DeltaSpin (Self-Adaptive Spin Constraint in VASP)

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Contents

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2 Docker Image User Guide

For the following commands, replace the values within the angle brackets (`<>`) with relevant values for your system:

1. Pull the DeltaSpin image from ghcr.io:

```
docker pull ghcr.io/caizefeng/deltaspin:latest
```

2. Start a container using this image. Please note that you should allocate at least 16 GB of shared memory when starting the container from this image:

```
docker run -d --shm-size=<size of shared memory> \
--name=deltaspin ghcr.io/caizefeng/deltaspin:latest
```

3. Enter the container:

```
docker exec -it deltaspin bash
```

4. Navigate to your calculation directory and place the activation code in the same directory as the `INCAR`, `POSCAR`, `POTCAR`, and `KPOINTS` files:

```
cd /root/DeltaSpin/examples/metal/Fe
cp <path to your activation code file> .
```

5. Run `vasp_deltaspin`:

```
mpirun -np <number of threads> /root/DeltaSpin/bin/vasp_deltaspin
```

3 Getting Started

1. *Determine Wigner-Seitz radius (in Å) for each atom type in the system.*

   Read the `RWIGS` entry presented in `POTCAR` and use that value for INCAR. Note that this value should **remain constant** in every single calculation of this system.

   (OPTIONAL) Though tedious and not recommended, you can also determine the Wigner-Seitz radius through a Bader-type **magnetization density** analysis.

2. *Determine the magnetic ground state of the system.*

   Set up `INCAR` with the following parameters:

```
RWIGS = <values from the 1st step>
I_CONSTRAINED_M = 2
SCTYPE = 0
LAMBDA = 0
```

   and run `vasp_deltaspin`. When the calculation is finished, the last `MW_current` in `OSZICAR` represents the new ground-state `RWIGS`-defined magnetic moments.

   Note that `MAGMOM` should be close to the ground-state.

   You can add the following two tags to increase precision in ANY non-collinear calculation.

```
LASPH = .TRUE.
GGA_COMPAT = .FALSE.
```

3. *Constrain the system to the desired magnetic configuration.*

   Refer to the `INCAR` template in `DeltaSpin/templates` directory. Set `M_CONSTR` to any value you want to constrain the magnetic moments to.

   Run `vasp_deltaspin`.

   Note that the ground-state magnetic configuration for reference, throughout all DeltaSpin calculations of this specific material system, is what we obtained in the 2nd step instead of any empirical value or `magnetization` in `OUTCAR` [1]. Different definitions of "magnetization" used in VASP will be discussed in another section.

4. *(OPTIONAL) Non-Self-Consistent (NSCF) calculation.*

   Set `INCAR-LAMDA` to the last value of `OSZICAR-lambda` from the same **SCF** calculation that `CHGCAR` is from.

   Also, complete the standard procedure for a typical VASP NSCF calculation, like preparing a `CHGCAR` and setting `INCAR-ICHARG >= 10`.

   Run `vasp_deltaspin`.

[1] This means that if you set `M_CONSTR` to the last `MW_current` in `OSZICAR` from the 2nd step and apply the constraining, the energy will be exactly the same.

