DeltaSpin (Self-Adaptive Spin Constraint in VASP)

Zefeng Cai

July 2021

1 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 **spin 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_ncl`. 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_ncl`.

   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_ncl`.

[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.

2 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}) F_{I}(|\mathbf{r}|) d\mathbf{r}

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
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 Lagragian coefficients are coupled   0: coupled  1: decoupled between atoms  2: decoupled
      between every Cartesian component
LAMBDA   = 12*0               # Initial Lagragian 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. Lagragian 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 Lagragian 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 Lagragian 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 mom.
#LORBMOM = T

#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
```

3 Magnetization in VASP

"*OUTCAR magnetization*" is the integral of spin density in an area $\Omega_{\mathbf{R}}^{\mathrm{AE}}$, where AE partial wave has integer occupancy, which can be slightly different from the augmentation sphere $\Omega_{\mathbf{R}}$.

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

$$\rho^1(r) = \sum_{n,(i,j)} f_n \langle \tilde{\Psi}_n | \tilde{p}_i \rangle \langle \phi_i | r \rangle \sigma \langle r | \phi_j \rangle \langle \tilde{p}_j | \tilde{\Psi}_n \rangle \tag{2}$$

where

$$\int_{\Omega_{\mathbf{R}}^{\mathrm{AE}}} \langle \phi_i | r \rangle \langle r | \phi_j \rangle = \delta_{ij} \tag{3}$$

Because

$$\begin{aligned} \rho(r) &= \tilde{\rho}(r) + \rho^1(r) - \tilde{\rho}^1(r) \\ &= \sum_n f_n \langle \tilde{\Psi}_n | r \rangle \sigma \langle r | \tilde{\Psi}_n \rangle + \rho^1(r) - \sum_{n,(i,j)} f_n \langle \tilde{\Psi}_n | \tilde{p}_i \rangle \langle \tilde{\phi}_i | r \rangle \langle r | \tilde{\phi}_j \rangle \sigma \langle \tilde{p}_j | \tilde{\Psi}_n \rangle \end{aligned} \tag{4}$$

and

$$\sum_i |\tilde{\phi}_i\rangle\langle\tilde{p}_i| = 1 \text{ within } \Omega_{\mathbf{R}} \tag{5}$$

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

$$\mathbf{M}_{\mathrm{OUTCAR}} = \int_{\Omega_{\mathbf{R}}^{\mathrm{AE}}} \rho^1(r) \approx \int_{\Omega_{\mathbf{R}}} \rho(r) \tag{6}$$

"*OSZICAR magnetization*", is the integral of spin density in a user-defined real-space sphere.

$$\mathbf{M}_{\mathrm{OSZICAR}} = \int_{\Omega'_{\mathbf{R}}} \rho(r) \tag{7}$$

"*Weighted OSZICAR magnetization*", is the weighted integral of spin density in a user-defined real-space sphere. Prevalent weight functions are Bessel functions.

$$\mathbf{M}_{\mathrm{OSZICAR}}^{\mathrm{W}} = \int_{\Omega'_{\mathbf{R}}} W(r)\, \rho(r) \tag{8}$$

If we define $\Omega'_{\mathbf{R}}$ using WS radii (`RWIGS`) in `POTCAR`, that is $\Omega'_{\mathbf{R}} \approx \Omega_{\mathbf{R}}$,"*OUTCAR magnetization*" and "*OSZICAR magnetization*" will be closer (closer than partial core radius `RPCOR` and outmost cutoff radius `RCORE`), but still have a notable difference. That's because these two have totally different origins.

Most importantly, both of them are based on the muffin-tin approximation and the summation

of all atomic magnetic moments won't be equal to total magnetization. It's because there always exist some gaps in between those spheres.

The only well-defined quantity in DFT is "total magnetization", the all-space integral of charge density, which is `mag` in `OSZICAR`.

To get access to a new definition of atomic magnetization, the summation of which is exactly the all-space integral, one could apply Bader analysis on spin density.