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YPHON: A package for calculating phonons of polar materials☆

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ABSTRACT

In our recent works, we have developed a mixed-space approach within the framework of direct method for the first-principle calculation of phonon properties. It makes full use of the accuracy of the force constants calculated in the real space and the dipole–dipole interactions in the reciprocal space, making the accurate phonon calculation possible with the direct method for polar materials. In this paper, an efficient C++ implementation of the mixed-space approach, YPHON, is provided as open source, including demos and Linux scripts for extracting input data to YPHON from the output of VASP.5. The functions of the current package include the calculations of: (1) the phonon dispersions; (2) the phonon density of states; (3) the neutron scattering section weighted phonon density of state; (4) the phonons of the high symmetry structure using the force constants from low symmetry structure; (5) the phonon dispersions of random alloys; and (6) the analysis of the vibrational modes using the point group theory.

Program summary

Program title: YPHON

Catalogue identifier: AETS_v1_0

Program summary URL: http://cpc.cs.qub.ac.uk/summaries/AETS_v1_0.html

Program obtainable from: CPC Program Library, Queen's University, Belfast, N. Ireland

Licensing provisions: Standard CPC licence, http://cpc.cs.qub.ac.uk/licence/licence.html

No. of lines in distributed program, including test data, etc.: 567815

No. of bytes in distributed program, including test data, etc.: 9763594

Distribution format: tar.gz

Programming language: C++, Linux scripts.

Computer: Linux systems with a g++ or C++ compiler.

Operating system: Linux.

RAM: Ranges from a few Mbytes to a few Gbytes, dynamically depending on the system size.

Classification: 7.8.

External routines: GSL—the GNU Scientific Library (GSL) is a numerical library for C and C++ programmers. VASP.5 or later for the calculations of force constants and dielectric constants and Born effective charge for polar materials.

Nature of problem:
This package has the purpose of computing accurately phonon properties of polar materials within the small displacement approach.

Solution method:
Mixed-space approach to the vibration-induced dipole–dipole interaction.

☆ This paper and its associated computer program are available via the Computer Physics Communication homepage on ScienceDirect (http://www.sciencedirect.com/science/journal/00104655).

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Running time:
In the scale of a common Linux command.

1. Introduction

Currently there are essentially two methods in use for the first-principles calculations of phonon frequencies: (i) the linear response theory and (ii) the direct approach. The direct approach is also referred to as the small displacement approach, the supercell method, or the frozen phonon approach. The linear response theory directly evaluates the dynamical matrix through the density functional perturbation theory without the approximation of the cutoff in neighboring interactions. In comparison, the direct method is conceptually simple and straightforward to implement with a new potential or approximation, such as strong correlation [1,2], the explicit treatment of the semi-core electrons, and the hybrid potential [3,4]. Other advantage of the direct approach is that the phonon frequencies at the exact wave vectors that are commensurable with the supercell are calculated exactly with no further approximation. However, most of the previous implementations of the supercell approach are unable to accurately handle the long range dipole–dipole interactions when calculating phonon properties.

Our mixed-space approach [5,6] has resolved the long-standing problem within the direct approach on how to account for the effects of vibration-induced dipole–dipole interactions on the phonons of an ionic crystal or a polar material. By explicitly taking into account the effects of vibration-induced dipole–dipole interactions between periodic supercells, it has made it possible to determine accurately the lattice dynamics of polar materials within the direct approach. We demonstrate that the vibration-induced dipole–dipole interactions lead to a constant contribution to the interatomic force constant in real space. The approach has been demonstrated within our group [7–12] and outside our group [13,14] for a variety of materials. The rest of this paper is organized as follows:

- Section 2: The mixed-space approach.
- Section 3: Installation of YPHON.
- Section 4: Contents in YPHON package.
- Section 5: Command line options and files used by YPHON.
- Section 6: Demos of YPHON.
- Section 7: Quick start to run YPHON.
- Section 8: Summary.
- Section 9: Troubleshooting.

2. The mixed-space approach

2.1. Notations in the formulism

Below we summarize the notations in the theoretical formulations of the following subsections.

$\hbar$	Planck constant
j, k	Index for atoms in the primitive unit cell
μ_j	Atomic mass of atom j
α, β	Index the Cartesian direction of x, y, z.
$\mathbf{a}$	3×3 matrix whose rows are made of the lattice vectors of the primitive unit cell.
V	Volume of the primitive unit cell.
M	Number of atoms in the primitive unit cell.
$\mathbf{S} = \mathbf{Na}$	Represent a supercell with $\mathbf{N}$ $\mathbf{a}$ 3×3 matrix made of integers defining the supercell in the unit of the lattice vectors of the primitive unit cell
N	Number of primitive unit cells within the supercell **S.**
$\mathbf{r}^j$	Atomic position of atom j within the primitive unit cell, $\mathbf{r}^j = r^j_\alpha \mathbf{a}_\alpha$ (Einstein notations).
P, Q	Index for the primitive unit cell in the supercell **S.**
$\mathbf{R}_P$	Position the Pth primitive unit cell within the supercell, $\mathbf{R}_P = P_\alpha \mathbf{a}_\alpha$ (Einstein notations).
$\mathbf{b}$	3×3 matrix made of the reciprocal lattice vectors of the primitive unit cell, $\mathbf{a}_\alpha \cdot \mathbf{b}^\beta = \delta_{\alpha\beta}$ where α and β label the rows (lattice vectors) of $\mathbf{a}$ or $\mathbf{b}$.
$\mathbf{q} = 2\pi q^\alpha \mathbf{b}_\alpha$	Wave vector (Einstein notations).
$\mathbf{q}_S$	Exact wave vector point satisfying the condition of $\mathbf{q_S} \cdot \mathbf{S}_\alpha = 2\pi * Integer$.
$C^{jk}_{\alpha\beta}(P, Q)$	Real-space interatomic force constants.
$u^j_\alpha(P)$	Cartesian displacement from its static position of the jth atom within the Pth primitive unit cell.
$D^{jk}_{\alpha\beta}(\mathbf{q})$	Dynamical matrix in the wave vector space.
ω	Phonon frequency.
$\mathbf{Z}^j$	3×3 matrix defining the Born effective charge tensor of the jth atom within the primitive unit cell.
$\boldsymbol{\varepsilon}^{(\infty)}$	3×3 matrix defining the macroscopic dielectric constant tensor.

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2.2. Summary of the phonon theory

The essential part of the phonon theory is to solve the eigenvalue problem of the vibrational Hamiltonian of a periodic crystal system in the harmonic approximation [15,16]:

$$H = -\frac{1}{2}\sum_{P}^{\infty}\sum_{j}^{M}\frac{\hbar^2}{\mu_j}\nabla^2 + \frac{1}{2}\sum_{P,Q}^{\infty}\sum_{j,k}^{M}\sum_{\alpha,\beta}^{3} C_{\alpha\beta}^{jk}(P,Q)u_{\alpha}^{j}(P)u_{\beta}^{k}(Q). \tag{1}$$

In principle, Eq. (1) cannot be solved directly due to its infinite dimension as indicated by the summation over the index P or Q indexing the positions of infinite number of primitive unit cells repeated in the infinite crystal space. One approach [16,17] is the **periodic supercell approach**, namely, the infinite crystal system is represented by an infinite exact repeating of a supercell **S** with finite size to fill the infinite crystal space, including values of the atom displacement $u_{\alpha}^{j}(P)$ due to vibration. As a result, one gets the accurate solution for a special set of vibrations at the exact wave vector point, $\mathbf{q_S}$, which satisfy

$$\mathbf{q_S}\cdot\mathbf{S}_{\alpha} = 2\pi * Integer. \tag{2}$$

The rationale is given as follows. Based on the repeating feature of the atomic displacements in the periodic supercell system, we can separate the P dependent part of $u_{\alpha}^{j}(P)$ in Eq. (1) by rewriting

$$u_{\alpha}^{j}(P) = u_{\alpha}^{j}\exp(i\mathbf{q_S}\cdot\mathbf{R}_P). \tag{3}$$

Substituting Eq. (3) into Eq. (1) together with Eq. (2), the phonon frequency can be calculated by finding the roots, $\omega^2(\boldsymbol{q})$, of the secular determinant with a dimension of $3M$ [16,17]:

$$\det|D_{\alpha\beta}^{jk}(\mathbf{q}) - \omega^2(\mathbf{q})| = 0 \tag{4}$$

where the dynamical matrix

$$D_{\alpha\beta}^{jk}(\mathbf{q_S}) = \sum_{P}^{N} e^{i\mathbf{q_S}\cdot\mathbf{R}_P}\frac{1}{\sqrt{\mu_j\mu_k}}\Phi_{st}^{\alpha\beta}(P,0) \tag{5}$$

and

$$\Phi_{st}^{\alpha\beta}(P,0) = \sum_{S}^{\infty} C_{\alpha\beta}^{jk}(P+S,0). \tag{6}$$

$\Phi_{st}^{\alpha\beta}(P,0)$ is called the accumulative interatomic force constants [18] since it represents the summation of $C_{\alpha\beta}^{jk}(P,0)$'s, counting P and all its images resulted from repeating the supercell **S** over the whole crystal space.

