

# DCDFTBMD USER MANUAL

Version 1.0

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## **Abstract**

DCDFTBMD allows the users to perform quantum mechanical molecular dynamics (MD) simulation for large systems by using divide-and-conquer (DC) density functional tight-binding (DFTB) method. The scaling of DC-DFTB with respect to system size is close to linear. A high parallel efficiency has been achieved in the implementation with the hybrid of MPI and OpenMP. The program has functionalities not only for MD but also for geometry optimization, vibrational frequency calculation, and property evaluation.

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# 1 Input for DCDFTBMD

To run the program **DCDFTBMD**, you need to prepare the input file and parameters (Slater-Koster files). The input file, which is named “**dftb.inp**” by default, mainly consists of 4 parts below:

- Information about the calculation (Keyword section)
- Brief description of the calculation (Title section)
- Specification of Slater-Koster parameters (Parameter section)
- Specification of molecular coordinates (Geometry section)

Each section is terminated with blank line (starting new line is indicated as ↵ in this document). For example, the input file looks like as follows in the case of SCC-DFTB single point energy calculation of water molecule:

```
SCC=TRUE_DC=FALSE↵ Keyword section
↵
TITLE↵ Title section
↵
2↵ Parameter section
O_2↵
oo.spl_oh.spl↵
H_1↵
ho.spl_hh.spl↵
↵
3_0_1↵ Geometry section
O _ 0.06275391_ 0.06275391_ 0.00000000↵
H _ 1.01910555_-0.08185946_ 0.00000000↵
H _-0.08185946_ 1.01910555_ 0.00000000↵
↵
```

Additional input may be required after geometry section if subsystems are specified manually in DC approach (see [section 1.4.3](#)). Note that blank area (represented as \_ in this document) should be written with spaces (**DO NOT USE TABS!**).

## 1.1 Keyword section

SCC=TRUE\_DC=FALSE ↵

*SCC and DC keywords are  
turned on and off*

The present example specifies two keywords in a single line. The same specification is possible by giving two separated lines as follows:

SCC=TRUE ↵

DC=FALSE ↵

*SCC keyword is turned on*

*DC keyword is turned off*

The following subsections account for the available keywords at present. Two keywords SCC and DC are activated if no keywords are given in the input. Other keywords can be turned on using the following forms:

*keyword=TRUE*

The above specification employs the default value of options in curly bracket. Several keywords have options for controlling the calculation. The italic type in parenthesis stands for the data type of option. The options can be specified by the following forms:

- *keyword=(option1=value1\_option2=value2\_...)*
- *keyword=(option1=value1)*  
*keyword=(option2=value2\_...)*

If option is written in **red** or **blue** character, that option is available in only serial and OpenMP or MPI/OpenMP version, respectively.

### 1.1.1 SCC

The keyword SCC sets whether a self consistent charge (SCC) calculation is made or not. As already mentioned, the default is `SCC=TRUE`, namely the SCC-DFTB (DFTB2) [1] calculation is performed.

- `MAXITER` (*integer*) {200}  
Maximum number of SCC cycles to reach convergence.
- `BROYITER/MAXBROYDEN` (*integer*) {70}  
Maximum number of charge mixing using Broyden method.
- `ENERGYCONV/ECONV` (*real*) {1.0e-09}  
Convergence criteria for the energy change between two SCC cycles in atomic unit.
- `DENSITYCONV/DCONV` (*real*) {1.0e-06}  
Convergence criteria for the maximum difference in any charge density between two SCC cycles in atomic unit.
- `READCHARGE` (*method*) {NONE}  
Enables to read initial charge information from file.
  - NONE constructs initial Mulliken population based on the number of valence electrons.
  - BINARY reads Mulliken population from the binary file “**chrgfile**”. If this option is set but “**chrgfile**” is not found in the same directory as the one including the input file, this option will be turned off.
  - ASCII reads net atomic charges from the ASCII file “**charge.dat**”. If this option is set but “**charge.dat**” is not found in the same directory as the one including the input file, this option will be turned off.

Examples for water molecule where oxygen corresponds to atom label 1 are given below.

For serial and OpenMP versions, the first line of “**charge.dat**” is either “**ALLATOM**” or “**ATOMSPECIES**”.

- ✓ **ALLATOM** lists the net charge for every atom from the second line.

```
ALLATOM.┘
```

-0.6	<i>Mulliken population of O (atom label 1) is 6.6</i>
0.3	<i>Mulliken population of H (atom label 2) is 0.7</i>
0.3	<i>Mulliken population of H (atom label 3) is 0.7</i>

- ✓ **ATOMSPECIES** lists the chemical symbol and the net charge in the first and second columns from the second line until detecting blank line if any.

ATOMSPECIES	
O <sub>1</sub> -0.6	<i>Mulliken population of O (atom label 1) is 6.6</i>
H <sub>2</sub> 0.3	<i>Mulliken population of H (atom labels 2 and 3) is 0.7</i>

For MPI/OpenMP version, the first line of “**charge.dat**” is either “**ALLSHELL**” or “**SHELLSPECIES**”.

- ✓ **ALLSHELL** lists the net charge for every shell from the second line.

ALLSHELL	
0.2	<i>Mulliken population of s shell of O (atom label 1) is 1.8</i>
-0.8	<i>Mulliken population of p shell of O (atom label 1) is 4.8</i>
0.3	<i>Mulliken population of s shell of H (atom label 2) is 0.7</i>
0.3	<i>Mulliken population of s shell of H (atom label 3) is 0.7</i>

- ✓ **SHELLSPECIES** lists the chemical symbol, subshell label (s/p/d), and

the net charge in the first, second, and third columns from the second line until detecting blank line if any. If system is open-shell, alpha/beta spin (a/b) needs to be specified in the fourth column.

#### SHELLSPECIES↓

O\_s\_ 0.2↓

Mulliken population of s shell of O (atom label 1) is 1.8

O\_p\_-0.8↓

Mulliken population of p shell of O (atom label 1) is 4.8

H\_s\_ 0.3↓

Mulliken population of s shell of H (atom labels 2 and 3) is 0.7

- DAMPXH (*logical*) {FALSE}
 

Enables the use of damped  $\gamma$ -function for X–H pairs. [2–4]
- DAMPXHZETA (*real*) {4.0e+00}
 

Empirical parameter for the damped  $\gamma$ -function for X–H pairs.
- THIRDDIAG (*logical*) {FALSE}
 

Enables the use of third order corrections only for diagonal terms. [3]

Hubbard derivatives need to be specified in Parameter section (see [section 1.3.2](#)).
- THIRDFULL (*logical*) {FALSE}
 

Enables the use of full third order corrections. [4]

Hubbard derivatives need to be specified in Parameter section (see [section 1.3.2](#)).  
The so-called DFTB3 [4] requires DAMPXH=TRUE.
- SPIN (*logical*) {FALSE}
 

Enables to perform collinear spin polarized calculation. [5, 6]

Spin constants have to be stored in different file for each atom (see [section 1.3.2](#)).
- ALMIX (*real*) {0.2e+00}
 

Charge mixing parameter used in charge mixing method ( $0 \leq \text{ALMIX} \leq 1$ ).
- MIXER (*integer*) {1}
 

Charge mixing method to accelerate SCC convergence.

Note that modified Broyden mixing is used to solve coupled perturbed DFTB equation in analytical Hessian calculation.

- 1: Modified Broyden [\[7\]](#)
- 2: Simple mixing
- 3: Modified Anderson [\[8\]](#)
- 4: DIIS [\[9\]](#)
  
- NGEN (*integer*) {4}  
Number of iteration to start Anderson and DIIS charge mixing.
  
- SOLVER (*integer*) {1}  
Eigensolver for diagonalization of Hamiltonian matrix.
  - 1: DSYGVD
  - 2: DSYGV
  - 3: DM\_VGEVPH  
This may be available if the platform is the K computer, Fujitsu FX10, and Fujitsu FX100.
  
- EFIELD (*logical*) {FALSE}  
Enables to include external static electric field.  
This is available for only cluster models.  
Static electric field vector is set after atomic coordinates starting from the character “EF” (see [section 1.4.1](#)).
  
- POLAR (*logical*) {FALSE}  
Enables to calculate dipole moment and static polarizability using external static electric field.  
This is available for only cluster models.
  
- POLEFSTR (*real*) {2.0e−03}  
Strength of external static electric field used in dipole moment and static polarizability calculation.
  
- POINTCHARGE (*logical*) {FALSE}  
Enables to include point charges.  
Coordinates and charge value are set after atomic coordinates starting from the character “PC” (see [section 1.4.1](#)).
  
- NUMPC (*integer*) {0}  
Number of point charges in system.



- **MAYER** (*logical*) {FALSE}  
Enables to perform Mayer bond order analysis. [[10](#)]
- **CM3** (*logical*) {FALSE}  
Enables to perform CM3 charge analysis. [[11](#), [12](#)]
- **ORBITALPOP/ORBPOP** (*logical*) {FALSE}  
Enables to print Mulliken population of each orbital.
- **ZEROCHARGE** (*logical*) {FALSE}  
Enables to permanently set values of neutral atoms as initial charge guess in SCC calculation during geometry optimization and molecular dynamics.  
Deactivation uses the converged charges of previous geometry step as initial guess (better than zero charge guess).
- **PSEUDODIAG/PDIAG** (*logical*) {FALSE}  
Enables to perform psuedodiagonalization using Jacobi rotations. [[13](#)]  
After reaching the SCC convergence with psuedodiagonalization, full diagonalization is performed to obtain final eigenvalues and eigenvectors.
- **PDIAGTHRESH** (*real*) {1.0e-02}  
Threshold to activate psuedodiagonalization.  
Full diagonalization is performed to update reference eigenvalues until charge density difference becomes less than the specified value.
- **PDIAGKAPPA** (*real*) {4.0e-02}  
Accuracy control parameter for psuedodiagonalization.
- **GAMMASPLINE** (*logical*) {FALSE}  
Enables to use cubic spline interpolation for exponentially decaying short-range part of  $\gamma$ -function.  
For MPI/OpenMP version, this is available in only RUNTYPE=ATOM.  
See reference [[14](#)] for theory, implementation, and numerical assessments of the present option.
- **GAMMASPLINEGRID** (*real*) {4.0e-02}  
Grid size of cubic spline interpolation for exponentially decaying short-range part of  $\gamma$ -function in atomic unit.
- **UNPAIREDELEC** (*real*) {0.0e+00}  
Number of unpaired electrons.

