Double Exchange *II*: *Anderson-Hasegawa Model*

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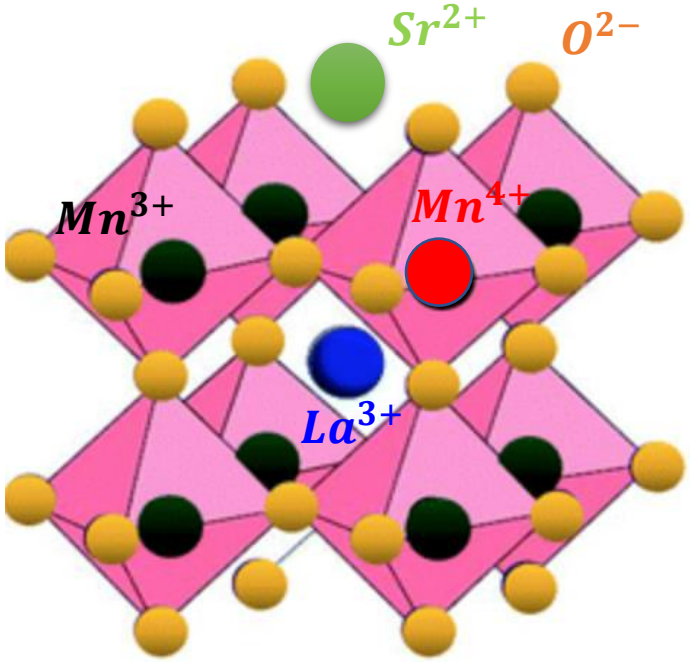
05/09/2020

W. Anderson and H. Hasegawa, Phys. Rev. 100, 675 (1955)

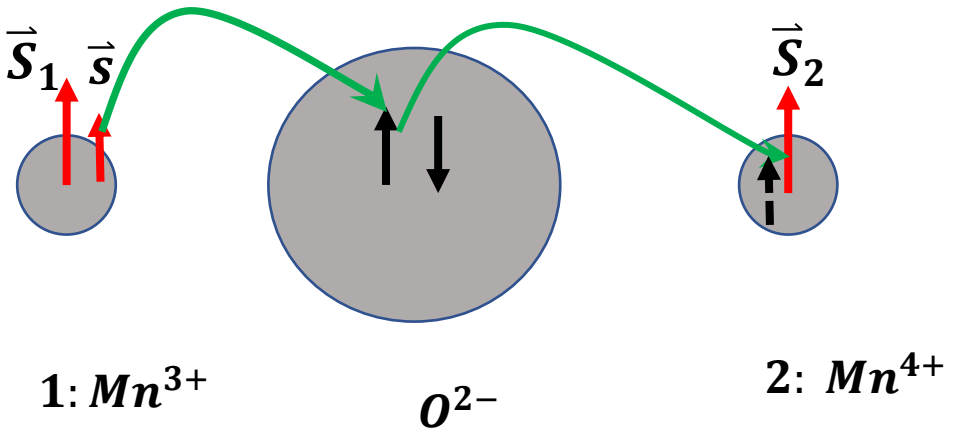
Retrospect: Indirect exchange via conduction electrons



$(La^{3+}_{1-x}Sr^{2+}_{x})(Mn^{3+}_{1-x}Mn^{4+}_{x})O_3$

$|R_1\rangle$: $\boldsymbol{Mn^{3+}O^{2-}Mn^{4+}}$

$|R_2\rangle$: $\boldsymbol{Mn^{4+}O^{2-}Mn^{3+}}$



$$\vec{S}_1 // \vec{s} // \vec{S}_2$$

$$\Delta H = \begin{bmatrix} -JS_1 & b \\ b & -JS_2 \end{bmatrix}$$

Eigenvalues:

