



ONE DIMENSIONAL SPIN CHAINS ON SURFACES

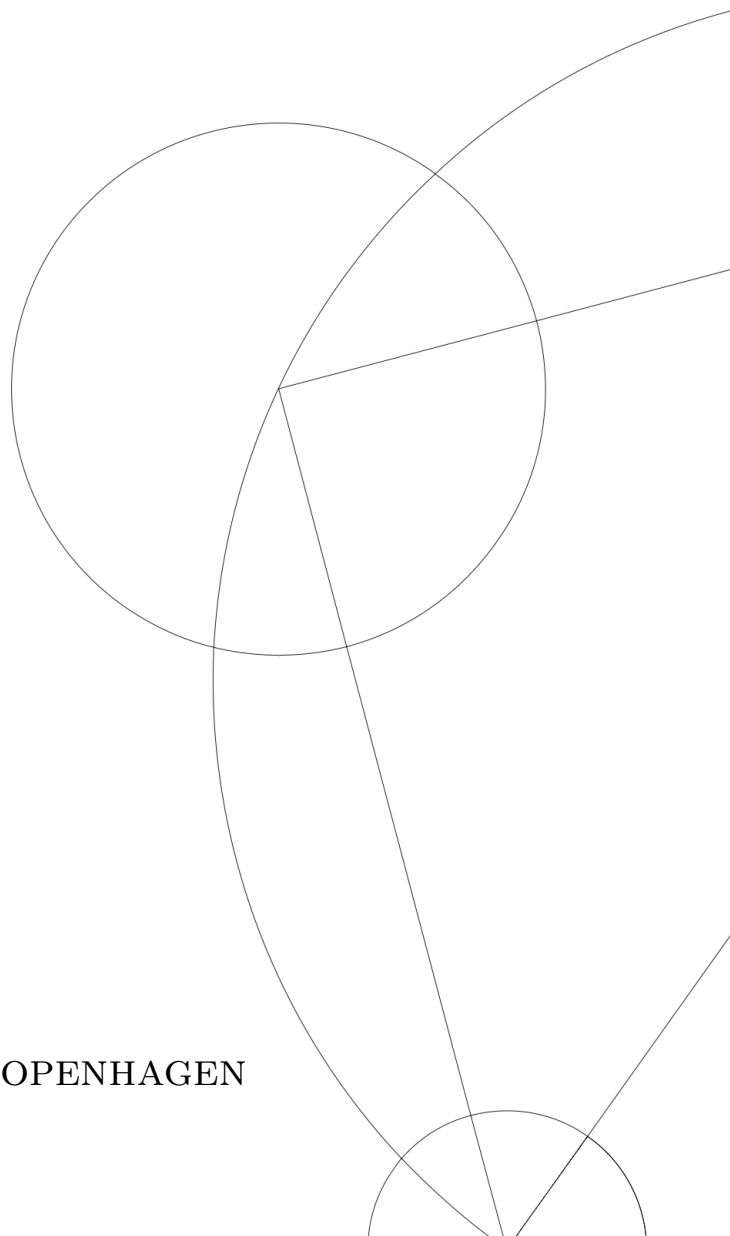
BACHELOR'S THESIS

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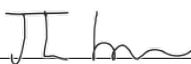




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Abstract

Describing and predicting some quantum systems can be difficult due to the sheer size of the system, especially when describing systems of higher spin atoms. This creates a need for a simplified description of such systems. This thesis explores one such system, by applying the Schrieffer Wolff transformation up to the second order [1] to a one dimensional spin chain of cobalt atoms with nearest neighbor antiferromagnetic isotropic Heisenberg exchange, an external transverse magnetic field, and an anisotropic spin coupling to the surface. The transformation results in a description of this system in a low energy setting as an effective spin-1/2 XXZ exchange spin chain with nearest and next nearest coupling in an external magnetic field. Additionally we demonstrate that in the third order of the transformation a third nearest neighbor coupling emerges.

Finally the last part of the thesis explores the energy predictions of spin chains of length 2, 3 and 4 by the original and transformed models, and discover the accuracy of the transformation to be best for excitation energies rather than direct state energy predictions.

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1 Introduction

Many textbook quantum systems have been entirely theoretical for a long time, but the ability to arrange and manipulate individual atoms has made it possible to realize a large portion of these in experimental settings [2]. Furthermore this ability becomes key in the creation of quantum computers and spintronic devices [3].

In order to apply and use these systems we need methods to predict their behaviors, which for some systems means simplifying their models for simpler computational systems. This thesis explores one such system, specifically a one dimensional spin chain of cobalt atoms with nearest neighbor antiferromagnetic isotropic Heisenberg exchange, an external transverse magnetic field, and an anisotropic spin coupling to the surface. Using the Schrieffer Wolff transformation we will try to simplify this system of spin-3/2 atoms, in order to describe it using a more well known model for the spin-1/2 XXZ Heisenberg chains in a transverse field.

Additionally we explore the limitations and strengths of the Schrieffer Wolff transformation on this particular system for the lower length spin chains, by exploring the energy developments in the original as well as the transformed model.

2 Cobalt spin chain

Consider a one dimensional spin chain of cobalt atoms with nearest neighbor antiferromagnetic isotropic Heisenberg exchange, an external transverse magnetic field, and an anisotropic spin coupling to the surface, the Hamiltonian can be written as [4]:

$$\mathcal{H}_{3/2} = J \sum_{i=1}^{N-1} \vec{S}_i \cdot \vec{S}_{i+1} + D \sum_{i=1}^N (S_i^z)^2 - g\mu_B B_x \sum_{i=1}^N S_i^x \quad (1)$$

Where J is the nearest neighbor exchange factor, D is the anisotropic spin coupling to the surface, and B_x is the transverse magnetic field, as visualized in Figure 1.

This Hamiltonian is prohibitively expensive to compute for a large N . Since every atom behaves as spin $S = 3/2$, the number of entries for the matrix representation of the Hamiltonian is 4^N . In low energy conditions one could imagine the system could be approximated with cobalt atoms behaving as spin $S = 1/2$, reducing the number of entries to 2^N .

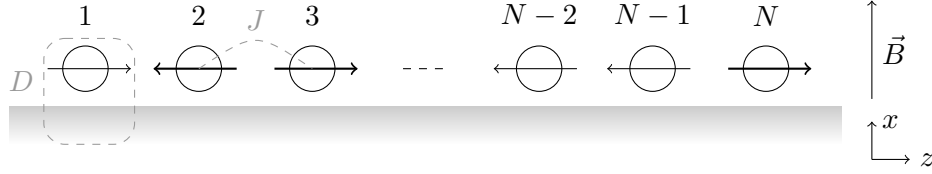


Figure 1: An example of a state of the Cobalt spin chain of length N , with a transverse magnetic field, $\vec{B}$. Here the thick arrows represent an atom spin corresponding to $|m_S| = 3/2$, and the thin represent $|m_S| = 1/2$. The nearest neighbor antiferromagnetic isotropic Heisenberg exchange, J , is represented as a dashed line between atoms number 2 and 3, and the anisotropic surface spin coupling, D as a dashed box encircling atom 1.

3 Schrieffer-Wolff transformation of the 1D spin chain

3.1 Schrieffer-Wolff transformation

In the Schrieffer-Wolff transformation a Hamiltonian is separated into two parts; one low energy and one high energy part. The purpose of this transformation is to evaluate a given system in a low energy state without having to consider the higher energy composites of the Hamiltonian.

The transformation is based on separating the Hamiltonian into three parts, as shown in Figure 2: the unperturbed matrix diagonal, H^0 , and the perturbation consisting of the diagonal blocks H^1 and the off-diagonal blocks H^2 . The quantum states are divided in low energy states, m , and high energy states, l . The blocks are then divided so that the diagonal blocks, H^1 , consists of two blocks; one only containing interactions between low energy states and one between high energy states. The interactions between high and low energy states are then contained in the off-diagonal blocks, H^2 , thereby only containing the bra-ket Hamiltonian product between high and low energy states, (e.g. $\langle m|H|l\rangle$ and $\langle l|H|m\rangle$). By demanding that the off-diagonal blocks equal zero after the unitary transformation, the Hamiltonian is successfully separated into a high energy block B and a low energy block A , with no effective coupling. The low energy part is given by $H_{mm'} = H_{mm'}^{(0)} + H_{mm'}^{(1)} + H_{mm'}^{(2)} + \dots$ where the zeroth, first and second order are given by:

$$\begin{aligned} H_{mm'}^{(0)} &= H_{mm'}^0 \\ H_{mm'}^{(1)} &= H'_{mm'} \\ H_{mm'}^{(2)} &= \frac{1}{2} \sum_l H'_{ml} H'_{lm'} \left[\frac{1}{E_m - E_l} + \frac{1}{E_{m'} - E_l} \right] \end{aligned} \quad (2)$$

with $H_{mm'}^X = \langle m|H^X|m'\rangle$ and $H' = H^1 + H^2$. [1]

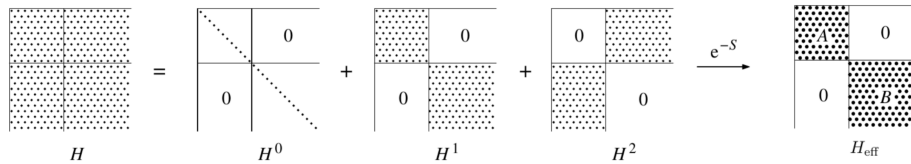


Figure 2: The three parts of the original Hamiltonian, the unperturbed diagonal part H^0 and the perturbation consisting of a block diagonal term H^1 and a block off-diagonal term H^2 . The Schrieffer-Wolff transformation (the unitary transformation e^{-S}) leaves us with a block diagonal effective Hamiltonian, with no coupling between the blocks A and B. (Figure is adapted from [1] appendix B, page 202)

3.2 Spin matrices

In order to correctly go from the $S = 3/2$ space to the $S = 1/2$ space we need some insight on the spin matrices used. These matrices in $\mathcal{H}_{3/2}$ are in the $S = 3/2$ space and are found using the equations from ref. [5].

First of we get the $S^{z,3/2}$ matrix from the equation $S_z |s, m\rangle = \hbar m |s, m\rangle$, yielding matrix elements:

$$S_{n,m}^z = \langle s, n | S_z | s, m \rangle = \hbar m \langle s, n | s, m \rangle = \hbar m \delta_{n,m}$$

And thereby the matrix:

$$S^{z,3/2} = \hbar \begin{pmatrix} \frac{3}{2} & 0 & 0 & 0 \\ 0 & \frac{1}{2} & 0 & 0 \\ 0 & 0 & -\frac{1}{2} & 0 \\ 0 & 0 & 0 & -\frac{3}{2} \end{pmatrix}$$

The $S^{\pm,3/2}$ matrices can likewise be found from the equation, $S_{\pm} |s, m\rangle = \hbar \sqrt{s(s+1) - m(m \pm 1)} |s, m \pm 1\rangle$, giving us matrix elements:

$$S_{n,m}^{\pm} = \langle s, n | S_{\pm} | s, m \rangle = \hbar \sqrt{(s \mp m)(s \pm m + 1)} \delta_{n,m \pm 1}$$

Corresponding to the matrices:

$$S^{+,3/2} = \hbar \begin{pmatrix} 0 & \sqrt{3} & 0 & 0 \\ 0 & 0 & 2 & 0 \\ 0 & 0 & 0 & \sqrt{3} \\ 0 & 0 & 0 & 0 \end{pmatrix} \quad S^{-,3/2} = \hbar \begin{pmatrix} 0 & 0 & 0 & 0 \\ \sqrt{3} & 0 & 0 & 0 \\ 0 & 2 & 0 & 0 \\ 0 & 0 & \sqrt{3} & 0 \end{pmatrix}$$

The spin operators $S^{x,3/2}$ and $S^{y,3/2}$ are then found using $S_{\pm} = S_x \pm iS_y$

$$\Rightarrow S^{x,3/2} = \frac{1}{2}(S^{+,3/2} + S^{-,3/2}) \quad \& \quad S^{y,3/2} = \frac{1}{2i}(S^{+,3/2} - S^{-,3/2}) \quad (3)$$

Giving us the matrices:

$$S^{x,3/2} = \frac{\hbar}{2} \begin{pmatrix} 0 & \sqrt{3} & 0 & 0 \\ \sqrt{3} & 0 & 2 & 0 \\ 0 & 2 & 0 & \sqrt{3} \\ 0 & 0 & \sqrt{3} & 0 \end{pmatrix}, \quad S^{y,3/2} = \frac{\hbar}{2i} \begin{pmatrix} 0 & \sqrt{3} & 0 & 0 \\ -\sqrt{3} & 0 & 2 & 0 \\ 0 & -2 & 0 & \sqrt{3} \\ 0 & 0 & -\sqrt{3} & 0 \end{pmatrix}$$

And in the desired $\mathcal{H}_{1/2}$ the spin operators are in the $S = 1/2$ space, and are written as follows:

$$S^{x,1/2} = \frac{\hbar}{2} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad S^{y,1/2} = \frac{\hbar}{2i} \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix} \quad S^{z,1/2} = \hbar \begin{pmatrix} \frac{1}{2} & 0 \\ 0 & -\frac{1}{2} \end{pmatrix}$$

This gives us a factor of two when projecting $S^{x,3/2}$ and $S^{y,3/2}$ on to the $S = 1/2$ space, but only a factor of one when projecting $S^{z,3/2}$ on to the same space.