It is possible to set non-integer positive value.

Note that the specified spin multiplicity is discarded except for zero unpaired electrons.

- **SPINRELAX** (*logical*) {FALSE}  
Enables to relax total spin in spin-polarized calculation.
- **NELECTHRESH** (*real*) {1.0e-06}  
Threshold for conservation of total number of electrons in **SPINRELAX=TRUE**.
- **RESTRICT** (*logical*) {FALSE}  
Enables to use restricted approach in open-shell doublet calculation.  
Default is unrestricted approach.
- **UDERIVGAUSS** (*logical*) {FALSE}  
Enables to add charge dependent Gaussian function to Hubbard derivative in **THIRDDIAG=TRUE** [15].
- **UDERIVGAUSSD0** (*real*) {-9.0e-02}  
Parameter of Gaussian function (height) used in **UDERIVGAUSS=TRUE**.
- **UDERIVGAUSSG0** (*real*) {1.61e+01}  
Parameter of Gaussian function (width) used in **UDERIVGAUSS=TRUE**.
- **UDERIVGAUSSQ0** (*real*) {7.5e-01}  
Parameter of Gaussian function (position of center) used in **UDERIVGAUSS=TRUE**.
- **RUNTYPE** (*method*) {ATOM}  
Selects how to resolve DFTB calculations.  
Note that serial and OpenMP versions employ atom resolved calculation.
  - **SHELL0**: Based on valence shells. This is incompatible with **THIRDFULL=TRUE** and **THIRDDIAG=TRUE**.
  - **SHELL**: Based on valence shells. This is incompatible with **THIRDDIAG=TRUE**.
  - **ATOM**: Based on atoms. This is incompatible with **ORSCC=TRUE**.
- **ORSCC** (*logical*) {FALSE}  
Enables the use of orbitally resolved SCC calculation.  
Different Hubbard parameters for different atomic angular momentum shells are used instead of the use of *s*-shell only.

The DFTB2 [6] and DFTB3 [16] methods can be extended with this treatment.

- **GSDOMAIN** (*logical*) {FALSE}  
Enables the use of cell-linked list (CLL) algorithm for accelerating short-range part of charge-charge interaction calculation in GAMMASPLINE=TRUE.
- **GSDOMAINBLOCKXYZ/GSDOMAINBLOCK** (*integer*) {5}  
Block size in each of  $x$ -,  $y$ -, and  $z$ -directions used in GSDOMAIN=TRUE.
- **GSDOMAINBLOCKX** (*integer*) {5}  
Block size in  $x$ -direction used in GSDOMAIN=TRUE.
- **GSDOMAINBLOCKY** (*integer*) {5}  
Block size in  $y$ -direction used in GSDOMAIN=TRUE.
- **GSDOMAINBLOCKZ** (*integer*) {5}  
Block size in  $z$ -direction used in GSDOMAIN=TRUE.
- **GSDOMAINNEIGHBOR** (*integer*) {1}  
Number of linked neighbors referred to as adjacent cells in GSDOMAIN=TRUE.
- **MIXERSAVEMEMORY** (*logical*) {FALSE}  
Enables to reduce memory usage for charge mixing methods. This is implemented for atom resolved calculations (RUNTYPE=ATOM) and for closed-shell singlet calculations.

### 1.1.2 DC

The keyword DC sets whether a divide-and-conquer approach [17, 18] is employed or not. This keyword is turned on without any specification as well as SCC keyword. DC=FALSE is necessary if DC-DFTB calculation is deactivated.

- **BETA** (*real*) {8.0e+02}  
Inverse temperature parameter for the Fermi function in atomic unit.
- **BUFRAD/BUFFERRADIUS** (*real*) {5.0e+00}  
Buffer radius to construct buffer region in Angstrom (Å) unit.  
Atoms included in spheres centered at atoms in the central region are selected.
- **SUBTYPE** (*method*) {AUTO}  
Method to construct central region.
  - AUTO constructs subsystems automatically by dividing total system with

the specified cubic or rectangular grid.

Default is 5.0 Å cubic box.

- SPECIFY defines subsystems one by one for each atom (see [section 1.4.3](#)).
  - ATOM defines one atom as each subsystem.
  - SYSTEM defines all atoms as one subsystem.
  - SEMIAUTO defines subsystems as the hybrid of AUTO and SPECIFY (see [section 1.4.3](#)).
  - AUTOXH constructs subsystems similarly to AUTO without cutting covalent bonds involving hydrogen atom.
  - SEMIAUTOXH constructs subsystems similarly to SEMIAUTO without cutting covalent bonds involving hydrogen atom.
- DELTARXYZ/DELTAR (*real*) {5.0e+00}  
Grid length of cubic grid used in SUBTYPE=AUTO in Å unit.
  - DELTARX (*real*) {5.0e+00}  
Grid length of *x*-component used in SUBTYPE=AUTO in Å unit.
  - DELTARY (*real*) {5.0e+00}  
Grid length of *y*-component used in SUBTYPE=AUTO in Å unit.
  - DELTARZ (*real*) {5.0e+00}  
Grid length of *z*-component used in SUBTYPE=AUTO in Å unit.
  - NOFERMI (*logical*) {FALSE}  
Common Fermi level is not determined during DC calculation.
  - TRANSCOORD (*logical*) {TRUE}  
Transformation of Cartesian coordinate in SUBTYPE=AUTO.
    - TRUE: Standard orientation is used.
    - FALSE: Initial geometry is used as it is.
  - SUBXHSCALE (*real*) {1.1e+00}  
Scaling factor for the sum of covalent radii [19] to judge the presence of covalent bonds involving hydrogen atom in SUBTYPE=AUTOXH and SUBTYPE=SEMIAUTOXH.
  - FERMITYPE (*method*) {PARALLEL}  
Algorithm to determine Fermi level. See reference [20] for details.
    - PARALLEL: CRI (compute-reduce-interpolate) algorithm is used.

- **SERIAL**: GSS (gather-sort-solve) algorithm is used.
- **DOMAIN** (*logical*) {FALSE}
 

Enables the use of CLL algorithm for accelerating assignment of subsystems to localization regions.
- **DOMAINBLOCKXYZ/DOMAINBLOCK** (*integer*) {5}
 

Block size in each of *x*-, *y*-, and *z*-directions used in DOMAIN=TRUE.
- **DOMAINBLOCKX** (*integer*) {5}
 

Block size in *x*-direction used in DOMAIN=TRUE.
- **DOMAINBLOCKY** (*integer*) {5}
 

Block size in *y*-direction used in DOMAIN=TRUE.
- **DOMAINBLOCKZ** (*integer*) {5}
 

Block size in *z*-direction used in DOMAIN=TRUE.
- **DOMAININDEXTYPE** (*method*) {ATOM}
 

Definition of particle for the index of linked list in DOMAIN=TRUE.

  - **SUBSYSTEM**: The index of subsystem will be given if all constituent atoms are located in the same domain.
  - **ATOM**: The index will be assigned to all atoms.
- **DOMAINNEIGHBOR** (*integer*) {1}
 

Number of linked neighbors referred to as adjacent cells in DOMAIN=TRUE.
- **SINGLEPREC** (*logical*) {FALSE}
 

Enables the use of single precision floating point number for distance calculations in assignment of subsystems to localization regions.
- **XHDOMAIN** (*logical*) {FALSE}
 

Enables the use of CLL algorithm for judging the presence of covalent bonds involving hydrogen atom in SUBTYPE=AUTOXH.
- **XHDOMAINBLOCKXYZ/XHDOMAINBLOCK** (*integer*) {5}
 

Block size in each of *x*-, *y*-, and *z*-directions used in XHDOMAIN=TRUE.
- **XHDOMAINBLOCKX** (*integer*) {5}
 

Block size in *x*-direction used in XHDOMAIN =TRUE.
- **XHDOMAINBLOCKY** (*integer*) {5}

Block size in  $y$ -direction used in XHDOMAIN =TRUE.

- **XHDOMAINBLOCKZ** (*integer*) {5}  
Block size in  $z$ -direction used in XHDOMAIN =TRUE.
- **XHDOMAINNEIGHBOR** (*integer*) {1}  
Number of linked neighbors referred to as adjacent cells in XHDOMAIN =TRUE.

### 1.1.3 OPT/OPTIMIZE

The keyword OPT sets whether the geometry optimization is performed or not.

- **MAXITER** (*integer*) {50}  
Maximum number of optimization steps.
- **GRADCONV/GCONV** (*real*) {1.0e-04}  
Convergence criteria for the maximum absolute value of force component in atomic unit.
- **OPTTYPE** (*integer*) {1}  
Method for geometry optimization.
  - 1: BFGS  
This uses covalent radius of atoms to construct initial Hessian matrix, which are taken from reference [19].
  - 2: Steepest descent
  - 3: Conjugate gradient
- **STEPWISE** (*real*) {1.0e+02}  
Step size along the forces for steepest descent method in atomic unit.
- **CONSTRAINT** (*logical*) {FALSE}  
Enables to specify geometry constraints.  
Constraint vector of  $x$ ,  $y$ ,  $z$  components is necessary for atoms to be constrained (see [section 1.4.2](#)).
- **LATTICEOPT** (*logical*) {FALSE}  
Enables to optimize translation vectors.  
It is not allowed to impose partial geometry constraints and to set external point charges.

