Workshop and International conference on Electronic Structure Theory of Emergent Spin Orbit Driven Phenomenon

Tutorial 4

Calculation of magnetic exchange interactions

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Introduction:

$$H = -\sum_{i>j} J_{ij} \vec{S}_i \cdot \vec{S}_j$$

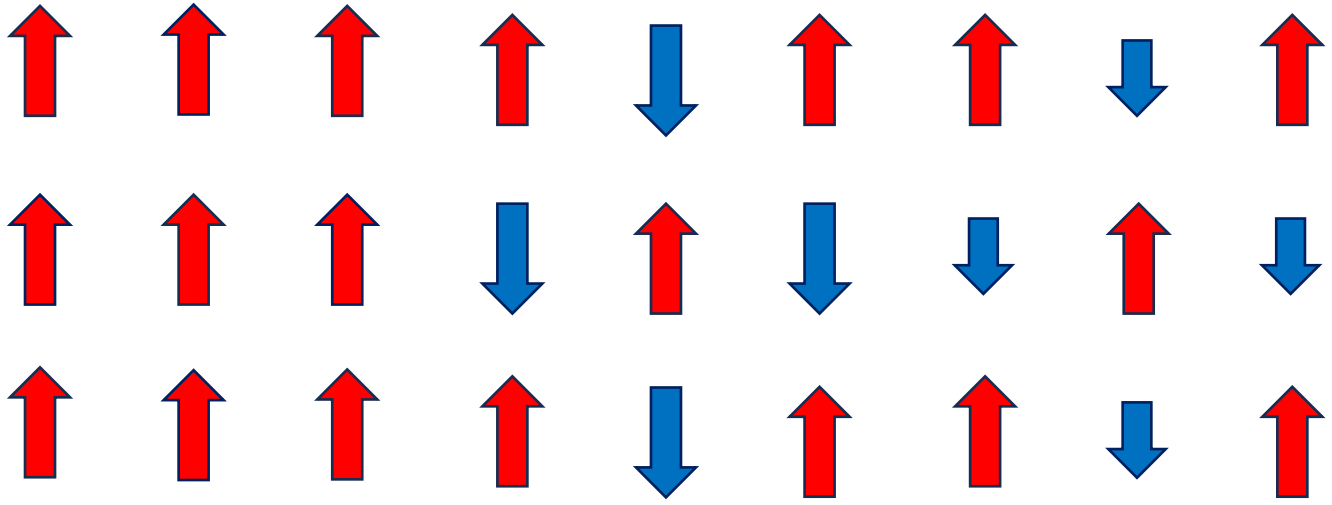
J_{ij} = interatomic exchange interaction constant