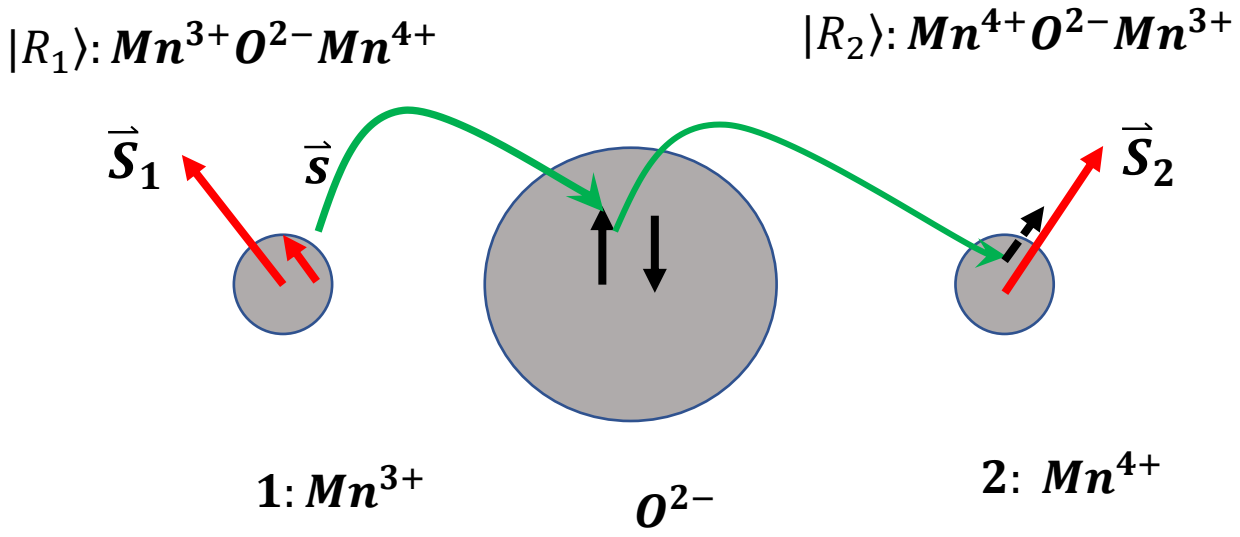
$$\Delta E = -JS \pm b$$

Eigenvectors:

$$\varphi_{\pm} = \frac{1}{\sqrt{2}}[|R_1\rangle \pm |R_2\rangle]$$

C. Zener, Phys.Rev. 82, 403 (1951)
P. W. Anderson and H. Hasegawa, Phys. Rev. 100, 675 (1955)

Double Exchange: Semi-classical model in two-ion case



$$\langle \vec{S}_1, \vec{S}_2 \rangle = \theta$$

$$\Delta H: \begin{array}{c|cc} & |R_1\rangle & |R_2\rangle \\ \hline |R_1\rangle & -2J\vec{S}_1\cdot\vec{s} & \hat{b} \\ |R_2\rangle & \hat{b}^\dagger & -2J\vec{S}_2\cdot\vec{s} \end{array}$$

***Symmetry* 1 ↔ 2:** $\hat{b} = \hat{b}^\dagger = \langle R_1|\Delta H|R_2\rangle$

State space:

$$\{|R_1\rangle, |R_2\rangle\} \otimes \{|S, S_z, s, s_z\rangle\}$$

Mutually Commuting Observables:

$$\{\hat{S}_i^2, \hat{S}_{iz}^2, \hat{s}^2, \hat{s}_z\}, \; i = 1, 2.$$

Uncoupled Basis: $\hbar \equiv 1$

$$\hat{s}^2|S, S_{iz}, s, s_z\rangle = \frac{1}{2}\left(\frac{1}{2}+1\right)|S, S_{iz}, s, s_z\rangle$$

$$\hat{S}_1^2\,|S, S_{1z}, s, s_z\rangle = S(S+1)|S, S_{1z}, s, s_z\rangle$$

$$\hat{S}_2^2\,|S, S_{2z}, s, s_z\rangle = S(S+1)|S, S_{2z}, s, s_z\rangle$$

Double Exchange: Semi-classical model in two-ion case

Step 1: Diagonalize diagonal blocks in spin subspace

$$\Delta H: \begin{array}{c|cc} & |R_1\rangle & |R_2\rangle \\ \hline |R_1\rangle & \boxed{-2J\vec{S}_1\cdot\vec{s}} & \hat{b} \\ |R_2\rangle & \hat{b}^\dagger & \boxed{-2J\vec{S}_2\cdot\vec{s}} \end{array}$$

