Density Matrix Renormalization Group for the Heisenberg Model

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I. THE FORMULAS

Some special cases of using the wigner-Eckart theorem:

$\langle J,J|X_0^{(0)}|J,J\rangle = \frac{1}{\sqrt{2J+1}}\langle J||X_0^{(0)}||J\rangle$

$\langle J_1',J_2',J'||X_{1,2}^{(0)}||J_1,J_2,J\rangle = (-1)^{J_1+J_2'+J+k}\frac{\sqrt{2J+1}}{\sqrt{2k+1}}\begin{Bmatrix} J_1' & J_1 & k \\ J_2 & J_2' & J \end{Bmatrix}\langle J_1'||T_i^{(1)}||J_1\rangle\langle J_2'||T_j^{(1)}||J_2\rangle$

$\langle J_{sys}',S',J',\alpha'||T_i^{(1)}||J_{sys},S,J,\alpha\rangle = (-1)^{J_{sys}'+S'+J+1}\delta_{S',S}\sqrt{(2J'+1)(2J+1)}\begin{Bmatrix} J' & 1 & J \\ J_{sys} & S' & J_{sys}' \end{Bmatrix}\langle J_{sys}',\alpha'||T_i^{(1)}||J_{sys},\alpha\rangle$

$\langle J_{sys}',S',J',\alpha'||T_i^{(1)}||J_{sys},S,J,\alpha\rangle = (-1)^{J_{sys}+S+J'+1}\delta_{J_{sys}',J_{sys}}\sqrt{(2J'+1)(2J+1)}\begin{Bmatrix} J' & 1 & J \\ S & J_{sys}' & S' \end{Bmatrix}\langle S',\alpha'||T_i^{(1)}||S,\alpha\rangle$

$\langle J_{sys}',S',J',\alpha'||X_{ij}^{(1)}||J_{sys},S,J,\alpha\rangle =$

$\sqrt{(2J'+1)(2K+1)(2J+1)}\begin{Bmatrix} J_{sys}' & J_{sys} & 1 \\ S' & S & 1 \\ J' & J & 1 \end{Bmatrix}\langle J_{sys}',\alpha'||T_i^{(1)}||J_{sys},\alpha\rangle\langle S'||T_j^{(1)}||S\rangle(k=1)$

II. IRREDUCIBLE TENSOR OPERATORS OF THE HEISENBERG TERMS

The product of two irreducible tensor operators (ITOs) is:
$\sum_{q_1,q_2}\langle k_1q_1k_2q_2|kq\rangle T_{q_1}^{(k_1)}T_{q_2}^{(k_2)} = T_q^{(k)}$ $(k=k_1+k_2,...,|k_1-k_2|)$
The scalar product of two irreducible tensor operators (ITOs) is:
$Q=(U^{(L)},V^{(L)})=\sum_{M=-L}^{L}(-1)^M U_M^{(L)}V_{-M}^{(L)}$, so:
$Q=(U^{(1)},V^{(1)})=\sum_{M=-1}^{1}(-1)^M U_M^{(1)}V_{-M}^{(1)} = -U_{-1}^{(1)}V_1^{(1)} - U_1^{(1)}V_{-1}^{(1)} + U_0^{(1)}V_0^{(1)}$
as the product of two irreducible tensor operators $X_0^{(0)}=[U_L\times V_L]_0=\frac{(-1)^L}{\sqrt{2L+1}}Q$, so: $X_0^{(0)}=[U_1\times V_1]_0=\frac{(-1)}{\sqrt{3}}Q$

For the Heisenberg spin interaction terms, we use the rank-1 ITO $T_{i,q}^{(1)}(q=+1,0,-1)$ to reproduce spin operators, which is:
$T_{i,1}^{(1)}=-\frac{1}{\sqrt{2}}S_i^{\dagger}, T_{i,0}^{(1)}=S_i^z, T_{i,-1}^{(1)}=\frac{1}{\sqrt{2}}S_i^-$
The Hamiltonian of Heisenberg term H_J can be expressed as:
$H_J=\sum_{i,j}J_{ij}(S_i^xS_j^x+S_i^yS_j^y+S_i^zS_j^z)=\sum_{i,j}J_{ij}(\frac{1}{2}S_i^{\dagger}S_j^-+\frac{1}{2}S_i^-S_j^{\dagger}+S_i^zS_j^z)=\sum_{i,j}J_{ij}(-T_{i,1}^{(1)}T_{j,-1}^{(1)}-T_{i,-1}^{(1)}T_{j,1}^{(1)}+T_{i,0}^{(1)}T_{j,0}^{(1)})=\sum_{i,j}J_{ij}Q$
Thus, the Hamiltonian H_J can be presented as:
$H_J=-\sqrt{3}\sum_{i,j}J_{ij}X_{0,ij}^{(0)}$

A. Initialization

There are only one site in system and environment. We need to calculate $\langle J||T^{(1)}||J\rangle$ for initialization.
$\langle JJ|T_0^{(1)}|JJ\rangle = \frac{1}{\sqrt{2J+1}}\langle JJ10|JJ\rangle\langle J||T^{(1)}||J\rangle = \frac{1}{\sqrt{2J+1}}\frac{J}{\sqrt{J(J+1)}}\langle J||T^{(1)}||J\rangle = J$
Thus, $\langle J||T^{(1)}||J\rangle = \sqrt{J(J+1)(2J+1)}$.
If $J=\frac{1}{2}$, $\langle J||T^{(1)}||J\rangle=\sqrt{\frac{3}{2}}$.
If $J=1$, $\langle J||T^{(1)}||J\rangle=\sqrt{6}$.

B. Increasing a new site

In this section, we add a new site in the system block. The irreducible basis is different from before. We need to calculate $\langle J_{sys}',S',J',\alpha'||T_i^{(1)}||J_{sys},S,J,\alpha\rangle$ in the new irreducible basis. There, we need the product of two irreducible tensor operators. There, $|J-J'|=0,1$.
If the site i is in the system block, then

$\langle J'_{sys}, S', J', \alpha' || T_i^{(1)} || J_{sys}, S, J, \alpha\rangle = (-1)^{J'_{sys}+S'+J+1}\delta_{S',S}\sqrt{(2J'+1)(2J+1)} \begin{Bmatrix} J' & 1 & J \\ J_{sys} & S' & J'_{sys} \end{Bmatrix} \langle J'_{sys}, \alpha' || T_i^{(1)} || J_{sys}, \alpha\rangle$
if $J = J'$
$\langle J'_{sys}, S', J', \alpha' || T_i^{(1)} || J_{sys}, S, J, \alpha\rangle = (-1)^{J'_{sys}+S'+J+1}\delta_{S',S}(2J+1) \begin{Bmatrix} J & 1 & J \\ J_{sys} & S' & J'_{sys} \end{Bmatrix} \langle J'_{sys}, \alpha' || T_i^{(1)} || J_{sys}, \alpha\rangle$
$= (-1)^{J'_{sys}+S+J+1}(2J+1) \begin{Bmatrix} J & 1 & J \\ J_{sys} & S & J'_{sys} \end{Bmatrix} \langle J'_{sys}, \alpha' || T_i^{(1)} || J_{sys}, \alpha\rangle$
which would be non-zero only for $J \neq 0$. If $J = 0$, the $6j-$ coefficient would be zero.

If the site i is in the added new site, then
$\langle J'_{sys}, S', J', \alpha' || T_i^{(1)} || J_{sys}, S, J, \alpha\rangle = (-1)^{J_{sys}+S+J'+1}\delta_{J'_{sys},J_{sys}}\sqrt{(2J'+1)(2J+1)} \begin{Bmatrix} J' & 1 & J \\ S & J'_{sys} & S' \end{Bmatrix} \langle S', \alpha' || T_i^{(1)} || S, \alpha\rangle$
For the diagonal part with $J = J'$, the result can be simplified as
$\langle J'_{sys}, S', J', \alpha' || T_i^{(1)} || J_{sys}, S, J, \alpha\rangle = (-1)^{J_{sys}+S+J+1}\delta_{J'_{sys},J_{sys}}(2J+1) \begin{Bmatrix} J & 1 & J \\ S & J'_{sys} & S' \end{Bmatrix} \langle S', \alpha' || T_i^{(1)} || S, \alpha\rangle$
which would be non-zero only for $J \neq 0$. If $J = 0$, the $6j-$ coefficient would be zero.

C. Calculating the Hamiltonian

Suppose the site i belong to the system block and the site j is the added new site. Then the Hamiltonian H_J is
$H_J = -\sqrt{3}\sum_{i,j} J_{i,j} X^0_{0,ij}$
The matrix element of the Hamiltonian H_J is
$-\sqrt{3}J_{i,j}\langle J'_{sys}, S', J', \alpha' | X^0_{0,ij} | J_{sys}, S, J, \alpha\rangle = -\frac{\sqrt{3}}{\sqrt{2J+1}} J_{i,j}\langle J'_{sys}, S', J', \alpha' || X^0_{ij} || J_{sys}, S, J, \alpha\rangle$
as $\langle J'_{sys}, S', J', \alpha' || X^0_{ij} || J_{sys}, S, J, \alpha\rangle = \sqrt{\frac{2J+1}{3}}(-1)^{J_{sys}+S'+1+J} \begin{Bmatrix} J'_{sys} & J_{sys} & 1 \\ S & S' & J \end{Bmatrix} \langle J'_{sys}, \alpha' || T_i^{(1)} || J_{sys}, \alpha\rangle\langle S' || T_j^{(1)} || S\rangle$
Thus, the matrix element of the Hamiltonian H_J is
$-\sqrt{3}J_{i,j}\langle J'_{sys}, S', J', \alpha' | X^0_{0,ij} | J_{sys}, S, J, \alpha\rangle = -J_{i,j}(-1)^{J_{sys}+S'+1+J} \begin{Bmatrix} J'_{sys} & J_{sys} & 1 \\ S & S' & J \end{Bmatrix} \langle J'_{sys}, \alpha' || T_i^{(1)} || J_{sys}, \alpha\rangle\langle S' || T_j^{(1)} || S\rangle$