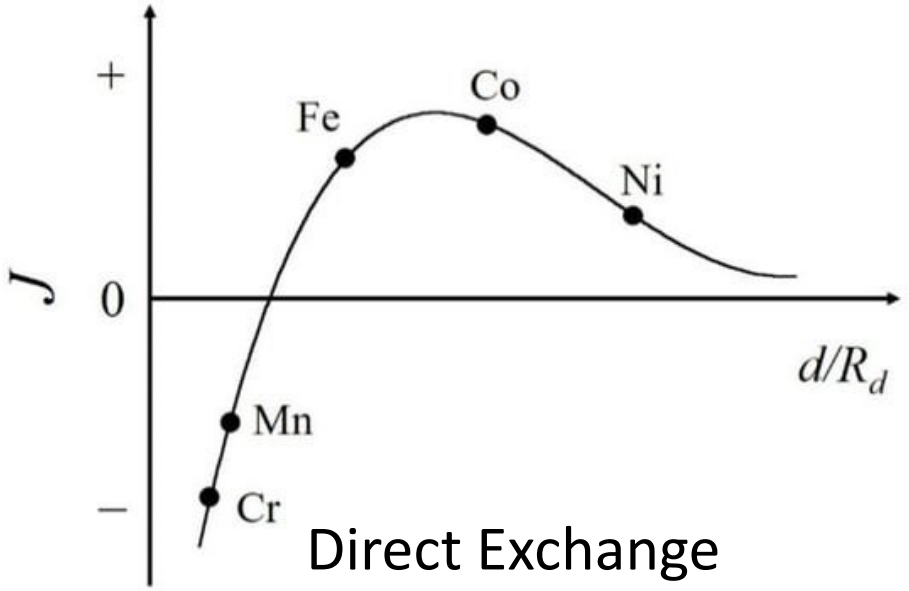
S_i and S_j are spins at site i and j

J > 0 Ferromagnet

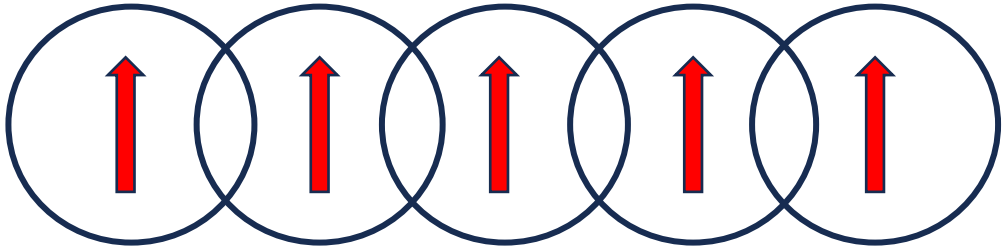
J < 0 Antiferromagnet

Ferrimagnet

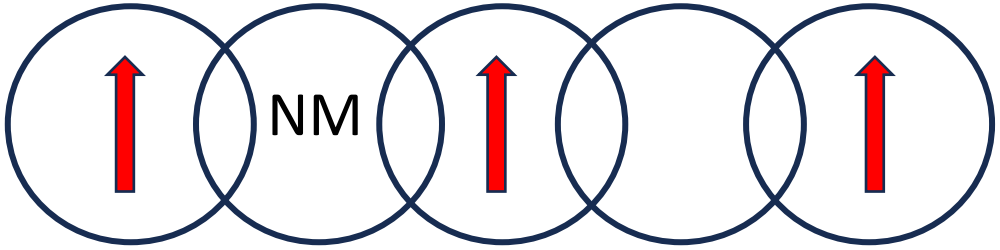




Direct Exchange



Indirect Exchange

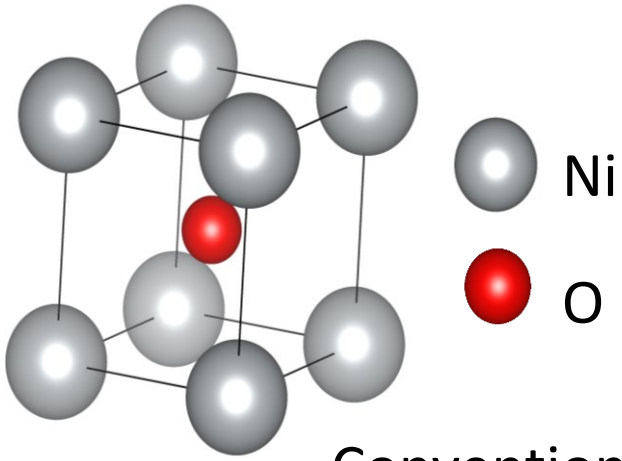


Indirect exchange (Super-exchange and double exchange) mechanism explains the long range magnetic ordering while direct exchange interaction is short range.

