run the following short script to get the irreducible representations of phonon system:

```python
calcPhononIrep("supercell") -> size of supercell, 
"unitcell" -> path of unit cell file, 
"force" -> path of force constants file, 
"kset" -> list of k points, 
"symprec" -> \( \epsilon_1 \), 
"degeneracytolerance" -> \( \epsilon_2 \), 
"showRep" -> True or False
```

calcPhononIrep is the function to get the irreducible representations which have six options. "supercell" is a list including three integers represent the size of the supercell in phonon calculation. "unitcell" is the full path of unit cell structure file. "force" is the full path of unit force constants file. "kset" is a \( n \times 3 \) array that tell PhononIrep which \( k \) points will be calculated. "showRep" tell PhononIrep whether to show the results in a Grid form. "symprec" and "degeneracytolerance" are tolerance of determining the symmetry and the degeneracy in phononpy, the default values of "symprec" and "degeneracytolerance" are 0.0001, which are fine for most of calculations (see Appendix A for installation and running of PhononIrep).

As an example, we use PhononIrep to calculate irreducible representations of diamond (Fig.(2)). The irreducible representations are consistent with Ref. [3]. Therefore, PhononIrep can avoid tedious derivation and get the result directly.

**An application — CCDP.** The CCDP with cubic leading order dispersion on the four body diagonals of the cube Brillouin zone is merged by two C-4 Weyl points. According to the encyclopedia of emergent particles [13], CCDP can only appear in cubic structure with space group 218, 220, 222, 223, 230 in spinless system. The real materials which host CCDP has not been reported before. Here, we take Ti\(_3\)Au as an example to confirm the CCDP can exist in phonon system. Ti\(_3\)Au host simple cubic structure. Ti atom locate at 6\( c \) Wyckoff position and Au locate at 2\( a \) Wyckoff position. The crystal structure of Ti\(_3\)Au is shown in Fig. 3(a). After the force constants is calculated, we can immediately get the irreducible representations of \( R(\frac{1}{2}, \frac{1}{2}, \frac{1}{2}) \) by PhononIrep. As shown in table I, three exist three six-fold, one and one four-fold degeneracy points. The four-fold point is a CCDP which locate at 5.80 THz formed by \( R_2 \oplus R_3 \). One can easily verify the degeneracies by checking the phonon dispersions [see Fig. 3(b)].

Within the calculated irreducible representations, we can do a more detailed analysis of this material. A set of generators and representation matrices of the symmetry

![FIG. 2. Phonon spectrum of diamond. The little representations are labelled by Γ notation.](image1)

![FIG. 3. (a) Crystal structures of X\(_3\)Y. (b) The phonon dispersion of Ti\(_3\)Au along high symmetry path, the CCDP marked within the green rectangles are zoomed in around the zero frequency. (c-d) The frequency difference of CCDP along different path, along path \( R-X \) the dispersion are quadratic, along path \( R-Γ \) the dispersion are cubic.](image2)

| band  | frequency | dim | irreducible representations |
|-------|-----------|-----|-----------------------------|
| start | end       |     |                             |
| 1     | 6         | 3.74 | \( H \) \( R_4 \)          |
| 7     | 12        | 4.81 | \( H \) \( R_4 \)          |
| 13    | 16        | 5.80 | \( 1F \oplus 2F \) \( R_2 \oplus R_3 \) |
| 17    | 22        | 7.34 | \( H \) \( R_4 \)          |
| 23    | 24        | 7.46 | \( E \) \( R_1 \)          |
where \( \Gamma_{i,j} = \sigma_i \otimes \sigma_j \) and \( \sigma_i(i = 0, 1, 2, 3) \) are identity matrix and three Pauli matrices.

We then can construct the \( k \cdot p \) model of \( R \) point that satisfies the symmetry constrains

\[
P(Q)H(k)P^{-1}(Q) = H(Qk)
\]

where \( P(Q) \) is the representation matrix of the little group for \( R \). Consider the Hamiltonian up to third order. The obtained model can be written as

\[
H = \begin{pmatrix}
\lambda^* \alpha k_x^2 + \alpha k_y^2 + \lambda \alpha k_z^2 & \beta k_x k_y k_z \\
-\beta k_x k_y k_z & \lambda^* \alpha k_z^2 + \lambda \alpha k_y^2 + \alpha k_x^2
\end{pmatrix}
\]

\[
H_{CCDP}^{223,R} = \varepsilon + ck^2 + \begin{pmatrix} 0 & H \\ H^\dagger & 0 \end{pmatrix}
\]