The above procedures provide the accurate solutions for the phonon frequencies at those nonzero exact **q** points, i.e., all $\mathbf{q_S}$'s except for the point of $\mathbf{q}\to 0$ for polar material. For the calculation of phonon frequency at a general **q** point, the usual practice [15] is to generalize the usage of Eqs. (4) and (5) to the case where **q** is away from the $\mathbf{q_S}$ for the already defined supercell **S**, as

$$D_{\alpha\beta}^{jk}(\mathbf{q}) = \sum_{P}^{N} e^{i\mathbf{q}\cdot\mathbf{R}_P}\frac{1}{\sqrt{\mu_j\mu_k}}\Phi_{st}^{\alpha\beta}(P,0) \tag{7}$$

and

$$\det|D_{\alpha\beta}^{jk}(\mathbf{q}) - \omega^2(\mathbf{q})| = 0. \tag{8}$$

2.3. Polar materials

For a polar material or an ionic insulator, when the periodic supercell approach is employed, one has to distinguish two cases in calculating phonon frequencies:

(i) At the nonzero $\mathbf{q_S}$ — the lattice vibration does not result in macroscopic electric fields; and
(ii) Away from the $\mathbf{q_S}$ — the lattice vibration results in a macroscopic electric field for the atoms within the predefined supercell, which could invalidate the accurate application of the Fourier interpolation by Eqs. (7) and (8).

The effects of macroscopic electric fields on phonons is not yet well accounted for in previous implementations of the supercell method [18,19] until the mixed-space approach [5,6] developed by the present authors. This is due to the fact [20] that the condition of zero macroscopic electric fields required by the periodic boundary conditions of the periodic supercell approach is not satisfied by the previous implementation of the direct approach. Using the mixed-space approach [5,6], at an arbitrary wave vector **q,** it is proved that it simply requires recasting the accumulative interatomic force constants in Eq. (7) as

$$\Phi_{\alpha\beta}^{jk}(P,0) = \phi_{\alpha\beta}^{jk}(P,0) + \varphi_{\alpha\beta}^{jk}(\mathbf{q}) \tag{9}$$

where $\phi_{\alpha\beta}^{jk}(P,0)$ is the accumulative interatomic force constants calculated by the first-principles supercell method [19]. $\varphi_{\alpha\beta}^{jk}$ accounts for the contribution due to the vibration-induced dipole–dipole interaction as

$$\varphi_{\alpha\beta}^{jk}(\mathbf{q}) = \frac{1}{\sqrt{\mu_j\mu_k}}\frac{4\pi}{NV}\frac{(\mathbf{Z}^j\cdot\mathbf{q})_{\alpha}(\mathbf{Z}^k\cdot\mathbf{q})_{\beta}}{(\mathbf{q}\cdot\boldsymbol{\varepsilon}^{(\infty)}\cdot\mathbf{q})}. \tag{10}$$

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3. Installation of YPHON

3.1. Operating systems

YPHON works well in the Linux environment.

3.2. General software requirements

YPHON is written using C++. The enclosed precompiled executable binaries should work for most Linux systems. If one wants to recompile YPHON, the only external package required is the GNU Scientific Library (GSL) which is a numerical library for C and C++ programmers. If you are just interested in the phonon dispersions, phonon density-of-states (PDOS), and the neutron scattering cross section weighted PDOS — so-called generalized phonon density-of-states (GPDOS), this is enough.

Otherwise, for best performance, it is strongly recommended that the gnuplot be installed (most possibly, gnuplot is already installed in your system).

3.3. Scientific software requirements

(A) VASP.5 or later: VASP is a package for performing ab-initio quantum-mechanical molecular dynamics (MD) using pseudopotentials and a plane wave basis set. On how to get VASP, please go to http://cms.mpi.univie.ac.at/vasp/vasp/vasp.html.
(B) Otherwise, any first-principles codes which can calculate the static energy and force constants as long as you can manage to organize output data of that first-principles code into the YPHON input formats (text formats as detailed later).

3.4. Get and unpack YPHON

Download the package, a gzipped tar file "`yphon.tar.gz`", followed by unpacking the package and installing YPHON by the following series of Linux commands:

```
tar zxf yphon.tar.gz
cd YPHON; make
```

Notes: Mostly you do not need to recompile the codes with "`make`". The precompiled executable binaries enclosed with the package work quite well in a number of computer centers and until the present time we have not seen any exceptions.

For `csh` user, the Linux command search `PATH` should be modified by inserting the line (assume that `$HOME/YPHON` is the location of the `YPHON` folder after unpacking the package. Of course, you can move the `YPHON` folder into wherever you wish) below into the proper location within the `.cshrc (.tcshrc or.cshrc.ext)` file, depending on the specific management of your Linux system.

```
set PATH = (. $HOME/YPHON $PATH)
```

For `bsh` user, the Linux command search `PATH` should be changed by inserting the two lines below into the `.bash_profile (.bashrc)` file

```
PATH=.: $HOME/YPHON:$PATH
export PATH
```

4. Contents in YPHON package

`Yphon` – A C++ code to calculate the phonon density of states and the phonon dispersion based on the force constants calculated by VASP.5. `Yphon` is the central code of the YPHON package doing the phonon calculation.

`Ycell` – A C++ code to build supercell in the VASP.5 `POSCAR` format.

`vasp_fij` – A Linux script to collect the force constant matrix from the `OUTCAR` and `CONTCAR` files of VASP.5 or the Hessian matrix from the `vasprun.xml` file of VASP.5. `vasp_fij` collects the force constants into the `superfij.out` file which is the input to `Yphon`.

`vaspfijxml` – A C++ code to convert the Hessian matrix from the `vasprun.xml` file into the force constant matrix. `vaspfijxml` is only called by `vasp_fij` when it sees the vasprun.xml file in the current folder.

`vasp_BE` – A Linux script to collect the data of Born effective charge and high frequency dielectric tensors from the `OUTCAR` and `CONTCAR` files of VASP.5. `vasp_BE` collects the dielectric constant and Born effective charge tensor into the `dielecfij.out` file which is the input to `Yphon` when calculating polar materials.

`pos2s` – A Linux script for making the `symmetry.mode` file for the vibrational mode analysis, in terms of irreducible representations by group theory, for phonons at the Γ point, and the `Rotation.sym` file for the space group operations according to the symmetry of the crystal.

`Ymode` – A C++ code to support the script `pos2s`.

`findsym`, `smodes` – precompiled executable binaries from ISOTROPY which is a software package applying group theoretical methods to the analysis of phase transitions in crystalline solids. Using `pos2s` implicitly uses these two modes of the ISOTROPY software. When using `pos2s` results in published papers, you may include a reference which contains the following information:
H.T. Stokes, D.M. Hatch, and B.J. Campbell, (2007). ISOTROPY, stokes.byu.edu/isotropy.html.

`poscar2findsym.rb` – Python module from the *phonopy* package by Atsushi Togo. Using `pos2s` implicitly uses this Python module. When using `pos2s` results in published papers, you may cite the paper:

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"First-principles calculations of the ferroelastic transition between rutile-type and CaCl2-type SiO2 at high pressures", Atsushi Togo, Fumiyasu Oba, and Isao Tanaka, Phys. Rev. B, 78, 134106 (2008).

`Ydemo` – A folder contains several subfolders as demos of the YPHON package.

5. Command line options and files used by YPHON

The usage of YPHON follows the command style of the Linux operating system. YPHON are composed of a number of Linux commands. A YPHON command is followed by a series of keywords and parameters. Different keywords and parameters are separated by a space character in the command. All keywords in YPHON commands are case sensitive.

5.1. Ycell

Usage: `Ycell [options]`
`<yourprimitiveposcarfile >yoursupercellposcarfile`
Where `yourprimitiveposcarfile` is a file from the standard input and `yoursupercellposcarfile` is a file for the standard output. Both files are in the VASP.5 `POSCAR` format.
The options are:
`-bc2`
Make a supercell by a kind of doubling the primitive unit cell.
`-bc3`
Make a supercell by a kind of tripling the primitive unit cell.
`-bc4`
Make a supercell with a size of four times of the primitive unit cell.
`-mat matrix3x3`
Make a supercell by transforming the primitive unit cell with a 3×3 matrix (9 parameters), for example '`-mat 2 -2 0 2 2 0 0 2`'
`-ss` n
Make a $n \times n \times n$ supercell of the primitive unit cell.

5.2. Yphon

Usage: `Yphon [options] <superfij.out`
Where `superfij.out` is the name of the file created by `vasp_fij` containing crystal structure as well as force constant information. Be default, `Yphon` calculate the PDOS which is outputted into the file of `vdos.out`.
The options are:

Note: You may see many other options in the `Yphon` source code not explained below. The unexplained options are either for the debug purpose or in the test stage.