4 INCAR Template

```
SYSTEM = NiO
#GGA = PE

#Electronic minimization
PREC = Accurate               # Accurate is always preferred since non-collinear magnetization is very sensitive to the completeness
      of FFT grid.
ENCUT = 600
EDIFF = 1E-9
LORBIT  = 11
LREAL = .FALSE.
ISTART  = 0
NPAR  = 4
#ICHARG =  1
VOSKOWN = 1
LWAVE = .FALSE.
LCHARG = .FALSE.
NELMIN = 10
NELM = 500

#IBRION = 2 #OPT
#EDIFFG = -1E-3 #OPT
#ISIF = 3 #OPT
#NSW = 500 #OPT
#POTIM = 0.2  #OPT

#DOS
ISMEAR = -5
SIGMA = 0.2
#NEDOS = 3000 #DOS

#Magnetism
# The DEFINITION of atomic spins which are constrained (M_CONSTR in INCAR, MW in constrmag.F/OSZICAR):
# \vec{M}_{I} =
# \int_{\Omega_{I}} \vec{m}(\mathbf{r}) W_{I}(\mathbf{r}) d\mathbf{r}

#External magneitc field (Zeeman field)
Bfield = 0 0 0                # External field in Cartesian coordinate (X,Y,Z), The unit is Tesla.                !IMPORTANT!

#Non-collinear
ISYM = -1
RWIGS = 1.286 0.820           # Wigner-Seitz radius (in angstrom) for each atom type in the system.              !IMPORTANT!
GGA_COMPAT = .FALSE.          # Restore the full lattice symmetry for gradient corrected functionals. Recommended.
LASPH = .TRUE.                # Non-spherical contribution to the gradient of the density. Recommended.
ISPIN = 2                     # Does not matter in non-collinear calculation.
LNONCOLLINEAR = .TRUE.
LSORBIT  = .TRUE.

#SASC
I_CONSTRAINED_M = 2
MAGMOM = 0.75914 0.16146 -1.00688 -0.67954 -0.91284 0.56668 0.00000 0.00000 0.00000 0.00000 0.00000 0.00000
M_CONSTR = 0.75914 0.16146 -1.00688 -0.67954 -0.91284 0.56668 0.00000 0.00000 0.00000 0.00000 0.00000 0.00000      !IMPORTANT!
SCTYPE = 1                    # Type of SASC calculation   0: noSASC  1: SASC(L)  2: SASC(Q)  3: SASC(QL)         !IMPORTANT!

#SASC(L)
IDECOSC = 0                   # Whether Lagrangian coefficients are coupled   0: coupled  1: decoupled between atoms (deprecated)  2:
       decoupled between every Cartesian component (deprecated)
LAMBDA   = 12*0               # Initial Lagrangian coefficients for SASC(L)
CONSTRL  = 6*1 6*0            # Whether the component is constrained or not   0: not constrained  1: constrained  !IMPORTANT!
NSC = 100                     # Maximum number of steps for each inner optimization
NSCMIN = 2                    # Minimum number of steps for each inner optimization
SCDIFF = 1E-8                 # Convergence criterion of iteration (RMS)
SCCONVB_GRAD = 1.9 -1         # Break condition for local gradients of spins w.r.t. Lagrangian coefficients (local field)   -1: off
      for the element

IDECAY_GRAD = 2               # Decay policy of gradient break condition   0: no decay  1: exponential decay  2: step down
SCDECAY_GRAD = 0.9            # Exponetial base of decayed gradient break condition (IDECAY_GRAD = 1)
SCGRADB = 0.1                 # Lower bound of exponetially decayed gradient break condition
NGRAD = 2                     # Number of "steps" the gradient break condition curve includes (IDECAY_GRAD = 2)
NGRAD_STEP = 200 400          # Indices in the outer loop where the gradient break condition discontinuously decay
NGRAD_VALUE = 1.7 -1 \        # SCCONVB_GRAD for corresponding "steps"
              1.5 -1

SCDECAY = -1                  # Exponential decay of convergence criterion   -1: no decay  >0,<1: exponential base
SCDIFFB = 1E-7                # Lower bound of criterion decay, not gonna work if SCDECAY = -1
SCCONVB = -1                  # Break condition for Lagrangian coefficients difference between two inner steps   -1: this condition
      is off
INISC = 0.01                  # Initial trial step size
SCCUT = 3                     # Restriction of step size
NELMSCI = 1                   # Number of electronic steps BEFORE SASC(L)
NELMSCT = 0                   # Number of INTERMEDIATE normal electronic steps
IALGOSC = 1                   # Inner optimization algorithm   1: CG(F-R) 2: CG(P-R) 3: CG(H-S) 4: CG(D-Y)
IALGOSC_DIAG = 1              # Inner diagonalization algorithm   1: sub-space rotation  2: Blocked-Davidson  3: sub-space rotation
      with B-D for the last step
LCUTSC_TRIAL = .TRUE.         # Whether trial step size is updated acoording to last-step optimal value   TRUE: update  FALSE: do not
       update
LDESC = .FALSE.               # Debug mode   TRUE: on  FALSE: off

#SASC(Q)
```

```
LAMBDA_Q   = 10 10 10 10 10 10 # Initial Lagrangian coefficients for SASC(Q)
CONSTRL_Q  = 1 1 1 1 1 1       # Whether the component is constrained or not   0: not constrained  1: constrained
NSC_Q = 500                    # Maximum number of steps for SASC(Q) iteration
SCDIFF_Q = 3E-4                # Convergence criterion of iteration (+Inf-Norm)
EDIFF_Q = 1E-6                 # Global break condition (EDIFF) for SASC(Q)
INISC_Q = 0.1                  # Initial trial step size
LDESC_Q = .FALSE.              # Debug mode   TRUE: on  FALSE: off

#Orbital momemts
#LORBMOM = .TRUE.

#Mixer
AMIX      = 0.2
BMIX      = 0.0001
AMIX_MAG  = 0.8
BMIX_MAG  = 0.0001

#LSDA+U
LDAU      = .TRUE.
LDAUTYPE  = 2
LDAUL     = 2 -1
LDAUU     = 5.3 0.0
LDAUJ     = 0.0 0.0
LDAUPRINT = 2
LMAXMIX   = 4
```

