Second Harmonic Generation (SHG) calculations using ABINIT
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Nonlinear optical (NLO) effects, such as second harmonic generation (SHG), play a crucial role in technologies such as lasers, modulators, and on-chip photonic and optoelectronic applications [https://pubs.acs.org/doi/10.1021/acsnano.1c00344]. SHG is also important in identifying crystal symmetries and their orientations. Exploring 2D materials, such as graphene and TMDs sparked significant interest as potential candidates for NLO devices. The atomically thin nature and reduced dimensionality of 2D materials enhance the interactions between photons and electrons, leading to stronger nonlinear responses than bulk materials. Thus, it will be interesting to investigate the novel 2D materials that exhibit high SHG.

Below is a step-by-step process for using the ABINIT package to calculate the SHG spectra. It involves relaxing the structure, calculating the band gap, and finally the SHG spectra. I have used monolayer B2Te3 as an example. For detailed information, please visit the ABINIT wiki here: https://docs.abinit.org/.

Step 1. Relax the structure
There may be multiple steps to relax the structure based on how complicated the crystal structure is. For monolayer B2Te3, I am doing the relaxation in 4 steps:

(a) optcell = 0
This is equivalent to setting ISIF = 2 in the VASP. i.e. relax the ions keeping the cell size and shape unchanged. This step can be skipped if the initial starting structure is very close to its ground state. However, if we don't have prior information about the final structure, it is recommended to go through this step first. For each calculation, we need three input files: name_of_the_file.abi (main input file), job submission script, and pseudopotential files. Below are the input parameters of the main input file named **mono_B2Te3.abi**.

acell: The magnitude of the lattice parameters in either Angstrom or Bohr unit. The Abinit default is Bohr. So, **acell** of the relaxed structure printed by Abinit will be in Bohr.

rprim: Primitive lattice vectors, with a format similar to the POSCAR file. See the examples below.

For hexagonal structures:
rprim 0.8660254 -0.5000000 0.0000000
   0.0000000 1.0000000 0.0000000

0.0000000 0.0000000 1.0000000

Similarly, for square lattices:
rprim 1.0000000 0.0000000 0.0000000
                0.0000000 1.0000000 0.0000000
                0.0000000 0.0000000 1.0000000

It should be noted that these vectors will be multiplied by the **acell** parameter to get the exact length of lattice parameters along each axis.

xred: Ionic positions (x,y,z) in reduced or fractional format. We can also use **xcart** if we want to use them in the Cartesian format.

optcell: As mentioned above, this tag is similar to setting ISIF in the VASP. Setting **optcell** to 0 means relaxing the ions keeping the cell size and shape unchanged (ISIF = 2).

nsym: Number of symmetry operations. Its default value is zero. i.e., all the atomic coordinates must be explicitly given: the code will then find automatically the symmetry operations that leave the lattice and each atomic sublattice invariant. It also checks whether the cell is primitive. Note that the tolerance on symmetric atomic positions and lattice is rather stringent. i.e. for a symmetry operation to be admitted, the lattice and atomic positions must map on themselves within 1.0e-8. I have set **nsym** = 0 in the calculation.

ionmov: It controls the displacements of ions. 0 means no movement of ions at all. I have used **ionmov** = 2 for the structural optimization using the BFGS algorithm. If **optcell** = 0, only the ions are moved. If **optcell** > 0, the cell parameters also change based on the value of **optcell**.

ecutsm: Energy cutoff smearing with the default value of 0.0. This is important when performing relaxation of unit cell size and shape (**optcell** > 0). Using a non-zero **ecutsm**, the total energy curves as a function of **ecut**, or **acell**, can be smoothed, keeping consistency with the stress. The recommended value is 0.5 Hartree (Ha). When **optcell** is not 0, ABINIT requires **ecutsm** to be larger than zero. I have set **ecutsm** = 0.5 Hartree in the calculation.

getwfk: Get the wavefunctions from _WFK file. It is used when **ndtset** > 0 (in the multi-dataset mode), to indicate starting wavefunctions. The default value is 0, meaning that no use of the previously computed output wavefunction file appended with _DSx_WFK is done. If **getwfk** > 0, its value gives the index of the dataset for which the output wavefunction file appended with _WFK must be used. If **getwfk** is a negative number, it indicates the number of datasets to go backward to find the needed wavefunction file. I have used **getwfk** = 0 in the relax calculation.

chksymtnons: It is related to the symmetry checking. It governs the behavior of the code when there is a potential symmetry breaking, related to the presence of non-symmorphic translations not leaving the FFT exchange-correlation grid invariant. If **chksymtnons** = 1, the code stops if the non-symmorphic translation part of the symmetry operations has components that are not zero, or simple fractions with 2, 3, 4, 5, 6, 8, 9, 10, or 12 as denominators. Also, suggestions to bypass the problem are made in the output file. If **chksymtnons** = 2, the code makes a similar check but does not stop after providing in the output file suggestions to bypass the problem. If **chksymtnons** = 3, the code acts as with **chksymtnons** = 1, but then generates an FFT grid which is left invariant under the action of the spatial symmetry operations, which might enlarge it. If **chksymtnons** = 0, the code skips the check. If we encounter the problem outlined above, we may have two choices: (1) change the atomic positions (translate them) such that the origin appears as the most symmetric point, (2) ignore the problem, and set **chksymtnons** = 2 or 0 (only the latter for cDFT). If **chksymtnons** = 2, ABINIT suggests a possible global translation and corresponding translated atomic positions. I have used **chksymtnons** = 0 in the calculation.

nstep: Max number of electronic steps to be performed in an ionic step. This is equivalent to the NELM tag in the VASP. I have used **nstep** = 60 in the calculation, which is the default value.

ntime: Max number of ionic steps (time steps) to be performed in the calculations. It works if **ionmov** is set to non-zero. Its default value is 1000. This is equivalent to the NSW tag in the VASP. I have used **ntime** =1000 in the calculation.