`-nq` *`nqx nqy nqz`*
Instruct `Yphon` to calculate the PDOS using a `nqx` × `nqy` × `nqz` mesh in the wave vector space. The default values of *`nqx nqy nqz`* are those can provide ∼3, 000, 000 phonon frequencies. Do not be afraid! The high efficiency of `Yphon` makes the calculation of millions of frequencies done in minutes.
`-DebCut f`
Instruct `Yphon` to fit the PDOS using the Debye expression in the form of $C\omega^2$ for the phonon frequency range lower than the given frequency f. The typical value of f should be around 0.1–1.0 (in the unit of THz), depending your system and the used **q** mesh. One should always plot the calculated PDOS and inspect the region at around the frequency f for the smoothness of the curve to decide the proper value of f. This option is useful for the case if one wants to use the PDOS to calculate the heat capacity or Debye temperature at temperature lower than ∼ 10 K, in particular, for superconductor etc.
`-Born dielecfij.out`
Consider the vibration-induced dipole–dipole interaction (called LO–TO splitting in the literature) in the calculation, where `dielecfij.out` is the name of the file created by `vasp_BE` containing all information about Born effective charge and high frequency dielectric tensors.
`-pdis` *`yourdisfile`*
This option instructs `Yphon` to calculate the phonon dispersion instead of the PDOS. *`yourdisfile`* is a file defining the directions for the dispersion calculation, see the subsection of **"File for dispersion calculation"** for instruction on how to prepare the file *`yourdisfile`*.
`-pvdos`
Calculate also the generalized PDOS (GPDOS, the neutron scattering cross section weighted PDOS), followed Zbiri et al. [21], i.e., GPDOS $= \sum_i \frac{\sigma_i}{\mu_i}$pDOS$_i$ where σ_i and pDOS$_i$ represent respectively the atomic scattering cross section [22] and the partial phonon density-of-states projected into the individual atoms. The results are saved in the file pvdos.out.
`-bvec`
For polar materials only, use lattice vectors the primitive unit cell from the `dielecfij.out` file to define the wave vector space.
Note: sometimes, the primitive lattice vectors in the `dielecfij.out` file may be different from those defined in `superfij.out`.
`-noNA -nof`
Use together with `-bvec`. Use the primitive lattice vectors from the `dielecfij.out` file to define wave vector space, but not consider the effects of the vibration-induced dipole–dipole interaction.

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Note: In some cases it makes sense to calculate the phonons of a high symmetry structure using the force constant matrix calculated under a low symmetry structure. For this purpose, one can use a different lattice structure through the `dielecfij.out` file from that from the `superfij.out` file. The option `-noNA` is for the case of conductor where no dielectric information available. The option `-nof` tells `Yphon` to ignore some error check steps such as the atomic type mismatch in the case of SQS calculation explained below.

`-sqs`

Used together with `-noNA -nof`. Calculate the phonon dispersions of a random alloy with respect to the wave vector space of the ideal lattice, by averaging over the force constants calculated using a special quasirandom structure (SQS). The detailed formulism can be found in our previous publication [23].

`-mall`

Make an average of the dynamical matrix over all the primitive unit cells within the supercell, i.e, replacing Eq. (7) by the following equation:

$$D^{jk}_{\alpha\beta}(\mathbf{q}) = \frac{1}{N}\sum_{P,Q}^{N} e^{i\mathbf{q}\cdot(\mathbf{R}_P-\mathbf{R}_Q)}\frac{1}{\sqrt{\mu_j\mu_k}}\Phi^{\alpha\beta}_{st}(P,Q). \tag{11}$$

When there are no atomic position distortions within the supercell with respect to the primitive unit cell, this option does not change the results, except for taking more computer time. This option is only useful when the supercell is allowed to relax or to calculate the phonon dispersions of high symmetry structure using the force constants calculated from a low symmetry structure. The detailed formulism can be found in our previous publications [1,11].

`-thr2 parameter`

Define the threshold on how to determine the atomic position relation between the high symmetry structure and the low symmetry structure. Care should be taken in this kind of calculation. One should gradually increase the value of the `parameter` from 0.01 to 0.15.

`-Mass` *yourmassfile*

This option tells `Yphon` to redefine the atomic mass, being required for the SQS phonon dispersion calculation. The context in the *yourmassfile* file contains lines like

```
Cu 96.8975
Au 96.8975
```

`-Gfile symmetry.mode`

This option tells `Yphon` to make the vibrational mode analysis, in terms of irreducible representations for phonons at the Γ point. The `symmetry.mode` file must be made by using the enclosed script `pos2s`, containing the information about the group theory representations of different vibrational modes.

`-Rfile Rotation.sym`

In some cases it makes sense to calculate the phonons of a high symmetry structure using the force constant matrix calculated under a low symmetry structure. This option tells `Yphon` to "restore" the symmetry of the high symmetry crystal using

$$\Phi^{\alpha\beta}_{st}(\mathrm{HS}) = \frac{1}{S}\sum_{r=1}^{S}\mathbf{O}_r^{-1}\Phi^{\alpha\beta}_{st}(\mathrm{LS})\mathbf{O}_r \tag{12}$$

where $\Phi^{\alpha\beta}_{st}(\mathrm{HS})$ represents the force constant matrix of high symmetry structure (for instance, the paramagnetic phase or the high temperature cubic structure of a Perovskite), $\Phi^{\alpha\beta}_{st}(\mathrm{LS})$ the force constant matrix calculated under the low symmetry structure (for example, antiferromagnetic structure or the low temperature tetragonal or rhombohedral structure of a Perovskite), $\mathbf{O}_r$ the space group operation of the high symmetry structure, and S the number of $\mathbf{O}_r$'s. The `Rotation.sym` file must be made by using the enclosed script `pos2s`, containing the rotation operations of the high symmetry structure from group theory.

Note: Caution should be taken for this kind of calculation. In particular, large supercell is needed to delimit the effects of symmetry loss of the supercell.

`-plot`

Instructs `Yphon` to display the plot in the terminal using gnuplot for one to check the calculated results.

`-expt exp01.dat`

Instructs `Yphon` to plot the experimental data contained in the file "`exp01.dat`" together with the calculations.

5.3. pos2s

Usage: `pos2s Symmetry.pos`

`Symmetry.pos` is a file produced by `Yphon` before running the `pos2s` script.

5.4. Files used by YPHON

5.4.1. *superfij.out file*

`superfij.out` is the central input file of `Yphon` through the standard input stream `stdin` in the Linux environment. It contains information about the lattice vectors of the primitive unit cell, the lattice vectors of the supercell, the atomic positions in the supercell, and the matrix of the negative values of the force constants. You do not have to prepare this file by hand if you use VASP.5 since the enclosed script `vasp_fij` can make this file by extracting data from the output files of VASP.5, namely, `CONTCAR`, `OUTCAR`, and `vasprun.xml`.

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The format of the data contained in `superfij.out` are illustrated below:

```
0.0000000000 2.1060000000 2.1060000000
2.1060000000 0.0000000000 2.1060000000
2.1060000000 2.1060000000 0.0000000000
   0.000000    4.212000    4.212000
   4.212000    0.000000    4.212000
   4.212000    4.212000    0.000000
16 8
Direct
   0.00000000   0.00000000   0.00000000   Mg
   0.00000000   0.00000000   0.50000000   Mg
   0.00000000   0.50000000   0.00000000   Mg
   0.00000000   0.50000000   0.50000000   Mg
   0.50000000   0.00000000   0.00000000   Mg
   0.50000000   0.00000000   0.50000000   Mg
   0.50000000   0.50000000   0.00000000   Mg
   0.50000000   0.50000000   0.50000000   Mg
   0.25000000   0.25000000   0.25000000   O
   0.25000000   0.25000000   0.75000000   O
   0.25000000   0.75000000   0.25000000   O
   0.25000000   0.75000000   0.75000000   O
   0.75000000   0.25000000   0.25000000   O
   0.75000000   0.25000000   0.75000000   O
   0.75000000   0.75000000   0.25000000   O
   0.75000000   0.75000000   0.75000000   O
-10.944494    0.000000     0.000000   1.260786    3.229127    0.000000
1.260786    0.000000    3.229127    -0.377911   0.000000    0.000000   -
0.377911    0.000000    0.000000     1.260786   0.000000   -3.229127
1.260786   -3.229127    0.000000     1.697668   0.000000    0.000000   -
0.151592    0.000000    0.000000     0.788701   0.000000    0.000000
0.788701    0.000000    0.000000     1.065411   0.000000    0.000000
1.065411    0.000000    0.000000     0.788701   0.000000    0.000000
0.788701    0.000000    0.000000    -0.151592   0.000000    0.000000
    0.000000   -10.944494   0.000000     3.229127   1.260786    0.000000
0.000000    -0.377911   0.000000     0.000000   1.260786   -3.229127
0.000000     1.260786   3.229127     0.000000  -0.377911    0.000000   -
3.229127     1.260786   0.000000     0.000000   1.697668    0.000000
0.000000    -0.151592   0.000000     0.000000   0.788701    0.000000
0.000000     1.065411   0.000000     0.000000   0.788701    0.000000
0.000000     0.788701   0.000000     0.000000   1.065411    0.000000
0.000000     0.788701   0.000000     0.000000  -0.151592    0.000000
...
```

Where,

line 1–line 3: lattice vectors of the primitive unit cell
line 4–line 6: lattice vectors of the supercell
line 7: Number of atoms in supercell, and number of primitive unit cell in the supercell (serve as error check)
line 8: the same meaning as VASP
line 9 — line of 8+"Number of atoms in supercell": internal atomic positions in the supercell, which again have the same meaning as VASP and the data after these lines are the negative values of the force constant matrix.