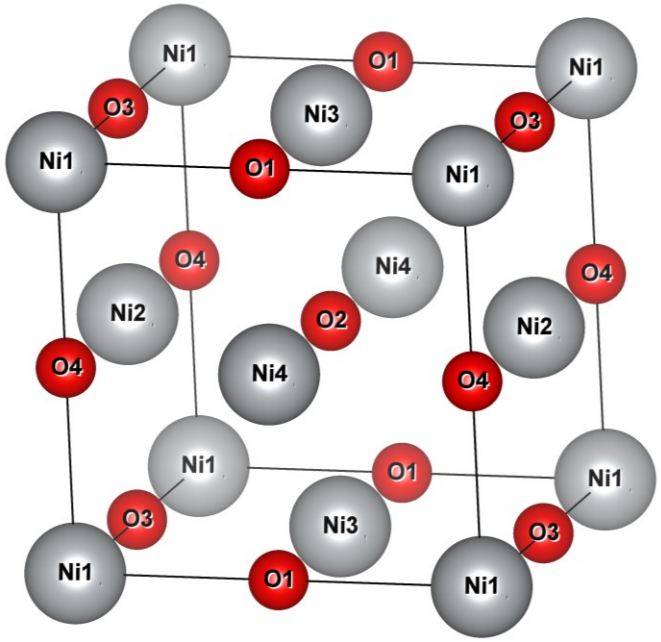
How to calculate exchange coupling constant ?

Structure of NiO

Primitive cell



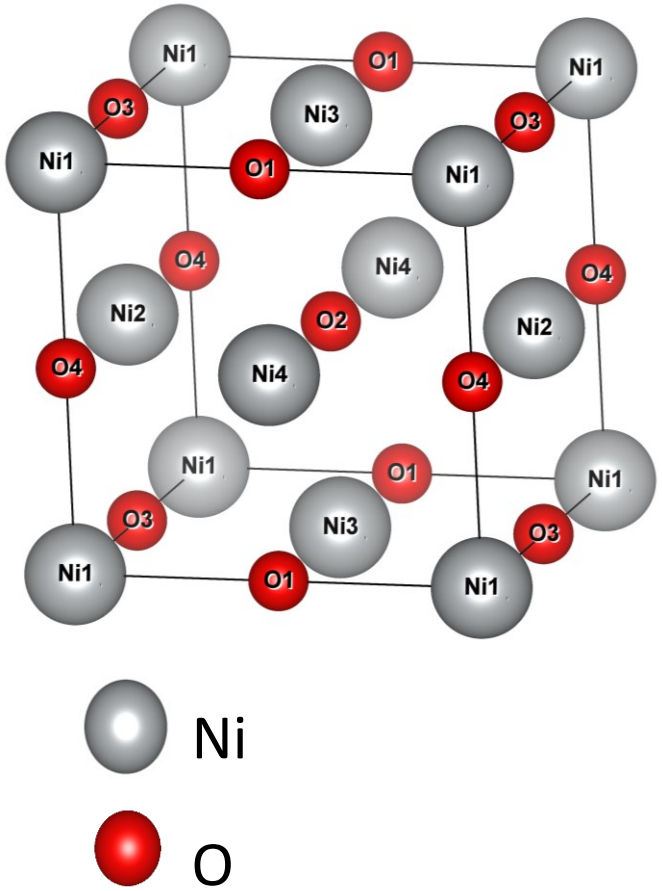
Conventional cell



Structure of conventional cell

```
Ni4 O4
1.00000000000000
4.1760000000000002    0.0000000000000000  0.0000000000000000
0.0000000000000000    4.1760000000000002  0.0000000000000000
0.0000000000000000    0.0000000000000000  4.1760000000000002
Ni   O
 4    4
Direct
0.0000000000000000  0.0000000000000000  0.0000000000000000
0.0000000000000000  0.5000000000000000  0.5000000000000000
0.5000000000000000  0.0000000000000000  0.5000000000000000
0.5000000000000000  0.5000000000000000  0.0000000000000000
0.5000000000000000  0.0000000000000000  0.0000000000000000
0.5000000000000000  0.5000000000000000  0.5000000000000000
0.0000000000000000  0.0000000000000000  0.5000000000000000
0.0000000000000000  0.5000000000000000  0.0000000000000000
```

Input for spin polarized calculation




```
 1 System=bulk_NiO
 2 ISTART=0
 3 ICHARG=2
 4 ISMEAR =0
 5 SIGMA = 0.05
 6 ENCUT= 500
 7 ISIF=2
 8 IBRION =-1
 9 ISPIN=2
10 MAGMOM= 3 -3 -3 3 4*0
11 NSW=0
12 Prec=Accurate
13 ALGO =Normal
14 EDIFF = 0.1E-05
15 EDIFFG = -0.001
16 LORBIT=11
17 LMAXMIX=4
18 LREAL= Auto
```

