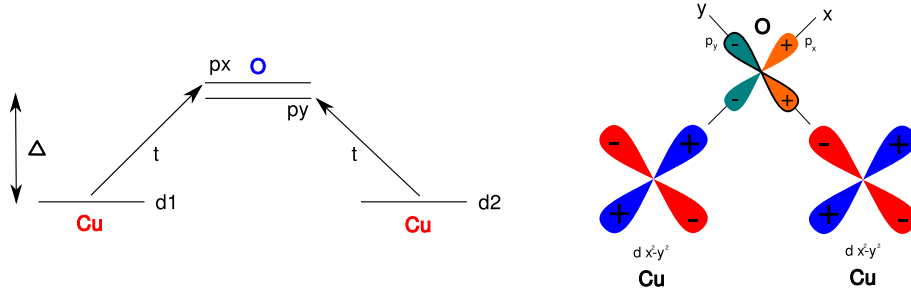


(A.) In this exercise, we propose to describe an exchange mechanism which, unlike the simple two-site computation, leads to a ferromagnetic coupling. We consider a system composed of two atoms of copper and one atom of oxygen.



Each copper brings an electron to the system (total of two electrons in the system). For reasons of symmetry, one of the electrons can hop only between the orbital d_1 and the orbital p_x while the other can hop only between the orbital d_2 and the orbital p_y . The Hamiltonian that describes such a system is given by

$$\mathcal{H} = \mathcal{H}_\Delta + \mathcal{H}_H + \mathcal{H}_t \quad (1)$$

The term $\mathcal{H}_\Delta$ takes into account that the d levels of copper have a lower energy than the p orbitals of oxygen. $\mathcal{H}_\Delta$ is written as:

$$\mathcal{H}_\Delta = \Delta [n_{p_x, \uparrow} + n_{p_x, \downarrow} + n_{p_y, \uparrow} + n_{p_y, \downarrow}] \quad \Delta > 0 \quad (2)$$

The term $\mathcal{H}_H$ is the Hund coupling between two electrons on the orbitals of oxygen. This is a ferromagnetic Heisenberg coupling. In terms of the creation and annihilation operators, it takes the form (for simplicity, $\alpha^\dagger$ means the creation operator for the state α):

$$\mathcal{H}_H = -J_H \left[\frac{1}{4} (n_{p_x, \uparrow} - n_{p_x, \downarrow}) (n_{p_y, \uparrow} - n_{p_y, \downarrow}) + \frac{1}{2} (p_{x, \uparrow}^\dagger p_{x, \downarrow} p_{y, \downarrow}^\dagger p_{y, \uparrow} + p_{x, \downarrow}^\dagger p_{x, \uparrow} p_{y, \uparrow}^\dagger p_{y, \downarrow}) \right] \quad J_H > 0 \quad (3)$$

The kinetic term $\mathcal{H}_t$ is given by:

$$\mathcal{H}_t = -t \sum_{\sigma} p_{x, \sigma}^\dagger d_{1, \sigma} + d_{1, \sigma}^\dagger p_{x, \sigma} + p_{y, \sigma}^\dagger d_{2, \sigma} + d_{2, \sigma}^\dagger p_{y, \sigma} \quad (4)$$

In this problem Δ sets the energy scale and we consider the case $t \ll J_H \ll \Delta$. $\mathcal{H}_t$ will thus be treated as a perturbation of $\mathcal{H}_0 = \mathcal{H}_\Delta + \mathcal{H}_H$. In degenerate perturbation theory, the effective Hamiltonian up to 4th order reads:

$$\begin{aligned} \mathcal{H}_{\text{eff}}^{(4)} = & E_0 P_0 + P_0 V P_0 + P_0 V S V P_0 + P_0 V S V S V P_0 - \frac{1}{2} P_0 V P_0 V S^2 V P_0 - \frac{1}{2} P_0 V S^2 V P_0 V P_0 \\ & + P_0 V S V S V S V P_0 - \frac{1}{2} P_0 V S^2 V P_0 V S V P_0 - \frac{1}{2} P_0 V S V P_0 V S^2 V P_0 + \frac{1}{2} P_0 V P_0 V P_0 V S^3 V P_0 \\ & + \frac{1}{2} P_0 V S^3 V P_0 V P_0 V P_0 - \frac{1}{2} P_0 V P_0 V S^2 V S V P_0 - \frac{1}{2} P_0 V S V S^2 V P_0 V P_0 - \frac{1}{2} P_0 V P_0 V S V S^2 V P_0 \\ & - \frac{1}{2} P_0 V S^2 V S V P_0 V P_0 \end{aligned} \quad (5)$$