$$\vec{S}'_i = \vec{S}_i + \vec{s}, \quad S'_i = S \pm \frac{1}{2}$$

In coupled basis: $|S'_i, S'_{iz}, s, S\rangle$

$$-2J\vec{S}_1\cdot\vec{s}: \begin{array}{c|cc} & |R_1\rangle|S+\frac{1}{2}, S'_{1z}\rangle & |R_1\rangle|S-\frac{1}{2}, S'_{1z}\rangle \\ \hline |R_1\rangle|S+\frac{1}{2}, S'_{1z}\rangle & -2JS & 0 \\ |R_1\rangle|S-\frac{1}{2}, S'_{1z}\rangle & 0 & J(S+1) \end{array}$$

$$-2J\vec{S}_2\cdot\vec{s}: \begin{array}{c|cc} & |R_2\rangle|S+\frac{1}{2}, S'_{2z}\rangle & |R_2\rangle|S-\frac{1}{2}, S'_{2z}\rangle \\ \hline |R_2\rangle|S+\frac{1}{2}, S'_{2z}\rangle & -2JS & 0 \\ |R_2\rangle|S-\frac{1}{2}, S'_{2z}\rangle & 0 & J(S+1) \end{array}$$

Mutually Commuting Observables:

$$\{\hat{S}'^2_i, \hat{S}'_z, \hat{S}^2_i, \hat{s}^2\}$$

$$\hat{S}'^2_i|S'_i, S'_{iz}\rangle = S'_i(S'_i+1)|S'_i, S'_{iz}\rangle$$

$$\hat{S}^2_i|S'_i, S'_{iz}\rangle = S(S+1)|S'_i, S'_{iz}\rangle$$

$$\hat{s}^2|S'_i, S'_{iz}\rangle = \frac{1}{2}\left(\frac{1}{2}+1\right)|S'_i, S'_{iz}\rangle$$

$$|S'_i, S'_{iz}\rangle \equiv |S'_i, S'_{iz}, s=\tfrac{1}{2}, S\rangle; \quad i = 1, 2$$

$$2\vec{S}_i\cdot\vec{s} = S'^2_i - s^2 - S^2_i = \begin{cases} S, & S'_i = S + \frac{1}{2} \\ -(S+1), & S'_i = S - \frac{1}{2} \end{cases}$$

$$\vec{S}_i\cdot\vec{s}|S'_i, S'_{iz}, s, S\rangle = \frac{S'^2_i - s^2 - S^2_i}{2}|S'_i, S'_{iz}\rangle = \frac{\left(S\pm\frac{1}{2}\right)\left(S\pm\frac{1}{2}+1\right) - \frac{1}{2}\left(\frac{1}{2}+1\right) - S(S+1)}{2}|S'_i, S'_{iz}\rangle$$

$$= \frac{\left(S\pm\frac{1}{2}\right)\left(S\pm\frac{1}{2}+1\right) - \frac{1}{2}\left(\frac{1}{2}+1\right) - S(S+1)}{2}|S'_i, S'_{iz}\rangle = \frac{S}{2}\ \mathbf{or}\ \frac{-(S+1)}{2}|S'_i, S'_{iz}\rangle$$

Double Exchange: Semi-classical model in two-ion case

Step 2: Get off-diagonal blocks in spin subspace

In coupled basis: $\left| S'_i, S'_{iz}, s = \frac{1}{2}, S \right\rangle$

$$\Delta H: \begin{array}{c|cc} & |R_1\rangle & |R_2\rangle \\ \hline |R_1\rangle & -2J\vec{S}_1\cdot\vec{s} & \hat{b} \\ |R_2\rangle & \hat{b}^\dagger & -2J\vec{S}_2\cdot\vec{s} \end{array}$$

$$\begin{array}{c|cccc} & |R_1\rangle|S+\frac{1}{2},S'_{1z}\rangle & |R_1\rangle|S-\frac{1}{2},S'_{1z}\rangle & |R_2\rangle|S+\frac{1}{2},S'_{2z}\rangle & |R_2\rangle|S-\frac{1}{2},S'_{2z}\rangle \\ \hline |R_1\rangle|S+\frac{1}{2},S'_{1z}\rangle & -2JS & 0 & B & \\ |R_1\rangle|S-\frac{1}{2},S'_{1z}\rangle & 0 & J(S+1) & & \\ |R_2\rangle|S+\frac{1}{2},S'_{2z}\rangle & & & -2JS & 0 \\ |R_2\rangle|S-\frac{1}{2},S'_{2z}\rangle & B^\dagger & & 0 & J(S+1) \end{array}$$