Need to change this configuration for various magnetic configurations

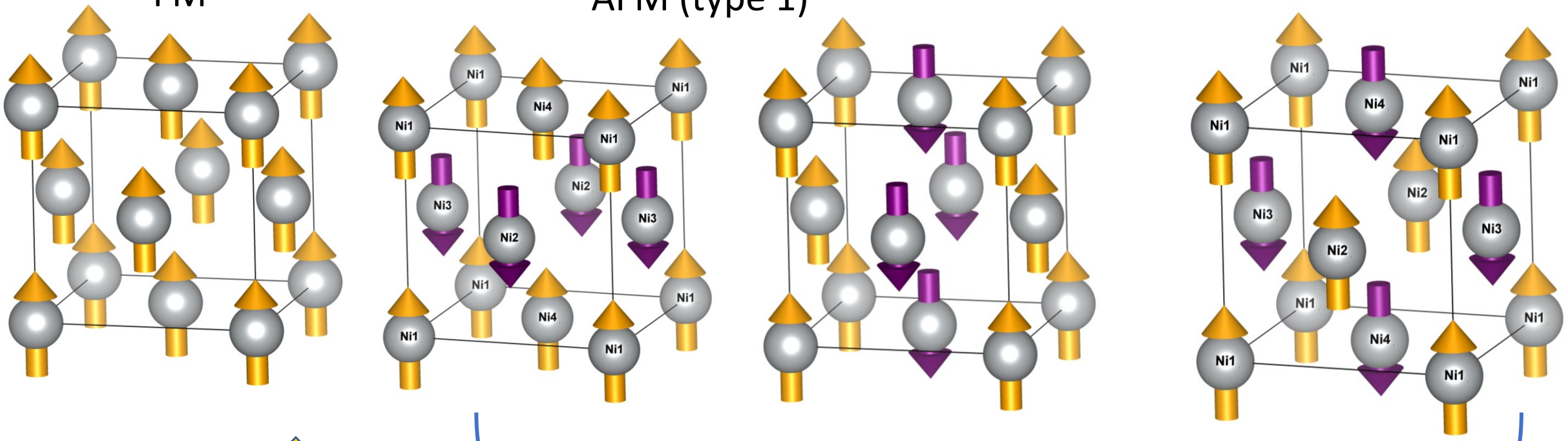
DFT + U

```
 1 System=bulk_NiO
 2 ISTART=0
 3 ICHARG=2
 4 ISMEAR =0
 5 SIGMA = 0.05
 6 ENCUT= 500
 7 ISIF=2
 8 IBRION =-1
 9 ISPIN=2
10 MAGMOM= 3 -3 -3 3 4*0
11 NSW=0
12 Prec=Accurate
13 ALGO =Normal
14 EDIFF = 0.1E-05
15 EDIFFG = -0.001
16 LORBIT=11
17 LMAXMIX=4
18 LREAL= Auto
19 # GGA +U
20 LDAU= .TRUE.
21 LDAUTYPE=2
22 LDAUL=2 0
23 LDAUU=6.2 0
24 LDAUJ=0 0
25 LDAUPRINT=0
```

Magnetic configurations

FM

AFM (type 1)



