Investigation of the Inelastic Tunneling Current for Electron Tunneling Spectroscopy on a Chain of Magnetic Atoms

by

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Abstract

The aim of this report is to investigate the inelastic tunneling current that would be measured during an inelastic electron tunneling spectroscopy experiment performed on a one dimensional chain of magnetic atoms.

A chain consisting of \( n \) magnetic iron atoms with spin 2 has been examined. The spins of the iron atoms in the chain have been treated quantum mechanically. The spins of the atoms in the chain are governed by the time independent spin Hamiltonian. A method has been developed to find the eigenstates and eigenenergies of the Hamiltonian. The eigenstates of the Hamiltonian live in a \( 5^n \) dimensional Hilbert space. The eigenstates and eigenenergies of the chain have been used to predict the inelastic tunneling current that would be measured during an inelastic electron tunneling spectroscopy experiment. Furthermore, the eigenstates have been investigated for 'spin waves'. Note that these 'spin waves' are not waves in time, but wavelike 'motions' in Hilbert space.

Results show that the current increases when the voltage increases. The differential conductance exhibits a jump if a state transition occurs. A system with a high temperature has a higher current and the current starts increasing for lower voltages than a system with lower temperature. Changing the position of the STM tip changes the interaction with the chain, resulting in different state transitions to occur. The influence of the location of the STM tip is the same for symmetric atoms in the chain. Increasing the number of atoms in the chain causes more jumps in the differential conductance, but the jumps are smaller. Decreasing the number of atoms causes less jumps in the differential conductance, but the jumps are bigger. The eigenstates are superpositions of z-basis elements and wavelike 'motions' occur in the eigenstates.
Magnets are used in many different devices, for example there are magnets in compasses, in electric motors and magnets are even used as nice decorations for fridges. But magnets are used to store data too, a good example is the hard disk drive in a computer. A hard disk drive is a big device used for storing data, but is it possible to store data in smaller objects? One of the smallest objects known to mankind is the atom. It would be fantastic if data could be stored in something as small as an atom or a one dimensional chain of atoms. Loth et al. already managed to store eight bits using eight side by side one dimensional chains with a total of only 96 iron atoms. [3]

It is possible to make chains of magnetic atoms using a Scanning Tunneling Microscope. Inelastic electron tunneling spectroscopy experiments have been performed on chains of magnetic atoms and a model has been described by J. Fernández-Rossier. [2] Furthermore, spin waves in an chain of iron atoms have been observed during an experiment performed by Spinelli et al.. [4]

During this research a one dimensional chain of magnetic iron atoms will be examined in equilibrium. The spins of the iron atoms in the chain will be treated quantum mechanically. The spins of the atoms in the chain are governed by the time independent spin Hamiltonian. A method will be developed to find the eigenstates and eigenenergies of the Hamiltonian. Then the eigenstates and eigenenergies of the chain will be used to predict the inelastic tunneling current that would be measured during an inelastic electron tunneling spectroscopy experiment. Furthermore, the eigenstates will be investigated for ‘spin waves’. Note that these ‘spin waves’ are not waves in time.

J. Assendelft
Delft, July 2016
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<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>$s$</td>
<td>Spin of an atom</td>
<td></td>
</tr>
<tr>
<td>$m_s$</td>
<td>Z-component of the spin</td>
<td></td>
</tr>
<tr>
<td>$</td>
<td>s m_s\rangle$</td>
<td>Representation of the spin and z-component of the spin for a single atom</td>
</tr>
<tr>
<td>$</td>
<td>2 m_{s_1}\rangle...</td>
<td>2 m_{s_n}\rangle$</td>
</tr>
<tr>
<td>$</td>
<td>m_1 m_2 \ldots m_n\rangle$</td>
<td>Abbreviation of the representation of a chain consisting of $n$ atoms with spin 2</td>
</tr>
<tr>
<td>$\tilde{H}$</td>
<td>Hamiltonian operator</td>
<td></td>
</tr>
<tr>
<td>$\tilde{H}_S$</td>
<td>Single-spin Hamiltonian operator</td>
<td>[J]</td>
</tr>
<tr>
<td>$D$</td>
<td>Single-spin longitudinal magnetocrystalline anisotropy</td>
<td>[J]</td>
</tr>
<tr>
<td>$E$</td>
<td>Transverse magnetic anisotropy</td>
<td>[J]</td>
</tr>
<tr>
<td>$g^*$</td>
<td>Landé g-factor</td>
<td></td>
</tr>
<tr>
<td>$\mu_B$</td>
<td>Bohr magneton</td>
<td>[J/T]</td>
</tr>
<tr>
<td>$B = B\hat{z}$</td>
<td>Magnetic field in the z-direction</td>
<td>[T]</td>
</tr>
<tr>
<td>$E_z$</td>
<td>Equal to $g^* \mu_B B$</td>
<td>[J]</td>
</tr>
<tr>
<td>$\hat{S}$</td>
<td>Spin operator</td>
<td></td>
</tr>
<tr>
<td>$\hat{S}_z$</td>
<td>Z-component of the spin operator</td>
<td></td>
</tr>
<tr>
<td>$\hat{S}_+(s m_s)$</td>
<td>Z-component of the spin operator acting on a single atom which is equal to $m_s</td>
<td>s m_s\rangle$</td>
</tr>
<tr>
<td>$\hat{S}_+$</td>
<td>Spin raising operator</td>
<td></td>
</tr>
<tr>
<td>$\hat{S}_-(s m_s)$</td>
<td>Spin lowering operator acting on a single atom which is equal to $\sqrt{s(s+1)} - m_s(m_s+1)\hat{S}_z</td>
<td>[J]$</td>
</tr>
<tr>
<td>$\hat{S}_-$</td>
<td>Spin lowering operator acting on a single atom which is equal to $\sqrt{s(s+1)} - m_s(m_s-1)\hat{S}_z</td>
<td>[J]$</td>
</tr>
<tr>
<td>$\hat{S}_x$</td>
<td>X-component of the spin operator and equal to $\frac{1}{2} (\hat{S}<em>+ - \hat{S}</em>-)</td>
<td>[J]$</td>
</tr>
<tr>
<td>$\hat{S}_y$</td>
<td>Y-component of the spin operator and equal to $\frac{1}{2} i (\hat{S}<em>+ - \hat{S}</em>-)</td>
<td>[J]$</td>
</tr>
<tr>
<td>$</td>
<td>M\rangle$</td>
<td>Eigenstate of the Hamiltonian</td>
</tr>
<tr>
<td>$E_M$</td>
<td>Eigenenergy of eigenstate $M</td>
<td>[J]$</td>
</tr>
<tr>
<td>$M_{tot}$</td>
<td>Total number of eigenstates</td>
<td></td>
</tr>
<tr>
<td>$\langle M</td>
<td>M'\rangle$</td>
<td>Inner product of eigenstate $M$ and $M'</td>
</tr>
<tr>
<td>$I_{n\times n}$</td>
<td>$n$ by $n$ identity matrix</td>
<td></td>
</tr>
<tr>
<td>$V$</td>
<td>Voltage</td>
<td>[V]</td>
</tr>
<tr>
<td>$I$</td>
<td>Current</td>
<td>[A]</td>
</tr>
<tr>
<td>$\frac{dI}{dV}$</td>
<td>Differential conductance</td>
<td>[A/V]</td>
</tr>
<tr>
<td>$\Delta E_{M,M'}$</td>
<td>Energy difference between eigenstate $M$ and eigenstate $M'</td>
<td>[J]$</td>
</tr>
<tr>
<td>$P_M$</td>
<td>Probability for a system to occupy eigenstate $M</td>
<td>[J]$</td>
</tr>
<tr>
<td>$e$</td>
<td>Elementary charge</td>
<td>[C]</td>
</tr>
<tr>
<td>$\hbar$</td>
<td>Planck constant</td>
<td>[J s]</td>
</tr>
<tr>
<td>$eG_S$</td>
<td>Quantum conductance</td>
<td>[A/V]</td>
</tr>
<tr>
<td>$k_B$</td>
<td>Boltzmann constant</td>
<td>[J/K]</td>
</tr>
<tr>
<td>$T$</td>
<td>Temperature</td>
<td>[K]</td>
</tr>
<tr>
<td>$\beta$</td>
<td>Equal to $(k_B T)^{-1}$</td>
<td>[J$^{-1}$]</td>
</tr>
</tbody>
</table>
In this chapter a one dimensional chain, an IETS experiment and a two dimensional Hilbert space are visualized.

1.1. One dimensional chain
In this report one dimensional chains of magnetic atoms are researched. A chain can be constructed with a scanning tunneling microscope (STM). The STM places the magnetic atoms on a substrate next to each other forming a chain. In figure 1.1 a schematic representation of a three particle system is displayed. The red circles represent the atoms and the yellow rectangle represents the substrate.

![Figure 1.1: Schematic representation of the three particle system. Three atoms deposited on a substrate. The red circles represent the atoms and the yellow rectangle represents the substrate.](image1.jpg)

1.2. Inelastic electron tunneling spectroscopy experiment
An inelastic electron tunneling spectroscopy (IETS) experiment is performed with a STM. In figure 1.2 a schematic representation of an IETS experiment performed on a three particle system is displayed. The tip of the STM is represented by the blue circles. The STM tip is placed above an atom of the chain, in figure 1.2 above the left atom of the chain. A voltage is applied between the tip and the chain. It is possible for electrons to tunnel from the tip to the atom below the tip. The tunneling electrons cause a small current. The tunneling electrons interact with the atom below the tip. The interaction can result in an increase in current, this will be explained in section 2.5.
1. Introduction

Figure 1.2: Schematic representation of an IETS experiment performed on a three particle system. Three atoms deposited on a substrate. The red circles represent the atoms and the yellow rectangle represents the substrate. The tip of the STM is represented by the blue circles and is positioned above the left atom of the chain. A voltage is applied between the tip and the chain. It is possible for electrons to tunnel from the tip to the atom below the tip causing a small current.

1.3. Hilbert space

Hilbert space is a vector space. Hilbert space is a complete space with an inner product. Hilbert space can have any finite number of dimensions. The spin of an atom is a vector, see section 2.1. The spin can be displayed in z-basis elements. The z-basis elements form an orthonormal set. Thus the z-basis element form an orthonormal basis for the Hilbert space. For example, the z-basis elements of a chain of two iron atom with spin 2 form an orthonormal basis for a 25 dimensional Hilbert space, see section 2.2. The Hilbert space can be visualized in the following way, using the representation described with formula 2.1:

\[
\begin{align*}
| −2 & −2 \rangle & | −2 & −1 \rangle & | −2 & 0 \rangle & | −2 & 1 \rangle & | −2 & 2 \rangle \\
| −1 & −2 \rangle & | −1 & −1 \rangle & | −1 & 0 \rangle & | −1 & 1 \rangle & | −1 & 2 \rangle \\
| 0 & −2 \rangle & | 0 & −1 \rangle & | 0 & 0 \rangle & | 0 & 1 \rangle & | 0 & 2 \rangle \\
| 1 & −2 \rangle & | 1 & −1 \rangle & | 1 & 0 \rangle & | 1 & 1 \rangle & | 1 & 2 \rangle \\
| 2 & −2 \rangle & | 2 & −1 \rangle & | 2 & 0 \rangle & | 2 & 1 \rangle & | 2 & 2 \rangle 
\end{align*}
\]

with each z-basis element representing a different vector in the 25 dimensional Hilbert space.
During the research a one dimensional chain of magnetic atoms with spin 2 will be examined. Magnetic atoms are examined because they are affected by an applied magnetic field. Some of the magnetic atoms have spin 2, for example iron. Moreover, iron atoms can be used to create a one dimensional chain. [4]

2.1. Spin
Spin is the rotation of the atom around its own axis. Spin is a quantum mechanical concept. Spin is a vector, it has a magnitude and a direction. Spin 2 denotes the magnitude of the spin is 2. Due to the uncertainty principle it is only possible to know the spin component in one direction. During the research the z-component of the spin will be used. The representation of the spin of a particle is $|s m_s\rangle$, where $s$ equals the spin of the particle and $m_s$ equals the spin in the z-direction. $m_s$ ranges from $-s$ to $s$ with integer steps, because the direction of the spin is quantized. Thus for particles with spin 2: $m_s = -2, -1, 0, 1$ or 2. For example when the spin is pointed upwards along the z-axis then the representation of the spin of the particle is $|2 2\rangle$.

2.2. Spins of a one dimensional chain of magnetic atoms
A one dimensional chain of magnetic atoms with spin 2 can be created with a scanning tunneling microscope (STM). [4] The representation of a chain of $n$ atoms with spin 2 is $|2 m_{s_1}\rangle|2 m_{s_2}\rangle...|2 m_{s_{n-1}}\rangle|2 m_{s_n}\rangle$. In this report $|m_1 m_2 ... m_n\rangle$ will be used as a shorter notation for a chain of $n$ atoms. The chain of atoms must be considered as one system. The system will be examined in equilibrium. The system has many z-basis elements, a chain of $n$ particles has $5^n$ different z-basis elements, because each particle has spin 2. The z-basis elements are orthonormal. The z-basis elements form an orthonormal basis for the $5^n$ dimensional Hilbert space.

2.3. Hamiltonian
The system can change over time from element to element or to a superposition of multiple elements. How the system changes elements is governed by the Hamiltonian. The Hamiltonian is the energy operator. The Hamiltonian used in this research for a chain of $n$ atoms equals: [1] [2] [4]

$$\mathcal{H} = \sum_{i=1}^{n} \mathcal{H}_S^{(i)} + \sum_{i=1}^{n-1} J \hat{S}_i^{(i)} \cdot \hat{S}_i^{(i+1)}$$

with

$$\mathcal{H}_S^{(i)} = D \left( s_{z}^{(i)} \right)^2 + E \left( \left( s_{x}^{(i)} \right)^2 - \left( s_{y}^{(i)} \right)^2 \right) - g^* \mu_B B \cdot \hat{S}_i^{(i)}.$$

$\mathcal{H}_S$ is the single-spin Hamiltonian, $D$ represents the single-spin longitudinal magnetocrystalline anisotropy, $E$ the transverse magnetic anisotropy, $g^*$ the Landé g-factor, $\mu_B$ the Bohr magneton, B the external magnetic
field and $J$ the exchange energy between neighbouring atoms. $\hat{S}^{(i)}$ is the spin operator acting on atom $i$ of the system, with $\hat{S}_x^{(i)}$, $\hat{S}_y^{(i)}$ and $\hat{S}_z^{(i)}$ the $x$, $y$ and $z$ components of the spin operator acting on atom $i$ of the system. [1]

The first sum of the $\hat{H}_S^{(i)}$ accounts for the individual spin of atom $i$ reacting to the external magnetic field and anisotropies. The second sum accounts for the influence of the neighbouring atoms on each other. This sum is known as the Heisenberg Hamiltonian and is frequently used in describing magnetic systems where the spins of the atoms are treated quantum mechanically.

An unperturbed system will always be in an eigenstate of the Hamiltonian. $|M\rangle$ is an eigenstate of the Hamiltonian if $\hat{H}(M) = E_M|M\rangle$ and $\langle M|M\rangle = 1$ hold, with $E_M$ the eigenenergy of eigenstate $|M\rangle$. How to find the eigenstates and eigenenergies will be discussed in chapter 3.

2.4. Inner product and matrix properties

In chapter 3 and in the appendix many inner products and matrices are used, thus in this section some inner product properties and matrix properties will be discussed. These properties will be used to find the eigenstates and eigenenergies will be discussed in chapter 3.

The $z$-basis elements form an orthonormal basis for the $5^n$ dimensional Hilbert space, thus:

$$\langle m_{a_1} \ldots m_{a_i} \ldots m_{a_n} | m_{b_1} \ldots m_{b_i} \ldots m_{b_n} \rangle = \begin{cases} 1, & \text{if } m_{a_i} = m_{b_i} \text{ for } i = 1,2,\ldots,n. \\ 0, & \text{otherwise.} \end{cases}$$

Matrix $A$ is a real symmetric $n$ by $n$ matrix if each matrix element $a_{ij}$ is real and $a_{ij} = a_{ji}$ for each matrix element $a_{ij}$. Some important properties of a real symmetric $n$ by $n$ matrix are: there are $n$ real eigenvalues and eigenvectors from different eigenvalues are orthogonal. For a real symmetric $n$ by $n$ matrix the set of eigenvectors is orthogonal, thus by scaling the eigenvectors they can be made orthonormal.

The $n$ by $n$ identity matrix is denoted by $I_{n \times n}$. The $n$ eigenvalues of $I_{n \times n}$ are all 1 and every vector with dimension $n$ is an eigenvector of $I_{n \times n}$. Thus for a constant $c$, then the eigenvalues of $cI_{n \times n}$ are all $c$ and every vector with dimension $n$ is an eigenvector of $cI_{n \times n}$. Let $A$ and $B$ be $n$ by $n$ matrices and $c$ a constant such that $A = cI_{n \times n} + B$. Let $\lambda_B$ be an eigenvalue of matrix $B$, thus $\det (B - \lambda_B I_{n \times n}) = 0$, where $\det (B - \lambda_B I_{n \times n})$ means the determinant of $B - \lambda_B I_{n \times n}$. Now $\lambda_A = c + \lambda_B$ is an eigenvalue of matrix $A$, because

$$\det (A - \lambda_A I_{n \times n}) = \det (cI_{n \times n} + B - (c + \lambda_B) I_{n \times n}) = \det (cI_{n \times n} + B - cI_{n \times n} - \lambda_B I_{n \times n}) = \det (B - \lambda_B I_{n \times n}) = 0.$$

If $v$ is an eigenvector of matrix $B$, then $v$ is an eigenvector of matrix $A$, because $A = cI_{n \times n} + B$ and $v$ is an eigenvector of $cI_{n \times n}$.

2.5. Inelastic tunneling current

The spins of the atoms cannot be measured directly, but the spin dynamics of the system can be examined with an inelastic electron tunneling spectroscopy (IETS) experiment. In an IETS experiment, the spin of an electron tunneling from the tip of a STM interacts via exchange with the spin of the atom below the tip. When the energy provided by the bias voltage matches the energy of an atomic spin transition, the transition can occur. For a chain of $n$ magnetic atoms and the STM tip located above atom $j$ in the chain, then the inelastic tunneling current $I(V)$ in an IETS experiment is given by: [1] [2]

$$I(V) = G_S \sum_{M,M',M''=x,y,z} P_M |\langle M|\hat{S}_a^{(j)}|M'\rangle|^2 F_{M,M',a}(V)$$

with

$$F_{M,M',a}(V) = \frac{eV - s\Delta_{M',M}}{1 - e^{-\beta e(V - s\Delta_{M',M})}} + \frac{eV + s\Delta_{M',M}}{1 - e^{\beta e(V + s\Delta_{M',M})}}.$$  

Here $|M\rangle$ and $|M'\rangle$ are eigenstates of the system and the sum is over all possible combinations of two eigenstates. And

$$\Delta_{M',M} = E_{M'} - E_M$$

is the energy difference between eigenstate $|M\rangle$ and $|M'\rangle$, with $E_{M'}$ the eigenenergy of eigenstate $|M'\rangle$ and $E_M$ the eigenenergy of eigenstate $|M\rangle$. $s$ is the spin of the atoms, in this research atoms with spin 2 are used, thus $s = 2$. $e$ is the elementary charge, $V$ is the applied voltage and $\beta = (k_B T)^{-1}$. $\hat{S}_a^{(j)}$ with $\alpha = x, y$ or $z$ is the
2.5. Inelastic tunneling current

The $\alpha$ component of the spin operator acting on atom $j$. $P_M$ is the probability to occupy eigenstate $|M\rangle$. Because the system is in equilibrium, the probability follows from the Boltzmann distribution. Hence

$$P_M = \frac{e^{-\beta E_M}}{\sum_{i=1}^{M_{\text{tot}}} e^{-\beta E_i}}$$

with $M_{\text{tot}}$ the total number of eigenstates. $eG_S$ is the quantum conductance with $eG_S = T_S^2 \rho_T(\epsilon_F) \rho_S(\epsilon_F)$. [2] Because the system is at equilibrium, it can be simplified to $eG_S = \frac{2e^2}{h}$ and $G_S = \frac{2e}{h}$.

The current calculated with formula 2.3 is a double sum over all possible combinations of the eigenstates of the system. The probability that the system occupies eigenstate $|M\rangle$ is represented by $P_M$. The STM tip which is located above atom $j$ couples with atom $j$ resulting in a factor $|\langle M| S_{\alpha}^{(j)} |M'\rangle|^2$ which describes the exchange spin interaction between the spin of the tunneling electron and the atomic spin. [1] If $|\langle M| S_{\alpha}^{(j)} |M'\rangle|^2 = 0$ the exchange interaction cannot change the eigenstate of the system from eigenstate $|M\rangle$ to eigenstate $|M'\rangle$. Also $F_{M,M',s}(V)$ evaluates if there is enough energy for the transition from state $|M\rangle$ to state $|M'\rangle$, besides $F_{M,M',s}(V)$ is a temperature dependent function. And $G_S$ is the conductance, it is useful to change Joule to Ampère.

Summarizing: for each eigenstate $|M\rangle$, formula 2.3 calculates the probability for the system occupying eigenstate $|M\rangle$, then the formula evaluates which eigenstates $|M'\rangle$ are accessible from eigenstate $|M\rangle$ when the exchanging spin interaction occurs, afterwards the formula calculates if there is enough energy to change the eigenstate of the system from eigenstate $|M\rangle$ to eigenstate $|M'\rangle$, finally the dimension is changed to Ampère with $G_S$. 
3

Method

When an IETS experiment is performed on the system of \( n \) particles, as described in section 2.5, a voltage is applied and the current is measured. The current can be predicted with formula 2.3, but to use this formula the eigenstates and eigenenergies of the system have to be known. How to find the terms for formula 2.3 will be explained now.

3.1. Analyse a three particle system as an example

The formulas from the theory are for a general \( n \) particle systems. For large \( n \) calculations get complicated quickly, so start with a simple but still interesting system of three particles with spin 2. Moreover, a system of two particles with spin 2 has been worked out in appendix A.3.

3.2. Analyse the Hamiltonian and make approximations

The representation of a system consisting of three atoms is \(|m_1 \ m_2 \ m_3\rangle\). To analyse the Hamiltonian use formula 2.2 with \( n = 3 \) and let it act on \(|m_1 \ m_2 \ m_3\rangle\), which is an element of a three particle system with spin 2 for each particle. Some assumptions are going to be made in formula 2.2, the external magnetic field will be assumed to point in the \( \hat{z} \)-direction, so \( B = B \hat{z} \). And zero transverse anisotropy \( E = 0 \) will be assumed to make it easier to find the eigenstates of the Hamiltonian. In section 3.5 the results of the Hamiltonian with \( E = 0 \) will be used to analyse the Hamiltonian with \( E \neq 0 \).

After some work, which can be found in appendix A.1, the result is:

\[
\mathcal{H}|m_1 \ m_2 \ m_3\rangle = \left(\left((m_1)^2 + (m_2)^2 + (m_3)^2\right)D - (m_1 + m_2 + m_3)E_z + (m_1 m_2 + m_2 m_3) J\right)|m_1 \ m_2 \ m_3\rangle
\]

\[
+ \frac{J}{2} \sqrt{6 - m_1 (m_1 + 1) \sqrt{6 - m_2 (m_2 - 1) \sqrt{6 - m_3 (m_3 - 1)}}} |m_1 + 1 \ m_2 - 1 \ m_3\rangle
\]

\[
+ \frac{J}{2} \sqrt{6 - m_1 (m_1 - 1) \sqrt{6 - m_2 (m_2 + 1) \sqrt{6 - m_3 (m_3 + 1)}}} |m_1 - 1 \ m_2 + 1 \ m_3\rangle
\]

\[
+ \frac{J}{2} \sqrt{6 - m_2 (m_2 + 1) \sqrt{6 - m_3 (m_3 - 1) \sqrt{6 - m_1 (m_1 + 1)}}} |m_2 + 1 \ m_3 - 1 \ m_1\rangle
\]

\[
+ \frac{J}{2} \sqrt{6 - m_2 (m_2 - 1) \sqrt{6 - m_3 (m_3 + 1) \sqrt{6 - m_1 (m_1 - 1)}}} |m_2 - 1 \ m_3 + 1 \ m_1\rangle
\]

(3.1)

with \( E_z = g^* \mu_B B \).

3.3. Find the eigenstates of the three particle system

The Hamiltonian acting on an element \(|m_1 \ m_2 \ m_3\rangle\) of the three particle system returns a maximum of five different \( z \)-basis elements of the system, so multiple elements are needed to form an eigenstate. The total spin of each particle is 2, thus the maximum value of \( m_1 = 2 \) and the minimum value of \( m_1 = -2 \), because the \( z \)-component of the spin of a particle cannot be greater than the total spin. The spin operators account for this and when the \( m_1 \) value is raised to 3 or lowered to -3 the square root in front of that element prevents this, so the math is consistent with the physics.
When the Hamiltonian acts on an element \(|m_1 m_2 m_3\rangle\) of the three particle system, the same element is returned and a maximum of four other elements are returned if their z-components of the spin are smaller or equal to the total spin. All five elements do have something in common, sum the z-components of each element and for each element it is equals to \(m_1 + m_2 + m_3\). Thus collect all the elements with the same \(m_1 + m_2 + m_3\) and when the Hamiltonian acts on this group, the same group is the result. Because each atom has spin 2 the sum of \(m_1 + m_2 + m_3\) ranges from -6 to 6, thus there are 13 different groups with the same \(m_1 + m_2 + m_3\).

### 3.4. Eigenenergies of the three particle system

\(|M\rangle\) is an eigenstate of the Hamiltonian if \(\mathcal{H}|M\rangle = E_M|M\rangle\) and \(\langle M|M\rangle = 1\) hold, with \(E_M\) the eigenenergy of eigenstate \(|M\rangle\). In the previous section 13 groups were found, time to investigate them. Start with group 1, the group with \(m_1 + m_2 + m_3 = -6\). Because the spin of each atom is 2, there is only one element in group 1, which is \(|-2-2-2\rangle\).

\(|-2-2-2\rangle\) is an eigenstate, because \((-2-2-2)(-2-2-2) = 1\) and from formula 3.1 follows:

\[
\mathcal{H}|-2-2-2\rangle = \left((4+4+4)D - (2-2-2)E_z + (4+4)J\right)|-2-2-2\rangle
\]

\[
+ \left(\frac{1}{2} (2)(0)|-1-3-2\rangle + \frac{1}{2} (0)(2)|-3-1-2\rangle
\]

\[
+ \frac{1}{2} (2)(0)|-2-1-3\rangle + \frac{1}{2} (0)(2)|-2-3-1\rangle
\]

\[
= (12D + 6E_z + 8J)|-2-2-2\rangle.
\]

Thus \(|M_1\rangle = |−2−2−2\rangle\) is an eigenstate of the Hamiltonian with eigenenergy \(E_{M_1} = 12D + 6E_z + 8J\).