Assumptions:

$$b \equiv \left\langle S \pm \frac{1}{2}, S'_{1z} \middle| \hat{b} \middle| S \pm \frac{1}{2}, S'_{1z} \right\rangle$$

$$B_{jk} \equiv \left\langle j, S'_{1z} \middle| \hat{b} \middle| k, S'_{2z} \right\rangle;\ j,k = S + \frac{1}{2}, S - \frac{1}{2}$$

$$0 \equiv \left\langle S \pm \frac{1}{2}, S'_{1z} \middle| \hat{b} \middle| S \mp \frac{1}{2}, S'_{1z} \right\rangle$$

$$|k, S'_{2z}\rangle \xRightarrow{c_{jk} \equiv \langle j, S'_{1z} | k, S'_{2z} \rangle} |j, S'_{1z}\rangle \quad ????$$

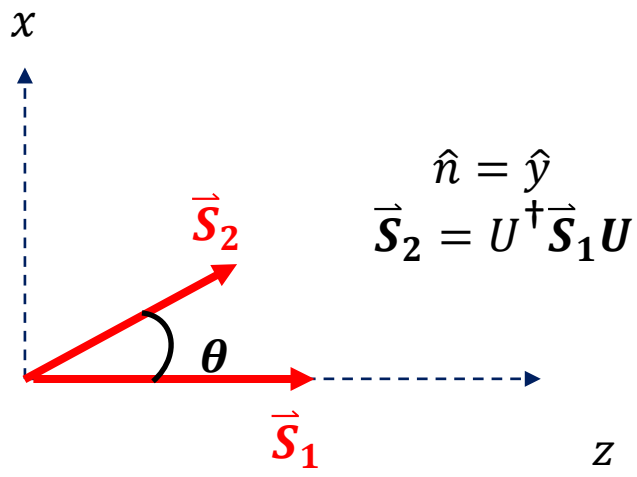
We can solve c_{jk} both **classically** and *quantum mechanically*

Double Exchange: Semi-classical model in two-ion case

Step 2: Get off-diagonal blocks in spin subspace

If $\langle \vec{S}_1, \vec{S}_2 \rangle = \theta$

Classical rotation as unitary transformation in Hilbert space:



$$\hat{n} = \hat{y}$$
$$\vec{S}_2 = U^{\dagger} \vec{S}_1 U$$

$$U = e^{i\frac{\theta}{2}\hat{n}\cdot\vec{\sigma}} = \cos\frac{\theta}{2} + i\hat{n}\cdot\vec{\sigma}\sin\frac{\theta}{2}$$

In 2-D Hilbert space, σ_z representation

$$\sigma_x = \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}, \sigma_y = \begin{bmatrix} 0 & -i \\ i & 0 \end{bmatrix}, \sigma_z = \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix}$$

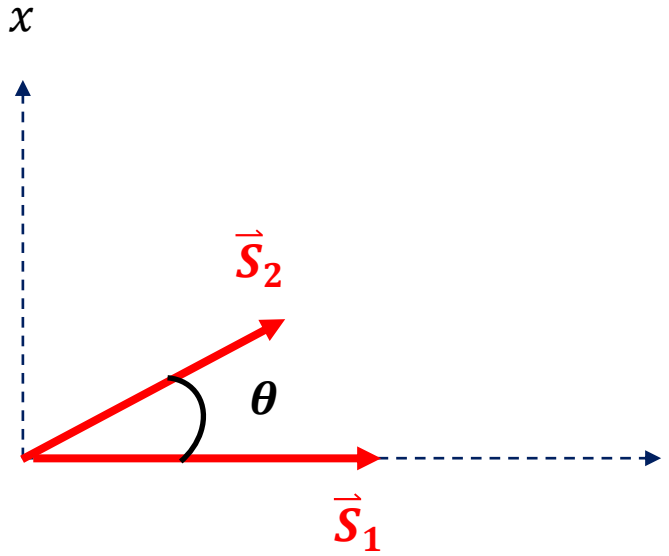
$$\begin{pmatrix} \left|S + \frac{1}{2}, S'_{2z}\right\rangle \\ \left|S - \frac{1}{2}, S'_{2z}\right\rangle \end{pmatrix} = U^{\dagger} \begin{pmatrix} \left|S + \frac{1}{2}, S'_{1z}\right\rangle \\ \left|S - \frac{1}{2}, S'_{1z}\right\rangle \end{pmatrix}$$

