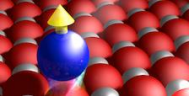
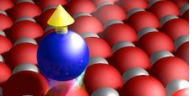


3.1 Monolayer with high Curie temperature

The Curie temperature of bulk Fe, Co and Ni is respectively 1043 K, 1388 K, and 627 K.
- Fe has a bcc structure with lattice constant a = 0.287 nm;
- Co has fcc structure (β-Co) with a = 0.355nm (for simplicity; in reality, hcp is the most frequent structure for Co);
- Ni has an fcc structure with a = 0.352 nm.

1) Calculate J_{ex} between a pair of atoms in each of the mentioned systems.

2) We want to grow a single atomic layer (monolayer) on Cu(100) with the highest Curie temperature. Assume that the bulk lattice constants are preserved and no hybridization takes place at the interface: which element among Fe, Co and Ni you chose?



3.1 Monolayer with high Curie temperature - Solution

1) We know that $T_C = \frac{2\,S(S+1)\,N\,J_{ex}}{3\,k_B}$

Fe has a bcc structure; thus each atom has N = 8 nearest neighbors.

Fe has a $4s^2\ 3d^6$ electronic configuration; thus, S = 2 -> $J_{ex} = \frac{3k_BT_C}{2\,S(S+1)N}$ = 2.8 meV

Co has a fcc structure; thus each atom has N = 12 nearest neighbors.

Co has a $4s^2\ 3d^7$ electronic configuration; thus, S = 3/2 -> $J_{ex} = \frac{3k_BT_C}{2\,S(S+1)N}$ = 4 meV

Ni has a fcc structure; thus each atom has N = 12 nearest neighbors.

Ni has a $4s^2\ 3d^8$ electronic configuration; thus, S = 1 -> $J_{ex} = \frac{3k_BT_C}{2\,S(S+1)N}$ = 3.4 meV

2) In a monolayer on a (100) surface, the number of nearest neighbor atoms is N = 4. We can use the previously calculated values of J_{ex} to calculate T_C for each element

We have T_C(Fe) = 521 K; T_C(Co) = 465 K; T_C(Ni) = 211 K.

Then in the monolayer regime Fe is the element with the highest Curie temperature (while in bulk it is Co)

3.2 Making a bit with 3 exchange coupled spins

Adapted from: *Inorg. Chem.* 2022, 61, 31, 12138–12148

We want to form a bit using 3 spins $\boldsymbol{S}_i$ coupled by an Heisenberg like exchange interaction. To be able to store information, a bit must be a binary system corresponding to the "0" or "1" stored information.

We arrange the three spins at the corners of a equilateral triangle such that each spin is coupled by an exchange constant J_{ex} to the two neighbors.

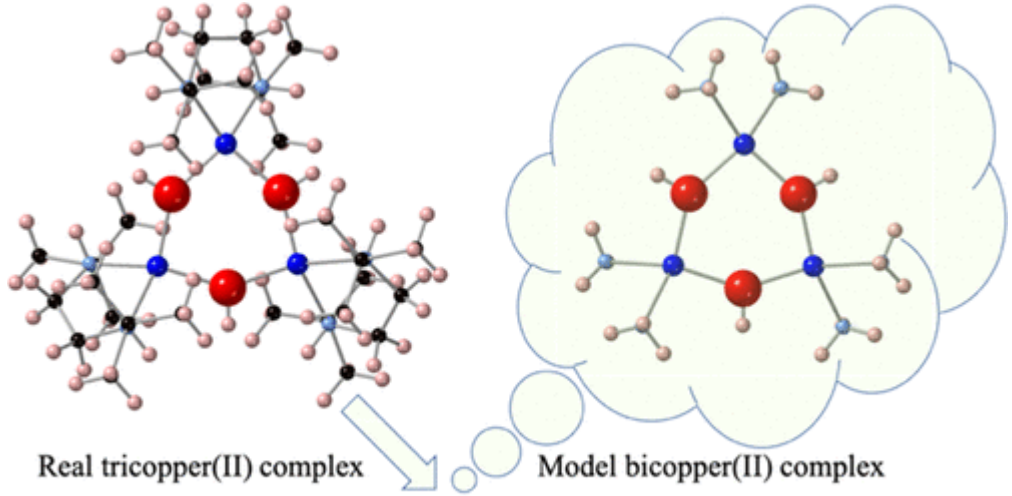
1) Assume that you can form a molecule like the one sketched in the figure with both Cu (S_i = ½) and Ni (S_i = 1), which species has to be used?