Note that conjugate gradient optimizer is used when OPTTYPE=1 (BFGS).

- EXTPRESSURE (*real*) {0.0e+00}  
External pressure during optimization of translation vectors in atomic unit.  
If value is not zero, enthalpy ( $E + pV$ ) of the system is additionally printed.
- ISOSCALING (*logical*) {FALSE}  
Enables to scale translation vectors uniformly during optimization of translation vectors.  
This is incompatible with FIXANGLE=TRUE.
- FIXANGLE (*logical*) {FALSE}  
Enables to fix angles between translation vectors during optimization of translation vectors.  
This is incompatible with ISOSCALING=TRUE.
- FIXANGLE1 (*logical*) {FALSE}  
Enables to fix length of first translation vector during optimization of translation vectors with FIXANGLE=TRUE.
- FIXANGLE2 (*logical*) {FALSE}  
Enables to fix length of second translation vector during optimization of translation vectors with FIXANGLE=TRUE.
- FIXANGLE3 (*logical*) {FALSE}  
Enables to fix length of third translation vector during optimization of translation vectors with FIXANGLE=TRUE.
- LATTICEONLY (*logical*) {FALSE}  
Enables to fix atomic coordinates during optimization of translation vectors.  
It is possible to apply the same option by putting a single asterisk (\*) after z-coordinate for all atoms (see [section 1.4.2](#)).
- STEPGRATOM (*real*) {1.0e-01}  
Maximum step size of atom relaxation for conjugate gradient method in atomic unit.
- STEPGRLATTICE (*real*) {1.0e-01}  
Maximum step size of lattice relaxation for conjugate gradient method in atomic unit.

- BFGSSOLVER (*integer*) {1}  
Eigensolver for diagonalization of Hessian matrix in BFGS method.
  - 1: DSYEVD
  - 2: DSYEV
- PRINTCOORD (*logical*) {TRUE}  
Enables to print molecular coordinate at each geometry step in the standard output file.  
Deactivation (PRINTCOORD=FALSE) corresponds to print only the final molecular coordinate in the standard output file.
- PRINTGRAD (*logical*) {TRUE}  
Enables to print DFTB energy gradient at each geometry step in the standard output file.  
In the case of PRINTGRAD=FALSE, maximum and root mean square forces are written in the standard output file.

#### 1.1.4 MD

The keyword MD sets whether the molecular dynamics (MD) using velocity Verlet integration scheme is performed or not.

- NSTEP/MAXSTEP (*integer*) {10000}  
Number of MD steps.
- DELTAT/DELTASTEP (*real*) {1.0e-15}  
Time interval between two MD steps in second (s) unit.
- BATHTEMP/TEMPERATURE (*real*) {2.9815e+02}  
Target temperature of thermostat in Kelvin (K) unit.
- NVE (*logical*) {TRUE}  
Enables to run MD with NVE ensemble.  
If deactivated, NVT ensemble will be used.
- NVT (*logical*) {FALSE}  
Enables to run MD with NVT ensemble.  
If deactivated, NVE ensemble will be used.
- NVTTYPE (*integer*) {3}  
Method to control temperature for achieving NVT or NPT ensemble.



- 1: Velocity scaling [21]
  - 2: Nosé-Hoover chain [22]
  - 3: Nosé-Hoover chain [22]
    - In comparison to NVTTYPE=2, NVTTYPE=3 allows to control the coupling strength.
  - 4: Berendsen [23]
  - 5: Andersen [24]
- RESTART (*logical*) {FALSE}
    - Enables to read information for restarting MD from the binary file “**restart**”.
  - SOFT/SOFTPOT (*logical*) {FALSE}
    - Enables to use soft potential.
  - SOFTRANGE (*real*) {1.0e+01}
    - Radius of soft potential in Å unit.
  - SOFTK (*real*) {1.5e+00}
    - Hardness of soft potential in kcal/(mol·Å<sup>2</sup>) unit.
  - INITTEMP (*real*) {2.9815e+02}
    - Target temperature of initial velocities in K unit.
    - Initial velocities are generated following the Maxwell-Boltzmann distribution.
  - SCALESTEP (*integer*) {20}
    - Interval to scale velocities for velocity scaling method (NVTTYPE=1).
  - ERRORTEMP (*real*) {1.0e+02}
    - Threshold of temperature for scaling velocities in K unit.
    - If the absolute value of difference between the target temperature of thermostat and the MD temperature is greater than ERRORTEMP, the velocities are scaled so that the temperature becomes BATHTEMP for NVT methods except for velocity scaling (NVTTYPE=2–5).
  - READVELOCITY (*logical*) {FALSE}
    - Enables to read velocities for all atoms from the ASCII file.
      - TRUE: Read the file “**veloc.dat**” in Å/fs unit (fs is femtosecond). The ASCII file should have 3 columns (*x*, *y*, *z* velocity components) for each atom of the given structure in Geometry section.
      - FALSE: No file to be read.

- NOSECLEAN (*logical*) {FALSE}
 

Enables to initialize internal parameters of Nosé-Hoover chain thermostat when MD is restarted.
- PRINT (*integer*) {1}
 

Interval to write coordinate, velocity, and Mulliken atomic charges to the files “**traject**”, “**velocity**”, and “**mulliken**”, respectively.
- NBATH (*integer*) {3}
 

Number of particles for Nosé-Hoover chain thermostat (NVTTYPE=3).
- COUPLINGSTR (*real*) {1.5e+03}
 

Frequency of oscillation of the thermostat particles in  $\text{cm}^{-1}$  unit for Nosé-Hoover chain thermostat (NVTTYPE=3).
- SEEDTYPE (*integer*) {1}
 

Method of random number generation for initial velocities.

  - 1: No specification (the same initial velocities may be reproduced on the same platform for the same given structure in Geometry section)
  - 2: Generation of random number from the current time.
  - 3: Specification of arbitrary random number using RANDOMSEED option.
- RANDOMSEED (*integer*) {0}
 

Seed to generate random number for initial velocities using SEEDTYPE=3.
- TAUTEMP (*real*) {1.0e-13}
 

Time constant for Berendsen thermostat (NVTTYPE=4) in s unit.
- COLLISIONFREQ (*real*) {1.0e-02}
 

Random collision frequency for Andersen thermostat (NVTTYPE=5) in  $\text{fs}^{-1}$  unit.
- MASSIVE (*logical*) {FALSE}
 

Method of scaling velocities for Andersen thermostat (NVTTYPE=5).

  - TRUE: All velocities are scaled simultaneously with the specified probability.
  - FALSE: Each atomic velocity is scaled individually with the specified probability.
- LIMULLIKEN (*logical*) {FALSE}
 

Enables to predict better initial Mulliken charges using Lagrange interpolation to accelerate SCC convergence.

- **LIDEGREE** (*integer*) {0}  
Degree of Lagrange polynomial in LIMULLIKEN=TRUE.  
The default setting 0 employs the previous converged charges for initial guess (conventional approach).
- **LIMINDEG** (*logical*) {FALSE}  
Enables to obtain the appropriate degree of Lagrange polynomial within a range from 0 to the value set in LIDEGREE in LIMULLIKEN=TRUE.  
Degree which gives the smallest absolute error of the predicted charges from the converged values at  $N$  step is used at  $N + 1$  step.
- **RMROTATION** (*logical*) {FALSE}  
Removal of center of mass rotation in addition to translation.  
This option is available in only cluster model simulations.
- **RMSTEP** (*integer*) {10000}  
Interval to remove center of mass translation (and rotation if RMROTATION=TRUE) for NVTTYPE=1–4.
- **RATTLE** (*method*) {NONE}  
Constraint MD simulation using RATTLE algorithm [25] for bond lengths.
  - NONE corresponds to no constraints of bond lengths.
  - COVALENTXH fixes X–H covalent bonds as the sum of their covalent radii, which are taken from reference [19].
  - INITGEOMXH fixes X–H covalent bond distance as the value of initial structure.
  - SPECIFY sets any bond constraints using the file “**rattle.dat**”.  
The first and second columns specify atom labels whose distance is fixed.  
The third column is an upper-case letter to determine how the specified bond distance is fixed.

1\_2\_C\_↓

“C” fixes to the sum of covalent radii

2\_3\_I\_↓

“I” fixes to the value of initial structure

1\_3\_S\_0.95\_↓

“S” fixes to the specified value (0.95 Å in this example)

*Note that the sum of covalent radii is used if no fourth column*

- RATTLEXH (*real*) {1.2e+00}  
Threshold of X–H pair as the covalent bond in RATTLE=COVALENTXH and RATTLE=INITGEOMXH in Å unit.
- RATTLEITER (*integer*) {100}  
Maximum number of cycles to satisfy RATTLE position and velocity constraints.
- RATTLECONV (*real*) {1.0e–06}  
Convergence criteria for the RATTLE constraints in atomic unit.
- ANNEAL (*logical*) {FALSE}  
Enables to perform simulated annealing simulation.  
Temperature scheduling is specified using the file “**anneal.dat**” up to 99 profiles.