3.3 Zeroth and first order

Writing $\mathcal{H}_{3/2}$ in terms of S^x, S^y and S^z in order to determine off diagonal elements, the Hamiltonian becomes:

$$\mathcal{H}_{3/2} = J \sum_{i=1}^{N-1} (S_i^x \cdot S_{i+1}^x + S_i^y \cdot S_{i+1}^y + S_i^z \cdot S_{i+1}^z) + D \sum_{i=1}^N (S_i^z)^2 - g\mu_B B_x \sum_{i=1}^N S_i^x$$

The diagonal of the matrix representation of $\mathcal{H}_{3/2}$ in the S^z basis, H^0 , is then the S^z -elements of this Hamiltonian:

$$H^0 = J \sum_{i=1}^{N-1} S_i^{z,3/2} \cdot S_{i+1}^{z,3/2} + D \sum_{i=1}^N (S_i^{z,3/2})^2$$

The remainder of $\mathcal{H}_{3/2}$ then constitutes the off-diagonal elements of the same matrix representation, H' :

$$H' = J \sum_{i=1}^{N-1} (S_i^{x,3/2} \cdot S_{i+1}^{x,3/2} + S_i^{y,3/2} \cdot S_{i+1}^{y,3/2}) - g\mu_B B_x \sum_{i=1}^N S_i^{x,3/2}$$

The zeroth order of the Schrieffer-Wolff transformation is then retrieved from the projection of H^0 on to the $S = 1/2$ space:

$$\begin{aligned} H^{(0)} &= H^0|_{|m_S|=1/2} = J \sum_{i=1}^{N-1} S_i^{z,3/2}|_{|m_S|=1/2} \cdot S_{i+1}^{z,3/2}|_{|m_S|=1/2} \\ &\quad + D \sum_{i=1}^N (S_i^{z,3/2}|_{|m_S|=1/2})^2 \\ &= J \sum_{i=1}^{N-1} S_i^{z,1/2} \cdot S_{i+1}^{z,1/2} + D \sum_{i=1}^N \frac{\hbar^2}{4} \\ &= J \sum_{i=1}^{N-1} S_i^{z,1/2} \cdot S_{i+1}^{z,1/2} + \frac{DN\hbar^2}{4} \end{aligned}$$

Since $\frac{DN\hbar^2}{4}$ is a constant, we find that:

$$H^{(0)} = J \sum_{i=1}^{N-1} S_i^{z,1/2} \cdot S_{i+1}^{z,1/2} \quad (4)$$

The first order of the Schrieffer-Wolff transformation is then retrieved from the projection of H' on to the $S = 1/2$ space by application of the factors

found in the Spin matrices subsection:

$$\begin{aligned}
H^{(1)} &= H' |_{|m_S|=1/2} \\
&= J \sum_{i=1}^{N-1} \left(S_i^{x,3/2} |_{|m_S|=1/2} \cdot S_{i+1}^{x,3/2} |_{|m_S|=1/2} + S_i^{y,3/2} |_{|m_S|=1/2} \cdot S_{i+1}^{y,3/2} |_{|m_S|=1/2} \right) \\
&\quad - \mu_B B_x g \sum_{i=1}^N S_i^{x,1/2} |_{|m_S|=1/2} \\
&= 4J \sum_{i=1}^{N-1} \left(S_i^{x,1/2} \cdot S_{i+1}^{x,1/2} + S_i^{y,1/2} \cdot S_{i+1}^{y,1/2} \right) - 2\mu_B B_x g \sum_{i=1}^N S_i^{x,1/2}
\end{aligned} \tag{5}$$

3.4 Second order

The second order of the transformation is given by equation (2):

$$H_{mm'}^{(2)} = \frac{1}{2} \sum_l H'_{ml} H'_{lm'} \left[\frac{1}{E_m - E_l} + \frac{1}{E_{m'} - E_l} \right]$$

Where the l states contain one or more high energy atoms, e.g. $|m_S| = 3/2$, and the m states only contain low energy atoms.

Considering the l states with n_l high energy atoms, and all energy scales staying well below the anisotropy level $2D$ we can write:

$$\begin{aligned}
E_m - E_l &= \frac{ND\hbar^2}{4} - \left(\frac{(N - n_l)D\hbar^2}{4} + n_l D\hbar^2 \frac{9}{4} \right) + \mathcal{O}(J, \mu_B B_x) \\
&= -2n_l D\hbar^2 + \mathcal{O}(J, \mu_B B_x)
\end{aligned} \tag{6}$$

When $D \gg J, \mu_B B_x$ this reduces to $-2n_l D\hbar^2$, giving us:

$$\left[\frac{1}{E_m - E_l} + \frac{1}{E_{m'} - E_l} \right] \approx -\frac{1}{n_l D\hbar^2} \tag{7}$$

Using equation (3) we can write H' in terms of $S^{+,3/2}$ and $S^{-,3/2}$:

$$\begin{aligned}
 H' = & J \sum_{i=1}^{N-1} \frac{1}{2} \left(S_i^{+,3/2} + S_i^{-,3/2} \right) \cdot \frac{1}{2} \left(S_{i+1}^{+,3/2} + S_{i+1}^{-,3/2} \right) \\
 & + \frac{1}{2I} \left(S_i^{+,3/2} - S_i^{-,3/2} \right) \cdot \frac{1}{2I} \left(S_{i+1}^{+,3/2} - S_{i+1}^{-,3/2} \right) \\
 & - g\mu_B B_x \sum_{i=1}^N \frac{1}{2} \left(S_i^{+,3/2} + S_i^{-,3/2} \right) \\
 = & \frac{J}{2} \sum_{i=1}^{N-1} \left(S_i^{+,3/2} S_{i+1}^{-,3/2} + S_i^{-,3/2} S_{i+1}^{+,3/2} \right) \\
 & - g\mu_B B_x \sum_{i=1}^N \frac{1}{2} \left(S_i^{+,3/2} + S_i^{-,3/2} \right)
 \end{aligned} \tag{8}$$

Now $H'_{ml} H'_{lm'}$ can be evaluated, while implicitly in the $S = 3/2$ space:

$$\begin{aligned}
 H'_{ml} H'_{lm'} &= \langle m | H' | l \rangle \langle l | H' | m' \rangle \\
 &= \langle m | \left(\frac{J}{2} \sum_{i=1}^{N-1} (S_i^+ S_{i+1}^- + S_i^- S_{i+1}^+) - g\mu_B B_x \sum_{i=1}^N \frac{1}{2} (S_i^+ + S_i^-) \right) | l \rangle \\
 &\quad \langle l | \left(\frac{J}{2} \sum_{j=1}^{N-1} (S_j^+ S_{j+1}^- + S_j^- S_{j+1}^+) - g\mu_B B_x \sum_{j=1}^N \frac{1}{2} (S_j^+ + S_j^-) \right) | m' \rangle \\
 &= \frac{J^2}{4} \sum_{i,j=1}^{N-1} \left(\langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_j^+ S_{j+1}^- | m' \rangle + \langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_j^- S_{j+1}^+ | m' \rangle \right. \\
 &\quad \left. + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_j^+ S_{j+1}^- | m' \rangle + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_j^- S_{j+1}^+ | m' \rangle \right) \\
 &\quad - Jg\mu_B B_x \frac{1}{4} \sum_{i=1}^{N-1} \sum_{j=1}^N \left(\langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_j^+ | m' \rangle + \langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_j^- | m' \rangle \right. \\
 &\quad \left. + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_j^+ | m' \rangle + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_j^- | m' \rangle \right) \\
 &\quad - Jg\mu_B B_x \frac{1}{4} \sum_{j=1}^{N-1} \sum_{i=1}^N \left(\langle m | S_i^+ | l \rangle \langle l | S_j^+ S_{j+1}^- | m' \rangle + \langle m | S_i^+ | l \rangle \langle l | S_j^- S_{j+1}^+ | m' \rangle \right. \\
 &\quad \left. + \langle m | S_i^- | l \rangle \langle l | S_j^+ S_{j+1}^- | m' \rangle + \langle m | S_i^- | l \rangle \langle l | S_j^- S_{j+1}^+ | m' \rangle \right) \\
 &\quad + g^2 \mu_B^2 B_x^2 \frac{1}{4} \sum_{i=1}^N \sum_{j=1}^N \left(\langle m | S_i^+ | l \rangle \langle l | S_j^+ | m' \rangle + \langle m | S_i^+ | l \rangle \langle l | S_j^- | m' \rangle \right. \\
 &\quad \left. + \langle m | S_i^- | l \rangle \langle l | S_j^+ | m' \rangle + \langle m | S_i^- | l \rangle \langle l | S_j^- | m' \rangle \right)
 \end{aligned} \tag{9}$$

In order to reduce this expression to a Hamiltonian in the $S = 1/2$ space, we must first evaluate the four summations in equation (9).

Starting with the second summation in equation (9):

$$-Jg\mu_B B_x \frac{1}{4} \sum_{i=1}^{N-1} \sum_{j=1}^N \left(\langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_j^+ | m' \rangle + \langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_j^- | m' \rangle \right. \\ \left. + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_j^+ | m' \rangle + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_j^- | m' \rangle \right)$$

We only get contributions when the spin of $|m'\rangle$ is either raised or lowered to match the spin of $\langle l|$ and the spin of $|l\rangle$ simultaneously is lowered or raised to match $\langle m|$. Otherwise one of the bra-ket products would be zero, since there at least is one atom in l with a spin value $|m_S| = 3/2$, which therefore would be perpendicular to the corresponding atom in m' and m .

The only contributions are therefore from $j = i$ and $j = i + 1$ where we get:

$$-Jg\mu_B B_x \sum_{i=1}^{N-1} \frac{1}{4} \left(\langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_i^- | m' \rangle + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_i^+ | m' \rangle \right)$$

and

$$-Jg\mu_B B_x \sum_{i=1}^{N-1} \frac{1}{4} \left(\langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_{i+1}^+ | m' \rangle + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_{i+1}^- | m' \rangle \right)$$

Only l states with one atom in the $S = 3/2$ will give a contribution, since only one atom is raised or lowered before multiplying with an m or m' state. There will both be a raising or lowering of $|m'\rangle$ to this state and a lowering or raising of $|l\rangle$ back to $\langle m|$, giving us a factor of $3\hbar^2$. In order to ensure that we get the same contributions as above we require that the atoms, that are raised in $|m'\rangle$ and lowered in $\langle l|$ have spin $m_S = +1/2$ in $|m'\rangle$, and the opposite for the ones lowered and then raised. This is done by reversing the order of operations in the $S = 1/2$ space, thereby ensuring that only the $\langle m|$ and $|m'\rangle$ states, that gave contributions in the $S = 3/2$ space, give contributions in the $S = 1/2$ space. When adjusting factors to make the contributions equal we get:

$$-Jg\mu_B B_x \sum_{i=1}^{N-1} \frac{1}{4} \left(6 \langle m | S_i^- S_i^+ S_{i+1}^- | m' \rangle + 6 \langle m | S_i^+ S_i^- S_{i+1}^+ | m' \rangle \right)$$

and

$$-Jg\mu_B B_x \sum_{i=1}^{N-1} \frac{1}{4} \left(6 \langle m | S_i^+ S_{i+1}^+ S_{i+1}^- | m' \rangle + 6 \langle m | S_i^- S_{i+1}^- S_{i+1}^+ | m' \rangle \right)$$

where $n_l = 1$ and the factor of 6 comes from $S_i^{\pm,3/2} |l\rangle \langle l| S_i^{\mp,3/2} |m'\rangle = 3 S_i^{\mp,1/2} S_i^{\pm,1/2} |m'\rangle$ and $S_i^{\pm,3/2} = 2 S_i^{\pm,1/2}$. A similar deduction is visualized in Figure 3.