where P_0 is the projector on the fundamental subspace, $S = \frac{1-P_0}{E_0-\mathcal{H}_0}$ and $V = \mathcal{H}_t$.

We choose to work in the basis $\mathcal{B} = \{|T_{Cu,Cu}^1\rangle, |T_{Cu,Cu}^0\rangle, |T_{Cu,Cu}^{-1}\rangle, |S_{Cu,Cu}\rangle, |T_{Cu,O}^1\rangle, \dots\}$ defined as follows:

$$\begin{aligned}
 |T_{Cu,Cu}^1\rangle &= d_{1,\uparrow}^\dagger d_{2,\uparrow}^\dagger |0\rangle & |T_{Cu,O}^1\rangle &= d_{1,\uparrow}^\dagger p_{y,\uparrow}^\dagger |0\rangle \\
 |T_{Cu,Cu}^0\rangle &= \frac{1}{\sqrt{2}}(d_{1,\uparrow}^\dagger d_{2,\downarrow}^\dagger + d_{1,\downarrow}^\dagger d_{2,\uparrow}^\dagger) |0\rangle & |T_{Cu,O}^0\rangle &= \frac{1}{\sqrt{2}}(d_{1,\uparrow}^\dagger p_{y,\downarrow}^\dagger + d_{1,\downarrow}^\dagger p_{y,\uparrow}^\dagger) |0\rangle \\
 |T_{Cu,Cu}^{-1}\rangle &= d_{1,\downarrow}^\dagger d_{2,\downarrow}^\dagger |0\rangle & |T_{Cu,O}^{-1}\rangle &= d_{1,\downarrow}^\dagger p_{y,\downarrow}^\dagger |0\rangle \\
 |S_{Cu,Cu}\rangle &= \frac{1}{\sqrt{2}}(d_{1,\uparrow}^\dagger d_{2,\downarrow}^\dagger - d_{1,\downarrow}^\dagger d_{2,\uparrow}^\dagger) |0\rangle & |S_{Cu,O}\rangle &= \frac{1}{\sqrt{2}}(d_{1,\uparrow}^\dagger p_{y,\downarrow}^\dagger - d_{1,\downarrow}^\dagger p_{y,\uparrow}^\dagger) |0\rangle
 \end{aligned} \tag{6}$$

$$\begin{aligned}
 |T_{O,Cu}^1\rangle &= p_{x,\uparrow}^\dagger d_{2,\uparrow}^\dagger |0\rangle & |T_{O,O}^1\rangle &= p_{x,\uparrow}^\dagger p_{y,\uparrow}^\dagger |0\rangle \\
 |T_{O,Cu}^0\rangle &= \frac{1}{\sqrt{2}}(p_{x,\uparrow}^\dagger d_{2,\downarrow}^\dagger + p_{x,\downarrow}^\dagger d_{2,\uparrow}^\dagger) |0\rangle & |T_{O,O}^0\rangle &= \frac{1}{\sqrt{2}}(p_{x,\uparrow}^\dagger p_{y,\downarrow}^\dagger + p_{x,\downarrow}^\dagger p_{y,\uparrow}^\dagger) |0\rangle \\
 |T_{O,Cu}^{-1}\rangle &= p_{x,\downarrow}^\dagger d_{2,\downarrow}^\dagger |0\rangle & |T_{O,O}^{-1}\rangle &= p_{x,\downarrow}^\dagger p_{y,\downarrow}^\dagger |0\rangle \\
 |S_{O,Cu}\rangle &= \frac{1}{\sqrt{2}}(p_{x,\uparrow}^\dagger d_{2,\downarrow}^\dagger - p_{x,\downarrow}^\dagger d_{2,\uparrow}^\dagger) |0\rangle & |S_{O,O}\rangle &= \frac{1}{\sqrt{2}}(p_{x,\uparrow}^\dagger p_{y,\downarrow}^\dagger - p_{x,\downarrow}^\dagger p_{y,\uparrow}^\dagger) |0\rangle
 \end{aligned}$$