Group 2 has elements with \(m_1 + m_2 + m_3 = -5\). Thus \(|-1-2-2\rangle\), \(|-2-1-2\rangle\) and \(|-2-2-1\rangle\) are the elements of group 2. All three elements are needed to form an eigenstate, so the eigenstate will be a superposition. Define a superposition \(|G_2\rangle\) with all elements of group 2,

\[
|G_2\rangle = a|-1-2-2\rangle + b|-2-1-2\rangle + c|-2-2-1\rangle
\]

and find the coefficients \(a, b\) and \(c\) wherefore the superposition is an eigenstate of the Hamiltonian. Check the properties of an eigenstate and get the requirements for the coefficients. Using the inner product properties discussed in section 2.4 the first requirement for the coefficients follows:

\[
\langle G_2|G_2\rangle = 1 \Leftrightarrow |a|^2 + |b|^2 + |c|^2 = 1
\]

For the other requirements let the Hamiltonian act on \(|G_2\rangle\):

\[
\mathcal{H}|G_2\rangle = \mathcal{H}\left(a|-1-2-2\rangle + b|-2-1-2\rangle + c|-2-2-1\rangle\right)
\]

\[
= \left(1 + 4 + 4\right)D - \left(-1-2-2\right)E_z + \left(2+4\right)J|a|-1-2-2\rangle + 0 + \frac{1}{2} \left(2\right) \left(2\right) a|2-1-2\rangle + 0 + 0
\]

\[
+ \left(1 + 4 + 4\right)D - \left(-2-1-2\right)E_z + \left(2+2\right)J|b|-2-1-2\rangle + 0 + 0 + \frac{1}{2} \left(2\right) \left(2\right) b|2-2-1\rangle
\]

\[
+ \left(1 + 4 + 1\right)D - \left(-2-2-1\right)E_z + \left(4+2\right)J|c|-2-2-1\rangle + 0 + 0 + \frac{1}{2} \left(2\right) \left(2\right) c|2-1-2\rangle + 0
\]

\[
= \left(9D + 5E_z + 6J\right)a + 2Jb\left|-1-2-2\rangle\right) + \left(9D + 5E_z + 4J\right)b + 2Ja + 2Jc\left|-2-1-2\rangle\right)
\]

\[
+ \left(9D + 5E_z + 6J\right)c + 2Jb\left|-2-2-1\rangle\right) = E_{G_2}|G_2\rangle = E_{G_2}a|-1-2-2\rangle + E_{G_2}b|-2-1-2\rangle + E_{G_2}c|-2-2-1\rangle.
\]
Thus we have found
\[
\begin{align*}
(9D + 5E_Z + 6J) |a + 2Jb| - 2 - 2) &= E_{G_2} |a - 1 - 2 - 2) \\
(9D + 5E_Z + 4J) |b + 2Ja + 2Jc| - 2 - 1 - 2) &= E_{G_3} |b - 2 - 1 - 2) \\
(9D + 5E_Z + 6J) |c + 2Jb| - 2 - 2 - 1) &= E_{G_4} |c - 2 - 2 - 1).
\end{align*}
\]

These equations can be transformed into one eigenvalue equation:
\[
(9D + 5E_Z + 4J) \mathbf{I}_{3 \times 3} \begin{bmatrix} a \\ b \\ c \end{bmatrix} + 2J \begin{bmatrix} 1 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 1 \end{bmatrix} \begin{bmatrix} a \\ b \\ c \end{bmatrix} = E_{G_2} \begin{bmatrix} a \\ b \\ c \end{bmatrix}.
\]

Following the matrix properties discussed in section 2.4, the eigenvalues are \( E_{G_2} = 9D + 5E_Z + 4J + 2J \lambda_{A_2} \) with \( \lambda_{A_2} \) the eigenvalues of matrix \( A_2 \). With \( A_2 = \begin{bmatrix} 1 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 1 \end{bmatrix} \). The next step is finding the eigenvalues and eigenvectors of \( A_2 \). The eigenvalues and eigenvectors of \( A_2 \) can be found by hand, but during the search for the other eigenstates, the eigenstates and eigenvalues of more complicated matrices have to be determined. Matlab is used to find the eigenvalues and eigenvectors, the code can be found in appendix A.7. From the matrix properties discussed in section 2.4, it is known that there will be three real eigenvalues and an orthogonal set of eigenvectors. By scaling the eigenvectors to an orthonormal set the first requirement for the coefficients \( a, b \) and \( c \) is satisfied. The result from using the matlab code is: \( \lambda_{A_2} = -1 \) with \( \begin{bmatrix} a \\ b \\ c \end{bmatrix} = \frac{1}{\sqrt{2}} \begin{bmatrix} -1 \\ 0 \\ 1 \end{bmatrix} \) and \( \lambda_{A_2} = 2 \) with \( \begin{bmatrix} a \\ b \\ c \end{bmatrix} = \frac{1}{\sqrt{3}} \begin{bmatrix} 1 \\ 1 \\ 1 \end{bmatrix} \). Thus three eigenstates have been found:
\[
\begin{align*}
|M_2\rangle &= \frac{1}{\sqrt{6}} [-1 - 2 - 2) - \frac{2}{\sqrt{6}} [-2 - 1 - 2) + \frac{1}{\sqrt{6}} [-2 - 2 - 1) \\
|M_3\rangle &= \frac{1}{\sqrt{2}} [-1 - 2 - 2) + \frac{1}{\sqrt{2}} [-2 - 2 - 1) \\
|M_4\rangle &= \frac{1}{\sqrt{3}} [-1 - 2 - 2) + \frac{1}{\sqrt{3}} [-2 - 1 - 2) + \frac{1}{\sqrt{3}} [-2 - 2 - 1).
\end{align*}
\]

with eigenenergies \( E_{M_2} = 9D + 5E_Z + 2J, E_{M_3} = 9D + 5E_Z + 6J \) and \( E_{M_4} = 9D + 5E_Z + 8J \). How to find all the other eigenstates and eigenvalues has been worked out in appendix A.2.

There are 125 eigenstates in total, because there are 125 z-basis elements \( |m_1 m_2 m_3\rangle \) and every z-basis element belongs to a group with the same \( m_1 + m_2 + m_3 \). To find the eigenenergies these groups are used to construct symmetric matrices as described above and a \( n \) by \( n \) has \( n \) eigenvalues with \( n \) orthogonal eigenstates. Thus there are 125 eigenstates in total for the 3 particle system. For the same reason a system containing \( n \) particles with spin 2 has \( 5^n \) z-basis elements, thus the \( n \) particle system has \( 5^n \) eigenstates and eigenenergies, thus \( M_{tot} = 5^n \).

### 3.5. Hamiltonian including transverse magnetic anisotropy

When the Hamiltonian was analysed and the eigenstates and eigenenergies were found in the sections above there was an assumption about the transverse magnetic anisotropy \( E \). The assumption was \( E = 0 \) to make it easier to find the eigenstates of the system. But in reality \( E \neq 0 \), \( E \) is of the same order as \( D \). The previous results with the assumption \( E = 0 \) can be used to find the eigenstates and eigenenergies when \( E \neq 0 \).

Start with exploring the additional term in the Hamiltonian when \( E \neq 0 \). Then for a \( n \) particle system
\[
\sum_{i=1}^{n} E \left( \left( \mathbf{S}_x^{(i)} \right)^2 - \left( \mathbf{S}_y^{(i)} \right)^2 \right) \tag{3.2}
\]
has to be investigated. After some work which can be found in appendix A.4, the result is:
\[
\sum_{i=1}^{n} E \left( \mathbf{S}_+^{(i)} \mathbf{S}_-^{(i)} + \mathbf{S}_-^{(i)} \mathbf{S}_+^{(i)} \right) \tag{3.3}
\]
Now we let this operator act on an element of the three particle system \( |m_1 \, m_2 \, m_3 \rangle \). After some work which can be found in appendix A.4, this can be expressed as:

\[
\sum_{i=1}^{3}\frac{E}{2}\left(S_z^{+}(i)S_z^{-}(i) + S_z^{-}(i)S_z^{+}(i)\right)|m_1 \, m_2 \, m_3 \rangle = \frac{E}{2}\sqrt{6 - (m_1 + 1)(m_1 + 2)}\sqrt{6 - m_1(m_1 + 1)}(m_1 + 2)\, m_2 \, m_3 \right) \tag{3.4}
\]

Thus the transverse magnetic anisotropy term of the Hamiltonian acting on \( |m_1 \, m_2 \, m_3 \rangle \) contributes to six other z-basis elements of the three particle system. Look at the z-components of the spin and sum them, the sums are \( m_1 + m_2 + m_3 \). Thus the transverse magnetic anisotropy term of the Hamiltonian acting on \( |m_1 \, m_2 \, m_3 \rangle \) group \( m_1 + m_2 + m_3 \) contributes to group \( m_1 + m_2 + m_3 + 2 \) and \( m_1 + m_2 + m_3 - 2 \). Thus the 13 groups of the Hamiltonian with the assumption \( E = 0 \) are merged into two groups when \( E \neq 0 \), the odd groups are merged into one group containing 63 elements and the even groups are merged into the other group containing 62 elements. These two new groups are eigenstates of the Hamiltonian if they are normalized.

Normalizing these big groups would be a lot of work if it had to be done by hand. A matlab code was made, see appendix A.9, to find the eigenstates and eigenenergies. As described above, which elements are in the eigenstates is known, but not the coefficients in front of the elements. To find the eigenstates and eigenenergies a similar process has been used as described in section 3.4. Let the full Hamiltonian act on an element of the three particle system, then:

\[
\mathcal{H}|m_1 \, m_2 \, m_3 \rangle = \left(\left((m_1)^2 + (m_2)^2 + (m_3)^2\right)D - (m_1 + m_2 + m_3)E_z + (m_1m_2 + m_2m_3)F\right)|m_1 \, m_2 \, m_3 \rangle
\]

because this is equal to adding formula 3.1 and formula 3.4 together. The equations that follow from the Hamiltonian acting on an element of the three particle system can be written as a column vector with length 125, with each place of the vector representing an element of the three particle system. For example the first place in the vector represents \( |2 \, -2 \, -2 \rangle \) and the third place of the vector represents \( |0 \, -2 \, -2 \rangle \) and every other place represents an other element of the three particle system.
Now the process of finding the eigenstates and eigenenergies will be explained. Let the full Hamiltonian act on every element of the three particle system and transform all equations to column vectors, resulting in 125 column vectors, because there are 125 elements in the three particle system. The 125 column vectors will be added into a 125 by 125 matrix, but the order of the column vectors is important. In the example $| -2 \ -2 \ -2 \rangle$ is represented by the first place of the column vector and $| 0 \ -2 \ -2 \rangle$ is represented by the third place of the column vector, then the first column of the matrix has to be the vector of the Hamiltonian acting on $| -2 \ -2 \ -2 \rangle$ and the third column of the matrix has to be the vector of the Hamiltonian acting on $| 0 \ -2 \ -2 \rangle$. If the vectors are ordered in this way the matrix will be symmetric just like the matrices in the process described in section 3.4 and appendix A.2. The equations following from the Hamiltonian have been transformed into a symmetric matrix and the eigenvalues of this matrix are the eigenenergies and the eigenvectors of this matrix are the eigenstates. There are 125 eigenstates and eigenenergies, because of the 125 by 125 symmetric matrix.

The eigenstates contain either the 63 elements of the odd groups or the 62 elements of the even groups, to see the specific coefficients of each element in every eigenstate see appendix A.9 on how to access the eigenstates in the code. And the eigenenergies are shown in figure 3.1.

![Eigenenergy spectrum of a 3 particle system](image)

Figure 3.1: Eigenenergy spectrum of the three particle system.

This process can be used for a chain consisting of $n$ particles. The $n$ particle system has $5^n$ z-basis elements. Let the Hamiltonian act on every element of the $n$ particle system and transform all equations to column vectors with length $5^n$. Order the vectors the right way as described above into a $5^n$ by $5^n$ matrix. The symmetric matrix has $5^n$ eigenvalues and eigenvectors, thus the $n$ particle system has $5^n$ eigenenergies and $5^n$ eigenstates. However, for large $n$ a lot of computing power and storage is needed, because $5^n$ eigenstates and eigenenergies have to be calculated and stored and these numbers increase exponentially for large $n$.

### 3.6. The inelastic tunneling current

The eigenstates of the Hamiltonian are made of elements in the z-spin basis, thus write out the sum and work out the $\hat{S}^+_x(j)$ and $\hat{S}^+_y(j)$ operators as combinations of $\hat{S}^+_x(j)$ and $\hat{S}^-_y(j)$ in formula 2.3 from section 2.5. After some work which can be found in appendix A.5, the inelastic current in an IETS experiment with the STM tip located above atom $j$ in the chain is given by:

$$I(V) = G_S \sum_{M,M'} P_M \frac{1}{2} \left( \langle M | \hat{S}^+_x(j) | M' \rangle \right)^2 F_{M,M',\alpha}(V)$$

$$+ G_S \sum_{M,M'} P_M \frac{1}{2} \left( \langle M | \hat{S}^-_y(j) | M' \rangle \right)^2 F_{M,M',\alpha}(V)$$

$$+ G_S \sum_{M,M'} P_M \left( \langle M | \hat{S}^+_z(j) | M' \rangle \right)^2 F_{M,M',\alpha}(V)$$

(3.6)
with

$$F_{M,M',\Delta}(V) = \frac{eV - s\Delta_{M',M}}{1 - e^{-s\beta(eV - s\Delta_{M',M})}} + \frac{eV + s\Delta_{M',M}}{1 - e^{s\beta(eV + s\Delta_{M',M})}}.$$

For the definition of all symbols take a look at section 2.5 or the list of symbols. To obtain this formula we used that the coefficients of the eigenstates are real. If the coefficients are not real use formula A.1, which can be found in appendix A.5.

How to find the eigenstates and eigenenergies is described in section 3.5. Set a temperature and the location of the STM tip above an atom of the chain. Now everything is known for the formula 3.6. A matlab script has been written to calculate the current, the script can be found in appendix A.8, A.9 and A.10. The temperature and the STM tip location can be varied, as well as the longitudinal magnetocrystalline anisotropy $D$, the transverse magnetic anisotropy $E$ and the the exchange energy between neighbouring atoms $J$, because they vary depending on the type of atoms in the chain. With the calculated current $I(V)$ graphs and $\frac{dI}{dV}$ graphs can be made. Some graphs are listed in the results, see chapter 4.
Results and Discussion

The spins of the atoms cannot be measured directly, but the spin dynamics of the system can be examined with an inelastic electron tunneling spectroscopy (IETS) experiment. Formula 2.3 from the theory predicts the inelastic tunneling current $I(V)$ in an IETS experiment. The different terms in the current formula have been explained in the theory, see section 2.5.

In the next sections realistic experimental conditions for an IETS experiment will be used. The plots are made with matlab, see appendix A.8, A.9 and A.10 for the code. In this section $D = -1.29 \text{ meV}$, $E = 0.31 \text{ meV}$ and $J = -0.73 \text{ meV}$ are used, unless specified otherwise. These values are from an IETS experiment performed on a chain of magnetic iron atoms with spin 2. The values were obtained with no external magnetic field $\mathbf{B} = \mathbf{0}$. [4] In this chapter the STM tip will be positioned above the first atom of the chain, a temperature of 1 K and a magnetic field of 300 mT pointing in the z-direction will be used, unless specified otherwise. The magnetic field of 300 mT results in $E_z = 0.035 \text{ meV}$. The values of $D$, $E$ and $J$ can be used with an applied magnetic field of 300 mT. [4]

4.1. Inelastic tunneling current of a three particle system

In Figure 4.1 the current is plotted for a three particle system. For voltages lower than $-0.1 \text{ mV}$ there is almost no current, because there is not enough energy in the system for a state transition. When there is enough energy for a state transition to occur, the inelastic tunneling current will increase. For voltages of $-0.1 \text{ mV}$ or higher the current increases, because the applied voltage increased the energy of the system enough for a state transition to occur.

![IV plot of a 3 particle system](image)

Figure 4.1: The tunneling current versus the applied voltage for a three particle system.
4.2. Differential conductance of a three particle system

In Figure 4.1 an increase in current was observed for an increasing voltage, but how fast does the current increase? In Figure 4.2 the first derivative of the current is displayed, it gives information about how fast the current changes. The first derivative of the current is the differential conductance. The differential conductance is almost zero for voltages below \(-0.3 \text{ mV}\). The differential conductance increases fast for higher voltages, until \(0.3 \text{ mV}\) is reached and from there the differential conductance increases slowly. This means that around \(-0.3 \text{ mV}\) there is enough energy in the system for a state transition. When the voltage is increased from \(-0.3 \text{ mV}\) there will be enough energy for more possible state transitions. And more possible state transitions mean more current, thus an increase in the differential conductance. For voltages of \(0.3 \text{ mV}\) and higher the differential conductance increases slowly. But why does the differential conductance increase fast and after \(0.3 \text{ mV}\) it increases slowly? As described above an increase in voltage between \(-0.3 \text{ mV}\) and \(0.3 \text{ mV}\) causes more possible state transitions. But there is a limited amount of eigenstates and thus a maximum amount of possible state transitions and for each state transition another amount of energy is needed. Thus the differential conductance increases slowly for voltages above \(0.3 \text{ mV}\), because for these voltages there is not enough energy for another state transition to occur. The differential conductance still increases, because increasing the voltage changes the exponents in the denominator of \(F_{M,M',s}(V)\) resulting in a small increase in the differential conductance. The maximum energy required for a state transition is the energy needed to transition the ground state into the state with the highest eigenenergy, this is around \(27.6 \text{ meV}\). Thus for voltages above \(27.6 \text{ mV}\) practically all state transitions can occur.

Summarizing, the current increases for higher voltages. How fast the current increases can be seen in the differential conductance. The differential conductance increases slowly due to the influence of \(F_{M,M',s}(V)\). And the differential conductance increases fast when more state transitions can occur.

![differential conductance plot](image)

**Figure 4.2**: The first derivative of the tunneling current versus the applied voltage for a three particle system.

4.3. Influence of the temperature on the current

The current is influenced by the temperature. As described in the theory \(P_M\) and \(F_{M,M',s}(V)\) are temperature dependent. \(P_M\) represents the probability that the system occupies eigenstate \(M\). Investigate the low temperature limit \(T \to 0 \text{ K}\), then the system has minimum energy, thus the system is in its ground state. Thus only state transitions from the ground state can be made if \(T \to 0 \text{ K}\). Is this low temperature limit a good approximation for low temperatures? In Figure 4.3 both the current at \(1 \text{ K}\) and the low temperature limit for \(P_M\) are displayed. The two overlap, so Figure 4.4 is zoomed in on a part of Figure 4.3. The difference is about \(5 \times 10^{-13} \text{ A}\), thus it is a good approximation for low temperatures. Notice that the low temperature limit has a higher current than the three particle system. In the three particle system the probability the system is in the ground state is about 99.2 % and the probability the system is in another state than the ground state is 0.8 %. Thus the current of the three particle system consists of contributions primarily from the ground state, but other states contribute too. Apparently state transitions from states other than the ground state contribute so little to the current, that the low temperature limit has a higher current than the three particle system.
4.3. Influence of the temperature on the current

Investigate higher temperatures, for example room temperature $T = 293 \, K$. In Figure 4.5 the current can be seen for a three particle system at $T = 293 \, K$. The current is almost zero for voltages below $-50 \, meV$ and increases for voltages higher than $-50 \, meV$. Compare this to Figure 4.1 where the system is at $T = 1 \, K$. The current increase in Figure 4.1 starts at $-0.3 \, meV$, this difference is caused by the temperature. Because at $T = 1 \, K$ the probability that the system is occupying the ground state is 99.2 % and the probability the system is occupying another state is only 0.8 %. At $T = 293 \, K$ the probability that the system is occupying the ground state is 1.36 % and the probability for every other individual state is lower, ranging from 1.34 % to 0.46 %, because the ground state has the lowest energy. Thus at $T = 293 \, K$ the system could be occupying any of the 125 eigenstates with a reasonable probability: this means that there are many potential state transitions. Because the system could be occupying any of its eigenstates with reasonable chance there are state transitions possible which require less energy than state transitions from the ground state, thus the current increases for lower voltages than the system at $T = 1 \, K$. This can be seen in Figure 4.6, where the current of the system at $T = 293 \, K$ increases at a lower voltage than the system at $T = 1 \, K$. Another difference between high and low temperatures that can be seen in Figure 4.6 is: a system with a higher temperature has a higher current.

Summarizing, for low temperatures the system has a very high probability to occupy the ground state and thus a good approximation is the low temperature limit where the system is in the ground state. For
high temperatures the system has a reasonable probability to occupy any of the 125 eigenstates, thus there are more potential state transitions. Resulting in a start of the current increase at lower voltages than a low temperature system. Overall a high temperature system has a higher current than a low temperature system.

![Image](image1.png)

**Figure 4.5:** The tunneling current versus the applied voltage for a three particle system at room temperature $T = 293 \, K$.

![Image](image2.png)

**Figure 4.6:** The tunneling current versus the applied voltage for a three particle system at room temperature $T = 293 \, K$ and at $T = 1 \, K$. The red line is the current of the system at $T = 293 \, K$ and the blue line is the current of the system at $T = 1 \, K$. The red line is above the blue line, thus the current of the system at $T = 293 \, K$ is higher than the current of the system at $T = 1 \, K$.

### 4.4. Influence of the location of the STM tip on the current

The current depends on the location of the STM tip, because the location of the STM tip determines with which atom there will be spin exchange interaction. In Figure 4.7 the current of the system is displayed with the tip above atom 1 in blue and the tip above atom 2 in red. In Figure 4.7 a small difference in the current can be seen due to a difference in STM tip location. There is a difference in current because the tip interacts with the atom and the spin interaction can change the state of the system if there is enough energy resulting in a current. Other state transition can occur depending on the location of the tip of the STM, but since the system is at $T = 1 \, K$ the probability the system is occupying the ground state is 99.2 %, thus there are not many new state transitions possible when the STM tip changes its position, this results in the small current difference. Increase the temperature, for example to room temperature $T = 293 \, K$. The system has reasonable probability to occupy any of the 125 states. When the STM tip interacts with an atom there are many state
4.4. Influence of the location of the STM tip on the current

transitions possible. In Figure 4.8 the two currents of the system at $T = 293 \, K$ are plotted. The current with the tip above atom 1 is blue and the current with the tip above atom 2 is red. The difference in current is now easy to see and in this case the current is higher with the tip above atom 1. The difference depends on which states were made accessible by the tip interaction, because different state transitions have a different effect on the current.

Figure 4.7: The tunneling current versus the applied voltage for a three particle system, with the STM tip above atom 1 and above atom 2. The blue line is the current with the tip above atom 1 and the red line is the current with the tip above atom 2. The difference is small, but in this instance the red line is above the blue line, with the current about 1.1 $nA$ higher at high voltages when the tip is above atom 2.

Figure 4.8: The tunneling current versus the applied voltage for a three particle system at room temperature $T = 293 \, K$, with the STM tip above atom 1 and above atom 2. The blue line is the current with the tip above atom 1 and the red line is the current with the tip above atom 2. The blue line is above the red line, thus the current of the system at $T = 293 \, K$ is higher when the tip is located above atom 1.

The tip can be positioned above the third atom in the chain too. When the tip is above the third atom, the tunneling current is the same as if the tip was located above the first atom. This makes sense because the chain consists of three identical magnetic iron atoms and the chain is symmetrical. It can be explained with the eigenstates too, the symmetric elements of the eigenstates have the same coefficient. In appendix A.9 is explained how to access the eigenstates with the matlab code. In general within all eigenstates $|m_a m_b m_c\rangle$ and $|m_c m_b m_a\rangle$ are a symmetric pair, this is caused by the Hamiltonian, see formula 3.5 in section 3.5. Look at formula 3.5, when $m_1$ and $m_3$ are switched, thus $\mathcal{H}|m_3 m_2 m_1\rangle$, then the numbers in front of the symmetric elements are the same, resulting in the same symmetric matrix and thus the same eigenstates follow from this.
matrix. For some examples see appendix A.2, for example in the superposition of group 3: $a|0 − 2 − 2\rangle$ and $f|−2 \quad 2 \quad 0\rangle$ are symmetric and the other symmetric pair is $b|−1 \quad 1 \quad 1 − 2\rangle$ and $e|−2 \quad 1 \quad 1 \quad 1\rangle$.

Summarizing, the location of the STM tip influences which state transitions are possible and different state transitions have a different effect on the current. The influence of the location of the STM tip is the same for symmetric atoms, for example in the three particle system the current is the same when the STM tip is above atom 1 or atom 3. This also holds for chains of even length, for example for a four particle system the symmetric pairs of atoms are 1 with 4 and 2 with 3.

4.5. Influence of the number of particles in the chain

To investigate the influence of the number of atoms in the chain, a two, three and four particle system will be examined. In Figure 4.9 the tunneling currents of these three systems are plotted. The currents overlap a lot, thus all the systems have roughly the same tunneling current. But why are the currents almost the same for systems with different amounts of particles? The eigenstates of a system are important to determine the current. As discussed in chapter 3, the amount of eigenstates depends on the number of atoms in the chain. For a chain containing $n$ magnetic atoms with spin 2 the amount of eigenstates equals $5^n$, see section 3.5. Thus the two particle system has 25 eigenstates, the three particle system has 125 eigenstates and the four particle system has 625 eigenstates. When the STM tip interacts with the chain a state transition can occur. A system with many particles has many possible state transitions and a system with fewer particles allows fewer possible state transitions. However, a system with many particles, thus many eigenstates has a lower probability to occupy a specific eigenstate than a system with fewer particles, because there are fewer eigenstates. The probability to occupy an eigenstate and the amount of possible state transitions are both important for the inelastic tunneling current, which can be seen in formula 2.3.

![Figure 4.9: The tunneling current versus the applied voltage for a system with two, three and four particles. The current of the two particle system is red, the current of the three particle system is blue and the current of the four particle system is yellow. The currents of the three systems overlap and the difference between them is a few nanoampère. For voltages between 5 meV and 15 meV the currents differ more.](image)

We now investigate the differential conductance to see the influence of the probabilities and state transitions. In Figure 4.10 the differential conductances of the three systems are plotted. The differential conductances exhibit a few jumps, the jumps are caused by state transitions as described in section 4.2. The differential conductances overlap a bit, but differ for voltages between 5 meV and 15 meV. Because this interval is interesting, a close up on this interval has been made and can be seen in Figure 4.11. Look at Figure 4.11, notice that the two particle system makes the biggest jumps and the four particle system makes the smallest jumps, but the four particle system makes the most jumps. This is caused by the probability of the eigenstates and the possible state transitions. The two particle system has the least amount of eigenstates, thus the differential conductance makes the least amount of jumps. However, because the two particle system has the least amount of eigenstates, the probability to occupy a specific eigenstate of the two particle system is higher than the probability to occupy a specific eigenstate of the four particle system, thus the two...
4.5. Influence of the number of particles in the chain

A particle system makes bigger jumps than the four particle system. Because the differential conductances of the currents overlap for a large range of voltages, the currents overlap often too. But there is a bigger difference between the currents when one system has a jump in its differential conductance and the other systems have no jump in their differential conductances. These differences occur, because the two particle system makes a few big jumps in its differential conductance, the three particle system makes some jumps in its differential conductance and the four particle system makes many small jumps in its differential conductance.

Summarizing, the current is determined by the number of possible state transitions and the probability to occupy an eigenstate. When the number of atoms in a chain increases, the number of eigenstates increases too. Thus there is an increase in the total number of possible state transitions, because there are more states to transition to. But the probability to occupy a specific eigenstate decreases, because the number of eigenstates was increased. Thus the differential conductance makes more jumps, because there are more state transitions possible. But the jumps are smaller, because the probability to occupy a specific state is smaller. When the number of atoms decreases, the differential conductance makes fewer jumps, because there are fewer state transitions possible. But the jumps are bigger, because the probability to occupy a specific state is bigger.

Figure 4.10: The first derivative of the tunneling current versus the applied voltage for a system with two, three and four particles. The first derivative of the tunneling current is the differential conductance. The differential conductance of the two particle system is red, the differential conductance of the three particle system is blue and the differential conductance of the four particle system is yellow. For a large range of voltages the differential conductances overlap, but there is a big difference between the differential conductances for voltages between 5 mV and 15 mV, for a close up see Figure 4.11. In this interval the three systems exhibit differences in the height and amount of jumps. The two particle system exhibits two large jumps, the three particle system exhibits two jumps and the four particle system exhibits three small jumps.
4.6. Influence of the anisotropies, the exchange energy and the applied magnetic field

The single-spin longitudinal magnetocrystalline anisotropy represented by $D$, the transverse magnetic anisotropy represented by $E$ and the exchange energy between neighbouring atoms represented by $J$ are parameters of the Hamiltonian. The applied magnetic field in the $z$-direction represented by $B = B \hat{z}$ influences the Hamiltonian too, because $E_z = g^* \mu_B B$, with $g^*$ the Landé g-factor and $\mu_B$ the Bohr magneton, see formula 2.2. The Hamiltonian is important, because the eigenstates and eigenenergies follow from the Hamiltonian. The anisotropies $D$ and $E$ are determined by the type of the atoms in the chain, because the chain can be built with magnetic atoms and every type of magnetic atom has different values for $D$ and $E$. The exchange energy between neighbouring atoms $J$ depends on the distance between the atoms. During this research iron atoms were used as magnetic atoms, but other magnetic atoms can be used as well. However, during this research iron atoms with spin 2 were used, if the other magnetic atoms have different spin, then the method explained in section 3.5 has to be modified.

Different values of $D$, $E$, $J$ and $E_z$ lead to different eigenenergies and eigenstates. The eigenenergies and eigenstates are important to calculate the tunneling current, see formula 2.3. The eigenenergies influence the probability to occupy an eigenstate, because $P_M = \frac{e^{-\beta E_M}}{\sum_{i=1}^{M_{tot}} e^{-\beta E_i}}$ with $E_M$ the eigenenergy of eigenstate $|M\rangle$ and $M_{tot} = 5^9$ the total number of eigenstates. And the eigenenergies influence $F_{M,M',s}(V)$, see formula 2.3. The eigenstates influence the possible state transitions and the eigenstates influence $F_{M,M',s}(V)$ too. If different magnetic atoms with spin 2 are used, the different eigenstates will still be superpositions of the $z$-basis elements as described in the method, but the coefficients in front of the $z$-basis elements change when different values of $D$, $E$, $J$ and $E_z$ are used.

Summarizing, if new values for $D$, $E$, $J$ and $E_z$ are used, then the new eigenstates and eigenenergies have to be calculated for these new parameters. When the new eigenstates and eigenenergies are known the inelastic tunneling current formula can be used to calculate the current.