From the third summation in equation (9) we use the same approach, only for $j = i$ and $j = i - 1$ and get:

$$-Jg\mu_B B_x \sum_{i=1}^{N-1} \frac{1}{4} (6 \langle m | S_i^+ S_i^- S_{i+1}^- | m' \rangle + 6 \langle m | S_i^- S_i^+ S_{i+1}^+ | m' \rangle)$$

and

$$-Jg\mu_B B_x \sum_{i=2}^N \frac{1}{4} (6 \langle m | S_{i-1}^+ S_i^- S_i^+ | m' \rangle + 6 \langle m | S_{i-1}^- S_i^+ S_i^- | m' \rangle)$$

Collecting the results from the two summations we get:

$$\begin{aligned} -Jg\mu_B B_x \sum_{i=1}^{N-1} \frac{3}{2} & (\langle m | S_i^+ (S_{i+1}^- S_{i+1}^+ + S_{i+1}^+ S_{i+1}^-) | m' \rangle \\ & + \langle m | S_i^- (S_{i+1}^+ S_{i+1}^- + S_{i+1}^- S_{i+1}^+) | m' \rangle \\ & + \langle m | (S_i^+ S_i^- + S_i^- S_i^+) S_{i+1}^- | m' \rangle \\ & + \langle m | (S_i^- S_i^+ + S_i^+ S_i^-) S_{i+1}^+ | m' \rangle) \end{aligned}$$

Using the reduction $(S_i^- S_i^+ + S_i^+ S_i^-) |m'\rangle = \hbar^2 |m'\rangle$ in the $S = 1/2$ space yields:

$$\begin{aligned} & -Jg\mu_B B_x \sum_{i=1}^{N-1} \frac{3}{2} \hbar^2 (\langle m | S_i^+ + S_i^- + S_{i+1}^- + S_{i+1}^+ | m' \rangle) \\ & = -3\hbar^2 Jg\mu_B B_x \sum_{i=1}^{N-1} (S_i^x + S_{i+1}^x) \end{aligned} \tag{10}$$

The two middle summations in equation (9) evaluated in the $S = 1/2$ space, with $n_l = 1$, thereby becomes equation (10).

Next from the fourth summation in equation (9) (in the $S = 3/2$ space);

$$\begin{aligned} g^2 \mu_B^2 B_x^2 \frac{1}{4} \sum_{i=1}^N \sum_{j=1}^N & \left(\langle m | S_i^+ | l \rangle \langle l | S_j^+ | m' \rangle + \langle m | S_i^+ | l \rangle \langle l | S_j^- | m' \rangle \right. \\ & \left. + \langle m | S_i^- | l \rangle \langle l | S_j^+ | m' \rangle + \langle m | S_i^- | l \rangle \langle l | S_j^- | m' \rangle \right) \end{aligned}$$

We similarly only get contributions from $j = i$ which reduces to:

$$g^2 \mu_B^2 B_x^2 \frac{3\hbar^2}{4} \sum_{i=1}^N \langle m | m' \rangle$$

Leaving us with only a constant, and thereby an energy offset, rendering this part without interest.

Finally we consider the first summation in equation (9):

$$\begin{aligned} \frac{J^2}{4} \sum_{i,j=1}^{N-1} & \left(\langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_j^+ S_{j+1}^- | m' \rangle + \langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_j^- S_{j+1}^+ | m' \rangle \right. \\ & \left. + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_j^+ S_{j+1}^- | m' \rangle + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_j^- S_{j+1}^+ | m' \rangle \right) \end{aligned}$$

For $j = i + 1$ we get:

$$\begin{aligned} \frac{J^2}{4} \sum_{i=1}^{N-2} & \left(\langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_{i+1}^+ S_{i+2}^- | m' \rangle + \langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_{i+1}^- S_{i+2}^+ | m' \rangle \right. \\ & \left. + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_{i+1}^+ S_{i+2}^- | m' \rangle + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_{i+1}^- S_{i+2}^+ | m' \rangle \right) \end{aligned}$$

in the $S = 3/2$ space. By going to the $S = 1/2$ space, as we did in the deduction of equation (10), we get:

$$\frac{J^2}{4} \sum_{i=1}^{N-2} \langle m | 12 (S_i^+ S_{i+1}^+ S_{i+1}^- S_{i+2}^- + S_i^- S_{i+1}^- S_{i+1}^+ S_{i+2}^+) | m' \rangle$$

and for $j = i - 1$ we get:

$$\begin{aligned} \frac{J^2}{4} \sum_{i=2}^{N-1} & \left(\langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_{i-1}^+ S_i^- | m' \rangle + \langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_{i-1}^- S_i^+ | m' \rangle \right. \\ & \left. + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_{i-1}^+ S_i^- | m' \rangle + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_{i-1}^- S_i^+ | m' \rangle \right) \end{aligned}$$

in the $S = 3/2$. And in the $S = 1/2$ space we get:

$$\frac{J^2}{4} \sum_{i=1}^{N-2} \langle m | 12 (S_i^+ S_{i+1}^- S_{i+1}^+ S_{i+2}^- + S_i^- S_{i+1}^+ S_{i+1}^- S_{i+2}^+) | m' \rangle$$

By adding both contributions from $j = i + 1$ and $j = i - 1$ we get:

$$\begin{aligned} & \frac{J^2}{4} \sum_{i=1}^{N-2} \langle m | 12 (S_i^+ (S_{i+1}^+ S_{i+1}^- + S_{i+1}^- S_{i+1}^+) S_{i+2}^- \\ & \quad + S_i^- (S_{i+1}^- S_{i+1}^+ + S_{i+1}^+ S_{i+1}^-) S_{i+2}^+) | m' \rangle \\ & = 6\hbar^2 J^2 \sum_{i=1}^{N-2} \langle m | \frac{1}{2} (S_i^+ S_{i+2}^- + S_i^- S_{i+2}^+) | m' \rangle \end{aligned}$$

and using the reduction in equation (8) we then get:

$$6\hbar^2 n_l J^2 \sum_{i=1}^{N-2} \langle m | (S_i^x S_{i+2}^x + S_i^y S_{i+2}^y) | m' \rangle \quad (11)$$

where $n_l = 1$ as in the deduction of equation (10).

The final contribution comes from $j = i$:

$$\begin{aligned} \frac{J^2}{4} \sum_{i=1}^{N-1} & (\langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_i^+ S_{i+1}^- | m' \rangle + \langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_i^- S_{i+1}^+ | m' \rangle \\ & + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_i^+ S_{i+1}^- | m' \rangle + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_i^- S_{i+1}^+ | m' \rangle) \end{aligned}$$

This immediately reduces to:

$$\frac{J^2}{4} \sum_{i=1}^{N-1} (\langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_i^- S_{i+1}^+ | m' \rangle + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_i^+ S_{i+1}^- | m' \rangle)$$

since the other parts are not be able to simultaneously give a contribution from both brackets in their product.

Each of the remaining parts now give a contribution for three unique l states. One where atom i has spin $|m_S| = 3/2$ another where atom $i + 1$ has spin $|m_S| = 3/2$, and one where both atoms, i and $i + 1$, have spin $|m_S| = 3/2$. Going to the $S = 1/2$ space we rewrite these parts as:

$$\begin{aligned} \frac{J^2}{4} \sum_{i=1}^{N-1} & (12 \langle m | S_i^- S_i^+ S_{i+1}^- S_{i+1}^+ | m' \rangle |_{n_l=1} + 12 \langle m | S_i^+ S_i^- S_{i+1}^- S_{i+1}^+ | m' \rangle |_{n_l=1} \\ & + 9 \langle m | S_i^- S_i^+ S_{i+1}^+ S_{i+1}^- | m' \rangle |_{n_l=2} + 12 \langle m | S_i^+ S_i^- S_{i+1}^+ S_{i+1}^- | m' \rangle |_{n_l=1} \\ & + 12 \langle m | S_i^- S_i^+ S_{i+1}^- S_{i+1}^+ | m' \rangle |_{n_l=1} + 9 \langle m | S_i^+ S_i^- S_{i+1}^- S_{i+1}^+ | m' \rangle |_{n_l=2}) \end{aligned}$$

The only n_l dependency is from equation (7), where the dependency is the inverse of n_l . Since equation (7) is an expression for the factor of $H'_{ml} H'_{lm'}$ we can already take the n_l dependency into consideration, giving us:

$$\begin{aligned} \frac{J^2}{4} \sum_{i=1}^{N-1} & \left(24 \langle m | (S_i^- S_i^+ S_{i+1}^- S_{i+1}^+ + S_i^+ S_i^- S_{i+1}^+ S_{i+1}^-) | m' \rangle \right. \\ & \left. + \frac{9}{2} \langle m | (S_i^- S_i^+ S_{i+1}^+ S_{i+1}^- + S_i^+ S_i^- S_{i+1}^- S_{i+1}^+) | m' \rangle \right) \end{aligned}$$

For equal factors of all parts we get a constant offset since it will give the same result independent of m and m' . This means we can add a constant to all factors, giving us:

$$\begin{aligned} \frac{J^2}{4} \sum_{i=1}^{N-1} & \left(\frac{39}{4} \langle m | (S_i^- S_i^+ S_{i+1}^- S_{i+1}^+ + S_i^+ S_i^- S_{i+1}^+ S_{i+1}^-) | m' \rangle \right. \\ & \left. - \frac{39}{4} \langle m | (S_i^- S_i^+ S_{i+1}^+ S_{i+1}^- + S_i^+ S_i^- S_{i+1}^- S_{i+1}^+) | m' \rangle \right) \end{aligned}$$

We now recognize the product of commutators for S^+ and S^- :

$$\frac{J^2}{4} \sum_{i=1}^{N-1} \frac{39}{4} \langle m | [S_i^+, S_i^-] \cdot [S_{i+1}^+, S_{i+1}^-] | m' \rangle$$

And since $[S_i^+, S_i^-] = 2\hbar S_i^z$ we get:

$$\frac{39J^2\hbar^2}{4} \sum_{i=1}^{N-1} \langle m | S_i^z S_{i+1}^z | m' \rangle \quad (12)$$

3.5 Solution of the Schrieffer-Wolff transformation

We can now write an expression for $H'_{ml}H'_{lm'}$ from equation (10), (11) and (12) in the $S = 1/2$ space:

$$\begin{aligned} H'_{ml}H'_{lm'} = & 6\hbar^2 J^2 \sum_{i=1}^{N-2} \langle m | (S_i^x S_{i+2}^x + S_i^y S_{i+2}^y) | m' \rangle \\ & + \frac{39J^2\hbar^2}{4} \sum_{i=1}^{N-1} \langle m | S_i^z S_{i+1}^z | m' \rangle \\ & - 3\hbar^2 J g \mu_B B_x \sum_{i=1}^{N-1} \langle m | (S_{i+1}^x + S_i^x) | m' \rangle \end{aligned}$$

for $n_l = 1$.

And we can finally evaluate $H_{mm'}^{(2)}$ from equation (2):

$$\begin{aligned} H_{mm'}^{(2)} = & \frac{1}{2} \sum_l H'_{ml}H'_{lm'} \left[\frac{1}{E_m - E_l} + \frac{1}{E_{m'} - E_l} \right] \\ \simeq & \frac{1}{2} \sum_l \frac{-1}{n_l D \hbar^2} H'_{ml}H'_{lm'} \end{aligned}$$

The sum over l is accounted for in equation (10) and (11) since every contribution to these equations come from a unique l state with $n_l = 1$. The n_l dependency along with the summation over l has explicitly been dealt with in equation (12), e.g. $\sum_l \frac{H'_{ml}H'_{lm'}}{n_l}$ is the sum of equation (10), (11) and (12), leaving us with:

$$\begin{aligned} H^{(2)} \simeq & -\frac{3J^2}{D} \sum_{i=1}^{N-2} (S_i^x S_{i+2}^x + S_i^y S_{i+2}^y) \\ & - \frac{39J^2}{8D} \sum_{i=1}^{N-1} S_i^z S_{i+1}^z \\ & + \frac{3J}{2D} g \mu_B B_x \sum_{i=1}^{N-1} (S_{i+1}^x + S_i^x) \end{aligned} \quad (13)$$

Any higher order in the Schrieffer-Wolff transformation is a higher order in $1/D$.