Ni up spin

Ni down spin

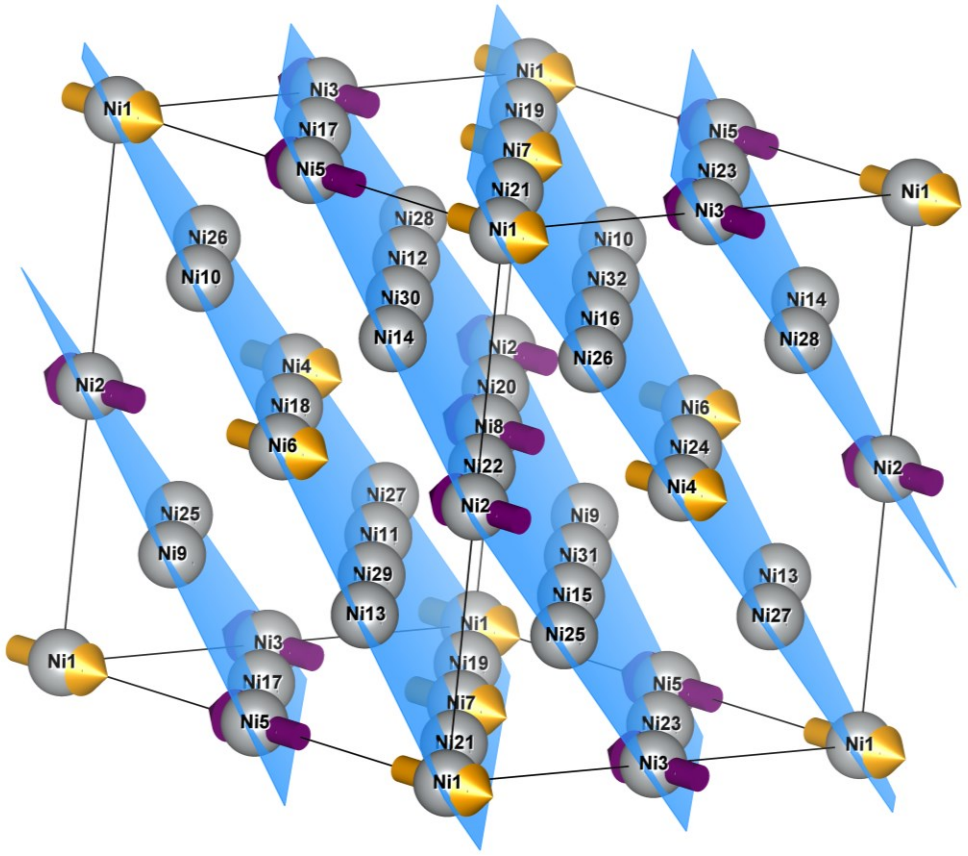
Degenerate energy states

Play in VESTA with different spin configurations

Is there any other nondegenerate configurations possible?

Magnetic configurations

AFM (type 2)



AFM-2 is found as magnetically ground state

INCAR

```
System=bulk_NiO
ISTART=0
ICHARG=2
ISMEAR =0
SIGMA = 0.05
ENCUT= 400
ISIF=2
IBRION =-1
ISPIN=2
MAGMOM= 3 -3 -3 3 -3 3 3 -3 -3 3 3 -3 3 -3 -3 3
     -3 3 3 -3 3 -3 -3 3 -3 3 3 -3 3 -3 -3 3 32*0
NSW=0
EDIFF = 0.1E-06
NELM=180
EDIFFG = -0.001
Prec=Accurate
ALGO =Normal
#IALGO=38
LORBIT=11
LCHARG=.TRUE.
LWAVE=.TRUE.
LREAL= Auto
LDAU= .TRUE.
LDAUTYPE=2
LDAUL=2 0
LDAUU= 6.2 0
LDAUJ=0 0
LMAXMIX= 4
LDAUPRINT=0
```

