Supplementary Information
Half-metallic and ferromagnetic phases in CrSH monolayer using DFT+U and BO-MD calculations

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Magnetocrystalline Anisotropy Energy (MAE) calculation

we performed the MAE calculations to determine the preferred magnetic easy axis for the FM CrSH. The MAE was calculated using the Force-Theorem approximation (Phys. Rev. B 90, 205409 (2014) and Phys. Chem. Chem. Phys., 2024, 26, 10111-10119). The MAE is defined as:

$$MAE = E_{\parallel} - E_{\perp},$$

where $E_{\parallel}$ represents the energy when the spin moments are aligned along the x-y plane, and $E_{\perp}$ represents energy when the spin moments are aligned along the z-axis.

Our calculated MAE results

For FM 1T-CrSH:

$$MAE_{1T} = (-550.3159eV)\text{–}(-550.3161eV) = +0.172meV/fu,$$

The positive MAE_{1T} value indicates that the magnetic easy axis lies in a perpendicular (z-axis) direction.

For 2H-CrSH:

$$MAE_{2H} = (-549.5262eV)\text{–}(-549.5244eV) = -1.775meV/fu.$$

The negative MAE_{2H} value suggests a preference for the magnetic easy axis to lie in the x-y plane.

Examples of Quantum ESPRESSO (QE) input files

This section provides examples of input files such as the self-consistent field energy (SCF), Geometry optimization, and Born-Oppenheimer molecular dynamics (BO-MD) simulations for QE packages.

2x2x1 supercell for FM and AFM CrSH monolayers

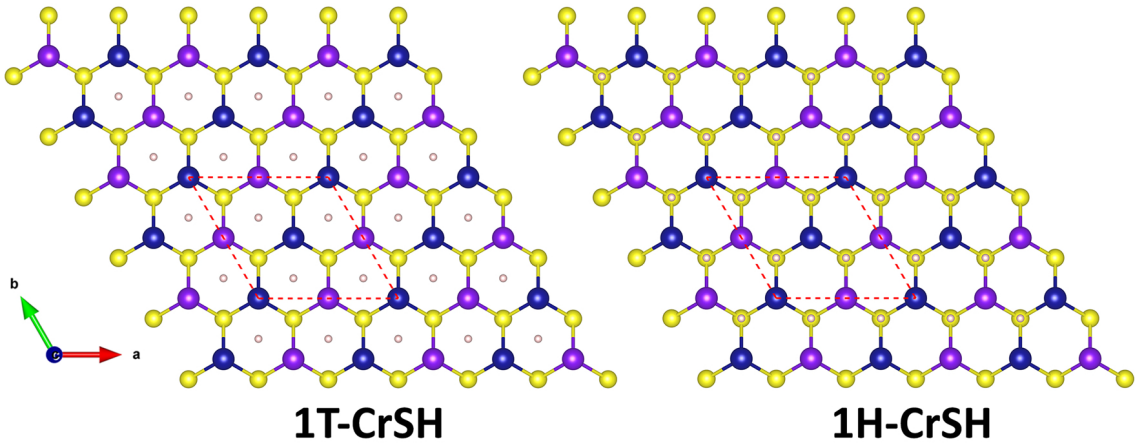

Figure S1: 2x2x1 supercell for CrSH monolayers: (Left) 1T-CrSH and (Right) 1H-CrSH.

`SCF.in` for FM 1T-CrSH (12 atoms)