2) What is the value $\boldsymbol{S}$ of the spin molecule in the ground state?

3) What quantity can be used to define the states "0" and "1"?

4) Assume to be at T = 0 . Let define as "1" the value of the spin in the ground state when the external field is B = 0; what has to be the sign of J_{ex} to be able to write the bit applying a magnetic field?

5) Assuming $|J_{ex}|$ = 0.1 meV, what is the value of the magnetic field we need to apply to put the system in state "0"? Assume to be at T = 0 .



J and ***D*** effective coupling

3.2 Making a bit with 3 exchange coupled spins - solution

The Hamiltonian of the system is $H = -2J_{ex}(\boldsymbol{S}_1 \cdot \boldsymbol{S}_2 + \boldsymbol{S}_2 \cdot \boldsymbol{S}_3 + \boldsymbol{S}_3 \cdot \boldsymbol{S}_1)$

Putting $\boldsymbol{S} = \boldsymbol{S}_1 + \boldsymbol{S}_2 + \boldsymbol{S}_3$ we find that $\boldsymbol{S}^2 = \boldsymbol{S}_1^2 + \boldsymbol{S}_2^2 + \boldsymbol{S}_3^2 + 2(\boldsymbol{S}_1 \cdot \boldsymbol{S}_2 + \boldsymbol{S}_2 \cdot \boldsymbol{S}_3 + \boldsymbol{S}_3 \cdot \boldsymbol{S}_1)$

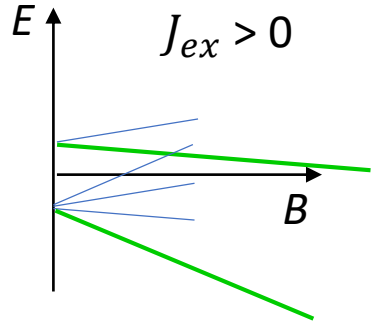
Then $H = -J_{ex}(\boldsymbol{S}^2 - \boldsymbol{S}_1^2 - \boldsymbol{S}_2^2 - \boldsymbol{S}_3^2)$ with $\boldsymbol{S}^2 = S(S+1)$

We also note that we can define $\boldsymbol{S}_{1,2} = \boldsymbol{S}_1 + \boldsymbol{S}_2$ running from $|S_1 - S_2|$ to $S_1 + S_2$ and rewrite $\boldsymbol{S} = \boldsymbol{S}_1 + \boldsymbol{S}_2 + \boldsymbol{S}_3 = \boldsymbol{S}_{1,2} + \boldsymbol{S}_3$

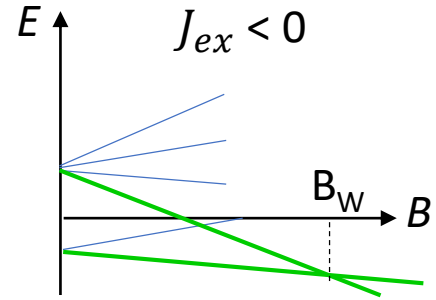
With this in mind we have that:

$S_1 = S_2 = S_3 = 1/2$ implying $\boldsymbol{S}_i^2 = 3/4$ and $S = 3/2, 1/2$ i.e. $\boldsymbol{S}^2 = \frac{15}{4}, \frac{3}{4}$. Then $E = -\frac{3}{2}J_{ex}, \frac{3}{2}J_{ex}$

$S_1 = S_2 = S_3 = 1$ implying $\boldsymbol{S}_i^2 = 2$ and $S = 3, 2, 1, 0$ i.e. $\boldsymbol{S}^2 = 12, 6, 2, 0$. Then $E = -6J_{ex}, 0, 4J_{ex}, 6J_{ex}$



1) Three spins S_i = ½ produce the required binary system, while 4 states are found with S_i = 1. Cu is the correct choice
2) The spin value in the ground state is S = 3/2 for $J_{ex} > 0$, while it is S = 1/2 for $J_{ex} < 0$
3) The bit value is defined by the high/low spin molecular state. The choice for "0" and "1" is arbitrary.
4) The energy splitting in an external magnetic field is sketched on the side for the two cases. At T = 0 only the lowest state of each multiplet is occupied. We see that only when the ground multiplet has S = ½ a crossing between the lowest states of the two multiplets is possible, meaning that we need $J_{ex} < 0$.
5) The lowest states of the two multiplets cross when $3J_{ex} = \mu_B g_e \left(\frac{3}{2} - \frac{1}{2}\right) B_W = 2\mu_B B_W$ i.e $B_W \approx 2.6\ T$



3.3 Spin structure in a trimer

The relative orientation of the spins $\boldsymbol{S}_i$ of the atoms in a trimer depends on the interactions at play.