$$U = e^{i\frac{\theta}{2}\hat{n}\cdot\vec{\sigma}} = \cos\frac{\theta}{2} + i\,\sigma_y \sin\frac{\theta}{2} = \begin{bmatrix} \cos\frac{\theta}{2} & \sin\frac{\theta}{2} \\ -\sin\frac{\theta}{2} & \cos\frac{\theta}{2} \end{bmatrix} = \begin{pmatrix} \cos\frac{\theta}{2}\left|S + \frac{1}{2}, S'_{1z}\right\rangle - \sin\frac{\theta}{2}\left|S - \frac{1}{2}, S'_{1z}\right\rangle \\ \sin\frac{\theta}{2}\left|S + \frac{1}{2}, S'_{1z}\right\rangle + \cos\frac{\theta}{2}\left|S - \frac{1}{2}, S'_{1z}\right\rangle \end{pmatrix}$$

Double Exchange: Semi-classical model in two-ion case

Step 2: Get off-diagonal blocks in spin subspace

If $\langle \vec{S}_1, \vec{S}_2 \rangle = \theta \neq 0$,



$$\begin{pmatrix} \left|S+\frac{1}{2}, S'_{2z}\right\rangle \\ \left|S-\frac{1}{2}, S'_{2z}\right\rangle \end{pmatrix} = \begin{pmatrix} \cos\frac{\theta}{2}\left|S+\frac{1}{2}, S'_{1z}\right\rangle - \sin\frac{\theta}{2}\left|S-\frac{1}{2}, S'_{1z}\right\rangle \\ \sin\frac{\theta}{2}\left|S+\frac{1}{2}, S'_{1z}\right\rangle + \cos\frac{\theta}{2}\left|S-\frac{1}{2}, S'_{1z}\right\rangle \end{pmatrix}$$

$$B_{jk} \equiv \langle \boldsymbol{j}, \boldsymbol{S'_{1z}} | \hat{b} | \boldsymbol{k}, \boldsymbol{S'_{2z}} \rangle;\ j,k = \boldsymbol{S}+\frac{1}{2}, \boldsymbol{S}-\frac{1}{2}$$

$$B = \begin{bmatrix} b\cos\frac{\theta}{2} & b\sin\frac{\theta}{2} \\ -b\sin\frac{\theta}{2} & b\cos\frac{\theta}{2} \end{bmatrix}$$

$$b \equiv \left\langle S \pm \frac{1}{2}, S'_{1z} \middle| \hat{b} \middle| S \pm \frac{1}{2}, S'_{1z} \right\rangle$$

$$\Delta H: \begin{array}{c} \\ |R_1\rangle|S+\frac{1}{2}, S'_{1z}\rangle \\ |R_1\rangle|S-\frac{1}{2}, S'_{1z}\rangle \\ |R_2\rangle|S+\frac{1}{2}, S'_{2z}\rangle \\ |R_2\rangle|S-\frac{1}{2}, S'_{2z}\rangle \end{array} \begin{array}{c} \begin{array}{cccc} |R_1\rangle|S+\frac{1}{2}, S'_{1z}\rangle & |R_1\rangle|S-\frac{1}{2}, S'_{1z}\rangle & |R_2\rangle|S+\frac{1}{2}, S'_{2z}\rangle & |R_2\rangle|S-\frac{1}{2}, S'_{2z}\rangle \end{array} \\ \begin{bmatrix} -2JS & 0 & b\cos\frac{\theta}{2} & b\sin\frac{\theta}{2} \\ 0 & J(S+1) & -b\sin\frac{\theta}{2} & b\cos\frac{\theta}{2} \\ b\cos\frac{\theta}{2} & -b\sin\frac{\theta}{2} & -2JS & 0 \\ b\sin\frac{\theta}{2} & b\cos\frac{\theta}{2} & 0 & J(S+1) \end{bmatrix} \end{array}$$