C\_ 1\_ 300.0\_

*The first step is set to constant value of 300 K (C) INITTEMP is updated with the specified value*

L\_1000\_1000.0\_

*Next 1000 steps increase from 300 K to 1000 K linearly (L)*

C\_1000\_1000.0\_

*Next 1000 steps keep the temperature 1000 K (equilibration)*

E\_1000\_ 500.0\_3.0\_

*Next 1000 steps decrease from 1000 K to 500 K exponentially (E)*

*The fourth column controls the shape of exponential function*

*Note that the value of 5.0 is used if no fourth column*

C\_1000\_ 500.0\_

*Next 1000 steps keep the temperature 500 K*

S\_1000\_ 300.0\_2.0\_

Next 1000 steps decrease  
from 500 K to 300 K  
sigmoidally (S)

The fourth column controls  
the shape of sigmoid  
function

Note that the value of 3.0 is  
used if no fourth column

- SOFTCENTERTYPE (*method*) {ATOM}
  - ATOM defines one of atoms as center of soft potential.
  - SPECIFY manually sets the position of xyz coordinate.
  - COC is the abbreviation of “center of coordinate”.
  - COM is the abbreviation of “center of mass”.
- SOFTATOMNO (*integer*) {1}  
Atom label to be the center of soft potential used in SOFTCENTERTYPE=ATOM.
- SOFTPOINTXYZ/SOFTPOINT (*real*) {0.0e+00}  
Cartesian coordinate of the center of soft potential satisfying  $x = y = z$  used in SOFTCENTERTYPE=SPECIFY in Å unit.
- SOFTPOINTX (*real*) {0.0e+00}  
 $x$ -component of the center of soft potential used in SOFTCENTERTYPE=SPECIFY in Å unit.
- SOFTPOINTY (*real*) {0.0e+00}  
 $y$ -component of the center of soft potential used in SOFTCENTERTYPE=SPECIFY in Å unit.
- SOFTPOINTZ (*real*) {0.0e+00}  
 $z$ -component of the center of soft potential used in SOFTCENTERTYPE=SPECIFY in Å unit.
- SOFTSHAPETYPE (*method*) {SPHERE}  
Method to specify shape of soft potential.
  - SPHERE is sphere.
  - SQUARE is cubic or rectangular box.

- ELLIPSOID is ellipsoid.
- SOFTSIDEXYZ/SOFTSIDE (*real*) {1.0e+01}  
Grid length of cubic grid used in SOFTSHAPETYPE=SQUARE in Å unit.
- SOFTSIDEX (*real*) {1.0e+01}  
Grid length of  $x$ -component used in SOFTSHAPETYPE=SQUARE in Å unit.
- SOFTSIDEY (*real*) {1.0e+01}  
Grid length of  $y$ -component used in SOFTSHAPETYPE=SQUARE in Å unit.
- SOFTSIDEZ (*real*) {1.0e+01}  
Grid length of  $z$ -component used in SOFTSHAPETYPE=SQUARE in Å unit.
- SOFTSEMIAXISX (*real*) {1.0e+01}  
Length of  $x$ -semi-principal axis used in SOFTSHAPETYPE=ELLIPSOID in Å unit.
- SOFTSEMIAXISY (*real*) {1.0e+01}  
Length of  $y$ -semi-principal axis used in SOFTSHAPETYPE=ELLIPSOID in Å unit.
- SOFTSEMIAXISZ (*real*) {1.0e+01}  
Length of  $z$ -semi-principal axis used in SOFTSHAPETYPE=ELLIPSOID in Å unit.
- NPH (*logical*) {FALSE}  
Enables to run MD with NPH ensemble.  
If deactivated, NVE ensemble will be used.
- NPT (*logical*) {FALSE}  
Enables to run MD with NPT ensemble.  
If deactivated, NVE ensemble will be used.
- NPHTYPE (*integer*) {1}  
Method to control pressure for achieving NPH or NPT ensemble.
  - 1: Berendsen [\[23\]](#)
- NPPTYPE (*integer*) {1}  
Method to control pressure and temperature for achieving (isotropic) NPT ensemble.
  - 1: Hoover-Andersen [\[22\]](#)

- **EXTPRESSURE** (*real*) {1.01325e+05}  
Target pressure during NPH or NPT ensemble simulation in Pascal (Pa) unit.
- **TAUPRESS** (*real*) {2.0e-12}  
Time constant for Berendsen barostat (NPHTYPE=1) in s unit.
- **ISOSCALING** (*logical*) {FALSE}  
Enables to scale translation vectors uniformly during NPH or NPT ensemble simulation.  
Default is anisotropic change of unit cell.
- **CALCPRESSURE** (*logical*) {FALSE}  
Enables to calculate virial pressure in NVE or NVT ensemble.  
This option is supported for only periodic boundary condition.
- **COUPLINGSTRLAT** (*real*) {1.5e+01}  
Frequency of oscillation of the thermostat particles in  $\text{cm}^{-1}$  unit for Hoover-Andersen NPT ensemble (NPPTYPE=1).
- **CHECKPOINT** (*integer*) {100}  
Interval to write the second restart file “**restart\_chk**”.
- **RANDTYPE** (*integer*) {1}  
Method to generate random numbers.
  - 1: Fortran intrinsic random number generator  
Different platform may generate different random numbers using the same seed.
  - 2: Random number generator taken from reference [26].

### 1.1.5 NCC

The keyword NCC sets whether a non-SCC calculation (NCC-DFTB or DFTB1) [27, 28] is performed or not. NCC=TRUE is identical to set SCC=FALSE.

- **PRINTMULLIKEN** (*logical*) {FALSE}  
Enables to perform Mulliken population analysis.  
This option is required for further analysis of NCC Mulliken population.
- **MAYER** (*logical*) {FALSE}  
Enables to perform Mayer bond order analysis. [10]

Activation of PRINTMULLIKEN is required.

- **CM3** (*logical*) {FALSE}  
Enables to perform CM3 charge analysis. [11, 12]  
Activation of PRINTMULLIKEN is required.  
Note that the fitted parameters are taken from DFTB2.
- **ORBITALPOP/ORBPOP** (*logical*) {FALSE}  
Enables to print Mulliken population of each orbital.  
Activation of PRINTMULLIKEN is required.
- **SOLVER** (*integer*) {1}  
Eigensolver for diagonalization of Hamiltonian matrix.
  - 1: DSYGVD
  - 2: DSYGV
  - 3: DM\_VGEVPH  
This may be available if the platform is the K computer, Fujitsu FX10, and Fujitsu FX100.
- **RESTRICT** (*logical*) {FALSE}  
Enables to use restricted approach in open-shell doublet calculation.  
Default is unrestricted approach.

#### 1.1.6 FREQ/FREQUENCY

The keyword FREQ sets whether the harmonic vibrational frequency analysis is performed or not. If this keyword is turned on together with geometry optimization (OPT=TRUE), frequency calculation is carried out for the last optimization step of given structures. The frequency calculation is currently available for cluster models.

- **FREQTYPE** (*integer*) {1}  
Method of Hessian matrix construction.
  - 1: Analytical second derivatives.  
This is not implemented for including point charges (POINTCHARGE=TRUE in SCC keyword), for spline interpolation of  $\gamma$ -function (GAMMASPLINE=TRUE in SCC keyword), for open shell systems, for including chemical-potential equalization correction (CPE=TRUE in SCC keyword), and for any shell resolved calculations in MPI/OpenMP version (RUNTYPE=SHELL/SHELL0 in SCC keyword)



and ORSCC=TRUE in SCC keyword).

- 2: Numerical second derivatives using analytically calculated first derivatives.
- STEPSIZE (*real*) {1.0e-04}  
Step size for numerical differentiation of analytically calculated first derivatives in atomic unit.
- PRINTHESS (*logical*) {FALSE}  
Enables to print the lower triangle of Hessian matrix in the standard output file. Additionally, Hessian matrix information is stored in the binary file “**restart\_hess**” for restarting purpose.
- THERMOTEMP (*real*) {2.9815e+02}  
Target temperature for thermochemical analysis in K unit.
- THERMOPRES (*real*) {1.0e+00}  
Target pressure for thermochemical analysis in standard atmosphere (atm) unit.
- THERMOSCALE (*real*) {1.0e+00}  
Frequency scaling factor for thermochemical analysis.
- PRECFORMAT (*logical*) {FALSE}  
Enables to print high precision format for frequency eigenvectors.
- READHESS (*method*) {NONE}  
Enables to read Hessian matrix information from file.
  - NONE calculates the Hessian matrix either analytically or numerically.
  - BINARY reads Hessian matrix information from the binary file “**restart\_hess**”.
  - ASCII reads Hessian matrix information from the ASCII file “**hess.dat**”. Each line consists of three columns: the first and second integers are used to specify the index of Hessian matrix and the third value is an element of Hessian matrix.
- READISOTOPE (*logical*) {FALSE}  
Enables to specify arbitrary atomic mass.  
The specification form (ISO=*value*) is necessary for atoms to be modified from the default mass in Slater-Koster parameter file (see [section 1.4.2](#)).
- PROJECTION (*logical*) {TRUE}

Projection out translational and rotational modes from normal coordinate analysis.

Deactivation (`PROJECTION=FALSE`) corresponds to the diagonalization of full Hessian matrix.

- `CPCONV` (*real*) {1.0e-06}  
Convergence criteria for coupled-perturbed DFTB equations in atomic unit.
- `EFSTR` (*real*) {2.0e-03}  
Strength of external static electric field used in infrared (IR) and Raman intensities calculation.
- `INTERRUPTCP` (*logical*) {FALSE}  
Enables to interrupt solving coupled-perturbed DFTB equations.  
The partially constructed Hessian matrix is stored in the ASCII file “**hess.dat**” for restarting purpose.
- `CPSTART` (*integer*) {1}  
Atom index to specify the beginning of partial Hessian matrix to be constructed in `INTERRUPTCP=TRUE`.
- `CPEND` (*integer*) {1}  
Atom index to specify the end of partial Hessian matrix to be constructed in `INTERRUPTCP=TRUE`.
- `HESSSOLVER` (*integer*) {1}  
Eigensolver for diagonalization of Hessian matrix.
  - 1: DSYEVD
  - 2: DSYEV
- `SAVEMEMORYANA` (*logical*) {FALSE}  
Enables to reduce memory usage for computation of analytical Hessian in SCC calculation in compensation for the increased computational cost.