The input file `SCF.in` is used to calculate the ground-state energy calculation for FM 1T-CrSH (12 atoms).

```
&CONTROL
    calculation    = "scf"
    forc_conv_thr =  1.00000e-04
    nstep          = 100
    outdir         = "/temp"
    prefix         = "prefix"
    pseudo_dir     = "./pseudo"
    restart_mode   = "from_scratch"
    tprnfor        = .TRUE.
    tstress        = .TRUE.
/

&SYSTEM
    degauss                    =  1.0000e-02
    ecutwfc                    =  8.00000e+01
    ibrav                      = 4
a = 6.5133618
c = 20.000d-0
    nat                        = 12
    nspin                      = 2
    ntyp                       = 4
    assume_isolated            = "2D"
    occupations                = "smearing"
    smearing                   = "marzari-vanderbilt"
    starting_magnetization(1) =  5.0d-1
    starting_magnetization(2) =  5.0d-1
/

&ELECTRONS
    conv_thr         =  1.00000e-08
    diago_david_ndim = 4
```

```
    diagonalization  = "david"
    electron_maxstep = 250
    mixing_beta      =  7.00000e-01
    mixing_mode      = "plain"
    startingpot      = "atomic"
    startingwfc      = "atomic+random"
/

K_POINTS {automatic}
10 10 2 0 0 0

ATOMIC_SPECIES
Cr1     51.99610  Cr.pbe-spn-kjpaw_psl.1.0.0.UPF
Cr2     51.99610  Cr.pbe-spn-kjpaw_psl.1.0.0.UPF
H        1.00794  H.pbe-kjpaw_psl.1.0.0.UPF
S       32.06500  S.pbe-n-kjpaw_psl.1.0.0.UPF

HUBBARD {ortho-atomic}
U Cr1-3d 5.520e-0
U Cr2-3d 5.520e-0

ATOMIC_POSITIONS {crystal}
Cr1 0.000000    0.000000    0.500000
Cr1 0.500000    0.000000    0.500000
Cr2 0.500000    0.500000    0.500000
Cr2 0.000000    0.500000    0.500000
H   0.333333    0.166667    0.543345
H   0.333333    0.666667    0.543345
H   0.833333    0.166667    0.543345
H   0.833333    0.666667    0.543345
S   0.166667    0.333333    0.428455
S   0.166667    0.833333    0.428455
S   0.666667    0.333333    0.428455
S   0.666667    0.833333    0.428455
```

`OPT.in` for Geometry optimization

The input file `OPT.in` is used to calculate the variable-cell relaxation optimization for FM and AFM 1H-CrSH (12 atoms). To switch between FM and AFM configurations, modify the sign of starting_magnetization for Cr2.

```
&CONTROL
    calculation   = "vc-relax"
    forc_conv_thr =  1.00000e-04
    nstep         = 100
    outdir        = "./temp/"
2H/02Prop/temp_1FM/"
    prefix        = "prefix"
    pseudo_dir    = "./PP"
    restart_mode  = "from_scratch"
    tprnfor       = .TRUE.
    tstress       = .TRUE.
/

&SYSTEM
    a                           =  6.509350
    c                           =  2.00000e+01
    degauss                     =  1.0000e-02
    ecutwfc                     =  8.00000e+01
    ibrav                       = 4
    nat                         = 12
    nspin                       = 2
    ntyp                        = 4
    assume_isolated             = "2D"
    occupations                 = "smearing"
    smearing                    = "marzari-vanderbilt"
    starting_magnetization(1) =  5.0d-1
    starting_magnetization(2) =  5.0d-1  ! +0.5 for FM and -0.5 for AFM
/
```

```
&ELECTRONS
    conv_thr         =  1.00000e-08
    diago_david_ndim = 4
    diagonalization  = "david"
    electron_maxstep = 250
    mixing_beta      =  7.00000e-01
    mixing_mode      = "plain"
    startingpot      = "atomic"
    startingwfc      = "atomic+random"
/
 &IONS
    ion_dynamics='bfgs'
/
&CELL
   cell_dofree= '2Dxy'
   cell_dynamics = 'bfgs' ,
   press = 0.0D-0 ,
   press_conv_thr= 0.5D-2,
 /

K_POINTS {automatic}
10 10 1 0 0 0

ATOMIC_SPECIES
Cr1     51.99610  Cr.pbe-spn-kjpaw_psl.1.0.0.UPF
Cr2     51.99610  Cr.pbe-spn-kjpaw_psl.1.0.0.UPF
H        1.00794  H.pbe-kjpaw_psl.1.0.0.UPF
S       32.06500  S.pbe-n-kjpaw_psl.1.0.0.UPF

HUBBARD {ortho-atomic}
U Cr1-3d 5.520e-0
U Cr2-3d 5.520e-0

ATOMIC_POSITIONS {crystal}
Cr1 0.000000    0.000000    0.500000
Cr1 0.500000    0.000000    0.500000
Cr2 0.500000    0.500000    0.500000
Cr2 0.000000    0.500000    0.500000
H   0.166667    0.333333    0.543270
H   0.166667    0.833333    0.543270
H   0.666667    0.333333    0.543270
H   0.666667    0.833333    0.543270
S   0.166667    0.333333    0.428240
S   0.166667    0.833333    0.428240
S   0.666667    0.333333    0.428240
S   0.666667    0.833333    0.428240
```