5.4.2. *`dielecfij.out` file*

When one wants to consider the effects of vibration-induced dipole–dipole interaction on phonons, `dielecfij.out` is required as the command line option of `-Born dielecfij.out` in running `Yphon. dielecfij.out` is a file containing the information about the lattice vectors of the primitive unit cell, the atomic positions, the high frequency dielectric tensor, and the Born effective charge tensor. You do not have to prepare this file by hand if you use VASP.5 since the enclosed script `vasp_BE` can make this file by extracting data from the `OUTCAR` of VASP.5, if you have setting of "`LEPSILON =.T.`" and "`NSW=0`" in the `INCAR` file. The format of the data contained in `dielecfij.out` are illustrated below:

```
 0.000000      2.106000      2.106000
 2.106000      0.000000      2.106000
 2.106000      2.106000      0.000000
0.0000000000000000   0.0000000000000000   0.0000000000000000   Mg
0.5000000000000000   0.5000000000000000   0.5000000000000000   O
         3.147       0.000       0.000
         0.000       3.147       0.000
```

```
         0.000      0.000      3.147
ion   1
   1     1.96085     0.00000     0.00000
   2     0.00000     1.96085     0.00000
   3     0.00000     0.00000     1.96085
ion   2
   1    -1.96142     0.00000     0.00000
   2     0.00000    -1.96141     0.00000
   3     0.00000     0.00000    -1.96141
```

where,

line 1–line 3: lattice vectors of the primitive unit cell.

line 4 — line of 3+"Number of atoms in the primitive unit cell": internal atomic positions in the primitive unit cell, which again have the same meaning as VASP. The lines followed the atomic positions are the dielectric constant tensor and the Born effective charge tensor.

Note: the calculation of the dielectric properties should be performed using the primitive unit cell separately.

5.4.3. *vdos.plt file*

`vdos.plt` is a gnuplot script made by `Yphon` when no "`-pdis`" command line option is defined. Its purpose is to use gnuplot to plot the PDOS, typically using the Linux command "`gnuplot -persist vdos.plt`". The content of this file typically contains:

```
reset
#set terminal postscript landscape enhanced color "Times_Roman" 20
set encoding iso_{8859}_{1}
set pointsize 1.2
set size 0.95,0.95

#set output "vdos.eps"

funit=1.000000
set xlabel "Frequency (THz)"
set ylabel "(THz^[19])"
plot 'vdos.out' using ($1*funit*1.e-12):($2/funit*1.e12) notitle w l lt -1
```

Note: the default unit for the phonon frequency in the "vdos.out" file is Hz. To convert into the THz unit, a convert factor of 1.e−12 is needed.

5.4.4. *vdos.out file*

`vdos.out` is a default output file of `Yphon` when no "`-pdis`" command line option is defined. It contains the PDOS data.

Note: The PDOS in `vdos.out` has been normalized to $3M$ where M is the number of atoms in the primitive unit cell. Also be careful that the unit in the `vdos.out` file for the frequency in is Hz. Therefore, if you want to compare the calculated phonon with experiment, you need the proper conversion factor.

A characteristic feature of `Yphon` is concerned with the PDOS calculation. `Yphon` does not follow the conventional Gaussian smearing approach to calculate the PDOS, since it is a headache for us on how to determine the value of the empirical Gaussian broadening parameter. Instead, `Yphon` uses the following convolution average to calculate the PDOS.

$$D(\omega) = \frac{3M}{N_\omega} \frac{1}{T(T+1)\Delta\omega} \sum_{n=0}^{T} [A(\omega + n\Delta\omega) - A(\omega + n\Delta\omega - T\Delta\omega)] \tag{13}$$

where $D(\omega)$ is the PDOS, N_ω the total number of phonon frequencies calculated by a uniform **q** mesh in the wave vector space, $A(\omega)$ the total number of phonon frequencies below the frequency ω, $\Delta\omega$ the frequency interval of the numerical expression of the PDOS, and T an integer playing the role of smoothening the PDOS. By default, `Yphon` calculate $D(\omega)$ with a mesh of 10,001 with $T = 40$. As mentioned in the Section 5.2, with the **q** mesh that provides $N_\omega = \sim 3000000$, the calculated $D(\omega)$ is rather reasonable from our experience.

Note: Using the option of "`-nq nqx nqy nqz`", `Yphon` is still affordable for even denser **q** mesh that can provide $\sim$ 30,000,000 frequencies if one wants more accurate PDOS.

5.4.5. *pvdos.out file*

`pvdos.out` is a output file of `Yphon` when the key "`-pvdos`" is defined in the command line. It contains the neutron scattering crossing section weighted phonon density of state (GPDOS). The format of the data contained in this file are:

column 1: phonon frequency (in the unit of THz).
column 2: PDOS (in the unit of THz^{-1}).
column 3: weight factor due to the different atomic neutron scattering crossing sections. Therefore, (column 2)*(column 3) in this file is the GPDOS.
column 4: not useful.
column 5: partial PDOSs of the atoms following their orders in the primitive unit cell.

Note: when using gnuplot, one can plot GPDOS by

```
plot "pvdos.out" using 1:($2*$3) w l.
```

5.4.6. vdis.plt file

vdis.plt is a gnuplot script made by Yphon when the "-pdis" command line option is defined. It is used by gnuplot to plot the phonon dispersions. After you run Yphon, you can always check the phonon dispersion plot in the screen using

```
gnuplot -persist vdis.plt
```

Particularly, if you delete the comment sign "#" in the vdis.plt file and then run it by "gnuplot vdis.plt", it will produce a postscript file almost ready for your publication.

5.4.7. vdis.out file

vdis.out is a file made by Yphon when the "-pdis" command line option is defined. It contains phonon dispersion data.

5.4.8. File for dispersion calculation

To calculate the phonon dispersion, one is needed to use the command line option of "-pdis *yourdisfile*" with Yphon. *yourdisfile* is a file defining the direction for the dispersion calculation, and typically its format is (from our unpublished calculation of GaAs)

```
0 0 0 0 0 .5 Gamma X 0 $1 2 0 $1 3
0 .5 .5 0 0 0 X Gamma 1 (1.-$1) 2 1 (1.-$1) 3 1 (1.-$1) 4
0 0 0 .25 .25 .25 Gamma L 2 (2*$1) 2 2 (2*$1) 3
.25 .25 .25 0 0 .5 L X 3 (2*$1) 2 3 (2*$1) 3 3 (2*$1) 4
0 0 .5 0 .25 .5 X W 4 (2*$1) 2 4 (2*$1) 3 4 (2*$1) 4
0 .25 .5 .25 .25 .25 W L 5 (2*$1) 2 5 (2*$1) 3 5 (2*$1) 4
```

Data columns 1–3: the reciprocal reduced coordinate of the starting **q** point along the dispersion path
Data columns 4–6: the reciprocal reduced coordinate of the end **q** point along the dispersion path
Data column 7: the label of the starting **q** point along the dispersion path
Data column 8: the label of the end **q** point along the dispersion path
Data column 9: multi sets of data each set containing three columns wherein the first column is the index of the data group (separated by two blank lines following the convention of the gnuplot) in the experimental data file, the second column tells gnuplot which column and how to transform the column into the **q** point following the convention of the gnuplot, and the third column tells gnuplot which column of experimental data will be used as the frequency data. This can help you save a lot of time if you can learn it. The calculated phonon dispersions are contained in the file of vdis.out.