toldfe: Tolerance on the difference of total energy (EDIFF in VASP). It forbids the use of **tolwfr** or **toldff** or **tolrff** or **tolvrs**. Its default value is 0.0. I have used 1E-7, 1E-8, and 1E-9 Ha in different types of calculations based on the level of accuracy required to get accurate results.

toldff: Tolerance on the difference of forces (EDIFFG in VASP). It forbids the use of **toldfe** or **tolrff** or **tolvrs**. Its default value is 0.0. It is ineffective for very symmetric structures, so **toldfe** should be used in that case. Therefore, I haven't used this tag in the calculation.

tolmxf: Tolerance on the maximal force. Its default value is 5E-5 Ha/Bohr. It sets a maximal absolute force tolerance below which BFGS structural relaxation iterations will stop. This tolerance applies to any particular cartesian component of any atom, excluding fixed ones. This is to be used when trying to equilibrate a structure to its lowest energy configuration. So instead of **toldff**, I have used **tolmxf** in the calculation. This is because **toldff** didn't help with the convergence and **tolmxf** did instead. The default value is used in my calculation.

natom: Number of atoms in the structure. For example, for the monolayer MoS2 unit cell, its value is 3, for monolayer B2Te3, its value is 5.

ntypat: Number of types of atoms in the structure. For example, for both MoS2 and B2Te3, its value is 2 because there are only two types of atoms in the cell.

znucl: Atomic number of the different types of atoms in the cell. For example, for MoS2, **znucl** = 42 16. For B2Te3, **znucl** = 5 52. There should be a space between two **znucl** values.

typat: Type and numbers of atoms should be mentioned explicitly. For example, for MoS2, there are 1 Mo and 2 S atoms. So its value is 1 2 2. For BaTiO3: 1 2 3 3 3. For MnBi2Te4: 1 2 2 3 3 3 3 OR 1 2*2 4*3. Similarly, for B2Te3, this will be 1 1 2 2 2, each value separated by a comma.

nsppol: Number of spin polarization. nsppol = 1 (default) means no spin polarization.

ecut: Energy cutoff for the wavefunction expansion. This is equivalent to the ENCUT tag in the VASP. It is expressed in Hartree (Ha) and has no default value. I have used ecut = 15 Ha (equivalent to $15 * 27.2113845$ eV = 408.17 eV) in the calculation.

ngkpt: Number of grid points for K-points generation. This is equivalent to the KPOINTS file in VASP. I have used a 12 x 12 x 1 grid in the ground state calculation. However, the SHG calculation needs more denser grid, so I have used a 24 x 24 x 1 during the SHG calculation.

nshiftk: Number of shifts for K-points grids. The default value is 1. It gives the number of shifted grids to be used concurrently to generate the full grid of k points. It can be used with primitive grids defined either from **ngkpt** or **kptrlatt**. The maximum allowed value of **nshiftk** is 8. The values of the shifts are given by **shiftk**. The use of **nshiftk** =1, 2, or 4 is quite common, see the values suggested in the description of **shiftk**. The other values are either for debugging purposes by experts or can indicate an error. I have used the default value (i.e. 1) in the calculation.

shiftk: As mentioned in **nshiftk**, it determines the shift for K-points.The default value is None if **nshiftk** > 1 and [0.5, 0.5, 0.5] otherwise. The choice of **shiftk** depends upon the symmetry of the lattice. For the FCC lattice, we use **nshiftk** = 4 and **shiftk** will be
 0.5 0.5 0.5
 0.5 0.0 0.0
 0.0 0.5 0.0
 0.0 0.0 0.5
For the BCC lattice, we use **nshiftk** = 2 and **shiftk** will be
 0.25 0.25 0.25
 -0.25 -0.25 -0.25
For the hexagonal lattice, we use **nshiftk** = 1 and **shiftk** = 0.0 0.0 0.5. I have used the last option (i.e. hexagonal) in the calculation. Easy back-up: use **nshiftk** = 1 and **shiftk** = 0.0 0.0 0.0 to get a

non-shifted Monkhorst-Pack grid, which will be compatible with all symmetries, and is necessary for some features such as k-point interpolation.

kptopt: K-points option. The default is 4 if **nspden** = 4, and 1 otherwise. This means relying on **ngkpt** (grid) and taking fully into account the symmetry to generate the K-points in the Irreducible Brillouin Zone with the appropriate weights. (This is the usual mode for GS calculations). This value should be changed to a negative number when doing the band structure calculation.

diemac: Model dielectric macroscopic constant. The default value is 1000000.0. A rough knowledge of the macroscopic dielectric constant **diemac** of the system is a useful help to speed up the SCF procedure. The value of **diemac** should usually be bigger than 1.0, on physical grounds. For metals, simply put **diemac** to a very large value (the default is OK). For silicon, use 12.0. A similar value is likely to work well for other semiconductors. For wider gap insulators, use 2.0 … 4.0. For molecules in an otherwise empty big box, try 1.5 … 3.0. I have used its value of 12 in the calculation.

pawecutdg: PAW energy cutoff for the double grid and MUST be specified for PAW calculations. It is relevant only if PAW potentials are used. Its default value is -1. **pawecutdg** must be set larger or equal to **ecut**. If it is equal to **ecut**, then no fine grid is used. The results are not very accurate, but the computations proceed quite fast. For typical PAW computations, where **ecut** is of the order of 15 Ha, **pawecutdg** must be tested according to what we wanted to do. For calculations that do not require high accuracy (eg, molecular dynamics calculations), a value of 20 Ha is enough. For calculations that require high accuracy (eg, response functions calculations), it should be on the order of 30 Ha. I have used 30 Ha in the calculation to be on the safe side.

pp_dirpath: Address to the PAW pseudopotential files. For example, I have set **pp_dirpath** = "/home/rbhttri1/data-trevor42/rbhttri1/ABINIT_PPs/PAW/ATOMICDATA/" on rockfish cluster.

pseudos: Name of the pseudopotential files. For B2Te3 monolayer using LDA potentials, this will be "B.LDA_PW-JTH.xml, Te.LDA_PW-JTH.xml". It will be different for GGA PPs.