Here, \( \lambda = e^{\frac{2\pi i}{3}} \), \( k^2 = k_x^2 + k_y^2 + k_z^2 \), \( \varepsilon, c \) are real parameters and \( \alpha, \beta \) are complex parameters. Notice in four body diagonals of the cube Brillouin zone, the leading order dispersion in the band splitting is cubic, i.e. for \( q^2 = k_x^2 = k_y^2 = k_z^2 \), the spectrum of this Hamiltonian (4) is

\[
E(q) = \varepsilon + 3c q^2 \pm |\beta| q^3
\]

where the \((-\cdots)\) represent the lower (higher) two bands of CCDP. However, in six face diagonals the dispersion are quadratic, e.g. along \( R-X \) high symmetry, \( p = k_x = k_z, k_y = 0 \), the spectrum of this Hamiltonian (4) is

\[
E(q) = \varepsilon + 2c q^2 \pm |\beta| q^3
\]

which is different from the conventional cubic Dirac point composed of two C-3 Weyl fermions [33]. Actually the CCDP composed of two C-4 Weyl fermions, when the time reversal symmetry or inversion is broken, the CCDP will split into two C-4 Weyl fermions with opposite topological charge [34]. Such difference will play a dominant role in the physical properties of the system [35].

For the phonon calculations of diamond and Ti\textsubscript{3}Au, the second-order force constants is based on the density functional perturbation theory within the phonony and VASP package [1, 28]. A \( 2 \times 2 \times 2 \) supercell of the conventional unit cell with \( 3 \times 3 \times 3 \) \( k \)-mesh and 300 eV for energy cutoff are applied. It should be emphasized that CCDP always exist in proper frequency for materials with space group 218, 220, 222, 223, 230. Therefore, we partially list a series of materials \( X_3Y \) (\( X=\text{Ti}, \text{Nb}, \text{Ta}, Y=\text{Au}, \text{Sb} \)) in Materials project [36] (as shown in Appendix B) which have same symmetry with Ti\textsubscript{3}Au. Such materials must have CCDP in \( R \) point of Brillouin zone.

In conclusion, we have developed a package PhononIrep which can get the irreducible representations of phonon system. As an application, it confirms that the CCDP can exist in realistic phonon systems. Our work provide a useful tool to analyse the phonon system. An exciting direction for future is to use PhononIrep to search for topological materials.

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Appendix A: Installation and running of PhononIrep

Before install PhononIrep, users should install SpacegroupIrep and Phonopy. Then, the steps of installing PhononIrep is exactly same as installing MagneticTB [9]. Unzip the "PhononIrep-main.zip" file and copy the PhononIrep directory to any of directory in SPath, e.g. copy the PhononIrep directory to FileNameJoin[{SysUserBaseDirectory, "Applications"}]. Then one can use the package after running Needs["PhononIrep"]]. The version of Mathematica should ≥ 11.2. The source code of PhononIrep are available on https://github.com/zhangzeyingvv/PhononIrep.

One can run the following script to get the irreducible representations of R point for Ti3 Au

1 SetEnvironment["PATH" -> Environment["PATH"] <> ";", "D:\Anaconda3\Library\bin"];
2 RegisterExternalEvaluator["Python", "D:\Anaconda3\python.exe"];
3 FindExternalEvaluators["Python"];
4 Needs["PhononIrep"];

1 SetEnvironment["PATH" -> Environment["PATH"] <> ";", "D:\\Anaconda3\\Library\\bin"];
2 RegisterExternalEvaluator["Python", "D:\\Anaconda3\\python.exe"];
3 FindExternalEvaluators["Python"];
4 Needs["PhononIrep"];}
Line 1-3 is to set the Python environment in Mathematica. Users have to specify the Python installation path in `RegisterExternalEvaluator` and `SetEnvironment`, see [https://support.wolfram.com/52852](https://support.wolfram.com/52852) for detail. Line 4 is to load PhononIrep package. See main text for a detailed description of line 5-8. The output of above script is:

To directly compare the irreducible representations between the output of PhononIrep and the results in Ref. [13], one can use `checkLGrepLabel` in `SpaceGroupIrep` to get the abstract groups' irreducible representations label:

the output of above command is:

### Appendix B: List of $X_3 Y$

TABLE II. List of mp-ID, lattice constant and ICSD for $X_3 Y$ (X=Ti, Nb, Ta, Y=Au, Sb), the structure data are available on [www.materialsproject.org](http://www.materialsproject.org).

| Material | mp-ID | lattice constant (Å) | ICSD(s) |
|----------|-------|----------------------|---------|
| Ti$_3$Au | mp-1786 | 5.113 | 612419 612405 58605 612417 612420 612418 |
| Nb$_3$Au | mp-2752 | 5.256 | 612199 58557 612192 612186 612203 612198 612185 |
| Ta$_3$Au | mp-569249 | 5.246 | 58599 |
| V$_3$Au | mp-839 | 4.881 | 612456 58612 612451 612459 |
| Ti$_3$Sb | mp-1412 | 5.217 | 651683 96137 651684 106035 651685 43356 657034 |
| Nb$_3$Sb | mp-2053 | 5.312 | 645347 76572 645349 190178 645357 645352 |
| Ta$_3$Sb | mp-541 | 5.303 | 52310 651601 |
| V$_3$Sb | mp-1555 | 4.935 | 651731 52330 106037 651717 |