The final transformation to the first order in $1/D$ is then the sum of equations (4), (5) and (13):

$$\begin{aligned} \mathcal{H}_{1/2} = & \sum_{i=1}^{N-1} J_{\perp} (S_i^x S_{i+1}^x + S_i^y S_{i+1}^y) + J_z S_i^z S_{i+1}^z \\ & + J_{\perp}^{nnn} \sum_{i=1}^{N-2} S_i^x S_{i+2}^x + S_i^y S_{i+2}^y - \mu_B B_x \sum_{i=1}^N g_i S_i^x \end{aligned} \quad (14)$$

with nearest and next-nearest neighbor exchange parameters and bulk/boundary g -factors, given by:

$$\begin{aligned} J_{\perp} &= 4J, & J_z &= J - \frac{39J^2}{8D}, & J_{\perp}^{nnn} &= -\frac{3J^2}{D}, \\ g_i &= \begin{cases} 2g \left(1 - \frac{3J}{2D}\right) & \text{if } i = 2, \dots, N-1 \\ 2g \left(1 - \frac{3J}{4D}\right) & \text{if } i = 1, N \end{cases} \end{aligned} \quad (15)$$

This means that a $S = 3/2$ nearest neighbor Heisenberg exchange spin chain in an external magnetic field can be described as a XXZ exchange spin chain with nearest and next nearest coupling in an external magnetic field, in a low energy setting where $k_B T$, $\mu_B B$ and J are significantly smaller than the anisotropy energy, D .

3.6 Third order

For good measure we can discuss the impact of the third order term in the transformation, given by: [1]

$$\begin{aligned} H_{mm'}^{(3)} = & -\frac{1}{2} \sum_{l,m''} \left[\frac{H'_{ml} H'_{lm''} H'_{m''m'}}{(E_{m'} - E_l)(E_{m''} - E_l)} + \frac{H'_{mm''} H'_{m''l} H'_{lm'}}{(E_{m'} - E_l)(E_{m''} - E_l)} \right] \\ & + \frac{1}{2} \sum_{l,l'} H'_{ml} H'_{ll'} H'_{l'm'} \left[\frac{1}{(E_m - E_l)(E_m - E_{l'})} + \frac{1}{(E_{m'} - E_l)(E_{m'} - E_{l'})} \right] \end{aligned}$$

Using equation (6) this becomes:

$$\begin{aligned} H_{mm'}^{(3)} = & -\frac{1}{2} \sum_{l,m''} \frac{H'_{ml} H'_{lm''} H'_{m''m'} + H'_{mm''} H'_{m''l} H'_{lm'}}{(-2n_l D \hbar^2)^2} \\ & + \frac{1}{2} \sum_{l,l'} \frac{2H'_{ml} H'_{ll'} H'_{l'm'}}{(-2n_l D \hbar^2)(-2n_{l'} D \hbar^2)} \end{aligned} \quad (16)$$

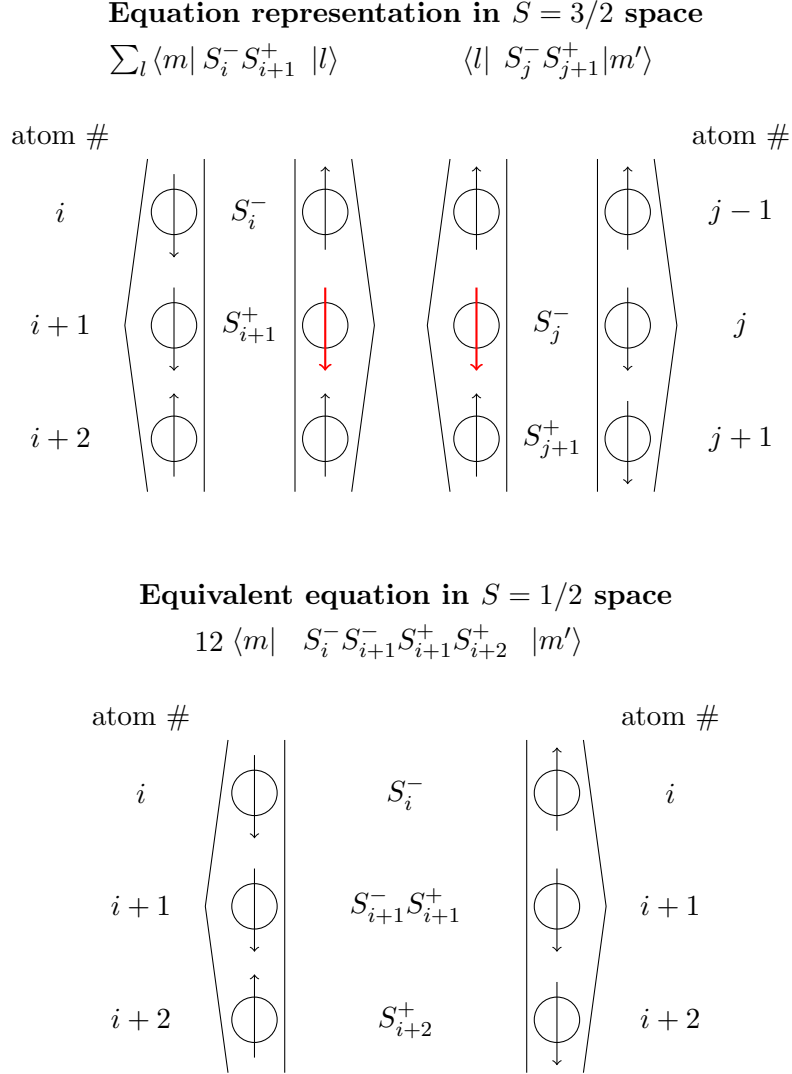


Figure 3: A visual representation of a part of the second order of the Schrieffer-Wolff transformation. Here the thick and red arrows represent an atom spin corresponding to $|m_S| = 3/2$, and the thin black arrows represent $|m_S| = 1/2$. It shows how the equation $\sum_l \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_j^- S_{j+1}^+ | m' \rangle$ in the $S = 3/2$ space corresponds to the equation $12 \langle m | (S_i^+ S_{i+1}^- S_{i+1}^- S_{i+2}^+) | m' \rangle$ in the $S = 1/2$ space. All atoms in the chain that are not shown in the representation above must necessarily have equal spins in m , l and m' , otherwise this equation renders 0, making it necessary that only a single l -state in the sum gives a contribution.

By introducing a third Hamiltonian evaluation a possibility for a new ex-

change operation emerges, the third nearest neighbor coupling. In order to determine if this term ultimately cancels out, all contributions to this coupling must be found.

The only part of the Hamiltonian capable of contributing to such a term is the original nearest neighbor exchange coupling. Only regarding this term in $\sum_{l,l'} H'_{ml} H'_{ll'} H'_{l'm'}$ renders:

$$\begin{aligned} \frac{J^3}{8} \sum_{i,j,k=1}^{N-1} & \left(\langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_j^+ S_{j+1}^- | l' \rangle \langle l' | S_k^+ S_{k+1}^- | m' \rangle \right. \\ & + \langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_j^+ S_{j+1}^- | l' \rangle \langle l' | S_k^- S_{k+1}^+ | m' \rangle \\ & + \langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_j^- S_{j+1}^+ | l' \rangle \langle l' | S_k^- S_{k+1}^+ | m' \rangle \\ & + \langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_j^- S_{j+1}^+ | l' \rangle \langle l' | S_k^+ S_{k+1}^- | m' \rangle \\ & + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_j^+ S_{j+1}^- | l' \rangle \langle l' | S_k^+ S_{k+1}^- | m' \rangle \\ & + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_j^+ S_{j+1}^- | l' \rangle \langle l' | S_k^- S_{k+1}^+ | m' \rangle \\ & + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_j^- S_{j+1}^+ | l' \rangle \langle l' | S_k^+ S_{k+1}^- | m' \rangle \\ & \left. + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_j^- S_{j+1}^+ | l' \rangle \langle l' | S_k^- S_{k+1}^+ | m' \rangle \right) \end{aligned}$$

Which, for $i = j + 1 = k + 2$ and $i = j - 1 = k - 2$, quickly becomes

$$\begin{aligned} \frac{J^3}{8} \sum_{i,j,k=1}^{N-1} & \left(\langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_j^+ S_{j+1}^- | l' \rangle \langle l' | S_k^+ S_{k+1}^- | m' \rangle \right. \\ & \left. + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_j^- S_{j+1}^+ | l' \rangle \langle l' | S_k^- S_{k+1}^+ | m' \rangle \right) \end{aligned} \quad (17)$$

For $\sum_{l,l'} H'_{ml} H'_{ll'} H'_{l'm'}$ this is the limit in difference between, i, j and k , since at least one atom in l and l' is in a high energy state. For $\sum_{l,m''} (H'_{ml} H'_{lm''} H'_{m''m'} + H'_{mm''} H'_{m''l} H'_{lm'})$ it is not as trivial, but ultimately there will only be a third nearest neighbor coupling if the nearest and next nearest neighbor is both raised and lowered, leaving us with:

$$\begin{aligned} \frac{J^3}{8} \sum_{i,j,k=1}^{N-1} & \left(\left(\langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_j^+ S_{j+1}^- | m'' \rangle \langle m'' | S_k^+ S_{k+1}^- | m' \rangle \right. \right. \\ & + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_j^- S_{j+1}^+ | m'' \rangle \langle m'' | S_k^- S_{k+1}^+ | m' \rangle \Big) \\ & + \left(\langle m | S_i^+ S_{i+1}^- | m'' \rangle \langle m'' | S_j^+ S_{j+1}^- | l \rangle \langle l | S_k^+ S_{k+1}^- | m' \rangle \right. \\ & \left. \left. + \langle m | S_i^- S_{i+1}^+ | m'' \rangle \langle m'' | S_j^- S_{j+1}^+ | l \rangle \langle l | S_k^- S_{k+1}^+ | m' \rangle \right) \right) \end{aligned} \quad (18)$$

Reducing these to the $H_{mm'}^{(3)}$ form, with intermediate results in Appendix A equations (22) to (27). We can write equation (17) as:

$$\begin{aligned} & \frac{J^3}{8} \sum_{i=1}^{N-3} 36 (\langle m | S_i^+ (S_{i+1}^+ S_{i+1}^- S_{i+2}^+ S_{i+2}^- + S_{i+1}^- S_{i+1}^+ S_{i+2}^- S_{i+2}^+) S_{i+3}^- \\ & + S_i^- (S_{i+1}^- S_{i+1}^+ S_{i+2}^- S_{i+2}^+ + S_{i+1}^+ S_{i+1}^- S_{i+2}^+ S_{i+2}^-) S_{i+3}^+ | m' \rangle) |_{n_l=n_{l'}=1} \end{aligned} \quad (19)$$

And equation (18) as:

$$\begin{aligned} & 2 \frac{J^3}{8} \sum_{i=1}^{N-3} 48 ((\langle m | S_i^+ (S_{i+1}^+ S_{i+1}^- S_{i+2}^- S_{i+2}^+ + S_{i+1}^- S_{i+1}^+ S_{i+2}^+ S_{i+2}^-) S_{i+3}^- \\ & + S_i^- (S_{i+1}^- S_{i+1}^+ S_{i+2}^+ S_{i+2}^- + S_{i+1}^+ S_{i+1}^- S_{i+2}^- S_{i+2}^+) S_{i+3}^+ | m' \rangle) |_{n_l=1} \end{aligned} \quad (20)$$

Using these contributions in equation (16) will allow us to see the total contribution to third nearest neighbor coupling, with intermediate results in Appendix A equation (28) to (31):

$$\begin{aligned} H_{mm'}^{(3)} |_{\mathcal{O}(S_i, S_{i+3})} &= -\frac{3J^3}{16D^2} \sum_{i=1}^{N-3} \langle m | (S_i^x S_{i+3}^x + S_i^y S_{i+3}^y) | m' \rangle \\ &+ \frac{21J^3}{4\hbar^2 D^2} \sum_{i=1}^{N-3} \langle m | (S_i^x S_{i+3}^x + S_i^y S_{i+3}^y) (S_{i+1}^z S_{i+2}^z) | m' \rangle \end{aligned} \quad (21)$$

This means we get a third nearest neighbor exchange by itself and one coupled with the exchange of the two spins in between.

