The Exchange Interaction

So far, we considered independent magnetic moments but in reality magnetic moments interact with each other. This interaction can induce magnetic phase transitions and magnetic order at low temperatures.

One possible interaction between magnetic moments is the dipole-dipole interaction, which, however, is much too weak.

An effective interaction between moments, the exchange interaction, is generated by the indistinguishability of particles in quantum mechanics.

Consider two one-particle states $|a\rangle$, $|b\rangle$ with the wavefunctions $\varphi_a(\underline{x})$, $\varphi_b(\underline{x})$:

- each of these states can be occupied by an electron with two spin states.

→ The Fock space with particle numbers 0 to 4 has dimension $2^4 = 16$

The subspace with 2 particles has dimension 6

We expect that it is energetically favorable to fill each of the states $|a\rangle$, $|b\rangle$ with one electron.

→ 4 possibilities

$|a\uparrow, b\uparrow\rangle$, $|a\uparrow, b\downarrow\rangle$, $|a\downarrow, b\uparrow\rangle$, $|a\downarrow, b\downarrow\rangle$

Which of these spin states is favored?

$$\mathcal{H} = \varepsilon_a \left(c^+_{a\uparrow} c_{a\uparrow} + c^+_{a\downarrow} c_{a\downarrow} \right) + \varepsilon_b \left(c^+_{b\uparrow} c_{b\uparrow} + c^+_{b\downarrow} c_{b\downarrow} \right)$$

$$+ U_a \, c^+_{a\uparrow} c_{a\uparrow} \, c^+_{a\downarrow} c_{a\downarrow} + U_b \, c^+_{b\uparrow} c_{b\uparrow} \, c^+_{b\downarrow} c_{b\downarrow}$$

$$+ V \left(c^+_{a\uparrow} c_{a\uparrow} \, c^+_{b\uparrow} c_{b\uparrow} + c^+_{a\downarrow} c_{a\downarrow} \, c^+_{b\downarrow} c_{b\downarrow} \right.$$

$$\left. + c^+_{a\uparrow} c_{a\uparrow} \, c^+_{b\downarrow} c_{b\downarrow} + c^+_{a\downarrow} c_{a\downarrow} \, c^+_{b\uparrow} c_{b\uparrow} \right)$$

$$+ J \left(c^+_{a\uparrow} c^+_{b\uparrow} c_{a\uparrow} c_{b\uparrow} + c^+_{a\uparrow} c^+_{b\downarrow} c_{a\downarrow} c_{b\uparrow} \right.$$

$$+ c^+_{a\downarrow} c^+_{b\uparrow} c_{a\uparrow} c_{b\downarrow} + c^+_{a\downarrow} c^+_{b\downarrow} c_{a\downarrow} c_{b\downarrow}$$

$$\left. + c^+_{a\uparrow} c^+_{a\downarrow} c_{b\downarrow} c_{b\uparrow} + c^+_{b\uparrow} c^+_{b\downarrow} c_{a\downarrow} c_{a\uparrow} \right)$$

U_a, U_b are the intra-orbital Coulomb interactions

V is the direct Coulomb interaction between electrons in different orbitals

J is the exchange interaction, i.e., the Coulomb interaction where the interaction is accompanied with a change $a \to b$ (or vice versa)

The exchange interaction has no classical counterpart. It originates from the indistinguishability of identical particles.

The 2-particle Hilbert space in the problem above is spanned by

$$c^+_{a\uparrow} c^+_{b\uparrow} |0\rangle,\ c^+_{a\downarrow} c^+_{b\downarrow} |0\rangle,\ c^+_{a\uparrow} c^+_{b\downarrow} |0\rangle,\ c^+_{a\downarrow} c^+_{b\uparrow} |0\rangle,$$

$$c^+_{a\uparrow} c^+_{a\downarrow} |0\rangle,\ c^+_{b\uparrow} c^+_{b\downarrow} |0\rangle$$

The first two basis states are eigenstates of the Hamiltonian with eigenenergies $E_{1,2} = \varepsilon_a + \varepsilon_b + V - J$

The remaining 4 2-particle states are not eigenstates. One finds

$$\begin{pmatrix} 2\varepsilon_a + U_a & J & 0 & 0 \\ J & 2\varepsilon_b + U_b & 0 & 0 \\ 0 & 0 & \varepsilon_a + \varepsilon_b + V & J \\ 0 & 0 & J & \varepsilon_a + \varepsilon_b + V \end{pmatrix} \begin{matrix} c^+_{a\uparrow} c^+_{a\downarrow} |0\rangle \\ c^+_{b\uparrow} c^+_{b\downarrow} |0\rangle \end{matrix}$$

which yields the eigenvalues

$$E_{3,4} = \frac{1}{2}\left(2\varepsilon_a + U_a + 2\varepsilon_b + U_b \pm \sqrt{(2\varepsilon_a + U_a - 2\varepsilon_b - U_b)^2 + 4J^2}\right)$$

$$E_{5,6} = \varepsilon_a + \varepsilon_b + V \pm J$$

If $|a\rangle$ and $|b\rangle$ are similar orbitals, we have

$$\varepsilon_a = \varepsilon_b = \varepsilon \quad \text{and} \quad U_a = U_b = U$$

and therefore

$$E_{1,2} = 2\varepsilon + V - J$$

$$E_{3,4} = 2\varepsilon + U \pm J \quad , \quad E_{5,6} = 2\varepsilon + V \pm J$$

In general, $U \gg V \gg |J|$.