#### 1.1.7 DISP/DISPERSION

The keyword `DISP` sets whether the dispersion interaction is empirically corrected or not.

- `DISPTYPE` (*integer*) {1}  
Method of dispersion correction.
  - 1: Grimme’s DFT-D2 [\[29\]](#)

- 2: Slater-Kirkwood [30]  
The required dispersion constants have to be stored in different file for each element (see [section 1.3.2](#)).
  - 3: Lennard-Jones [31]  
The default values of Lennard-Jones parameters are taken from values of UFF force field [32].  
It is also possible to specify Lennard-Jones parameters manually in different file for each element (see [section 1.3.2](#)).
  - 4: Grimme's DFT-D3 [33]  
This is so-called zero-damping.
  - 5: Grimme's DFT-D3(BJ) [34]  
This is so-called Becke and Johnson damping.
  - 6: Řezáč's D3H4 [35]  
Since this corrects hydrogen bonding interaction empirically, the damped  $\gamma$ -function may not be combined together (DAMPXH=FALSE in SCC keyword). See, for example, reference [36].
  - 7: DFT-ulg [37]
  - 8: dDMC [38]
  - 9: Řezáč's D3H5 [39]  
Since H5 correction is designated to modify the original  $\gamma$ -function, DAMPXH=FALSE needs to be used in SCC keyword.  
The correction is not available for non-SCC calculations (NCC=TRUE or SCC=FALSE).
  - 10: Kubillus's D3X [40]
- UPDATESKPARAMS (*logical*) {FALSE}  
Update Slater-Kirkwood dispersion parameters in each DFTB calculation.  
Deactivation (UPDATESKPARAMS=FALSE) corresponds to keep dispersion parameters fixed with those of initial geometry during the entire calculation.
  - READUFF (*logical*) {FALSE}  
Enables to specify Lennard-Jones parameters manually for Lennard-Jones dispersion (see [section 1.3.2](#)).
  - S6 (*real*) {1.0e+00}  
Global scaling factor for Grimme's DFT-D2.
  - ZEROS8 (*real*) {6.73e-01}

Global scaling factor for  $r^{-8}$  term of Grimme's DFT-D3.

- ZERORS6 (*real*) {1.235e+00}  
Parameter for damping function of Grimme's DFT-D3.
- BJS8 (*real*) {5.883e-01}  
Global scaling factor for  $r^{-8}$  term of Grimme's DFT-D3(BJ).
- BJA1 (*real*) {5.719e-01}  
Parameter for Becke and Johnson damping of Grimme's DFT-D3(BJ).
- BJA2 (*real*) {3.6017e+00}  
Parameter for Becke and Johnson damping of Grimme's DFT-D3(BJ).
- THREEBODY (*logical*) {FALSE}  
Inclusion of three-body term for Grimme's DFT-D3 and DFT-D3(BJ).
- D3H4S6 (*real*) {1.0e+00}  
Parameter for Řezáč's D3H4 correction (D3 dispersion).
- D3H4RS6 (*real*) {1.215e+00}  
Parameter for Řezáč's D3H4 correction (D3 dispersion).
- D3H4ALPHA (*real*) {3.0e+01}  
Parameter for Řezáč's D3H4 correction (D3 dispersion).
- D3H4SHH (*real*) {3.0e-01}  
Parameter for Řezáč's D3H4 correction (H–H repulsion).
- D3H4EHH (*real*) {1.431e+01}  
Parameter for Řezáč's D3H4 correction (H–H repulsion).
- D3H4R0HH (*real*) {2.35e+00}  
Parameter for Řezáč's D3H4 correction (H–H repulsion).
- D3H4CNN (*real*) {2.01e+00}  
Parameter for Řezáč's D3H4 correction (H4 correction).
- D3H4CNO (*real*) {8.0e-01}  
Parameter for Řezáč's D3H4 correction (H4 correction).
- D3H4CON (*real*) {2.58e+00}  
Parameter for Řezáč's D3H4 correction (H4 correction).

- D3H4COO (*real*) {1.11e+00}  
Parameter for Řezáč's D3H4 correction (H4 correction).
- D3H4CWAT (*real*) {1.32e+00}  
Parameter for Řezáč's D3H4 correction (H4 correction).
- D3H4CSNHR3 (*real*) {2.33e+00}  
Parameter for Řezáč's D3H4 correction (H4 correction).
- D3H4CSCOO (*real*) {1.22e+00}  
Parameter for Řezáč's D3H4 correction (H4 correction).
- ULGS (*real*) {7.012e−01}  
Parameter for DFT-ulg.  
Note that default value is adjusted to PBE functional.
- ULGB (*real*) {6.966e−01}  
Parameter for DFT-ulg.  
Note that default value is adjusted to PBE functional.
- DDMCA (*real*) {1.018e+00}  
Parameter for dDMC.
- DDMCB0 (*real*) {1.857e+00}  
Parameter for dDMC.
- DDMCS (*real*) {4.6e+01}  
Parameter for dDMC.
- D3H5S6 (*real*) {1.0e+00}  
Parameter for Řezáč's D3H5 correction (D3 dispersion).
- D3H5S8 (*real*) {4.9e−01}  
Parameter for Řezáč's D3H5 correction (D3 dispersion).
- D3H5ALPHA (*real*) {2.961e+01}  
Parameter for Řezáč's D3H5 correction (D3 dispersion).
- D3H5SHH (*real*) {3.0e−01}  
Parameter for Řezáč's D3H5 correction (H–H repulsion).
- D3H5EHH (*real*) {1.431e+01}  
Parameter for Řezáč's D3H5 correction (H–H repulsion).

- D3H5R0HH (*real*) {2.35e+01}  
Parameter for Řezáč's D3H5 correction (H–H repulsion).
- D3H5SR (*real*) {7.14e−01}  
Parameter for Řezáč's D3H5 correction (H5 correction).
- D3H5SW (*real*) {2.5e−01}  
Parameter for Řezáč's D3H5 correction (H5 correction).
- D3H5KNH (*real*) {1.8e−01}  
Parameter for Řezáč's D3H5 correction (H5 correction).
- D3H5KOH (*real*) {6.0e−02}  
Parameter for Řezáč's D3H5 correction (H5 correction).
- D3H5KSH (*real*) {2.1e−01}  
Parameter for Řezáč's D3H5 correction (H5 correction).
- D3XS8 (*real*) {3.209e+00}  
Parameter for Kubillus's D3X correction (D3 dispersion).
- D3XA1 (*real*) {7.46e−01}  
Parameter for Kubillus's D3X correction (D3 dispersion).
- D3XA2 (*real*) {4.191e+00}  
Parameter for Kubillus's D3X correction (D3 dispersion).
- D3XC1 (*real*) {7.761e+00}  
Parameter for Kubillus's D3X correction (X correction).
- D3XC2 (*real*) {5.0e−02}  
Parameter for Kubillus's D3X correction (X correction).
- D3XC3 (*real*) {4.518e+00}  
Parameter for Kubillus's D3X correction (X correction).
- D3XDNCL (*real*) {1.526e+00}  
Parameter for Kubillus's D3X correction (X correction).
- D3XDNBR (*real*) {1.349e+00}  
Parameter for Kubillus's D3X correction (X correction).
- D3XDNI (*real*) {1.521e+00}

Parameter for Kubillus's D3X correction (X correction).

- **D3XDOCL** (*real*) {1.237e+00}  
Parameter for Kubillus's D3X correction (X correction).
- **D3XDOBR** (*real*) {1.099e+00}  
Parameter for Kubillus's D3X correction (X correction).
- **D3XDOI** (*real*) {1.313e+00}  
Parameter for Kubillus's D3X correction (X correction).
- **H4IDOMAIN** (*logical*) {FALSE}  
Enables the use of CLL algorithm for index of hydrogen atoms for accelerating Řezáč's D3H4 calculation (H4 correction) in DISPTYPE=6.
- **H4IDOMAINBLOCKXYZ/H4IDOMAINBLOCK** (*integer*) {5}  
Block size in each of *x*-, *y*-, and *z*-directions used in H4IDOMAIN=TRUE.
- **H4IDOMAINBLOCKX** (*integer*) {5}  
Block size in *x*-direction used in H4IDOMAIN =TRUE.
- **H4IDOMAINBLOCKY** (*integer*) {5}  
Block size in *y*-direction used in H4IDOMAIN =TRUE.
- **H4IDOMAINBLOCKZ** (*integer*) {5}  
Block size in *z*-direction used in H4IDOMAIN =TRUE.
- **H4IDOMAINNEIGHBOR** (*integer*) {1}  
Number of linked neighbors referred to as adjacent cells in H4IDOMAIN =TRUE.
- **H4VDOMAIN** (*logical*) {FALSE}  
Enables the use of CLL algorithm for evaluation of atom valence for accelerating Řezáč's D3H4 calculation (H4 correction) in DISPTYPE=6.
- **H4VDOMAINBLOCKXYZ/H4VDOMAINBLOCK** (*integer*) {5}  
Block size in each of *x*-, *y*-, and *z*-directions used in H4VDOMAIN=TRUE.
- **H4VDOMAINBLOCKX** (*integer*) {5}  
Block size in *x*-direction used in H4VDOMAIN =TRUE.
- **H4VDOMAINBLOCKY** (*integer*) {5}  
Block size in *y*-direction used in H4VDOMAIN =TRUE.

- **H4VDOMAINBLOCKZ** (*integer*) {5}  
Block size in *z*-direction used in H4VDOMAIN =TRUE.
- **H4VDOMAINNEIGHBOR** (*integer*) {1}  
Number of linked neighbors referred to as adjacent cells in H4VDOMAIN =TRUE.

### 1.1.8 PBC

The keyword PBC sets some parameters to control the calculation in periodic boundary condition. Note that the system is treated as periodic if three translation vectors are detected in Geometry section even if PBC=TRUE is not explicitly written (see [section 1.4.1](#)).