4.7. Spin waves

When the spin of an atom makes a wavelike motion with time it is called a spin wave. For an example of a spin wave in the $z$-direction see Figure 4.12, the $z$-component of the spin changes with time in a wavelike motion. Spin waves are not always periodic like the example in Figure 4.12. A spin wave can occur also in a one dimensional chain. [4] Then the spin of one or more atoms of the chain make a wavelike motion with time.
As described in section 2.3, an unperturbed system is in an eigenstate of the Hamiltonian. The eigenstates of the Hamiltonian have been found with a method described in section 3.5. The eigenstates are superpositions of elements expressed in the z-basis. For example an eigenstate of the two particle system is:

\[
\begin{align*}
&x_1|\!-\!2\!-\!2\rangle + x_{13}|\!-\!2\!-\!0\rangle + x_{12}|\!-\!2\!-\!2\rangle \\
&+ x_2|\!-\!1\!-\!1\rangle + x_{10}|\!1\!-\!1\rangle \\
&+ x_3|0\!-\!2\rangle + x_9|0\!0\rangle + x_{11}|0\!2\rangle \\
&+ x_4|1\!-\!1\rangle + x_8|1\!1\rangle \\
&+ x_5|2\!-\!2\rangle + x_6|2\!0\rangle + x_7|2\!2\rangle.
\end{align*}
\] (4.1)

To see the exact numbers of the coefficients take a look at appendix A.8 how to run the code and access the eigenstates. The eigenstate in this example is a superposition of 13 elements in the z-basis. A superposition means the system is multiple things at once, thus in this example the spins of the system are 13 elements in the z-basis at once. Thus the superposition of these 13 elements can be seen as a ‘spin wave’. The reason ‘spin wave’ is in quotation marks is because the ‘spin wave’ in the superposition is not a wave with time, but a wavelike ‘motion’ in Hilbert space. To be all 13 elements at the same time imagine that time is frozen and the system changes from element to element. If the system goes through all the states, the z-components of the spins make a wavelike motion: this is the ‘spin wave’. In Figure 4.13 the ‘spin wave’ of example 4.1 is displayed. In Figure 4.13 the wave starts in the element with coefficient \(x_1\) and changes from element to element in order to \(x_{13}\). This order was chosen at random, it is used to visualize the ‘spin wave’.
What happens to the eigenstates with time? The time dependency of the system is described by the Schrödinger equation. The Hamiltonian used in this report is time independent. Thus the solutions to the Schrödinger equation are the eigenstates of the time independent Hamiltonian multiplied by a time dependent constant. Thus an undisturbed system occupying an eigenstate keeps occupying that eigenstate when time passes by.

Summarizing, the eigenstates can be seen as 'spin waves'. These 'spin waves' are not waves with time, but wavelike 'motions' in Hilbert space. The eigenstates are superpositions of elements in the z-basis. Thus the eigenstate is all the elements at the same time. If time would be frozen, being in a superposition is equal to changing from element to element and this can be seen as a 'spin wave'. However, the Hamiltonian is time independent, thus an undisturbed system keeps occupying the same eigenstate when time passes by.
Conclusions

In this report a one dimensional chain consisting of \( n \) magnetic iron atoms with spin 2 has been examined. The spins of the \( n \) particle system were treated quantum mechanically. The spins of the atoms in the chain are governed by the time independent spin Hamiltonian. The parameters of the Hamiltonian are: the single-spin longitudinal magnetocrystalline anisotropy represented by \( D \), the transverse magnetic anisotropy represented by \( E \) and the exchange energy between neighbouring atoms represented by \( J \). The applied magnetic field in the \( z \)-direction represented by \( B = B \hat{z} \) influences the Hamiltonian too, where \( E_z = g^* \mu_B B \), with \( g^* \) the Landé g-factor and \( \mu_B \) the Bohr magneton. A method has been developed to find the eigenstates and eigenenergies of the time independent spin Hamiltonian resulting in \( 5^n \) eigenstates and eigenenergies. The eigenstates are superpositions of \( z \)-basis elements. The eigenstates thus live in a \( 5^n \) dimensional Hilbert space.

The inelastic tunneling current was predicted for an inelastic electron tunneling spectroscopy (IETS) experiment performed under realistic experimental conditions on a three particle system. An IETS experiment is performed with a scanning tunneling microscope (STM). The tip of the STM is located above an atom in the one dimensional chain. The spin of an electron tunneling from the tip of a STM interacts via exchange with the spin of the atom located beneath the tip. When the energy provided by the bias voltage matches the energy of a possible state transition, the transition can occur. The current is determined by the probability to occupy an eigenstate, the possible state transitions the eigenstate can make and if there is enough energy supplied by the voltage to make the state transition.

The current increases for higher voltages. How fast the current increases can be seen in the differential conductance which is the first derivative of the current. If there is enough energy for new possible state transitions to occur, then the differential conductance increases by making a jump.

For low temperatures, the system has a very high probability to occupy the ground state and thus a good approximation is the limit \( T \to 0 \) \( K \) where the system is in the ground state. For high temperatures the system has a reasonable probability to occupy any of the 125 eigenstates, thus there are more potential state transitions. This results in a start of the current increase at lower voltages than a low temperature system. Overall a high temperature system has a higher current than a low temperature system.

The location of the STM tip influences which state transitions are possible and different state transitions have a different effect on the current. The influence of the location of the STM tip is the same for symmetric atoms, for example in the three particle system the current is the same when the STM tip is above atom 1 or atom 3. This also holds for chains of even length, for example for a four particle system the symmetric pairs of atoms are 1 with 4 and 2 with 3.

When the number of atoms in a chain increases, the number of eigenstates increases too. Thus there is an increase in the total number of possible state transitions, because there are more states to transition to. But the probability to occupy a specific eigenstate decreases, because the number of eigenstates was increased. Thus the differential conductance makes more jumps, because there are more possible state transitions. But the jumps are smaller, because the probability to occupy a specific state is smaller. When the number of atoms decreases, the differential conductance makes less jumps, because there are less possible state transitions. But the jumps are bigger, because the probability to occupy a specific state is bigger.

The anisotropies \( D \) and \( E \) are determined by the type of the atoms in the chain, the exchange energy between neighbouring atoms \( J \) depends on the distance between the atoms and \( E_z \) is determined by the
strength of the applied magnetic field. Different values of $D$, $E$, $J$ and $E_z$ lead to different eigenenergies and eigenstates. If new values for $D$, $E$, $J$ and $E_z$ are used, then the new eigenstates and eigenenergies have to be calculated for these new parameters. When the new eigenstates and eigenenergies are known the new inelastic tunneling current can be calculated.

The Hamiltonian is time independent, thus an undisturbed system keeps occupying the same eigenstate when time passes by. The eigenstates are superpositions of $z$-basis elements. To be all elements of the superposition at the same time imagine that time is frozen and the system changes from element to element. If the system goes through all the elements, the $z$-components of the spins make a wavelike motion: this is the 'spin wave'. The reason 'spin wave' is in quotation marks is because the 'spin wave' in the superposition is a wavelike 'motion' in Hilbert space and not a wave with time.

In this report two, three and four particle systems of magnetic atoms with spin 2 have been examined. The developed method to find the eigenstates and eigenenergies of the Hamiltonian can be used for any $n$ particle system. However, for large one dimensional chains a lot of computing power and storage is needed, because $5^n$ eigenstates and eigenenergies have to be calculated and stored and these numbers increase rapidly for large $n$. Keep this in mind when researching large one dimensional chains of magnetic atoms with spin 2.

The developed method can be used for one dimensional chains containing magnetic atoms with spins other than 2, but then some modifications have to be made.
Bibliography


Appendix

In this appendix formulas will be derived step by step and each step will be explained. Some calculations are performed by matlab codes, the codes are listed in this appendix too, feel free to use them.

A.1. Derivation of formula 3.1
Let formula 2.2 act on \(|m_1 m_2 m_3\rangle\), with \(n = 3\). Remember each particle has spin 2. Then

\[
\mathcal{H}|m_1 m_2 m_3\rangle = \sum_{i=1}^{3} \left( D(S_{z}^{(i)})^2 + E \left( J(S_{x}^{(i)})^2 - \left( S_{y}^{(i)} \right)^2 \right) - g^* \mu_B B \cdot \hat{\mathcal{S}}^{(i)} \right) |m_1 m_2 m_3\rangle
\]

From now on \(E_2 = g^* \mu_B B\) will be used. Take a look at the second sum above, the inner product will be simplified as follows:

\[
\mathcal{H}|m_1 m_2 m_3\rangle = \sum_{i=1}^{3} \left( D(S_{z}^{(i)})^2 - g^* \mu_B B \hat{S}_{z}^{(i)} \right) |m_1 m_2 m_3\rangle + \sum_{i=1}^{3} \hat{J} \hat{S}^{(i)} \cdot \hat{S}^{(i+1)} |m_1 m_2 m_3\rangle
\]

The external magnetic field will be assumed to point in the \(\hat{z}\)-direction, so \(B = B \hat{z}\). And zero transverse anisotropy \(E = 0\) will be assumed to make it easier to find the eigenstates of the Hamiltonian. In section 3.5 the results of the Hamiltonian with the \(E = 0\) assumption will be used to analyse the Hamiltonian with \(E \neq 0\). Then

\[
\mathcal{H}|m_1 m_2 m_3\rangle = \sum_{i=1}^{3} \left( D(S_{z}^{(i)})^2 - g^* \mu_B B \hat{S}_{z}^{(i)} \right) |m_1 m_2 m_3\rangle + \sum_{i=1}^{3} \hat{J} \hat{S}^{(i)} \cdot \hat{S}^{(i+1)} |m_1 m_2 m_3\rangle
\]

From now on \(E_z = g^* \mu_B B\) will be used. Take a look at the second sum above, the inner product will be simplified as follows:

\[
\hat{S}^{(i)} \cdot \hat{S}^{(i+1)} = S_{x}^{(i)} S_{x}^{(i+1)} + S_{y}^{(i)} S_{y}^{(i+1)} + S_{z}^{(i)} S_{z}^{(i+1)}
\]

\[
= \frac{1}{4} \left( S_{+}^{(i)} S_{+}^{(i+1)} + S_{-}^{(i)} S_{-}^{(i+1)} \right)
\]

\[
= \frac{1}{4} \left( S_{+}^{(i)} - S_{-}^{(i)} \right) \left( S_{+}^{(i+1)} - S_{-}^{(i+1)} \right) + S_{z}^{(i)} S_{z}^{(i+1)}
\]

\[
= \frac{1}{4} \left( S_{+}^{(i)} S_{+}^{(i+1)} + S_{-}^{(i)} S_{-}^{(i+1)} \right) + \frac{1}{4} \left( S_{+}^{(i)} - S_{-}^{(i)} \right) \left( S_{+}^{(i+1)} - S_{-}^{(i+1)} \right) + S_{z}^{(i)} S_{z}^{(i+1)}
\]

\[
= \frac{1}{2} \left( S_{+}^{(i)} S_{-}^{(i+1)} + S_{-}^{(i)} S_{+}^{(i+1)} \right) + S_{z}^{(i)} S_{z}^{(i+1)}.
\]

Because \(S_{x} = \frac{1}{2} \left( \hat{S}_{x} + \hat{S}_{-} \right)\) and \(S_{y} = \frac{1}{2} \left( \hat{S}_{y} - \hat{S}_{-} \right)\), with \(i\) the imaginary number with \(i^2 = -1\), not the index of the sum.

Now the second sum can be simplified:

\[
\mathcal{H}|m_1 m_2 m_3\rangle = \sum_{i=1}^{3} \left( D(S_{z}^{(i)})^2 - E_z S_{z}^{(i)} \right) |m_1 m_2 m_3\rangle
\]

\[
+ \sum_{i=1}^{3} \frac{1}{2} \left( S_{+}^{(i)} S_{-}^{(i+1)} + S_{-}^{(i)} S_{+}^{(i+1)} \right) + S_{z}^{(i)} S_{z}^{(i+1)} |m_1 m_2 m_3\rangle.
\]
The next step is to write down all the elements of both sums:

\[
\mathcal{H}|m_1 m_2 m_3\rangle = D\left(\hat{S}_z^{(1)}\right)^2|m_1 m_2 m_3\rangle - E_z\hat{S}_z^{(1)}|m_1 m_2 m_3\rangle \\
+ D\left(\hat{S}_z^{(2)}\right)^2|m_1 m_2 m_3\rangle - E_z\hat{S}_z^{(2)}|m_1 m_2 m_3\rangle \\
+ D\left(\hat{S}_z^{(3)}\right)^2|m_1 m_2 m_3\rangle - E_z\hat{S}_z^{(3)}|m_1 m_2 m_3\rangle \\
+ \frac{J}{2}\hat{S}_+^{(1)}\hat{S}_-^{(2)}|m_1 m_2 m_3\rangle + \frac{J}{2}\hat{S}_-^{(1)}\hat{S}_+^{(2)}|m_1 m_2 m_3\rangle + f\hat{S}_z^{(1)}\hat{S}_z^{(2)}|m_1 m_2 m_3\rangle \\
+ \frac{J}{2}\hat{S}_+^{(2)}\hat{S}_-^{(3)}|m_1 m_2 m_3\rangle + \frac{J}{2}\hat{S}_-^{(2)}\hat{S}_+^{(3)}|m_1 m_2 m_3\rangle + f\hat{S}_z^{(2)}\hat{S}_z^{(3)}|m_1 m_2 m_3\rangle.
\]

Now let the spin operators act on \(|m_1 m_2 m_3\rangle\), remember the total spin of every atom is 2, so \(s(s+1) = 6\).

\[
\mathcal{H}|m_1 m_2 m_3\rangle = D\hat{S}_z^{(1)}|m_1 m_2 m_3\rangle - E_z m_1|m_1 m_2 m_3\rangle \\
+ D\hat{S}_z^{(2)}|m_1 m_2 m_3\rangle - E_z m_2|m_1 m_2 m_3\rangle \\
+ D\hat{S}_z^{(3)}|m_1 m_2 m_3\rangle - E_z m_3|m_1 m_2 m_3\rangle \\
+ \frac{J}{2}\hat{S}_+^{(1)}\sqrt{6-m_2(m_2-1)}|m_1 (m_2-1) m_3\rangle \\
+ \frac{J}{2}\hat{S}_-^{(1)}\sqrt{6-m_2(m_2+1)}|m_1 (m_2+1) m_3\rangle + f\hat{S}_z^{(1)}|m_1 m_2 m_3\rangle \\
+ \frac{J}{2}\hat{S}_+^{(2)}\sqrt{6-m_3(m_3-1)}|m_1 m_2 (m_3-1)\rangle \\
+ \frac{J}{2}\hat{S}_-^{(2)}\sqrt{6-m_3(m_3+1)}|m_1 m_2 (m_3+1)\rangle + f\hat{S}_z^{(2)}|m_1 m_2 m_3\rangle \\
= D(m_1)^2|m_1 m_2 m_3\rangle - E_z m_1|m_1 m_2 m_3\rangle \\
+ D(m_2)^2|m_1 m_2 m_3\rangle - E_z m_2|m_1 m_2 m_3\rangle \\
+ D(m_3)^2|m_1 m_2 m_3\rangle - E_z m_3|m_1 m_2 m_3\rangle \\
+ \frac{J}{2}\sqrt{6-m_2(m_2+1)}\sqrt{6-m_2(m_2-1)}|m_1 (m_2+1) (m_2-1) m_3\rangle \\
+ \frac{J}{2}\sqrt{6-m_2(m_2+1)}\sqrt{6-m_2(m_2-1)}|m_1 (m_2+1) (m_2-1) m_3\rangle \\
+ J m_1 m_2|m_1 m_2 m_3\rangle \\
+ \frac{J}{2}\sqrt{6-m_2(m_2+1)}\sqrt{6-m_3(m_3+1)}|m_1 (m_2+1) (m_3+1)\rangle \\
+ \frac{J}{2}\sqrt{6-m_2(m_2-1)}\sqrt{6-m_3(m_3+1)}|m_1 (m_2-1) (m_3+1)\rangle \\
+ J m_2 m_3|m_1 m_2 m_3\rangle.
\]

The final step is sorting element by element.

\[
\mathcal{H}|m_1 m_2 m_3\rangle = \left(\left((m_1)^2 + (m_2)^2 + (m_3)^2\right) D - (m_1 + m_2 + m_3) E_z + (m_1 m_2 + m_2 m_3) f\right) |m_1 m_2 m_3\rangle \\
+ \frac{J}{2}\sqrt{6-m_2(m_2+1)}\sqrt{6-m_2(m_2-1)}|m_1 (m_2+1) (m_2-1) m_3\rangle \\
+ \frac{J}{2}\sqrt{6-m_2(m_2+1)}\sqrt{6-m_2(m_2-1)}|m_1 (m_2+1) (m_2-1) m_3\rangle \\
+ \frac{J}{2}\sqrt{6-m_2(m_2+1)}\sqrt{6-m_3(m_3+1)}|m_1 (m_2+1) (m_3+1)\rangle \\
+ \frac{J}{2}\sqrt{6-m_2(m_2-1)}\sqrt{6-m_3(m_3+1)}|m_1 (m_2-1) (m_3+1)\rangle.
\]
A.2. Find the other eigenstates and eigenenergies of the three particle system with zero transverse magnetic anisotropy

In the section 3.3 13 groups were found. With these groups the eigenstates and eigenenergies of the Hamiltonian can be found. Group 1 and 2 were discussed in section 3.4, the process used for the remaining 11 groups is similar as the process for group 2, but formula 3.1 will be used to quickly determine the Hamiltonian of an element from the superposition. Remember $M$ is an eigenstate of the Hamiltonian if $\mathcal{H}M = E_M M$ and $\langle M| M \rangle = 1$, hold, with $E_M$ the eigenenergy of eigenstate $M$.

Group 3 has $m_1 + m_2 + m_3 = -4$. Define a superposition $G_3$ with the elements of group 3,

$$G_3 = a |0\rangle - 2 - 2 \rangle + b |1\rangle - 1 - 1 \rangle + d |1\rangle - 1 - 2 \rangle + f |2\rangle - 2 - 2 \rangle$$

and find the coefficients for which the superposition is an eigenstate of the Hamiltonian. Check the properties of an eigenstate and the requirements for the coefficients. Using the inner product properties discussed in section 2.4 the first requirement for the coefficients follows:

$$\langle G_3|G_3 \rangle = 1 \iff |a|^2 + |b|^2 + |c|^2 + |d|^2 + |e|^2 + |f|^2 = 1$$

For the other requirements let the Hamiltonian act on $G_3$.

$$\mathcal{H}G_3 = \mathcal{H} \left( a |0\rangle - 2 - 2 \rangle + b |1\rangle - 1 - 1 \rangle + c |1\rangle - 1 - 2 \rangle + d |1\rangle - 2 - 1 \rangle + e |2\rangle - 2 - 1 \rangle + f |2\rangle - 2 - 2 \rangle \right)$$

This can be written as an eigenvalue problem.

$$\begin{pmatrix} a \\ b \\ c \\ d \\ e \\ f \end{pmatrix} = E_{G_3} \begin{pmatrix} a \\ b \\ d \\ e \\ f \end{pmatrix}$$

Determine the eigenvalues $\lambda_{A_3}$ and corresponding eigenvectors of matrix $A_3$ with the matlab code, see appendix A.7. Matlab finds six eigenvalues and six orthonormal eigenvectors, because $A_3$ is a symmetric matrix, thus satisfying the first requirement. With

$$A_3 = \begin{pmatrix} 2D + 4J & \sqrt{6}J & 0 & 0 & 0 & 0 \\ \sqrt{6}J & 3J & 2J & \sqrt{6}J & 0 & 0 \\ 0 & 2J & 4J & 0 & 2J & 0 \\ 0 & \sqrt{6}J & 0 & 2D & \sqrt{6}J & 0 \\ 0 & 0 & 2J & \sqrt{6}J & 3J & \sqrt{6}J \\ 0 & 0 & 0 & 0 & \sqrt{6}J & 2D + 4J \end{pmatrix}$$

Then the eigenvalues are $E_{G_3} = 6D + 4E_z + \lambda_{A_3}$. And the eigenstates are the corresponding eigenvectors of $\lambda_{A_3}$. To see the exact numbers take a look at appendix A.7 how to run the code and access the exact numbers of the coefficients.
Group 4 has $m_1 + m_2 + m_3 = -3$. Define a superposition $G_4$ with the elements of group 4,

\[
G_4 = a|1 - 2 - 2\rangle + b|0 - 1 - 2\rangle + c|0 - 2 - 1\rangle + d|1 0 - 2\rangle + e|1 - 1 - 1\rangle + f|1 - 2 0\rangle + g|-2 1 - 2\rangle + h|-2 0 - 1\rangle + i|-2 1 0\rangle + j|-2 2 1\rangle
\]

and find the coefficients for which the superposition is an eigenstate of the Hamiltonian. Check the properties of an eigenstate and get the requirements for the coefficients. Using the inner product properties discussed in section 2.4 the first requirement for the coefficients follows:

\[
\langle G_4 | G_4 \rangle = 1 \iff |a|^2 + |b|^2 + |c|^2 + |d|^2 + |e|^2 + |f|^2 + |g|^2 + |h|^2 + |i|^2 + |j|^2 = 1
\]

For the other requirements let the Hamiltonian act on $G_4$.

\[
\mathcal{H}G_4 = \mathcal{H} \left( a|1 - 2 - 2\rangle + b|0 - 1 - 2\rangle + c|0 - 2 - 1\rangle + d|1 0 - 2\rangle + e|1 - 1 - 1\rangle + f|1 - 2 0\rangle + g|-2 1 - 2\rangle + h|-2 0 - 1\rangle + i|-2 1 0\rangle + j|-2 2 1\rangle \right)
\]

\[
= \left( (9D + 3E_z + 2f) \left( a + \sqrt{6}Jb \right)|1 - 2 - 2\rangle + (5D + 3E_z + 2f) \left( b + \sqrt{6}Ja + 3Jd + 2Jc \right)|0 - 1 - 2\rangle \\
+ (5D + 3E_z + 2f) \left( c + \sqrt{6}Je + 2Jb \right)|0 - 2 - 1\rangle + (5D + 3E_z + 2f) \left( d + 3Jb + \sqrt{6}Jg + \sqrt{6}Je \right)|1 - 0 - 2\rangle \\
+ (3D + 3E_z + 2f) \left( e + \sqrt{6}Jc + \sqrt{6}Jf + \sqrt{6}Jd \right)|1 - 1 - 1\rangle \\
+ (5D + 3E_z + 2f) \left( f + 2Ji + \sqrt{6}Je \right)|1 - 2 - 0\rangle + (9D + 3E_z + 2f) \left( g + \sqrt{6}Jd + \sqrt{6}Jh \right)|2 - 1 - 1\rangle \\
+ (5D + 3E_z + 2f) \left( h + \sqrt{6}Je + \sqrt{6}Jg + 3Jf \right)|2 - 0 - 1\rangle \\
+ (5D + 3E_z + 2f) \left( i + 2Jf + 3Jh + \sqrt{6}Jf \right)|2 - 1 0\rangle + (9D + 3E_z + 2f) \left( j + \sqrt{6}Ji \right)|2 - 2 1\rangle \right)
\]

\[
= E_{G_4} a|1 - 2 - 2\rangle + E_{G_4} b|0 - 1 - 2\rangle + E_{G_4} c|0 - 2 - 1\rangle + E_{G_4} d|1 0 - 2\rangle + E_{G_4} e|1 - 1 - 1\rangle \\
+ E_{G_4} f|1 - 2 0\rangle + E_{G_4} g|-2 1 - 2\rangle + E_{G_4} h|-2 0 - 1\rangle + E_{G_4} i|-2 1 0\rangle + E_{G_4} j|-2 2 1\rangle
\]

This can be written as an eigenvalue problem.

\[
(3D + 3E_z) I_{10 \times 10} \begin{bmatrix}
   a \\
   \vdots \\
   j
\end{bmatrix} + A_4 \begin{bmatrix}
   a \\
   \vdots \\
   j
\end{bmatrix} = E_{G_4} \begin{bmatrix}
   a \\
   \vdots \\
   j
\end{bmatrix}
\]

With

\[
A_4 = \begin{bmatrix}
   6D + 2f & \sqrt{6}J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
   \sqrt{6}J & 2D + 2f & 2J & 3J & 0 & 0 & 0 & 0 & 0 & 0 \\
   0 & 2J & 2D + 2f & 0 & \sqrt{6}J & 0 & 0 & 0 & 0 & 0 \\
   0 & 3J & 0 & 2D & \sqrt{6}J & 0 & \sqrt{6}J & 0 & 0 & 0 \\
   0 & 0 & \sqrt{6}J & \sqrt{6}J & 2J & \sqrt{6}J & 0 & \sqrt{6}J & 0 & 0 \\
   0 & 0 & 0 & \sqrt{6}J & 0 & \sqrt{6}J & 0 & \sqrt{6}J & 0 & 0 \\
   0 & 0 & 0 & 0 & \sqrt{6}J & 0 & \sqrt{6}J & 0 & \sqrt{6}J & 0 \\
   0 & 0 & 0 & 0 & 0 & \sqrt{6}J & 0 & \sqrt{6}J & 0 & \sqrt{6}J \\
   0 & 0 & 0 & 0 & 0 & 0 & \sqrt{6}J & 0 & \sqrt{6}J & 0 \\
   0 & 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{6}J & 0 & \sqrt{6}J
\end{bmatrix}
\]

Determine the eigenvalues $\lambda_{A_4}$ and corresponding eigenvectors of matrix $A_4$ with the matlab code, see appendix A.7. Matlab finds ten eigenvalues and ten orthonormal eigenvectors, because $A_4$ is a symmetric matrix, thus satisfying the first requirement. Then the eigenenergies are $E_{G_4} = 3D + 3E_z + \lambda_{A_4}$. And the eigenstates are the corresponding eigenvectors of $\lambda_{A_4}$. To see the exact numbers take a look at appendix A.7 how to run the code and access the exact numbers of the coefficients.
Group 5 has $m_1 + m_2 + m_3 = -2$. Define a superposition $G_5$ with the elements of group 5,

$$G_5 = a|2 - 2 - 2\rangle + b|1 - 1 - 2\rangle + c|1 - 2 - 1\rangle + d|0 0 - 2\rangle + e|0 - 1 - 1\rangle + f|0 - 2 0\rangle + g|1 - 1 - 2\rangle + h|1 - 0 - 1\rangle + i|1 - 1 0\rangle + j|1 - 1 - 2\rangle + k|2 - 2 - 2\rangle + l|2 1 - 1\rangle + m|2 0 0\rangle + n|2 - 1 1\rangle + o|2 - 2 2\rangle$$

and find the coefficients for which the superposition is an eigenstate of the Hamiltonian. Check the properties of an eigenstate and get the requirements for the coefficients. Using the inner product properties discussed in section 2.4 the first requirement for the coefficients follows:

$$\langle G_5 | G_2 \rangle = 1 \Leftrightarrow |a|^2 + |b|^2 + |c|^2 + |d|^2 + |e|^2 + |f|^2 + |g|^2 + |h|^2 + |i|^2 + |j|^2 + |k|^2 + |l|^2 + |m|^2 + |n|^2 + |o|^2 = 1$$

For the other requirements let the Hamiltonian act on $G_5$.

$$\mathcal{H} G_5 = \mathcal{H} \left[ a|2 - 2 - 2\rangle + b|1 - 1 - 2\rangle + c|1 - 2 - 1\rangle + d|0 0 - 2\rangle + e|0 - 1 - 1\rangle + f|0 - 2 0\rangle + g|1 - 1 - 2\rangle + h|1 - 0 - 1\rangle + i|1 - 1 0\rangle + j|1 - 1 - 2\rangle + k|2 - 2 - 2\rangle + l|2 1 - 1\rangle + m|2 0 0\rangle + n|2 - 1 1\rangle + o|2 - 2 2\rangle \right]$$

This can be written as an eigenvalue problem.

$$(2D + 2E_z) \mathbf{f}_{15 \times 15} \begin{bmatrix} a \\ \vdots \\ a \\ \vdots \\ o \\ \vdots \\ o \end{bmatrix} + A_5 \begin{bmatrix} a \\ \vdots \\ a \\ \vdots \\ o \\ \vdots \\ o \end{bmatrix} = E G_5 \begin{bmatrix} a \\ \vdots \\ a \\ \vdots \\ o \\ \vdots \\ o \end{bmatrix}$$
With $A_5 = \begin{pmatrix} 10D & 2J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 2J & 4D + J & 2J & 3J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 2J & 4D & 0 & \sqrt{6}J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 3J & 0 & 2D & \sqrt{6}J & 0 & 3J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & \sqrt{6}J & \sqrt{6}J & J & \sqrt{6}J & 0 & 3J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \sqrt{6}J & 2D & 0 & 0 & \sqrt{6}J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 3J & 0 & 0 & 4D - 3J & \sqrt{6}J & 0 & 0 & 2J & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & \sqrt{6}J & 0 & 3J & J & \sqrt{6}J & 0 & 0 & \sqrt{6}J & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{6}J & 0 & 0 & 2J & 4D - 3J & 3J & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{6}J & 0 & 0 & 3J & 2D & 3J & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 2J & 0 & 0 & 3J & 4D + J & 2J \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 2J & 10D \end{pmatrix}$

Determine the eigenvalues $\lambda_{A_5}$ and corresponding eigenvectors of matrix $A_5$ with the matlab code, see appendix A.7. Matlab finds fifteen eigenvalues and fifteen orthonormal eigenvectors, because $A_5$ is a symmetric matrix, thus satisfying the first requirement. Then the eigenenergies are $E_{G_5} = 2D + 2E_{z} + \lambda_{A_5}$. And the eigenstates are the corresponding eigenvectors of $\lambda_{A_5}$. To see the exact numbers take a look at appendix A.7 how to run the code and access the exact numbers of the coefficients.