$\Rightarrow$ It is energetically not favored to doubly occupy the same orbital

This leaves three states with $E_{1,2,5} = 2\varepsilon + V - J$ and 1 state with $E_6 = 2\varepsilon + V + J$

For $J > 0$, the triplet state is energetically favored over the singlet state and forms the ground state

The three degenerate eigenstates are

$$c^+_{a\downarrow} c^+_{a\downarrow} |0\rangle, \quad c^+_{a\uparrow} c^+_{b\uparrow} |0\rangle, \quad \frac{1}{\sqrt{2}} \left(c^+_{a\uparrow} c^+_{b\downarrow} |0\rangle + c^+_{a\downarrow} c^+_{b\uparrow} |0\rangle \right)$$

$\rightarrow$ spin-symmetric states with total spin $S = 1$

The forth state, energetically $2J$ above the triplet state is

$$\frac{1}{\sqrt{2}} \left(c^+_{a\uparrow} c^+_{b\downarrow} |0\rangle - c^+_{a\downarrow} c^+_{b\uparrow} |0\rangle \right)$$

and has $S = 0$

For degenerate orbitals, the spins will tend to align → compare with 1. Hund's rule.

When $|a\rangle$ and $|b\rangle$ are from different atoms, they will not necessarily be orthogonal but can be used to construct orthogonal states (which we used in the Hamiltonian)

positive J will then again favor the triplet state

This can be captured by an effective spin-spin Hamiltonian:

$$\vec{S}^2 = (\vec{s}_a + \vec{s}_b)^2 = \vec{s}_a^2 + \vec{s}_b^2 + 2\vec{s}_a\vec{s}_b \quad \text{or}$$

$$\vec{s}_a\vec{s}_b = \frac{1}{2}(\vec{S}^2 - \vec{s}_a^2 - \vec{s}_b^2)$$

This leads to the eigenvalue of $\vec{s}_a\vec{s}_b$:

$$\frac{1}{2}[S(S+1) - s_a(s_a+1) - s_b(s_b+1)]$$ with $S=1$ for the triplet and $S=0$ for the singlet state

and $s_a = s_b = \frac{1}{2}$

Therefore, $S=1 \Rightarrow EV = \frac{1}{4}$

$S=0 \Rightarrow EV = -\frac{3}{4}$

The effective Spin-Spin Hamiltonian becomes

$$H_{eff} = 2\varepsilon + V - \frac{J}{2} - 2J\,\vec{S}_a \cdot \vec{S}_b$$

which reproduces $E_{1,2,3} = 2\varepsilon + V - J$ for $S=1$

and $E_4 = 2\varepsilon + V + J$ for $S=0$

The effective exchange interaction between spins is therefore $H_{eff} = -2J\,\vec{S}_a \cdot \vec{S}_b + \text{const}$

which is of the same form as the dipole-dipole interaction, which for atomic moments, however, is of order 10^{-3}-10^{-4} eV. The exchange interaction discussed here is of the order of eV.

Consider now a lattice of localized moments located at the lattice sites.
This leads us to the Hamiltonian

$$\mathcal{H} = -\sum_{i,j} J_{ij}\,\vec{S}_i \cdot \vec{S}_j + g\mu_B \sum_i \vec{B} \cdot \vec{S}_i$$

↑ external magnetic field

This is the Heisenberg model to describe magnetism.

The Heisenberg model is a quantum mechanical model as the spin operators do not commute.

Instead

$$[S_{i\alpha}, S_{j,\beta}] = i \delta_{ij} \varepsilon_{\alpha\beta\gamma} S_{i\gamma}$$

J_{ij} describes the exchange coupling between spin i and spin j

Because of the translational invariance

$$J_{ij} = J(|\vec{R}_i - \vec{R}_j|) = J_{ji}$$

Often, one only considers nearest-neighbor interaction

$$J_{ij} = \begin{cases} J & \text{for } R_i, R_j \text{ nearest neighbors} \\ 0 & \text{otherwise} \end{cases}$$

If $J>0$: leads to parall aligned spins as $\vec{S}_i \cdot \vec{S}_j$ will be positive in this case and the ground state energy becomes smaller (than in the anti-parallel aligned case).

This is also called ferromagnetic coupling.

If $J<0$: anti-alignment is favored.

This is also called anti-ferromagnetic coupling.

For $J<0$ we expect in a nearest-neighbor Heisenberg model a ground state where nearest neighbors are anti-aligned. As a result, a superlattice with inequivalent sublattices A and B forms.
This is not for all lattice types possible.
On a triangular lattice this is e.g. not possible and leads to <u>fustration</u>:

The exchange coupling constant J can also be considered a random variable with given probability distribution.
These systems are called <u>spin glasses</u> and arise e.g. in alloys of magnetic and non-magnetic constituents.
In a <u>classical Heisenberg model</u> spins are taken to be classical variables, i.e. vectors of fixed length.
A generalization of the Heisenberg model is obtained by allowing for anisotropy in spin space.

This leads to

$$H = -\sum_{i,j} J_{ij}\left(\alpha\left(S_i^x S_j^x + S_i^y S_j^y\right) + \beta S_i^z S_j^z\right) + g\mu_B \sum_i B S_i^z$$

where we assumed that $\vec{B}$ points along $\vec{z}$.

For $\alpha=\beta=1$ we recover the Heisenberg model.

For $\alpha=0, \beta=1$ we obtain the <u>Ising model</u>:

$$H = -\sum_{ij} J_{ij} S_i^z S_j^z + g\mu_B B \sum_i S_i^z$$

This is a classical model as it only contains S^z.
In 1 and 2 dimensions, it is exactly solvable.
The two-dimensional solution was found by L. Onsager and shows that the model has a phase transition in 2D.

For $\alpha=1, \beta=0$ we obtain the <u>XY-model</u>

$$\mathcal{H} = -\sum_{ij} J_{ij}\left(S_i^x S_j^x + S_i^y S_j^y\right) + g\mu_B \vec{B} \sum_i \vec{S}_i$$