Below is the example file of monolayer B2Te3 (**mono_B2Te3.abi)** for relax calculation using **optcell** = 0.

acell   6.454601 6.454601 20.011829 angstrom

rprim   0.8660254 -0.5000000 0.0000000
*   0.0000000 1.0000000 0.0000000*

0.0000000 0.0000000 1.0000000

xred   0.666667 0.333333 0.163702
*   0.333333 0.666667 0.163557*
*   0.000000 0.323492 0.163629*
*   0.676508 0.676508 0.163629*
*   0.323492 0.000000 0.163629*

#nsym 0
optcell 0
ionmov 2
ecutsm 0.5
getwfk 0
chksymtnons 0

nstep 60
ntime 1000
 # toldff 1.0d-6
toldfe 1.0d-7
tolmxf 5.0d-5
natom 5
ntypat 2
znucl 5 52
typat 1 1 2 2 2

ecut 15
ngkpt 12 12 1
nshiftk 1
shiftk 0.0 0.0 0.5

kptopt 1
diemac 12
#pseudo PPs
pawecutdg 30
pp_dirpath "/home/rbhttri1/data-trevor42/rbhttri1/ABINIT_PPs/PAW/ATOMICDATA/"
pseudos "B.LDA_PW-JTH.xml, Te.LDA_PW-JTH.xml"

After the calculation is finished, search the string '**Calculation completed**' in the output file **mono_B2Te3.abo** and ***.out** file. Also, search for the line '**At Broyd/MD step XXX, gradients are converged:**' and make sure the MD step (**XXX** in the above line) is less than 1000 (max

value of **ntime** tag). This tells us that the calculation is converged at **XXX** ionic steps out of 1000 (**ntime** in the input file). If **XXX** = 1000, it means the convergence is reached and is only finished because the max number of **nstep** is reached. In that case, copy the **rprim**, and **xred** parameters from the output file (see at the end of the file), replace the initial values in the input file with the new ones, and rerun the same job. After the calculation is finally converged, go to the second step.

(b) optcell = 3
Copy the previous input files from **optcell = 0** run (i.e. **mono_B2Te3.abi**, and **submission script**), replace the parameters **rprim**, and **xred** with those obtained from the output of the previous run (from **mono_B2Te3.abo** file), and set **optcell** = 3 (equivalent to ISIF=4 in VASP). Submit the new job and see if it runs well or crashes instead. This job may crash if **dilatmax** is exceeded many times, meaning that the lattice vectors are changed a lot from the initial guess. We will see an error like this in the output file (i.e. *.out file)

> *--- !DilatmxError*
> *src_file: m_precpred_1geo.F90*
> *src_line: 256*
> *mpi_rank: 0*
> *message: |*
> *Dilatmx has been exceeded too many times (4)*
> *See the description of **dilatmx** and **chkdilatmx** input variables.*
> *Action: either first do a calculation with **chkdilatmx=0**, or restart your calculation with a larger **dilatmx**, or larger lattice vectors.*
> *Warning: With **chkdilatmx** = 0 the final computation of lattice parameters might be inaccurate.*

This error can be solved by agin following a two-step relax process using **optcell** = 3. First, run a job by setting **chkdilatmx** = 0 in the input file. After the job finishes, make a new input file by replacing the three parameters **acell, rprim, and xred** from the output file, and set **chkdilatmx** = 0 and **dilatmx** = 1.05 in the input file. The parameter **optcell** = 3 is already set in the previous step, so don't change it. Submit the job and see the output if the structure is relaxed. So in total, there are three steps to relax the structure: one with **optcell** = 0 and 2 with **optcell** = 3.

dilatmx: This is the max value of lattice dilation. I have used 1.05 in the calculation as per the ABINIT suggestion.

Step 2: Band structure calculations
The band structure calculation using ABINIT is a straightforward process. Unlike two separate input files and two separate runs in VASP, ABINIT uses a single input file and produces two separate datasets which should be included in the input file by setting **ndtset** = 2. Most of the

parameters are the same as in the relax calculation. I have explained the additional input tags below. We ask the ABINIT to first do a SCF calculation in the first run and produce the first set of datasets. Later, use those files in the second run (i.e. NSCF run) and produce the second set of datasets. The SCF run is defined by the following parameters: The numbers attached to the input tags (eg, the number 1 in the **kptopt1** tag) represent which dataset they belong to. It is only applicable for the multi-dataset ABINIT calculations.

#dataset 1: scf calculation
kptopt1 1 # (defined above)

nshiftk1 1 # (defined above)

shiftk1 0.0 0.0 0.5 # (defined above)

ngkpt1 12 12 1 # (defined above)

prtden1 1 # Print the charge density files. If set to 1 or a larger value, ABINIT provides the output of electron density in real space rho(r), in units of electrons/Bohr^3. If **ionmov** = 0, the name of the density file will be the root output name, followed by _DEN.
T
oldfe1 1.0d-10 # (defined above)

The NSCF run is defined by the following parameters:

##dataset 2: band structure calculation (Number 2 associated with each tags represent the second step of the calculation which produces the second set of datasets.)
iscf2 -2 # (iscf2 < 0 is for non-SCF calculation, -2 for band structure calculation)

getden2 -1 # (use from charge density from the previous dataset, equivalent to ICHARG = 1/11. Its default value is 0, meaning that the previous charge density file is not used)

kptopt2 -3 # (number of segments in the K-path. For hexagonal lattices, the K-path is M-G-K-M. So there are 3 segments, M-G, G-K, and K-M in the irreducible Brillouin zone.

nband2 36 # (search **nband** in the output file of relax calculation and multiply that value by at least 1.4. The main purpose is to make sure we provide a sufficient number of empty bands.)

ndivsm2 20 # (number of points or divisions in each segment M-G, G-K, K-M. If more dense points are needed in the band structure, just increase that value to 40,50…)

nshiftk2 1 # (defined above)

shiftk2 0 0 0 # (do not shift the K-points in the second step)

kptbounds2 0.50000 0.00000 0.00000 # M point
            0.00000 0.00000 0.00000 # Gamma point
            0.33333 0.33333 0.00000 # K point
            0.50000 0.00000 0.00000 # M point

kptbounds represents the actual high-symmetric points in the Brillouin zone for band structure calculation. Thus it depends upon the type of lattices.