Group 6 has $m_1 + m_2 + m_3 = -1$. Define a superposition $G_6$ with the elements of group 6,

$$G_6 = x_1|2 - 1 - 2\rangle + x_2|2 - 2 - 1\rangle + x_3|1 0 - 2\rangle + x_4|1 - 1 - 1\rangle + x_5|1 - 2 0\rangle + x_6|0 1 - 2\rangle + x_7|0 0 - 1\rangle + x_8|0 0 - 1\rangle + x_9|0 - 2 1\rangle + x_{10}|1 - 2 - 2\rangle + x_{11}|1 1 - 1\rangle + x_{12}|1 0 0\rangle + x_{13}|1 - 1 - 1\rangle + x_{14}|1 - 1 - 2\rangle + x_{15}|2 2 - 1\rangle + x_{16}|2 1 0\rangle + x_{17}|2 0 1\rangle + x_{18}|2 - 1 - 2\rangle$$

and find the coefficients for which the superposition is an eigenstate of the Hamiltonian. Check the properties of an eigenstate and get the requirements for the coefficients. Using the inner product properties discussed in section 2.4 the first requirement for the coefficients follows:

$$\langle G_6|G_6 \rangle = 1 \iff |x_1|^2 + |x_2|^2 + |x_3|^2 + |x_4|^2 + |x_5|^2 + |x_6|^2 + |x_7|^2 + |x_8|^2 + |x_9|^2 + |x_{10}|^2 + |x_{11}|^2 + |x_{12}|^2 + |x_{13}|^2 + |x_{14}|^2 + |x_{15}|^2 + |x_{16}|^2 + |x_{17}|^2 + |x_{18}|^2 = 1$$
A.2. Find the other eigenstates and eigenenergies of the three particle system with zero transverse magnetic anisotropy

For the other requirements let the Hamiltonian act on $G_6$.

\[ \mathcal{H} G_6 = \mathcal{H} \left( x_1 |2 - 1 - 2\rangle + x_2 |2 - 2 - 1\rangle + x_3 |1 0 - 2\rangle + x_4 |1 - 1 - 1\rangle + x_5 |1 - 2 0\rangle + x_6 |0 1 - 2\rangle 
+ x_7 |0 0 - 1\rangle + x_8 |0 - 1 0\rangle + x_9 |0 - 2 1\rangle + x_{10} |- 1 2 - 2\rangle + x_{11} |- 1 1 - 1\rangle + x_{12} |- 1 0 0\rangle 
+ x_{13} |1 - 1 1\rangle + x_{14} |- 1 - 2 2\rangle + x_{15} |- 2 2 - 1\rangle + x_{16} |- 2 1 0\rangle + x_{17} |- 2 0 1\rangle + x_{18} |- 2 - 2 1\rangle \right) \]

\[ = (9D + 2E_z) x_1 + 2J x_2 + \sqrt{6}J x_3 |2 - 1 - 2\rangle + (9D + 2E_z - 2J) x_2 + 2J x_1 + 2J x_4 |2 - 2 - 1\rangle \]

\[ + (5D + E_z) x_3 + \sqrt{6}J x_1 + 3J x_4 + 3J x_6 |1 0 - 2\rangle \]

\[ + (3D + E_z) x_4 + 2J x_2 + \sqrt{6}J x_3 + 3J x_5 + 3J x_7 |1 - 1 - 1\rangle \]

\[ + (5D + E_z - 2J) x_5 + \sqrt{6}J x_4 + 3J x_6 |1 - 2 0\rangle \]

\[ + (5D + E_z - 2J) x_6 + 3J x_3 + \sqrt{6}J x_5 + \sqrt{6}J x_7 |1 0 - 2\rangle \]

\[ + (D + E_z) x_7 + 3J x_4 + \sqrt{6}J x_6 + 3J x_8 + 3J x_1 |0 0 - 1\rangle \]

\[ + (D + E_z) x_8 + \sqrt{6}J x_5 + 3J x_7 + \sqrt{6}J x_9 + 3J x_{12} |0 - 1 0\rangle \]

\[ + (5D + E_z - 2J) x_9 + \sqrt{6}J x_8 + \sqrt{6}J x_{13} |0 - 2 1\rangle + (9D + E_z - 6J) x_{10} + \sqrt{6}J x_6 + 2J x_{11} |1 - 2 - 2\rangle \]

\[ + (3D + E_z - 2J) x_{11} + 3J x_7 + 2J x_{10} + 3J x_{12} + 2J x_{15} |1 1 - 1\rangle \]

\[ + (D + E_z) x_{12} + 3J x_9 + 3J x_{11} + 3J x_{13} + \sqrt{6}J x_{16} |1 0 0\rangle \]

\[ + (3D + E_z) x_{13} + \sqrt{6}J x_9 + 3J x_{12} + 2J x_{14} + \sqrt{6}J x_{17} |1 - 1 1\rangle \]

\[ + (9D + E_z - 2J) x_{14} + 2J x_{13} + 2J x_{18} |1 - 1 2\rangle + (9D + E_z - 6J) x_{15} + 2J x_{11} + \sqrt{6}J x_{16} |2 2 - 1\rangle \]

\[ + (5D + E_z - 2J) x_{16} + \sqrt{6}J x_{12} + \sqrt{6}J x_{15} + 3J x_{17} |1 1 0\rangle \]

\[ + (5D + E_z) x_{17} + \sqrt{6}J x_{13} + 3J x_{16} + \sqrt{6}J x_{18} |2 0 1\rangle + (9D + E_z) x_{18} + 2J x_{14} + \sqrt{6}J x_{17} |2 - 2 1\rangle \]

\[ = E_{G_6} x_1 |2 - 1 - 2\rangle + E_{G_6} x_2 |2 - 2 - 1\rangle + E_{G_6} x_3 |1 0 - 2\rangle + E_{G_6} x_4 |1 - 1 - 1\rangle + E_{G_6} x_5 |1 - 2 0\rangle 
+ E_{G_6} x_6 |0 1 - 2\rangle + E_{G_6} x_7 |0 0 - 1\rangle + E_{G_6} x_8 |0 - 1 0\rangle + E_{G_6} x_9 |0 - 2 1\rangle + E_{G_6} x_{10} |- 1 2 - 2\rangle 
+ E_{G_6} x_{11} |- 1 1 - 1\rangle + E_{G_6} x_{12} |- 1 0 0\rangle + E_{G_6} x_{13} |- 1 - 1 1\rangle + E_{G_6} x_{14} |- 1 - 2 2\rangle 
+ E_{G_6} x_{15} |- 2 2 - 1\rangle + E_{G_6} x_{16} |- 2 1 0\rangle + E_{G_6} x_{17} |- 2 0 1\rangle + E_{G_6} x_{18} |- 2 - 2 1\rangle \]

This can be written as an eigenvalue problem.

\[
(D + E_z) \begin{bmatrix} x_1 \\ \vdots \\ x_{18} \end{bmatrix} + A_6 \begin{bmatrix} x_1 \\ \vdots \\ x_{18} \end{bmatrix} = E_{G_6} \begin{bmatrix} x_1 \\ \vdots \\ x_{18} \end{bmatrix}
\]
With $A_6 = \begin{bmatrix} 8D & 2I & \sqrt{\bar{J}} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 2I & 8D-2J & 0 & 2I & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ \sqrt{\bar{J}} & 0 & 4D & \sqrt{\bar{J}} & 0 & 3J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 2I & \sqrt{\bar{J}} & 2D & \sqrt{\bar{J}} & 0 & 3J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & \sqrt{\bar{J}} & 4D-2J & 0 & 0 & \sqrt{\bar{J}} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \sqrt{\bar{J}} & 0 & 3J & 0 & 0 & 0 & 3J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & \sqrt{\bar{J}} & 0 & 3J & 0 & 0 & 3J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & \sqrt{\bar{J}} & 0 & 0 & 3J & 0 & 3J & 0 & 0 & \sqrt{\bar{J}} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{\bar{J}} & 0 & 0 & 3J & 2D & 2J & 0 & 0 & \sqrt{\bar{J}} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 2J & 4D-2J & 0 & 0 & 0 & 2J & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 2J & 0 & 0 & 0 & \sqrt{\bar{J}} & 0 & 0 & \sqrt{\bar{J}} & 4D-2J & 3J & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{\bar{J}} & 0 & 0 & \sqrt{\bar{J}} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{\bar{J}} & 0 & 0 & \sqrt{\bar{J}} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{\bar{J}} & 0 & 0 & \sqrt{\bar{J}} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{\bar{J}} & 0 & 0 & \sqrt{\bar{J}} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 2J & 0 & 0 & 0 & 0 & \sqrt{\bar{J}} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{\bar{J}} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{\bar{J}} & 0 & 0 & \sqrt{\bar{J}} \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{\bar{J}} & 0 & 0 & \sqrt{\bar{J}} \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{\bar{J}} & 0 & 0 & \sqrt{\bar{J}} \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{\bar{J}} & 0 & 0 & \sqrt{\bar{J}} \end{bmatrix}$

Determine the eigenvalues $\lambda_{\xi_6}$ and corresponding eigenvectors of matrix $A_6$ with the matlab code, see appendix A.7. Matlab finds eighteen eigenvalues and eighteen orthonormal eigenvectors, because $A_6$ is a symmetric matrix, thus satisfying the first requirement. Then the eigenenergies are $E_{\xi_6} = D + E_2 + \lambda_{\xi_6}$. And the eigenstates are the corresponding eigenvectors of $\lambda_{\xi_6}$. To see the exact numbers take a look at appendix A.7 how to run the code and access the exact numbers of the coefficients.

Group 7 has $m_1 + m_2 + m_3 = 0$. Define a superposition $G_7$ with the elements of group 7,

$$G_7 = x_1|20-2\rangle + x_2|2-1\rangle + x_3|2-2\rangle + x_4|11-2\rangle + x_5|10-1\rangle + x_6|1-1\rangle + x_7|1-2\rangle + x_8|02-2\rangle + x_9|01-1\rangle + x_{10}|000\rangle + x_{11}|0-1\rangle + x_{12}|0-2\rangle + x_{13}|1-2\rangle + x_{14}|110\rangle + x_{15}|101\rangle + x_{16}|1-1\rangle + x_{17}|2-2\rangle + x_{18}|211\rangle + x_{19}|202\rangle$$

and find the coefficients for which the superposition is an eigenstate of the Hamiltonian. Check the properties of an eigenstate and get the requirements for the coefficients. Using the inner product properties discussed in section 2.4 the first requirement for the coefficients follows:

$$\langle G_7|G_7 \rangle = 1 \iff |x_1|^2 + |x_2|^2 + |x_3|^2 + |x_4|^2 + |x_5|^2 + |x_6|^2 + |x_7|^2 + |x_8|^2 + |x_9|^2 + |x_{10}|^2 + |x_{11}|^2 + |x_{12}|^2 + |x_{13}|^2 + |x_{14}|^2 + |x_{15}|^2 + |x_{16}|^2 + |x_{17}|^2 + |x_{18}|^2 + |x_{19}|^2 = 1$$
For the other requirements let the Hamiltonian act on $G_7$.

$$\mathcal{H}G_7 = \mathcal{H} \left[ x_1 | 2 \ 0 \ -2 \right] + x_2 | 2 \ 1 \ -1 \ + x_3 | 2 \ 2 \ -2 \ + x_4 | 1 \ 1 \ -2 \ + x_5 | 1 \ 0 \ -1 \ + x_6 | 1 \ -1 \ -2 \ -2 \ (2 \ -2) \ + x_13 | -1 \ -2 \ -2 \ -2 \ (2 \ -2) \ + x_14 | -1 \ 1 \ 0 \ + x_15 | -1 \ 1 \ 0 \ + x_16 | 1 \ -1 \ -2 \ -2 \ (2 \ -2) \ + x_17 | -2 \ 2 \ 0 \ + x_18 | -2 \ 1 \ 1 \ + x_19 | -2 \ 0 \ 2 \right]$$

$$= \left( BD x_1 + \sqrt{6} x_2 \ + \sqrt{6} x_4 \right) | 2 \ 0 \ -2 \right) + \left( (6D - J) x_3 + \sqrt{6} x_1 + \sqrt{6} x_3 + \sqrt{6} x_5 \right) | 2 \ 1 \ -1 \ -2 \ (2 \ -2) \ + (2D - J) x_4 \ + \sqrt{6} x_4 + \sqrt{6} x_5 \ + \sqrt{6} x_8 \right) | 1 \ 1 \ -2 \$$

$$= \left( (6D - J) x_5 + \sqrt{6} x_5 + 3J x_4 + 3J x_9 \right) | 1 \ 0 \ -1 \$$

This can be written as an eigenvalue problem.

$$A_7 \left[ \begin{array}{c} x_1 \\ \vdots \\ x_{19} \end{array} \right] = E_{G_7} \left[ \begin{array}{c} x_1 \\ \vdots \\ x_{19} \end{array} \right]$$

With $A_7 = \left[ \begin{array}{cccccccccccccccccc} 8D & \sqrt{6} J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ \sqrt{6} J & 6D - J & \sqrt{6} J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & \sqrt{6} J & 8D - 4J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ \sqrt{6} J & 0 & 6D - J & \sqrt{6} J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & \sqrt{6} J & 0 & \sqrt{6} J & 2D & 3J & \sqrt{6} J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 2J & 0 & 3J & 2D - J & \sqrt{6} J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{6} J & 6D - 4J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 3J & 0 & 3J & 3J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 3J & 0 & 3J & 2D - J & 2J & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 3J & 3J & 2D - J & 2J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 3J & 2D & 3J & 3J & 2D - J & 2J & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{6} J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 3J & 0 & 3J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{array} \right]$$

Determine the eigenvalues $\lambda_{G_7}$ and corresponding eigenvectors of matrix $A_7$ with the matlab code, see appendix A.7. Matlab finds nineteen eigenvalues and nineteen orthonormal eigenvectors, because $A_7$ is a
symmetric matrix, thus satisfying the first requirement. Then the eigenenergies are \( E_G = \lambda A \). And the eigenstates are the corresponding eigenvectors of \( \lambda A \). To see the exact numbers take a look at appendix A.7 how to run the code and access the exact numbers of the coefficients.

Group 8 has \( m_1 + m_2 + m_3 = 1 \). Define a superposition \( G_8 \) with the elements of group 8,

\[
G_8 = x_{18}[2 \cdot 1 - 2] + x_{17}[2 \cdot 0 - 1] + x_{16}[2 \cdot 1 - 0] + x_{15}[2 \cdot 0 - 1]
+ x_{14}[1 \cdot 2 - 2] + x_{13}[1 \cdot 1 - 1] + x_{12}[1 \cdot 0 0] + x_{11}[1 \cdot 0 1]
+ x_{10}[2 \cdot 0 - 1] + x_9[0 \cdot 1 0] + x_8[0 \cdot 0 1] + x_7[0 \cdot 0 1]
+ x_6[0 \cdot 1 0] + x_5[1 \cdot 1 0]
+ x_4[1 \cdot 2 0] + x_3[1 \cdot 0 2] + x_2[1 \cdot 0 2] + x_1[2 \cdot 2 1]
\]

and find the coefficients for which the superposition is an eigenstate of the Hamiltonian. Check the properties of an eigenstate and get the requirements for the coefficients. Using the inner product properties discussed in section 2.4 the first requirement for the coefficients follows:

\[
\langle G_8 | G_8 \rangle = 1 \iff |x_1|^2 + |x_2|^2 + |x_3|^2 + |x_4|^2 + |x_5|^2 + |x_6|^2 + |x_7|^2 + |x_8|^2 + |x_9|^2 + |x_{10}|^2
+ |x_{11}|^2 + |x_{12}|^2 + |x_{13}|^2 + |x_{14}|^2 + |x_{15}|^2 + |x_{16}|^2 + |x_{17}|^2 + |x_{18}|^2 = 1
\]

For the other requirements let the Hamiltonian act on \( G_8 \). The equations that follow can be written as an eigenvalue problem.

\[
(D - E_z) I_{18x18} \begin{bmatrix} x_1 \\ \vdots \\ x_{18} \end{bmatrix} + A_6 \begin{bmatrix} x_1 \\ \vdots \\ x_{18} \end{bmatrix} = E_G \begin{bmatrix} x_1 \\ \vdots \\ x_{18} \end{bmatrix}
\]

Matrix \( A_6 \) appears, because of how the coefficients are placed in front of the states of group 8. Now there is no need to calculate the Hamiltonian of \( G_8 \) and to find the eigenstates and eigenvectors, because the eigenstates and eigenvectors of matrix \( A_6 \) are known.

Take a look at the superpositions of group 6 and 8. Look at the same coefficient, the element of group 8 has the same numbers, but they are minus the numbers of group 6. For example look at the element with coefficient \( x_1 \), in group 6 \( x_1[2 \cdot 1 - 2] \) and in group 8 \( x_1[2 \cdot -1 -2] \). Look at the Hamiltonian acting on a element, formula 3.1. The \( m_i \) in front of \( D \) are squared, thus the difference in minus sign between the elements of group 6 and 8 does not matter. For example let the Hamiltonian act on \( x_1[2 \cdot 1 - 2] \) from group 6, there is a factor \( 9Dx_1[2 \cdot -1 -2] \) and when the Hamiltonian acts on \( x_1[2 \cdot -1 -2] \) from group 8 there is a factor \( 9Dx_1[2 \cdot -1 -2] \). The \( m_i \) in front of \( E_z \) are not squared and there will be a minus sign difference between group 6 and 8 for an element with the same coefficient. For example let the Hamiltonian act on \( x_1[2 \cdot 1 - 2] \) from group 6, there is a factor \( E_z x_1[2 \cdot -1 -2] \) and when the Hamiltonian acts on \( x_1[2 \cdot -1 -2] \) from group 8 there is a factor \( -E_z x_1[2 \cdot -1 -2] \). The \( m_i m_{i+1} \) in front of \( J \) do not have a difference, because for the same coefficient the numbers of group 8 are minus the numbers of group 8. For example let the Hamiltonian act on \( x_1[2 \cdot 1 - 2] \) from group 6, there is a factor \( (-2 + 2) Jx_1[2 \cdot -1 -2] \) and when the Hamiltonian acts on \( x_1[2 \cdot -1 -2] \) from group 8 there is a factor \( (-2 + 2) Jx_1[2 \cdot -1 -2] \) = 0. The Hamiltonian contributes to other elements too. For example when the Hamiltonian acts on \( x_1[2 \cdot 1 - 2] \) of group 6, the result is:

\[
\mathcal{H} x_1[2 \cdot 1 - 2] = (9D + E_z) x_1[2 \cdot 1 - 2] + 2Jx_1[2 \cdot -1 -2] + \sqrt{6} Jx_1[1 \cdot 0 -2]
\]

Now let the Hamiltonian act on \( x_1[2 \cdot -1 -2] \) of group 8.

\[
\mathcal{H} x_1[2 \cdot -1 -2] = (9D - E_z) x_1[2 \cdot -1 -2] + 2Jx_1[2 \cdot 1 -2] + \sqrt{6} Jx_1[1 \cdot 0 -2]
\]

Notice that group 6 contains \( [2 \cdot -2 -1] \) with coefficient \( x_2 \). When the Hamiltonian acts on \( x_1[2 \cdot -1 -2] \) from group 6 there is a factor \( 2Jx_1[2 \cdot -2 -1] \). And notice that group 8 contains \( [2 \cdot -2 -1] \) with coefficient \( x_2 \). When the Hamiltonian acts on \( x_1[2 \cdot -1 -2] \) from group 8 there is a factor \( 2Jx_1[2 \cdot -2 -1] \). Thus for both groups the same factor \( 2Jx_1 \) stands in front of the element corresponding to coefficient \( x_2 \). Thus almost the same equations follow from the Hamiltonian acting on superposition 6 and 8, the explanation for the general case see below. The only difference is superposition 6 has \( E_z \) and superposition 8 has \(-E_z\). But the difference is in all equations, thus can be placed in front of an identity matrix in the eigenvalue problem. The remainder
is the same, hence why the eigenvalue problem of superposition 8 can be written with matrix $A_6$ and a lot of calculations will be prevented.

Above was an example with the coefficient $x_1$, but it is true for all coefficients. See the table below, these are the square roots in front of the additional shifted elements.

<table>
<thead>
<tr>
<th>$m_i$</th>
<th>$\sqrt{6 - m_i (m_i + 1)}$</th>
<th>$\sqrt{6 - m_i (m_i - 1)}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>0</td>
<td>2</td>
</tr>
<tr>
<td>1</td>
<td>2</td>
<td>$\sqrt{6}$</td>
</tr>
<tr>
<td>0</td>
<td>$\sqrt{6}$</td>
<td>$\sqrt{6}$</td>
</tr>
<tr>
<td>-1</td>
<td>$\sqrt{6}$</td>
<td>2</td>
</tr>
<tr>
<td>-2</td>
<td>2</td>
<td>0</td>
</tr>
</tbody>
</table>

Take look at the table, $m_i$ plugged in $\sqrt{6 - m_i (m_i + 1)}$ has the same value as $-m_i$ plugged in $\sqrt{6 - m_i (m_i - 1)}$.

For example if $m_i = 1$ then $\sqrt{6 - m_i (m_i + 1)} = 2$ and $-m_i = -1$ with $\sqrt{6 - m_i (m_i - 1)} = 2$.

In general for coefficient $x_i$, the corresponding element in group 6 equals $|m_{i_1}, m_{i_2}, m_{i_3}\rangle$ and the corresponding element in group 8 equals $|-m_{i_1} - m_{i_2} - m_{i_3}\rangle$. In formula 3.1 look at $\frac{\sqrt{6 - m_1 (m_1 + 1)}}{\sqrt{6 - m_2 (m_2 - 1)}} |(m_1 + 1) (m_2 - 1) m_3\rangle$ and $\frac{\sqrt{6 - m_1 (m_1 - 1)}}{\sqrt{6 - m_2 (m_2 + 1)}} |(m_1 - 1) (m_2 + 1) m_3\rangle$. The plus and minus sign square roots are 'switched'.

Thus together with the observation underneath the table:

$$\sqrt{6 - m_i (m_i + 1)} \sqrt{6 - m_i (m_i - 1)} = \sqrt{6 - (-m_i) (-m_i - 1)} \sqrt{6 - (-m_i) (-m_i + 1)}$$

Thus for a element in group 6, the number in front of $|(m_1 + 1) (m_2 - 1) m_3\rangle$ is the same as the number in front of $|(-m_1 - 1) (-m_2 + 1) - m_3\rangle$ in group 8. And for a element in group 6, the number in front of $|(m_1 - 1) (m_2 + 1) m_3\rangle$ is the same as the number in front of $|(-m_1 + 1) (-m_2 - 1) m_3\rangle$ in group 8. This trick with the square roots can be used with $\sqrt{6 - m_2 (m_2 + 1)} \sqrt{6 - m_3 (m_3 - 1)} |m_1 (m_2 + 1) (m_3 - 1)\rangle$ and $\sqrt{6 - m_2 (m_2 - 1)} \sqrt{6 - m_3 (m_3 + 1)} |m_1 (m_2 - 1) (m_3 + 1)\rangle$ too.

Thus for a element in group 6, the number in front of $|m_1 (m_2 + 1) (m_3 - 1)\rangle$ is the same as the number in front of $|(-m_1 - 1) (-m_2 - 1) - m_3\rangle$ in group 8. And for a element in group 6, the number in front of $|m_1 (m_2 - 1) (m_3 + 1)\rangle$ is the same as the number in front of $|(-m_1 + 1) (-m_2 + 1) m_3\rangle$. Take another look at the superpositions of group 6 and 8. See that the coefficients are the same for the 'linked' elements of group 6 and 8.

Thus the equations that follow from the Hamiltonian can indeed be written as:

$$(D - E_z) I_{18x18} \begin{pmatrix} x_1 \\ x_2 \\ \vdots \\ x_{18} \end{pmatrix} + A_6 \begin{pmatrix} x_1 \\ x_2 \\ \vdots \\ x_{18} \end{pmatrix} = E_{G_6} \begin{pmatrix} x_1 \\ x_2 \\ \vdots \\ x_{18} \end{pmatrix}$$

The eigenvalues $\lambda_{A_6}$ and the corresponding eigenvectors of matrix $A_6$ are known. The eigenvectors of $A_6$ are orthonormal, because $A_6$ is a symmetric matrix, thus satisfying the first requirement. Thus the eigenenergies are $E_{G_6} = D - E_z + \lambda_{A_6}$. And the eigenstates are the corresponding eigenvectors of $\lambda_{A_6}$. To see the exact numbers take a look at appendix A.7 how to run the code and access the exact numbers of the coefficients.

Group 9 has $m_1 + m_2 + m_3 = 2$. Define a superposition $G_9$ with the elements of group 9,

$$G_9 = x_{13} |2 2 -2\rangle + x_{14} |2 1 -1\rangle + x_{13} |2 0 0\rangle + x_{12} |2 -1 1\rangle + x_{11} |2 -2 2\rangle + x_{10} |1 2 -1\rangle + x_{9} |1 1 0\rangle + x_{8} |1 0 1\rangle + x_{7} |1 -1 2\rangle + x_{6} |0 2 0\rangle + x_{5} |0 1 1\rangle + x_{4} |0 0 2\rangle + x_{3} |-1 2 1\rangle + x_{2} |-1 1 2\rangle + x_{1} |2 2 2\rangle$$

and find the coefficients for which the superposition is an eigenstate of the Hamiltonian. Check the properties of an eigenstate and get the requirements for the coefficients. Using the inner product properties described in section 2.4 the first requirement for the coefficients follows:

$$\langle G_9 | G_9 \rangle = 1 \iff |x_1|^2 + |x_2|^2 + |x_3|^2 + |x_4|^2 + |x_5|^2 + |x_6|^2 + |x_7|^2 + |x_8|^2 + |x_9|^2 + |x_{10}|^2 + |x_{11}|^2 + |x_{12}|^2 + |x_{13}|^2 + |x_{14}|^2 + |x_{15}|^2 = 1$$

$$+ |x_{16}|^2 + |x_{17}|^2 + |x_{18}|^2 = 1$$
The superpositions of group 9 and 5 are linked in the same way as the superpositions of group 6 and 8. Thus there is no need to calculate the Hamiltonian of the superposition of group 9 and the eigenvalues and eigenstates. The equations that follow from the Hamiltonian acting on $G_9$ can be written as an eigenvalue problem.

$$(2D - 2E_z) I_{15 \times 15} \left[ \begin{array}{c} x_1 \\ \vdots \\ x_{15} \end{array} \right] + A_5 \left[ \begin{array}{c} x_1 \\ \vdots \\ x_{15} \end{array} \right] = E_{G_9} \left[ \begin{array}{c} x_1 \\ \vdots \\ x_{15} \end{array} \right]$$

The eigenvalues $\lambda_{A_5}$ and the corresponding eigenvectors of matrix $A_5$ are known. The eigenvectors of $A_5$ are orthonormal, because $A_5$ is a symmetric matrix, thus satisfying the first requirement. Thus the eigenenergies are $E_{G_9} = 2D - 2E_z + \lambda_{A_5}$. And the eigenstates are the corresponding eigenvectors of $\lambda_{A_5}$. To see the exact numbers take a look at appendix A.7 how to run the code and access the exact numbers of the coefficients.

Group 10 has $m_1 + m_2 + m_3 = 3$. Define a superposition $G_{10}$ with the elements of group 10,

$$G_{10} = x_1|2\ 2\ -1\rangle + x_5|2\ 1\ 0\rangle + x_6|2\ 0\ 1\rangle + x_7|2\ -1\ 2\rangle + x_9|1\ 2\ 0\rangle + x_5|1\ 1\ 1\rangle + x_4|1\ 0\ 2\rangle + x_3|0\ 2\ 1\rangle + x_2|0\ 1\ 2\rangle + x_1|1\ -1\ 2\rangle$$

and find the coefficients for which the superposition is an eigenstate of the Hamiltonian. Check the properties of an eigenstate and get the requirements for the coefficients. Using the inner product properties discussed in section 2.4 the first requirement for the coefficients follows:

$$\langle G_{10}|G_{10} \rangle = 1 \iff |x_1|^2 + |x_2|^2 + |x_3|^2 + |x_4|^2 + |x_5|^2 + |x_6|^2 + |x_7|^2 + |x_8|^2 + |x_9|^2 + |x_{10}|^2 = 1$$

The superpositions of group 10 and 4 are linked in the same way as the superpositions of group 6 and 8. Thus there is no need to calculate the Hamiltonian of the superposition of group 10 and the eigenvalues and eigenstates. The equations that follow from the Hamiltonian acting on $G_{10}$ can be written as an eigenvalue problem.

$$(3D - 3E_z) I_{10 \times 10} \left[ \begin{array}{c} x_1 \\ \vdots \\ x_{10} \end{array} \right] + A_4 \left[ \begin{array}{c} x_1 \\ \vdots \\ x_{10} \end{array} \right] = E_{G_{10}} \left[ \begin{array}{c} x_1 \\ \vdots \\ x_{10} \end{array} \right]$$

The eigenvalues $\lambda_{A_4}$ and the corresponding eigenvectors of matrix $A_4$ are known. The eigenvectors of $A_4$ are orthonormal, because $A_4$ is a symmetric matrix, thus satisfying the first requirement. Thus the eigenenergies are $E_{G_{10}} = 3D - 3E_z + \lambda_{A_4}$. And the eigenstates are the corresponding eigenvectors of $\lambda_{A_4}$. To see the exact numbers take a look at appendix A.7 how to run the code and access the exact numbers of the coefficients.