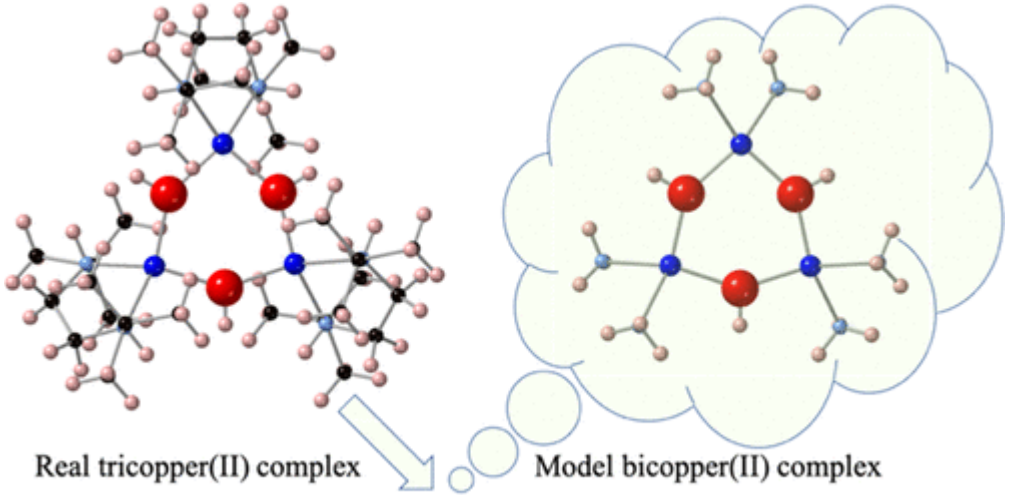
Consider 3 identical spins $\boldsymbol{S}_i$=1/2, forced to stay in the molecule plain, and the spin alignments sketched below. Which is the ground state in case of:

1) a FM Heisenberg like exchange ($-2J_{ex}\ \boldsymbol{S}_i \cdot \boldsymbol{S}_j$) interaction and $\boldsymbol{D}$ = 0

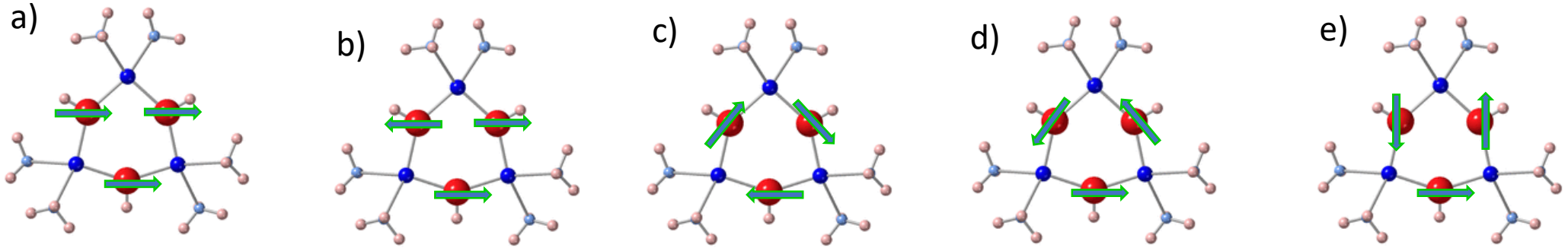
2) a DMI interaction ($\boldsymbol{D} \cdot (\boldsymbol{S}_i \wedge \boldsymbol{S}_j)$) with the $\boldsymbol{D}$ vector entering the sheet and J_{ex} = 0

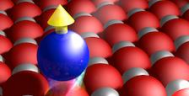
3) both *D* and J_{ex} are not zero.