- **STRESS** (*logical*) {FALSE}  
Enables to calculate stress tensors and lattice derivatives.
- **COULOMBTYPE** (*integer*) {1}  
Method of Coulomb interaction calculation.
  - 1: Ewald summation
  - 2: Multipole expansion [\[41\]](#)  
See reference [\[14\]](#) for theory, implementation, and numerical assessments of the present option.  
There are several limitations at present.
    - ✓ The stress tensor calculation is not supported.
    - ✓ Point charges should not be present (i.e. POINTCHARGE=FALSE in SCC keyword).
    - ✓ Atom resolved energies cannot be evaluated (i.e. PRINTATOME=FALSE in MISC keyword).
    - ✓ Shell resolved calculation in MPI/OpenMP version (RUNTYPE=SHELL, RUNTYPE=SHELL0, or ORSCC=TRUE in SCC keyword) is not available.
    - ✓ Only cubic unit cell has been tested.
- **EWALDALPHA** (*real*) {0.0e+00}  
Fraction of Ewald summation between reciprocal and real space.  
If value is larger than zero and smaller than one, manually specified value is used (in only single point calculation).



Otherwise, value is automatically determined (default and recommended).

- **MAXGAMMA** (*integer*) {7}  
Maximum number of unit cell repetition for Ewald summation.
- **FMMP** (*integer*) {8}  
Expansion order for multipole expansion in COULOMBTYPE=2.
- **FMMN** (*integer*) {3}  
Number of aggregated units at each level of telescoping procedure in COULOMBTYPE=2.
- **FMMK** (*integer*) {5}  
Maximum number of level for multipole expansion in COULOMBTYPE=2.
- **EWALDSAVEMEMORY** (*logical*) {FALSE}  
Enables to reduce memory usage for computation of reciprocal space summation in Ewald method in compensation for the increased computational cost.
- **EWALDMEMORYGB** (*real*) {8.0e+00}  
Maximum memory size for temporary arrays used in reciprocal space summation in Ewald method in gigabyte (GB) unit.  
If the estimated memory size for temporary arrays is larger than the specified value, the reduced memory usage approach is used in computations irrespective of EWALDSAVEMEMORY=FALSE.
- **FMMSAVEMEMORY** (*logical*) {FALSE}  
Enables to reduce memory usage for computation of solid harmonics in multipole-based algorithm (COULOMBTYPE=2) in compensation for the increased computational cost.

### 1.1.9 MISC

The keyword MISC sets other miscellaneous options.

- **PRINTFORCE/FORCE** (*logical*) {FALSE}  
Enables to perform the force calculation after single point energy calculation.  
If system includes external point charges, forces of point charges are additionally printed.  
If STRESS=TRUE in PBC keyword, stress tensor, lattice derivatives, volume and pressure are additionally printed.

- **PRINTRESOLVEDFORCE** (*logical*) {FALSE}
 

Enables to evaluate force contributions (electronic, repulsive, and dispersion term if included).

If **STRESS=TRUE** in PBC keyword, corresponding contributions of stress tensor are additionally evaluated.

Note that total forces are printed in the standard output while the respective contributions are printed in the detailed data file.
- **PRINTMO** (*logical*) {FALSE}
 

Enables to print eigenvectors.

Note that this option is available when the number of subsystem is equal to 1.
- **PRINTATOME** (*logical*) {FALSE}
 

Enables to evaluate atomic energy contributions (electronic, repulsive, and dispersion term if included).
- **PRINTHS** (*logical*) {FALSE}
 

Enables to print the lower triangle of non-perturbed Hamiltonian and overlap matrices in the standard output.

This option is available when the number of subsystem is equal to 1.

Note that the program terminates immediately without proceeding further DFTB computational steps.
- **PRINTDENSITY** (*logical*) {FALSE}
 

Enables to print the lower triangle of density matrix in the standard output.
- **PRINTLOCALREGION** (*logical*) {FALSE}
 

Enables to print number of subsystems, atoms, and basis functions in localization regions in the standard output.

Note that the program terminates immediately without proceeding further DFTB computational steps.
- **PRINTLAPTIME** (*logical*) {FALSE}
 

Enables to print wall clock time for DFTB computational steps.

Total wall clock time is additionally printed at the end of program run.
- **WRITEDAT** (*method*) {APPEND}
 

Enables to control dumping of the detailed data file.

  - NONE never generates the data file.
  - APPEND opens the data file in the beginning of program run and writes the

information of each DFTB calculation. The size of detailed data file may become huge if a large number of geometry steps is specified in geometry optimization or MD.

- REPLACE opens/closes the data file at the beginning/end of DFTB calculation. Namely, in geometry optimization or MD, the data file contains the information of the last geometry step.
  
- WRITECHRGFILE (*logical*) {TRUE}  
Enables to control dumping of the binary file “**chrgfile**” containing Mulliken charge information which is used in READCHARGE=BINARY in SCC keyword.
  
- USESCRATCH (*logical*) {TRUE}  
Enables to use disk I/O for storing some intermediate data.  
If USESCRATCH=FALSE, all intermediate data are stored in memory.

## 1.2 Title section

TITLE┆

*Comment line*

This is typically used for the comment of compound name, symmetry, electronic state, other relevant information, and so on. Each line should be less than 128 characters. All comments will be written in the standard output file as it is.

## 1.3 Parameter section

### 1.3.1 General overview

2	<i>Number of unique elements</i>
O_2	<i>Atom symbol and its highest angular momentum for valence shells</i>
oo.spl_oh.spl	<i>Slater-Koster parameters: atom1–atom1 and atom1–atom2</i>
H_1	<i>Atom symbol and its highest angular momentum for valence shells</i>
ho.spl_hh.spl	<i>Slater-Koster parameters: atom2–atom1 and atom2–atom2</i>

This section starts from the line giving the number of elements in the given structure. The present example is 2 since water molecule contains oxygen and hydrogen. Next, atomic symbol and its maximum angular momentum are specified for the first element (*atom1*). The integer 1, 2, 3 corresponds to the maximum angular momentum *s*, *p*, *d*, respectively. Then, the paths to Slater-Koster parameter files including the first element are specified in a single line. The order has to be consistent in the order of elements specified in this section, namely *atom1* with *atom1*, *atom1* with *atom2* (the second element), *atom1* with *atom3* (the third element), ... These two lines are repeated until all elements in the given structure are specified. Note that the present example assumes 4 Slater-Koster parameter files (**oo.spl**, **oh.spl**, **ho.spl**, **hh.spl**) are located in the same directory as the one containing the input file.

### 1.3.2 Specification of additional constants

Several options request atom dependent constants which are not hard-coded or written in Slater-Koster files. Such constants are specified in two ways:

- Direct specification of employed values

This is necessary for Hubbard derivatives in third order corrections (THIRDFULL=TRUE and THIRDDIAG=TRUE in SCC keyword).

The Hubbard derivative should be specified after the maximum angular momentum.

In the case of orbitally resolved calculation (ORSCC=TRUE in SCC keyword) with Multi version, values of *s*-, *p*-, and *d*-shells need to be provided separately.

O\_2\_-0.1575\_↓

*Atom resolved case (usual)*

Cu\_3\_-0.20\_-0.0575\_-0.0575\_↓

*Orbitally resolved case  
(order is d, p, s)*

- Preparation of additional ASCII files

This is necessary for the following three options:

- Spin constants for spin polarized calculation (SPIN=TRUE in SCC keyword)
- Atomic polarizabilities, van der Waals radii, and effective charge for Slater-Kirkwood dispersion (DISPTYPE=2 in DISP keyword)
- Lennard-Jones parameters for Lennard-Jones dispersion (DISPTYPE=3 and READUFF=TRUE in DISP keyword)

The path of additional file should be specified after the maximum angular momentum (or Hubbard derivatives if they exist). In the case of specifying both files, the path of dispersion constants file should come first, followed by the path of spin constants file:

O\_2\_o.wll\_↓

*Only spin constants file*

O\_2\_o.dispsk\_↓

*Only dispersion constants  
file*

O\_2\_o.dispsk\_o.wll\_↓

*Both spin and dispersion  
constants files*

Here is an example of the spin constants file for carbon:

-0.031\_↓

-0.025\_↓

-0.025\_↓

-0.023\_↓

The number of lines of spin constants file depends on the maximum angular momentum (1, 4, 9 for *s*, *p*, *d*). Each line contains the atomic spin constant in atomic unit in the following order:

- *s*: *ss*
- *p*: *ss, sp, ps, pp*

➤ *d: ss, sp, sd, ps, pp, pd, ds, dp, dd*

For reference, PBE values for H, C, N, O, S, Fe, and Ni listed in DFTB+ manual [42] are stored into the file with suffix “.wll” in the slko directory.

Here is an example of the Slater-Kirkwood dispersion constants file for carbon:

```
1.382_1.382_1.382_1.064_3.8_3.8_3.8_3.8_2.5_
```

There should exist nine columns in a single line. The first to fourth columns specify atomic polarizabilities in  $\text{\AA}^3$  unit, the fifth to eighth columns specify van der Waals radii in  $\text{\AA}$  unit, and the last column specifies effective charge. The values for atomic polarizabilities and van der Waals radii are given for zero, one, two, and three hydrogen neighbors. For reference, H, C, N, O, and S dispersion constants listed in DFTB+ manual [42] are stored into the file with suffix “.dispsk” in the slko directory.

To manually specify the Lennard-Jones parameters, values of non-bonded distance (in  $\text{\AA}$  unit) and energy (in kcal/mol unit) are given in a single line.

```
3.851_0.105_
```

*The order of specification  
is distance then energy*

For reference, H, C, N, O, S, Fe, and Ni Lennard-Jones parameters listed in reference [32] are stored into the file with suffix “.displj” in the slko directory.