Ni up spin

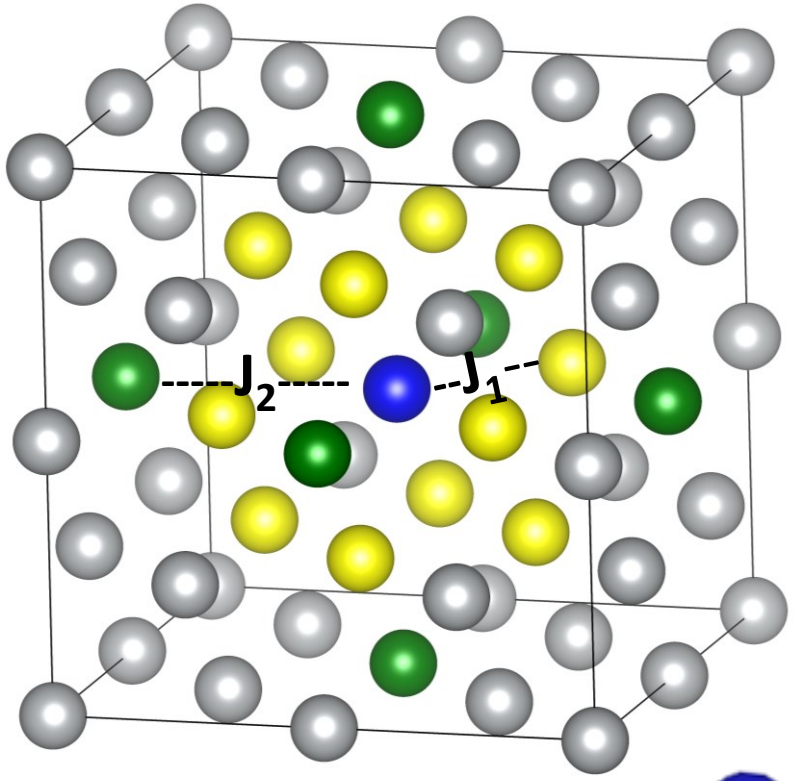
Ni down spin

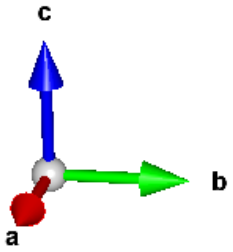
Phase	Energy (eV/f.u)	Δ E (meV / f.u.)
FM	-9.34	0
AFM1	-9.35	10
AFM2	**-9.45**	−110

Nickle oxide (NiO): Exchange coupling

- Make a supercell of 2 × 2 × 2 from the conventional cell
- Visualize it on VESTA
- Supercell contains: 64 atoms
- Ni: 32
- O: 32

2 × 2 × 2 supercell
(O atoms are not shown)





Number of nearest neighbors (NN) = 12
Number of next nearest neighbors (NNN) = 6

Reference atom

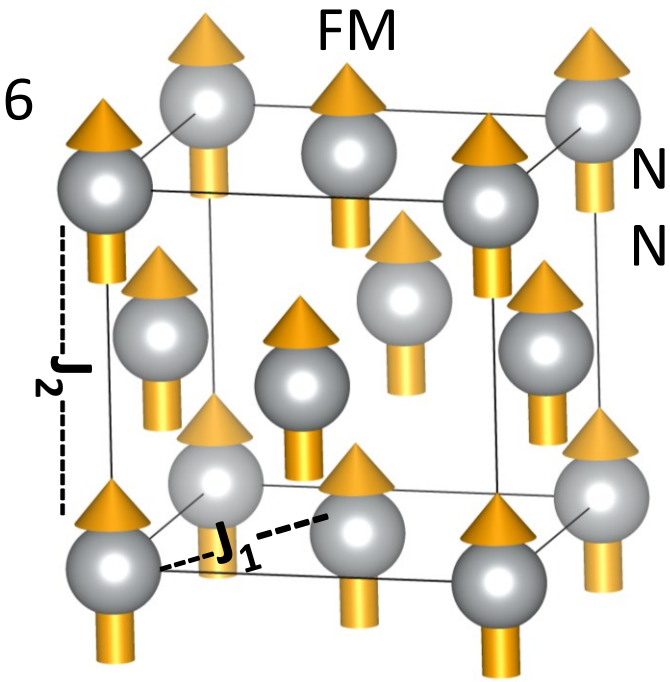
Nearest neighbor atom

Next nearest neighbor atom

FM configuration

- Number of nearest neighbors (NN) = 12
- Number of next nearest neighbors (NNN) = 6

- J_1: NN exchange & J_2: NNN exchange
- E (FM) = $-6\,J_1 - 3\,J_2 + E_0$



NN bond length =2.952 Å
NNN bond length = 4.176 Å

magnetization (x)