5 Selected Multi-step Use Cases

5.1 Calculate the effective field of the original ground-state configuration under Zeeman field

1. Relax the system to reach the ground-state magnetic configuration without an external field.

```
Bfield = 0 0 0
I_CONSTRAINED_M = 2
SCTYPE = 0
LAMBDA = 0
```

2. Apply a finite external field B. Constrain the size and direct of the magnetic moments using DeltaSpin.

```
Bfield = <B_x> <B_y> <B_z>
I_CONSTRAINED_M = 2
SCTYPE = 1
LAMBDA = <number of atoms>*0
M_CONSTR = <the last "M_current" from step 1>
```

5.2 Calculate the effective field of a direction-constrained magnetic configuration

1. Constrain the direction of the magnetic moments using the original VASP algorithm that is compatible with DeltaSpin.

```
I_CONSTRAINED_M = 1
SCTYPE = 0
LAMBDA = <some positive real number>
```

2. Constrain the size and direct of the magnetic moments using DeltaSpin.

```
I_CONSTRAINED_M = 2
SCTYPE = 1
LAMBDA = <number of atoms>*0
M_CONSTR = <the last "M_current" from step 1>
```

5.3 Calculate the electronic structure of a fully-constrained configuration

1. Self-consistently constrain the size and direct of the magnetic moments using DeltaSpin.

```
ISTART = 0
ICHARG = 2
LAMBDA = <number of atoms>*0
LWAVE = .TRUE.
LCHARG = .TRUE.
```

2. Change to a desired k-point grid/path. Run DeltaSpin non-self-consistently.

```
ISTART = 1
ICHARG = 11
LAMBDA = <the last "lambda" from step 1>
```

6 Strategies

When the energy difference (dE) keeps fluctuating around a value above the normal EDIFF (approximately 1E+02 eV ⇔ 1E-02 eV), consider the following steps:

1. TEST FOR A SUITABLE GRADIENT CRITERION, i.e. `SCCONVB_GRAD`. Modifying this value might have a significant impact on convergence. Some values of `SCCONVB_GRAD` may result in a complete corruption of the diagonalization process.
   Note that the same element in different systems may have different optimal gradient criteria. For example, the optimal `SCCONVB_GRAD` for chromium (Cr) is 0.5 (μ_B^2/eV) in a monolayer CrI_3, but it is 2.0 in CrB_2. Normally, a simple GRID SEARCH is sufficient to find a decent value for `SCCONVB_GRAD`.

2. SLOW DOWN THE MIXING. Decrease the values of `AMIX` and `AMIX_MAG`, and usually, it is necessary to increase `NELM` at the same time. To avoid out-of-memory issues caused by the increasing number of electronic steps, you may also need to allocate more memory when specifying the computing resources.

7 Compatibility

Using `SCTYPE=0`, *DeltaSpin* is now compatible with almost all functionalities from the original VASP, including

1. Constrained local moments approach (direction, `I_CONSTRAINED_M = 1`)
2. Constrained local moments approach (size and direction, `I_CONSTRAINED_M = 2`)
3. Spin spiral (`LSPIRAL=.TRUE.`)

8 Discussion on Magnetization in VASP

8.1 Total magnetization

Total magnetization (`mag=` in `stdout`, which is usually redirected to a log file, or `OSZICAR`), is the integral of magnetization density $\rho(r)$ **over the entire periodic box $\boldsymbol{\Omega}$**.