Group 11 has $m_1 + m_2 + m_3 = 4$. Define a superposition $G_{11}$ with the elements of group 11,

$$G_{11} = x_6|2\ 2\ 0\rangle + x_5|2\ 1\ 1\rangle + x_4|2\ 0\ 2\rangle + x_3|1\ 2\ 1\rangle + x_2|1\ 1\ 2\rangle + x_1|0\ 2\ 2\rangle$$

and find the coefficients for which the superposition is an eigenstate of the Hamiltonian. Check the properties of an eigenstate and get the requirements for the coefficients. Using the inner product properties discussed in section 2.4 the first requirement for the coefficients follows:

$$\langle G_{11}|G_{11} \rangle = 1 \iff |x_1|^2 + |x_2|^2 + |x_3|^2 + |x_4|^2 + |x_5|^2 + |x_6|^2 = 1$$

The superpositions of group 11 and 3 are linked in the same way as the superpositions of group 6 and 8. Thus there is no need to calculate the Hamiltonian of the superposition of group 11 and the eigenvalues and eigenstates. The equations that follow from the Hamiltonian acting on $G_{11}$ can be written as an eigenvalue problem.

$$(6D - 4E_z) I_{6 \times 6} \left[ \begin{array}{c} x_1 \\ \vdots \\ x_6 \end{array} \right] + A_3 \left[ \begin{array}{c} x_1 \\ \vdots \\ x_6 \end{array} \right] = E_{G_{11}} \left[ \begin{array}{c} x_1 \\ \vdots \\ x_6 \end{array} \right]$$
A.3. Find the eigenstates and eigenenergies of the two particle system with zero transverse magnetic anisotropy

The eigenvalues $\lambda_{A_i}$ and the corresponding eigenvectors of matrix $A_3$ are known. The eigenvectors of $A_3$ are orthonormal, because $A_3$ is a symmetric matrix, thus satisfying the first requirement. Thus the eigenenergies are $E_{G_{12}} = 6D - 4E_z + \lambda_{A_1}$. And the eigenstates are the corresponding eigenvectors of $\lambda_{A_1}$. To see the exact numbers take a look at appendix A.7 how to run the code and access the exact numbers of the coefficients.

Group 12 has $m_1 + m_2 + m_3 = 5$. Define a superposition $G_{12}$ with the elements of group 12,

$$G_{12} = x_3 |2 2 1\rangle + x_2 |2 1 2\rangle + x_3 |1 2 2\rangle$$

and find the coefficients for which the superposition is an eigenstate of the Hamiltonian. Check the properties of an eigenstate and get the requirements for the coefficients. Using the inner product properties discussed in section 2.4 the first requirement for the coefficients follows:

$$\langle G_{12} | G_{12} \rangle = 1 \iff |x_1|^2 + |x_2|^2 + |x_3|^2 = 1$$

The superpositions of group 12 and 2 are linked in the same way as the superpositions of group 6 and 8. Thus there is no need to calculate the Hamiltonian of the superposition of group 12 and the eigenvalues and eigenstates. The equations that follow from the Hamiltonian acting on $G_{12}$ can be written as an eigenvalue problem.

$$(9D - 5E_z + 4J) I_{3 \times 3} \begin{bmatrix} x_1 \\ x_2 \\ x_3 \end{bmatrix} + 2JA_2 \begin{bmatrix} x_1 \\ x_2 \\ x_3 \end{bmatrix} = E_{G_{12}} \begin{bmatrix} x_1 \\ x_2 \\ x_3 \end{bmatrix}$$

The eigenvalues $\lambda_{A_2}$ and the corresponding eigenvectors of matrix $A_2$ are known. The eigenvectors of $A_2$ are orthonormal, because $A_2$ is a symmetric matrix, thus satisfying the first requirement. Thus the eigenenergies are $E_{G_{12}} = 9D - 5E_z + 4J + 2J\lambda_{A_2}$. And the eigenstates are the corresponding eigenvectors of $\lambda_{A_2}$. To see the exact numbers take a look at appendix A.7 how to run the code and access the exact numbers of the coefficients.

Group 13 has $m_1 + m_2 + m_3 = 6$. Define a superposition $G_{13}$ with the elements of group 13,

$$G_{13} = |2 2 2\rangle$$

and check if it is and eigenstate. Using the inner product properties discussed in section 2.4 the first requirement is satisfied:

$$\langle G_{13} | G_{13} \rangle = 1$$

For the other requirement let the Hamiltonian act on $G_{13}$.

$$\mathcal{H}G_{13} = \mathcal{H}|2 2 2\rangle = (12D - 6E_z + 8J)|2 2 2\rangle = E_{G_{13}}|2 2 2\rangle = E_{G_{13}}G_{13}$$

Thus $|2 2 2\rangle$ is an eigenstate with eigenenergy $E_{G_{13}} = 12D - 6E_z + 8J$.

This was the last eigenstate, now all eigenstates of the 3 particle system are known. There are 125 eigenstates in total.

A.3. Find the eigenstates and eigenenergies of the two particle system with zero transverse magnetic anisotropy

A system of two particles with spin 2 will be worked out. The process will be similar to the process of a three particle system. The representation of a system consisting of two atoms is $|m_1 m_2\rangle$. Start with analysing the Hamiltonian. Let the Hamiltonian act on $|m_1 m_2\rangle$.

$$\mathcal{H}|m_1 m_2\rangle = \sum_{i=1}^2 \left(D \left(S_z^{(i)}\right)^2 + E \left(S_x^{(i)}\right)^2 - \left(S_z^{(i)}\right)^2 \right) - g^* \mu_B \mathbf{B} \cdot \mathbf{S}^{(i)} |m_1 m_2\rangle$$

$$+ \sum_{i=1}^1 J \mathbf{S}^{(i)} \cdot \mathbf{S}^{(i+1)} |m_1 m_2\rangle$$
Thus

\[ \mathcal{H}(m_1 m_2) = \sum_{i=1}^{2} \left( D \left( \hat{S}_z^i \right)^2 - g^* \mu_B B \hat{S}_z^i \right) |m_1 m_2\rangle + J \hat{S}^{(1)} \cdot \hat{S}^{(2)} |m_1 m_2\rangle. \]

From now on \( E_z = g^* \mu_B B \) will be used. From appendix A.1 it is known that

\[ \hat{S}^{(i)} \cdot \hat{S}^{(i+1)} = \frac{1}{2} \left( \hat{S}_+^{(i)} \hat{S}_-^{(i+1)} + \hat{S}_-^{(i)} \hat{S}_+^{(i+1)} \right) + \hat{S}_z^{(i)} \hat{S}_z^{(i+1)} \]

Thus

\[ \mathcal{H}(m_1 m_2) = \sum_{i=1}^{2} D \left( \hat{S}_z^i \right)^2 |m_1 m_2\rangle - E_z \hat{S}_z^i |m_1 m_2\rangle + \frac{J}{2} \left( \hat{S}_+^{(1)} \hat{S}_-^{(2)} + \hat{S}_-^{(1)} \hat{S}_+^{(2)} \right) + J \hat{S}_z^{(1)} \hat{S}_z^{(2)} |m_1 m_2\rangle. \]

The step next is sorting as well as down all the elements of the sum.

\[ \mathcal{H}(m_1 m_2) = D \left( \hat{S}_z^1 \right)^2 |m_1 m_2\rangle - E_z \hat{S}_z^1 |m_1 m_2\rangle + D \left( \hat{S}_z^2 \right)^2 |m_1 m_2\rangle - E_z \hat{S}_z^2 |m_1 m_2\rangle \\
+ \frac{J}{2} \hat{S}_+^{(1)} \hat{S}_-^{(2)} |m_1 m_2\rangle + \frac{J}{2} \hat{S}_-^{(1)} \hat{S}_+^{(2)} |m_1 m_2\rangle + J \hat{S}_z^{(1)} \hat{S}_z^{(2)} |m_1 m_2\rangle. \]

Now let the spin operators act on \( |m_1 m_2\rangle \), remembering the total spin of every atom is 2, so \( s = 2 \), thus \( s(s+1) = 6 \).

\[ \mathcal{H}(m_1 m_2) = D \hat{S}_z^1 |m_1 m_2\rangle - E_z m_1 m_2 |m_1 m_2\rangle + D \hat{S}_z^2 |m_1 m_2\rangle - E_z m_2 |m_1 m_2\rangle \\
+ \frac{J}{2} \hat{S}_+^{(1)} \hat{S}_-^{(2)} |m_1 m_2\rangle + \frac{J}{2} \hat{S}_-^{(1)} \hat{S}_+^{(2)} |m_1 m_2\rangle + J \hat{S}_z^{(1)} \hat{S}_z^{(2)} |m_1 m_2\rangle. \]

The final step is sorting as well as element by element.

\[ \mathcal{H}(m_1 m_2) = \left( (m_1^2 + m_2^2) D - (m_1 + m_2) E_z + m_1 m_2 J \right) |m_1 m_2\rangle \\
+ \frac{J}{2} \sqrt{6 - m_1 (m_1 + 1)} \sqrt{6 - m_2 (m_2 - 1)} |m_1 m_2\rangle (m_1 + 1) (m_2 - 1) \]

There are 9 groups with the same \( m_1 + m_2 \), because \( m_1 + m_2 \) ranges from -4 to 4. Now determine the eigenstates and eigenenergies. Remember \( M \) is an eigenstate of the Hamiltonian if \( \mathcal{H} M = E_M M \) and \( \langle M | M \rangle = 1 \) hold, with \( E_M \) the eigenenergy of eigenstate \( M \).

Group 1 has \( m_1 + m_2 = -4 \). Define a superposition \( G_1 \) with the elements of group 1,

\[ G_1 = \{|-2 -2\rangle\} \]

and check if it is an eigenstate. Using the inner product properties discussed in section 2.4 the first requirement is satisfied:

\[ \langle G_1 | G_1 \rangle = 1 \]
A.3. Find the eigenstates and eigenenergies of the two particle system with zero transverse magnetic anisotropy

For the other requirement let the Hamiltonian act on $G_1$.

$$\mathcal{H} G_1 = \mathcal{H} | -2 \rangle = (8D + 4E_z + 4J) | -2 \rangle = E_{G_1} | -2 \rangle = E_{G_1} G_1$$

Thus $| -2 \rangle$ is an eigenstate with eigenenergy $E_{G_1} = 8D + 4E_z + 4J$.

Group 2 has $m_1 + m_2 + m_3 = -3$. Define a superposition $G_2$ with the elements of group 2,

$$G_2 = a | -1 \rangle - 2 b | -2 \rangle - 1$$

and find the coefficients for which the superposition is an eigenstate of the Hamiltonian. Check the properties of an eigenstate and get the requirements for the coefficients. Using the inner product properties discussed in section 2.4 the first requirement for the coefficients follows:

$$\langle G_2 | G_2 \rangle = 1 \iff |a|^2 + |b|^2 = 1$$

For the other requirements let the Hamiltonian act on $G_2$.

$$\mathcal{H} G_2 = (5D + 3E_z + 2J) a | -2 \rangle + a | -1 \rangle - 2 b | -2 \rangle - 1$$

This can be written as an eigenvalue problem.

$$(5D + 3E_z + 2J) I_{2 \times 2} \begin{bmatrix} a \\ b \end{bmatrix} + 2J A_2 \begin{bmatrix} a \\ b \end{bmatrix} = E_{G_2} \begin{bmatrix} a \\ b \end{bmatrix}$$

With

$$A_2 = \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}$$

Determine the eigenvalues $\lambda_{A_2}$ and corresponding eigenvectors of matrix $A_2$ with the matlab code, see appendix A.6. Matlab finds two eigenvalues and two orthonormal eigenvectors, because $A_2$ is a symmetric matrix, thus satisfying the first requirement. Then the eigenenergies are $E_{G_2} = 5D + 3E_z + 2J + 2J \lambda_{A_2}$. And the eigenstates are the corresponding eigenvectors of $\lambda_{A_2}$. To see the exact numbers take a look at appendix A.6 how to run the code and access the exact numbers of the coefficients.

Group 3 has $m_1 + m_2 + m_3 = -2$. Define a superposition $G_3$ with the elements of group 3,

$$G_3 = a | 0 \rangle - 2 b | -1 \rangle - 1 c | -2 \rangle$$

and find the coefficients for which the superposition is an eigenstate of the Hamiltonian. Check the properties of an eigenstate and get the requirements for the coefficients. Using the inner product properties discussed in section 2.4 the first requirement for the coefficients follows:

$$\langle G_3 | G_3 \rangle = 1 \iff |a|^2 + |b|^2 + |c|^2 = 1$$

For the other requirements let the Hamiltonian act on $G_3$.

$$(2D + 2E_z) I_{3 \times 3} \begin{bmatrix} a \\ b \\ c \end{bmatrix} + A_3 \begin{bmatrix} a \\ b \\ c \end{bmatrix} = E_{G_3} \begin{bmatrix} a \\ b \\ c \end{bmatrix}$$
Determine the eigenvalues $\lambda_{A_3}$ and corresponding eigenvectors of matrix $A_3$ with the matlab code, see appendix A.6. Matlab finds three eigenvalues and three orthonormal eigenvectors, because $A_3$ is a symmetric matrix, thus satisfying the first requirement. Then the eigenergies are $E_{G_3} = 2D + 2E_z + \lambda_{A_3}$. And the eigenstates are the corresponding eigenvectors of $\lambda_{A_3}$. To see the exact numbers take a look at appendix A.6 how to run the code and access the exact numbers of the coefficients.

Group 4 has $m_1 + m_2 + m_3 = -1$. Define a superposition $G_4$ with the elements of group 4,

$$G_4 = a|1 - 2\rangle + b|0 - 1\rangle + c|1 - 0\rangle + d|2 - 2\rangle$$

and find the coefficients for which the superposition is an eigenstate of the Hamiltonian. Check the properties of an eigenstate and get the requirements for the coefficients. Using the inner product properties discussed in section 2.4 the first requirement for the coefficients follows:

$$\langle G_4 | G_4 \rangle = 1 \iff |a|^2 + |b|^2 + |c|^2 + |d|^2 = 1$$

For the other requirements let the Hamiltonian act on $G_4$,

$$\mathcal{H}G_4 = \left( (5D + E_z - 2J) a + \sqrt{6} jb \right)|1 - 2\rangle + \left( (D + E_z) b + \sqrt{6} ja + 3Jc \right)|0 - 1\rangle$$

$$+ \left( (D + E_z) c + 3Jb + \sqrt{6} jd \right)|1 - 0\rangle + \left( (5D + E_z - 2J) d + \sqrt{6} jc \right)|2 - 1\rangle$$

$$= E_{G_4} a|1 - 2\rangle + E_{G_4} b|0 - 1\rangle + E_{G_4} c|1 - 0\rangle + E_{G_4} d|2 - 1\rangle$$

This can be written as an eigenvalue problem.

$$(D + E_z) I_{4x4} \begin{bmatrix} a \\ \vdots \\ d \end{bmatrix} + A_4 \begin{bmatrix} a \\ \vdots \\ d \end{bmatrix} = E_{G_4} \begin{bmatrix} a \\ \vdots \\ d \end{bmatrix}$$

With

$$A_4 = \begin{bmatrix} 4D - 2J & \sqrt{6} J & 0 & 0 \\ \sqrt{6} J & 0 & 3J & 0 \\ 0 & 3J & 0 & \sqrt{6} J \\ 0 & 0 & \sqrt{6} J & 4D - 2J \end{bmatrix}$$

Determine the eigenvalues $\lambda_{A_4}$ and corresponding eigenvectors of matrix $A_4$ with the matlab code, see appendix A.6. Matlab finds four eigenvalues and four orthonormal eigenvectors, because $A_4$ is a symmetric matrix, thus satisfying the first requirement. Then the eigenergies are $E_{G_4} = D + E_z + \lambda_{A_4}$. And the eigenstates are the corresponding eigenvectors of $\lambda_{A_4}$. To see the exact numbers take a look at appendix A.6 how to run the code and access the exact numbers of the coefficients.

Group 5 has $m_1 + m_2 + m_3 = 0$. Define a superposition $G_5$ with the elements of group 5,

$$G_5 = a|2 - 2\rangle + b|1 - 1\rangle + c|0 0\rangle + d|1 - 1\rangle + e|- 2 2\rangle$$

and find the coefficients for which the superposition is an eigenstate of the Hamiltonian. Check the properties of an eigenstate and get the requirements for the coefficients. Using the inner product properties discussed in section 2.4 the first requirement for the coefficients follows:

$$\langle G_5 | G_5 \rangle = 1 \iff |a|^2 + |b|^2 + |c|^2 + |d|^2 + |e|^2 = 1$$

For the other requirements let the Hamiltonian act on $G_5$.

$$\mathcal{H}G_5 = \left( (8D - 4J) a + 2J b \right)|2 - 2\rangle + \left( (2D - J) b + 2Ja + 3Jc \right)|1 - 1\rangle + \left( 3Jb + 3Jd \right)|0 0\rangle$$

$$+ \left( (2D - J) d + 3Jc + 2Je \right)|1 - 1\rangle + \left( (8D - 4J) e + 2Jd \right)|2 - 2\rangle$$

$$= E_{G_5} a|2 - 2\rangle + E_{G_5} b|1 - 1\rangle + E_{G_5} c|0 0\rangle + E_{G_5} d|1 - 1\rangle + E_{G_5} e|- 2 2\rangle$$

With
Determine the eigenvalues $E$ and orthonormal eigenvectors of matrix $A_5$ with the matlab code, see appendix A.6. Matlab finds five eigenvalues and five orthonormal eigenvectors, because in section 2.4 the first requirement for the coefficients follows:

This can be written as an eigenvalue problem.

$$A_5 \begin{bmatrix} a \\ \vdots \\ e \end{bmatrix} = E G_6 \begin{bmatrix} a \\ \vdots \\ e \end{bmatrix}$$

With

$$A_5 = \begin{bmatrix} 8D - 4J & 2J & 0 & 0 & 0 \\ 2J & 2D - J & 3J & 0 & 0 \\ 0 & 3J & 0 & 3J & 0 \\ 0 & 0 & 3J & 2D - J & 2J \\ 0 & 0 & 0 & 2J & 8D - 4J \end{bmatrix}$$

Determine the eigenvalues $\lambda_{A_5}$ and corresponding eigenvectors of matrix $A_5$ with the matlab code, see appendix A.6. Matlab finds five eigenvalues and five orthonormal eigenvectors, because $A_5$ is a symmetric matrix, thus satisfying the first requirement. Then the eigenenergies are $E G_6 = \lambda_{A_5}$. And the eigenstates are the corresponding eigenvectors of $\lambda_{A_5}$. To see the exact numbers take a look at appendix A.6 how to run the code and access the exact numbers of the coefficients.

Group 6 has $m_1 + m_2 + m_3 = 1$. Define a superposition $G_6$ with the elements of group 6,

$$G_6 = a|1\ 2\rangle + b|0\ 1\rangle + c|1\ 0\rangle + d|2\ 1\rangle$$

and find the coefficients for which the superposition is an eigenstate of the Hamiltonian. Check the properties of an eigenstate and get the requirements for the coefficients. Using the inner product properties discussed in section 2.4 the first requirement for the coefficients follows:

$$\langle G_6|G_6 \rangle = 1 \iff |a|^2 + |b|^2 + |c|^2 + |d|^2 = 1$$

For the other requirements let the Hamiltonian act on $G_6$.

$$\mathcal{H} G_6 = (5D - E_z - 2J) a + \sqrt{6} b |0\ 1\rangle - \langle 1\ 0| + \left( (D - E_z) b + \sqrt{6} a + 3J c \right) |0\ 1\rangle + \left( (D - E_z) c + 3J b + \sqrt{6} d \right) |1\ 0\rangle + \left( (5D - E_z - 2J) d + \sqrt{6} c \right) |2\ 1\rangle$$

This can be written as an eigenvalue problem.

$$\begin{bmatrix} a \\ \vdots \\ d \end{bmatrix} + A_6 \begin{bmatrix} a \\ \vdots \\ d \end{bmatrix} = E G_6 \begin{bmatrix} a \\ \vdots \\ d \end{bmatrix}$$

With

$$A_6 = \begin{bmatrix} 4D - 2J & \sqrt{6} J & 0 & 0 \\ \sqrt{6} J & 0 & 3J & 0 \\ 0 & 3J & 0 & \sqrt{6} J \\ 0 & 0 & \sqrt{6} J & 4D - 2J \end{bmatrix}$$

The eigenvalues $\lambda_{A_6}$ and corresponding eigenvectors of matrix $A_4$ are known. The eigenvectors of $A_4$ are orthonormal, because $A_4$ is a symmetric matrix, thus satisfying the first requirement. Then the eigenenergies are $E G_6 = \lambda_{A_5}$. And the eigenstates are the corresponding eigenvectors of $\lambda_{A_5}$. To see the exact numbers take a look at appendix A.6 how to run the code and access the exact numbers of the coefficients.

Group 7 has $m_1 + m_2 + m_3 = 2$. Define a superposition $G_7$ with the elements of group 7,

$$G_7 = a|0\ 2\rangle + b|1\ 1\rangle + c|2\ 0\rangle$$
and find the coefficients for which the superposition is an eigenstate of the Hamiltonian. Check the properties of an eigenstate and get the requirements for the coefficients. Using the inner product properties discussed in section 2.4 the first requirement for the coefficients follows:

\[(G_7|G_7) = 1 \iff |a|^2 + |b|^2 + |c|^2 = 1\]

For the other requirements let the Hamiltonian act on \(G_7\).

\[\mathcal{H}G_7 = \left[ (4D - 2E_z) a + \sqrt{6} J b |0\ 2) + \left( (2D - 2E_z + J) b + \sqrt{6} J a \right) |1\ 1) \right.\]
\[+ \left( (4D - 2E_z) c + \sqrt{6} J b |2\ 0)\right.\]
\[= E_{G_7} a|0\ 2) + E_{G_7} b|1\ 1) + E_{G_7} c|2\ 0)\]

This can be written as an eigenvalue problem.

\[(2D - 2E_z) I_{3\times3} \begin{bmatrix} a \\ b \\ c \end{bmatrix} + A_7 \begin{bmatrix} a \\ b \\ c \end{bmatrix} = E_{G_7} \begin{bmatrix} a \\ b \\ c \end{bmatrix}\]

With

\[A_7 = \begin{bmatrix} 2D & \sqrt{6} J & 0 \\ \sqrt{6} J & J & \sqrt{6} J \\ 0 & \sqrt{6} J & 2D \end{bmatrix} = A_3\]

The eigenvalues \(\lambda_{A_3}\) and corresponding eigenvectors of matrix \(A_3\) are known. The eigenvectors of \(A_3\) are orthonormal, because \(A_3\) is a symmetric matrix, thus satisfying the first requirement. Then the eigenenergies are \(E_{G_7} = 2D - 2E_z + \lambda_{A_3}\). And the eigenstates are the corresponding eigenvectors of \(\lambda_{A_3}\). To see the exact numbers take a look at appendix A.6 how to run the code and access the exact numbers of the coefficients.

Group 8 has \(m_1 + m_2 + m_3 = 3\). Define a superposition \(G_8\) with the elements of group 8,

\[G_8 = a|1\ 2) + b|2\ 1)\]

and find the coefficients for which the superposition is an eigenstate of the Hamiltonian. Check the properties of an eigenstate and get the requirements for the coefficients. Using the inner product properties discussed in section 2.4 the first requirement for the coefficients follows:

\[(G_8|G_8) = 1 \iff |a|^2 + |b|^2 = 1\]

For the other requirements let the Hamiltonian act on \(G_8\).

\[\mathcal{H}G_8 = \left[ (5D - 3E_z + 2J) a + 2J b |1\ 2) + \left( (5D - 3E_z + 2J) b + 2J a \right) |2\ 1) \right.\]
\[= E_{G_8} a|1\ 2) + E_{G_8} b|2\ 1)\]

This can be written as an eigenvalue problem.

\[(5D - 3E_z + 2J) I_{2\times2} \begin{bmatrix} a \\ b \end{bmatrix} + 2J A_8 \begin{bmatrix} a \\ b \end{bmatrix} = E_{G_8} \begin{bmatrix} a \\ b \end{bmatrix}\]

With

\[A_8 = \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} = A_2\]

The eigenvalues \(\lambda_{A_2}\) and corresponding eigenvectors of matrix \(A_2\) are known. The eigenvectors of \(A_2\) are orthonormal, because \(A_2\) is a symmetric matrix, thus satisfying the first requirement. Then the eigenenergies are \(E_{G_8} = 5D - 3E_z + 2J + 2J \lambda_{A_2}\). And the eigenstates are the corresponding eigenvectors of \(\lambda_{A_2}\). To see the exact numbers take a look at appendix A.6 how to run the code and access the exact numbers of the coefficients.
A.4. The transverse magnetic anisotropy term acting on an element of the three particle system

Group 9 has $m_1 + m_2 = 4$. Define a superposition $G_9$ with the elements of group 9,

$$G_9 = |2 2\rangle$$

and check if it is an eigenstate. Using the inner product properties discussed in section 2.4 the first requirement is satisfied:

$$\langle G_9 | G_9 \rangle = 1$$

For the other requirement let the Hamiltonian act on $G_9$.

$$\mathcal{H}G_9 = \mathcal{H}|2 2\rangle = (8D - 4E_x + 4J)|2 2\rangle = E_{G_9}|2 2\rangle = E_{G_9}G_9$$

Thus $|2 2\rangle$ is an eigenstate with eigenenergy $E_{G_9} = 8D - 4E_x + 4J$.

This was the last eigenstate, now all eigenstates of the 2 particle system are known. There are 25 eigenstates in total.