D. Hamiltonian from the Heisenberg interactions

In this section, we need the matrix-vector multiplication calculations
$H_{n+1} = H_{A_n} + H_{B_n} + S_{A_n}\cdot S_{A_n+1} + S_{A_n+1}\cdot S_{B_n+1} + S_{B_n+1}\cdot S_{B_n}$
$H_{n+1}|f\rangle = |g\rangle$, thus
$(H_{A_n} + H_{B_n} + S_{A_n}\cdot S_{A_n+1} + S_{A_n+1}\cdot S_{B_n+1} + S_{B_n+1}\cdot S_{B_n})|f\rangle = |g\rangle$
$(H_{A_n})|f\rangle = |g_1\rangle$, $(H_{B_n})|f\rangle = |g_2\rangle$, $(S_{A_n}\cdot S_{A_n+1})|f\rangle = |g_3\rangle$, $(S_{A_n+1}\cdot S_{B_n+1})|f\rangle = |g_4\rangle$, $(S_{B_n+1}\cdot S_{B_n})|f\rangle = |g_5\rangle$
thus, $|g\rangle = |g_1\rangle + |g_2\rangle + |g_3\rangle + |g_4\rangle + |g_5\rangle$
generally, $J_{ij}\vec{S}_i\cdot\vec{S}_j|f\rangle = J_{ij}\vec{S}_i\cdot\vec{S}_j|J_{sys}, J_{ns}, J_{sysnew}, J_{env}, J_{ne}, J_{envnew}, J\rangle f(J_{sys}, J_{ns}, J_{sysnew}, J_{env}, J_{ne}, J_{envnew}, J)$
$= \sum_{J'_{sys},J'_{ns},J'_{sysnew},J'_{env},J'_{ne},J'_{envnew},J'} |J'_{sys}, J'_{ns}, J'_{sysnew}, J'_{env}, J'_{ne}, J'_{envnew}, J'\rangle\langle J'_{sys}, J'_{ns}, J'_{sysnew}, J'_{env}, J'_{ne}, J'_{envnew}, J'|$
$J_{ij}\vec{S}_i\cdot\vec{S}_j|J_{sys}, J_{ns}, J_{sysnew}, J_{env}, J_{ne}, J_{envnew}, J\rangle f(J_{sys}, J_{ns}, J_{sysnew}, J_{env}, J_{ne}, J_{envnew}, J)$
$= \sum_{J'_{sys},J'_{ns},J'_{sysnew},J'_{env},J'_{ne},J'_{envnew},J'} \langle J'_{sys}, J'_{ns}, J'_{sysnew}, J'_{env}, J'_{ne}, J'_{envnew}, J'|J_{ij}\vec{S}_i\cdot\vec{S}_j|J_{sys}, J_{ns}, J_{sysnew}, J_{env}, J_{ne}, J_{envnew}, J\rangle$
$f(J_{sys}, J_{ns}, J_{sysnew}, J_{env}, J_{ne}, J_{envnew}, J)|J'_{sys}, J'_{ns}, J'_{sysnew}, J'_{env}, J'_{ne}, J'_{envnew}, J'\rangle$
$= g(J_{sys}, J_{ns}, J_{sysnew}, J_{env}, J_{ne}, J_{envnew}, J)|J_{sys}, J_{ns}, J_{sysnew}, J_{env}, J_{ne}, J_{envnew}, J\rangle$

1. *Hamiltonian from the Heisenberg interactions in the system block*

The Hamiltonian contribution is $J_{ij}\vec{S}_i\cdot\vec{S}_j$, where $\vec{S}_i$ and $\vec{S}_j$ belong to the system block. The matrix-vector multiplication is explicitly written as
$\langle J'_{sys}, J'_{ns}, J'_{sysnew}, J'_{env}, J'_{ne}, J'_{envnew}, J'|J_{ij}\vec{S}_i\cdot\vec{S}_j|J_{sys}, J_{ns}, J_{sysnew}, J_{env}, J_{ne}, J_{envnew}, J\rangle$
$f(J_{sys}, J_{ns}, J_{sysnew}, J_{env}, J_{ne}, J_{envnew}, J) = g(J_{sys}, J_{ns}, J_{sysnew}, J_{env}, J_{ne}, J_{envnew}, J)$
As $\vec{S}_i$ and $\vec{S}_j$ belong to the system block. Thus
$\langle J'_{sys}, J_{ns}, J'_{sysnew}, J_{env}, J_{ne}, J_{envnew}, 0|J_{ij}\vec{S}_i\cdot\vec{S}_j|J_{sys}, J_{ns}, J_{sysnew}, J_{env}, J_{ne}, J_{envnew}, 0\rangle$
$f(J_{sys}, J_{ns}, J_{sysnew}, J_{env}, J_{ne}, J_{envnew}, 0) = g(J_{sys}, J_{ns}, J_{sysnew}, J_{env}, J_{ne}, J_{envnew}, 0)$

We can compute the matrix element as
$J_{ij}\langle J'_{sys},J_{ns},J'_{sysnew},J_{env},J_{ne},J_{envnew},0|\vec{S}_i\cdot\vec{S}_j|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$=-\sqrt{3}J_{ij}\langle J'_{sys},J_{ns},J'_{sysnew},J_{env},J_{ne},J_{envnew},0|X^0_{0,ij}|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$=-J_{ij}\frac{\sqrt{3}}{\sqrt{2J_{sysnew}+1}}\langle J'_{sys},J_{ns},J'_{sysnew},J_{env},J_{ne},J_{envnew},0||X^0_{ij}||J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$=-J_{ij}\frac{\sqrt{3}}{\sqrt{2J_{sysnew}+1}}\langle J'_{sys}||X^0_{ij}||J_{sys}\rangle$

2. Hamiltonian from the Heisenberg interactions between the system block and the added sites

The Hamiltonian contribution is $J_{ij}\vec{S}_i\cdot\vec{S}_j$, where $\vec{S}_i$ and $\vec{S}_j$ belong to the system block and the added site, respectively. The matrix-vector multiplication is explicitly written as
$\langle J'_{sys},J'_{ns},J'_{sysnew},J'_{env},J'_{ne},J'_{envnew},J'|J_{ij}\vec{S}_i\cdot\vec{S}_j|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J\rangle$
$f(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J)=g(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J)$
As $\vec{S}_i$ and $\vec{S}_j$ belong to the system block and the added site, respectively. Thus
$\langle J'_{sys},J_{ns},J'_{sysnew},J_{env},J_{ne},J_{envnew},0|J_{ij}\vec{S}_i\cdot\vec{S}_j|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$f(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0)=g(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0)$
We can compute the matrix element as
$J_{ij}\langle J'_{sys},J_{ns},J'_{sysnew},J_{env},J_{ne},J_{envnew},0|\vec{S}_i\cdot\vec{S}_j|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$=-\sqrt{3}J_{ij}\langle J'_{sys},J_{ns},J'_{sysnew},J_{env},J_{ne},J_{envnew},0|X^0_{0,ij}|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$=-J_{ij}\frac{\sqrt{3}}{\sqrt{2J_{sysnew}+1}}\langle J'_{sys},J_{ns},J'_{sysnew},J_{env},J_{ne},J_{envnew},0||X^0_{ij}||J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$=-J_{ij}\frac{\sqrt{3}}{\sqrt{2J_{sysnew}+1}}\frac{\sqrt{2J_{sysnew}+1}}{\sqrt{3}}(-1)^{J_{sys}+J_{ns}+1+J_{sysnew}}\begin{Bmatrix}J'_{sys}&J_{sys}&1\\J_{ns}&J_{ns}&J_{sysnew}\end{Bmatrix}\langle J'_{sys},\alpha'||T^{(1)}_i||J_{sys},\alpha\rangle\langle J_{ns}||T^{(1)}_j||J_{ns}\rangle$
$=-J_{ij}(-1)^{J_{sys}+J_{ns}+1+J_{sysnew}}\begin{Bmatrix}J'_{sys}&J_{sys}&1\\J_{ns}&J_{ns}&J_{sysnew}\end{Bmatrix}\langle J'_{sys},\alpha'||T^{(1)}_i||J_{sys},\alpha\rangle\langle J_{ns}||T^{(1)}_j||J_{ns}\rangle$
If $J_{sys}=J'_{sys}$, then $J_{sys}\neq 0$

3. Hamiltonian from the Heisenberg interactions between the environment block and the added sites

The Hamiltonian contribution is $J_{ij}\vec{S}_i\cdot\vec{S}_j$, where $\vec{S}_i$ and $\vec{S}_j$ belong to the environment block and the added site, respectively. The matrix-vector multiplication is explicitly written as
$\langle J'_{sys},J'_{ns},J'_{sysnew},J'_{env},J'_{ne},J'_{envnew},J'|J_{ij}\vec{S}_i\cdot\vec{S}_j|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J\rangle$
$f(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J)=g(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J)$
As $\vec{S}_i$ and $\vec{S}_j$ belong to the environment block and the added site, respectively. Thus
$\langle J_{sys},J_{ns},J_{sysnew},J'_{env},J_{ne},J'_{envnew},0|J_{ij}\vec{S}_i\cdot\vec{S}_j|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$f(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0)=g(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0)$
We can compute the matrix element as
$J_{ij}\langle J_{sys},J_{ns},J_{sysnew},J'_{env},J_{ne},J'_{envnew},0|\vec{S}_i\cdot\vec{S}_j|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$=-\sqrt{3}J_{ij}\langle J_{sys},J_{ns},J_{sysnew},J'_{env},J_{ne},J'_{envnew},0|X^0_{0,ij}|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$=-J_{ij}\frac{\sqrt{3}}{\sqrt{2J_{envnew}+1}}\langle J_{sys},J_{ns},J_{sysnew},J'_{env},J_{ne},J'_{envnew},0||X^0_{ij}||J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$=-J_{ij}\frac{\sqrt{3}}{\sqrt{2J_{envnew}+1}}\frac{\sqrt{2J_{envnew}+1}}{\sqrt{3}}(-1)^{J_{env}+J_{ne}+1+J_{envnew}}\begin{Bmatrix}J'_{env}&J_{env}&1\\J_{ne}&J_{ne}&J_{envnew}\end{Bmatrix}\langle J'_{env},\alpha'||T^{(1)}_i||J_{env},\alpha\rangle\langle J_{ne}||T^{(1)}_j||J_{ne}\rangle$
$=-J_{ij}(-1)^{J_{env}+J_{ne}+1+J_{envnew}}\begin{Bmatrix}J'_{env}&J_{env}&1\\J_{ne}&J_{ne}&J_{envnew}\end{Bmatrix}\langle J'_{env},\alpha'||T^{(1)}_i||J_{env},\alpha\rangle\langle J_{ne}||T^{(1)}_j||J_{ne}\rangle$

4. Hamiltonian from the Heisenberg interactions between the added sites

The Hamiltonian contribution is $J_{ij}\vec{S}_i\cdot\vec{S}_j$, where $\vec{S}_i$ and $\vec{S}_j$ belong to the added sites, respectively. The matrix-vector multiplication is explicitly written as
$\langle J'_{sys},J'_{ns},J'_{env},J'_{ne},J'|J_{ij}\vec{S}_i\cdot\vec{S}_j|J_{sys},J_{ns},J_{env},J_{ne},J\rangle$

$f(J_{sys}, J_{ns}, J_{env}, J_{ne}, J) = g(J_{sys}, J_{ns}, J_{env}, J_{ne}, J)$
As $\vec{S}_i$ and $\vec{S}_j$ belong to the added sites, respectively. Thus
$\langle J_{sys}, J_{ns}, J_{env}, J_{ne}, J_{ns,ne}, 0|J_{ij}\vec{S}_i \cdot \vec{S}_j|J_{sys}, J_{ns}, J_{env}, J_{ne}, J_{ns,ne}, 0\rangle$
$f(J_{sys}, J_{ns}, J_{env}, J_{ne}, J_{ns,ne}, 0) = g(J_{sys}, J_{ns}, J_{env}, J_{ne}, J_{ns,ne}, 0)$
We can compute the matrix element as
$\langle J_{sys}, J_{ns}, J_{env}, J_{ne}, J_{ns,ne}|J_{ij}\vec{S}_i \cdot \vec{S}_j|J_{sys}, J_{ns}, J_{env}, J_{ne}, J_{ns,ne}\rangle$
$= -\sqrt{3}J_{ij}\langle J_{sys}, J_{ns}, J_{env}, J_{ne}, J_{ns,ne}|X^0_{0,ij}|J_{sys}, J_{ns}, J_{env}, J_{ne}, J_{ns,ne}\rangle$
$= -J_{ij}\frac{\sqrt{3}}{\sqrt{2J_{ns,ne}+1}}\langle J_{ns}, J_{ne}, J_{ns,ne}||X^0_{0,ij}||J_{ns}, J_{ne}, J_{ns,ne}\rangle$
$= -J_{ij}\frac{\sqrt{3}}{\sqrt{2J_{ns,ne}+1}}\frac{\sqrt{2J_{ns,ne}+1}}{\sqrt{3}}(-1)^{J_{ns}+J_{ne}+1+J_{ns,ne}}\begin{Bmatrix} J_{ns} & J_{ns} & 1 \\ J_{ne} & J_{ne} & J_{ns,ne}\end{Bmatrix}\langle J_{ns}, ||T^{(1)}_i||J_{ns}\rangle\langle J_{ne}||T^{(1)}_j||J_{ne}\rangle$
$= -J_{ij}(-1)^{J_{ns}+J_{ne}+1+J_{ns,ne}}\begin{Bmatrix} J_{ns} & J_{ns} & 1 \\ J_{ne} & J_{ne} & J_{ns,ne}\end{Bmatrix}\langle J_{ns}, ||T^{(1)}_i||J_{ns}\rangle\langle J_{ne}||T^{(1)}_j||J_{ne}\rangle$