Double Exchange: Semi-classical model in two-ion case

Step 3: Diagonalize the whole ΔH

$$\Delta H: \quad \begin{array}{c|cccc} & |R_1\rangle|S+\frac{1}{2},S'_{1z}\rangle & |R_1\rangle|S-\frac{1}{2},S'_{1z}\rangle & |R_2\rangle|S+\frac{1}{2},S'_{2z}\rangle & |R_2\rangle|S-\frac{1}{2},S'_{2z}\rangle \\ \hline |R_1\rangle|S+\frac{1}{2},S'_{1z}\rangle & -2JS & 0 & b\cos\frac{\theta}{2} & -b\sin\frac{\theta}{2} \\ |R_1\rangle|S-\frac{1}{2},S'_{1z}\rangle & 0 & J(S+1) & b\sin\frac{\theta}{2} & b\cos\frac{\theta}{2} \\ |R_2\rangle|S+\frac{1}{2},S'_{2z}\rangle & b\cos\frac{\theta}{2} & b\sin\frac{\theta}{2} & -2JS & 0 \\ |R_2\rangle|S-\frac{1}{2},S'_{2z}\rangle & -b\sin\frac{\theta}{2} & b\cos\frac{\theta}{2} & 0 & J(S+1) \end{array}$$

$$\|\Delta \boldsymbol{H} - \Delta \boldsymbol{E}\| = \mathbf{0}$$

$$\Delta \boldsymbol{E} = \frac{\mathbf{1}}{\mathbf{2}}\boldsymbol{J} \pm \sqrt{\boldsymbol{J}^{\mathbf{2}}\left(\boldsymbol{S}+\frac{\mathbf{1}}{\mathbf{2}}\right)^{\mathbf{2}} + \boldsymbol{b}^{\mathbf{2}} \pm \mathbf{2}\boldsymbol{Jb}\left(\boldsymbol{S}+\frac{\mathbf{1}}{\mathbf{2}}\right)\boldsymbol{\cos}\frac{\boldsymbol{\theta}}{\mathbf{2}}}$$

If $kT_c \ll |\boldsymbol{J}|, |\boldsymbol{b}|,$

$$\Delta \boldsymbol{E} = \frac{\mathbf{1}}{\mathbf{2}}\boldsymbol{J} - \sqrt{\boldsymbol{J}^{\mathbf{2}}\left(\boldsymbol{S}+\frac{\mathbf{1}}{\mathbf{2}}\right)^{\mathbf{2}} + \boldsymbol{b}^{\mathbf{2}} \pm \mathbf{2}\boldsymbol{Jb}\left(\boldsymbol{S}+\frac{\mathbf{1}}{\mathbf{2}}\right)\boldsymbol{\cos}\frac{\boldsymbol{\theta}}{\mathbf{2}}}$$

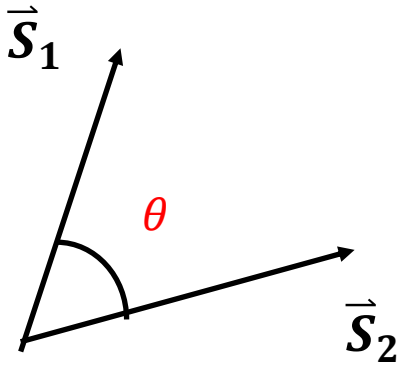
Double Exchange: Semi-classical model in two-ion case

Discussions: (1) $J \gg b$

$$\Delta E = \frac{1}{2}J - \sqrt{J^2\left(S+\frac{1}{2}\right)^2 + b^2 \pm 2Jb\left(S+\frac{1}{2}\right)\cos\frac{\theta}{2}}$$

$$\cong \frac{1}{2}J - \sqrt{J^2\left(S+\frac{1}{2}\right)^2 + b^2\cos^2\frac{\theta}{2} \pm 2Jb\left(S+\frac{1}{2}\right)\cos\frac{\theta}{2}}$$

$$= \frac{1}{2}J - \left[|J|\left(S+\frac{1}{2}\right) \pm |b|\cos\frac{\theta}{2}\right] = \frac{1}{2}(J-|J|) - |J|S \pm |b|\cos\frac{\theta}{2}$$



$$\varphi_\pm \cong \frac{1}{\sqrt{2}}\left[|R_1\rangle\left|S+\frac{1}{2}, S'_{1z}\right\rangle \pm |R_2\rangle\left|S+\frac{1}{2}, S'_{2z}\right\rangle\right]$$

$$kT_c \ll b,\ \Delta E = Const. + \Delta E(\vec{s}, \vec{S}_i) + \Delta E(\vec{S}_1, \vec{S}_2)$$

$$\Delta E(\vec{s}, \vec{S}_i) \equiv -|J|S \qquad \Delta E(\vec{S}_1, \vec{S}_2) \equiv -|b|\cos\frac{\theta}{2}$$