## 1.4 Geometry section

### 1.4.1 General overview

```
3_0_1_↓
O _ 0.06275391_ 0.06275391_ 0.00000000_↓
H _ 1.01910555_-0.08185946_ 0.00000000_↓
H _ -0.08185946_ 1.01910555_ 0.00000000_↓
```

*Number of atoms, charge,  
spin multiplicity*

*Cartesian coordinates of  
atom label 1*

*Cartesian coordinates of  
atom label 2*

*Cartesian coordinates of  
atom label 3*

This section begins with the line giving three integers; number of atoms, charge, and spin multiplicity for the given structure. Then the position of atom is specified with Cartesian coordinates in Å unit with floating point number (**DO NOT USE INTEGERS!**).

If the given system is periodic, the unit cell can be specified by adding three translation vectors with symbol TV after the last Cartesian coordinates (TV should be an upper-case). The following example is for diamond crystal structure (primitive unit cell).

```
2_0_1_↓
C _ 0.00000000_ 0.00000000_ 0.00000000_↓
C _ 0.89169750_ 0.89169750_ 0.89169750_↓
TV_ 0.00000000_ 1.78339500_ 1.78339500_↓
TV_ 1.78339500_ 0.00000000_ 1.78339500_↓
TV_ 1.78339500_ 1.78339500_ 0.00000000_↓
```

If the external static electric field is included, the electric field vector can be specified by adding one line with symbol EF after the last Cartesian coordinates (EF should be an upper-case). The example of water molecule with electric field components of  $x = 0.001$ ,  $y = -0.001$ , and  $z = 0.000$  is given below.

```
3_0_1_↓
O _ 0.06275391_ 0.06275391_ 0.00000000_↓
H _ 1.01910555_-0.08185946_ 0.00000000_↓
```



```
H  -0.08185946  1.01910555  0.00000000
EF  0.00100000 -0.00100000  0.00000000
```

If the point charges are included, the coordinates and charge value can be specified by adding lines with symbol PC after the last Cartesian coordinates (PC should be an uppercase). The example of water molecule with point charge of 1.0 positioned at  $x = 5.0 \text{ \AA}$ ,  $y = 5.0 \text{ \AA}$ , and  $z = 5.0 \text{ \AA}$  is given below.

```
3_0_1
O  0.06275391  0.06275391  0.00000000
H  1.01910555 -0.08185946  0.00000000
H  -0.08185946  1.01910555  0.00000000
PC  5.00000000  5.00000000  5.00000000  1.00000000
```

#### 1.4.2 Specification of additional information

Additional columns after Cartesian coordinates can be read when you would like to specify atoms to be moved or atomic masses to be changed to investigate the isotope effects of vibrational properties.

- Freeze the coordinate in geometry optimization or MD  
If a single asterisk (\*) is put, the position of atom will not be moved.
- Constraint geometry optimization (CONSTRAINT=TRUE in OPT keyword)  
Constraint vector of  $x$ ,  $y$ ,  $z$  components is either 0.0 (fixed) or 1.0 (moved). For example, the vector “0.0\_0.0\_1.0” will update only  $z$  component. The vector “0.0\_0.0\_0.0” is identical to put a single asterisk.
- Specification of different atomic masses in harmonic vibrational frequency analysis (READISOTOPE=TRUE in FREQ keyword)  
The form ISO=*value* changes the default atomic mass to *value*. No space between ISO= and *value*. This specification should come later than the single asterisk or constraint vector when both options are activated.

Here is an example of the mixed specifications for water molecule.

```
3_0_1
O  0.06275391  0.06275391  0.00000000
```

```
H 1.01910555 -0.08185946 0.00000000 0.0 1.0 0.0
H -0.08185946 1.01910555 0.00000000 *ISO=2.014102
```

When the constraint geometry optimization is performed, the hydrogen with atom label 2 can be moved along only the y-axis and the coordinate of hydrogen with atom label 3 is never updated. If `FREQ=(READISOTOPE=TRUE)` is additionally specified, the atomic mass of hydrogen with atom label 3 is changed to 2.014102, namely the harmonic vibrational frequencies and corresponding thermochemical properties of HOD molecule will be obtained.

#### 1.4.3 Manual construction of subsystems in DC-DFTB

This section supplements how to specify the subsystems using `SUBTYPE=SPECIFY` and `SUBTYPE=SEMIAUTO` options in DC keyword. After the blank line of Geometry section, the indices of subsystems for each atom are listed.

Here is an example of specification of 5 subsystems for the given structure containing 22 atoms:

```
1_1_1_1_1_1      Index of subsystem for atom label 1, 2, 3, 4, 5
2_2_2_2_2      Index of subsystem for atom label 6, 7, 8, 9
3_3_3_3_3      Index of subsystem for atom label 10, 11, 12, 13
4_4_4_4_4      Index of subsystem for atom label 14, 15, 16, 17
5_5_5_5_5      Index of subsystem for atom label 18, 19, 20, 21, 22
```

where atoms with label 1–5 belong to subsystem 1, atoms with label 6–9 belong to subsystem 2, atoms with label 10–13 belong to subsystem 3, and so on. It is not necessarily to use the same number of lines as the number of subsystems. The new line will be read until the total number of indices is equal to the number of atoms in the given structure.

In the case of `SUBTYPE=SEMIAUTO`, the integer “0” is used as an index for atoms to be assigned to subsystems automatically by using the grid box. The following is an example of system containing 22 atoms (using analogous format from the previous one for comparison):

```
1_1_1_1_1_1      Index of subsystem for atom label 1, 2, 3, 4, 5
```

0_0_0_0_↓	<i>Index of subsystem for atom label 6, 7, 8, 9</i>
0_0_0_0_↓	<i>Index of subsystem for atom label 10, 11, 12, 13</i>
0_0_0_0_↓	<i>Index of subsystem for atom label 14, 15, 16, 17</i>
2_2_2_2_2_↓	<i>Index of subsystem for atom label 18, 19, 20, 21, 22</i>

where atoms with label 1–5 belong to subsystem 1, atoms with label 18–22 belong to subsystem 2, and atoms with label 6–17 are assigned to subsystems 3–X automatically (X depends on the given structure and size of grid box). If all indices are zero and positive integer, the option is identical to SUBTYPE=AUTO and SUBTYPE=SPECIFY, respectively.

## 2 Running DCDFTBMD

Once the input file “**dftb.inp**” is created, other requisites such as Slater-Koster parameters and restart files are needed to be placed in the appropriate directory. For example, in the case of SCC-DFTB single point energy calculation of water molecule shown in [section 1](#), 4 Slater-Koster parameter files (oo.spl, oh.spl, ho.spl, and hh.spl) must be placed in the same directory as the input file. The executable is used without any arguments.

Serial version	<installation path>/dftb_serial.00.x
OpenMP version	<installation path>/dftb_openmp.00.x
MPI/OpenMP version	mpirun -np (number of processes) <installation path>/dftb_mpiomp.00.x

The header of input file may be specified arbitrarily by setting it as the first argument. The example of using “**file.inp**” is as follows.

Serial version	<installation path>/dftb_serial.00.x file
OpenMP version	<installation path>/dftb_openmp.00.x file
MPI/OpenMP version	mpirun -np (number of processes) <installation path>/dftb_mpiomp.00.x file

For OpenMP parallelization, number of threads must be set before starting the calculation.

bash	export OMP_NUM_THREADS=(number of threads)
csh, tcsh	setenv OMP_NUM_THREADS (number of threads)

If a large-scale calculation stops, one may need to increase the stack size.

bash	ulimit -s unlimited; export OMP_STACKSIZE=(stack size value)
csh, tcsh	unlimit; setenv OMP_STACKSIZE (stack size value)

Note that the maximum number of basis sets in serial and OpenMP versions is limited to 16384.

### 3 Output of DCDFTBMD

If the calculation terminates normally, the standard output file and the detailed data file will be appeared. By default (using “**dftb.inp**” as the input file), the corresponding filenames are “**dftb.out**” and “**dftb.dat**”, respectively. When the header of input file is specified arbitrarily (e.g. “**file.inp**” as described in [section 2](#)), the header of output and data files will be changed appropriately (e.g. “**file.out**” and “**file.dat**”). Here, the output files of serial and OpenMP versions for SCC-DFTB single point energy calculation of water molecule shown in [section 1](#) are briefly outlined. Those of MPI/OpenMP version are similar to serial and OpenMP versions although the format of detailed data file is slightly different for the information of subsystem and Mulliken population.