5. *Hamiltonian from the Heisenberg interactions between the system and environment*

The Hamiltonian contribution is $J_{ij}\vec{S}_i \cdot \vec{S}_j$, where $\vec{S}_i$ and $\vec{S}_j$ belong to the system and environment, respectively. The matrix-vector multiplication is explicitly written as
$\langle J'_{sys}, J'_{ns}, J'_{env}, J'_{ne}, J'|J_{ij}\vec{S}_i \cdot \vec{S}_j|J_{sys}, J_{ns}, J_{env}, J_{ne}, J\rangle$
$f(J_{sys}, J_{ns}, J_{env}, J_{ne}, J) = g(J_{sys}, J_{ns}, J_{env}, J_{ne}, J)$
As $\vec{S}_i$ and $\vec{S}_j$ belong to the system and environment, respectively. Thus
$\langle J'_{sys}, J_{ns}, J'_{env}, J_{ne}, J_{sys,env}, 0|J_{ij}\vec{S}_i \cdot \vec{S}_j|J_{sys}, J_{ns}, J_{env}, J_{ne}, J_{sys,env}, 0\rangle$
$f(J_{sys}, J_{ns}, J_{env}, J_{ne}, J_{sys,env}, 0) = g(J_{sys}, J_{ns}, J_{env}, J_{ne}, J_{sys,env}, 0)$
We can compute the matrix element as
$\langle J'_{sys}, J_{ns}, J'_{env}, J_{ne}, J_{sys,env}|J_{ij}\vec{S}_i \cdot \vec{S}_j|J_{sys}, J_{ns}, J_{env}, J_{ne}, J_{sys,env}\rangle$
$= -\sqrt{3}J_{ij}\langle J'_{sys}, J_{ns}, J'_{env}, J_{ne}, J_{sys,env}|X^0_{0,ij}|J_{sys}, J_{ns}, J_{env}, J_{ne}, J_{sys,env}\rangle$
$= -J_{ij}\frac{\sqrt{3}}{\sqrt{2_{sys,env}+1}}\langle J'_{sys}, J_{ns}, J'_{env}, J_{ne}, J_{sys,env}||X^0_{ij}||J_{sys}, J_{ns}, J_{env}, J_{ne}, J_{sys,env}\rangle$
$= -J_{ij}\frac{\sqrt{3}}{\sqrt{2J_{sys,env}+1}}\frac{\sqrt{2J_{sys,env}+1}}{\sqrt{3}}(-1)^{J_{sys}+J'_{env}+1+J_{sys,env}}\begin{Bmatrix} J'_{sys} & J_{sys} & 1 \\ J_{env} & J'_{env} & J_{sys,env}\end{Bmatrix}\langle J'_{sys}, \alpha'||T^{(1)}_i||J_{sys}, \alpha\rangle\langle J'_{env}, \beta'||T^{(1)}_j||J_{env}, \beta\rangle$
$= -J_{ij}(-1)^{J_{sys}+J'_{env}+1+J_{sys,env}}\begin{Bmatrix} J'_{sys} & J_{sys} & 1 \\ J_{env} & J'_{env} & J_{sys,env}\end{Bmatrix}\langle J'_{sys}, \alpha'||T^{(1)}_i||J_{sys}, \alpha\rangle\langle J'_{env}, \beta'||T^{(1)}_j||J_{env}, \beta\rangle$

III. IRREDUCIBLE TENSOR OPERATORS OF THE K TERMS

$$
\begin{aligned}
H = \sum_{\langle i,j\rangle} K(\mathbf{S}_i \cdot \mathbf{S}_j)^2 = \sum_{\langle i,j\rangle} & K(1/2(S^\dagger_i S^-_j + S^-_i S^\dagger_j) + (S^z_i S^z_j))(1/2(S^\dagger_i S^-_j + S^-_i S^\dagger_j) + (S^z_i S^z_j)) \\
= \sum_{\langle i,j\rangle} & K(1/4(S^\dagger_i S^-_j S^\dagger_i S^-_j) + 1/4(S^\dagger_i S^-_j S^-_i S^\dagger_j) + 1/2(S^\dagger_i S^-_j S^z_i S^z_j) \\
& +1/4(S^-_i S^\dagger_j S^\dagger_i S^-_j) + 1/4(S^-_i S^\dagger_j S^-_i S^\dagger_j) + 1/2(S^-_i S^\dagger_j S^z_i S^z_j) \\
& +1/2(S^z_i S^z_j S^\dagger_i S^-_j) + 1/2(S^z_i S^z_j S^-_i S^\dagger_j) + (S^z_i S^z_j S^z_i S^z_j)) \\
= \sum_{\langle i,j\rangle} & K(1/4(S^\dagger_i S^\dagger_i S^-_j S^-_j) + 1/4(S^\dagger_i S^-_i S^-_j S^\dagger_j) + 1/2(S^\dagger_i S^z_i S^-_j S^z_j) \\
& +1/4(S^-_i S^\dagger_i S^\dagger_j S^-_j) + 1/4(S^-_i S^-_i S^\dagger_j S^\dagger_j) + 1/2(S^-_i S^z_i S^\dagger_j S^z_j) \\
& +1/2(S^z_i S^\dagger_i S^z_j S^-_j) + 1/2(S^z_i S^-_i S^z_j S^\dagger_j) + (S^z_i S^z_i S^z_j S^z_j))
\end{aligned}
\tag{1}
$$

as,
$T^{(0)}_0 = \langle 111-1|00\rangle T^{(1)}_1 T^{(1)}_{-1} + \langle 1-111|00\rangle T^{(1)}_{-1} T^{(1)}_1 + \langle 1010|00\rangle T^{(1)}_0 T^{(1)}_0 = \frac{1}{\sqrt{3}} T^{(1)}_1 T^{(1)}_{-1} + \frac{1}{\sqrt{3}} T^{(1)}_{-1} T^{(1)}_1 - \frac{1}{\sqrt{3}} T^{(1)}_0 T^{(1)}_0$

$$T_0^{(1)} = \langle 111-1|10\rangle T_1^{(1)}T_{-1}^{(1)} + \langle 1-111|10\rangle T_{-1}^{(1)}T_1^{(1)} + \langle 1010|10\rangle T_0^{(1)}T_0^{(1)} = \tfrac{1}{\sqrt{2}}T_1^{(1)}T_{-1}^{(1)} - \tfrac{1}{\sqrt{2}}T_{-1}^{(1)}T_1^{(1)}$$
$$T_1^{(1)} = \langle 1011|11\rangle T_0^{(1)}T_1^{(1)} + \langle 1110|11\rangle T_1^{(1)}T_0^{(1)} = -\tfrac{1}{\sqrt{2}}T_0^{(1)}T_1^{(1)} + \tfrac{1}{\sqrt{2}}T_1^{(1)}T_0^{(1)}$$
$$T_{-1}^{(1)} = \langle 101-1|1-1\rangle T_0^{(1)}T_{-1}^{(1)} + \langle 1-110|1-1\rangle T_{-1}^{(1)}T_0^{(1)} = \tfrac{1}{\sqrt{2}}T_0^{(1)}T_{-1}^{(1)} - \tfrac{1}{\sqrt{2}}T_{-1}^{(1)}T_0^{(1)}$$
$$T_0^{(2)} = \langle 111-1|20\rangle T_1^{(1)}T_{-1}^{(1)} + \langle 1-111|20\rangle T_{-1}^{(1)}T_1^{(1)} + \langle 1010|20\rangle T_0^{(1)}T_0^{(1)} = \tfrac{1}{\sqrt{6}}T_1^{(1)}T_{-1}^{(1)} + \tfrac{1}{\sqrt{6}}T_{-1}^{(1)}T_1^{(1)} + \sqrt{\tfrac{2}{3}}T_0^{(1)}T_0^{(1)}$$
$$T_1^{(2)} = \langle 1011|21\rangle T_0^{(1)}T_1^{(1)} + \langle 1110|21\rangle T_1^{(1)}T_0^{(1)} = \tfrac{1}{\sqrt{2}}T_0^{(1)}T_1^{(1)} + \tfrac{1}{\sqrt{2}}T_1^{(1)}T_0^{(1)}$$
$$T_{-1}^{(2)} = \langle 101-1|2-1\rangle T_0^{(1)}T_{-1}^{(1)} + \langle 1-110|2-1\rangle T_{-1}^{(1)}T_0^{(1)} = \tfrac{1}{\sqrt{2}}T_0^{(1)}T_{-1}^{(1)} + \tfrac{1}{\sqrt{2}}T_{-1}^{(1)}T_0^{(1)}$$
$$T_2^{(2)} = \langle 1111|22\rangle T_1^{(1)}T_1^{(1)} = T_1^{(1)}T_1^{(1)}$$
$$T_{-2}^{(2)} = \langle 1-11-1|2-2\rangle T_{-1}^{(1)}T_{-1}^{(1)} = T_{-1}^{(1)}T_{-1}^{(1)}$$

so,

$$\begin{aligned} H = \sum_{\langle i,j\rangle} K_{i,j}(&T_{i,1}^{(1)}T_{i,1}^{(1)}T_{j,-1}^{(1)}T_{j,-1}^{(1)} + T_{i,1}^{(1)}T_{i,-1}^{(1)}T_{j,-1}^{(1)}T_{j,1}^{(1)} - T_{i,1}^{(1)}T_{i,0}^{(1)}T_{j,-1}^{(1)}T_{j,0}^{(1)} \\ &+T_{i,-1}^{(1)}T_{i,1}^{(1)}T_{j,1}^{(1)}T_{j,-1}^{(1)} + T_{i,-1}^{(1)}T_{i,-1}^{(1)}T_{j,1}^{(1)}T_{j,1}^{(1)} - T_{i,-1}^{(1)}T_{i,0}^{(1)}T_{j,1}^{(1)}T_{j,0}^{(1)} \\ &-T_{i,0}^{(1)}T_{i,1}^{(1)}T_{j,0}^{(1)}T_{j,-1}^{(1)} - T_{i,0}^{(1)}T_{i,-1}^{(1)}T_{j,0}^{(1)}T_{j,1}^{(1)} + T_{i,0}^{(1)}T_{i,0}^{(1)}T_{j,0}^{(1)}T_{j,0}^{(1)}) \end{aligned} \tag{2}$$