4 Energy developments in 1D Cobalt spin chain

Comparing the original Hamiltonian with the transformed Hamiltonian for short 1D Cobalt spin chains, allows us to check the accuracy of the Schrieffer-Wolff transformation for this system.

In Wolfram Mathematica 11 this is done by creating Matrices for the exact Hamiltonian, $\mathcal{H}_{3/2}$, as well as the transformed Hamiltonian, $\mathcal{H}_{1/2}$. The eigenvalues of the matrices are then determined for a range of the external magnetic field, and the energy spectra are compared for the two Hamiltonians.

The advantage of the Schrieffer-Wolff transformation already appears in the evaluation of the eigenvalues of the Hamiltonians, where we can determine these with a variable magnetic field, as opposed to the necessary numerical evaluation needed for the original Hamiltonian.

In Figure 4 we see the direct prediction of energies for one dimensional Cobalt spin chains of lengths two, three and four, by the original and the transformed models. For the longest spin chain it quickly becomes difficult to distinguish which energy levels correlate between the two models, because of the apparent inaccuracy of the transformation. And for the lower length spin chains it is clear that the transformed Hamiltonian begins to overestimate the energies of the different states. This inaccuracy is not unanticipated, since the whole transformation is based on the division of low and high energy states, which clearly does not hold for higher magnetic fields. This is apparent in the plots for the two longer spin chains, where the energies of the assumed high energy states are going down to the levels of the assumed low energy states, making this basic assumption invalid. The excitation energies in Figure 5 evidently correlate much better between the two models, while naturally becoming less accurate for higher magnetic fields. It is, however, worth noting that we see a significant offset in the magnetic field value at the phase shift for the spin chain of length 2, which seems to only appear at this chain length.

Ultimately we see that the Schrieffer-Wolff transformation works better for applications like determining excitation energies and ground states, rather than for calculating the energy developments of specific states of spin chains like this, since the slope of the direct energy calculations evidently are less accurate.

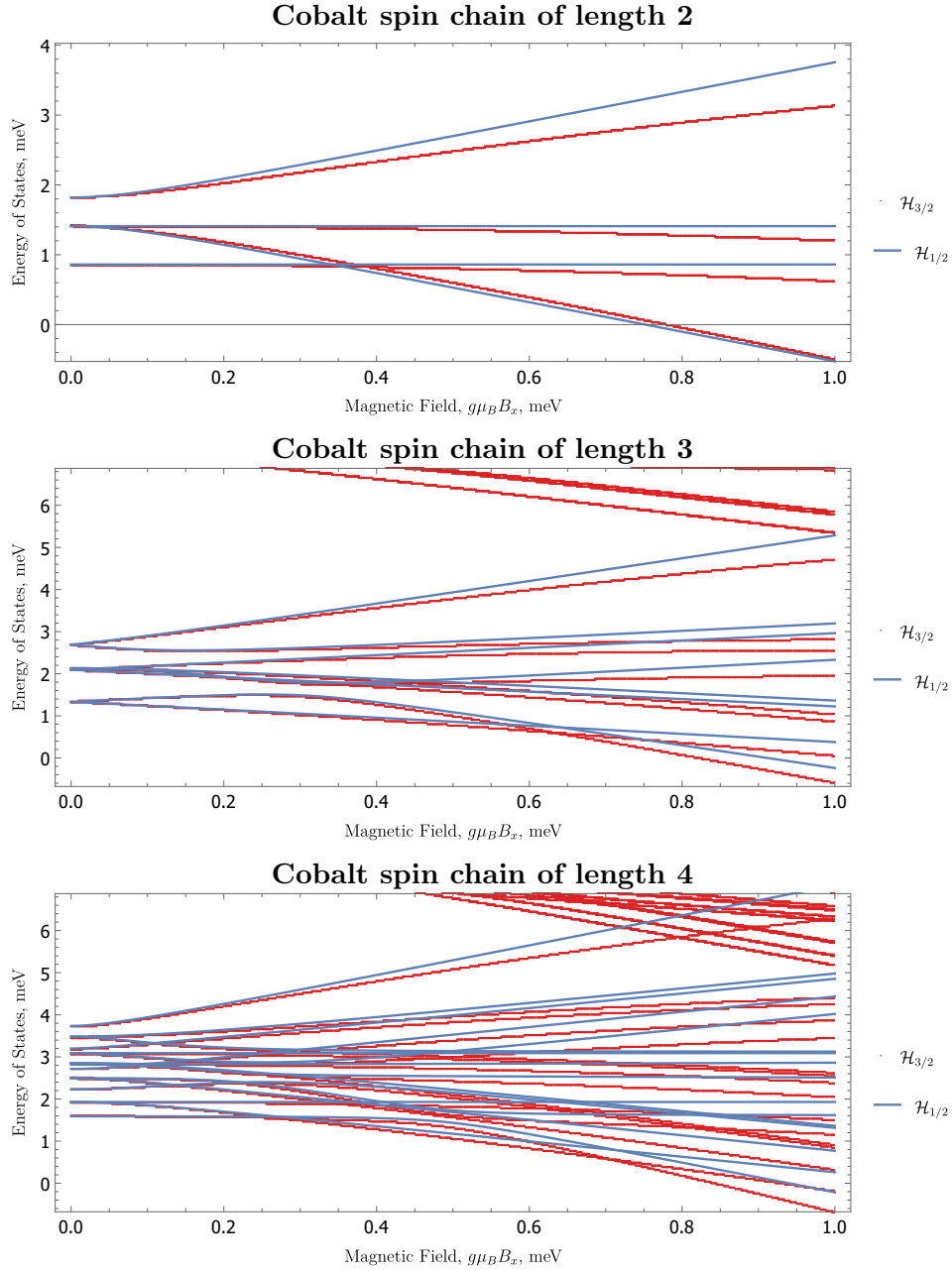


Figure 4: The energy spectrum for the 2^N lowest energy states of the cobalt spin chain of length $N = 2, 3$ and 4 , numerically evaluated with the matrix for the original Hamiltonian, $\mathcal{H}_{3/2}$, and analytically evaluated with the matrix for the Schrieffer-Wolff transformed Hamiltonian, $\mathcal{H}_{1/2}$. Since the g factor is 2.3 , and the Bohr magneton is approximately 5.788 eV/T, 1 meV corresponds to a magnetic field of 7.51 T. (For more details see Appendix B, p. 5)

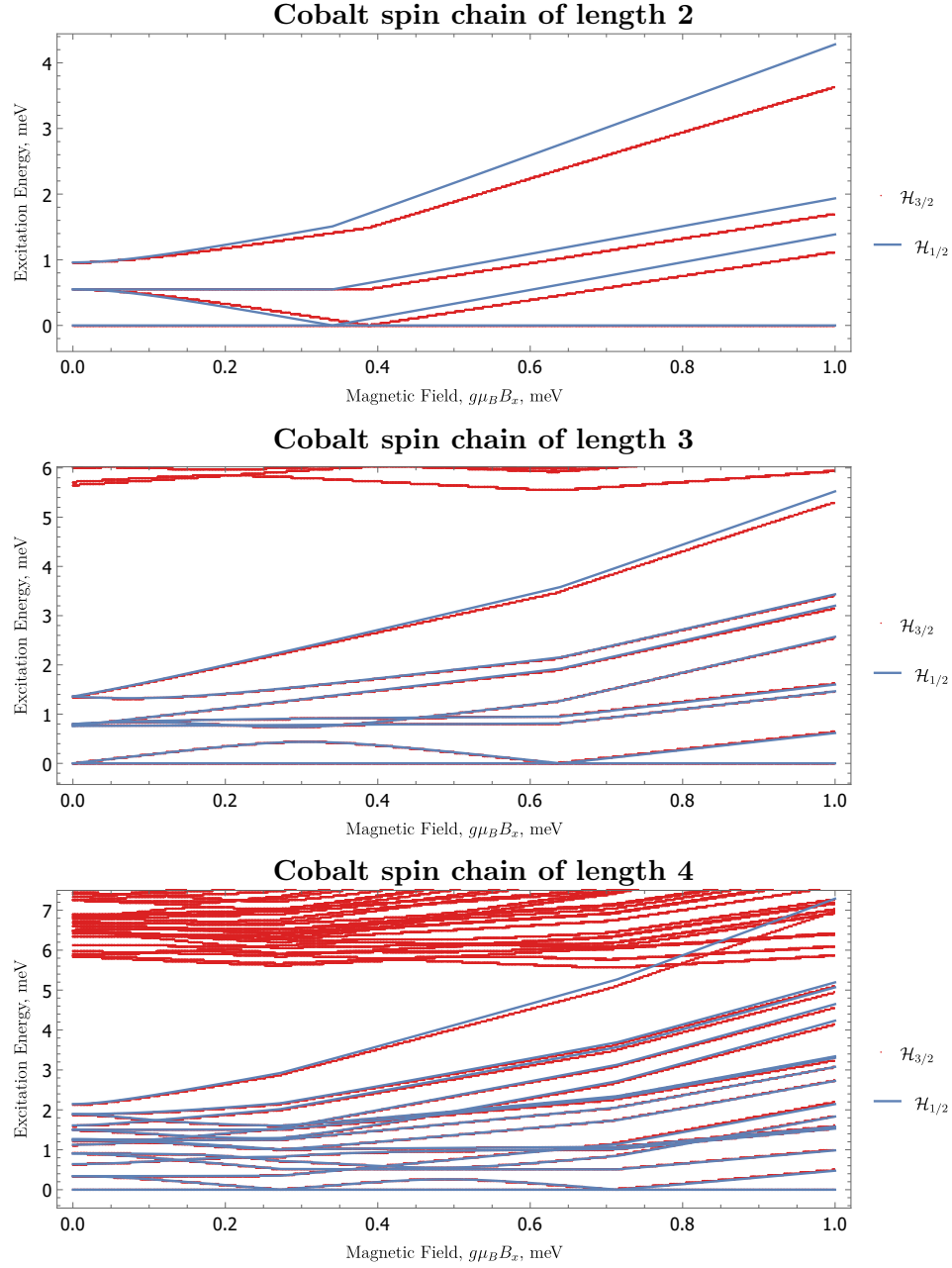


Figure 5: The excitation energies for the 2^N lowest energy states of the cobalt spin chain of length $N = 2, 3$ and 4 , at a varying magnetic field, B_x . Evaluated using the same methods as for Figure 4. This figure shows a much better correlation between the Schrieffer-Wolff transformed, and the original Hamiltonian.

5 Conclusion

Throughout this thesis we have thoroughly examined a Schrieffer Wolff transformation of a one dimensional spin chain of cobalt atoms with nearest neighbor antiferromagnetic isotropic Heisenberg exchange, an external transverse magnetic field, and an anisotropic spin coupling to the surface, with the original Hamiltonian: $\mathcal{H}_{3/2} = J \sum_{i=1}^{N-1} \vec{S}_i \cdot \vec{S}_{i+1} + D \sum_{i=1}^N (S_i^z)^2 - g\mu_B B_x \sum_{i=1}^N S_i^x$

Ending up with an effective spin-1/2 XXZ exchange spin chain with nearest and next nearest coupling in an external magnetic field, with the transformed Hamiltonian:

$$\begin{aligned} \mathcal{H}_{1/2} = & \sum_{i=1}^{N-1} J_{\perp} (S_i^x S_{i+1}^x + S_i^y S_{i+1}^y) + J_z S_i^z S_{i+1}^z \\ & + J_{\perp}^{nnn} \sum_{i=1}^{N-2} S_i^x S_{i+2}^x + S_i^y S_{i+2}^y - \mu_B B_x \sum_{i=1}^N g_i S_i^x \end{aligned}$$

In probing the third order of this transformation we even found evidence of an emerging third nearest neighbor coupling.