tolwfr2 1.0d-12 # (Tolerance on wavefunction squared residual. Its default value is 0. I have set 1E-12 Hartree as a criterion for non-SCF calculation)

enunit2 1 # (**enunit** is energy unit. 0 means in Hartree, 1 means eV and 2 means print the energy in both units. I am setting it to 1 to get the output eigenenergies in eV.)

prtebands2 2 # (Print the electronic bands. Its default value is 1 unless **nimage** > 1 is set. Setting **prtebands2** = 0 means disabling the output of band energies, and 1 means outputting in the Xmgrace format. A file with the extension EBANDS.agr is produced at the end of the run. Use Xmgrace file_EBANDS.agr to visualize the band energies. 2 is for Gnuplot format. The code produces an EBANDS.dat file with the eigenvalues and a file_EBANDS.gnuplot script. Use Gnuplot file_EBANDS.gnuplot to visualize the band energies. I chose the last option because ABINIT also prints the EBANDS.data file in that case and we can manually plot this data using any packages.

Below is an example file of monolayer B2Te3 (**mono_B2Te3_band.abi**) for calculating the band structure and band gap using ABINIT.

ndtset 2

acell   1.1120307982E+01 1.1120308019E+01 4.5497607328E+01 Bohr

rprim   8.6602540284E-01 -5.0000000164E-01 0.0000000000E+00
*   0.0000000000E+00 1.0000000000E+00 0.0000000000E+00*
*   0.0000000000E+00 0.0000000000E+00 1.0000000000E+00*

xred   6.6666673412E-01 3.3333323936E-01 1.6362949360E-01
*   3.3333340078E-01 6.6666657269E-01 1.6362949360E-01*
*   6.7449999975E-08 2.9900746157E-01 1.6362949360E-01*

```
        7.0099251190E-01  7.0099235047E-01  1.6362949360E-01
        2.9900762300E-01 -9.3976250040E-08  1.6362949360E-01


natom 5 # number of atoms
ntypat 2 # types of atoms
znucl 5 52   # atomic number Al->13, Sb->51
typat 1 1 2 2 2 # for BaTiO3: 1 2 3 3 3. Also 1 2*2 for MoS2 because Mo:1, S1 and S2:2. If there
is MnBi2Te4: 1 2*2 4*3
chksymtnons 0


ecut 15 # Encut in vasp (1 Ha = 27.2113845 eV)
diemac 12.0
pawecutdg 30
#pawfatbnd 1 # orbital separated band


#dataset 1: scf calculation
kptopt1 1
nshiftk1 1
shiftk1 0.0 0.0 0.5
ngkpt1 12 12 1 # KPOINTS


prtden1 1
toldfe1 1.0d-10


##dataset 2: band structure calculation
iscf2 -2 # non-SCF calculation
getden2 -1 # use from charge density from previous dataset
kptopt2 -3 # 3 segments, M-G, G-K, K-M
nband2 36 # 14 in general for MoS2, 14*1.4
ndivsm2 20


nshiftk2 1
shiftk2 0 0 0 # No shift
kptbounds2  0.50000  0.00000  0.00000 # M point
        0.00000  0.00000  0.00000 # Gamma point
        0.33333  0.33333  0.00000 # K point
        0.50000  0.00000  0.00000 # M point


tolwfr2  1.0d-12 # criterion for non-SCF
enunit2  1 # output eigenenergies in eV
```

prtebands2 2

#pseudo PPs
pp_dirpath   "/home/rbhttri1/data-trevor42/rbhttri1/ABINIT_PPs/PAW/ATOMICDATA/"   #
pseudo dir
pseudos "B.LDA_PW-JTH.xml, Te.LDA_PW-JTH.xml"

ndtset: If we want to run multiple calculations using a single input file, the ABINIT will add a subscript at the end of each file to separate them.

After the calculation finishes, a file named '**mono_B2Te3_bando_DS2_EBANDS.data**' will be generated. When I plot this file using Xmgrace , I get the following band structure:
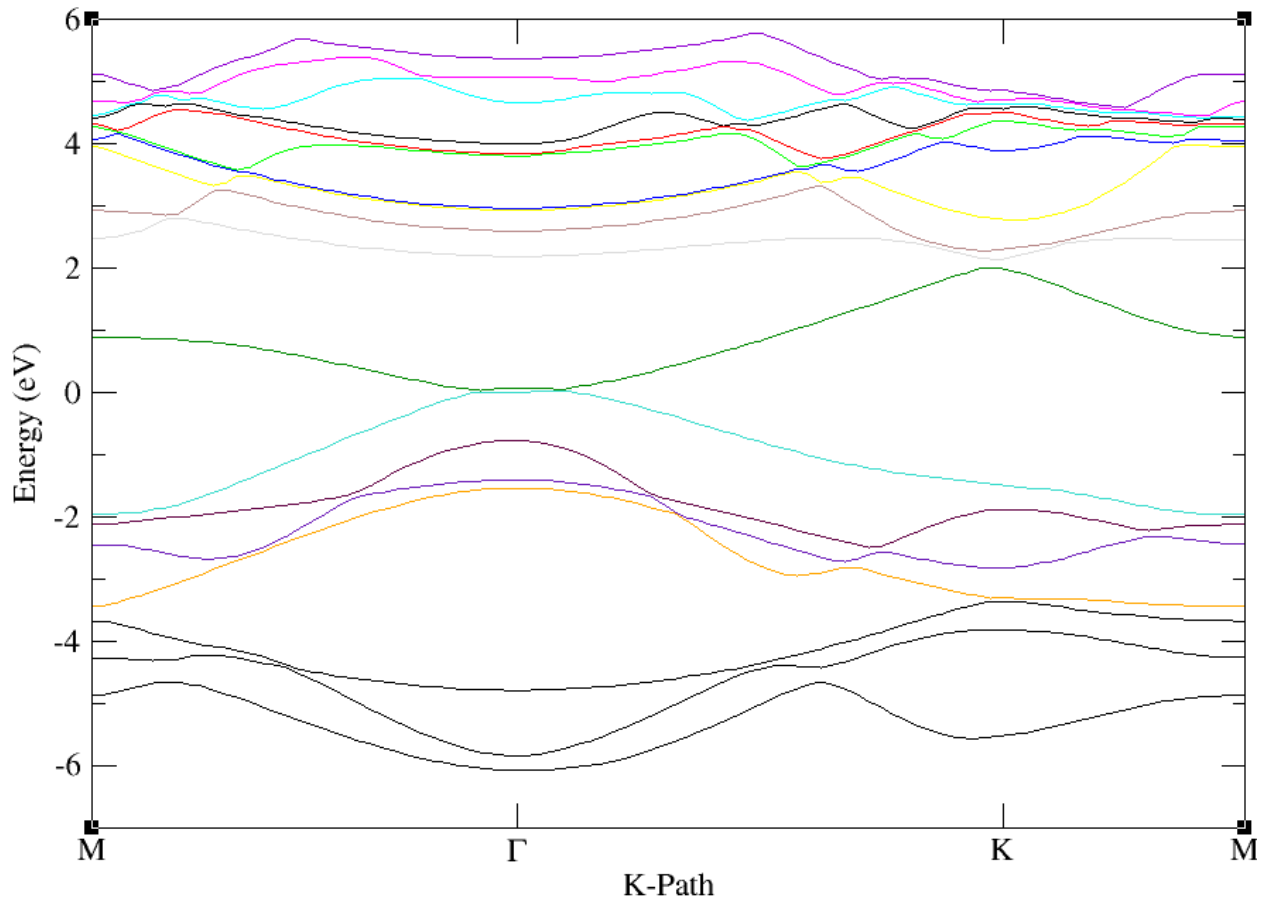