$$\begin{aligned} X_0^{(0)} &= ([T_i^{(0)} \times T_j^{(0)}]_0 + [T_i^{(1)} \times T_j^{(1)}]_0 + [T_i^{(2)} \times T_j^{(2)}]_0) \\ &= (Q_{ij}^{(0)} - \frac{1}{\sqrt{3}}Q_{ij}^{(1)} + \frac{1}{\sqrt{5}}Q_{ij}^{(2)}) \\ &= T_{i,0}^{(0)}T_{j,0}^{(0)} - \frac{1}{\sqrt{3}}(-T_{i,1}^{(1)}T_{j,-1}^{(1)} - T_{i,-1}^{(1)}T_{j,1}^{(1)} + T_{i,0}^{(1)}T_{j,0}^{(1)}) + \frac{1}{\sqrt{5}}(T_{i,2}^{(2)}T_{j,-2}^{(2)} - T_{i,1}^{(2)}T_{j,-1}^{(2)} + T_{i,0}^{(2)}T_{j,0}^{(2)} - T_{i,-1}^{(2)}T_{j,1}^{(2)} + T_{i,-2}^{(2)}T_{j,2}^{(2)}) \\ &= (\frac{1}{\sqrt{3}}T_{i,1}^{(1)}T_{i,-1}^{(1)} + \frac{1}{\sqrt{3}}T_{i,-1}^{(1)}T_{i,1}^{(1)} - \frac{1}{\sqrt{3}}T_{i,0}^{(1)}T_{i,0}^{(1)}) \\ &\quad(\frac{1}{\sqrt{3}}T_{j,1}^{(1)}T_{j,-1}^{(1)} + \frac{1}{\sqrt{3}}T_{j,-1}^{(1)}T_{j,1}^{(1)} - \frac{1}{\sqrt{3}}T_{j,0}^{(1)}T_{j,0}^{(1)}) \\ &\quad-\frac{1}{\sqrt{3}}(-(-\frac{1}{\sqrt{2}}T_{i,0}^{(1)}T_{i,1}^{(1)} + \frac{1}{\sqrt{2}}T_{i,1}^{(1)}T_{i,0}^{(1)}) \\ &\quad(\frac{1}{\sqrt{2}}T_{j,0}^{(1)}T_{j,-1}^{(1)} - \frac{1}{\sqrt{2}}T_{j,-1}^{(1)}T_{j,0}^{(1)}) - (\frac{1}{\sqrt{2}}T_{i,0}^{(1)}T_{i,-1}^{(1)} - \frac{1}{\sqrt{2}}T_{i,-1}^{(1)}T_{i,0}^{(1)})(-\frac{1}{\sqrt{2}}T_{j,0}^{(1)}T_{j,1}^{(1)} + \frac{1}{\sqrt{2}}T_{j,1}^{(1)}T_{j,0}^{(1)}) \\ &\quad+(\frac{1}{\sqrt{2}}T_{i,1}^{(1)}T_{i,-1}^{(1)} - \frac{1}{\sqrt{2}}T_{i,-1}^{(1)}T_{i,1}^{(1)})(\frac{1}{\sqrt{2}}T_{j,1}^{(1)}T_{j,-1}^{(1)} - \frac{1}{\sqrt{2}}T_{j,-1}^{(1)}T_{j,1}^{(1)})) \\ &\quad+\frac{1}{\sqrt{5}}((T_{i,1}^{(1)}T_{i,1}^{(1)})(T_{j,-1}^{(1)}T_{j,-1}^{(1)}) - (\frac{1}{\sqrt{2}}T_{i,0}^{(1)}T_{i,1}^{(1)} + \frac{1}{\sqrt{2}}T_{i,1}^{(1)}T_{i,0}^{(1)})(\frac{1}{\sqrt{2}}T_{j,0}^{(1)}T_{j,-1}^{(1)} + \frac{1}{\sqrt{2}}T_{j,-1}^{(1)}T_{j,0}^{(1)}) \\ &\quad(\frac{1}{\sqrt{6}}T_{i,1}^{(1)}T_{i,-1}^{(1)} + \frac{1}{\sqrt{6}}T_{i,-1}^{(1)}T_{i,1}^{(1)} + \sqrt{\frac{2}{3}}T_{i,0}^{(1)}T_{i,0}^{(1)})(\frac{1}{\sqrt{6}}T_{j,1}^{(1)}T_{j,-1}^{(1)} + \frac{1}{\sqrt{6}}T_{j,-1}^{(1)}T_{j,1}^{(1)} + \sqrt{\frac{2}{3}}T_{j,0}^{(1)}T_{j,0}^{(1)}) \\ &\quad-(\frac{1}{\sqrt{2}}T_{i,0}^{(1)}T_{i,-1}^{(1)} + \frac{1}{\sqrt{2}}T_{i,-1}^{(1)}T_{i,0}^{(1)})(\frac{1}{\sqrt{2}}T_{j,0}^{(1)}T_{j,1}^{(1)} + \frac{1}{\sqrt{2}}T_{j,1}^{(1)}T_{j,0}^{(1)}) + T_{i,-1}^{(1)}T_{i,-1}^{(1)}T_{j,1}^{(1)}T_{j,1}^{(1)}) \end{aligned} \tag{3}$$

$$\begin{aligned} &(Q_{ij}^{(0)} - Q_{ij}^{(1)} + Q_{ij}^{(2)}) = \\ &T_{i,0}^{(0)}T_{j,0}^{(0)} - (-T_{i,1}^{(1)}T_{j,-1}^{(1)} - T_{i,-1}^{(1)}T_{j,1}^{(1)} + T_{i,0}^{(1)}T_{j,0}^{(1)}) + (T_{i,2}^{(2)}T_{j,-2}^{(2)} - T_{i,1}^{(2)}T_{j,-1}^{(2)} + T_{i,0}^{(2)}T_{j,0}^{(2)} - T_{i,-1}^{(2)}T_{j,1}^{(2)} + T_{i,-2}^{(2)}T_{j,2}^{(2)}) = \\ &(T_{i,1}^{(1)}T_{i,1}^{(1)}T_{j,-1}^{(1)}T_{j,-1}^{(1)} + T_{i,1}^{(1)}T_{i,-1}^{(1)}T_{j,-1}^{(1)}T_{j,1}^{(1)} - T_{i,1}^{(1)}T_{i,0}^{(1)}T_{j,-1}^{(1)}T_{j,0}^{(1)} \\ &+T_{i,-1}^{(1)}T_{i,1}^{(1)}T_{j,1}^{(1)}T_{j,-1}^{(1)} + T_{i,-1}^{(1)}T_{i,-1}^{(1)}T_{j,1}^{(1)}T_{j,1}^{(1)} - T_{i,-1}^{(1)}T_{i,0}^{(1)}T_{j,1}^{(1)}T_{j,0}^{(1)} \\ &-T_{i,0}^{(1)}T_{i,1}^{(1)}T_{j,0}^{(1)}T_{j,-1}^{(1)} - T_{i,0}^{(1)}T_{i,-1}^{(1)}T_{j,0}^{(1)}T_{j,1}^{(1)} + T_{i,0}^{(1)}T_{i,0}^{(1)}T_{j,0}^{(1)}T_{j,0}^{(1)}) = H_K \end{aligned} \tag{4}$$

so, $H_K = \sum_{\langle i,j\rangle} K_{i,j}(Q^{(0)}_{ij} - Q^{(1)}_{ij} + Q^{(2)}_{ij}) = \sum_{\langle i,j\rangle} K_{i,j}([T^{(0)}_i \times T^{(0)}_j]_0 + \sqrt{3}[T^{(1)}_i \times T^{(1)}_j]_0 + \sqrt{5}[T^{(2)}_i \times T^{(2)}_j]_0) = \sum_{\langle i,j\rangle} K_{i,j}(X^0_{0,ij}(0) + \sqrt{3}X^0_{0,ij}(1) + \sqrt{5}X^0_{0,ij}(2))$

A. Initialization

There are only one site in system and environment. However, there are two operators in one site. The product of two 1 rank irreducible tensor operators can lead to 0 rank, 1 rank and 2 rank irreducible tensor operators.We need to calculate $\langle j||K^{(0)}||j\rangle$,$\langle j||K^{(1)}||j\rangle$,$\langle j||K^{(2)}||j\rangle$ for initialization.

$T^{(L)}_{M,i} = \sum_{M_1} T^{(L1)}_{M_1,i} T^{(L2)}_{M-M_1,i}\langle L_1 M_1 L_2 (M-M_1)|LM\rangle$

$\langle jm|T^{(L)}_{M,i}|jm'\rangle = (-1)^{j-m}\begin{Bmatrix} j & L & j \\ -m & M & m' \end{Bmatrix}$

$\langle j||T^{(L)}||j\rangle$

for $\langle j||K^{(0)}||j\rangle$,

$T^{(0)}_{0,i} = T^{(1)}_{0,i}T^{(1)}_{0,i}\langle 1010|00\rangle + T^{(1)}_{1,i}T^{(1)}_{-1,i}\langle 111-1|00\rangle + T^{(1)}_{-1,i}T^{(1)}_{1,i}\langle 1-111|00\rangle$

$= -\frac{1}{\sqrt{3}}(T^{(1)}_{0,i}T^{(1)}_{0,i} - T^{(1)}_{1,i}T^{(1)}_{-1,i} - T^{(1)}_{-1,i}T^{(1)}_{1,i}) = -\frac{1}{\sqrt{3}}(\frac{1}{2}S^{\dagger}_i S^{-}_i + \frac{1}{2}S^{-}_i S^{\dagger}_i + S^z_i S^z_i) = -\frac{1}{\sqrt{3}}(j(j+1))$

$T^{(L)}_{M,i} = \sum_{M_1} T^{(L1)}_{M_1,i} T^{(L2)}_{M-M_1,i}\langle L_1 M_1 L_2 (M-M_1)|LM\rangle$

$$\langle jm|T^{(0)}_{0,i}|jm\rangle = (-1)^{j-m}\begin{Bmatrix} j & 0 & j \\ -m & 0 & m \end{Bmatrix}$$

$\langle j||T^{(L)}||j\rangle = \frac{1}{\sqrt{2j+1}}\langle j||T^{(L)}||j\rangle$

so, the $\langle j||K^{(0)}_i||j\rangle = -\frac{1}{\sqrt{3}}(j(j+1)\sqrt{2j+1}$

as the same way,

$\langle jm|T^{(1)}_{0,i}|jm\rangle = (-1)^{j-m}\begin{Bmatrix} j & 1 & j \\ -m & 0 & m \end{Bmatrix}\langle j||T^{(1)}||j\rangle$

$= (-1)^{j-m} \times (-1)^{j-m}\frac{m}{\sqrt{(2j+1)(j+1)j}}\langle j||T^{(1)}||j\rangle = \frac{m}{\sqrt{(2j+1)(j+1)j}}\langle j||T^{(1)}||j\rangle$