$$\mathbf{M}_{\text{total}} = \int_{\Omega} \rho(r) \tag{1}$$

where

$$\rho(r) = \sum_n f_n \langle \Psi_n | r \rangle \, \sigma \, \langle r | \Psi_n \rangle \tag{2}$$

8.2 OUTCAR magnetization

`OUTCAR` magnetization (`magnetization (x,y,z)` in `OUTCAR`), is the integral of pseudo magnetization density $\rho^1(r)$ (defined in the PAW formalism) **over the area $\boldsymbol{\Omega}_{\mathbf{R}}^{\text{AE}}$**, where all-electron (AE) partial wave has integer occupancy. Note that the area can be "slightly" different from the augmentation sphere $\Omega_{\mathbf{R}}$.

$$\mathbf{M}_{\text{OUTCAR}} = \int_{\Omega_{\mathbf{R}}^{\text{AE}}} \rho^1(r) \tag{3}$$

where

$$\rho^1(r) = \sum_{n,(i,j)} f_n \langle \tilde{\Psi}_n | \tilde{p}_i \rangle \langle \phi_i | r \rangle \sigma \langle r | \phi_j \rangle \langle \tilde{p}_j | \tilde{\Psi}_n \rangle \tag{4}$$

$$\int_{\Omega_{\mathbf{R}}^{\text{AE}}} \langle \phi_i | r \rangle \langle r | \phi_j \rangle = \delta_{ij} \tag{5}$$

Given

$$\begin{aligned} \rho(r) &= \tilde{\rho}(r) + \rho^1(r) - \tilde{\rho}^1(r) \\ &= \sum_n f_n \langle \tilde{\Psi}_n | r \rangle \sigma \langle r | \tilde{\Psi}_n \rangle + \rho^1(r) - \sum_{n,(i,j)} f_n \langle \tilde{\Psi}_n | \tilde{p}_i \rangle \langle \tilde{\phi}_i | r \rangle \langle r | \tilde{\phi}_j \rangle \sigma \langle \tilde{p}_j | \tilde{\Psi}_n \rangle \end{aligned} \tag{6}$$

and

$$\sum_i |\tilde{\phi}_i\rangle\langle\tilde{p}_i| = 1 \text{ within } \Omega_{\mathbf{R}} \tag{7}$$

If we assume $\Omega_{\mathbf{R}}^{\text{AE}} \approx \Omega_{\mathbf{R}}$, we have

$$\mathbf{M}_{\text{OUTCAR}} = \int_{\Omega_{\mathbf{R}}^{\text{AE}}} \rho^1(r) \approx \int_{\Omega_{\mathbf{R}}} \rho(r) \tag{8}$$

that is, the integral of magnetization density $\rho(r)$ **over the augmentation sphere**.

8.3 OSZICAR magnetization

`OSZICAR` magnetization (`M_int`, `M_current`(w/ DeltaSpin) in `OSZICAR`), is the integral of magnetization density **over a user-defined real-space sphere**.

$$\mathbf{M}_{\mathrm{OSZICAR}} = \int_{\Omega'_{\mathbf{R}}} \rho(r) \tag{9}$$

Weighted `OSZICAR` magnetization (`MW_int`, `MW_current`(w/ DeltaSpin) in `OSZICAR`), is the weighted integral of magnetization density in a user-defined real-space sphere. Prevalent weight functions are Bessel functions.

$$\mathbf{M}^{\mathrm{W}}_{\mathrm{OSZICAR}} = \int_{\Omega'_{\mathbf{R}}} W(r)\,\rho(r) \tag{10}$$

8.4 Relation between different definitions

If we define $\Omega'_{\mathbf{R}}$ using Wigner–Seitz (WS) radii (`RWIGS`) in `POTCAR`, such that $\Omega'_{\mathbf{R}} \approx \Omega_{\mathbf{R}}$, "OUTCAR magnetization" and "OSZICAR magnetization" can be close (closer than using the partial core radius `RPCOR` or the outmost cutoff radius `RCORE`), but there will still be a notable difference. That's because these these two values originate from entirely different sources.

Most importantly, both of them are based on the muffin-tin approximation, meaning that sum of all atomic magnetic moments won't be equal to the "total magnetization", which is the all-space integral. This discrepancy arises because there are always "gaps" between these "spheres."

To obtain a new definition of atomic magnetization, the summation of which exactly equals the all-space integral, one could apply Bader analysis on the magnetization density.

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