Figure 1. Band structure of monolayer B2Te3 using ABINIT and LDA pseudopotential.

Step 3: SHG calculations

There are two steps in the SHG calculation. In the first step, we create the necessary files that will be used in the second step while producing the specified linear and non-linear optical

components. The first calculation will go through six sub-steps and will create output files in each sub-step based on the input tags provided in the **mono_B2Te3_shg.abi** file.

ndtset: The number of datasets to be created. We use **ndtset** = 6 in the SHG calculations. That is, six steps of calculations with six different output datasets will be performed.

Input tags for the first dataset

nbands1: Search '**nband**' in the output file of the relax calculation. Use **nbands1** at least more than 2 times **nband** to consider enough unoccupied bands for accurate SHG results. I have used **nbands1** = 30 (> 13 (nbands)*2) in the calculation.

nstep1: NELM, number of electronic steps. I have already described this above. Make this value sufficient enough so that there won't be any convergence issue because **nstep1** is smaller.

kptopt1: Already defined above.

nbdbuf1: It gives the number of bands, the highest in energy, that, among the **nband** bands, are to be considered as part of a buffer. A negative value is interpreted as a percentage of **nband**. The purpose of this tag is to facilitate the convenience. It is recommended to set 10% of **nband1**. I have used 3 (i.e. 10% of 30) in the calculation.

prtden1: Print the charge density file, which is already explained above. The file created in this step will be used in the following steps.

getden1: Get the density from. For the first set of calculations, it should be set to 0 because there is no such file to read at the beginning. If **getden** = 0, no such use of the previously computed output density file is done. If **getden** is positive, its value gives the index of the dataset from which the output density is to be used as input. However, if the first dataset is treated, -1 is equivalent to 0, since no dataset has been computed in the same run. If **getden** is -1, the output density of the previous dataset must be taken, which is a frequently occurring case. If **getden** is a negative number, it indicates the number of datasets to go backward to find the needed file. Going back beyond the first dataset is equivalent to using zero for the get variable.

getwfk1: Get the wave functions from the WFK file. It also works for the wavefunction the same way that **getden1** works for the density.

Input tags for the second dataset

iscf2: Already described above. Since this step is an NSCF step, so iscf should be set to -2.

nstep2: Same as step 1

kptopt2: Same as step 1

getwfk2: Get the wave functions from the WFK file. Starting this step, we will need the wavefunctions file. So it should be set to 1.

getden2: Same as getwfk2 for charge density. It is set to 1.

Input tags for the third dataset

Everything is the same as in step 2, except the number 3 is used as an extension instead of 2 in the input tags.

iscf3 -2

nstep3 200

getwfk3 2

getden3 1

Starting with the 4th dataset, the ddk response functions (d/dk) will be calculated along three different axes.

Input tags for the fourth dataset:

iscf4: It is already explained above. For the response functions calculation, we should use -3. It is similar to -2 but initializes **occ** and **wtk**, directly or indirectly depending on the value of **occopt**. This option is needed to compute the response to ddk perturbation.

nstep4: Explained above. Starting from the fourth step, we just run one NSCF calculation. So it should be set to 1.

nline4: The number of line minimization. Its default value is 4. We use **nline** = 0 to diagonalize the Hamiltonian matrix in the subspace spanned by the input wavefunctions. This is recommended for the response function calculations.

prtwf4: Print the wavefunctions. We use **prtwf** = 3. In this case, the file that is created is nearly the same as with **prtwf** = 1, except that the records that should contain the wavefunction are empty (so, such records exist, but store nothing). This is useful to generate size-reduced ddk files, to perform an optic run. So, only matrix elements are needed and no wavefunction, but possibly a large number of conduction bands.

nqpt4: Number of q points. It determines whether one q-point value should be read, having a default value of 0. We set **nqpt4** = 1 in the SHG calculation. It defines the wavevector of the perturbation in the response function calculations.

rfdir4: Direction of response function. 1: direction considered, 0: not considered. We set **rfdir4** to 1 0 0 in the fourth step of the SHG calculation. It means d\dk along the axis is computed.

rfelfd4: Response function with respect to the electric field. The default value is 0, meaning no electric field perturbation. 1 means full calculation, with first the derivative of the ground-state wavefunction with respect to k (i.e. *d/dk*) calculation, by a non-self-consistent calculation, then generation of the first-order response to a homogeneous electric field. 2 means only d/dk calculation, 3 means only the generation of the first-order response to the electric field, assuming that the data on d/dk is available. **In case a scissor shift is to be used, it is usually not applied for the response.** We have used 2 in the calculation.