`BOMD.in` for FM 1H-CrSH (27 atoms)

he input file `BOMD.in` is used to calculate the Bo-MD calculation for 3x3x1 supercell of FM 1H-CrSH (27 atoms).

```
&CONTROL
  calculation = 'md'
  restart_mode = 'from_scratch'
  outdir = './temp'
  prefix = 'prefix'
  pseudo_dir = './pseudo'
  nstep = 10000
  dt = 10.0
/
&SYSTEM
  ecutwfc =   60.0
  ibrav = 0
  nat   = 27
  ntyp  = 3
  occupations= 'smearing'
  smearing = "marzari-vanderbilt"
  degauss = 0.02
```

```
  nosym = .true.
/
&electrons
  conv_thr =1.0d-8
  mixing_beta =   0.4
  electron_maxstep = 500000
/
&ions
  ion_dynamics = 'verlet',
  ion_temperature = 'berendsen'
  tempw = 300.0
  pot_extrapolation = 'second-order'
  wfc_extrapolation = 'second-order'
/
ATOMIC_SPECIES
Cr       51.99610  Cr.pbe-spn-kjpaw_psl.1.0.0.UPF
H        1.00794  H.pbe-kjpaw_psl.1.0.0.UPF
S        32.06500  S.pbe-n-kjpaw_psl.1.0.0.UPF

ATOMIC_POSITIONS crystal
Cr            0.0000000000       0.0000000000       0.5087720000
Cr            0.0000000000       0.3333333333       0.5087720000
Cr            0.0000000000       0.6666666667       0.5087720000
Cr            0.3333333333       0.0000000000       0.5087720000
Cr            0.3333333333       0.3333333333       0.5087720000
Cr            0.3333333333       0.6666666667       0.5087720000
Cr            0.6666666667       0.0000000000       0.5087720000
Cr            0.6666666667       0.3333333333       0.5087720000
Cr            0.6666666667       0.6666666667       0.5087720000
H             0.2222220000       0.1111110000       0.5576730000
H             0.2222220000       0.4444443333       0.5576730000
H             0.2222220000       0.7777776667       0.5576730000
H             0.5555553333       0.1111110000       0.5576730000
H             0.5555553333       0.4444443333       0.5576730000
H             0.5555553333       0.7777776667       0.5576730000
H             0.8888886667       0.1111110000       0.5576730000
H             0.8888886667       0.4444443333       0.5576730000
H             0.8888886667       0.7777776667       0.5576730000
S             0.2222220000       0.1111110000       0.4335550000
S             0.2222220000       0.4444443333       0.4335550000
S             0.2222220000       0.7777776667       0.4335550000
S             0.5555553333       0.1111110000       0.4335550000
S             0.5555553333       0.4444443333       0.4335550000
S             0.5555553333       0.7777776667       0.4335550000
S             0.8888886667       0.1111110000       0.4335550000
S             0.8888886667       0.4444443333       0.4335550000
S             0.8888886667       0.7777776667       0.4335550000
CELL_PARAMETERS angstrom
      8.5164000000       0.0000000000       0.0000000000
     -4.2582010298       7.3754181542       0.0000000000
      0.0000000000       0.0000000000      20.0000000000
K_POINTS automatic
2 2 1 0 0 0
```

Listing 1: Input file of 1H-CrSH phase (27 atoms) for the BO-MD simulation.

Phonon Calculation Using PHONOPY and HIPHIVE packages

This section provides details of the workflow used for phonon calculations, including steps performed with the PHONOPY and HIPHIVE packages. Example input files and references are provided to assist readers in replicating similar workflows.

Phonon Dispersion Using PHONOPY

The phonon dispersion calculations for CrSH monolayer were conducted using a combination of QE and PHONOPY packages. The detailed workflow is based on the tutorial available at `https://phonopy.github.io/phonopy/qe.html`. Below is an outline of the workflow:

- The initial input file (`SCF_uc.in`) for FM 1T-CrSH unit cell (3 atoms) including atomic positions was required to generate displacement structures in 3x3x1 supercell (27 atoms). This file specifies the atomic positions, pseudopotentials, and computational parameters. The `SCF_uc.in` file was also compatible with the QE package.