$T^{(1)}_{0,i} = \frac{1}{\sqrt{2}}T^{(1)}_{1,i}T^{(1)}_{-1,i} - \frac{1}{\sqrt{2}}T^{(1)}_{-1,i}T^{(1)}_{1,i} = \frac{1}{\sqrt{2}}(T^{(1)}_{1,i}T^{(1)}_{-1,i} - T^{(1)}_{-1,i}T^{(1)}_{1,i}) = \frac{1}{\sqrt{2}}(-\frac{1}{\sqrt{2}}S^{\dagger}_i\frac{1}{\sqrt{2}}S^{-}_i - \frac{1}{\sqrt{2}}S^{-}_i(-\frac{1}{\sqrt{2}}S^{\dagger}_i)) = -\frac{1}{\sqrt{2}}(\frac{1}{2}S^{\dagger}_i S^{-}_i - S^{-}_i S^{\dagger}_i) = -\frac{1}{\sqrt{2}}(\frac{1}{2}\times 2S^z_i) = -\frac{1}{\sqrt{2}}(S^z_i)$

$\langle jm|T^{(1)}_{0,i}|jm\rangle = \langle jm| - \frac{1}{\sqrt{2}}(S^z_i)|j,m\rangle = -\frac{1}{\sqrt{2}}m$

so, $\langle j||K^{(1)}_i||j\rangle = -\sqrt{\frac{j(j+1)(2j+1)}{2}}$.

for $\langle j||K^{(2)}_i||j\rangle$,

$\langle jm|T^{(2)}_{0,i}|jm\rangle = (-1)^{j-m}\begin{Bmatrix} j & 2 & j \\ -m & 0 & m \end{Bmatrix}\langle j||T^{(2)}||j\rangle$

$= (-1)^{j-m} \times (-1)^{j-m}\frac{3m^2-j(j+1)}{\sqrt{(2j+3)(2j+1)(j+1)j(2j-1)}}\langle j||T^{(2)}||j\rangle = \frac{3m^2-j(j+1)}{\sqrt{(2j+3)(2j+1)(j+1)j(2j-1)}}\langle j||T^{(2)}||j\rangle$

$T^{(2)}_{0,i} = \frac{1}{\sqrt{6}}T^{(1)}_{1,i}T^{(1)}_{-1,i} + \frac{1}{\sqrt{6}}T^{(1)}_{-1,i}T^{(1)}_{1,i} + \sqrt{\frac{2}{3}}T^{(1)}_{0,i}T^{(1)}_{0,i} = \frac{1}{\sqrt{6}}(-\frac{1}{\sqrt{2}}S^{\dagger}_i\frac{1}{\sqrt{2}}S^{-}_i + \frac{1}{\sqrt{2}}S^{-}_i(-\frac{1}{\sqrt{2}}S^{\dagger}_i)) + \sqrt{\frac{2}{3}}S^z_i S^z_i = -\frac{1}{\sqrt{6}}(S^x_i S^x_i + S^y_i S^y_i + S^z_i S^z_i) + \frac{3}{\sqrt{6}}S^z_i S^z_i = -\frac{1}{\sqrt{6}}(j(j+1) - 3S^z_i S^z_i)$

so, $\langle jm|T^{(2)}_{0,i}|jm\rangle = \langle jm| - \frac{1}{\sqrt{6}}(j(j+1) - 3S^z_i S^z_i)|jm\rangle = -\frac{1}{\sqrt{6}}(j(j+1) - 3m^2)$

so $\langle j||K^{(2)}_i||j\rangle = \sqrt{\frac{(2j-1)j(j+1)(2j+1)(2j+3)}{6}}$.

B. Increasing a new site

In this section, we add a new site in the system block. The irreducible basis is different from before. We need to calculate $\langle J'_{sys}, S', J', \alpha'||T^{(1)}_i||J_{sys}, S, J, \alpha\rangle$,$\langle J'_{sys}, S', J', \alpha'||T^{(0)}_i||J_{sys}, S, J, \alpha\rangle$,$\langle J'_{sys}, S', J', \alpha'||T^{(2)}_i||J_{sys}, S, J, \alpha\rangle$ in the new irreducible basis. There, we need the product of two irreducible tensor operators.

for $K = 0, 1, 2$

If the site i is in the system block, then

$\langle J'_{sys}, S', J', \alpha'||T^{(K)}_i||J_{sys}, S, J, \alpha\rangle = (-1)^{J'_{sys}+S'+J+K}\delta_{S',S}\sqrt{(2J'+1)(2J+1)}\begin{Bmatrix} J' & K & J \\ J_{sys} & S' & J'_{sys} \end{Bmatrix}\langle J'_{sys}, \alpha'||T^{(K)}_i||J_{sys}, \alpha\rangle$

if $J = J'$
$\langle J'_{sys}, S', J', \alpha' || T_i^{(K)} || J_{sys}, S, J, \alpha\rangle = (-1)^{J'_{sys}+S'+J+K}\delta_{S',S}(2J+1)\begin{Bmatrix} J & K & J \\ J_{sys} & S' & J'_{sys}\end{Bmatrix}\langle J'_{sys}, \alpha' || T_i^{(K)} || J_{sys}, \alpha\rangle$
$= (-1)^{J'_{sys}+S+J+K}(2J+1)\begin{Bmatrix} J & K & J \\ J_{sys} & S & J'_{sys}\end{Bmatrix}\langle J'_{sys}, \alpha' || T_i^{(K)} || J_{sys}, \alpha\rangle$
which would be non-zero only for $J \neq 0$. If $J = 0$, the $6j-$ coefficient would be zero.

If the site i is in the added new site, then
$\langle J'_{sys}, S', J', \alpha' || T_i^{(K)} || J_{sys}, S, J, \alpha\rangle = (-1)^{J_{sys}+S+J'+K}\delta_{J'_{sys},J_{sys}}\sqrt{(2J'+1)(2J+1)}\begin{Bmatrix} J' & K & J \\ S & J'_{sys} & S'\end{Bmatrix}\langle S', \alpha' || T_i^{(K)} || S, \alpha\rangle$
For the diagonal part with $J = J'$, the result can be simplified as
$\langle J'_{sys}, S', J', \alpha' || T_i^{(K)} || J_{sys}, S, J, \alpha\rangle = (-1)^{J_{sys}+S+J+K}\delta_{J'_{sys},J_{sys}}(2J+1)\begin{Bmatrix} J & K & J \\ S & J'_{sys} & S'\end{Bmatrix}\langle S', \alpha' || T_i^{(K)} || S, \alpha\rangle$
which would be non-zero only for $J \neq 0$. If $J = 0$, the $6j-$ coefficient would be zero.

C. Calculating the Hamiltonian

Suppose the site i belong to the system block and the site j is the added new site. Then the Hamiltonian $H_K = \sum_{\langle i,j\rangle} K_{i,j}(Q_{ij}^{(0)} - Q_{ij}^{(1)} + Q_{ij}^{(2)}) = \sum_{\langle i,j\rangle} K_{i,j}([T_i^{(0)} \times T_j^{(0)}]_0 + \sqrt{3}[T_i^{(1)} \times T_j^{(1)}]_0 + \sqrt{5}[T_i^{(2)} \times T_j^{(2)}]_0) = \sum_{\langle i,j\rangle} K_{i,j}(X_{0,ij}^0(0) + \sqrt{3}X_{0,ij}^0(1) + \sqrt{5}X_{0,ij}^0(2))$
The matrix element of the Hamiltonian H_K is
for $X_{0,ij}^0(k), k = 0, 1, 2,$
$K_{i,j}\langle J'_{sys}, S', J', \alpha' |(X_{0,ij}^0(k)| J_{sys}, S, J, \alpha\rangle = \frac{1}{\sqrt{2J+1}}K_{i,j}\langle J'_{sys}, S', J', \alpha' || X_{ij}^0(k) || J_{sys}, S, J, \alpha\rangle$
as $\langle J'_{sys}, S', J', \alpha' || X_{ij}^0(k) || J_{sys}, S, J, \alpha\rangle = \sqrt{\frac{2J+1}{2k+1}}(-1)^{J_{sys}+S'+k+J}\begin{Bmatrix} J'_{sys} & J_{sys} & k \\ S & S' & J\end{Bmatrix}\langle J'_{sys}, \alpha' || T_i^{(k)} || J_{sys}, \alpha\rangle\langle S' || T_j^{(k)} || S\rangle$
Thus, the matrix element of the Hamiltonian H_K is
$H_K =$
$K_{i,j}(\langle J'_{sys}, S', J', \alpha' |(X_{0,ij}^0(0)| J_{sys}, S, J, \alpha\rangle + \sqrt{3}\langle J'_{sys}, S', J', \alpha' |(X_{0,ij}^0(1)| J_{sys}, S, J, \alpha\rangle + \sqrt{5}\langle J'_{sys}, S', J', \alpha' |(X_{0,ij}^0(2)| J_{sys}, S, J, \alpha\rangle)$
$= (K_{i,j}\frac{1}{\sqrt{2k+1}}(-1)^{J_{sys}+S'+0+J}\begin{Bmatrix} J'_{sys} & J_{sys} & 0 \\ S & S' & J\end{Bmatrix}\langle J'_{sys}, \alpha' || T_i^{(0)} || J_{sys}, \alpha\rangle\langle S' || T_j^{(0)} || S\rangle)$
$+ (\sqrt{3}K_{i,j}\frac{1}{\sqrt{2k+1}}(-1)^{J_{sys}+S'+1+J}\begin{Bmatrix} J'_{sys} & J_{sys} & 1 \\ S & S' & J\end{Bmatrix}\langle J'_{sys}, \alpha' || T_i^{(1)} || J_{sys}, \alpha\rangle\langle S' || T_j^{(1)} || S\rangle)$
$+ (\sqrt{5}K_{i,j}\frac{1}{\sqrt{2k+1}}(-1)^{J_{sys}+S'+2+J}\begin{Bmatrix} J'_{sys} & J_{sys} & 2 \\ S & S' & J\end{Bmatrix}\langle J'_{sys}, \alpha' || T_i^{(2)} || J_{sys}, \alpha\rangle\langle S' || T_j^{(2)} || S\rangle)$
$= (K_{i,j}(-1)^{J_{sys}+S'+0+J}(-1)^{J_{sys}+S'+J}\frac{1}{\sqrt{(2S+1)(2J_{sys}+1)}}\langle J'_{sys}, \alpha' || T_i^{(0)} || J_{sys}, \alpha\rangle\langle S' || T_j^{(0)} || S\rangle)$
$+ (\sqrt{3}K_{i,j}\frac{1}{\sqrt{3}}(-1)^{J_{sys}+S'+1+J}\begin{Bmatrix} J'_{sys} & J_{sys} & 1 \\ S & S' & J\end{Bmatrix}\langle J'_{sys}, \alpha' || T_i^{(1)} || J_{sys}, \alpha\rangle\langle S' || T_j^{(1)} || S\rangle)$
$+ (\sqrt{5}K_{i,j}\frac{1}{\sqrt{5}}(-1)^{J_{sys}+S'+2+J}\begin{Bmatrix} J'_{sys} & J_{sys} & 2 \\ S & S' & J\end{Bmatrix}\langle J'_{sys}, \alpha' || T_i^{(2)} || J_{sys}, \alpha\rangle\langle S' || T_j^{(2)} || S\rangle)$
$= (K_{i,j}\frac{1}{\sqrt{(2S+1)(2J_{sys}+1)}}\langle J_{sys}, \alpha' || T_i^{(0)} || J_{sys}, \alpha\rangle\langle S || T_j^{(0)} || S\rangle)$
$+ (K_{i,j}(-1)^{J_{sys}+S+1+J}\begin{Bmatrix} J'_{sys} & J_{sys} & 1 \\ S & S & J\end{Bmatrix}\langle J'_{sys}, \alpha' || T_i^{(1)} || J_{sys}, \alpha\rangle\langle S || T_j^{(1)} || S\rangle)$
$+ (K_{i,j}(-1)^{J_{sys}+S+2+J}\begin{Bmatrix} J'_{sys} & J_{sys} & 2 \\ S & S & J\end{Bmatrix}\langle J'_{sys}, \alpha' || T_i^{(2)} || J_{sys}, \alpha\rangle\langle S || T_j^{(2)} || S\rangle)$