Input tags for the fifth dataset

Same as step 4 except, the direction is 0 1 0 (i.e. y-axis) and the number 5 is used as the extensions instead of 4 in the input tags.
iscf5 -3
nstep5 1
nline5 0
prtwf5 3
nqpt5 1
rfdir5 0 1 0
rfelfd5 2

Input tags for the sixth dataset

Same as step 4 except, the direction is 0 0 1 (i.e. z-axis) and the number 6 is used as the extensions instead of 4 in the input tags.
iscf6 -3
nstep6 1
nline6 0
prtwf6 3
nqpt6 1
rfdir6 0 0 1
rfelfd6 2

Common input tags to all datasets:

In addition to the above dataset-specific tags, there are a lot of other input tags that are common to some datasets:

Input tags common to datasets 2-6

nband 100: The number of bands should be set high for SHG calculation. 100 should be enough for the B2Te3 monolayer according to the ABINIT manual.

chksymtnons 0: Already described above.

Input tags common to datasets 3-6

kptopt 3: According to ABINIT wiki, 3 means to rely on **ngkpt** as well as on **nshiftk** and **shiftk** to set up the K-points. Do not take into account any symmetry. K-points will be generated in the full Brillouin zone with appropriate weights. This is the usual mode when preparing or executing an RF calculation at non-zero q, or with non-collinear magnetism.

Input tags common to datasets 4-6

getwfk 3: Gives the index of the dataset (i.e. 3 here) for which the output wavefunction file appended with _WFK must be used. i.e. A file named **mono_B2Te3_shgo_DS3_WFK** will be generated in the third step (where **getwfk3** = 2 is set) and will be used in steps 4-6.

In addition, several input tags are common to all the datasets.

Input tags common to all datasets
The input tatgs common to all the datasets are **nshiftk, shiftk, ngkpt, qpt, acell, rprim, xred, natom, ntypat, znucl, typat, ecut, diemac, pawecutdg, nbdbuf, tolwfr, pp_dirpath, pseudos.** These tags have already been explained above.

Below is the example input file '**mono_B2Te3_shg.abi**' for the first step of SHG calculation:

ndtset 6 # (six steps of calculations with six different output datasets)

#First dataset : SC run with kpoints in the IBZ
nband1 30
nstep1 200
kptopt1 1
nbdbuf1 3 # 10 % of nband1

prtden1 1   getden1 0   getwfk1 0   # Usual file handling data

#Second dataset : NSC run with large number of bands, and points in the IBZ
  iscf2 -2
  nstep2 200
  kptopt2 1
  getwfk2 1   getden2 1   # Usual file handling data

#Third dataset : NSC run with large number of bands, and points in the full BZ
  iscf3 -2
  nstep3 200
  getwfk3 2   getden3 1   # Usual file handling data

#Fourth dataset : ddk response function along axis 1
  iscf4 -3
   nstep4   1   nline4   0   prtwf4   3   # Speed up the calculation : the first-order response wavefunction is not computed, only the matrix elements.

  nqpt4 1
  rfdir4 1 0 0
  rfelfd4 2

#Fifth dataset : ddk response function along axis 2
  iscf5 -3
   nstep5   1   nline5   0   prtwf5   3   # Speed up the calculation : the first-order response wavefunction is not computed, only the matrix elements.

  nqpt5 1
  rfdir5 0 1 0
  rfelfd5 2

#Sixth dataset : ddk response function along axis 3
  iscf6 -3
   nstep6   1   nline6   0   prtwf6   3   # Speed up the calculation : the first-order response wavefunction is not computed, only the matrix elements.

  nqpt6 1
  rfdir6 0 0 1
  rfelfd6 2

 # Data common to datasets 2-6

nband   100   # This number of bands might be too low for non-linear optics and real part of linear optics

chksymtnons 0

 # Data common to datasets 3-6

kptopt   3   # Need points in the full BZ for non-linear optical properties

 # Data common to datasets 4-6

getwfk 3

#Data common to all datasets

nshiftk 1

shiftk 0.0 0.0 0.5

ngkpt 24 24 1   # 48 in Kory Beach, but trying smaller first

qpt 0.0 0.0 0.0   # Not really needed, because it is the defaults, but mentioned here to keep track of its importance for datasets 4-6

acell   1.1120307982E+01 1.1120308019E+01 4.5497607328E+01 Bohr

rprim   8.6602540284E-01 -5.0000000164E-01 0.0000000000E+00
0.0000000000E+00 1.0000000000E+00 0.0000000000E+00
0.0000000000E+00 0.0000000000E+00 1.0000000000E+00

xred   6.6666673412E-01 3.3333323936E-01 1.6362949360E-01
3.3333340078E-01 6.6666657269E-01 1.6362949360E-01
6.7449999975E-08 2.9900746157E-01 1.6362949360E-01
7.0099251190E-01 7.0099235047E-01 1.6362949360E-01
2.9900762300E-01 -9.3976250040E-08 1.6362949360E-01

natom 5 # number of atoms

ntypat 2 # types of atoms

znucl 5 52   # atomic number

*typat 1 1 2 2 2 # for BaTiO3: 1 2 3 3 3. Also 1 2*2 for MoS2 because Mo:1, S1 and S2:2. If there is MnBi2Te4: 1 2*2 4*3*

ecut 15 # Encut in vasp (1 Ha = 27.2113845 eV)

diemac 12.0

pawecutdg 30

nbdbuf 10 # 10% of nband

tolwfr   1.e-20 # This tolerance factor should be made sufficiently small to get more accurate results.