Phonopy command:

```
phonopy --qe -d --dim="3 3 1" -c SCF_uc.in
```

`SCF_uc.in` for a FM 1T-CrSH unit cell (3 atoms)

```
    &CONTROL
    calculation   = "scf"
    forc_conv_thr =  1.00000e-04
    nstep         = 100
    outdir        = "./temp"
    prefix        = "prefix"
    pseudo_dir    = "./PP"
    restart_mode  = "from_scratch"
    tprnfor       = .TRUE.
    tstress       = .TRUE.
/

&SYSTEM
    degauss          =  1.0000e-02
    ecutwfc          =  8.00000e+01
    ecutrho          =  7.20000e+02
    ibrav            = 0
    nat              = 3
    nspin            = 2
    ntyp             = 3
    assume_isolated           = "2D"
    occupations               = "smearing"
    smearing                  = "marzari-vanderbilt"
    starting_magnetization(1) =  5.0d-1
/

&ELECTRONS
    conv_thr         =  1.00000e-08
    diago_david_ndim = 4
    diagonalization  = "david"
    electron_maxstep = 250
    mixing_beta      =  7.00000e-01
    mixing_mode      = "plain"
    startingpot      = "atomic"
    startingwfc      = "atomic+random"
```

```
/

K_POINTS {automatic}
20 20 2 0 0 0

ATOMIC_SPECIES
Cr     51.99610  Cr.pbe-spn-kjpaw_psl.1.0.0.UPF
H        1.00794  H.pbe-kjpaw_psl.1.0.0.UPF
S       32.06500  S.pbe-n-kjpaw_psl.1.0.0.UPF

HUBBARD {ortho-atomic}
U Cr1-3d 5.520e-0

CELL_PARAMETERS {bohr}
  6.154233   0.000000   0.000000
 -3.077117   5.329722   0.000000
  0.000000   0.000000  37.794523

ATOMIC_POSITIONS {crystal}
Cr      0.000000   0.000000   0.500000
H       0.666667   0.333333   0.543345
S       0.333333   0.666667   0.428455
```

- The PHONOPY package was used to generate and obtain the displacement supercell input files (3x3x1 supercell). The displacement supercells were created. (e.g., `supercell_1.in`, `supercell_2.in`, ...)