Note: care should be taken about the suffix of the *yourdisfile* file. The suffix .fcc, .bcc, .hcp, and .tet2 are reserved for the fcc, bcc, hcp, and tetragonal crystals only. For these crystal, Yphon internally converts the **q** vector of the primitive unit cell into that of the conventional unit cell using the following C++ statements

```
double bcc[9] = {-1.,1.,1.,1.,-1.,1.,1.,1.,-1};
double fcc[9] = {0.,1.,1.,1., 0.,1.,1.,1.,0.};
double hcp[9] = {1., 0.,0.,1.,-1.,0.,0.,0.,1.};
double tet2[9] = {-1.,1.,1.,1.,-1.,1.,1.,1.,-1};
```

For the cases of bcc and fcc crystals, most neutron scattering data are reported with respect to the cubic conventional cell instead of the primitive unit cell. Yphon did the conversion internally, assumed that you defined the shape of the primitive unit cell of the fcc crystal in the POSCAR file as

```
0.5 .5
.5 0 .5
.5 .5 0
```

and you defined the shape of the primitive unit cell of the bcc crystal in the POSCAR file as

```
-.5 .5 .5
.5 -.5 .5
.5 .5 -.5
```

For the case of hcp crystal, Yphon assumed that you defined the shape of the primitive unit cell of the crystal in the POSCAR file as (note that c in the third line below is the relative lattice parameter in *c* direction)

```
0.8660254037844 -.5 0.
0.0000000000000  1. 0.
0.0000000000000  0. c
```

For the case of tetragonal crystal, Yphon assumed that you defined the shape of the primitive unit cell of the crystal in the POSCAR file as

```
1.0 0. 0.
0. 1.0 0.
0.5 0.5 c
```

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If you do not use the suffix .fcc, .bcc, .hcp, and .tet2 for the *yourdisfile* file, Yphon will define the direction of wave vector using the reciprocal lattice vector of the primitive unit cell. The reciprocal lattice vector is printed out in the screen as the last three lines like

```
-0.000000000000        -0.362182366065        -0.000000000000
-0.313659560187        -0.181091751042        -0.000000000000
-0.000000000000        -0.000000000000        -0.192811598003
```

You can refer these (only the direction is important) to define the direction of your phonon dispersion calculation.

For the dispersion calculation, we strongly recommend one refer the web site http://www.cryst.ehu.es/ for the definition of the KVEC, i.e., the k-vector types and Brillouin zones of the space groups.

6. Demos of YPHON

The demos given below assume that you are using VASP.5. For start, make a demo folder named such as "mYdemo".

Even you do not have VASP, you can still try YPHON demos since the demo subfolders contain the already calculated force constants and the dielectric quantities (except Mg and Cu_3Au since they are conductors) by us. You can just skip into the Sections 6.1.7, 6.1.8, 6.2.9 and 6.3 if you do not have VASP.

6.1. Magnesium

Make a folder "Mg" under the folder of "Ydemo" and go to the folder "Mg". You need to prepare the following files and submit your VASP.5 job.

6.1.1. POSCAR file

First, prepare the POSCAR file for the primitive unit cell, name the file as "POSCAR.prm", and copy/paste the following lines into it

```
Mg HCP
 1.00
    1.594090000000       -2.761040000000        0.000000000000
   -3.188170000000        0.000000000000        0.000000000000
    0.000000000000        0.000000000000       -5.186410000000
   Mg
    2
D
 0 0 0
 0.66666667 0.33333333 0.5
```

Then make the supercell using "Ycell -ss 2 <POSCAR.prm >POSCAR" which builds the supercell POSCAR file containing these lines:

```
Supercell by Yi Wang
1.00
         3.1881800000         -5.5220800000          0.0000000000
        -6.3763400000          0.0000000000          0.0000000000
         0.0000000000          0.0000000000        -10.3728200000
 Mg
 16
D
   0.0000000000    0.0000000000    0.0000000000 Mg
   0.0000000000    0.0000000000    0.5000000000 Mg
   0.0000000000    0.5000000000    0.0000000000 Mg
   0.0000000000    0.5000000000    0.5000000000 Mg
   0.5000000000    0.0000000000    0.0000000000 Mg
   0.5000000000    0.0000000000    0.5000000000 Mg
   0.5000000000    0.5000000000    0.0000000000 Mg
   0.5000000000    0.5000000000    0.5000000000 Mg
   0.3333333350    0.1666666650    0.2500000000 Mg
   0.3333333350    0.1666666650    0.7500000000 Mg
   0.3333333350    0.6666666650    0.2500000000 Mg
   0.3333333350    0.6666666650    0.7500000000 Mg
   0.8333333350    0.1666666650    0.2500000000 Mg
   0.8333333350    0.1666666650    0.7500000000 Mg
   0.8333333350    0.6666666650    0.2500000000 Mg
   0.8333333350    0.6666666650    0.7500000000 Mg
```

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6.1.2. INCAR file

Make a INCAR file containing the following lines:

```
EDIFF=1.d-6
PREC = A
ISMEAR = 1
SIGMA = 0.2
IBRION = 6
ISIF = 0
NSW=1
```

6.1.3. KPOINTS file

Make a KPOINTS file containing the following lines:

```
Magnesium
 0
G
 3 3 3
 0 0 0
```

6.1.4. POTCAR file

Get POTCAR by the Linux command (depending on the VASP pseudopotential file location in your system)

```
zcat /usr/global/msc/vasp/potpaw_PBE/Mg/POTCAR.Z >POTCAR
```

6.1.5. Run VASP.5

Run VASP interactively (the run can be finished in less than one minute) or make a PBS batch job script containing the following lines and then submit your job (depending on your system environment for batch job)

```
       #PBS -q debug
       #PBS -l nodes=1:ppn=8
       #PBS -S /bin/tcsh
       #PBS -j oe
       #PBS -l walltime=00:30:00
       setenv VSPCMD "eval ‘/usr/common/nsg/opt/Modules/3.2.7/bin/modulecmd tcsh load
vasp/5.2.12‘; mpirun -np 8 /usr/common/usg/vasp/5.2.12/bin/vasp"
       cd $PBS_O_WORKDIR
       $VSPCMD
```

6.1.6. Collect the results and input to Yphon

After your job is done, first delete the not used files using "`rm CHG DYNMAT IBZKPT WAVECAR CHGCAR DOSCAR EIGENVAL REPORT XDATCAR PCDAT`", then type the following Linux command sequentially, namely to get the force constant, type

```
vasp_fij
```

You will see a file named "`superfij.out`" which contains lines like

```
1.5940900000 -2.7610400000 0.0000000000
-3.1881700000 0.0000000000 0.0000000000
0.0000000000 0.0000000000 -5.1864100000
   3.188180   -5.522080    0.000000
  -6.376340    0.000000    0.000000
   0.000000    0.000000   -10.372820
16 8
Direct
   0.00000000  0.00000000   0.00000000  Mg
   0.00000000  0.00000000   0.50000000  Mg
   0.00000000  0.50000000   0.00000000  Mg
   0.00000000  0.50000000   0.50000000  Mg
   0.50000000  0.00000000   0.00000000  Mg
   0.50000000  0.00000000   0.50000000  Mg
   0.50000000  0.50000000   0.00000000  Mg
   0.50000000  0.50000000   0.50000000  Mg
   0.33333333  0.16666667   0.25000000  Mg
   0.33333333  0.16666667   0.75000000  Mg
   0.33333333  0.66666667   0.25000000  Mg
   0.33333333  0.66666667   0.75000000  Mg
   0.83333333  0.16666667   0.25000000  Mg
```

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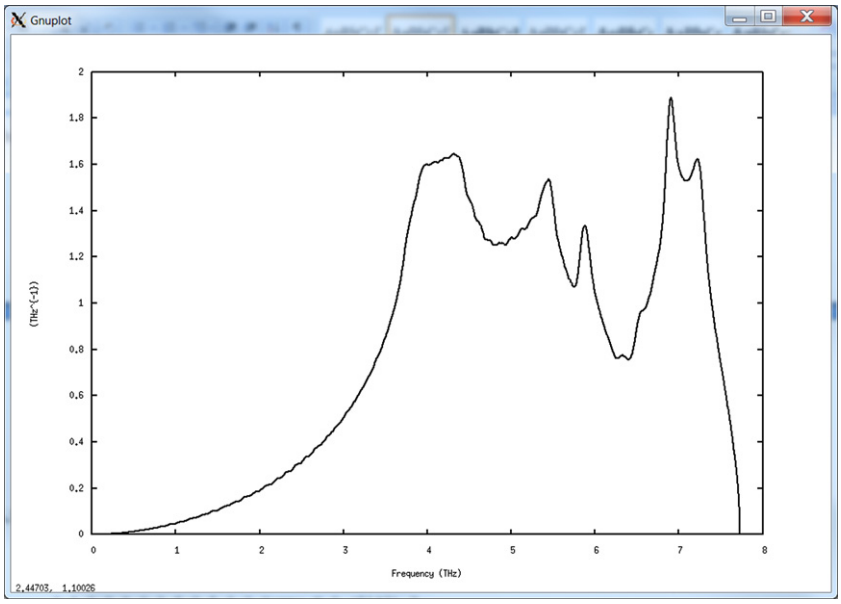

Fig. 1. PDOS of Mg.

```
   0.83333333  0.16666667   0.75000000  Mg
   0.83333333  0.66666667   0.25000000  Mg
   0.83333333  0.66666667   0.75000000  Mg
   -2.885402    0.000005     0.000000    0.056928    0.000000    0.000000
1.425033    0.000000    0.000000    -0.042757    0.000000    0.000000
0.356201   -0.617095    0.000000     0.016087    0.033974    0.000000
0.356196    0.617091    0.000000     0.016087   -0.033974    0.000000   -
0.027400    0.000000   -0.000001    -0.027400    0.000000    0.000001
0.003752    0.000000    0.000000     0.003752    0.000000    0.000000
0.187231   -0.123917    0.312727     0.187231   -0.123917   -0.312727
0.187230    0.123917   -0.312726     0.187230    0.123917    0.312726
...
```