# of ion	s	p	d	tot
1	0.011	0.010	1.764	1.785
2	0.011	0.010	1.764	1.785
3	0.011	0.010	1.764	1.785
4	0.011	0.010	1.764	1.785
5	0.022	0.167	0.000	0.189
6	0.022	0.167	0.000	0.189
7	0.022	0.167	0.000	0.189
8	0.022	0.167	0.000	0.189
tot	0.131	0.708	7.054	7.893

$$\mu_{Ni} = 1.785\,\mu_B$$

AFM-1 configuration

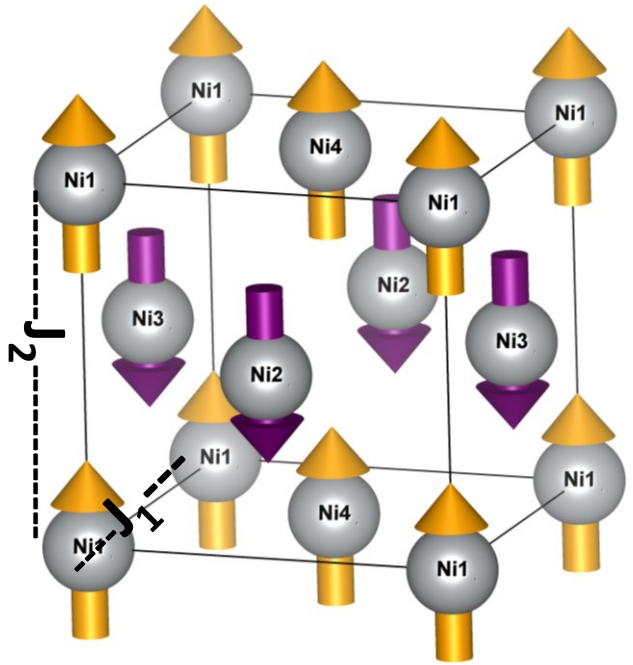
J_1: NN exchange & J_2: NNN exchange

magnetization (x)

# of ion	s	p	d	tot
1	0.013	0.013	1.769	1.795
2	-0.013	-0.013	-1.769	-1.795
3	0.013	0.013	1.769	1.795
4	-0.013	-0.013	-1.769	-1.795
5	0.008	0.056	0.000	0.064
6	-0.008	-0.056	0.000	-0.064
7	0.008	0.056	0.000	0.064
8	-0.008	-0.056	0.000	-0.064
tot	0.000	0.000	0.000	0.000