`supercell_1.in` for FM 1T-CrSH supercell (27 atoms)

```
&CONTROL
    calculation   = "scf"
    forc_conv_thr =  1.00000e-04
    nstep         = 100
    outdir        = ./temp01"
    prefix        = "prefix"
    pseudo_dir    = "./PP"
    restart_mode  = "from_scratch"
    tprnfor       = .TRUE.
    tstress       = .TRUE.
/

&SYSTEM
    degauss                   =  1.0000e-02
    ecutwfc                   =  6.00000e+01
    ecutrho                   =  5.40000e+02
   ibrav                     = 0
    nat                       = 27
    nspin                     = 2
    ntyp                      = 3
!    assume_isolated           = "2D"
    occupations               = "smearing"
    smearing                  = "marzari-vanderbilt"
    starting_magnetization(1) =  5.0d-1
/

&ELECTRONS
    conv_thr         =  1.00000e-08
    diago_david_ndim = 4
    diagonalization  = "david"
    electron_maxstep = 250
    mixing_beta      =  7.00000e-01
    mixing_mode      = "plain"
    startingpot      = "atomic"
    startingwfc      = "atomic+random"
/

K_POINTS {automatic}
10 10 1 0 0 0
```

```
CELL_PARAMETERS {bohr}
   18.4626990000     0.0000000000     0.0000000000
   -9.2313510000    15.9891660000     0.0000000000
    0.0000000000     0.0000000000    37.7945230000

ATOMIC_SPECIES
 Cr    51.99610    Cr.pbe-spn-kjpaw_psl.1.0.0.UPF
  H     1.00794    H.pbe-kjpaw_psl.1.0.0.UPF
  S    32.06500    S.pbe-n-kjpaw_psl.1.0.0.UPF

HUBBARD {ortho-atomic}
U Cr-3d 5.520e-0

ATOMIC_POSITIONS {crystal}
 Cr    0.0004754770917795  0.0000000000000000  0.5004754770917795
 Cr    0.3333333333333333  0.0000000000000000  0.5000000000000000
 Cr    0.6666666666666666  0.0000000000000000  0.5000000000000000
 Cr    0.0000000000000000  0.3333333333333333  0.5000000000000000
 Cr    0.3333333333333333  0.3333333333333333  0.5000000000000000
 Cr    0.6666666666666666  0.3333333333333333  0.5000000000000000
 Cr    0.0000000000000000  0.6666666666666666  0.5000000000000000
 Cr    0.3333333333333333  0.6666666666666666  0.5000000000000000
 Cr    0.6666666666666665  0.6666666666666666  0.5000000000000000
  H    0.2222223333333333  0.1111110000000000  0.5433450000000000
  H    0.5555556666666666  0.1111110000000000  0.5433450000000000
  H    0.8888890000000000  0.1111110000000000  0.5433450000000000
  H    0.2222223333333333  0.4444443333333333  0.5433450000000000
  H    0.5555556666666664  0.4444443333333333  0.5433450000000000
  H    0.8888890000000000  0.4444443333333333  0.5433450000000000
  H    0.2222223333333333  0.7777776666666667  0.5433450000000000
  H    0.5555556666666666  0.7777776666666667  0.5433450000000000
  H    0.8888890000000000  0.7777776666666667  0.5433450000000000
  S    0.1111110000000000  0.2222223333333333  0.4284549999999999
  S    0.4444443333333333  0.2222223333333333  0.4284549999999999
  S    0.7777776666666666  0.2222223333333333  0.4284549999999999
  S    0.1111110000000000  0.5555556666666666  0.4284549999999999
  S    0.4444443333333334  0.5555556666666666  0.4284549999999999
  S    0.7777776666666666  0.5555556666666666  0.4284549999999999
  S    0.1111109999999999  0.8888889999999999  0.4284549999999999
  S    0.4444443333333333  0.8888889999999999  0.4284549999999999
  S    0.7777776666666666  0.8888889999999999  0.4284549999999999
```

- The force on each atom was calculated using QE's `pw.x` module for each displacement input file, resulting in output files (e.g., `supercell_1.out`, `supercell_2.out`, ...).
- The forces from the output files were processed to generate the `FORCE_SETS` file, which serves as input for PHONOPY to compute the phonon dispersion.

  Phonopy command:
  `phonopy -f supercell_1.out supercell_2.out ...`

The phonon dispersion was calculated using PHONOPY, and the results are presented in Figure 4(c) of the main manuscript (red dashed line).

Incorporating Rotational Sum Rules with HIPHIVE package

To enhance the accuracy of the phonon dispersion and ensure compliance with physical invariance principles, rotational sum rules were applied using the HIPHIVE package. The methodology is detailed in the online documentation (eg. MoS2 monolayer and Graphene) at `https://hiphive.materialsmodeling.org/advanced_topics/rotational_sum_rules.html`

The procedure involves:

- The `FORCE_SETS` file was converted to the `extxyz` format to make it compatible with HIPHIVE package (`Forceset.extxyz`).
- The rotational invariance corrections were enforced using HIPHIVE. Python scripts and step-by-step instructions from the HIPHIVE tutorial were followed to apply these corrections.
- The corrected phonon dispersion, incorporating rotational invariance, resolves spurious imaginary frequencies near the Γ-point in the flexural ZA mode.