6.1.7. To calculate the PDOS

To calculate the PDOS, type (if your system have gnuplot installed, you will see the PDOS plotted as shown in Fig. 1)

```
Yphon <superfij.out -plot
```

6.1.8. To calculate the phonon dispersions

Continually, prepare the phonon dispersion file contains lines (copy and paste works fast)

```
0.0 0.0 0.0 0.5 0.5 0.0 Gamma K 0 ($1*2) 2
0.5 0.0 0.0 0.0 0.0 0.0 K Gamma 1 ((0.5-$1)*2) 2
0.0 0.0 0.0 0.0 0.0 0.5 Gamma A 2 (($1)*2) 2
0.0 0.0 0.5 0.5 0.5 0.5 A L 3 ($1*2) 2
0.5 0.0 0.5 0.5 0.0 0.0 L M 4 ((0.5-$1)*2) 2
```

Then execute Yphon as follows to see the phonon dispersion plot in Fig. 2.

```
Yphon <superfij.out -pdis dfile.hcp -expt exp01.dat -plot
```

Note: `dfile.hcp` file contains the instruction on how to plot the experimental data contained in the file "`exp01.dat`" together with the calculated phonon dispersions. You can get the `dfile.hcp` and `exp01.dat` files from the demo folder of "`Mg`".

Up to this step, you will find two useful files "`vdis.plt`" and "`vdos.plt`" which are gnuplot scripts produced by `Yphon` for usage by gnuplot to make figures of phonon dispersions and PDOS.

6.2. MgO

Make a folder "`MgO`" under the folder of "`mYdemo`" and go to the folder "`MgO`".

This demo just shows how to calculate the phonon dispersions of MgO. You need to prepare the following files and submit your VASP.5 job.

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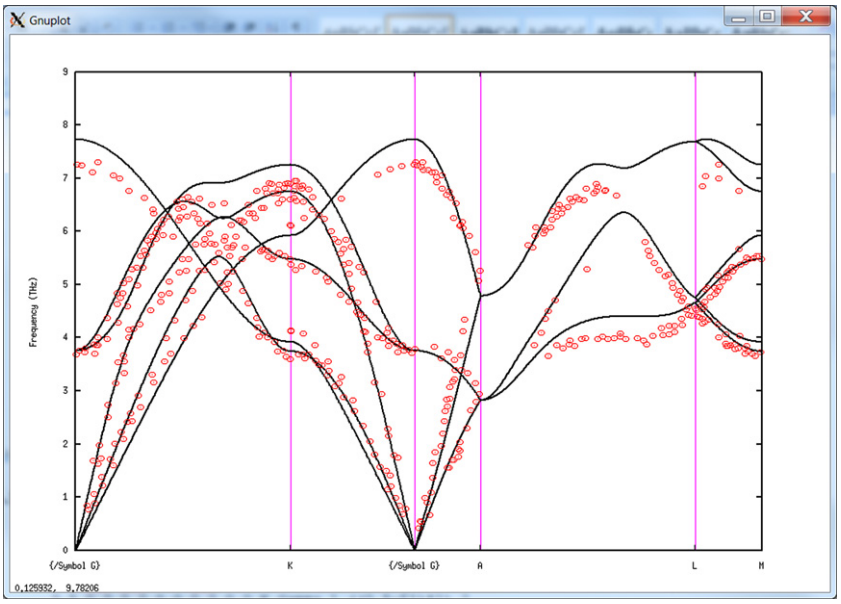

Fig. 2. Phonon dispersions of Mg.

6.2.1. POSCAR file

First, prepare the POSCAR file containing the follow lines for the primitive unit cell, name the file as "POSCAR.prm"

```
MgO-Born effective charge
4.212
.0 .5 .5
.5 .0 .5
.5 .5 .0
Mg O
1 1
D
.0 .0 .0 Mg
.5 .5 .5 O
```

Then type "cp POSCAR.prm POSCAR"

6.2.2. INCAR file

```
EDIFF=1.d-6
PREC = High
ISMEAR = -5
IBRION = -1
LEPSILON=.T.
NSW=0
```

6.2.3. KPOINTS file

```
Magnesium Oxide
0
G
9 9 9
 0 0 0
```

6.2.4. POTCAR file

Get POTCAR by the following Linux commands (depending on the VASP pseudopotential location in your system)

```
zcat /usr/global/msc/vasp/potpaw_LDA/Mg/POTCAR.Z >POTCAR
zcat /usr/global/msc/vasp/potpaw_LDA/O/POTCAR.Z >>POTCAR
```

6.2.5. Collect Born effective charge tensor and macroscopic dielectric tensor

Submit your batch job and after the batch job is done, type "vasp_BE" to collect Born effective charge and macroscopic dielectric tensor and then you will see the file named "dielecfij.out" which contains lines like

```
 0.000000    2.106000    2.106000
 2.106000    0.000000    2.106000
 2.106000    2.106000    0.000000
0.0000000000000000  0.0000000000000000 0.0000000000000000   Mg
0.5000000000000000  0.5000000000000000 0.5000000000000000   O
         3.147     0.000    0.000
         0.000     3.147    0.000
         0.000     0.000    3.147
ion   1
   1    1.96085     0.00000    0.00000
   2    0.00000     1.96085    0.00000
   3    0.00000     0.00000    1.96085
ion   2
   1   -1.96142     0.00000    0.00000
   2    0.00000    -1.96141    0.00000
   3    0.00000     0.00000   -1.96141
```

6.2.6. Modify INCAR for force constant calculation

```
EDIFF=1.d-6
PREC = A
ISMEAR = -5
IBRION = 6
ISIF = 0
NSW=1
```

6.2.7. Create supercell POSCAR file

```
Ycell -ss 2 <POSCAR.prm >POSCAR
```

You will see the POSCAR file like

```
Supercell by Yi Wang
1.00
         0.0000000000        4.2120000000         4.2120000000
         4.2120000000        0.0000000000         4.2120000000
         4.2120000000        4.2120000000         0.0000000000
 Mg O
 8 8
D
   0.0000000000    0.0000000000    0.0000000000 Mg
   0.0000000000    0.0000000000    0.5000000000 Mg
   0.0000000000    0.5000000000    0.0000000000 Mg
   0.0000000000    0.5000000000    0.5000000000 Mg
   0.5000000000    0.0000000000    0.0000000000 Mg
   0.5000000000    0.0000000000    0.5000000000 Mg
   0.5000000000    0.5000000000    0.0000000000 Mg
   0.5000000000    0.5000000000    0.5000000000 Mg
   0.2500000000    0.2500000000    0.2500000000 O
   0.2500000000    0.2500000000    0.7500000000 O
   0.2500000000    0.7500000000    0.2500000000 O
   0.2500000000    0.7500000000    0.7500000000 O
   0.7500000000    0.2500000000    0.2500000000 O
   0.7500000000    0.2500000000    0.7500000000 O
   0.7500000000    0.7500000000    0.2500000000 O
   0.7500000000    0.7500000000    0.7500000000 O
```

6.2.8. Modify KPOINTS file

```
Magnesium Oxide
0
G
 3 3 3
 0 0 0
```

6.2.9. Phonon for MgO

Run VASP interactively (the run can be finished in less one minute) or make a PBS batch job script and then submit your job. After your supercell job is done, you need to type the following Linux commands sequentially

```
vasp_fij
```

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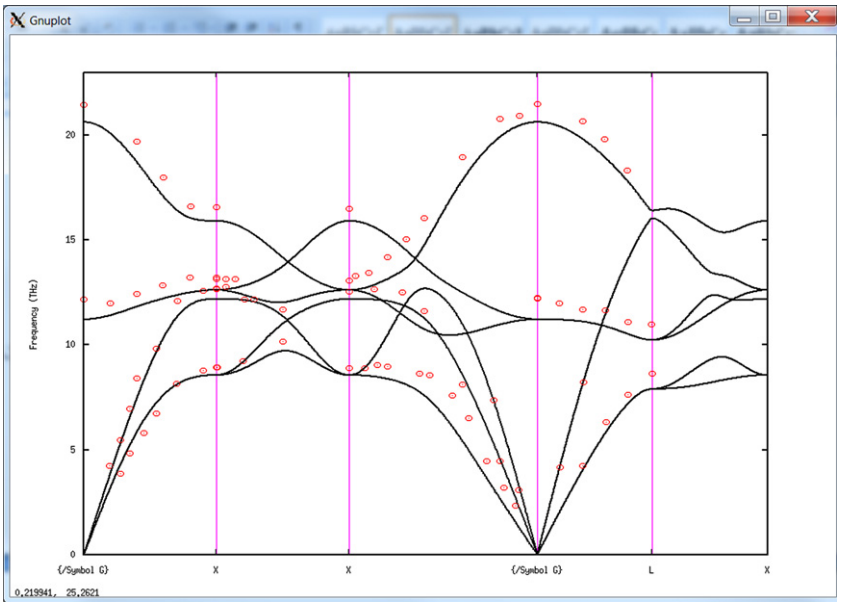

Fig. 3. Phonon dispersions of MgO.