A.4. The transverse magnetic anisotropy term acting on an element of the three particle system

First the magnetic anisotropy term has to be transformed, because $\hat{S}_x$ and $\hat{S}_y$ act on x-basis elements and y-basis elements respectively. Use that $\hat{S}_x = \frac{1}{2}(\hat{S}_+ + \hat{S}_-)$ and $\hat{S}_y = \frac{1}{2i}(\hat{S}_+ - \hat{S}_-)$, with $i$ the imaginary number with $i^2 = -1$, not the index of the sum. Then:

$$E \left( \left( \hat{S}_x^{(i)} \right)^2 - \left( \hat{S}_y^{(i)} \right)^2 \right) = E \left( \hat{S}_x^{(i)} \hat{S}_x^{(i)} - \hat{S}_y^{(i)} \hat{S}_y^{(i)} \right)$$

$$= E \frac{1}{4} \left( \hat{S}^{(i)}_+ + \hat{S}^{(i)}_- \right) \left( \hat{S}^{(i)}_+ + \hat{S}^{(i)}_- \right) - E \frac{1}{4} \left( \hat{S}^{(i)}_+ - \hat{S}^{(i)}_- \right) \left( \hat{S}^{(i)}_+ - \hat{S}^{(i)}_- \right)$$

$$= E \frac{1}{4} \left( \hat{S}^{(i)}_+ \hat{S}^{(i)}_+ + \hat{S}^{(i)}_- \hat{S}^{(i)}_- + \hat{S}^{(i)}_+ \hat{S}^{(i)}_- + \hat{S}^{(i)}_- \hat{S}^{(i)}_+ \right) - E \frac{1}{4} \left( \hat{S}^{(i)}_+ \hat{S}^{(i)}_- + \hat{S}^{(i)}_- \hat{S}^{(i)}_+ \right)$$

Let the transverse magnetic anisotropy term of the Hamiltonian act on an element of the three particle system and start with writing out the sum:

$$\sum_{i=1}^{3} \frac{E}{2} \left( \hat{S}^{(i)}_+ \hat{S}^{(i)}_+ + \hat{S}^{(i)}_- \hat{S}^{(i)}_- \right) |m_1 m_2 m_3\rangle = \frac{E}{2} \left( \hat{S}^{(1)}_+ \hat{S}^{(1)}_+ + \hat{S}^{(1)}_- \hat{S}^{(1)}_- \right) |m_1 m_2 m_3\rangle$$

$$+ \frac{E}{2} \left( \hat{S}^{(2)}_+ \hat{S}^{(2)}_+ + \hat{S}^{(2)}_- \hat{S}^{(2)}_- \right) |m_1 m_2 m_3\rangle$$

$$+ \frac{E}{2} \left( \hat{S}^{(3)}_+ \hat{S}^{(3)}_+ + \hat{S}^{(3)}_- \hat{S}^{(3)}_- \right) |m_1 m_2 m_3\rangle$$
Now let the $\hat{S}_+$ and $\hat{S}_-$ operators act on $|m_1 \ m_2 \ m_3\rangle$.

$$\sum_{i=1}^{3} \frac{E}{2} \left( S_+^{(i)} S_+^{(i)} + S_-^{(i)} S_-^{(i)} \right) |m_1 \ m_2 \ m_3\rangle = \frac{E}{2} \left( S_+^{(i)} \sqrt{6-m_1 (m_1+1)} (m_1+1) \ m_2 \ m_3\rangle 
+ \frac{E}{2} \left( S_-^{(i)} \sqrt{6-m_1 (m_1-1)} \ m_2 \ m_3\rangle 
+ \frac{E}{2} \left( S_+^{(i)} \sqrt{6-m_2 (m_2+1)} \ m_1 \ m_2 \ m_3\rangle 
+ \frac{E}{2} \left( S_-^{(i)} \sqrt{6-m_2 (m_2-1)} \ m_1 \ m_2 \ m_3\rangle 
+ \frac{E}{2} \left( S_+^{(i)} \sqrt{6-m_3 (m_3+1)} \ m_1 \ m_2 \ m_3\rangle 
+ \frac{E}{2} \left( S_-^{(i)} \sqrt{6-m_3 (m_3-1)} \ m_1 \ m_2 \ m_3\rangle
= \frac{E}{2} \sqrt{6-(m_1+1)(m_1+2)} \sqrt{6-m_1 (m_1+1)} \ m_2 \ m_3\rangle 
+ \frac{E}{2} \sqrt{6-(m_1-1)(m_1-2)} \sqrt{6-m_1 (m_1-1)} \ m_2 \ m_3\rangle 
+ \frac{E}{2} \sqrt{6-(m_2+1)(m_2+2)} \sqrt{6-m_2 (m_2+1)} \ m_1 \ m_2 \ m_3\rangle 
+ \frac{E}{2} \sqrt{6-(m_2-1)(m_2-2)} \sqrt{6-m_2 (m_2-1)} \ m_1 \ m_2 \ m_3\rangle 
+ \frac{E}{2} \sqrt{6-(m_3+1)(m_3+2)} \sqrt{6-m_3 (m_3+1)} \ m_1 \ m_2 \ m_3\rangle 
+ \frac{E}{2} \sqrt{6-(m_3-1)(m_3-2)} \sqrt{6-m_3 (m_3-1)} \ m_1 \ m_2 \ m_3\rangle

\textbf{A.5. Derivation of formula 3.6}

Start with formula 2.3 and write out the sum over $\alpha$.

$$I(V) = G_S \sum_{M,M',\alpha=x,y,z} P_M |\langle M | \hat{S}_\alpha^{(j)} | M' \rangle|^2 F_{M,M',s}(V)$$

$$= G_S \sum_{M,M'} P_M |\langle M | \hat{S}_x^{(j)} | M' \rangle|^2 F_{M,M',s}(V)$$

$$+ G_S \sum_{M,M'} P_M |\langle M | \hat{S}_y^{(j)} | M' \rangle|^2 F_{M,M',s}(V)$$

$$+ G_S \sum_{M,M'} P_M |\langle M | \hat{S}_z^{(j)} | M' \rangle|^2 F_{M,M',s}(V)$$

Now use that $\hat{S}_x = \frac{1}{2} (\hat{S}_+ + \hat{S}_-)$ and $\hat{S}_y = \frac{i}{2} (\hat{S}_+ - \hat{S}_-)$, with $i$ the imaginary number with $i^2 = -1$. Thus:

$$I(V) = G_S \sum_{M,M'} \frac{P_M}{4} |\langle M | \hat{S}_x^{(j)} + \hat{S}_y^{(j)} | M' \rangle|^2 F_{M,M',s}(V)$$

$$+ G_S \sum_{M,M'} \frac{P_M}{4} |\langle M | \hat{S}_x^{(j)} - \hat{S}_y^{(j)} | M' \rangle|^2 F_{M,M',s}(V)$$

$$+ G_S \sum_{M,M'} P_M |\langle M | \hat{S}_z^{(j)} | M' \rangle|^2 F_{M,M',s}(V).$$
Because $\hat{S}_+$ and $\hat{S}_-$ are operators $\langle M | \hat{S}_+^{(j)} + \hat{S}_-^{(j)} | M' \rangle = \langle M | \hat{S}_+^{(j)} | M' \rangle + \langle M | \hat{S}_-^{(j)} | M' \rangle$ and $\langle M | \hat{S}_+^{(j)} - \hat{S}_-^{(j)} | M' \rangle = \langle M | \hat{S}_+^{(j)} | M' \rangle - \langle M | \hat{S}_-^{(j)} | M' \rangle$, thus:

$$I(V) = G_S \sum_{M,M'} \frac{P_M}{4} \left( \langle M | \hat{S}_+^{(j)} | M' \rangle + \langle M | \hat{S}_-^{(j)} | M' \rangle \right)^2 F_{M,M',s}(V)$$

$$+ G_S \sum_{M,M'} \frac{P_M}{4} \left( \langle M | \hat{S}_+^{(j)} | M' \rangle - \langle M | \hat{S}_-^{(j)} | M' \rangle \right)^2 F_{M,M',s}(V)$$

$$+ G_S \sum_{M,M'} P_M \left( \langle M | \hat{S}_+^{(j)} | M' \rangle \right)^2 F_{M,M',s}(V).$$

If all the eigenstates $M$ have real coefficients, then $\langle M | \hat{S}_+^{(j)} | M' \rangle$, $\langle M | \hat{S}_-^{(j)} | M' \rangle$ and $\langle M | \hat{S}_z^{(j)} | M' \rangle$ are real. Because the spin operators multiply the eigenstate $M'$ by a real number and shift it and the inner product of eigenstate $M$ and shifted eigenstate $M'$ is real too, because the coefficients of the eigenstates are real. Now the absolute values can be simplified to:

$$I(V) = G_S \sum_{M,M'} P_M \left( \langle M | \hat{S}_+^{(j)} | M' \rangle \right)^2 F_{M,M',s}(V)$$

$$+ G_S \sum_{M,M'} \frac{P_M}{2} \left( \langle M | \hat{S}_+^{(j)} | M' \rangle \langle M | \hat{S}_-^{(j)} | M' \rangle \right) F_{M,M',s}(V)$$

$$+ G_S \sum_{M,M'} \frac{P_M}{4} \left( \langle M | \hat{S}_-^{(j)} | M' \rangle \right)^2 F_{M,M',s}(V)$$

$$+ G_S \sum_{M,M'} \frac{P_M}{4} \left( \langle M | \hat{S}_+^{(j)} | M' \rangle \right)^2 F_{M,M',s}(V)$$

$$- G_S \sum_{M,M'} \frac{P_M}{2} \left( \langle M | \hat{S}_+^{(j)} | M' \rangle \langle M | \hat{S}_-^{(j)} | M' \rangle \right) F_{M,M',s}(V)$$

$$+ G_S \sum_{M,M'} \frac{P_M}{4} \left( \langle M | \hat{S}_-^{(j)} | M' \rangle \right)^2 F_{M,M',s}(V)$$

$$+ G_S \sum_{M,M'} \frac{P_M}{2} \left( \langle M | \hat{S}_-^{(j)} | M' \rangle \right)^2 F_{M,M',s}(V)$$

$$= G_S \sum_{M,M'} \frac{P_M}{2} \left( \langle M | \hat{S}_+^{(j)} | M' \rangle \right)^2 F_{M,M',s}(V)$$

$$+ G_S \sum_{M,M'} \frac{P_M}{2} \left( \langle M | \hat{S}_-^{(j)} | M' \rangle \right)^2 F_{M,M',s}(V)$$

$$+ G_S \sum_{M,M'} \frac{P_M}{2} \left( \langle M | \hat{S}_z^{(j)} | M' \rangle \right)^2 F_{M,M',s}(V)$$

### A.6. Code for eigenstates of two particle system with zero transverse magnetic anisotropy

The code is listed below. The eigenstates are stored in a cell called 'Staten' and the eigenenergies are stored in a vector called 'Energie_check'. Staten[i,1] is eigenstate i, with corresponding eigenenergy 'Energie_check(i,1)'. Staten[i,1] is a seven by seven matrix with matrix element $(x, y)$ representing $|\langle x-4, y-4 \rangle|$ and the number of matrix element $(x, y)$ is the coefficient of $|\langle x-4, y-4 \rangle|$. 

$$\mathbf{Staten[i,1]}$$
%close all
clear
clc

% Constanten
D = -1.29*(10^(-3)); %-1.29 meV, uit projectomschrijving
Ez = 0.035*(10^(-3)); %0.035 meV, uit projectomschrijving
J = -0.73*(10^(-3)); %-0.73 meV, uit projectomschrijving

% We maken een cel met alle eigenstaten
Staten = cell([25,1]); % 25, want er zijn 25 deelstaten en 25 eigenwaarden, dus 25 eigenenergien
Ham_Staten = cell([25,1]); % De hamiltoniaan toegepast op de niet genormaliseerde eigenstaten
Eigen_energie = cell([25,1]); % De eigenenergie
Energie_check = zeros(25,1); % Check of de eigenergie klopt en dus check of het de goede eigenwaarden zijn.
for n = 1:25
    Staten{n,1} = zeros(7,7); % 7 bij 7, omdat we van -3 tot 3 moeten voor als we hamiltoniaan gaan doen, de rand is wordt dan wel nul, want die staten kunnen niet.
    Ham_Staten{n,1} = zeros(7,7);
    Eigen_energie{n,1} = zeros(7,7);
end

% Normeren van de eigenstaten
% Staat 1 heeft maar 1 element dus is al genormeerd.
for i = 1:1
    Staten{1,1}(2,2) = 1;
    Energie_check(1,1) = 8*D + 4*Ez + 4*J;
end % Energie klopt (1)

% Staat 2
for i = 1:1
    A2 = zeros(2,2);
    A2(1,2) = 1;
    A2(2,1) = 1;
    [EV2, labda2] = eig(A2);
    for n = 1:2
        Staten{((1+n),1)(3,2) = EV2(1,n);
        Staten{((1+n),1)(2,3) = EV2(2,n);
        Energie_check((1+n),1) = 5*D + 3*Ez + 2*J + 2*J*labda2(n,n);
    end
end % Energie klopt (2 t/m 3)

% Staat 3
for i = 1:1
    A3 = zeros(3,3);
    A3(1,1) = 2*D;
    A3(1,2) = sqrt(6)*J;
    A3(2,1) = sqrt(6)*J;
    A3(2,2) = J;
    A3(2,3) = sqrt(6)*J;
    A3(3,2) = sqrt(6)*J;
end
A3(3,3) = 2*D;

[EV3, lambda3] = eig(A3);

for n = 1:3
    Staten((3+n),1)(4,2) = EV3(1,n);  
    Staten((3+n),1)(3,3) = EV3(2,n);  
    Staten((3+n),1)(2,4) = EV3(3,n);  
    Energie_check((3+n),1) = 2*D + 2*Ez + lambda3(n,n);
end
end % Energie klopt (4 t/m 6)

% Staat 4
for i = 1:1
    A4 = zeros(4,4);
    A4(1,1) = 4*D - 2*J;
    A4(1,2) = sqrt(6)*J;  
    A4(2,1) = sqrt(6)*J;
    A4(2,3) = 3*J;
    A4(3,2) = 3*J;
    A4(3,4) = sqrt(6)*J;
    A4(4,3) = sqrt(6)*J;
    A4(4,4) = 4*D - 2*J;

    [EV4, lambda4] = eig(A4);

for n = 1:4
    Staten((6+n),1)(5,2) = EV4(1,n);
    Staten((6+n),1)(4,3) = EV4(2,n);
    Staten((6+n),1)(3,4) = EV4(3,n);
    Staten((6+n),1)(2,5) = EV4(4,n);
    Energie_check((6+n),1) = D + Ez + lambda4(n,n);
end
end % Energie klopt (7 t/m 10)

% Staat 5
for i = 1:1
    A5 = zeros(5,5);
    A5(1,1) = 8*D - 4*J;
    A5(1,2) = 2*J;
    A5(2,1) = 2*J;
    A5(2,2) = 2*D - J;
    A5(2,3) = 3*J;
    A5(3,2) = 3*J;
    A5(3,3) = 3*J;
    A5(3,4) = 3*J;
    A5(4,3) = 3*J;
    A5(4,4) = 2*D - J;
    A5(4,5) = 2*J;
    A5(5,4) = 2*J;
    A5(5,5) = 8*D - 4*J;

    [EV5, lambda5] = eig(A5);

for n = 1:5
    Staten((10+n),1)(6,2) = EV5(1,n);
    Staten((10+n),1)(5,3) = EV5(2,n);

Staten\(((10+n),1),(4,4)\) = EV5\((3,n)\);
Staten\(((10+n),1),(3,5)\) = EV5\((4,n)\);
Staten\(((10+n),1),(2,6)\) = EV5\((5,n)\);
Energie_check\(((10+n),1)\) = \(\lambda_5(n,n)\);

end
end % Energie klopt (11 t/m 15)

% Staat 6
for \(n =1:4\)
  Staten\(((15+n),1),(6,3)\) = EV4\((1,n)\);
  Staten\(((15+n),1),(5,4)\) = EV4\((2,n)\);
  Staten\(((15+n),1),(4,5)\) = EV4\((3,n)\);
  Staten\(((15+n),1),(3,6)\) = EV4\((4,n)\);
  Energie_check\(((15+n),1)\) = \(D - Ez + \lambda_4(n,n)\);
end % Energie klopt (16 t/m 19)

% Staat 7
for \(n =1:3\)
  Staten\(((19+n),1),(6,4)\) = EV3\((1,n)\);
  Staten\(((19+n),1),(5,5)\) = EV3\((2,n)\);
  Staten\(((19+n),1),(4,6)\) = EV3\((3,n)\);
  Energie_check\(((19+n),1)\) = \(2*D - 2*Ez + \lambda_3(n,n)\);
end % Energie klopt (20 t/m 22)

% Staat 8
for \(n =1:2\)
  Staten\(((22+n),1),(6,5)\) = EV2\((1,n)\);
  Staten\(((22+n),1),(5,6)\) = EV2\((2,n)\);
  Energie_check\(((22+n),1)\) = \(5*D - 3*Ez + 2*J + 2*J*\lambda_2(n,n)\);
end % Energie klopt (23 t/m 24)

% Staat 9 heeft maar 1 element dus is al genormeerd.
for \(i = 1:1\)
  Staten\((25,1),(6,6)\) = 1;
  Energie_check\((25,1)\) = \(8*D - 4*Ez + 4*J\);
end % Energie klopt (25)

% Doe nu de hamiltoniaan op de eigenstaten en bereken de eigenenergien
for \(n = 1:25\)
  for \(i = 2:6\)
    for \(j = 2:6\)
      Ham_Staten\((n,1),(i,j)\) = \(((i-4)^2 + (j-4)^2)*D - (i-4 + j-4)*Ez + (((i-4)\times(j-4))*J)*Staten\((n,1),(i,j)\) + (J/2) * sqrt\((6 - (i-4)\times(i-4) + (j-4)\times(j-4))*Staten\((n,1),(i+1,j-1)\) + (J/2) * sqrt\((6 - (i-4)\times(i-4) + (j-4)\times(j-4))*Staten\((n,1),(i-1,j+1)\);  
      if Staten\((n,1),(i,j)\) == 0
        else
          Eigen_energie\((n,1),(i,j)\) = \((1.602\times10^{-19})\times Ham_Staten\((n,1),(i,j)\) / 
                         Staten\((n,1),(i,j)\); % zodat het in joule staat
        end
      end
    end
  end
end

% Let op, als er een coefficient is met kans 0 in staat \(M_i\), dan is de eigen energie
0 in plaats van \(E_i\), let hier op als je de energieën gebruikt!
A.7. Code for eigenstates of the three particle system with zero transverse magnetic anisotropy

The code is listed below. The eigenstates are stored in a cell called 'Staten' and the eigenenergies are stored in a vector called 'Energie_check'. Staten{i,1} is eigenstate i, with corresponding eigenenergy 'Energie_check(i,1)'. Staten{i,1} is a seven by seven by seven matrix with matrix element $(x, y, z)$ representing $| (x - 4) (y - 4) (z - 4) \rangle$ and the number of matrix element $(x, y, z)$ is the coefficient of $| (x - 4) (y - 4) (z - 4) \rangle$.

```matlab
% close all
clear
clc

% Constanten
D = -1.29*(10^(-3)); % -1.29 meV, uit projectomschrijving
Ez = 0.035*(10^(-3)); % 0.035 meV, uit projectomschrijving
J = -0.73*(10^(-3)); % -0.73 meV, uit projectomschrijving

% We maken een cel met alle eigenstaten
Staten = cell([125,1]); % 125, want er zijn 125 deelstaten en 125 eigenwaarden, dus 125 eigenenergien
Ham_Staten = cell([125,1]); % De hamiltoniaan toegepast op de niet genormaliseerde eigenstaten
Eigen_energie = cell([125,1]); % De eigenenergie
Energie_check = zeros(125,1); % Check of de eigenenergie klopt en dus check of het de goede eigenwaarden zijn.

for n = 1:125
    Staten{n,1} = zeros(7,7,7); % 7 bij 7 bij 7, omdat we van -3 tot 3 moeten voor als we hamiltoniaan gaan doen, de rand is wordt dan wel nul, want die staten kunnen niet.
    Ham_Staten{n,1} = zeros(7,7,7);
    Eigen_energie{n,1} = zeros(7,7,7);
end

% Normeren van de eigenstaten
% Staat 1 heeft maar 1 element dus is al genormeerd.
for i = 1:1
```

% Close all
% clear
% clc

% Constants
D = -1.29*(10^(-3)); % -1.29 meV, from project notation
Ez = 0.035*(10^(-3)); % 0.035 meV, from project notation
J = -0.73*(10^(-3)); % -0.73 meV, from project notation

% We make a cell with all eigenstates
Staten = cell([125,1]); % 125, want there are 125 substates and 125 eigenvalues, thus 125 eigenvalues
Ham_Staten = cell([125,1]); % The hamiltonian applied to the unnormalized eigenstates
Eigen_energie = cell([125,1]); % The eigenenergy
Energie_check = zeros(125,1); % Check if the eigenenergy is correct and thus check if the good eigenvalues are there.

for n = 1:125
    Staten{n,1} = zeros(7,7,7); % 7 by 7 by 7, because we go from -3 to 3 when doing the hamiltonian, the boundary is thus zero, which is why these states cannot exist.
    Ham_Staten{n,1} = zeros(7,7,7);
    Eigen_energie{n,1} = zeros(7,7,7);
end

% Normalize the eigenstates
% State 1 has only 1 element so is already normalized.
for i = 1:1
\begin{verbatim}
Staten\{1,1\}(2,2,2) = 1;
Energie_check(1,1) = 12*D + 6*Ez + 8*J;
end % Energie klopt (1)

% Staat 2
for i = 1:1
    A2 = zeros(3,3);
    A2(1,1) = 1;
    A2(1,2) = 1;
    A2(2,1) = 1;
    A2(2,3) = 1;
    A2(3,2) = 1;
    A2(3,3) = 1;

    [EV2, labda2] = eig(A2);
    for n = 1:3
        Staten\{(1+n),1\}(3,2,2) = EV2(1,n);
        Staten\{(1+n),1\}(2,3,2) = EV2(2,n);
        Staten\{(1+n),1\}(2,2,3) = EV2(3,n);
        Energie_check((1+n),1) = 9*D + 5*Ez + 4*J + 2*J*labda2(n,n);
    end
end % Energie klopt (2 t/m 4)

% Staat 3
for i = 1:1
    A3 = zeros(6,6);
    A3(1,1) = 2*D + 4*J;
    A3(1,2) = sqrt(6)*J;
    A3(2,1) = sqrt(6)*J;
    A3(2,2) = 3*J;
    A3(2,3) = 2*J;
    A3(3,2) = 2*J;
    A3(3,3) = 4*J;
    A3(2,4) = sqrt(6)*J;
    A3(4,2) = sqrt(6)*J;
    A3(4,4) = 2*D;
    A3(3,5) = 2*J;
    A3(5,3) = 2*J;
    A3(4,5) = sqrt(6)*J;
    A3(5,4) = sqrt(6)*J;
    A3(5,5) = 3*J;
    A3(5,6) = sqrt(6)*J;
    A3(6,5) = sqrt(6)*J;
    A3(6,6) = 2*D + 4*J;

    [EV3, labda3] = eig(A3);
    for n = 1:6
        Staten\{(4+n),1\}(4,2,2) = EV3(1,n);
        Staten\{(4+n),1\}(3,3,2) = EV3(2,n);
        Staten\{(4+n),1\}(3,2,3) = EV3(3,n);
        Staten\{(4+n),1\}(2,4,2) = EV3(4,n);
        Staten\{(4+n),1\}(2,3,3) = EV3(5,n);
        Staten\{(4+n),1\}(2,2,4) = EV3(6,n);
        Energie_check((4+n),1) = 6*D + 4*Ez + labda3(n,n);
    end
\end{verbatim}
A.7. Code for eigenstates of the three particle system with zero transverse magnetic anisotropy

```matlab
end % Energie klopt (5 t/m 10)

% Staat 4
for i = 1:1
    A4 = zeros(10,10);
    A4(1,1) = 6*D + 2*J;
    A4(2,1) = sqrt(6)*J;
    A4(1,2) = sqrt(6)*J;
    A4(2,2) = 2*D + 2*J;
    A4(2,3) = 2*J;
    A4(3,2) = 2*J;
    A4(3,3) = 2*D + 2*J;
    A4(2,4) = 3*J;
    A4(4,2) = 3*J;
    A4(4,4) = 2*D;
    A4(3,5) = sqrt(6)*J;
    A4(5,3) = sqrt(6)*J;
    A4(4,5) = sqrt(6)*J;
    A4(5,4) = sqrt(6)*J;
    A4(5,5) = 2*J;
    A4(5,6) = sqrt(6)*J;
    A4(6,5) = sqrt(6)*J;
    A4(6,6) = 2*D + 2*J;
    A4(4,7) = sqrt(6)*J;
    A4(7,4) = sqrt(6)*J;
    A4(7,7) = 6*D - 4*J;
    A4(5,8) = sqrt(6)*J;
    A4(8,5) = sqrt(6)*J;
    A4(8,7) = sqrt(6)*J;
    A4(7,8) = sqrt(6)*J;
    A4(8,8) = 2*D;
    A4(6,9) = 2*J;
    A4(9,6) = 2*J;
    A4(8,9) = 3*J;
    A4(9,8) = 3*J;
    A4(9,9) = 2*D + 2*J;
    A4(9,10) = sqrt(6)*J;
    A4(10,9) = sqrt(6)*J;
    A4(10,10) = 6*D + 2*J;
end

[EV4, labda4] = eig(A4);

for n = 1:10
    Staten((10+n),1)(5,2,2) = EV4(1,n);
    Staten((10+n),1)(4,3,2) = EV4(2,n);
    Staten((10+n),1)(4,2,3) = EV4(3,n);
    Staten((10+n),1)(3,4,2) = EV4(4,n);
    Staten((10+n),1)(3,3,3) = EV4(5,n);
    Staten((10+n),1)(3,2,4) = EV4(6,n);
    Staten((10+n),1)(2,5,2) = EV4(7,n);
    Staten((10+n),1)(2,4,3) = EV4(8,n);
    Staten((10+n),1)(2,3,4) = EV4(9,n);
    Staten((10+n),1)(2,2,5) = EV4(10,n);
end

Energie_check((10+n),1) = 3*D + 3*Ez + labda4(n,n);
end
```
A5 = zeros(15,15);
A5(1,1) = 10*D;
A5(1,2) = 2*J;
A5(2,1) = 2*J;
A5(2,2) = 4*D + J;
A5(2,3) = 2*J;
A5(3,2) = 2*J;
A5(3,3) = 4*D;
A5(2,4) = 3*J;
A5(4,2) = 3*J;
A5(4,4) = 2*D;
A5(3,5) = sqrt(6)*J;
A5(5,3) = sqrt(6)*J;
A5(4,5) = sqrt(6)*J;
A5(5,4) = sqrt(6)*J;
A5(5,5) = J;
A5(5,6) = sqrt(6)*J;
A5(6,5) = sqrt(6)*J;
A5(6,6) = 2*D;
A5(4,7) = 3*J;
A5(7,4) = 3*J;
A5(7,7) = 4*D - 3*J;
A5(5,8) = 3*J;
A5(8,5) = 3*J;
A5(7,8) = sqrt(6)*J;
A5(8,7) = sqrt(6)*J;
A5(6,9) = sqrt(6)*J;
A5(9,6) = sqrt(6)*J;
A5(9,9) = J;
A5(9,10) = sqrt(6)*J;
A5(10,9) = sqrt(6)*J;
A5(10,10) = 4*D;
A5(7,11) = 2*J;
A5(11,7) = 2*J;
A5(11,11) = 10*D - 8*J;
A5(12,8) = sqrt(6)*J;
A5(8,12) = sqrt(6)*J;
A5(12,11) = 2*J;
A5(11,12) = 2*J;
A5(12,12) = 4*D - 3*J;
A5(9,13) = sqrt(6)*J;
A5(13,9) = sqrt(6)*J;
A5(12,13) = 3*J;
A5(13,12) = 3*J;
A5(13,13) = 2*D;
A5(10,14) = 2*J;
A5(14,10) = 2*J;
A5(14,13) = 3*J;
A5(13,14) = 3*J;
A.7. Code for eigenstates of the three particle system with zero transverse magnetic anisotropy

\[ A5(14,14) = 4D + J; \]
\[ A5(15,14) = 2J; \]
\[ A5(14,15) = 2J; \]
\[ A5(15,15) = 10D; \]

\[ [EV5, \lambda 5] = \text{eig}(A5); \]

\begin{verbatim}
for n = 1:15
    Staten{(20+n),1}(6,2,2) = EV5(1,n);
    Staten{(20+n),1}(5,3,2) = EV5(2,n);
    Staten{(20+n),1}(5,2,3) = EV5(3,n);
    Staten{(20+n),1}(4,4,2) = EV5(4,n);
    Staten{(20+n),1}(4,3,3) = EV5(5,n);
    Staten{(20+n),1}(4,2,4) = EV5(6,n);
    Staten{(20+n),1}(3,5,2) = EV5(7,n);
    Staten{(20+n),1}(3,4,3) = EV5(8,n);
    Staten{(20+n),1}(3,3,4) = EV5(9,n);
    Staten{(20+n),1}(3,2,5) = EV5(10,n);
    Staten{(20+n),1}(2,6,2) = EV5(11,n);
    Staten{(20+n),1}(2,5,3) = EV5(12,n);
    Staten{(20+n),1}(2,4,4) = EV5(13,n);
    Staten{(20+n),1}(2,3,5) = EV5(14,n);
    Staten{(20+n),1}(2,2,6) = EV5(15,n);
    Energie_check((20+n),1) = 2D + 2Ez + labda5(n,n);
end
end % Energie klopt (21 t/m 35)
\end{verbatim}

% Staat 6

\begin{verbatim}
for i = 1:1
    A6 = zeros(18,18);
    A6(1,1) = 8D;
    A6(1,2) = 2J;
    A6(2,1) = 2J;
    A6(2,2) = 8D - 2J;
    A6(1,3) = sqrt(6)*J;
    A6(3,1) = sqrt(6)*J;
    A6(3,3) = 4D;
    A6(2,4) = 2J;
    A6(4,2) = 2J;
    A6(4,3) = sqrt(6)*J;
    A6(3,4) = sqrt(6)*J;
    A6(4,4) = 2D;
    A6(4,5) = sqrt(6)*J;
    A6(5,4) = sqrt(6)*J;
    A6(5,5) = 4D - 2J;
    A6(3,6) = 3J;
    A6(6,3) = 3J;
    A6(6,6) = 4D - 2J;
    A6(4,7) = 3J;
    A6(7,4) = 3J;
    A6(6,7) = sqrt(6)*J;
    A6(7,6) = sqrt(6)*J;
    A6(5,8) = sqrt(6)*J;
    A6(8,5) = sqrt(6)*J;
    A6(8,7) = 3J;
    A6(7,8) = 3J;
\end{verbatim}
A6(9,8) = sqrt(6)*J;
A6(8,9) = sqrt(6)*J;
A6(9,9) = 4*D - 2*J;
A6(6,10) = sqrt(6)*J;
A6(10,6) = sqrt(6)*J;
A6(10,10) = 8*D - 6*J;
A6(11,7) = 3*J;
A6(7,11) = 3*J;
A6(10,11) = 2*J;
A6(11,10) = 2*J;
A6(11,11) = 3*J;
A6(9,13) = sqrt(6)*J;
A6(13,9) = sqrt(6)*J;
A6(13,12) = 3*J;
A6(12,13) = 3*J;
A6(13,13) = 2*D;
A6(13,14) = 2*J;
A6(14,13) = 2*J;
A6(14,14) = 8*D - 2*J;
A6(11,15) = 2*J;
A6(15,11) = 2*J;
A6(15,15) = 8*D - 6*J;
A6(12,16) = sqrt(6)*J;
A6(16,12) = sqrt(6)*J;
A6(15,16) = sqrt(6)*J;
A6(16,15) = sqrt(6)*J;
A6(16,16) = 4*D - 2*J;
A6(13,17) = sqrt(6)*J;
A6(17,13) = sqrt(6)*J;
A6(16,17) = 3*J;
A6(17,16) = 3*J;
A6(17,17) = 4*D;
A6(14,18) = 2*J;
A6(18,14) = 2*J;
A6(18,17) = sqrt(6)*J;
A6(17,18) = sqrt(6)*J;
A6(18,18) = 8*D;