`Forceset.extxyz` for force set calculation in HIPHIVE package

```
    27
Lattice="9.77004 0.0 0.0  -4.88502 8.46110284 0.0 0.0 0.0 20.0" Properties=species:S:1:pos:R:3:displacements:R:3:forces:R:3 pbc="T T T"
Cr  0.0000000000   0.0000000000   10.0000000000  0.00438930  0.00000000  0.00898521  -0.0474209460  0.0000251865  -0.0734513171
Cr  3.2566800000   0.0000000000   10.0000000000  0.00000000  0.00000000  0.00000000  0.0066732824  -0.0039597676  0.0051280489
Cr  6.5133600000   0.0000000000   10.0000000000  0.00000000  0.00000000  0.00000000  0.0036764036  -0.0018473485  0.0064974189
Cr  -1.6283400000  2.8203680000   10.0000000000  0.00000000  0.00000000  0.00000000  0.0018930859  -0.0011819468  0.0073176011
Cr  1.6283400000   2.8203680000   10.0000000000  0.00000000  0.00000000  0.00000000  -0.0014629762  0.0066082412  0.0041564287
Cr  4.8850200000   2.8203680000   10.0000000000  0.00000000  0.00000000  0.00000000  0.0001177386  0.0002085062  0.0080976741
Cr  -3.2566800000  5.6407350000   10.0000000000  0.00000000  0.00000000  0.00000000  0.0032675981  -0.0016642859  0.0066195464
Cr  0.0000000000   5.6407350000   10.0000000000  0.00000000  0.00000000  0.00000000  0.0001539911  0.0000398418  0.0079472645
Cr  3.2566800000   5.6407350000   10.0000000000  0.00000000  0.00000000  0.00000000  -0.0028318319  0.0018699537  0.0049879238
H   1.6283400000   0.9401230000   10.8669000000  0.00000000  0.00000000  0.00000000  0.0024674705  -0.0010485065  0.0007206625
H   4.8850200000   0.9401230000   10.8669000000  0.00000000  0.00000000  0.00000000  -0.0002406733  0.0001447429  -0.0072150500
H   8.1417000000   0.9401230000   10.8669000000  0.00000000  0.00000000  0.00000000  0.0009376636  0.0019167477  -0.0008757360
H   0.0000000000   3.7604900000   10.8669000000  0.00000000  0.00000000  0.00000000  0.0001300799  0.0001869090  -0.0073752298
H   3.2566800000   3.7604900000   10.8669000000  0.00000000  0.00000000  0.00000000  0.0001318796  -0.0002306383  -0.0071142628
H   6.5133600000   3.7604900000   10.8669000000  0.00000000  0.00000000  0.00000000  -0.0000673809  0.0001246883  -0.0073202082
H   -1.6283400000  6.5808580000   10.8669000000  0.00000000  0.00000000  0.00000000  0.0000310924  0.0000316143  -0.0074073686
H   1.6283400000   6.5808580000   10.8669000000  0.00000000  0.00000000  0.00000000  -0.0003041796  -0.0002393801  -0.0073490046
H   4.8850200000   6.5808580000   10.8669000000  0.00000000  0.00000000  0.00000000  0.0043078668  -0.0008399900  -0.0001095470
S   0.0000000000   1.8802450000   8.5691000000   0.00000000  0.00000000  0.00000000  0.0024990951  -0.0224596322  0.0233288941
S   3.2566800000   1.8802450000   8.5691000000   0.00000000  0.00000000  0.00000000  -0.0003252627  -0.0001324221  -0.0001514560
S   6.5133600000   1.8802450000   8.5691000000   0.00000000  0.00000000  0.00000000  0.0009574611  -0.0004988045  -0.0001877086
S   -1.6283400000  4.7006130000   8.5691000000   0.00000000  0.00000000  0.00000000  0.0000017818  -0.0000370342  -0.0009721524
S   1.6283400000   4.7006130000   8.5691000000   0.00000000  0.00000000  0.00000000  0.0001439638  -0.0000370342  -0.0006710761
S   4.8850200000   4.7006130000   8.5691000000   0.00000000  0.00000000  0.00000000  0.0000781436  0.0007106429  -0.0001619975
S   -3.2566800000  7.5209800000   8.5691000000   0.00000000  0.00000000  0.00000000  -0.0068422400  0.0053121477  0.0133272996
S   0.0000000000   7.5209800000   8.5691000000   0.00000000  0.00000000  0.00000000  0.0001475634  0.0000303287  -0.0008525961
```

```
S    3.2566800000    7.5209800000    8.5691000000    0.00000000  0.00000000  0.00000000
    0.0318793576    0.0169672192    0.0330859767
27
Lattice="9.77004 0.0 0.0  -4.88502 8.46110284 0.0 0.0 0.0 20.0" Properties=species:S:1:
    pos:R:3:displacements:R:3:forces:R:3 pbc="T T T"
Cr   0.0000000000    0.0000000000    10.0000000000   -0.00438930 0.00000000  -0.00898521
    0.0488561182    0.0000840854    0.0902102794
...
```

These resources and example files enable readers to replicate similar workflows to calculate phonon dispersion and apply rotational sum rules.