1. Show that the states $|S_{\alpha,\beta}\rangle$ $\alpha, \beta \in \{Cu, O\}$ are singlets. This amounts to showing that $(S_\alpha^- + S_\beta^-)|S_{\alpha,\beta}\rangle = 0$ and that $(S_\alpha^z + S_\beta^z)|S_{\alpha,\beta}\rangle = 0$.
2. Show that the states $|T_{\alpha,\beta}^\gamma\rangle$ $\alpha, \beta \in \{Cu, O\}$, $\gamma \in \{1, 0, -1\}$ are triplets. This amounts to showing that:

$$\begin{aligned}
 (S_\alpha^z + S_\beta^z)|T_{\alpha,\beta}^1\rangle &= |T_{\alpha,\beta}^1\rangle \\
 (S_\alpha^z + S_\beta^z)|T_{\alpha,\beta}^{-1}\rangle &= -1|T_{\alpha,\beta}^{-1}\rangle \\
 (S_\alpha^z + S_\beta^z)|T_{\alpha,\beta}^0\rangle &= 0 \quad \text{and} \quad \langle S_{\alpha,\beta}|T_{\alpha,\beta}^0\rangle = 0.
 \end{aligned} \tag{7}$$

3. Show that in the $\mathcal{B}$ basis, the Hamiltonian $\mathcal{H}_0$ is diagonal. What are the eigenenergies and eigenvectors?
4. Convince yourself that:

$$\begin{aligned}
 V|T_{Cu,Cu}^\alpha\rangle &= -t(|T_{O,Cu}^\alpha\rangle + |T_{Cu,O}^\alpha\rangle) & \text{and that} & & V|S_{Cu,Cu}\rangle &= -t(|S_{O,Cu}\rangle + |S_{Cu,O}\rangle) \\
 V|T_{Cu,O}^\alpha\rangle &= -t(|T_{O,O}^\alpha\rangle + |T_{Cu,Cu}^\alpha\rangle) & & & V|S_{Cu,O}\rangle &= -t(|S_{O,O}\rangle + |S_{Cu,Cu}\rangle) \\
 V|T_{O,Cu}^\alpha\rangle &= -t(|T_{O,O}^\alpha\rangle + |T_{Cu,Cu}^\alpha\rangle) & & & V|S_{O,Cu}\rangle &= -t(|S_{O,O}\rangle + |S_{Cu,Cu}\rangle) \\
 V|T_{O,O}^\alpha\rangle &= -t(|T_{Cu,O}^\alpha\rangle + |T_{O,Cu}^\alpha\rangle) & & & V|S_{O,O}\rangle &= -t(|S_{Cu,O}\rangle + |S_{O,Cu}\rangle)
 \end{aligned} \tag{8}$$

5. In the previous question it has been shown that the perturbation does not couple the ground subspace to itself. Show that $P_0 V P_0 = 0$ and that the terms of order 3 in V in (5) vanish. The effective Hamiltonian is reduced to:

$$\mathcal{H}_{\text{eff}}^{(4)} = E_0 P_0 + P_0 V S V P_0 + P_0 V S V S V S V P_0 - \frac{1}{2}(P_0 V S^2 V P_0 V S V P_0 + P_0 V S V P_0 V S^2 V P_0) \tag{9}$$

6. Calculate $\mathcal{H}_{\text{eff}}^{(2)} = P_0 V S V P_0$
7. Calculate $P_0 V S^2 V P_0 V S V P_0$ and by symmetry $P_0 V S V P_0 V S^2 V P_0$
8. Calculate $P_0 V S V S V S$
9. Deduce $\mathcal{H}_{\text{eff}}^{(4)}$. What are the spin configurations that are favored?