#### 3.1 Standard output file

The standard output file begins with the summary of calculation type.

```
*****
DDDDDD   CCCCCC   DDDDDD   FFFFFFFF TTTTTTTTTT BBBBBBBB   MM       MM DDDDD
DDDDDDDD CCCCCCCC DDDDDDDD FFFFFFFF TTTTTTTTTT BBB   BBB MMM   MMM DDDDDDD
DDD  DDD CCC      DDD  DDD FF          TTT  BBB   BBB MM  M M  MM DDD  DDD
DDD  DDD CCC      DDD  DDD FFFFFFFF   TTT  BBBBBBBB MM  M  MM DDD  DDD
DDD  DDD CCC      DDD  DDD FFFFFFFF   TTT  BBB   BBB MM          MM DDD  DDD
DDDDDDDD CCCCCCCC DDDDDDDD FF          TTT  BBB   BB MM          MM DDDDDDD
DDDDDD   CCCCCC   DDDDDD   FF          TTT  BBBBBBBB MM       MM DDDDD
VERSION 1.0 (Nov 2018)
*****
Execution of DCDFTBMD begun %a %b %d %H:%M:%S %Y
-----
TITLE
-----
2 kinds of elements will be used
    Pair of element 1 and element 1 : {path}/oo.spl
    Pair of element 1 and element 2 : {path}/oh.spl
    Pair of element 2 and element 1 : {path}/ho.spl
    Pair of element 2 and element 2 : {path}/hh.spl
SCC      = True
```

```

-----
Max cycle           = 200
Energy convergence = 1.0E-09
Density convergence = 1.0E-06
Charge mixing method = Broyden
  Broyden iteration = 70
Mixing parameter   = 2.0E-01
Eigensolver        = DSYGVD
Damping gamma X-H = False
Orbital resolved SCC = False
Third order diagonal = False
Third order full    = False
Spin contribution   = False
Mayer bond order    = False
Polarizability      = False
External field      = False
Point charge        = False
Pseudodiagonalization = False
Cubic splined gamma = False
Direct SCC          = False
Read charge         = NONE
DC                 = False
-----
Optimize           = False
-----
MD                 = False
-----
Frequency          = False
-----
Dispersion         = False
-----
PBC                = False
-----
MISC               = False
-----
Total number of basis set shells = 4

```

Number of basis functions = 6  
Number of electrons = 8  
Charge of system = 0  
Spin multiplicity = 1  
Number of occupied orbitals = 4  
Total number of atoms = 3

\*\*\* Start single point energy calculation \*\*\*

Then, the SCC-DFTB single point calculation starts in the present example.

```
*****  
*** Start SCC-DFTB calculation ***  
*****  
Iter. Total energy Energy diff. Density diff. Fermi level  
-----  
1 0.0726464067 0.0726464067 2.0000000000 0.0025000000  
2 -4.0712418245 -4.1438882313 0.0200187809 0.0025000000  
3 -4.0741988428 -0.0029570183 0.0590590622 0.3564583333  
4 -4.0779379555 -0.0037391127 0.0004103232 0.3525000000  
5 -4.0779381355 -0.0000001800 0.0000024570 0.3525000000  
6 -4.0779381355 0.0000000000 0.0000000001 0.3562500000  
Final SCC-DFTB Energy = -4.0779381355 Eh after 6 iterations  
Execution of DCDFTBMD terminated normally %a %b %d %H:%M:%S %Y
```

Here, the SCC converged total energy is  $-4.0779381355$  Hartree.

## 3.2 Detailed data file

The detailed data file contains the relevant properties of the given structure. The constructed subsystems and localization regions are initially printed. Since this calculation is `DC=FALSE`, all 3 atoms belong to subsystem 1.

```
SUBSYSTEM : 1
-----
  1    2    3
LOCALIZATION REGION : 1
-----
  1
```

If you use MPI/OpenMP version, this part looks concise excluding the case of `PRINTLOCALREGION=TRUE` in MISC keyword.

```
SUBSYSTEM
-----
  1    1    1
```

Then, the eigenvalues of the molecular orbitals (first column) and the electron occupation number of that level (second column) are shown.

```
EIGENVALUES [Eh] AND OCCUPANCIES
-----
-0.8489865909    2.0000000000
-0.4143390336    2.0000000000
-0.3137537414    2.0000000000
-0.2591755518    2.0000000000
 0.3992760730    0.0000000000
 0.5583952222    0.0000000000
```

If `PRINTMO=TRUE` in MISC keyword, the eigenvectors are additionally printed. The occupied and virtual orbitals are distinguished with `OCC.` and `VIRT.`, respectively.

```
EIGENVALUES [Eh], OCCUPANCIES, AND MO COEFFICIENTS
```



```

-----
1   OCC.   -0.8489865909   2.0000000000
   O   S   0   -0.8522740053
       P  -1   0.0017546133
       P   0   0.0000000000
       P   1   0.0017546133
2   H   S   0   -0.1530727458
3   H   S   0   -0.1530727458
2   OCC.   -0.4143390336   2.0000000000
   O   S   0   -0.0000000000
       P  -1   0.4626963766
       P   0   0.0000000000
       P   1  -0.4626963766
2   H   S   0   -0.3908041098
3   H   S   0   0.3908041098
...

```

Then, the total Mulliken charge on the atom (second column) is shown together with the corresponding net charges (third column).

```

MULLIKEN POPULATIONS AND NET ATOMIC CHARGES
-----
1   6.5926170634   -0.5926170634
2   0.7036914683   0.2963085317
3   0.7036914683   0.2963085317

```

If **ORBITALPOP=TRUE** in **SCC** or **NCC** keyword, the Mulliken charges on each atomic orbital are additionally printed.

```

MULLIKEN POPULATIONS AND NET ATOMIC CHARGES
-----
1   O   6.5926170634   -0.5926170634
       S   0   1.7342141199
       P  -1   1.4292014718
       P   0   2.0000000000
       P   1   1.4292014718

```

```

2   H   0.7036914683   0.2963085317
    S  0  0.7036914683
3   H   0.7036914683   0.2963085317
    S  0  0.7036914683

```

If you use MPI/OpenMP version, the shell resolved Mulliken population is printed.

MULLIKEN POPULATIONS

```

-----
1.73421412   4.85840294   0.70369147   0.70369147

```

Then, the  $x$ ,  $y$ ,  $z$  dipole moment components and the norm using the Mulliken charges are shown in both atomic unit and Debye unit. This is not printed if periodic boundary condition is used.

DIPOLE MOMENT [a.u.]

```

-----
          x           y           z
0.4545263289   0.4545263289   0.0000000000
NORM :    0.6427972988

```

DIPOLE MOMENT [Debye]

```

-----
          x           y           z
1.1552906835   1.1552906835   0.0000000000
NORM :    1.6338277531

```

If **MAYER=TRUE** in **SCC** or **NCC** keyword, atomic valences (diagonal elements) and Mayer bond orders (off-diagonal elements) are shown with matrix representation.

ATOMIC VALENCIES AND MAYER BOND ORDERS

```

-----
          1           2           3
1  1.796085   0.898042   0.898042
2  0.898042   0.912201   0.014159
3  0.898042   0.014159   0.912201

```

If **CM3=TRUE** in **SCC** or **NCC** keyword, the **CM3** net atomic charges and dipole moments are additionally printed.

CM3 NET ATOMIC CHARGES

```
-----  
1   -0.5833492665  
2    0.2916746332  
3    0.2916746332
```

CM3 DIPOLE MOMENT [a.u.]

```
-----  
      x           y           z  
0.4474181000   0.4474181000   0.0000000000  
NORM :    0.6327447451
```

CM3 DIPOLE MOMENT [Debye]

```
-----  
      x           y           z  
1.1372233679   1.1372233679   0.0000000000  
NORM :    1.6082767104
```

Then, the different energy contributions to the total energy are shown in Hartree unit. If you use **MPI/OpenMP** version, **PRINTATOME=TRUE** in **MISC** keyword is necessary to obtain the corresponding information.

ENERGY CONTRIBUTION [Eh]

```
-----  
ENERGY H0           = -4.1689461692  
ENERGY SCC          =  0.0183616273  
TOTAL ELECTRONIC ENERGY = -4.1505845420  
REPULSIVE ENERGY   =  0.0726464065  
TOTAL ENERGY       = -4.0779381355
```

If **PRINTATOME=TRUE** in **MISC** keyword, the atomic energy contributions to the total, electronic, and repulsive energies are additionally printed in Hartree unit.

ATOM RESOLVED ENERGY CONTRIBUTION [Eh]

	TOTAL	ELECTRONIC	REPULSIVE
1	-3.3763986686	-3.4127218718	0.0363232032
2	-0.3507697335	-0.3689313351	0.0181616016
3	-0.3507697335	-0.3689313351	0.0181616016

### 3.3 Other output files

Additional output files will be generated depending on the type of calculations:

- **“chrgfile”** (if SCC=TRUE)  
contains the information of Mulliken population for restarting purpose in binary format. The file may be used in READCHARGE=BINARY in SCC keyword.
- **“restart”** (if MD=TRUE)  
contains the information of last MD step for restarting purpose in binary format. The file may be used in RESTART=TRUE in MD keyword.
- **“restart\_chk”** (if MD=TRUE)  
contains the information of last checkpoint step for restarting purpose in binary format. The file may be used in RESTART=TRUE in MD keyword.
- **“traject”** (if MD=TRUE)  
contains the Cartesian coordinates during MD simulation in xyz format.
- **“velocity”** (if MD=TRUE)  
contains the velocity components during MD simulation in xyz format in Å/fs unit.
- **“mulliken”** (if MD=TRUE)  
contains the Mulliken net atomic charges during MD simulation. For serial and OpenMP versions, sequence of the following information is printed:
  - Line 1: Number of atoms
  - Line 2: Comments (simulation time and MD step)
  - Line 3–(Number of atoms + 2): Atom index (1st column), atom label (2nd column), and Mulliken net atomic charges (3rd column)

For MPI/OpenMP version, sequence of the following information is printed:

- Line 1: Number of shells (1st column) and number of atoms (2nd column)
  - Line 2: Comments (simulation time and MD step)
  - Line 3–(Number of shells + 2): Atom index (1st column), atom label (2nd column), angular momentum label (3rd column), and Mulliken net charges for the shell (4th column)
- **“dftb\_vib.out”** (if FREQ=TRUE)

contains the information of harmonic vibrational frequency analysis to visualize the results using GaussView.

- “**restart\_hess**” (if PRINTHESS=TRUE in FREQ keyword)  
contains the information of Hessian matrix elements for restarting purpose. The file may be used in RESTARTHESS=BINARY in FREQ keyword.
- “**hess.dat**” (if INTERRUPTCP=TRUE in FREQ keyword)  
contains the information of partial Hessian matrix elements for restarting purpose. The file may be used in RESTARTHESS=ASCII in FREQ keyword.

You may find other files (zacaz, zbcbz, zzszz) in serial and OpenMP versions if USESCRATCH=TRUE in MISC keyword but these are intermediate binary files and not important to examine the results.

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