To calculate the phonon dispersions, you run

```
Yphon -Born dielecfij.out -pdis dfile.fcc -bvec -expt
exp01.dat -plot <superfij.out
```

Note: `dfile.fcc` is a phonon dispersion file to instruct Yphon and `exp01.dat` contains the experimental neutron scattering data. You can get the `dfile.fcc` and `exp01.dat` files from the demo folder of "MgO". The data in `dfile.fcc` are like

```
0 0 0 0 0 .5 Gamma X 1 $1 2
0 .5 .5 0 .5 .0 X X 3 $1 2
0 .5 .5 0 0 0 X Gamma 0 (1-$1) 2
0 0 0 .25 .25 .25 Gamma L 2 (2*$1) 2
.25 .25 .25 0 .5 .5 L X
```

The key "`-expt`" instructs Yphon to plot the experimental data contained in the file "`exp01.dat`" together with the calculated phonon dispersions. The key "`-plot`" instructs Yphon to plot the figure in the terminal using gnuplot for you to check the calculated results, as shown in Fig. 3.

6.3. Fe_2O_3

The folder "Fe2O3" comes together with the YPHON package under the subfolder of "Ydemo". Go to the folder and you can play around by run

```
    pos2s Symmetry.pos
    Yphon -pdis dfile.rho -Born dielecfij.out -plot -Gfile
symmetry.mode <superfij.out
```

You can see some outputs from the screen, where the lines after the line "`Solving frequencies considering LO--TO splitting:`" contains the vibrational mode analysis showed like

```
  2  A1g Modes of raman_active
No irrep        THz                  (cm-1)          Z*(x)     Z*(y)      Zz(z)
 0 A1g    14.7222   14.7222  (    491.08    491.08)    0.0001    0.0001    -0.0000
 1 A1g     6.7666    6.7666  (    225.71    225.71)    0.0000   -0.0001    -0.0000

 3 A2g Modes of silent_mode
No irrep       THz                  (cm-1)          Z*(x)     Z*(y)      Zz(z)
 0  A2g 19.3966   19.3966   (    647.00    647.00)    0.0000   -0.0000     0.0000
 1  A2g 12.0187   12.0187   (    400.90    400.90)   -0.0000   -0.0000    -0.0000
 2  A2g  5.1938   5.1938    (    173.25    173.25)    0.0000    0.0000     0.0000

 5   Eg Modes of raman_active
No irrep       THz                  (cm-1)          Z*(x)     Z*(y)      Zz(z)
 0  Eg  18.0561   18.0561   (    602.29    602.29)    0.0001   -0.0000    -0.0000
 1  Eg  12.3172   12.3172   (    410.86    410.86)    0.0005    0.0000     0.0000
```

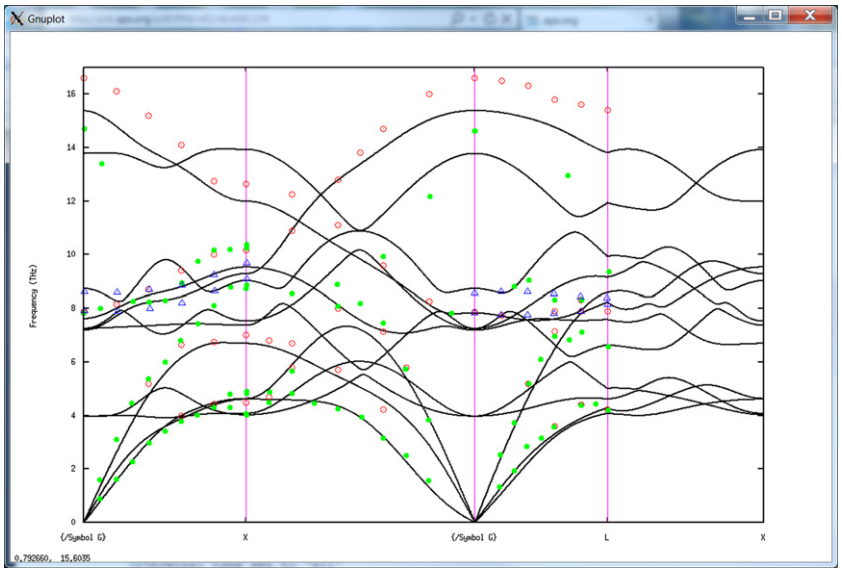

Fig. 4. Phonon dispersions of MnO under the hexagonal structure.

```
 2  Eg    9.1456    9.1456   (    305.06    305.06)   -0.0002   -0.0000    0.0000
 3  Eg    8.5509    8.5509   (    285.23    285.23)    0.0001   -0.0000    0.0000
 4  Eg    7.3436    7.3436   (    244.96    244.96)    0.0000    0.0000   -0.0000

 2 A1u Modes of silent_mode
No irrep       THz                    (cm-1)          Z*(x)     Z*(y)      Zz(z)
 0 A1u  16.9678   16.9678   (    565.99    565.99)    0.0000    0.0000    0.0000
 1 A1u  10.4414   10.4414   (    348.29    348.29)   -0.0000    0.0000   -0.0000

 3 A2u Modes of ir_active : P= 0.000, 0.000, 1.000 ( 0.577, 0.577, 0.577) with one
tranlational mode

No irrep       THz                    (cm-1)          Z*(x)     Z*(y)      Zz(z)
 0 A2u  15.3959   19.1088   (    513.55    637.40)    0.0000   -0.0000   -1.6197
 1 A2u   8.9912   11.5315   (    299.92    384.65)   -0.0000    0.0000    0.6251
 2 A2u  -0.0002   -0.0002   (     -0.01     -0.01)   -0.0000    0.0000   -0.0000

  5 Eu Modes of ir_active : P= 0.866, 0.500, 0.000 ( 0.000, 0.707,-0.707) with one
tranlational mode
No irrep       THz                    (cm-1)          Z*(x)     Z*(y)      Zz(z)
 0  Eu  15.4084   19.0377   (    513.97    635.03)   -1.6366   -0.9449    0.0000
 1  Eu  12.9891   14.7124   (    433.27    490.75)   -0.3671   -0.2119   -0.0000
 2  Eu   8.9470   10.9725   (    298.44    366.00)    0.3687    0.2129   -0.0000
 3  Eu   6.9920    7.0000   (    233.23    233.49)   -0.0170   -0.0098   -0.0000
 4  Eu   0.0000    0.0000   (      0.00      0.00)    0.0000   -0.0000    0.0000
```

Note: for the LO–TO splitting analysis, do not use the output lines after the line "`Frequencies in Gamma point without & with NA term`" that have been calculated by diagonalization of the force constant matrix in the real space using a polarization direction which might not be along the polarization direction of all infrared modes. For example, the polarization direction of the Eu mode is different from that of the A2u mode for Fe2O3 as shown above.

6.4. MnO

The salient feature of Yphon is best shown using the data contained in the subfolder MnO that we have published previously [1]. The physical thinking is that, for many materials, the measurements are usually done for the high symmetry structure which may not be mechanically stable at low temperature. And if one employs high symmetry structure in the calculation, one would get some imaginary phonon modes [11]. Other cases include the case that the magnetic ordering breaks the crystal symmetry. The Yphon solution is to restore the symmetry, or in another word, "unfold" the Brillouin zone.

First, one can run Yphon as follows and get result as in Fig. 4, showing that there are totally 12 dispersion curves along each direction since the primitive unit cell of MnO in the antiferromagnetic structure contains 4 atoms.

```
    Yphon -Born dielecfij.out -pdis dfile.scc -expt exp05.dat -
plot -thr2 0.01-bvec <superfij.out
```

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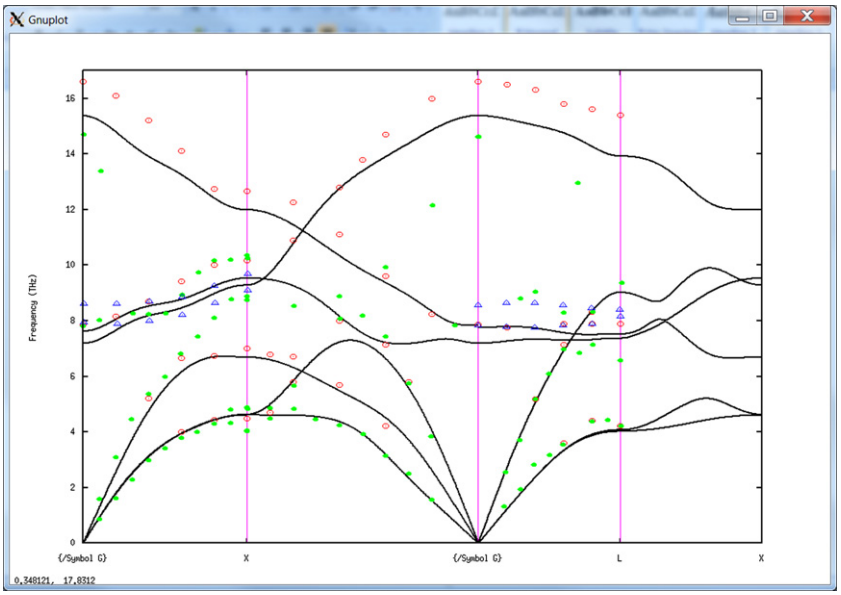

Fig. 5. Phonon dispersions of MnO under the cubic structure by the averaging force constant calculated with the hexagonal structure using Eq. (11).