$$\mu_{Ni} = 1.795\ \mu_B$$

$$E\ (AFM1) = 2J_1 - 3J_2 + E_0$$

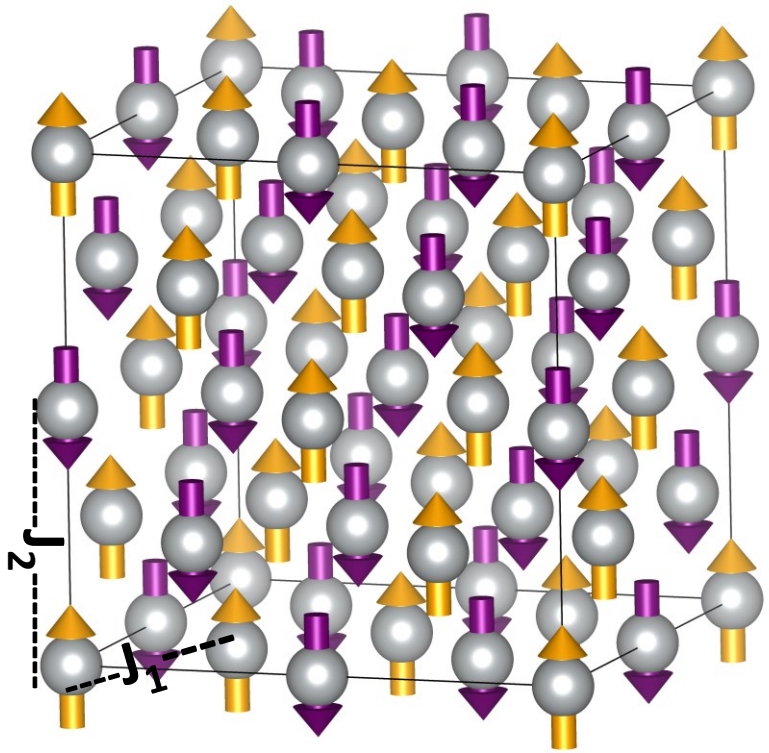


Ni up spin

Ni down spin

4 min (4 core)

AFM-2 configuration



E (AFM2) = $3J_2 + E_0$

magnetization (x)

# of ion	s	p	d	tot
1	-0.000	-0.002	1.722	1.720
2	0.000	0.002	-1.722	-1.720
3	0.000	0.002	-1.722	-1.720
4	-0.000	-0.002	1.722	1.720
5	0.000	0.002	-1.722	-1.720
6	-0.000	-0.002	1.722	1.720
7	-0.000	-0.002	1.722	1.720
8	0.000	0.002	-1.722	-1.720
9	0.000	0.002	-1.722	-1.720
10	-0.000	-0.002	1.722	1.720
11	-0.000	-0.002	1.722	1.720
12	0.000	0.002	-1.722	-1.720
13	-0.000	-0.002	1.722	1.720
14	0.000	0.002	-1.722	-1.720
15	0.000	0.002	-1.722	-1.720
16	-0.000	-0.002	1.722	1.720
17	0.000	0.002	-1.722	-1.720
18	-0.000	-0.002	1.722	1.720
19	-0.000	-0.002	1.722	1.720
20	0.000	0.002	-1.722	-1.720
21	-0.000	-0.002	1.722	1.720
22	0.000	0.002	-1.722	-1.720
23	0.000	0.002	-1.722	-1.720
24	-0.000	-0.002	1.722	1.720
25	0.000	0.002	-1.722	-1.720
26	-0.000	-0.002	1.722	1.720
27	-0.000	-0.002	1.722	1.720
28	0.000	0.002	-1.722	-1.720
29	-0.000	-0.002	1.722	1.720
30	0.000	0.002	-1.722	-1.720
31	0.000	0.002	-1.722	-1.720
32	-0.000	-0.002	1.722	1.720
33	0.000	0.000	0.000	0.000
34	0.000	0.000	0.000	0.000
35	0.000	0.000	0.000	0.000

$\mu_{Ni} = 1.720\ \mu_B$

NiO: Exchange parameter (J_1 &J_2 calculation)

Heisenberg Spin Hamiltonian

$$H = -\sum_{<i,j>} J_{ij} \vec{S}_i \cdot \vec{S}_j$$

$$H = -J_1 \sum_{<i,j>} \vec{S}_i \cdot \vec{S}_j - J_2 \sum_{\langle\langle i,j \rangle\rangle} \vec{S}_i \cdot \vec{S}_j$$

J_1: NN exchange, J_2: NNN exchange

J_{ij} =interatomic exchange interaction constant

$J_{ij} < 0$, FM

$J_{ij} > 0$, AFM

$E(FM) = -6J_1 - 3J_2 + E_0$

$E(AFM1) = 2J_1 - 3J_2 + E_0$

$E(AFM2) = 3J_2 + E_0$

$$J_1 = \frac{1}{8}[E(AFM1) - E(FM)]$$

$$J_2 = \frac{1}{24}[4 \times E(AFM2) - 3 \times E(AFM1) - E(FM)]$$

Exchange parameters	Cal. value (meV)	Reported value (meV)
J_1	1.25	1.4
J_2	-17.9	-19

- $J_1 > 0$ indicates 1 NN interaction is ferromagnetic
- $J_2 < 0$ indicates 2 NN interaction is antiferromagnetic
- $\frac{J_2}{J_1} \sim 14$ which means antiferromagnetic interaction dominates over ferromagnetic interaction.