D. Hamiltonian from the K interactions

In this section, we need the matrix-vector multiplication calculations
$H_{n+1} = H_{A_n} + H_{B_n} + (S_{A_n} \cdot S_{A_n+1})^2 + (S_{A_n+1} \cdot S_{B_n+1})^2 + (S_{B_n+1} \cdot S_{B_n})^2$
$H_{n+1}|f\rangle = |g\rangle$, thus
$(H_{A_n} + H_{B_n} + (S_{A_n} \cdot S_{A_n+1})^2 + (S_{A_n+1} \cdot S_{B_n+1})^2 + (S_{B_n+1} \cdot S_{B_n})^2)|f\rangle = |g\rangle$
$(H_{A_n})|f\rangle = |g_1\rangle$, $(H_{B_n})|f\rangle = |g_2\rangle$, $(S_{A_n} \cdot S_{A_n+1})^2|f\rangle = |g_3\rangle$, $(S_{A_n+1} \cdot S_{B_n+1})^2|f\rangle = |g_4\rangle$, $(S_{B_n+1} \cdot S_{B_n})^2|f\rangle = |g_5\rangle$
thus, $|g\rangle = |g_1\rangle + |g_2\rangle + |g_3\rangle + |g_4\rangle + |g_5\rangle$

generally, $J_{ij}(\vec{S}_i\cdot\vec{S}_j)^2|f\rangle = J_{ij}(\vec{S}_i\cdot\vec{S}_j)^2|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J\rangle f(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J)$
$= \sum_{J'_{sys},J'_{ns},J'_{sysnew},J'_{env},J'_{ne},J'_{envnew},J'} |J'_{sys},J'_{ns},J'_{sysnew},J'_{env},J'_{ne},J'_{envnew},J'\rangle\langle J'_{sys},J'_{ns},J'_{sysnew},J'_{env},J'_{ne},J'_{envnew},J'|$
$J_{ij}(\vec{S}_i\cdot\vec{S}_j)^2|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J\rangle f(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J)$
$= \sum_{J'_{sys},J'_{ns},J'_{sysnew},J'_{env},J'_{ne},J'_{envnew},J'} \langle J'_{sys},J'_{ns},J'_{sysnew},J'_{env},J'_{ne},J'_{envnew},J'|J_{ij}(\vec{S}_i\cdot\vec{S}_j)^2|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J\rangle$
$f(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J)|J'_{sys},J'_{ns},J'_{sysnew},J'_{env},J'_{ne},J'_{envnew},J'\rangle$
$= g(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J)|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J\rangle$

1. Hamiltonian from the K interactions between the system block and the added sites

The Hamiltonian contribution is $K_{ij}\vec{(}S_i\cdot\vec{S}_j)^2$, where $\vec{S}_i$ and $\vec{S}_j$ belong to the system block and the added site, respectively. The matrix-vector multiplication is explicitly written as
$\langle J'_{sys},J'_{ns},J'_{sysnew},J'_{env},J'_{ne},J'_{envnew},J'|K_{ij}(\vec{S}_i\cdot\vec{S}_j)^2|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J\rangle$
$f(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J) = g(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J)$
As $\vec{S}_i$ and $\vec{S}_j$ belong to the system block and the added site, respectively. Thus
$\langle J'_{sys},J_{ns},J'_{sysnew},J_{env},J_{ne},J_{envnew},0|K_{ij}(\vec{S}_i\cdot\vec{S}_j)^2|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$f(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0) = g(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0)$
We can compute the matrix element as
$K_{ij}\langle J'_{sys},J_{ns},J'_{sysnew},J_{env},J_{ne},J_{envnew},0|(\vec{S}_i\cdot\vec{S}_j)^2|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$= \beta K_{ij}\langle J'_{sys},J_{ns},J'_{sysnew},J_{env},J_{ne},J_{envnew},0|X^0_{0,ij}(k)|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$= \beta K_{ij}\frac{1}{\sqrt{2J_{sysnew}+1}}\langle J'_{sys},J_{ns},J'_{sysnew},J_{env},J_{ne},J_{envnew},0||X^0_{ij}(k)||J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$= \beta K_{ij}\frac{1}{\sqrt{2J_{sysnew}+1}}\frac{\sqrt{2J_{sysnew}+1}}{\sqrt{2k+1}}(-1)^{J_{sys}+J_{ns}+k+J_{sysnew}}\begin{Bmatrix} J'_{sys} & J_{sys} & k \\ J_{ns} & J_{ns} & J_{sysnew}\end{Bmatrix}\langle J'_{sys},\alpha'||T_i^{(k)}||J_{sys},\alpha\rangle\langle J_{ns}||T_j^{(k)}||J_{ns}\rangle$
$= \beta K_{ij}\frac{1}{\sqrt{2k+1}}(-1)^{J_{sys}+J_{ns}+k+J_{sysnew}}\begin{Bmatrix} J'_{sys} & J_{sys} & k \\ J_{ns} & J_{ns} & J_{sysnew}\end{Bmatrix}\langle J'_{sys},\alpha'||T_i^{(k)}||J_{sys},\alpha\rangle\langle J_{ns}||T_j^{(k)}||J_{ns}\rangle$
If $J_{sys} = J'_{sys}$, then $J_{sys} \neq 0$

IV. IRREDUCIBLE TENSOR OPERATORS OF THE CHIRAL TERMS

A. J_χ term

For $i<j<k$, the sites follow the clockwise direction.

$$\begin{aligned}
H=\sum_{\triangle} J_\chi(\mathbf{S}_i\times\mathbf{S}_j)\cdot\mathbf{S}_k &= \sum_{\triangle} J_\chi((S_i^yS_j^z-S_i^zS_j^y)\hat{x})+(S_i^zS_j^x-S_i^xS_j^z)\hat{y}+(S_i^xS_j^y-S_i^yS_j^x)\hat{z})\cdot(S_k^x\hat{x}+S_k^y\hat{y}+S_k^z\hat{z})\\
&=\sum_{\triangle} J_\chi((S_i^yS_j^z-S_i^zS_j^y)S_k^x+(S_i^zS_j^x-S_i^xS_j^z)S_k^y+(S_i^xS_j^y-S_i^yS_j^x)S_k^z)\\
&=\sum_{\triangle} J_\chi((S_i^yS_j^zS_k^x-S_i^zS_j^yS_k^x)+(S_i^zS_j^xS_k^y-S_i^xS_j^zS_k^y)+(S_i^xS_j^yS_k^z-S_i^yS_j^xS_k^z))\\
&=\sum_{\triangle} J_\chi((1/2i(S_i^\dagger-S_i^-)\cdot 1/2(S_k^\dagger+S_k^-))S_j^z-(1/2i(S_j^\dagger-S_j^-)\cdot 1/2(S_k^\dagger+S_k^-))S_i^z)\\
&\quad+((1/2(S_j^\dagger+S_j^-)\cdot 1/2i(S_k^\dagger-S_k^-))S_i^z-(1/2(S_i^\dagger+S_i^-)\cdot 1/2i(S_k^\dagger-S_k^-))S_j^z)\\
&\quad+((1/2(S_i^\dagger+S_i^-)\cdot 1/2i(S_j^\dagger-S_j^-))S_i^k-(1/2i(S_i^\dagger-S_i^-)\cdot 1/2(S_j^\dagger+S_j^-))S_k^z)\\
&+\sum_{\triangle} J_\chi(1/4i(S_i^\dagger S_k^\dagger+S_i^\dagger S_k^--S_i^-S_k^\dagger-S_i^-S_k^-)S_j^z-(S_j^\dagger S_k^\dagger+S_j^\dagger S_k^--S_j^-S_k^\dagger-S_j^-S_k^-)S_i^z)\\
&\quad+(1/4i(S_j^\dagger S_k^\dagger-S_j^\dagger S_k^-+S_j^-S_k^\dagger-S_j^-S_k^-)S_i^z-(S_i^\dagger S_k^\dagger-S_i^\dagger S_k^-+S_i^-S_k^\dagger-S_i^-S_k^-)S_j^z)\\
&\quad+(1/4i(S_i^\dagger S_j^\dagger-S_i^\dagger S_j^-+S_i^-S_j^\dagger-S_i^-S_j^-)S_i^k-(S_i^\dagger S_j^\dagger+S_i^\dagger S_j^--S_i^-S_j^\dagger-S_i^-S_j^-)S_k^z)\\
&=1/2i((S_i^\dagger S_k^--S_i^-S_k^\dagger)S_j^z+(-S_j^\dagger S_k^-+S_j^-S_k^\dagger)S_i^z+(-S_i^\dagger S_j^-+S_i^-S_j^\dagger)S_k^z)\\
&=1/2i(S_i^\dagger S_j^zS_k^--S_i^-S_j^zS_k^\dagger-S_i^zS_j^\dagger S_k^-+S_i^zS_j^-S_k^\dagger-S_i^\dagger S_j^-S_k^z+S_i^-S_j^\dagger S_k^z)
\end{aligned}\tag{5}$$

so,
$H=\sum_\triangle(J_{\chi,ijk})(i)(T^{(1)}_{1,i}T^{(1)}_{0,j}T^{(1)}_{-1,k}-T^{(1)}_{-1,i}T^{(1)}_{0,j}T^{(1)}_{1,k}-T^{(1)}_{0,i}T^{(1)}_{1,j}T^{(1)}_{-1,k}+T^{(1)}_{0,i}T^{(1)}_{-1,j}T^{(1)}_{1,k}-T^{(1)}_{1,i}T^{(1)}_{-1,j}T^{(1)}_{0,k}+T^{(1)}_{-1,i}T^{(1)}_{1,j}T^{(1)}_{0,k})$

For $i>j>k (k<j<i)$, the sites follow the clockwise direction(another situation).

$$\begin{aligned}
H=\sum_{\triangle} J_\chi(\mathbf{S}_k\times\mathbf{S}_j)\cdot\mathbf{S}_i &= \sum_{\triangle} J_\chi((S_k^yS_j^z-S_k^zS_j^y)\hat{x})+(S_k^zS_j^x-S_k^xS_j^z)\hat{y}+(S_k^xS_j^y-S_k^yS_j^x)\hat{z})\cdot(S_i^x\hat{x}+S_i^y\hat{y}+S_i^z\hat{z})\\
&=\sum_{\triangle} J_\chi((S_k^yS_j^z-S_k^zS_j^y)S_i^x+(S_k^zS_j^x-S_k^xS_j^z)S_i^y+(S_k^xS_j^y-S_k^yS_j^x)S_i^z)\\
&=\sum_{\triangle} J_\chi((S_k^yS_j^zS_i^x-S_k^zS_j^yS_i^x)+(S_k^zS_j^xS_i^y-S_k^xS_j^zS_i^y)+(S_k^xS_j^yS_i^z-S_k^yS_j^xS_i^z))\\
=\sum_{\triangle}(J_{\chi,kji})(i)(T^{(1)}_{1,k}T^{(1)}_{0,j}T^{(1)}_{-1,i}&-T^{(1)}_{-1,k}T^{(1)}_{0,j}T^{(1)}_{1,i}-T^{(1)}_{0,k}T^{(1)}_{1,j}T^{(1)}_{-1,i}+T^{(1)}_{0,k}T^{(1)}_{-1,j}T^{(1)}_{1,i}-T^{(1)}_{1,k}T^{(1)}_{-1,j}T^{(1)}_{0,i}+T^{(1)}_{-1,k}T^{(1)}_{1,j}T^{(1)}_{0,i})\\
&=-\sum_{\triangle} J_\chi(\mathbf{S}_i\times\mathbf{S}_j)\cdot\mathbf{S}_k
\end{aligned}\tag{6}$$