Next, one can run Yphon as follows and get result as in Fig. 5, showing 6 dispersion curves along each direction of the cubic MnO by averaging the force constants calculated from the hexagonal structure through "restore translational symmetry" using Eq. (11), resulting in 2 atoms (6 dispersion curves) in the primitive unit cell.

```
    Yphon -Born dielecfij.fcc -pdis dfile.fcc -expt exp05.dat -
plot -thr2 0.01 -bvec <superfij.out
```

Continually, one can run Yphon in the following form and get result as in Fig. 6, showing the results of the fcc MnO by further average of the force constants calculated from the hexagonal structure by means of "restore rotational symmetry" using Eq. (12), resulting in the degeneracy of the dispersions curves also recovered.

```
    Yphon -Born dielecfij.fcc -pdis dfile.fcc -expt exp05.dat -
plot -thr2 0.01 -bvec -Rfile Rotation.sym <superfij.out
```

6.5. Cu_3Au

This demo is for the phonon dispersion calculation of disordered Cu_3Au using the SQS structure with the following Yphon command:

```
    Yphon -pdis dfile.fcc -Born dielecfij.out -thr2 0.10 -expt
exp01.dat -plot -bvec -nof -noNA -mall -Mass mass.1
<superfij.out
```

The plot is shown in Fig. 7. For the detailed mechanism and formulations, see our previous publication [23].

6.6. Al_2O_3, $BiFeO_3$, *GaAs*, $MgAl_2O_4$, *NaCl*, ZrW_2O_8, *BeO, and* $ZrSiO_4$

These subfolders contain the already calculated force constants and the dielectric quantities by us. Some of them are not published yet. We hope that the users can find some useful settings from these subfolders on getting experiences running Yphon.

7. Quick start to run YPHON

Step 1. Make a project directory, say 'MgO', and go to the project directory by "cd MgO".
Step 2. Setup the POSCAR file with the primitive unit cell. Relax the calculation using ISIF=3 and IBRION=2 or whatever you can handle, followed by making a final static calculation, and then save the CONTCAR as 'Static.CON' and gzip the OUTCAR by 'gzip OUTCAR >Static.OUT.gz'. If you do not want consider the LO–TO splitting for the polar system, then skip to Step 4.
Step 3. For polar material, "cp CONTCAR POSCAR" and then delete the key IBRION and NSW in INCAR file and add three lines into INCAR as follows:

```
IBRION=-1
NSW=0
LEPSILON=.T.
```

Submit your job. After your job is done, collect the result using "vasp_BE". You may also want to save the CONCAR as 'Born.CON' and 'gzip OUTCAR >Born.OUT.gz' for your future reference.

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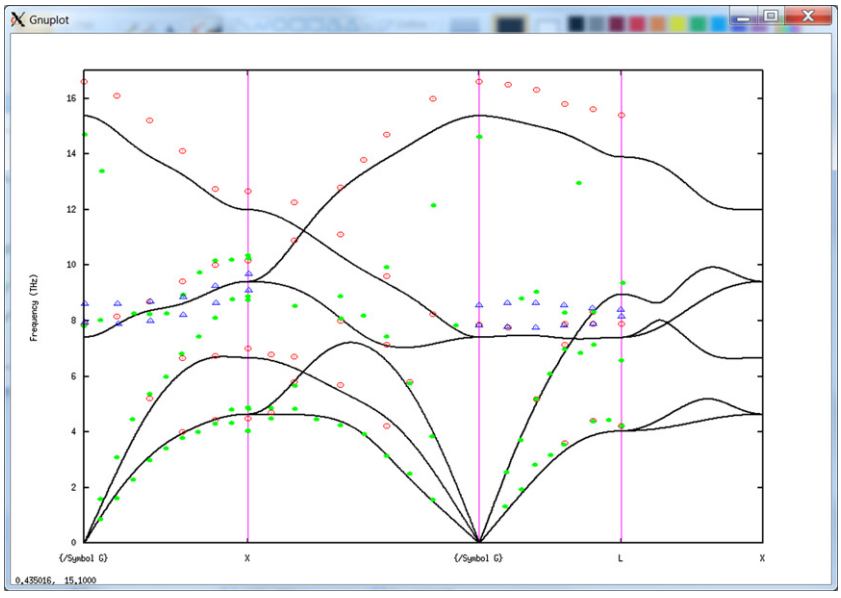

Fig. 6. Phonon dispersions of MnO under the cubic structure by the averaging force constant calculated with the hexagonal structure using Eq. (12).

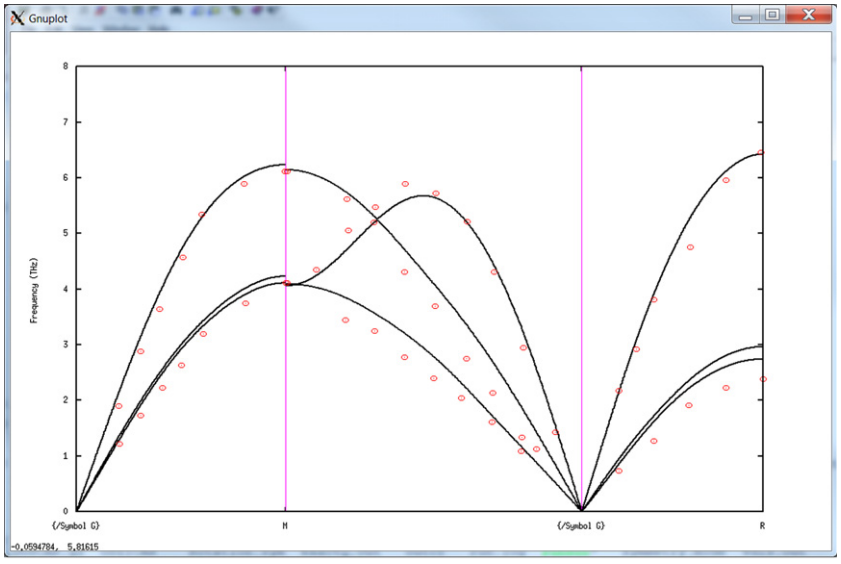

Fig. 7. Phonon dispersions of disordered Cu_3Au.

Step 4. Build supercell by '`Ycell -ss 2 <CONTCAR >POSCAR`'. Revise the INCAR file by

```
PREC=A
IBRION=6
NSW=1
ISIF=0
```

Note: ISIF = 0 instruct VASP.5 not calculate the stress and therefore can save plenty of computing time for you. Alternatively, you can also use IBRION=8.

Revise the KPOINTS file as follows

```
Magnesium Oxide
0
G
 3 3 3
 0 0 0
```

Submit your job. After your job is done, collect the result using "`vasp_fij`". Calculate the PDOS using "`Yphon <superfij.out -plot`" which produce the '`vdos.out`' file.

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8. Summary

The present paper had the aim to describe YPHON, a package for mixed-space approach to phonons of polar materials within the direct approach, in view of its use for user of VASP.5. YPHON makes full use of the accuracy of the force constants calculated in the real space and the dipole–dipole interactions in the reciprocal space, making the accurate phonon calculation possible with the direct method for polar materials besides the linear response method. After three years of internal test, we finally decide to distribute YPHON as an open source package.

9. Troubleshooting

Make sure that the Linux command search `PATH` has modified correctly to include `YPHON`, see Section 3.4.

Make sure Python is installed correctly. Otherwise, `pos2s` will not work.

After your VASP.5 job is done, always have a look at the OSZICAR file to make sure the VASP.5 calculation finished normally.

Before run `Yphon`, always check the `superfij.out` file (see Section 5.4.1 and the `dielecfij.out` file if for polar materials, see Section 5.4.2) for the data completeness and correctness. Nowadays the Linux clusters are not that stable and sometimes very weird results can be observed due to certain problems such as the RAM, disk space, and node sharing by different Linux processes. Of course, the most frequently happened problem is that your job has run over the allowed time limit in your VASP.5 batch jobs.

YPHON is program for lattice dynamical calculation of phonons of polar materials using the real-space force constants calculated within the first-principles supercell approach or direct method. The actual computer source code was written by Yi Wang. Questions about using YPHON may be directed to Yi Wang via e-mail at yuw3@psu.edu.

We understand that a lot of researchers have troubles in defining the high symmetry points in the phonon dispersion calculation. If this is the case for you, you can send Yi Wang your `superfij.out` file together with the experimental data in the form of two columns and tell him what kind of phonon dispersions you want. Then Yi Wang may help you when he has spare time.

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