➡ **Ferromagnetic**

Direction exchange:

$$\Delta E_d(\vec{S}_1, \vec{S}_2) = -J_d \vec{S}_1 \cdot \vec{S}_2, \quad J_d > 0$$

Superexchange:

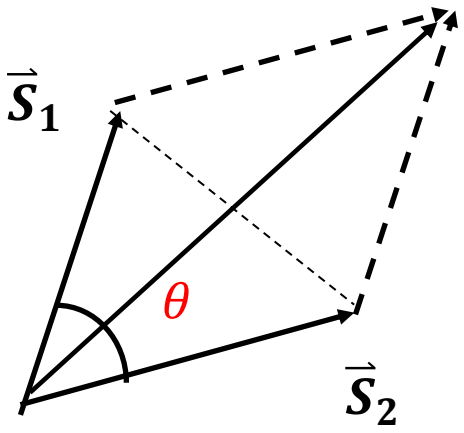
$$\Delta E_s(\vec{S}_1, \vec{S}_2) = J_s \vec{S}_1 \cdot \vec{S}_2, \qquad J_s > 0$$

$$\Delta E_d(\vec{S}_1, \vec{S}_2), \Delta E_s(\vec{S}_1, \vec{S}_2) \propto \cos\theta,$$

Double Exchange: Semi-classical model in two-ion case

Discussions: (1) $J \ll b$

$$\vec{S}_t = \vec{S}_1 + \vec{S}_2$$



$$\cos\frac{\theta}{2} = \frac{S_t}{2S}$$

$$\Delta E = \frac{1}{2}J - \sqrt{J^2\left(S+\frac{1}{2}\right)^2 + b^2 \pm 2Jb\left(S+\frac{1}{2}\right)\cos\frac{\theta}{2}}$$

$$\cong \frac{1}{2}J - \sqrt{J^2\left(S+\frac{1}{2}\right)^2\cos^2\frac{\theta}{2} + b^2 \pm 2Jb\left(S+\frac{1}{2}\right)\cos\frac{\theta}{2}}$$

$$= \frac{1}{2}J - \left[|b| \pm |J|\left(S+\frac{1}{2}\right)\cos\frac{\theta}{2}\right] = \frac{1}{2}J - |b| + \Delta E(\vec{s},\vec{S}_t)$$

$$\Delta E(\vec{s},\vec{S}_t) = \pm|J|\left(S+\frac{1}{2}\right)\cos\frac{\theta}{2}$$

$$= \pm|J|\left(S+\frac{1}{2}\right)\frac{S_t}{2S} \approx \pm|J|S_t \propto |J|\vec{S}_t\cdot\vec{s} = |J|(\vec{S}_1\cdot\vec{s} + \vec{S}_2\cdot\vec{s})$$

➡ **Conduction electron polarization in ferromagnetic metal**

Supplementary: Pure quantum method

$$\Delta H: \begin{array}{c|cc} & |R_1\rangle & |R_2\rangle \\ \hline |R_1\rangle & -2J\vec{\boldsymbol{S}}_{\boldsymbol{1}}\cdot\vec{\boldsymbol{s}} & \hat{b} \\ |R_2\rangle & \hat{b}^{\dagger} & -2J\vec{\boldsymbol{S}}_{\boldsymbol{2}}\cdot\vec{\boldsymbol{s}} \end{array}$$

$$B_{\boldsymbol{S}_1'\boldsymbol{S}_2'} \equiv \left\langle \boldsymbol{S}_0^2, \boldsymbol{S}_{0z}, \boldsymbol{S}_2'(\boldsymbol{S}_2, \boldsymbol{s}), \boldsymbol{S}_1 \middle| \hat{b} \middle| \boldsymbol{S}_0^2, \boldsymbol{S}_{0z}, \boldsymbol{S}_1'(\boldsymbol{S}_1, \boldsymbol{s}), \boldsymbol{S}_2 \right\rangle$$