pp_dirpath   "/home/rbhttri1/data-trevor42/rbhttri1/ABINIT_PPs/PAW/ATOMICDATA/"   # pseudo dir
pseudos "B.LDA_PW-JTH.xml, Te.LDA_PW-JTH.xml"

After this calculation finishes, we go to the second step. In this step, we use '**optic**' to calculate the SHG spectra. See the script file (in the example folder) for details. The input tags of "**mono_B2Te3_optic.abi**" are explained below (https://docs.abinit.org/variables/optic/):

The input tags are divided into three categories: &FILES, &PARAMETERS, and &COMPUTATIONS. The category '&FILES' contains the following tags:

ddkfile: This parameter specifies the name of the file containing the matrix elements of the d/dk operator in the specific directions. For example, ddkfile_1 belongs to the x-axis, and so on. This file should have been produced by the previous ABINIT run. This file may be generated using **prtwf** 3. In the previous run, I set **prtwf4**, **prtwf5**, and **prtwf6** to 3, asking the code to generate three different ddk files along three axes. These files will be used in the second step as follows:
ddkfile_1: The ddkfile produced in fourth step (100)
ddkfile_2: The ddkfile produced in fifth step (010)
ddkfile_3: The ddkfile produced in sixth step (001)

wfkfile: This is the wavefunction K file, which was also already generated in the previous run by setting **getwfk3** = 2 in the third step. This parameter specifies the name of the ground state wavefunction file. It should include both the valence and conduction states.

The category '&PARAMETERS' contains the following tags:

broadening: It applies a broadening to the spectrum and is used to avoid divergences in the sum-over-states approach. The sum-over-states approach to the linear and nonlinear susceptibilities inevitably includes resonant denominators of the form $(\omega - \omega_{nm})^{-1}$ that might lead to infinities. To avoid the singularities, we add a small imaginary contribution to the denominator. This addition prevents the denominator from ever going to 0 and acts as a broadening to the spectrum. I have used its value of 0.001 in the calculation.

domega: It specifies the step size $\Delta\omega$ for the grid over which the optic utility computes the susceptibilities. To capture more features, decrease $\Delta\omega$ to get a finer energy grid. I have used $\Delta\omega$ = 0.0001 in the calculation.

maxomega: It specifies the maximum energy for the grid over which the optic utility computes the susceptibilities. The grid step size is set by the companion parameter **domega**. The susceptibilities are thus computed at **maxomega/domega** energy points (zero excluded). To capture more features, decrease $\Delta\omega$ to get a finer energy grid. To include higher frequency, increase the maximum. I have used **maxomega** = 0.5 in the calculation.

scissor: Scissor correction. It provides a fixed shift to all the conduction bands. As LDA/GGA are known to underestimate the band-gap by a significant amount in some cases, to obtain a reasonable optical spectrum and make a realistic comparison with experiments one needs to correct for this. The scissors shift is normally chosen to be the difference between the experimental and theoretical band gap and simply shifts the conduction bands. Alternatively, one may determine the self-energy using the GW approach, in which case the opening of the gap due to the GW correction can be used as the scissor shift. In the example run, I haven't used the scissor correction. So I set this to 0.000.

tolerance: This parameter sets a scale for discarding small energy denominators. When energy denominators are smaller than **tolerance**, the term is discarded from the sum. I have set this value to 0.001in the calculations.

The category '&COMPUTATIONS' contains the following tags:

num_lin_comp: Total number of components of the linear optical dielectric tensor to calculate. There are a total of 9 such components. Some of these may be either equal to each other, or zero, depending upon the symmetry of the material. I have set all (9) in the calculation. Its default value is 0.

lin_comp: What are those linear components? In other words, it specifies the directions of the **num_lin_comp** requested components of the dielectric tensor. The components are specified in cartesian coordinates, where 1, 2, and 3 represent x, y, and z respectively. For example, 11 represents the xx component, and 32 represents zy. These components should match the number specified in **num_lin_comp**. I have set all 9 components in the calculation.

num_nonlin_comp: Similar to linear components, this tag sets the number of non-linear components of the second-order nonlinear optical tensor to calculate. There are a total of 27 such components. I have set all 27 in the calculation because it doesn't hurt to include all if you are

not sure what is the symmetry of the structure and what are the expected non-zero components. Its default value is 0.

nonlin_comp: Similar to linear components, It specifies the directions of the **num_nonlin_comp** requested components of the second-order nonlinear dielectric tensor. The components are specified in cartesian coordinates, where 1, 2, and 3 represent x, y, and z respectively. For example, 111 represents the xxx component, and 321 represents zyx. I have specified all 27 components in the calculation.

num_linel_comp: It specifies how many components (out of 27 possible) of the linear electro-optical susceptibility to calculate. I am not interested in calculating these components. So I set this to 0.

An example file of monolayer B2Te3 '**mono_B2Te3_optic.abi**' is shown below:

&FILES
ddkfile_1 = 'mono_B2Te3_shgo_DS4_1WF16',
ddkfile_2 = 'mono_B2Te3_shgo_DS5_1WF17',
ddkfile_3 = 'mono_B2Te3_shgo_DS6_1WF18',
wfkfile = 'mono_B2Te3_shgo_DS3_WFK'
/
&PARAMETERS
broadening = 0.001,
domega = 0.0001,
maxomega = 0.5,
scissor = 0.000,
tolerance = 0.001
/
&COMPUTATIONS
num_lin_comp = 9,
lin_comp = 11,22,33,12,21,13,31,23,32,
num_nonlin_comp = 27,
nonlin_comp = 111, 112,113,121,122,123,131,132,133,211,212,213,221,222,223,231,232,233,
311, 312, 313,321,322,323,331,332,333,
num_linel_comp = 0,
num_nonlin2_comp = 0,
/

After this calculation finishes, there will be 9 linear optical components (mono_B2Te3_optic_0001_0001-linopt.out for xx directions and so on) produced by the

ABINIT. Also, 27 non-linear components with each having 7 different files will also be produced. For example, the 7 filenames along xxx direction are mono_B2Te3_optic_0001_0001_0001-(ChiAbs.out, ChiImDec.out, ChiIm.out, ChiReDec.out, ChiRe.out, ChiTotIm.out, ChiTotRe.out). For calculating the absolute value of SHG spectra, we only look at the *-ChiAbs.out file. This file contains three columns: 1st column is energy in eV, the second column is Total Chi_2 in 10^(-7) esu or electrostatic unit, and the third column is Total Chi_2 in 10^(-12) m/V or pm/V. We plot the first (x-axis) and third columns (y-axis) to show the SHG results in SI units. We can also change it to nm/V by dividing the third column or y-axis by 1000. See this documentation for details: https://docs.abinit.org/guide/optic/

For calculating the interband and intraband transitions, we may need to play with *-(ChiIm.out and ChiRe.out) files. They seem to contain the interband and intraband information but their formatting looks confusing. Also, it is not clearly explained in the ABINIT manual.