Finally we used the transformation up to the second order to explore the energy developments of spin chains of length 2, 3 and 4, and discovered the accuracy of the transformation to be best for excitation energies rather than direct state energy predictions. This means that the model derived in this thesis is ideal for determining the magnetic field strengths for which the ground state transitions, and thereby phase transitions are observed, for longer spin chains. An example of this application can be seen in ref. [4]. One of the shortcomings of this transformation is the increasing complexity of the derivation of higher orders, making it harder to do more precise transformations.

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A Intermediate results in Schrieffer-Wolff Transformation

A.1 Third order

$$\begin{aligned} \frac{J^3}{8} \sum_{i,j,k=1}^{N-1} & \left(\langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_j^+ S_{j+1}^- | l' \rangle \langle l' | S_k^+ S_{k+1}^- | m' \rangle \right. \\ & \left. + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_j^- S_{j+1}^+ | l' \rangle \langle l' | S_k^- S_{k+1}^+ | m' \rangle \right) \end{aligned} \quad (22)$$

for $i = j + 1 = k + 2$ and $i = j - 1 = k - 2$. For $\sum_{l,l'} H'_{ml} H'_{ll'} H'_{l'm'}$ this is the limit in difference between, i, j and k , since at least one atom in l and l' is in a high energy state.

$$\begin{aligned} & \frac{J^3}{8} \sum_{i=1}^{N-3} \left(\langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_{i+1}^+ S_{i+2}^- | l' \rangle \langle l' | S_{i+2}^+ S_{i+3}^- | m' \rangle \right. \\ & \quad \left. + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_{i+1}^- S_{i+2}^+ | l' \rangle \langle l' | S_{i+2}^- S_{i+3}^+ | m' \rangle \right) \\ & + \frac{J^3}{8} \sum_{i=3}^{N-1} \left(\langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_{i-1}^+ S_i^- | l' \rangle \langle l' | S_{i-2}^+ S_{i-1}^- | m' \rangle \right. \\ & \quad \left. + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_{i-1}^- S_i^+ | l' \rangle \langle l' | S_{i-2}^- S_{i-1}^+ | m' \rangle \right) \\ & = \frac{J^3}{8} \sum_{i=1}^{N-3} 36 \left(\langle m | S_i^+ S_{i+1}^+ S_{i+1}^- S_{i+2}^+ S_{i+2}^- S_{i+3}^- | m' \rangle |_{n_l=n_{l'}=1} \right. \\ & \quad \left. + \langle m | S_i^- S_{i+1}^- S_{i+1}^+ S_{i+2}^- S_{i+2}^+ S_{i+3}^+ | m' \rangle |_{n_l=n_{l'}=1} \right) \\ & + \frac{J^3}{8} \sum_{i=3}^{N-1} 36 \left(\langle m | S_i^- S_{i+1}^- S_{i-1}^+ S_i^+ S_{i-2}^+ S_{i-1}^- | m' \rangle |_{n_l=n_{l'}=1} \right. \\ & \quad \left. + \langle m | S_i^+ S_{i+1}^+ S_{i-1}^- S_i^- S_{i-2}^- S_{i-1}^+ | m' \rangle |_{n_l=n_{l'}=1} \right) \\ & = \frac{J^3}{8} \sum_{i=1}^{N-3} 36 \left(\langle m | S_i^+ S_{i+1}^+ S_{i+1}^- S_{i+2}^+ S_{i+2}^- S_{i+3}^- | m' \rangle \right. \\ & \quad \left. + \langle m | S_i^- S_{i+1}^- S_{i+1}^+ S_{i+2}^- S_{i+2}^+ S_{i+3}^+ | m' \rangle |_{n_l=n_{l'}=1} \right) \\ & + \frac{J^3}{8} \sum_{i=3}^{N-1} 36 \left(\langle m | S_{i-2}^+ S_{i-1}^- S_{i-1}^+ S_i^- S_i^+ S_{i+1}^- | m' \rangle \right. \\ & \quad \left. + \langle m | S_{i-2}^- S_{i-1}^+ S_{i-1}^- S_i^+ S_i^- S_{i+1}^+ | m' \rangle |_{n_l=n_{l'}=1} \right) \\ & = \frac{J^3}{8} \sum_{i=1}^{N-3} 36 \left(\langle m | S_i^+ (S_{i+1}^+ S_{i+1}^- S_{i+2}^+ S_{i+2}^- + S_{i+1}^- S_{i+1}^+ S_{i+2}^- S_{i+2}^+) S_{i+3}^- \right. \\ & \quad \left. + S_i^- (S_{i+1}^- S_{i+1}^+ S_{i+2}^- S_{i+2}^+ + S_{i+1}^+ S_{i+1}^- S_{i+2}^+ S_{i+2}^-) S_{i+3}^+ | m' \rangle \right) |_{n_l=n_{l'}=1} \end{aligned} \quad (23)$$

$$\begin{aligned} & = \frac{J^3}{8} \sum_{i=1}^{N-3} 36 \left(\langle m | S_i^+ (S_{i+1}^+ S_{i+1}^- S_{i+2}^+ S_{i+2}^- + S_{i+1}^- S_{i+1}^+ S_{i+2}^- S_{i+2}^+) S_{i+3}^- \right. \\ & \quad \left. + S_i^- (S_{i+1}^- S_{i+1}^+ S_{i+2}^- S_{i+2}^+ + S_{i+1}^+ S_{i+1}^- S_{i+2}^+ S_{i+2}^-) S_{i+3}^+ | m' \rangle \right) |_{n_l=n_{l'}=1} \end{aligned} \quad (24)$$

And now for equation (18):

$$\begin{aligned}
& \frac{J^3}{8} \sum_{i,j,k=1}^{N-1} \left(\left(\langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_j^+ S_{j+1}^- | m'' \rangle \langle m'' | S_k^+ S_{k+1}^- | m' \rangle \right. \right. \\
& \quad + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_j^- S_{j+1}^+ | m'' \rangle \langle m'' | S_k^- S_{k+1}^+ | m' \rangle \Big) \\
& \quad + \left(\langle m | S_i^+ S_{i+1}^- | m'' \rangle \langle m'' | S_j^+ S_{j+1}^- | l \rangle \langle l | S_k^+ S_{k+1}^- | m' \rangle \right. \\
& \quad + \langle m | S_i^- S_{i+1}^+ | m'' \rangle \langle m'' | S_j^- S_{j+1}^+ | l \rangle \langle l | S_k^- S_{k+1}^+ | m' \rangle \Big) \Big) \quad (25)
\end{aligned}$$

$$\begin{aligned}
& \frac{J^3}{8} \sum_{i=1}^{N-3} \left(\left(\langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_{i+1}^+ S_{i+2}^- | m'' \rangle \langle m'' | S_{i+2}^+ S_{i+3}^- | m' \rangle \right. \right. \\
& \quad + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_{i+1}^- S_{i+2}^+ | m'' \rangle \langle m'' | S_{i+2}^- S_{i+3}^+ | m' \rangle \Big) \\
& \quad + \left(\langle m | S_i^+ S_{i+1}^- | m'' \rangle \langle m'' | S_{i+1}^+ S_{i+2}^- | l \rangle \langle l | S_{i+2}^+ S_{i+3}^- | m' \rangle \right. \\
& \quad + \langle m | S_i^- S_{i+1}^+ | m'' \rangle \langle m'' | S_{i+1}^- S_{i+2}^+ | l \rangle \langle l | S_{i+2}^- S_{i+3}^+ | m' \rangle \Big) \Big) \\
& + \frac{J^3}{8} \sum_{i=3}^{N-1} \left(\left(\langle m | S_i^+ S_{i+1}^- | l \rangle \langle l | S_{i-1}^+ S_i^- | m'' \rangle \langle m'' | S_{i-2}^+ S_{i-1}^- | m' \rangle \right. \right. \\
& \quad + \langle m | S_i^- S_{i+1}^+ | l \rangle \langle l | S_{i-1}^- S_i^+ | m'' \rangle \langle m'' | S_{i-2}^- S_{i-1}^+ | m' \rangle \Big) \\
& \quad + \left(\langle m | S_i^+ S_{i+1}^- | m'' \rangle \langle m'' | S_{i-1}^+ S_i^- | l \rangle \langle l | S_{i-2}^+ S_{i-1}^- | m' \rangle \right. \\
& \quad + \langle m | S_i^- S_{i+1}^+ | m'' \rangle \langle m'' | S_{i-1}^- S_i^+ | l \rangle \langle l | S_{i-2}^- S_{i-1}^+ | m' \rangle \Big) \Big) \quad (26)
\end{aligned}$$

$$\begin{aligned}
& = 2 \frac{J^3}{8} \sum_{i=1}^{N-3} 48 \left(\left(\langle m | S_i^+ (S_{i+1}^+ S_{i+1}^- S_{i+2}^- S_{i+2}^+ + S_{i+1}^- S_{i+1}^+ S_{i+2}^+ S_{i+2}^-) S_{i+3}^- \right. \right. \\
& \quad + S_i^- (S_{i+1}^- S_{i+1}^+ S_{i+2}^+ S_{i+2}^- + S_{i+1}^+ S_{i+1}^- S_{i+2}^- S_{i+2}^+) S_{i+3}^+ | m' \rangle \Big) \Big) |_{n_l=1} \quad (27)
\end{aligned}$$

And the total contribution to equation (16) is then:

$$\begin{aligned}
H_{mm'}^{(3)}|_{\mathcal{O}(S_i \cdot S_{i+3})} &= \frac{3J^3}{8D^2\hbar^4} \sum_{i=1}^{N-3} \\
&-4 \left(\langle m| S_i^+ (S_{i+1}^+ S_{i+1}^- S_{i+2}^- S_{i+2}^+ + S_{i+1}^- S_{i+1}^+ S_{i+2}^- S_{i+2}^+) S_{i+3}^- \right. \\
&\quad \left. + S_i^- (S_{i+1}^- S_{i+1}^+ S_{i+2}^- S_{i+2}^+ + S_{i+1}^+ S_{i+1}^- S_{i+2}^- S_{i+2}^+) S_{i+3}^+ |m'\rangle \right) \\
&+3 \left(\langle m| S_i^+ (S_{i+1}^+ S_{i+1}^- S_{i+2}^- S_{i+2}^+ + S_{i+1}^- S_{i+1}^+ S_{i+2}^- S_{i+2}^+) S_{i+3}^- \right. \\
&\quad \left. + S_i^- (S_{i+1}^- S_{i+1}^+ S_{i+2}^- S_{i+2}^+ + S_{i+1}^+ S_{i+1}^- S_{i+2}^- S_{i+2}^+) S_{i+3}^+ |m'\rangle \right) \quad (28) \\
&= \frac{3J^3}{8D^2\hbar^4} \sum_{i=1}^{N-3} \langle m| (S_i^+ S_{i+3}^- + S_i^- S_{i+3}^+) \\
&\quad (-4 (S_{i+1}^+ S_{i+1}^- S_{i+2}^- S_{i+2}^+ + S_{i+1}^- S_{i+1}^+ S_{i+2}^- S_{i+2}^+) \\
&\quad + 3 (S_{i+1}^+ S_{i+1}^- S_{i+2}^- S_{i+2}^+ + S_{i+1}^- S_{i+1}^+ S_{i+2}^- S_{i+2}^+)) |m'\rangle
\end{aligned}$$

The last parenthesis can be reduced using the product of commutators for S^+ and S^- , as done in the deduction of (12), and the reduction $(S_i^- S_i^+ + S_i^+ S_i^-) |m'\rangle = \hbar^2 |m'\rangle$ in the $S = 1/2$ space:

$$\begin{aligned}
&-4 (S_{i+1}^+ S_{i+1}^- S_{i+2}^- S_{i+2}^+ + S_{i+1}^- S_{i+1}^+ S_{i+2}^- S_{i+2}^+) \\
&+ 3 (S_{i+1}^+ S_{i+1}^- S_{i+2}^- S_{i+2}^+ + S_{i+1}^- S_{i+1}^+ S_{i+2}^- S_{i+2}^+) \\
&= \frac{7}{2} ((S_{i+1}^+ S_{i+1}^- S_{i+2}^- S_{i+2}^+ + S_{i+1}^- S_{i+1}^+ S_{i+2}^- S_{i+2}^+) \\
&\quad - (S_{i+1}^+ S_{i+1}^- S_{i+2}^- S_{i+2}^+ + S_{i+1}^- S_{i+1}^+ S_{i+2}^- S_{i+2}^+)) \\
&\quad - \frac{1}{2} (S_{i+1}^+ S_{i+1}^- + S_{i+1}^- S_{i+1}^+) (S_{i+2}^- S_{i+2}^+ + S_{i+2}^+ S_{i+2}^-) \\
&= \frac{7}{2} [S_{i+1}^+, S_{i+1}^-] [S_{i+2}^+, S_{i+2}^-] - \frac{\hbar^4}{2} \\
&= 14\hbar^2 S_{i+1}^z S_{i+2}^z - \frac{\hbar^4}{2} \quad (29)
\end{aligned}$$