Adapted from: *Inorg. Chem.* 2022, 61, 31, 12138–12148



J and ***D*** effective coupling





3.3 Spin structure in a trimer - Solution

1) The evaluation of $-2J_{ex}\boldsymbol{S}\cdot\boldsymbol{S}$ in the five cases gives:

a) $-6J_{ex}S^2$
b) $2J_{ex}S^2$
c) $3J_{ex}S^2$
d) $3J_{ex}S^2$
e) $2J_{ex}S^2$

Then a) is the ground state

2) The evaluation of $\boldsymbol{D}\cdot(\boldsymbol{S}\wedge\boldsymbol{S})$ in the five cases gives:

a) 0
b) 0
c) $\frac{3\sqrt{3}}{2}DS^2$
d) $-\frac{3\sqrt{3}}{2}DS^2$
e) $-2DS^2$

Then d) is the ground state

3) The energy in the five cases is:

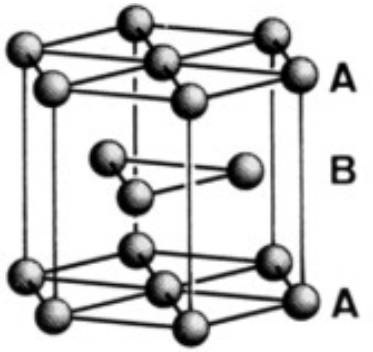
a) $-6J_{ex}S^2 + 0$
b) $2J_{ex}S^2 + 0$
c) $3J_{ex}S^2 + \frac{3\sqrt{3}}{2}DS^2$
d) $3J_{ex}S^2 - \frac{3\sqrt{3}}{2}DS^2$
e) $2J_{ex}S^2 - 2DS^2$

Comparing the energy of case a) and d) we find that for:

$-6J_{ex}S^2 < 3J_{ex}S^2 - 3/2\mathrm{D}\sqrt{3}S^2 \Rightarrow$

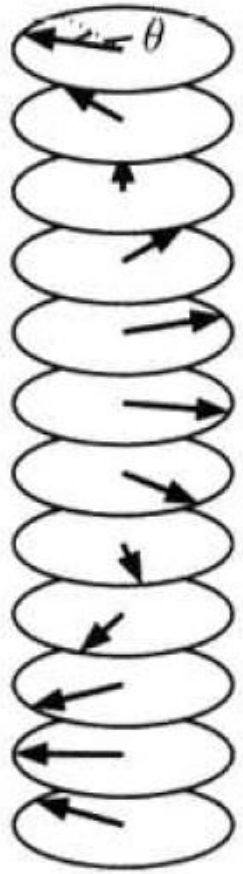
a) is the ground state for $J_{ex} > \frac{1}{2\sqrt{3}}D$; d) is the ground state for $J_{ex} < \frac{1}{2\sqrt{3}}D$

3.4 Helical order



Helical magnetic structures in rare-earths can be the basis of all-spin-based technology.
These structures can be formed also in absence of DMI interaction. They are essentially due to the presence of competing Heisenberg exchange interactions (RKKY type) but the structure is not chiral (i.e. there are coexisting domains with the two chirality).

Rare earths possess hexagonal-type structures , i.e. layered crystalline structures composed of stacked planes. For hcp the sequence is ABAB... The magnetic structure is characterized by a parallel alignment of the *N* spins S within each layer, i.e. each plane shows ferromagnetic behavior. A rotation of the magnetization by an angle θ occurs from layer to layer.
The interaction between the layers is described by a nearest-neighbor exchange constant J_1 and a next-nearest-neighbor exchange constant J_2 .



Depending on the values of θ, and of J_1 and J_2, the system can present ferromagnetic order (FM), antiferromagnetic order (AFM), or helical magnetic order (HM). The latter is represented in the figure.

1) Find the expression for the energy of layer 0 exchanged-coupled with layers 1 and 2
2) Find the conditions that minimize the exchange energy
3) Find the condition on $|J_1|$ and $|J_2|$ for helical order
4) Find the expression for the energies E_{FM}, E_{AFM} and E_{HM}
5) Find the sign of J_2 required for helical order

6) Consider now a system in which there is antiferromagnetic coupling between next-nearest neighbors, i.e. $J_2 < 0$, and $|J_2| = J_1\sqrt{3}/6$.
Show that this material is helimagnetic. Find the angle θ of the magnetization between adjacent layers.