The two rank-1 ITO on two sites respectively can be coupled as one rank-1 ITO.
$T_0^{(1)}=\langle 1,1,1,-1|1,0\rangle T_1^{(1)}T_{-1}^{(1)}+\langle 1,-1,1,1|1,0\rangle T_{-1}^{(1)}T_1^{(1)}+\langle 1,0,1,0|1,0\rangle T_0^{(1)}T_0^{(1)}=\sqrt{\tfrac12}T_1^{(1)}T_{-1}^{(1)}-\sqrt{\tfrac12}T_{-1}^{(1)}T_1^{(1)}$
$T_1^{(1)}=\langle 1,1,1,0|1,1\rangle T_1^{(1)}T_0^{(1)}+\langle 1,0,1,1|1,1\rangle T_0^{(1)}T_1^{(1)}+\langle 1,-1,1,2|1,1\rangle T_{-1}^{(1)}T_2^{(1)}=\sqrt{\tfrac12}T_1^{(1)}T_0^{(1)}-\sqrt{\tfrac12}T_0^{(1)}T_1^{(1)}$
$T_{-1}^{(1)}=\langle 1,1,1,-2|1,-1\rangle T_1^{(1)}T_{-2}^{(1)}+\langle 1,0,1,-1|1,-1\rangle T_0^{(1)}T_{-1}^{(1)}+\langle 1,-1,1,0|1,-1\rangle T_{-1}^{(1)}T_0^{(1)}=\sqrt{\tfrac12}T_0^{(1)}T_{-1}^{(1)}-\sqrt{\tfrac12}T_{-1}^{(1)}T_0^{(1)}$

For $i<j<k$. The new rank-1 ITO can be coupled to one rank-0 ITO with another rank-1 ITO on the third site.
$X_0^{(0)}=\langle 1,1,1,-1|0,0\rangle T_1^{(1)}T_{-1}^{(1)}+\langle 1,0,1,0|0,0\rangle T_0^{(1)}T_0^{(1)}+\langle 1,-1,1,1|0,0\rangle T_{-1}^{(1)}T_1^{(1)}$

$= \sqrt{\frac{1}{3}}T_1^{(1)}T_{-1}^{(1)} - \sqrt{\frac{1}{3}}T_0^{(1)}T_0^{(1)} + \sqrt{\frac{1}{3}}T_{-1}^{(1)}T_1^{(1)}$
$= \sqrt{\frac{1}{3}}(\sqrt{\frac{1}{2}}T_1^{(1)}T_0^{(1)} - \sqrt{\frac{1}{2}}T_0^{(1)}T_1^{(1)})T_{-1}^{(1)} - \sqrt{\frac{1}{3}}(\sqrt{\frac{1}{2}}T_1^{(1)}T_{-1}^{(1)} - \sqrt{\frac{1}{2}}T_{-1}^{(1)}T_1^{(1)})T_0^{(1)} + \sqrt{\frac{1}{3}}(\sqrt{\frac{1}{2}}T_0^{(1)}T_{-1}^{(1)} - \sqrt{\frac{1}{2}}T_{-1}^{(1)}T_0^{(1)})T_1^{(1)}$
$= \sqrt{\frac{1}{6}}T_1^{(1)}T_0^{(1)}T_{-1}^{(1)} - \sqrt{\frac{1}{6}}T_0^{(1)}T_1^{(1)}T_{-1}^{(1)} - \sqrt{\frac{1}{6}}T_1^{(1)}T_{-1}^{(1)}T_0^{(1)} + \sqrt{\frac{1}{6}}T_{-1}^{(1)}T_1^{(1)}T_0^{(1)} + \sqrt{\frac{1}{6}}T_0^{(1)}T_{-1}^{(1)}T_1^{(1)} - \sqrt{\frac{1}{6}}T_{-1}^{(1)}T_0^{(1)}T_1^{(1)}$

since, $([T_i^{(1)} \times T_j^{(1)}]_{10} \times T_{0,k}^{(1)} + [T_i^{(1)} \times T_j^{(1)}]_{11} \times T_{(-1),k}^{(1)} + [T_i^{(1)} \times T_j^{(1)}]_{1-1} \times T_{(1),k}^{(1)}) = X_{ijk}^0 = \frac{1}{\sqrt{6}}(T_{1,i}^{(1)}T_{0,j}^{(1)}T_{-1,k}^{(1)} - T_{-1,i}^{(1)}T_{0,j}^{(1)}T_{1,k}^{(1)} - T_{0,i}^{(1)}T_{1,j}^{(1)}T_{-1,k}^{(1)} + T_{0,i}^{(1)}T_{-1,j}^{(1)}T_{1,k}^{(1)} - T_{1,i}^{(1)}T_{-1,j}^{(1)}T_{0,k}^{(1)} + T_{-1,i}^{(1)}T_{1,j}^{(1)}T_{0,k}^{(1)})$,
so,$H = \sum_\triangle J_\chi(\mathbf{S}_i \times \mathbf{S}_j)\cdot\mathbf{S}_k = \sum_\triangle (J_{\chi,ijk})(i)\sqrt{6}X_{ijk}^0$

For $i > j > k (k < j < i)$. $([T_k^{(1)} \times T_j^{(1)}]_{10} \times T_{0,i}^{(1)} + [T_k^{(1)} \times T_j^{(1)}]_{11} \times T_{(-1),i}^{(1)} + [T_k^{(1)} \times T_j^{(1)}]_{1-1} \times T_{(1),i}^{(1)}) = X_{kji}^0 = \frac{1}{\sqrt{6}}(T_{1,k}^{(1)}T_{0,j}^{(1)}T_{-1,i}^{(1)} - T_{-1,k}^{(1)}T_{0,j}^{(1)}T_{1,i}^{(1)} - T_{0,k}^{(1)}T_{1,j}^{(1)}T_{-1,i}^{(1)} + T_{0,k}^{(1)}T_{-1,j}^{(1)}T_{1,i}^{(1)} - T_{1,k}^{(1)}T_{-1,j}^{(1)}T_{0,i}^{(1)} + T_{-1,k}^{(1)}T_{1,j}^{(1)}T_{0,i}^{(1)})$
so,$H = \sum_\triangle J_\chi(\mathbf{S}_k \times \mathbf{S}_j)\cdot\mathbf{S}_i = \sum_\triangle (J_{\chi,kji})(i)\sqrt{6}X_{kji}^0 = -\sum_\triangle (J_{\chi,ijk})(i)\sqrt{6}X_{ijk}^0$

B. Initialization

There are only one site in system and environment. As the Heisenberg terms. We need to calculate $\langle J||T^{(1)}||J\rangle$ for initialization.
$\langle JJ|T_0^{(1)}|JJ\rangle = \frac{1}{\sqrt{2J+1}}\langle JJ10|JJ\rangle\langle J||T^{(1)}||J\rangle = \frac{1}{\sqrt{2J+1}}\frac{J}{\sqrt{J(J+1)}}\langle J||T^{(1)}||J\rangle = J$
Thus, $\langle J||T^{(1)}||J\rangle = \sqrt{J(J+1)(2J+1)}$.
If $J = \frac{1}{2}$, $\langle J||T^{(1)}||J\rangle = \sqrt{\frac{3}{2}}$.
If $J = 1$, $\langle J||T^{(1)}||J\rangle = \sqrt{6}$.

C. Increasing a new site

As the J term, we add a new site in the system block. The irreducible basis is different from before. We need to calculate $\langle J'_{sys}, S', J', \alpha'||T_i^{(1)}||J_{sys}, S, J, \alpha\rangle$ in the new irreducible basis. There, we need the product of two irreducible tensor operators. There, $|J - J'| = 0, 1$.
If the site i is in the system block, then
$\langle J'_{sys}, S', J', \alpha'||T_i^{(1)}||J_{sys}, S, J, \alpha\rangle = (-1)^{J'_{sys}+S'+J+1}\delta_{S',S}\sqrt{(2J'+1)(2J+1)}\begin{Bmatrix} J' & 1 & J \\ J_{sys} & S' & J'_{sys}\end{Bmatrix}\langle J'_{sys}, \alpha'||T_i^{(1)}||J_{sys}, \alpha\rangle$
if $J = J'$
$\langle J'_{sys}, S', J', \alpha'||T_i^{(1)}||J_{sys}, S, J, \alpha\rangle = (-1)^{J'_{sys}+S'+J+1}\delta_{S',S}(2J+1)\begin{Bmatrix} J & 1 & J \\ J_{sys} & S' & J'_{sys}\end{Bmatrix}\langle J'_{sys}, \alpha'||T_i^{(1)}||J_{sys}, \alpha\rangle$
$= (-1)^{J'_{sys}+S+J+1}(2J+1)\begin{Bmatrix} J & 1 & J \\ J_{sys} & S & J'_{sys}\end{Bmatrix}\langle J'_{sys}, \alpha'||T_i^{(1)}||J_{sys}, \alpha\rangle$
which would be non-zero only for $J \neq 0$. If $J = 0$, the $6j-$ coefficient would be zero.
If the site i is in the added new site, then
$\langle J'_{sys}, S', J', \alpha'||T_i^{(1)}||J_{sys}, S, J, \alpha\rangle = (-1)^{J_{sys}+S+J'+1}\delta_{J'_{sys},J_{sys}}\sqrt{(2J'+1)(2J+1)}\begin{Bmatrix} J' & 1 & J \\ S & J'_{sys} & S'\end{Bmatrix}\langle S', \alpha'||T_i^{(1)}||S, \alpha\rangle$
For the diagonal part with $J = J'$, the result can be simplified as
$\langle J'_{sys}, S', J', \alpha'||T_i^{(1)}||J_{sys}, S, J, \alpha\rangle = (-1)^{J_{sys}+S+J+1}\delta_{J'_{sys},J_{sys}}(2J+1)\begin{Bmatrix} J & 1 & J \\ S & J'_{sys} & S'\end{Bmatrix}\langle S', \alpha'||T_i^{(1)}||S, \alpha\rangle$
which would be non-zero only for $J \neq 0$. If $J = 0$, the $6j-$ coefficient would be zero.