Now equation (28) can be written as:

$$\begin{aligned}
H_{mm'}^{(3)}|_{\mathcal{O}(S_i \cdot S_{i+3})} &= \frac{3J^3}{8D^2\hbar^4} \sum_{i=1}^{N-3} \langle m| (S_i^+ S_{i+3}^- + S_i^- S_{i+3}^+) \\
&\quad \left(14\hbar^2 S_{i+1}^z S_{i+2}^z - \frac{\hbar^4}{2} \right) |m'\rangle \quad (30)
\end{aligned}$$

Now using the reduction in equation (8), we get the final result:

$$\begin{aligned}
 H_{mm'}^{(3)}|_{\mathcal{O}(S_i \cdot S_{i+3})} = & -\frac{3J^3}{16D^2} \sum_{i=1}^{N-3} \langle m | (S_i^x S_{i+3}^x + S_i^y S_{i+3}^y) | m' \rangle \\
 & + \frac{21J^3}{4D^2 \hbar^2} \sum_{i=1}^{N-3} \langle m | (S_i^x S_{i+3}^x + S_i^y S_{i+3}^y) (S_{i+1}^z S_{i+2}^z) | m' \rangle
 \end{aligned} \tag{31}$$

B Wolfram Mathematica 11 Energy Developments

The Excitation Energies are found using the same script, but subtracting the lowest energy state of each set of energies from each set of energies.

```
Needs["PacletManager`"]
PacletInstall["C:/users/tai19/Downloads/MaTeX-1.7.3.paclet"]
In[298]:= Needs["MaTeX`"]
```

■ 2-chain of spin-1/2

```
In[299]:= n = 2; S = 1/2; (*n is the number of atoms of spin S*)
m = Table[Table[If[i == j, 1, 0], {i, 2 S + 1}], {j, 2 S + 1}];
(* m is the 2 states an atom can occupy,
e.g. spin up spin down, on the form (1,0) or (0,1)*)
bas = Tuples[m, n]; (*bas is the basis of the system, e.g. the states of the chain,
all the different combinations of n number of m states*)
dim = Dimensions[bas][[1]] (*dim is the dimension of bas,
e.g. the number of orthogonal states for the chain*)
dot[x_, y_] := Product[x[[i]].y[[i]], {i, n}]; (*dot is *)
Svec = Join[ $\frac{1}{2}$  PauliMatrix[#] & /@ {1, 2, 3}, (IdentityMatrix[2 S + 1])];
(*Svec is the spin matrices Sx, Sy, Sz and the identity matrix*)
Sop[k_, q_] [d_] := Table[If[i == k, Svec[[q]], Svec[[4]]].d[[i]], {i, n}]

Out[302]:= 4

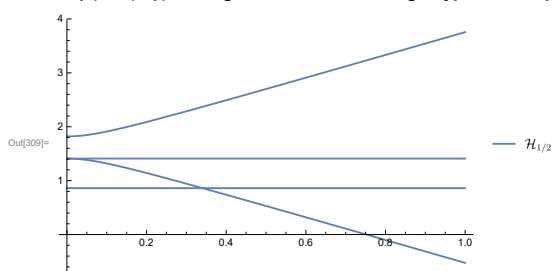
In[306]:= ClearAll[Ham21];
Ham21[Jay_, de_, h_] := IdentityMatrix[dim] n * de / 4 + 4 * Jay * Sum[
  Table[dot[bas[[i]], Sop[1, a] [Sop[2, a] [bas[[j]]]]], {i, dim}, {j, dim}], {a, 2}] +
  (Jay - (39 * Jay^2) / (8 * de)) * Table[dot[bas[[i]], Sop[1, 3] [Sop[2, 3] [bas[[j]]]]],
  {i, dim}, {j, dim}] - h * (2 + 3 * Jay / (2 * de)) * Sum[
  Table[dot[bas[[i]], Sop[1, 1] [Sop[2, 4] [bas[[j]]]]], {i, dim}, {j, dim}], {1, n}];

In[307]:= Eh2[x_] := Eigenvalues[Ham21[0.24, 2.75, x]] // Evaluate // Sort

In[308]:= mineh2 = Min[Eh2[0]]

Out[308]:= 0.860527

In[309]:= Sh2Spec = Plot[Eigenvalues[Ham21[0.24, 2.75, x]] // Evaluate // Sort,
  {x, 0.0, 1}, PlotLegends -> MaTeX /@ LineLegend[{"\\mathcal{H}_{1/2}"}]]
```



■ 3-chain of spin-1/2

```

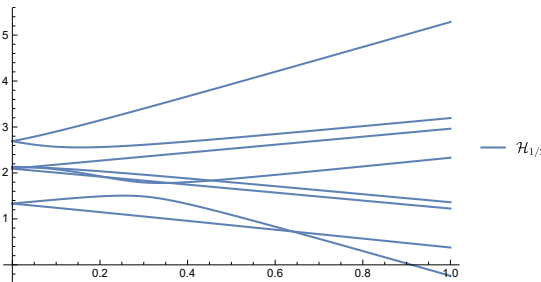
In[310]:= n = 3; S = 1/2;
m = Table[Table[If[i == j, 1, 0], {i, 2 S + 1}], {j, 2 S + 1}];
bas = Tuples[m, n];
dim = Dimensions[bas][[1]];
dot[x_, y_] := Product[x[[i]].y[[i]], {i, n}];
Svec = Join[ $\frac{1}{2}$  PauliMatrix[#] & /@ {1, 2, 3}, {IdentityMatrix[2]}];
Sop[k_, q_] [d_] := Table[If[i == k, Svec[[q]], Svec[[4]]].d[[i]], {i, n}]

In[317]:= Ham31[Jay_, de_, h_] = IdentityMatrix[dim] n * de / 4 +
4 * Jay * Sum[Table[dot[bas[[i]], Sop[1, a] [Sop[1 + 1, a] [bas[[j]]]]],
{i, dim}, {j, dim}], {a, 2}, {1, n - 1}] - (3 * Jay^2 / de) *
Sum[Table[dot[bas[[i]], Sop[1, a] [Sop[1 + 2, a] [bas[[j]]]]], {i, dim}, {j, dim}],
{a, 2}, {1, n - 2}] + (Jay - (39 * Jay^2 / (8 * de))) *
Sum[Table[dot[bas[[i]], Sop[1, 3] [Sop[1 + 1, 3] [bas[[j]]]]], {i, dim}, {j, dim}],
{1, n - 1}] - h * 2 * (1 - 3 * Jay / (2 * de)) *
Sum[Table[dot[bas[[i]], Sop[1, 1] [bas[[j]]]], {i, dim}, {j, dim}], {1, n}] -
(3 * Jay / (2 * de)) * h * (Table[dot[bas[[i]], Sop[1, 1] [bas[[j]]]], {i, dim},
{j, dim}] + Table[dot[bas[[i]], Sop[n, 1] [bas[[j]]]], {i, dim}, {j, dim}]);

In[318]:= EEE[h_] = Eigenvalues[Ham31[0.24, 2.75, h]];

In[319]:= Sh3Spec =
Plot[EEE[x], {x, 0, 1}, PlotLegends -> MaTeX /@ LineLegend[{" $\mathcal{H}_{1/2}$ "}]]

```



■ 4-chain of spin-1/2

```

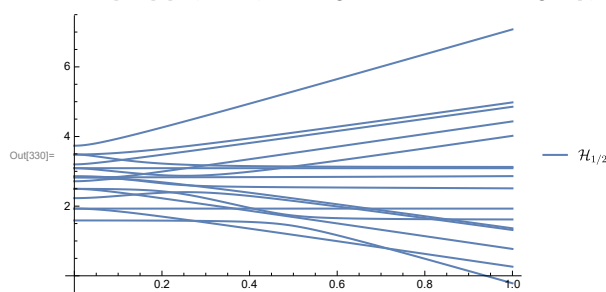
In[320]:= Clear[n, S, m, bas, dim]
n = 4; S = 1/2;
m = Table[Table[If[i == j, 1, 0], {i, 2 S + 1}], {j, 2 S + 1}];
bas = Tuples[m, n];
dim = Dimensions[bas][[1]];
dot[x_, y_] := Product[x[[i]].y[[i]], {i, n}];
Svec = Join[ $\frac{1}{2}$  PauliMatrix[#] & /@ {1, 2, 3}, {IdentityMatrix[2]}];
Sop[k_, q_] [d_] := Table[If[i == k, Svec[[q]], Svec[[4]]].d[[i]], {i, n}]

```

```
In[328]= Ham41[Jay_, de_, h_] = IdentityMatrix[dim] n * de / 4 +
4 * Jay * Sum[Table[dot[bas[[i]], Sop[1, a][Sop[1 + 1, a][bas[[j]]]]],
{i, dim}, {j, dim}], {a, 2}, {1, n - 1}] - (3 * Jay^2 / de) *
Sum[Table[dot[bas[[i]], Sop[1, a][Sop[1 + 2, a][bas[[j]]]]], {i, dim}, {j, dim}],
{a, 2}, {1, n - 2}] + (Jay - (39 * Jay^2 / (8 * de))) *
Sum[Table[dot[bas[[i]], Sop[1, 3][Sop[1 + 1, 3][bas[[j]]]]], {i, dim}, {j, dim}],
{1, n - 1}] - h * 2 * (1 - 3 * Jay / (2 * de)) *
Sum[Table[dot[bas[[i]], Sop[1, 1][bas[[j]]]], {i, dim}, {j, dim}], {1, n}] -
(3 * Jay / (2 * de)) * h * (Table[dot[bas[[i]], Sop[1, 1][bas[[j]]]], {i, dim},
{j, dim}] + Table[dot[bas[[i]], Sop[n, 1][bas[[j]]]], {i, dim}, {j, dim}]);

In[329]= Eh4[x_] := Eigenvalues[Ham41[0.24, 2.75, x]]

In[330]= Sh4Spec =
Plot[Eh4[x], {x, 0, 1}, PlotLegends -> MaTeX /@ LineLegend[{"\\mathcal{H}_{1/2}"]}]
```



■ 2-chain of spin-3/2 (Numerically evaluated)