From: M. Getzlaff: Fundamentals of Magnetism, Springer editor (2008)

the Hamiltonian leads to an energy of:

$$E = -2NS^2(J_1 \cos\theta + J_2 \cos 2\theta) \tag{5.131}$$

with N being the number of atoms per layer. The energy as a function of the rotation angle θ is minimum at $\partial E/\partial\theta = 0$ which leads to:

$$(J_1 + 4J_2 \cos\theta)\sin\theta = 0 \tag{5.132}$$

This equation can be solved on the one hand by $\sin\theta = 0$, i.e. $\theta = 0$ or $\theta = \pi$. In this situation we have a ferromagnetic or an antiferromagnetic alignment, respectively, between adjacent layers. On the other hand the equation is solved by:

$$\cos\theta = -\frac{J_1}{4J_2} \tag{5.133}$$

which characterizes helical order or helimagnetism.

Let us discuss the behavior if helical arrangement is present. Due to $|\cos\theta| \leq 1$ we can deduce:

$$|J_1| \leq 4|J_2| \tag{5.134}$$

The energies for ferro-, antiferro-, and helimagnetic arrangement amount to (see (5.131)):

$$E_{\mathrm{FM}} = -2NS^2(J_1 + J_2) \tag{5.135}$$

$$E_{\mathrm{AFM}} = -2NS^2(-J_1 + J_2) \tag{5.136}$$

$$E_{\mathrm{HM}} = -2NS^2\left(-\frac{J_1^2}{8J_2} - J_2\right) \tag{5.137}$$

The last equation is obtained using $\cos 2\theta = \cos^2\theta - \sin^2\theta = 2\cos^2\theta - 1$. For an energetic preference of helimagnetism two conditions must be fulfilled: (a) $E_{\mathrm{HM}} < E_{\mathrm{FM}}$ and (b) $E_{\mathrm{HM}} < E_{\mathrm{AFM}}$. From condition (a) we can conclude:

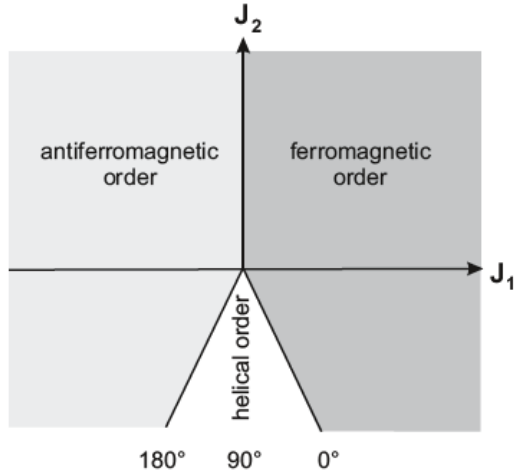


Fig. 5.14. Phase diagram for the model of planes being coupled by J_1 and J_2

$$-\frac{J_1^2}{8J_2} - J_2 > J_1 + J_2 \tag{5.138}$$

which implies:

$$2J_2 + J_1 + \frac{J_1^2}{8J_2} < 0 \tag{5.139}$$

Thus:

$$\frac{1}{8J_2}(16J_2^2 + 8J_1J_2 + J_1^2) < 0 \tag{5.140}$$

which leads to:

$$\frac{1}{8J_2}\left(J_2 + \frac{1}{4}J_1\right)^2 < 0 \tag{5.141}$$

We directly see that J_2 must be negative for the occurrence of helimagnetism. Condition (b) leads to:

$$\frac{1}{8J_2}\left(J_2 - \frac{1}{4}J_1\right)^2 < 0 \tag{5.142}$$

which results in the same conclusion.

Therefore, helical order requires an antiferromagnetic coupling between next nearest layers. The phase diagram shown in Fig. 5.14 summarizes our results.

5.4 Helimagnetism

(a) The prerequisites for helimagnetism (see (5.141) and (5.134)) are: $J_2 < 0$ and $|J_1| \leq 4|J_2|$. The first condition is fulfilled. The second condition can be written as:

$$\left|\frac{J_1}{J_2}\right| \leq 4 \tag{18.36}$$

Inserting results in:

$$\left|\frac{J_1}{J_2}\right| = \frac{6}{\sqrt{3}} = 3.46 \leq 4 \tag{18.37}$$

Thus, this material exhibits helical arrangement.

(b) The angle θ between adjacent layers can be calculated (see (5.133)) by:

$$\cos\theta = -\frac{J_1}{4J_2} = \frac{-6J_1}{-4\sqrt{3}J_1} = 0.866 \tag{18.38}$$

which leads to $\theta = 30°$.