A table showing all the nonvanishing tensor elements of $\chi^{(2)}$ for all 32 crystal classes is shown below. It should be noted that for some materials, such as monolayer B2Te3, using structural information in the default POSCAR format, where the lattice vector **a** is parallel to the x-axis, may print the Chi_2 components with x and y indices interchanged. In other words, the hexagonal systems with -6m2 symmetry should have the following non-zero Chi_2 components: yyy = yxx = xxy =xyx. This works perfectly for 2H-MoS2 and hBN monolayers. However, for monolayers B2Te3, B2Se3 B2S3, and all other structures that belong to this particular atomic configuration, the above rule doesn't seem to hold. Instead, we get the following non-zero Chi_2 components: xxx = yyx = xyy = yxy. To solve this issue, one needs to rotate the lattice parameters such that the lattice vector **b** is parallel to the y-axis instead of **a** parallel to the x-axis. See **rprim** in the above example file for details.

To convert the effective bulk susceptibility in nmV^{-1}. We should multiply the intensity of the SHG spectra (i.e. y-axis) by a dimensionless quantity L_z /L_{eff}, where L_z is the length of the simulation cell along the z-axis or lattice vector Cz. L_{eff} is the effective thickness of the cell and is obtained by subtracting the z-components of atoms (in cartesian coordinates) situating at the top and bottom of the crystal structure and adding the result to their respective van der Waals radius.

I have provided all the input files (relax calculations, band structure calculations, and SHG calculations) of monolayer B2Te3 and the job submission scripts in the example folder along with this manual. For the pseudopotential files, one can always download the ABINIT package from the website and unzip it.

Table 1: Nonvanishing tensor elements of $\chi^{(2)}$ for all 32 crystal classes denoted by Cartesian indices. Source: Korey Beach Dissertation.

Crystal System	Crystal Class	Nonvanishing Tensor Elements
Triclinic	$1 = C_1$	All elements are independent and nonzero
	$\bar{1} = S_2$	All elements vanish
Monoclinic	$2 = C_2$	$xyz, xzy, xxy, xyx, yxx, yyy, yzz,$
		$yzx, yxz, zyz, zzy, zxy, zyx$
	$m = C_{1h}$	$xxx, xyy, xzz, xzx, xxz, yyz, yzy,$
		$yxy, yyx, zxx, zyy, zzz, zzx, zxz$
	$2/m = C_{2h}$	All elements vanish
Orthorhombic	$222 = D2$	$xyz, xzy, yzx, yxz, zxy, zyx$
	$mm2 = C_{2v}$	$xzx, xxz, yyz, yzy, zxx, zyy, zzz$
	$mmm = D_{2h}$	All elements vanish
Tetragonal	$4 = C_4$	$xyz = yxz, xzy = yzx, xzx = yzy,$
		$xxz = yyz, zxx = zyy, zzz, zxy = zyx$
	$\bar{4} = S_4$	$xyz = yxz, xzy = yzx, xzx = yzy,$
		$xxz = yyz, zxx = zyy, zxy = zyx$
	$422 = D_4$	$xyz = yxz, xzy = yzx, zxy = zyx$
	$4mm = C_{4v}$	$xzx = yzy, xxz = yyz, zxx = zyy, zzz$
	$\bar{4}2m = D_{2d}$	$xyz = yxz, xzy = yzx, zxy = zyx$
	$4/m = C_{4h}$	All elements vanish
	$4/mmm = D_{4h}$	All elements vanish
Cubic	$432 = O$	$xyz = xzy = yzx = yxz = zxy = zyx$
	$\bar{4}3m = T_d$	$xyz = xzy = yzx = yxz = zxy = zyx$
	$23 = T$	$xyz = yzx = zxy, xzy = yxz = zyx$
	$m3 = T_h, m3m = O_h$	All elements vanish
Trigonal	$3 = C_3$	$xxx = xyy = yyz = yxy, xyz = yxz, xzy = yzx,$
		$xzx = yzy, xxz = yyz, yyy = yxx = xxy = xyx,$
		$zxx = zyy, zzz, zxy = zyx$
	$32 = D_3$	$xxx = xyy = yyx = yxy,$
		$xyz = yxz, xzy = yzx, zxy = zyx$
	$3m = C_{3v}$	$xzx = yzy, xxz = yyz, zxx = zyy,$
		$zzz, yyy = yxx = xxy = xyx$
	$\bar{3} = S_6, \bar{3}m = D_{3d}$	All elements vanish
Hexagonal	$6 = C_6$	$xyz = yxz, xzy = yzx, xzx = yzy,$
		$xxz = yyz, zxx = zyy, zzz, zxy = zyx$
	$\bar{6} = C_{3h}$	$xxx = xyy = yxy = yyx,$
		$yyy = yxx = xyx = xxy$
	$622 = D_6$	$xyz = yxz, xzy = yzx, zxy = zyx$
	$6mm = C_{6v}$	$xzx = yzy, xxz = yyz, zxx = zyy, zzz$
	$\bar{6}m2 = D_{3h}$	$yyy = yxx = xxy = xyx$
	$6/m = C_{6h}$	All elements vanish
	$6/mmm = D_{6h}$	All elements vanish