[EV6, labda6] = eig(A6);
for n = 1:18
    Staten((35+n),1){6,3,2} = EV6(1,n);
    Staten((35+n),1){6,2,3} = EV6(2,n);
    Staten((35+n),1){5,4,2} = EV6(3,n);
    Staten((35+n),1){5,3,3} = EV6(4,n);
    Staten((35+n),1){5,2,4} = EV6(5,n);
    Staten((35+n),1){4,5,2} = EV6(6,n);
    Staten((35+n),1){4,4,3} = EV6(7,n);
    Staten((35+n),1){4,3,4} = EV6(8,n);
    Staten((35+n),1){4,2,5} = EV6(9,n);
    Staten((35+n),1){3,6,2} = EV6(10,n);
    Staten((35+n),1){3,5,3} = EV6(11,n);
Staten \((35+n)_{\{1\}}(3,4,4) = EV6(12,n)\);
Staten \((35+n)_{\{1\}}(3,3,5) = EV6(13,n)\);
Staten \((35+n)_{\{1\}}(3,2,6) = EV6(14,n)\);
Staten \((35+n)_{\{1\}}(2,6,3) = EV6(15,n)\);
Staten \((35+n)_{\{1\}}(2,5,4) = EV6(16,n)\);
Staten \((35+n)_{\{1\}}(2,4,5) = EV6(17,n)\);
Staten \((35+n)_{\{1\}}(2,3,6) = EV6(18,n)\);
Energie_check \((35+n)_{\{1\}} = D + Ez + \lambda 6(n,n)\);
\( A7(12,12) = 8D - 4J; \)
\( A7(9,13) = \text{sqrt}(6) \cdot J; \)
\( A7(13,9) = \text{sqrt}(6) \cdot J; \)
\( A7(13,13) = 6D - 4J; \)
\( A7(14,10) = 3J; \)
\( A7(10,14) = 3J; \)
\( A7(14,13) = \text{sqrt}(6) \cdot J; \)
\( A7(13,14) = \text{sqrt}(6) \cdot J; \)
\( A7(14,14) = 2D - J; \)
\( A7(11,15) = 3J; \)
\( A7(15,11) = 3J; \)
\( A7(14,15) = 3J; \)
\( A7(15,14) = 3J; \)
\( A7(15,15) = 2D; \)
\( A7(12,16) = \text{sqrt}(6) \cdot J; \)
\( A7(16,12) = \text{sqrt}(6) \cdot J; \)
\( A7(15,16) = \text{sqrt}(6) \cdot J; \)
\( A7(16,15) = \text{sqrt}(6) \cdot J; \)
\( A7(16,16) = 6D - J; \)
\( A7(14,17) = 2J; \)
\( A7(17,14) = 2J; \)
\( A7(17,17) = 8D - 4J; \)
\( A7(15,18) = \text{sqrt}(6) \cdot J; \)
\( A7(18,15) = \text{sqrt}(6) \cdot J; \)
\( A7(17,18) = \text{sqrt}(6) \cdot J; \)
\( A7(18,17) = \text{sqrt}(6) \cdot J; \)
\( A7(18,18) = 6D - J; \)
\( A7(16,19) = \text{sqrt}(6) \cdot J; \)
\( A7(19,16) = \text{sqrt}(6) \cdot J; \)
\( A7(18,19) = \text{sqrt}(6) \cdot J; \)
\( A7(19,18) = \text{sqrt}(6) \cdot J; \)
\( A7(19,19) = 8D; \)

\[ [\text{EV7}, \text{labda7}] = \text{eig}(A7); \]

\textbf{for} n = 1:19

\( \text{Staten}((53+n),1)(6,4,2) = \text{EV7}(1,n); \)
\( \text{Staten}((53+n),1)(6,3,3) = \text{EV7}(2,n); \)
\( \text{Staten}((53+n),1)(6,2,4) = \text{EV7}(3,n); \)
\( \text{Staten}((53+n),1)(5,5,2) = \text{EV7}(4,n); \)
\( \text{Staten}((53+n),1)(5,4,3) = \text{EV7}(5,n); \)
\( \text{Staten}((53+n),1)(5,3,4) = \text{EV7}(6,n); \)
\( \text{Staten}((53+n),1)(5,2,5) = \text{EV7}(7,n); \)
\( \text{Staten}((53+n),1)(4,6,2) = \text{EV7}(8,n); \)
\( \text{Staten}((53+n),1)(4,5,3) = \text{EV7}(9,n); \)
\( \text{Staten}((53+n),1)(4,4,4) = \text{EV7}(10,n); \)
\( \text{Staten}((53+n),1)(4,3,5) = \text{EV7}(11,n); \)
\( \text{Staten}((53+n),1)(4,2,6) = \text{EV7}(12,n); \)
\( \text{Staten}((53+n),1)(3,6,3) = \text{EV7}(13,n); \)
\( \text{Staten}((53+n),1)(3,5,4) = \text{EV7}(14,n); \)
\( \text{Staten}((53+n),1)(3,4,5) = \text{EV7}(15,n); \)
\( \text{Staten}((53+n),1)(3,3,6) = \text{EV7}(16,n); \)
\( \text{Staten}((53+n),1)(2,6,4) = \text{EV7}(17,n); \)
\( \text{Staten}((53+n),1)(2,5,5) = \text{EV7}(18,n); \)
\( \text{Staten}((53+n),1)(2,4,6) = \text{EV7}(19,n); \)
\( \text{Energie_check}((53+n),1) = \text{labda7}(n,n); \)
end
end % Energie klopt (54 t/m 72)

% Staat 8
for n = 1:18
    % We gebruiken EV6, want A6 = A8 als je 'goed' nummert
    Staten([(72+n) , 1] (2, 5, 6) ) = EV6(1,n);
    Staten([(72+n) , 1] (2, 6, 5) ) = EV6(2,n);
    Staten([(72+n) , 1] (3, 4, 6) ) = EV6(3,n);
    Staten([(72+n) , 1] (3, 5, 5) ) = EV6(4,n);
    Staten([(72+n) , 1] (3, 6, 4) ) = EV6(5,n);
    Staten([(72+n) , 1] (4, 3, 6) ) = EV6(6,n);
    Staten([(72+n) , 1] (4, 4, 5) ) = EV6(7,n);
    Staten([(72+n) , 1] (4, 5, 4) ) = EV6(8,n);
    Staten([(72+n) , 1] (4, 6, 3) ) = EV6(9,n);
    Staten([(72+n) , 1] (5, 2, 6) ) = EV6(10,n);
    Staten([(72+n) , 1] (5, 3, 5) ) = EV6(11,n);
    Staten([(72+n) , 1] (5, 4, 4) ) = EV6(12,n);
    Staten([(72+n) , 1] (5, 5, 3) ) = EV6(13,n);
    Staten([(72+n) , 1] (5, 6, 2) ) = EV6(14,n);
    Staten([(72+n) , 1] (6, 2, 5) ) = EV6(15,n);
    Staten([(72+n) , 1] (6, 3, 4) ) = EV6(16,n);
    Staten([(72+n) , 1] (6, 4, 3) ) = EV6(17,n);
    Staten([(72+n) , 1] (6, 5, 2) ) = EV6(18,n);
    Energie_check([(72+n) , 1] ) = D - Ez + lambda6(n,n);
end % Energie klopt (73 t/m 90)

% Staat 9
for n = 1:15
    % We gebruiken EV5, want A5 = A9 als je 'goed' nummert
    Staten([(90+n) , 1] (2, 6, 6) ) = EV5(1,n);
    Staten([(90+n) , 1] (3, 5, 6) ) = EV5(2,n);
    Staten([(90+n) , 1] (3, 6, 5) ) = EV5(3,n);
    Staten([(90+n) , 1] (4, 4, 6) ) = EV5(4,n);
    Staten([(90+n) , 1] (4, 5, 5) ) = EV5(5,n);
    Staten([(90+n) , 1] (4, 6, 4) ) = EV5(6,n);
    Staten([(90+n) , 1] (5, 3, 6) ) = EV5(7,n);
    Staten([(90+n) , 1] (5, 4, 5) ) = EV5(8,n);
    Staten([(90+n) , 1] (5, 5, 4) ) = EV5(9,n);
    Staten([(90+n) , 1] (5, 6, 3) ) = EV5(10,n);
    Staten([(90+n) , 1] (6, 2, 6) ) = EV5(11,n);
    Staten([(90+n) , 1] (6, 3, 5) ) = EV5(12,n);
    Staten([(90+n) , 1] (6, 4, 4) ) = EV5(13,n);
    Staten([(90+n) , 1] (6, 5, 3) ) = EV5(14,n);
    Staten([(90+n) , 1] (6, 6, 2) ) = EV5(15,n);
    Energie_check([(90+n) , 1] ) = 2*D - 2*Ez + lambda5(n,n);
end % Energie klopt (91 t/m 105)

% Staat 10
for n = 1:10
    Staten([(105+n) , 1] (3, 6, 6) ) = EV4(1,n);
    Staten([(105+n) , 1] (4, 5, 6) ) = EV4(2,n);
    Staten([(105+n) , 1] (4, 6, 5) ) = EV4(3,n);
    Staten([(105+n) , 1] (5, 4, 6) ) = EV4(4,n);
    Staten([(105+n) , 1] (5, 5, 5) ) = EV4(5,n);
    Staten([(105+n) , 1] (5, 6, 4) ) = EV4(6,n);
Staten \{(105+n),1\}(6,3,6) = EV4(7,n); 
Staten \{(105+n),1\}(6,4,5) = EV4(8,n); 
Staten \{(105+n),1\}(6,5,4) = EV4(9,n); 
Staten \{(105+n),1\}(6,6,3) = EV4(10,n); 
Energie_check((105+n),1) = 3*D - 3*Ez + labda4(n,n); 

% Staat 11
for n = 1:6 
Staten \{(115+n),1\}(4,6,6) = EV3(1,n); 
Staten \{(115+n),1\}(5,5,6) = EV3(2,n); 
Staten \{(115+n),1\}(5,6,5) = EV3(3,n); 
Staten \{(115+n),1\}(6,4,6) = EV3(4,n); 
Staten \{(115+n),1\}(6,5,5) = EV3(5,n); 
Staten \{(115+n),1\}(6,6,4) = EV3(6,n); 
Energie_check((115+n),1) = 6*D - 4*Ez + labda3(n,n); 
end % Energie klopt (116 t/m 121)

% Staat 12
for n = 1:3 
Staten \{(121+n),1\}(5,6,6) = EV2(1,n); 
Staten \{(121+n),1\}(6,5,6) = EV2(2,n); 
Staten \{(121+n),1\}(6,6,5) = EV2(3,n); 
Energie_check((121+n),1) = 9*D - 5*Ez + 4*J + 2*J*labda2(n,n); 
end % Energie klopt (122 t/m 124)

% Staat 13 heeft maar 1 element dus is al genormeerd.
for i = 1:1 
Staten(125,1)(6,6,6) = 1; 
Energie_check(125,1) = 12*D - 6*Ez +8*J; 
end % Energie klopt (125)

% Doe nu de hamiltoniaan op de eigenstaten en vind de eigenenergien 
for n = 1:125 
for i = 2:6 
for j = 2:6 
for s = 2:6 
Ham_Staten\{n,1\}(i,j,s) = ( ((i-4)^2 + (j-4)^2 + (s-4)^2)*D -(i-4 +j -4 +s-4)*Ez + ((i-4)*(j-4) + (j-4)*(s-4))*J )*Staten\{n,1\}(i,j,s) + (J/2)*\sqrt(6 -(i-4)*(i-4+1))*\sqrt(6 -(j-4)*(j-4-1))*Staten\{n,1\}(i+1,j-1,s) + (J/2)*\sqrt(6 -(s-4)*(s-4-1))*Staten\{n,1\}(i,j+1,s-1) + (J/2)*\sqrt(6 -(j-4)*(j-4-1))*\sqrt(6 -(s-4)*(s-4-1))*Staten\{n,1\}(i,j,s+1) ;
if Staten\{n,1\}(i,j,s) == 0
else
Eigen_energie\{n,1\}(i,j,s) = (1.602*10^-19)*Ham_Staten\{n,1\}(i,j,s) / Staten\{n,1\}(i,j,s) ; % zodat het in joule staat
end
end
end 
% Sommige coefficienten zijn 0 of heel klein <10^-16 en dan wordt
% eigen_energie raar en niet zelfde als energie_check, maar dat komt dus
% door die coefficienten, want we delen daardoor bij de berekenening.

Energie_check = (1.602*10^-19)*Energie_check; % We hebben in elektron volt gerekend
en moeten nu naar joule

% Normerings check en orthogonaliteits check
Norm_check = zeros(125,125);
for i = 1:125
    for j = 1:125
        Norm_check(i,j) = sum(sum(sum(Staten{i,1} .* Staten{j,1})));
    end
end

% We zien dat alle staten goed genormeerd zijn, want inproduct met zichzelf is 1
% We zien ook dat het inproduct tussen verschillende staten niet altijd 1
% is, maar het is heel klein < 10^-15 en dit is verwaarloosbaar.
% Het is niet precies 0, omdat matlab het getal moet opslaan en dan moet
% hij het afronden op een aantal decimalen en daarom hebben we deze kleine 'fout'

A.8. Code for tunneling current, eigenstates and eigenenergies of the two particle system

The code is listed below. The eigenstates are stored in a cell called 'Staten' and the eigenenergies are stored in a vector called 'Energie'. Staten{i,1} is eigenstate i, with corresponding eigenenergy 'Energie(i,1)'. Staten{i,1} is a five by five matrix with matrix element \((x, y)\) representing \(|(x-3) \cdot (y-3)\rangle\) and the number of matrix element \((x, y)\) is the coefficient of \(|(x-3) \cdot (y-3)\rangle\).

In the second part of the code called 'Stroomformule' the location of the STM tip, the applied voltage range and the temperature can be changed. If variables were changed, run the code again. The new current is displayed in the same figure to make it easier to see the influences of the changes.

%close all
clear
clc

format long

% Constanten
D = -1.29*(10^-3); % -1.29 meV, uit projectomschrijving
E = 0.31*(10^-3);  % 0.31 meV, uit projectomschrijving
Ez = 0.035*(10^-3); % 0.035 meV, uit projectomschrijving
J = -0.73*(10^-3); % -0.73 meV, uit projectomschrijving

% Maak de kubussen, iedere kubus is 1 staat.
kubus = cell([25,1]);
teller_plek = 1;
for n = 1:25
    kubus{n,1} = zeros(5,5);
teller = 1;
    for b = 1:5
        for a = 1:5
            if teller == teller_plek
                kubus{n,1}(a,b) = 1;
            end
            teller = teller+1;
        end
    end


```matlab
end
teller_plek = teller_plek + 1;
end

% Doe de hamiltoniaan op de kubussen
ham_kubus = cell([25,1]);
teller_plek = 1;
for n = 1:25
    ham_kubus{n,1} = zeros(5,5);
teller = 1;
    for b = 1:5
        for a = 1:5
            if teller == teller_plek
                % ham_kubus = Ham( kubus )
                ham_kubus{n,1}(a,b) = ((a-3)^2 + (b-3)^2)*D - (a-3 + b-3)*Ez + ((a-3)*(b-3))*J)*kubus{n,1}(a,b);
            end
            if (a < 5) && (b > 1)
                ham_kubus{n,1}(a+1,b-1) = (J/2)*sqrt(6-(a-3)*(a-3+1))*sqrt(6-(b-3)*(b-3-1))*kubus{n,1}(a,b);
            end
            if (a > 1) && (b < 5)
                ham_kubus{n,1}(a-1,b+1) = (J/2)*sqrt(6-(a-3)*(a-3-1))*sqrt(6-(b-3)*(b-3+1))*kubus{n,1}(a,b);
            end
            if a < 4
                ham_kubus{n,1}(a+2,b) = (E/2)*sqrt(6-(a-3+1)*(a-3+2))*sqrt(6-(a-3+1))*kubus{n,1}(a,b);
            end
            if a > 2
                ham_kubus{n,1}(a-2,b) = (E/2)*sqrt(6-(a-3-1)*(a-3-2))*sqrt(6-(a-3-1))*kubus{n,1}(a,b);
            end
            if b < 4
                ham_kubus{n,1}(a,b+2) = (E/2)*sqrt(6-(b-3+1)*(b-3+2))*sqrt(6-(b-3+1))*kubus{n,1}(a,b);
            end
            if b > 2
                ham_kubus{n,1}(a,b-2) = (E/2)*sqrt(6-(b-3-1)*(b-3-2))*sqrt(6-(b-3-1))*kubus{n,1}(a,b);
            end
            teller = teller + 1;
        end
        teller_plek = teller_plek + 1;
    end
end

% Maak nu van de kubus een lange vector van 125 bij 1
vector = cell([25,1]);
for n = 1:25
    vector{n,1} = reshape(ham_kubus{n,1},25,1);
end

% Plak nu alle vectoren aan elkaar en maak zo een 125 bij 125 matrix, deze is symmetrisch
```
A.8. Code for tunneling current, eigenstates and eigenenergies of the two particle system

```matlab
matrix = zeros(25,25);
for n = 1:25
    matrix(:,n) = vector{n,1};
end

% Vind nu de eigenvectoren en eigenwaarden van deze grote matrix met het commando:
eig
[vector_eigenstaten, diagonaal_eigenwaarden] = eig(matrix);

% Haal nu de eigenwaarden van de diagonaal van diagonaal_eigenwaarden
Energie = zeros(25,1);
for n = 1:25
    Energie(n,1) = diagonaal_eigenwaarden(n,n);
end
Energie = (1.602*10^-19)*Energie; % We hebben in elektron volt gerekend en moeten nu naar joule

% De eigenstaten zijn de kolommen van vector_eigenstaten, haal deze
% vectoren er uit en bouw ze tot matrix.
Staten = cell([25,1]);
in_R = zeros(25,1);
for n = 1:25
    Staten{n,1} = reshape(vector_eigenstaten(:,n), 5, 5);
in_R(n,1) = isreal(Staten{n,1});
end

% Sommige coefficienten zijn heel klein <10^-17 in plaats van 0, komt door
% het eigenvectoren vinden van matlab
% check of de staten reeel zijn
if sum(in_R) == 25
    disp('Alle eigenstaten hebben reele coefficienten')
else
    disp('Er zijn staten met complexe coefficienten')
end

% Normerings check en orhtogonaliteits check
Norm_check = zeros(25,25);
for i = 1:25
    for j = 1:25
        Norm_check(i,j) = sum(sum(Staten{i,1}.*Staten{j,1}));
    end
end
% We zien dat alle staten goed genormeerd zijn, want inproduct met zichzelf is 1
% We zien ook dat het inproduct tussen verschillende staten niet altijd 1
% is, maar het is heel klein < 10^-15 en dit is verwaarloosbaar.
% Het is niet precies 0, omdat matlab het getal moet opslaan en dan moet
% hij het afronden op een aantal decimalen en daarom hebben we deze kleine 'fout'

% Stroomformule

% Tip van de STM boven atoom 1 of 2 (want we hebben maar 2 atomen)
Tip = 1;

% Voltage
Vmin = -5*10^(-4); % Begin voltage
Vmax = 5*10^(-4); % Eind voltage
```
deltaV = 10^{(-6)};  % Stapgrootte
V = (Vmin: deltaV: Vmax);  % van Vmin tot Vmax met stapjes van deltaV

% Temperatuur in kelvin
T = 1;  % [K]

% Constanten
kb = 1.38*10^{(-23)};  % constante van Boltzmann [J/K]
beta = 1/(kb*T);  % [1/J]
e = 1.602*10^{(-19)};  % [C] = [J/V] = [A*s]
h = 6.626*10^{(-34)};  % [J*s]
Gs = (2*e)/h;  % [C/(J*s)] = [A/J]

% i is de teller van M
% j is de teller van M'
delta = zeros(25,25);  % en delta = E_M' - E_M
S_plus_M = cell([25,1]);
S_min_M = cell([25,1]);
S_z_M = cell([25,1]);
F = cell([25,25]);  % Het ding met de voltages en e macht enzo.

for i = 1:25
    S_plus_M{i,1} = zeros(5,5);  % dit is alleen om te vullen met 0 matrices
    S_min_M{i,1} = zeros(5,5);  % dit is alleen om te vullen met 0 matrices
    S_z_M{i,1} = zeros(5,5);  % dit is alleen om te vullen met 0 matrices

    for j = 1:25
        delta(i,j) = (Energie(j,1) - Energie(i,1));
        % In F zit meteen +s, dus 2 en -2
        F{i,j} = (e*V - 2*delta(i,j))./(1 - exp(-2*beta*(e*V - 2*delta(i,j))) + (e*V + 2*delta(i,j))./(1 - exp(2*beta*(e*V + 2*delta(i,j)))));
    end
end

% Bereken de spin operator toegepast op het atoom waar de tip boven staat,
% daarom ook alle ifs om te checken welke operator gebruikt moet worden
if Tip == 1
    for n = 1:25
        for i = 1:5
            for j = 1:5
                if i < 5
                    S_plus_M{n,1}(i+1,j) = sqrt(6 - (i-3)*(i-3+1))*Staten{n,1}(i,j);
                end
                if i > 1
                    S_min_M{n,1}(i-1,j) = sqrt(6 - (i-3)*(i-3-1))*Staten{n,1}(i,j);
                end
                S_z_M{n,1}(i,j) = (i-3)*Staten{n,1}(i,j);
            end
        end
    end
elseif Tip == 2
    for n = 1:25
        for i = 1:5
            for j = 1:5
                if j < 5
                    S_plus_M{n,1}(i,j+1) = sqrt(6 - (j-3)*(j-3+1))*Staten{n,1}(i,j);
                end
                if j > 5
                    S_min_M{n,1}(i,j-1) = sqrt(6 - (j-3)*(j-3-1))*Staten{n,1}(i,j);
                end
                S_z_M{n,1}(i,j) = (j-3)*Staten{n,1}(i,j);
            end
        end
    end
A.8. Code for tunneling current, eigenstates and eigenenergies of the two particle system

```matlab
S_min_M{n, 1}(i, j−1) = sqrt(6 − (j−3)∗(j−3−1))∗Staten{n, 1}(i, j);
end
S_z_M{n, 1}(i, j) = (j−3)∗Staten{n, 1}(i, j);
end
else
disp('Vul 1 of 2 in bij Tip, want we hebben maar 2 atomen.');
end
boltz_som = 0;
for i = 1:25
    boltz_som = boltz_som + exp(-beta∗Energie(i, 1));
end
probability = zeros(25,1);
for i = 1:25
    probability(i, 1) = exp(-beta∗Energie(i, 1))/boltz_som;
end
% Bereken de stroom
I = zeros(1, length(V));
for i = 1:25
    for j = 1:25
        I = I + Gs∗(1/2)∗(exp(-beta∗Energie(i, 1))/boltz_som)∗((sum(sum(Staten{i, 1}∗S_plus_M{j, 1})))^2)∗F{i, j};
        I = I + Gs∗(1/2)∗(exp(-beta∗Energie(i, 1))/boltz_som)∗((sum(sum(Staten{i, 1}∗S_min_M{j, 1})))^2)∗F{i, j};
        I = I + Gs∗(exp(-beta∗Energie(i, 1))/boltz_som)∗((sum(sum(Staten{i, 1}∗S_z_M{j, 1})))^2)∗F{i, j};
    end
end
% Bereken de afgeleiden
dIdV = diff(I) ./ diff(V); % Eerste afgeleide
d2IdV2 = diff(dIdV) ./ diff(V(1, 1:length(V)−1)); % Tweede afgeleide
% Maak een plaatje
figure(1)
hold on
plot(V, I, ' . ')
hold off
xlabel('Voltage (V) ')
ylabel('Stroom (A) ')
title('IV plot van 2 deeltjes op een rij ')

% Maak een plaatje van dIdV
figure(2)
hold on
plot(V(:, 1:length(dIdV)), dIdV, ' . ')
hold off
xlabel('Voltage (V) ')
ylabel('Eerste afgeleide van de stroom (A/V) ')
title('dI/dV plot van 2 deeltjes op een rij ')

% Maak een plaatje van d2IdV2
figure(3)
hold on
```
```matlab
plot(V(:,1:length(d2IdV2)),d2IdV2,'.')
hold off
xlabel('Voltage (V)')
ylabel('Tweede afgeleide van de stroom (A/V^2)')
title('d^2I/dV^2 plot van 2 deeltjes op een rij')

plot(V(:,1:length(d2IdV2)),d2IdV2,'.')
hold off
xlabel('Voltage (V)')
ylabel('Tweede afgeleide van de stroom (A/V^2)')
title('d^2I/dV^2 plot van 2 deeltjes op een rij')
```

% P_M grond toestand approximatie
i_g = find(Energie == min(Energie));
Grond = Staten{i_g,1}; % Dit is de grondtoestand == de toestand met de laagste
% energie

% Find vindt de indices waarvoor geld dat Energie_check = zijn minimum, dus zo
% vinden we de grondtoestand

% Bereken de stroom met de P_M approximatie
I_grond = zeros(1,length(V));
for j = 1:25
    I_grond = I_grond + Gs*(1/2)*((sum(sum(Grond.*S_plus_M{j,1}))))^2)*F{i_g,j};
    I_grond = I_grond + Gs*(1/2)*((sum(sum(Grond.*S_min_M{j,1}))))^2)*F{i_g,j};
    I_grond = I_grond + Gs*((sum(sum(Grond.*S_z_M{j,1}))))^2)*F{i_g,j};
end

% Bereken de afgeleiden
dlgdV = diff(I_grond)./diff(V); % Eerste afgeleide
d2lgdV2 = diff(dlgdV)./diff(V(1:(length(V)-1))); % Tweede afgeleide

% Maak een plaatje
figure(4)
hold on
plot(V,I_grond,'.')
hold off
xlabel('Voltage (V)')
ylabel('Stroom (A)')
title('IV plot van 2 deeltjes op een rij, met P_M approximatie')

% Maak een plaatje van dIdV
figure(5)
hold on
plot(V(:,1:length(dlgdV)),dlgdV,'.')
hold off
xlabel('Voltage (V)')
ylabel('Eerste afgeleide van de stroom (A/V)')
title('dI/dV plot van 2 deeltjes op een rij, met P_M approximatie')

% Maak een plaatje van d2IdV
figure(6)
hold on
plot(V(:,1:length(d2lgdV2)),d2lgdV2,'.')
hold off
xlabel('Voltage (V)')
ylabel('Tweede afgeleide van de stroom (A/V^2)')
title('d^2I/dV^2 plot van 2 deeltjes op een rij, met P_M approximatie')
A.9. Code for tunneling current, eigenstates and eigenenergies of the three particle system

The code is listed below. The eigenstates are stored in a cell called 'Staten' and the eigenenergies are stored in a vector called 'Energie'. Staten{i,1} is eigenstate i, with corresponding eigenenergy 'Energie(i,1)'. Staten{i,1} is a five by five by five matrix with matrix element \((x,y,z)\) representing \(|(x-3)(y-3)(z-3)\rangle\) and the number of matrix element \((x,y,z)\) is the coefficient of \(|(x-3)(y-3)(z-3)\rangle\).