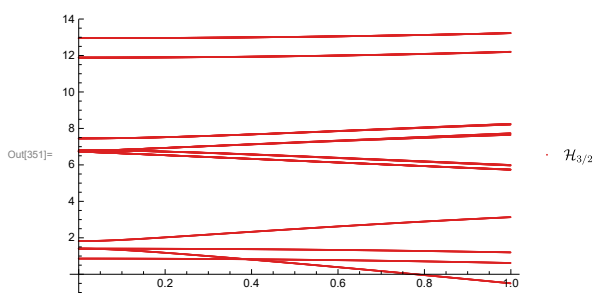
```
In[331]= n = 2; S = 3/2;
m = Table[Table[If[i == j, 1, 0], {i, 2 S + 1}], {j, 2 S + 1}];
bas = Tuples[m, n];
dim = Dimensions[bas][[1]];
dot[x_, y_] := Product[x[[i]].y[[i]], {i, n}];
Sx = 1/2 {{0, sqrt[3], 0, 0}, {sqrt[3], 0, 2, 0}, {0, 2, 0, sqrt[3]}, {0, 0, sqrt[3], 0}};
Sy = 1/(2 * I) {{0, sqrt[3], 0, 0}, {-sqrt[3], 0, 2, 0}, {0, -2, 0, sqrt[3]}, {0, 0, -sqrt[3], 0}};
Sz = {{3/2, 0, 0, 0}, {0, 1/2, 0, 0}, {0, 0, -1/2, 0}, {0, 0, 0, -3/2}};
Svec = {Sx, Sy, Sz, IdentityMatrix[4]} // N
Sop[k_, q_][d_] := Table[If[i == k, Svec[[q]], Svec[[4]].d[[i]], {i, n}]]

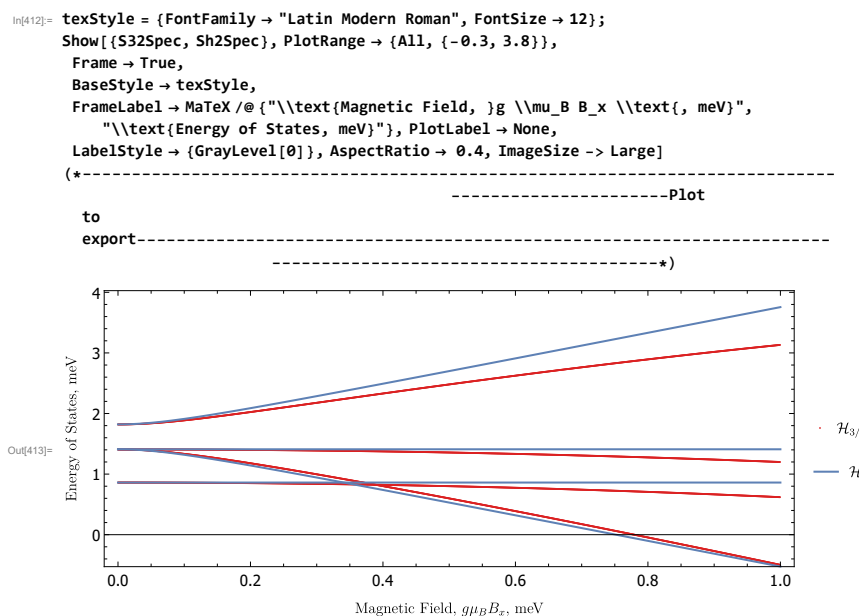
Out[339]= {{ {0., 0.866025, 0., 0.}, {0.866025, 0., 1., 0.},
{0., 1., 0., 0.866025}, {0., 0., 0.866025, 0.}},
{ {0., 0. - 0.866025 i, 0., 0.}, {0. + 0.866025 i, 0., 0. - 1. i, 0.},
{0., 0. + 1. i, 0., 0. - 0.866025 i}, {0., 0., 0. + 0.866025 i, 0.}},
{ {1.5, 0., 0., 0.}, {0., 0.5, 0., 0.}, {0., 0., -0.5, 0.}, {0., 0., 0., -1.5}},
{ {1., 0., 0., 0.}, {0., 1., 0., 0.}, {0., 0., 1., 0.}, {0., 0., 0., 1.}} }
```

4 | Spektrum_sammenligning_Final_AppendixVersion.nb

```
In[341]:= d = 2.75;
jay = 0.24;
ClearAll[b];
Ham23 =
  d * Sum[Table[dot[bas[[i]], Sop[1, 3][Sop[1, 3][bas[[j]]]]], {i, dim}, {j, dim}],
  {1, n}] + jay * Sum[Table[dot[bas[[i]], Sop[1, a][Sop[1 + 1, a][bas[[j]]]]],
  {i, dim}, {j, dim}], {a, 3}, {1, n - 1}] -
  b * Sum[Table[dot[bas[[i]], Sop[1, 1][bas[[j]]]], {i, dim}, {j, dim}], {1, n}];
Ham23 = Chop[Ham23, 10^-14];

In[346]:= Hamf23[b_] = Ham23;
nb = 1500;
rb = 1.;
Eners = Table[Eigenvalues[Hamf23[(b - 1) * rb / nb]] +
  mineh2 - Min[Eigenvalues[Hamf23[0]]], {b, nb}];
Colors = Table[Red, {nb}];
S32Spec =
  ListPlot[Table[Transpose[{Table[(1 - 1) * rb / nb, i = dim], Eners[[1]]}], {1, nb}],
  PlotStyle -> ColorData["Rainbow"],
  PlotLegends -> MaTeX /@ LineLegend[{"\\mathcal{H}_{3/2}"]]
```





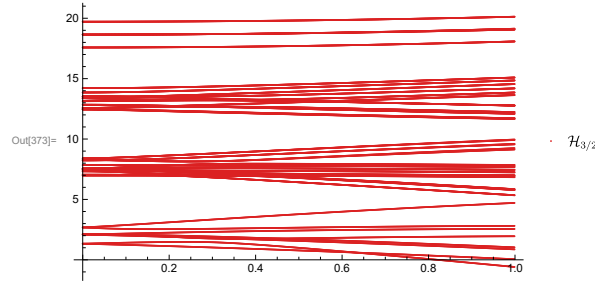
■ 3-chain of spin-3/2 (Numerically evaluated)

```
In[354]:= n = 3; S = 3/2;
m = Table[Table[If[i == j, 1, 0], {i, 2 S + 1}], {j, 2 S + 1}];
bas = Tuples[m, n];
dim = Dimensions[bas][[1]]
dot[x_, y_] := Product[x[[i]].y[[i]], {i, n}];
Sx = 1/2 {{0, sqrt[3], 0, 0}, {sqrt[3], 0, 2, 0}, {0, 2, 0, sqrt[3]}, {0, 0, sqrt[3], 0}};
Sy = 1/(2*I) {{0, sqrt[3], 0, 0}, {-sqrt[3], 0, 2, 0}, {0, -2, 0, sqrt[3]}, {0, 0, -sqrt[3], 0}};
Sz = {{3/2, 0, 0, 0}, {0, 1/2, 0, 0}, {0, 0, -1/2, 0}, {0, 0, 0, -3/2}};
Svec = {Sx, Sy, Sz, IdentityMatrix[4]} // N;
Sop[k_, q_] [d_] := Table[If[i == k, Svec[[q]], Svec[[4]]].d[[i]], {i, n}]
Out[357]= 64

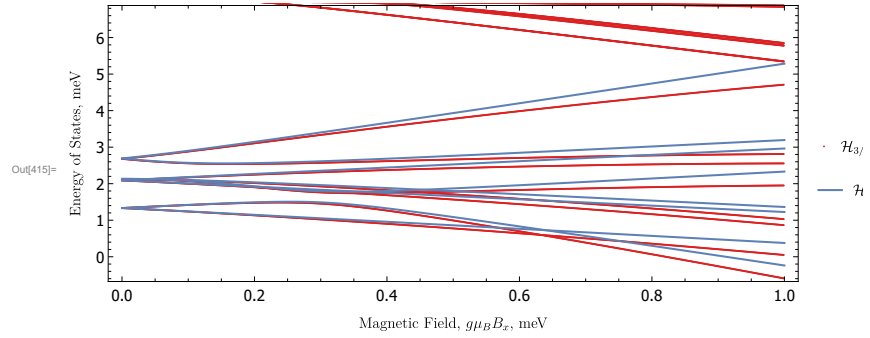
In[364]:= d = 2.75; (* <----- d*)
jay = 0.24;
ClearAll[b];
Ham33 =
d * Sum[Table[dot[bas[[i]], Sop[1, 3][Sop[1, 3][bas[[j]]]]], {i, dim}, {j, dim}],
{1, n}] + jay * Sum[Table[dot[bas[[i]], Sop[1, a][Sop[1 + 1, a][bas[[j]]]]],
{i, dim}, {j, dim}], {a, 3}, {1, n - 1}] -
b * Sum[Table[dot[bas[[i]], Sop[1, 1][bas[[j]]]], {i, dim}, {j, dim}], {1, n}];
Hamf33[b_] = Chop[Ham33, 10^-10];
```

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```
In[369]= nb = 1000;
rb = 1;
Eners33 = Table[Eigenvalues[Hamf33[(b - 1) * rb / nb]] +
  Min[EEE[0]] - Min[Eigenvalues[Hamf33[0]]], {b, nb}];
(*-----Her Skal der plusses-----*)
Colors = Table[Red, {nb}];
S33Spec = ListPlot[
  Table[Transpose[{Table[(1 - 1) * rb / nb, i = dim], Sort[Eners33[[1]]]}, {1, nb}],
  PlotStyle -> ColorData["Rainbow"],
  PlotLegends -> MaTeX /@ LineLegend[{"\\mathcal{H}_{3/2}"}]]
```



```
In[414]= textStyle = {FontFamily -> "Latin Modern Roman", FontSize -> 12};
Show[{S33Spec, Sh3Spec},
  PlotRange -> {{0, rb}, {-0.3, 6.5}},
  Frame -> True,
  BaseStyle -> textStyle,
  FrameLabel -> MaTeX /@ {"\\text{Magnetic Field, } g \\mu_B B_x \\text{ meV}",
    "\\text{Energy of States, meV}"},
  PlotLabel -> None,
  LabelStyle -> {GrayLevel[0]}, AspectRatio -> 0.4, ImageSize -> Large]
(*-----Plot
to export-----*)
```



■ 4-chain of spin-3/2 (Numerically evaluated)

```

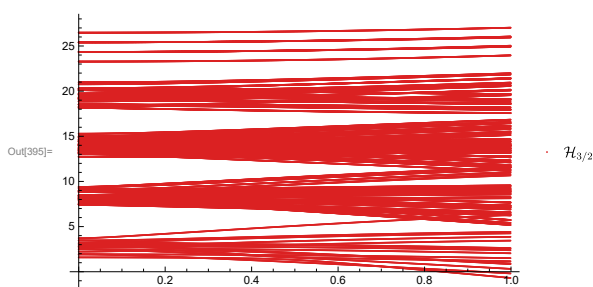
In[376]= n = 4; S = 3/2;
m = Table[Table[If[i == j, 1, 0], {i, 2 S + 1}], {j, 2 S + 1}];
bas = Tuples[m, n];
dim = Dimensions[bas][[1]]
dot[x_, y_] := Product[x[[i]].y[[i]], {i, n}];
Sx = 1/2 {{0, Sqrt[3], 0, 0}, {Sqrt[3], 0, 2, 0}, {0, 2, 0, Sqrt[3]}, {0, 0, Sqrt[3], 0}};
Sy = 1/(2 * I) {{0, Sqrt[3], 0, 0}, {-Sqrt[3], 0, 2, 0}, {0, -2, 0, Sqrt[3]}, {0, 0, -Sqrt[3], 0}};
Sz = {{3/2, 0, 0, 0}, {0, 1/2, 0, 0}, {0, 0, -1/2, 0}, {0, 0, 0, -3/2}};
Svec = {Sx, Sy, Sz, IdentityMatrix[4]} // N;
Sop[k_, q_] [d_] := Table[If[i == k, Svec[[q]], Svec[[4]]].d[[i]], {i, n}]

Out[379]= 256

In[380]= d = 2.75; (* <----- d *)
jay = 0.24;
ClearAll[b];
Ham43 =
  d * Sum[Table[dot[bas[[i]], Sop[1, 3][bas[[j]]]], {i, dim}, {j, dim}],
    {1, n}] + jay * Sum[Table[dot[bas[[i]], Sop[1, a][bas[[j]]]],
    {i, dim}, {j, dim}], {a, 3}, {1, n - 1}] -
  b * Sum[Table[dot[bas[[i]], Sop[1, 1][bas[[j]]]], {i, dim}, {j, dim}], {1, n}];
Hamf43[b_] = Chop[Ham43, 10^-10];

In[391]= nb = 1000;
rb = 1;
Eners43 = Table[Eigenvalues[Hamf43[(b - 1) * rb / nb]] +
  Min[Eh4[0]] - Min[Eigenvalues[Hamf43[0]]], {b, nb}];
(*-----Her Skal der plusses-----*)
Colors = Table[Red, {nb}];
S43Spec = ListPlot[
  Table[Transpose[{Table[(1 - 1) * rb / nb, i = dim], Sort[Eners43[[1]]]}], {1, nb}],
  PlotStyle -> ColorData["Rainbow"],
  PlotLegends -> MaTeX /@ LineLegend[{"\\mathcal{H}_{3/2}"}]]

```



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```
In[416]= texStyle = {FontFamily -> "Latin Modern Roman", FontSize -> 12};
Show[{$43Spec, $h4Spec},
PlotRange -> {{0, rb}, {-0.3, 6.5}},
Frame -> True,
BaseStyle -> texStyle,
FrameLabel -> MaTeX /@ {"\\text{Magnetic Field, } g \\mu_B B_x \\text{, meV}",
"\\text{Energy of States, meV}"},
PlotLabel -> None,
LabelStyle -> {GrayLevel[0]}, AspectRatio -> 0.4, ImageSize -> Large]
(*-----Plot to export-----*)
-----*)
```