D. Calculating the Hamiltonian

Suppose the site i, j belong to the system block and the site k is the added new site. As $i < j < k$. Then the Hamiltonian H_{J_χ} is
$H_{J_\chi} = \sum_\triangle J_\chi(\mathbf{S}_i \times \mathbf{S}_j)\cdot\mathbf{S}_k = \sum_\triangle (J_{\chi,ijk})(i)\sqrt{6}X_{ijk}^0$
The matrix element of the Hamiltonian H_{J_χ} is

$(J_{\chi,ijk})(i)\sqrt{6}\langle J'_{sys},S',J',\alpha'|X^0_{0,ijk}|J_{sys},S,J,\alpha\rangle = (J_{\chi,ijk})(i)\sqrt{6}\frac{1}{\sqrt{2J+1}}\langle J'_{sys},S',J',\alpha'||X^0_{ijk}||J_{sys},S,J,\alpha\rangle$
as $\langle J'_{sys},S',J',\alpha'||X^0_{ij}||J_{sys},S,J,\alpha\rangle = \sqrt{\frac{2J+1}{3}}(-1)^{J_{sys}+S'+1+J}\begin{Bmatrix} J'_{sys} & J_{sys} & 1 \\ S & S' & J \end{Bmatrix}\langle J'_{sys},\alpha'||T^{(1)}_i||J_{sys},\alpha\rangle\langle S'||T^{(1)}_j||S\rangle$
Thus, the matrix element of the Hamiltonian H_{J_χ} is
$(J_{\chi,ijk})(i)\sqrt{6}\langle J'_{sys},S',J',\alpha'|X^0_{0,ij}|J_{sys},S,J,\alpha\rangle = \sqrt{2}(J_{\chi,ijk})(i)(-1)^{J_{sys}+S'+1+J}\begin{Bmatrix} J'_{sys} & J_{sys} & 1 \\ S & S' & J \end{Bmatrix}$
$\langle J'_{sys},\alpha'||T^{(1)}_i||J_{sys},\alpha\rangle\langle S'||T^{(1)}_j||S\rangle$

Suppose the site i belong to the system block and the site j is the added new site. As $k<j<i$. Then
$\langle J'_{sys},S',J',\alpha'||X^{(1)}_{ij}||J_{sys},S,J,\alpha\rangle =$
$\sqrt{(2J'+1)(2K+1)(2J+1)}\begin{Bmatrix} J'_{sys} & J_{sys} & 1 \\ S' & S & 1 \\ J' & J & 1 \end{Bmatrix}\langle J'_{sys},\alpha'||T^{(1)}_i||J_{sys},\alpha\rangle\langle S'||T^{(1)}_j||S\rangle(k=1)$

E. Hamiltonian from the chiral interactions

In this section, we need the matrix-vector multiplication calculations
$H=\sum_\triangle J_\chi(\mathbf{S}_i\times\mathbf{S}_j)\cdot\mathbf{S}_k = \sum_\triangle (J_{\chi,ijk})(i)\sqrt{6}X^0_{ijk}$
$H_{n+1}|f\rangle = |g\rangle$, thus
$(H_{A_n,ijk}+H_{B_n,ijk}+S_{A_n,ij}\cdot S_{A_n+1,k}+S_{B_n+1,i}\cdot S_{B_n,jk})|f\rangle = |g\rangle$
$(H_{A_n,ijk})|f\rangle = |g_1\rangle$, $(H_{B_n,ijk})|f\rangle = |g_2\rangle$, $(S_{A_n,ij}\cdot S_{A_n+1,k})|f\rangle = |g_3\rangle$, $(S_{B_n+1,i}\cdot S_{B_n,jk})|f\rangle = |g_4\rangle$
thus, $|g\rangle = |g_1\rangle+|g_2\rangle+|g_3\rangle+|g_4\rangle$
generally, $X^0_{ijk}|f\rangle = X^0_{ijk}|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J\rangle f(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J)$
$=\sum_{J'_{sys},J'_{ns},J'_{sysnew},J'_{env},J'_{ne},J'_{envnew},J'}|J'_{sys},J'_{ns},J'_{sysnew},J'_{env},J'_{ne},J'_{envnew},J'\rangle\langle J'_{sys},J'_{ns},J'_{sysnew},J'_{env},J'_{ne},J'_{envnew},J'|$
$X^0_{ijk}|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J\rangle f(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J)$
$=\sum_{J'_{sys},J'_{ns},J'_{sysnew},J'_{env},J'_{ne},J'_{envnew},J'}\langle J'_{sys},J'_{ns},J'_{sysnew},J'_{env},J'_{ne},J'_{envnew},J'|X^0_{ijk}|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J\rangle$
$f(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J)|J'_{sys},J'_{ns},J'_{sysnew},J'_{env},J'_{ne},J'_{envnew},J'\rangle$
$=g(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J)|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},J\rangle$

1. *Hamiltonian from the chiral interactions between the system block and the added sites*

As $T^{(1)}_{ij}$ and $T^{(1)}_k$ belong to the system block and the added site, respectively. Thus
$\langle J'_{sys},J_{ns},J'_{sysnew},J_{env},J_{ne},J_{envnew},0|(J_{\chi,ijk})(i)\sqrt{6}X^0_{ijk}|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$f(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0) = g(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0)$
We can compute the matrix element as
$\langle J'_{sys},J_{ns},J'_{sysnew},J_{env},J_{ne},J_{envnew},0|(J_{\chi,ijk})(i)\sqrt{6}X^0_{ijk}|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$=(J_{\chi,ijk})(i)\sqrt{6}\langle J'_{sys},J_{ns},J'_{sysnew},J_{env},J_{ne},J_{envnew},0|X^0_{0,ijk}|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$=(J_{\chi,ijk})(i)\sqrt{6}\sqrt{\frac{1}{2J_{sysnew}+1}}\langle J'_{sys},J_{ns},J'_{sysnew},J_{env},J_{ne},J_{envnew},0||X^0_{ijk}||J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$=(J_{\chi,ijk})(i)\sqrt{6}\sqrt{\frac{1}{2J_{sysnew}+1}}\frac{\sqrt{2J_{sysnew}+1}}{\sqrt{3}}(-1)^{J_{sys}+J_{ns}+1+J_{sysnew}}\begin{Bmatrix} J'_{sys} & J_{sys} & 1 \\ J_{ns} & J_{ns} & J_{sysnew} \end{Bmatrix}\langle J'_{sys},\alpha'||T^{(1)}_{ij}||J_{sys},\alpha\rangle\langle J_{ns}||T^{(1)}_k||J_{ns}\rangle$
$=(J_{\chi,ijk})(i)\sqrt{2}(-1)^{J_{sys}+J_{ns}+1+J_{sysnew}}\begin{Bmatrix} J'_{sys} & J_{sys} & 1 \\ J_{ns} & J_{ns} & J_{sysnew} \end{Bmatrix}\langle J'_{sys},\alpha'||T^{(1)}_{ij}||J_{sys},\alpha\rangle\langle J_{ns}||T^{(1)}_k||J_{ns}\rangle$
If $J_{sys}=J'_{sys}$, then $J_{sys}\neq 0$

2. *Hamiltonian from the chiral interactions between the system block and the added sites (N_s and N_e)*

As $T^{(1)}_i$, $T^{(1)}_j$ and $T^{(1)}_k$ belong to the system block and the added sites (N_s and N_e), respectively. Thus
$\langle J'_{sys},J_{ns},J'_{sysnew},J_{env},J_{ne},J_{envnew},0|(J_{\chi,ijk})(i)\sqrt{6}X^0_{ijk}|J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0\rangle$
$f(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0) = g(J_{sys},J_{ns},J_{sysnew},J_{env},J_{ne},J_{envnew},0)$
We can compute the matrix element as

$$
\begin{aligned}
&\langle J'_{sys}, J_{ns}, J'_{sysnew}, J_{env}, J_{ne}, J_{envnew}, 0|(J_{\chi,ijk})(i)\sqrt{6}X^0_{ijk}|J_{sys}, J_{ns}, J_{sysnew}, J_{env}, J_{ne}, J_{envnew}, 0\rangle \\
&= (J_{\chi,ijk})(i)\sqrt{6}\langle J'_{sys}, J_{ns}, J'_{sysnew}, J_{env}, J_{ne}, J_{envnew}, 0|X^0_{0,ijk}|J_{sys}, J_{ns}, J_{sysnew}, J_{env}, J_{ne}, J_{envnew}, 0\rangle \\
&= (J_{\chi,ijk})(i)\sqrt{6}\sqrt{\tfrac{1}{2J+1}}\langle J'_{sys}, J_{ns}, J'_{sysnew}, J_{env}, J_{ne}, J_{envnew}, 0||X^0_{ijk}||J_{sys}, J_{ns}, J_{sysnew}, J_{env}, J_{ne}, J_{envnew}, 0\rangle \\
&= (J_{\chi,ijk})(i)\sqrt{6}\sqrt{\tfrac{1}{2J+1}}\tfrac{\sqrt{2J+1}}{\sqrt{3}}(-1)^{J_{sysnew}+J_{envnew}+1+J} \\
&\begin{Bmatrix} J'_{sysnew} & J_{sysnew} & 1 \\ J_{envnew} & J'_{envnew} & J \end{Bmatrix}\langle J'_{sysnew}, \alpha'||T^{(1)}_{ij}||J_{sysnew}, \alpha\rangle\langle J_{envnew}||T^{(1)}_{k}||J_{envnew}\rangle \\
&= (J_{\chi,ijk})(i)\sqrt{2}(-1)^{J_{sysnew}+J_{envnew}+1+J}\begin{Bmatrix} J'_{sysnew} & J_{sysnew} & 1 \\ J_{envnew} & J'_{envnew} & J \end{Bmatrix}\langle J'_{sysnew}, \alpha'||T^{(1)}_{ij}||J_{sysnew}, \alpha\rangle\langle J_{envnew}||T^{(1)}_{k}||J_{envnew}\rangle \\
&= (J_{\chi,ijk})(i)\sqrt{2}(-1)^{J_{sysnew}+J_{envnew}+1+J}\begin{Bmatrix} J'_{sysnew} & J_{sysnew} & 1 \\ J_{envnew} & J'_{envnew} & J \end{Bmatrix} \\
&\sqrt{(2J'_{sysnew}+1)(2K+1)(2J_{sysnew}+1)}\begin{Bmatrix} J'_{sys} & J_{sys} & 1 \\ S' & S & 1 \\ J'_{sysnew} & J_{sysnew} & 1 \end{Bmatrix}\langle J'_{sys}, \alpha'||T^{(1)}_{i}||J_{sys}, \alpha\rangle\langle S'||T^{(1)}_{j}||S\rangle\langle J_{envnew}||T^{(1)}_{k}||J_{envnew}\rangle \\
&= (J_{\chi,ijk})(i)\sqrt{2}(-1)^{J_{sysnew}+J_{envnew}+1+J}\begin{Bmatrix} J'_{sysnew} & J_{sysnew} & 1 \\ J_{envnew} & J'_{envnew} & J \end{Bmatrix} \\
&\sqrt{(2J'_{sysnew}+1)(2K+1)(2J_{sysnew}+1)}\begin{Bmatrix} J'_{sys} & J_{sys} & 1 \\ S' & S & 1 \\ J'_{sysnew} & J_{sysnew} & 1 \end{Bmatrix}\langle J'_{sys}, \alpha'||T^{(1)}_{i}||J_{sys}, \alpha\rangle\langle S'||T^{(1)}_{j}||S\rangle \\
&(-1)^{J_{env}+S+J_{envnew}+1}\sqrt{(2J'_{envnew}+1)(2J_{envnew}+1)}\begin{Bmatrix} J'_{envnew} & 1 & J_{envnew} \\ S & J'_{env} & S' \end{Bmatrix}\langle S', \alpha'||T^{(1)}_{i}||S, \alpha\rangle
\end{aligned}
$$