In the second part of the code called 'Stroomformule' the location of the STM tip, the applied voltage range and the temperature can be changed. If variables were changed, run the code again. The new current is displayed in the same figure to make it easier to see the influences of the changes.

```matlab
%close all
clear
clc
format long

% Constanten
D = -1.29*(10^(-3)); % -1.29 meV, uit projectomschrijving
E = 0.31*(10^(-3));  % 0.31 meV, uit projectomschrijving
Ez = 0.035*(10^(-3)); % 0.035 meV, uit projectomschrijving
J = -0.73*(10^(-3)); % -0.73 meV, uit projectomschrijving

% Maak de kubussen, iedere kubus is 1 staat.
kubus = cell([125,1,1]);
teller_plek = 1;
for n = 1:125
    kubus{n,1} = zeros(5,5,5);
teller = 1;
    for c = 1:5
        for b = 1:5
            for a = 1:5
                if teller == teller_plek
                    kubus{n,1}(a,b,c) = 1;
                end
                teller = teller+1;
            end
        end
    end
teller_plek = teller_plek + 1;
end

% Doe de hamiltoniaan op de kubussen
ham_kubus = cell([125,1,1]);
teller_plek = 1;
for n = 1:125
    ham_kubus{n,1} = zeros(5,5,5);
teller = 1;
    for c = 1:5
        for b = 1:5
            for a = 1:5
                if teller == teller_plek
                    % ham_kubus = Ham( kubus )
                    ham_kubus{n,1}(a,b,c) = ((a-3)^2 + (b-3)^2 + (c-3)^2) * D - (a-3 + b-3 + c-3) * Ez + ((a-3) * (b-3) + (b-3) * (c-3)) * J * kubus{n,1}(a,b,c);

```
if $(a < 5) \land (b > 1)$
    \[ \text{ham}_kubus\{n, 1\}(a+1, b-1, c) = \frac{E}{2} \cdot \sqrt{(6 - (a-3) \cdot (a-3+2)) \cdot \sqrt{6 - (a-3) \cdot (a-3+1)}} \cdot kubus\{n, 1\}(a, b, c); \]
end

if $(a > 1) \land (b < 5)$
    \[ \text{ham}_kubus\{n, 1\}(a-1, b+1, c) = \frac{E}{2} \cdot \sqrt{(6 - (a-3) \cdot (a-3-2)) \cdot \sqrt{6 - (a-3) \cdot (a-3+1)}} \cdot kubus\{n, 1\}(a, b, c); \]
end

if $(b < 5) \land (c > 1)$
    \[ \text{ham}_kubus\{n, 1\}(a, b+1, c-1) = \frac{E}{2} \cdot \sqrt{(6 - (b-3) \cdot (b-3+2)) \cdot \sqrt{6 - (b-3) \cdot (b-3+1)}} \cdot kubus\{n, 1\}(a, b, c); \]
end

if $(b > 1) \land (c < 5)$
    \[ \text{ham}_kubus\{n, 1\}(a, b-2, c) = \frac{E}{2} \cdot \sqrt{(6 - (b-3) \cdot (b-3-2)) \cdot \sqrt{6 - (b-3) \cdot (b-3+1)}} \cdot kubus\{n, 1\}(a, b, c); \]
end

if $a < 4$
    \[ \text{ham}_kubus\{n, 1\}(a+2, b, c) = \frac{E}{2} \cdot \sqrt{(6 - (a-3) \cdot (a-3+2)) \cdot \sqrt{6 - (a-3) \cdot (a-3+1)}} \cdot kubus\{n, 1\}(a, b, c); \]
end

if $a > 2$
    \[ \text{ham}_kubus\{n, 1\}(a-2, b, c) = \frac{E}{2} \cdot \sqrt{(6 - (a-3) \cdot (a-3-2)) \cdot \sqrt{6 - (a-3) \cdot (a-3+1)}} \cdot kubus\{n, 1\}(a, b, c); \]
end

if $b < 4$
    \[ \text{ham}_kubus\{n, 1\}(a, b+2, c) = \frac{E}{2} \cdot \sqrt{(6 - (b-3) \cdot (b-3+2)) \cdot \sqrt{6 - (b-3) \cdot (b-3+1)}} \cdot kubus\{n, 1\}(a, b, c); \]
end

if $b > 2$
    \[ \text{ham}_kubus\{n, 1\}(a, b-2, c) = \frac{E}{2} \cdot \sqrt{(6 - (b-3) \cdot (b-3-2)) \cdot \sqrt{6 - (b-3) \cdot (b-3+1)}} \cdot kubus\{n, 1\}(a, b, c); \]
end

if $c < 4$
    \[ \text{ham}_kubus\{n, 1\}(a, b, c+2) = \frac{E}{2} \cdot \sqrt{(6 - (c-3) \cdot (c-3+2)) \cdot \sqrt{6 - (c-3) \cdot (c-3+1)}} \cdot kubus\{n, 1\}(a, b, c); \]
end

if $c > 2$
    \[ \text{ham}_kubus\{n, 1\}(a, b, c-2) = \frac{E}{2} \cdot \sqrt{(6 - (c-3) \cdot (c-3-2)) \cdot \sqrt{6 - (c-3) \cdot (c-3+1)}} \cdot kubus\{n, 1\}(a, b, c); \]
end

teller = teller + 1;

teller_plek = teller_plek + 1;

% Maak nu van de kubus een lange vector van 125 bij 1
vector = cell([125, 1]);
for $n = 1:125$
    vector{n, 1} = reshape(ham_kubus\{n, 1\}(125, 1));
end

% Plak nu alle vectoren aan elkaar en maak zo een 125 bij 125 matrix, deze is symmetrisch
matrix = zeros(125,125);
for n = 1:125
    matrix(:,n) = vector{n,1};
end

% Vind nu de eigenvectoren en eigenwaarden van deze grote matrix met het commando:
eig
[vector_eigenstaten, diagonaal_eigenwaarden] = eig(matrix);

% Haal nu de eigenwaarden van de diagonaal van diagonaal_eigenwaarden
Energie = zeros(125,1);
for n = 1:125
    Energie(n,1) = diagonaal_eigenwaarden(n,n);
end

Energie = (1.602*10^-19)*Energie; % We hebben in elektron volt gerekend en moeten nu naar joule

% De eigenstaten zijn de kolommen van vector_eigenstaten, haal deze
% vectoren eruit en bouw ze tot matrix.
Staten = cell([125,1]);
in_R = zeros(125,1);
for n = 1:125
    Staten{n,1} = reshape(vector_eigenstaten(:,n),5,5,5);
in_R(n,1) = isreal(Staten{n,1});
end

% Sommige coefficienten zijn heel klein <10^-20 in plaats van 0, komt door
% het eigenvectoren vinden van matlab
if sum(in_R) == 125
    disp('Alle eigenstaten hebben reële coëfficiënten')
else
    disp('Er zijn staten met complexe coëfficiënten')
end

% Normerings check en orthogonaliteits check
Norm_check = zeros(125,125);
for i = 1:125
    for j = 1:125
        Norm_check(i,j) = sum(sum(sum(Staten{i,1}.*Staten{j,1})));
    end
end

% We zien dat alle staten goed genormeerd zijn, want inproduct met zichzelf is 1
% We zien ook dat het inproduct tussen verschillende staten niet altijd 1
% is, maar het is heel klein < 10^-15 en dit is verwaarloosbaar.
% Het is niet precies 0, omdat matlab het getal moet opslaan en dan moet
% hij het afronden op een aantal decimalen en daarom hebben we deze kleine 'fout'

% Stroomformule

% Tip van de STM boven atoom 1 of 2 of 3 (want we hebben maar 3 atomen)
Tip = 1;

% Voltage
Vmin = -5*10^(-4); % Begin voltage
Vmax = 5*10^(-4); % Eind voltage
\[ \text{deltaV} = 10^{(-6)}; \quad \% \text{Stapgrootte} \]
\[ V = (\text{Vmin}: \text{deltaV}: \text{Vmax}); \quad \% \text{van Vmin tot Vmax met stapjes van deltaV} \]

\% Temperatuur in kelvin
\[ T = 1; \quad \% [K] \quad \% \text{Voor temperaturen kleiner dan 0.4 K, dan doen de eerste 3 figuren er niet toe, want de boltzman som is dan bijna oneindig.} \]
\% En je zit in het 0 K limiet dat je alleen maar in de grondtoestand zit, dat is plaatje 4 t/m 6

\% Constanten
\[ \text{kb} = 1.38*10^{(-23)}; \quad \% \text{constante van Boltzmann [J/K]} \]
\[ \text{beta} = 1/(\text{kb} \cdot T); \quad \% [1/J] \]
\[ e = 1.602*10^{(-19)}; \quad \% [C] = [J/V] = [A*s] \]
\[ h = 6.626*10^{(-34)}; \quad \% [Js] \]
\[ \text{Gs} = (2\cdot e)/h; \quad \% [C/(Js)] = [A/J] \]

\% i is de teller van M
\% j is de teller van M'
\[ \text{delta} = \text{zeros}(125,125); \quad \% \text{en delta = E_M' - E_M} \]
\[ \text{S_plus_M} = \text{cell}([125,1]); \]
\[ \text{S_min_M} = \text{cell}([125,1]); \]
\[ \text{S_z_M} = \text{cell}([125,1]); \]
\[ \text{F} = \text{cell}([125,125]); \quad \% \text{Het ding met de voltages en e macht enzo.} \]
\% Bereken de spin operator toegepast op het atoom waar de tip boven staat,
\% daarom ook alle if's om te checken welke operator gebruikt moet worden
\% iif Tip == 1
\% \% iif Tip == 2

\% Bereken de spin operator toegepast op het atoom waar de tip boven staat,
for n = 1:125
    for i = 1:5
        for j = 1:5
            for s = 1:5
                if j < 5
                    S_plus_M{n,1}(i,j+1,s) = sqrt(6 - (j-3)*(j-3+1)) * Staten{n,1}(i,j,s);
                end
                if j > 5
                    S_min_M{n,1}(i,j-1,s) = sqrt(6 - (j-3)*(j-3-1)) * Staten{n,1}(i,j,s);
                end
                S_z_M{n,1}(i,j,s) = (j-3) * Staten{n,1}(i,j,s);
            end
        end
    end
else if Tip == 3
    for n = 1:125
        for i = 1:5
            for j = 1:5
                for s = 1:5
                    if s < 5
                        S_plus_M{n,1}(i,j,s+1) = sqrt(6 - (s-3)*(s-3+1)) * Staten{n,1}(i,j,s);
                    end
                    if s > 1
                        S_min_M{n,1}(i,j,s-1) = sqrt(6 - (s-3)*(s-3-1)) * Staten{n,1}(i,j,s);
                    end
                    S_z_M{n,1}(i,j,s) = (s-3) * Staten{n,1}(i,j,s);
                end
            end
        end
    end
else
    disp('Vul 1 of 2 of 3 in bij Tip, want we hebben maar 3 atomen.');
end

boltz_som = 0;
for i = 1:125
    boltz_som = boltz_som + exp(-beta * Energie(i,1));
end
probability = zeros(125,1);
for i = 1:125
    probability(i,1) = exp(-beta * Energie(i,1)) / boltz_som;
end

% Bereken de stroom
I = zeros(1,length(V));
for i = 1:125
    for j = 1:125
        I = 1 + Gs*(1/2) * (exp(-beta * Energie(i,1)) / boltz_som) * ((sum(sum(sum(Staten{i,1}.*S_plus_M{j,1}))))^2)*F{i,j};
    end
end
\[ I = I + G_s \left( \frac{\exp(-\beta E_{\text{Energie}(i,1)})}{\text{boltz_som}} \right)^2 \sum \left( \sum \left( \sum (S_t \cdot M_{j,1}) \right)^2 \right) F[i,j] \]

end
end

% Bereken de afgeleiden
dIdV = diff(I)/deltaV;   % Eerste afgeleide
d2IdV2 = diff(dIdV)/deltaV;   % Tweede afgeleide

% Maak een plaatje
figure(1)
hold on
plot(V,I, '.')
hold off
xlabel('Voltage (V)')
ylabel('Current (A)')
title('IV plot of a 3 particle system')

% Maak een plaatje van dIdV
figure(2)
hold on
plot(V(:,1:length(dIdV)),dIdV, '.')
hold off
xlabel('Voltage (V)')
ylabel('First derivative of the current (A/V)')
title('dI/dV plot of a 3 particle system')

% Maak een plaatje van d2IdV2
figure(3)
hold on
plot(V(:,1:length(d2IdV2)),d2IdV2, '.')
hold off
xlabel('Voltage (V)')
ylabel('Second derivative of the current (A/V^2)')
title('d^2I/dV^2 plot of a 3 particle system')

% P_M grond toestand approximatie
i_g = find(Energie == min(Energie));
Grond = Staten{i_g,1};   % Dit is de grondtoestand == de toestand met de laagste energie

% Bereken de stroom met de P_M approximatie
I_grond = zeros(1,length(V));
for j = 1:125
I_grond = I_grond + Gs*(1/2)*((sum(sum(Grond.*S_plus_M{j,1})))^2)*F{i_g,j};
I_grond = I_grond + Gs*(1/2)*((sum(sum(Grond.*S_min_M{j,1})))^2)*F{i_g,j};
I_grond = I_grond + Gs*((sum(sum(Grond.*S_z_M{j,1})))^2)*F{i_g,j};
end

% Bereken de afgeleiden
dIdgV = diff(I_grond)/deltaV;   % Eerste afgeleide
d2IdgV2 = diff(dIdgV)/deltaV;   % Tweede afgeleide

% Maak een plaatje
A.10. Code for tunneling current, eigenstates and eigenenergies of the four particle system

The code is listed below. The eigenstates are stored in a cell called 'Staten' and the eigenenergies are stored in a vector called 'Energie'. Staten{i,1} is eigenstate i, with corresponding eigenenergy 'Energie(i,1)' . Staten{i,1} is a five by five by five by five matrix with matrix element
\[
|w-3\rangle \langle x-3|y-3\rangle z-3\rangle
\]
and the number of matrix element
\[
|w-3\rangle \langle x-3|y-3\rangle z-3\rangle
\]
is the coefficient of \(|w-3\rangle \langle x-3|y-3\rangle z-3\rangle\).

In the second part of the code called 'Stroomformule' the location of the STM tip, the applied voltage range and the temperature can be changed. If variables were changed, run the code again. The new current is displayed in the same figure to make it easier to see the influences of the changes.

```
%close all
clear
clc

format long

% Constanten
D = -1.29*(10^(-3)); %-1.29 meV, uit projectomschrijving
E = 0.31*(10^(-3)); %0.31 meV, uit projectomschrijving
Ez = 0.035*(10^(-3)); %0.035 meV, uit projectomschrijving
J = -0.73*(10^(-3)); %-0.73 meV, uit projectomschrijving

% Maak de kubussen, iedere kubus is 1 staat.
kubus = cell([625,1]);
teller_plek = 1;
for n = 1:625
    kubus{n,1} = zeros(5,5,5,5);
teller = 1;
    for d = 1:5
```
for c = 1:5
    for b = 1:5
        for a = 1:5
            if teller == teller_plek
                kubus{n,1}{a,b,c,d} = 1;
            end
            teller = teller + 1;
        end
    end
end

% Doe de hamiltoniaan op de kubussen
ham_kubus = cell([625,1]);
teller_plek = 1;
for n = 1:625
    ham_kubus{n,1} = zeros(5,5,5,5);
teller = 1;
    for d = 1:5
        for c = 1:5
            for b = 1:5
                for a = 1:5
                    if teller == teller_plek
                        % ham_kubus = Ham( kubus )
                        ham_kubus{n,1}{a,b,c,d} = ((a-3)^2 + (b-3)^2 + (c-3)^2 + (d-3)^2)*D -(a-3 + b-3 + c-3 + d-3)*Ez + ((a-3)*(b-3) + (b-3)*(c-3) + (c-3)*(d-3) )*J )*kubus{n,1}{a,b,c,d};
                    end
                    if (a < 5) && (b > 1)
                        ham_kubus{n,1}{a+1,b-1,c,d} = (J/2)*sqrt(6-(a-3)+(a-3+1)) ) *sqrt(6-(b-3)+(b-3+1))*kubus{n,1}{a,b,c,d};
                    end
                    if (a > 1) && (b < 5)
                        ham_kubus{n,1}{a-1,b+1,c,d} = (J/2)*sqrt(6-(a-3)+(a-3-1)) ) *sqrt(6-(b-3)+(b-3+1))*kubus{n,1}{a,b,c,d};
                    end
                    if (b < 5) && (c > 1)
                        ham_kubus{n,1}{a,b+1,c-1,d} = (J/2)*sqrt(6-(b-3)+(b-3+1)) ) *sqrt(6-(c-3)+(c-3-1))*kubus{n,1}{a,b,c,d};
                    end
                    if (b > 1) && (c < 5)
                        ham_kubus{n,1}{a,b-1,c+1,d} = (J/2)*sqrt(6-(b-3)+(b-3-1)) ) *sqrt(6-(c-3)+(c-3+1))*kubus{n,1}{a,b,c,d};
                    end
                    if (c < 5) && (d > 1)
                        ham_kubus{n,1}{a,b,c+1,d-1} = (J/2)*sqrt(6-(c-3)+(c-3+1)) ) *sqrt(6-(d-3)+(d-3-1))*kubus{n,1}{a,b,c,d};
                    end
                    if (c > 1) && (d < 5)
                        ham_kubus{n,1}{a,b,c-1,d+1} = (J/2)*sqrt(6-(c-3)+(c-3-1)) ) *sqrt(6-(d-3)+(d-3+1))*kubus{n,1}{a,b,c,d};
                    end
                end
            end
        end
    end
end

if a < 4
A.10. Code for tunneling current, eigenstates and eigenenergies of the four particle system

```matlab
ham_kubus{n,1}(a+2,b,c,d) = (E/2)*sqrt(6-(a-3+1)*(a-3+2)) * sqrt(6-(a-3)*(a-3+1)) * kubus{n,1}(a,b,c,d);
end
if a > 2
  ham_kubus{n,1}(a-2,b,c,d) = (E/2)*sqrt(6-(a-3-1)*(a-3-2)) * sqrt(6-(a-3)*(a-3+1)) * kubus{n,1}(a,b,c,d);
end
if b < 4
  ham_kubus{n,1}(a,b+2,c,d) = (E/2)*sqrt(6-(b-3+1)*(b-3+2)) * sqrt(6-(b-3)*(b-3+1)) * kubus{n,1}(a,b,c,d);
end
if b > 2
  ham_kubus{n,1}(a,b-2,c,d) = (E/2)*sqrt(6-(b-3-1)*(b-3-2)) * sqrt(6-(b-3)*(b-3+1)) * kubus{n,1}(a,b,c,d);
end
if c < 4
  ham_kubus{n,1}(a,b,c+2,d) = (E/2)*sqrt(6-(c-3+1)*(c-3+2)) * sqrt(6-(c-3)*(c-3+1)) * kubus{n,1}(a,b,c,d);
end
if c > 2
  ham_kubus{n,1}(a,b,c-2,d) = (E/2)*sqrt(6-(c-3-1)*(c-3-2)) * sqrt(6-(c-3)*(c-3+1)) * kubus{n,1}(a,b,c,d);
end
if d < 4
  ham_kubus{n,1}(a,b,c,d+2) = (E/2)*sqrt(6-(d-3+1)*(d-3+2)) * sqrt(6-(d-3)*(d-3+1)) * kubus{n,1}(a,b,c,d);
end
if d > 2
  ham_kubus{n,1}(a,b,c,d-2) = (E/2)*sqrt(6-(d-3-1)*(d-3-2)) * sqrt(6-(d-3)*(d-3+1)) * kubus{n,1}(a,b,c,d);
end
teller = teller + 1;
end
teller_plek = teller_plek + 1;
end
% Maak nu van de kubus een lange vector van 625 bij 1
vector = cell(1,625);
for n = 1:625
  vector{n,1} = reshape(ham_kubus{n,1},625,1);
end
% Plak nu alle vectoren aan elkaar en maak zo een 625 bij 625 matrix, deze is symmetrisch
matrix = zeros(625,625);
for n = 1:625
  matrix(:,n) = vector{n,1};
end
% Vind nu de eigenvectoren en eigenwaarden van deze grote matrix met het commando:
eig
  [vector_eigenstaten, diagonaal_eigenwaarden] = eig(matrix);
```
% Haal nu de eigenwaarden van de diagonaal van diagonaal_eigenwaarden
Energie = zeros(625,1);
for n = 1:625
    Energie(n,1) = diagonaal_eigenwaarden(n,n);
end
Energie = (1.602*10^−19)*Energie; % We hebben in elektron volt gerekend en moeten nu naar joule

% De eigenstaten zijn de kolommen van vector_eigenstaten, haal deze % vectoren eruit en bouw ze tot matrix.
Staten = cell([625,1]);
in_R = zeros(625,1);
for n = 1:625
    Staten{n,1} = reshape(vector_eigenstaten(:,n),5,5,5,5);
in_R(n,1) = isreal(Staten{n,1});
end
% Sommige coefficienten zijn heel klein <10^−20 in plaats van 0, komt door % het eigenvectoren vinden van matlab
% check of de staten reeel zijn
if sum(in_R) == 625
    disp('Alle eigenstaten hebben reële coefficienten')
else
    disp('Er zijn staten met complexe coefficienten')
end
% Normerings check en orhtogonaliteits check
Norm_check = zeros(625,625);
for i = 1:625
    for j = 1:625
        Norm_check(i,j) = sum(sum(sum(sum(Staten{i,1}.*Staten{j,1}))));
    end
end
% We zien dat alle staten goed genormeerd zijn, want inproduct met zichzelf is 1 % We zien ook dat het inproduct tussen verschillende staten niet altijd 1 % is, maar het is heel klein < 10^−15 en dit is verwaarloosbaar. % Het is niet precies 0, omdat matlab het getal moet opslaan en dan moet % hij het afronden op een aantal decimalen en daarom hebben we deze kleine 'fout'

% Stroomformule (Voor nieuwe plaatjes run alleen dit deel, je wilt niet weer de ham uiterrekenen en matrizes maken enzo)
% Tip van de STM boven atoom 1 of 2 of 3 of 4 (want we hebben maar 4 atomen)
Tip = 1;
% Voltage
Vmin = -5*10^(-4); % Begin voltage
Vmax = 5*10^(-4); % Eind voltage
deltaV = 10^(-6); % Stapgrootte
V = (Vmin:deltaV:Vmax); % van Vmin tot Vmax met stapjes van deltaV
% Temperatuur in kelvin
T = 1; % [K]
% Constanten
\[ kb = 1.38 \times 10^{-23} \text{ J/K} \]
\[ e = 1.602 \times 10^{-19} \text{ C} = \text{J/V} = \text{A\cdot s} \]
\[ h = 6.626 \times 10^{-34} \text{ J s} \]
\[ G_s = \frac{2e}{h} \text{ C/J s} = \frac{1}{\text{A/J}} \]

% i is de teller van M
% j is de teller van M'
\[ \delta = \text{zeros}(625,625) \text{ en } \delta = E_{M'} - E_M \]
\[ S_{+M} = \text{cell}(625,1) \]
\[ S_{-M} = \text{cell}(625,1) \]
\[ F = \text{cell}(625,625) \text{ % Het ding met de voltages en e macht enzo.} \]

\textbf{for i = 1:625}
\textbf{for j = 1:625}
\[ \delta(i,j) = (Energie(j,1) - Energie(i,1)) \]
\[ F(i,j) = (e \times V - 2 \times \delta(i,j)) / (1 - \exp(-2 \times \beta \times (e \times V - 2 \times \delta(i,j))) + (e \times V + 2 \times \delta(i,j)) / (1 - \exp(2 \times \beta \times (e \times V + 2 \times \delta(i,j)))) ; \]
\textbf{end}
\textbf{end}

% Bereken de spin operator toegepast op het atoom waar de tip boven staat,
% daarom ook alle ifs om te checken welke operator gebruikt moet worden
\textbf{if Tip == 1}
\textbf{for n = 1:625}
\textbf{for i = 1:5}
\textbf{for j = 1:5}
\textbf{for s = 1:5}
\textbf{for d = 1:5}
\textbf{if i < 5}
\[ S_{+M}(n,1)(i+1,j,s,d) = \sqrt{6 - (i-3)*(i-3+1)} \times \text{Staten}\{n,1\}(i,j,s,d) ; \]
\textbf{end}
\textbf{if i > 1}
\[ S_{-M}(n,1)(i-1,j,s,d) = \sqrt{6 - (i-3)*(i-3-1)} \times \text{Staten}\{n,1\}(i,j,s,d) ; \]
\textbf{end}
\[ S_{z-M}(n,1)(i,j,s,d) = (i-3) \times \text{Staten}\{n,1\}(i,j,s,d) ; \]
\textbf{end}
\textbf{end}
\textbf{end}
\textbf{end}
\textbf{end}
\textbf{end}
\textbf{elseif Tip == 2}
\textbf{for n = 1:625}
\textbf{for i = 1:5}
\textbf{for j = 1:5}
\textbf{for s = 1:5}
\textbf{for d = 1:5}
\textbf{if j < 5}
\[ S_{+M}(n,1)(i,j+1,s,d) = \sqrt{6 - (j-3)*(j-3+1)} \times \text{Staten}\{n,1\}(i,j,s,d) ; \]
\textbf{end}
\textbf{end}
\textbf{end}
\textbf{end}
\textbf{end}
\textbf{end}
\begin{verbatim}
end
if j > 5
    S_min_M{n,1}(i,j-1,s,d) = sqrt(6 - (j-3)*(j-3-1))*Staten
    {n,1}(i,j,s,d);
end
S_z_M{n,1}(i,j,s,d) = (j-3)*Staten{n,1}(i,j,s,d);
end
end
end
end
else if Tip == 3
    for n = 1:625
        for i = 1:5
            for j = 1:5
                for s = 1:5
                    for d = 1:5
                        if s < 5
                            S_plus_M{n,1}(i,j,s+1,d) = sqrt(6 - (s-3)*(s-3+1))*
                            Staten{n,1}(i,j,s,d);
                        end
                        if s > 1
                            S_min_M{n,1}(i,j,s-1,d) = sqrt(6 - (s-3)*(s-3-1))*Staten
                            {n,1}(i,j,s,d);
                        end
                        S_z_M{n,1}(i,j,s,d) = (s-3)*Staten{n,1}(i,j,s,d);
                    end
                end
            end
        end
    end
else if Tip == 4
    for n = 1:625
        for i = 1:5
            for j = 1:5
                for s = 1:5
                    for d = 1:5
                        if d < 5
                            S_plus_M{n,1}(i,j,s,d+1) = sqrt(6 - (d-3)*(d-3+1))*
                            Staten{n,1}(i,j,s,d);
                        end
                        if d > 1
                            S_min_M{n,1}(i,j,s,d-1) = sqrt(6 - (d-3)*(d-3-1))*Staten
                            {n,1}(i,j,s,d);
                        end
                        S_z_M{n,1}(i,j,s,d) = (d-3)*Staten{n,1}(i,j,s,d);
                    end
                end
            end
        end
    end
else
disp('Vul 1 of 2 of 3 of 4 in bij Tip, want we hebben maar 4 atomen.');
end
boltz_som = 0;
\end{verbatim}
for i = 1:625
    boltz_som = boltz_som + exp(-beta*Energie(i,1));
end
probability = zeros(625,1);
for i = 1:625
    probability(i,1) = exp(-beta*Energie(i,1))/boltz_som;
end

% Bereken de stroom
I = zeros(1,length(V));
for i = 1:625
    for j = 1:625
        I = I + Gs*(1/2)*(exp(-beta*Energie(i,1))/boltz_som)*(sum(sum(sum(sum(Staten{i,1}.*S_plus_M{j,1}))))^2)*F{i,j};
        I = I + Gs*(1/2)*(exp(-beta*Energie(i,1))/boltz_som)*(sum(sum(sum(sum(Staten{i,1}.*S_min_M{j,1}))))^2)*F{i,j};
        I = I + Gs*(exp(-beta*Energie(i,1))/boltz_som)*(sum(sum(sum(sum(Staten{i,1}.*S_z_M{j,1}))))^2)*F{i,j};
    end
end

% Bereken de afgeleiden
dIdV = diff(I)/deltaV; % Eerste afgeleide
d2IdV2 = diff(dIdV)/deltaV; % Tweede afgeleide

% Maak een plaatje
figure(1)
hold on
plot(V,I,'.')
hold off
xlabel('Voltage (V)')
ylabel('Stroom (A)')
title('IV plot van 4 deeltjes op een rij')

% Maak een plaatje van dIdV
figure(2)
hold on
plot(V(:,1:length(dIdV)),dIdV,'.')
hold off
xlabel('Voltage (V)')
ylabel('Eerste afgeleide van de stroom (A/V)')
title('dI/dV plot van 4 deeltjes op een rij')

% Maak een plaatje van d2IdV2
figure(3)
hold on
plot(V(:,1:length(d2IdV2)),d2IdV2,'.')
hold off
xlabel('Voltage (V)')
ylabel('Tweede afgeleide van de stroom (A/V^2)')
title('d^2I/dV^2 plot van 4 deeltjes op een rij')

% P_M grond toestand approximatie
i_g = find(Energie == min(Energie));
Grond = Staten{i_g,1}; % Dit is de grondtoestand == de toestand met de laagste energie
% Find vindt de indices waarvoor geld dat Energie_check = zijn minimum, dus zo
vinden we de grondtoestand

% Bereken de stroom met de P,M approximatie
I_grond = zeros(1,length(V));
for j = 1:625
    I_grond = I_grond + Gs*(1/2)*((sum(sum(sum(sum(Grond.*S_plus_M(j,1)))))^2)*F{i_g,j});
    I_grond = I_grond + Gs*(1/2)*((sum(sum(sum(sum(Grond.*S_min_M(j,1)))))^2)*F{i_g,j});
    I_grond = I_grond + Gs*((sum(sum(sum(sum(Grond.*S_z_M(j,1)))))^2)*F{i_g,j});
end

% Bereken de afgeleiden
dIgdV = diff(I_grond)/deltaV; % Eerste afgeleide
d2lgdV2 = diff(diff(I_grond))/deltaV; % Tweede afgeleide

% Maak een plaatje
figure(4)
hold on
plot(V,I_grond,'. ')
hold off
xlabel('Voltage (V)')
ylabel('Stroom (A)')
title('IV plot van 4 deeltjes op een rij, met P,M approximatie')

% Maak een plaatje van dIdV
figure(5)
hold on
plot(V(:,1:length(dIgdV)),dIgdV,'. ')
hold off
xlabel('Voltage (V)')
ylabel('Eerste afgeleide van de stroom (A/V)')
title('dI/dV plot van 4 deeltjes op een rij, met P,M approximatie')

% Maak een plaatje van d2IdV2
figure(6)
hold on
plot(V(:,1:length(d2lgdV2)),d2lgdV2,'. ')
hold off
xlabel('Voltage (V)')
ylabel('Tweede afgeleide van de stroom (A/V^2)')
title('d^2I/dV^2 plot van 4 deeltjes op een rij, met P,M approximatie')