$$\boldsymbol{S}_1', \boldsymbol{S}_2' = \boldsymbol{S} + \frac{\boldsymbol{1}}{\boldsymbol{2}}, \boldsymbol{S} - \frac{\boldsymbol{1}}{\boldsymbol{2}}$$

$$\vec{\boldsymbol{S}}_{\boldsymbol{1}}' = \vec{\boldsymbol{S}}_{\boldsymbol{1}} + \vec{\boldsymbol{s}}, \vec{\boldsymbol{S}}_{\boldsymbol{0}} = \vec{\boldsymbol{S}}_{\boldsymbol{1}}' + \vec{\boldsymbol{S}}_{\boldsymbol{2}}$$

$$\vec{\boldsymbol{S}}_{\boldsymbol{2}}' = \vec{\boldsymbol{S}}_{\boldsymbol{2}} + \vec{\boldsymbol{s}}, \vec{\boldsymbol{S}}_{\boldsymbol{0}} = \vec{\boldsymbol{S}}_{\boldsymbol{2}}' + \vec{\boldsymbol{S}}_{\boldsymbol{1}}$$

$$\left|\boldsymbol{S}_0^2, \boldsymbol{S}_{0z}, \boldsymbol{S}_1'(\boldsymbol{S}_1, \boldsymbol{s}), \boldsymbol{S}_2\right\rangle$$

$$\left|\boldsymbol{S}_0^2, \boldsymbol{S}_{0z}, \boldsymbol{S}_2'(\boldsymbol{S}_2, \boldsymbol{s}), \boldsymbol{S}_1\right\rangle$$

$$\boldsymbol{S}_1'(\boldsymbol{S}_1, \boldsymbol{s}) = \boldsymbol{S}_1 - \frac{\boldsymbol{1}}{\boldsymbol{2}}, \boldsymbol{S}_1 + \frac{\boldsymbol{1}}{\boldsymbol{2}}$$

$$\boldsymbol{S}_2'(\boldsymbol{S}_2, \boldsymbol{s}) = \boldsymbol{S}_2 - \frac{\boldsymbol{1}}{\boldsymbol{2}}, \boldsymbol{S}_2 + \frac{\boldsymbol{1}}{\boldsymbol{2}}$$

Racah's Method: W coefficients

$$C_{S_1' S_2'} = \langle S_0^2, S_{0z}, S_2'(S_2, s), S_1 | S_0^2, S_{0z}, S_1'(S_1, s), S_2 \rangle \equiv \sqrt{(2S_1'+1)(2S_2'+1)}\, W(S_1 s S_0 S_2; S_1' S_2')$$

$$\Delta H: \begin{array}{c} \\ |R_1\rangle|S+\frac{1}{2}, S_{1z}'\rangle \\ |R_1\rangle|S-\frac{1}{2}, S_{1z}'\rangle \\ |R_2\rangle|S+\frac{1}{2}, S_{2z}'\rangle \\ |R_2\rangle|S-\frac{1}{2}, S_{2z}'\rangle \end{array} \begin{array}{c} \begin{array}{cccc} |R_1\rangle|S+\frac{1}{2}, S_{1z}'\rangle & |R_1\rangle|S-\frac{1}{2}, S_{1z}'\rangle & |R_2\rangle|S+\frac{1}{2}, S_{2z}'\rangle & |R_2\rangle|S-\frac{1}{2}, S_{2z}'\rangle \end{array} \\ \begin{bmatrix} -2JS & 0 & b\cos\frac{\theta}{2} & b\sin\frac{\theta}{2} \\ 0 & J(S+1) & -b\sin\frac{\theta}{2} & b\cos\frac{\theta}{2} \\ b\cos\frac{\theta}{2} & -b\sin\frac{\theta}{2} & -2JS & 0 \\ b\sin\frac{\theta}{2} & b\cos\frac{\theta}{2} & 0 & J(S+1) \end{bmatrix} \end{array}$$

$$\cos\frac{\theta}{2} \equiv (-1)^{1-2S_0}\frac{S_0+\frac{1}{2}}{2S+1}$$

$$S_0 = \frac{1}{2}, \frac{3}{2}, \ldots\ldots 2S+\frac{1}{2}$$

Biedenharn, Blatt, and Rose, Revs. Modern Phys. 24, 249 (1952).