Investigation of the magnetic phase diagram of the 2D ferrimagnetic $J_1$–$J_2$ model

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Abstract

The zero-temperature phase diagram and spin dynamics of the 2D ferrimagnetic $J_1$–$J_2$ model with $(S_1, S_2) = (1/2, 1)$ are investigated using the time-dependent cluster mean-field theory (t-CMFT). The t-CMFT enables the investigation of the quantum-mechanical as well as semi-classical phase diagram and spin dynamics by control of the entanglement. For the characterization of the ferrimagnetic system, the magnetization, the energy per atom, the cluster quantum states and the von Neumann entropy have been determined.

Keywords: ferrimagnet, frustration, time-dependent cluster mean field theory, Gisin Schrödinger equation

(Some figures may appear in colour only in the online journal)

1. Introduction

Magnetism is one of the oldest recognized material properties. In antique times loadstone, a naturally occurring magnetized ore of iron, was used in primitive compasses. Today, magnetism plays a role in data storage and information technology, as well as for logic devices. Therefore, magnetism is essential for the necessary trappings of modernity. With this in mind, scientists have pursued the discovery of materials with entirely new magnetic behaviors or states of matter with unique properties and potential benefits. One new material class suggested by Anderson is quantum spin liquids (QSLs) [1]. Anderson also suggested that low spatial dimension, low spin, and high frustration are the three main factors that lead to exotic spin liquid ground states [2]. One system which bears all three the three factors mentioned by Anderson is the two-dimensional $J_1$–$J_2$ model on a square lattice with antiferromagnetic nearest $J_1$ and next-nearest $J_2$ exchange interaction. Depending on the ratio of $J_2/J_1$, this model shows different ground states. In the classical $J_1$–$J_2$ model, there are two ground states the classical Néel state with a checkerboard-like spin configuration and the row-wise antiferromagnet. The $J_1$–$J_2$ model is the Néel state if the nearest-neighbor exchange $J_1$ is dominating and favors the row-wise antiferromagnet in the case of dominating next-nearest neighbor exchange $J_2$. If entanglement is taken into account (quantum-mechanical $J_1$–$J_2$ model) a third phase occurs between the Néel state and the row-wise antiferromagnet. This phase is most likely a resonating valence bond state (RVB), and therefore of relevance for the description of materials showing high-temperature superconductivity [3–6].

While the antiferromagnetic $J_1$–$J_2$ model has attracted much attention [7–18], the ferrimagnetic $J_1$–$J_2$ model has got less attention [19–21]. That does not mean that the investigation of ferrimagnets, in general, is not an active field. The contrary is the case [22–29]. In this publication, we use with the time-dependent cluster mean-field theory (t-CMFT) a quantum-mechanical method to describe the ferrimagnetic $J_1$–$J_2$ model. The idea behind the cluster mean-field theory (CMFT) is that most of the quantum spin systems contain too many spins to be treated precisely. Within the CMFT, the system is divided into smaller parts (clusters), which are treated exact with a quantum-mechanical method, e.g., exact diagonalization (ED). The clusters itself are connected by interactions where one of the two interacting spins is replaced by its spin expectation value (mean-field coupling). Ren et al [30] have shown that the CMFT...
shows a remarkable good agreement with other calculation methods, e.g., Exact Diagonalization (ED) ED [31], Density Matrix Renormalization Group Algorithm (DMRG) [32], or variational Quantum Monte Carlo (QMC) [33]. The only discrepancy is the fact that the magnetization within that theory is slightly too high. This can be explained by the couplings between the clusters, which act on the spins as effective fields stabilizing the system. Moreover, entanglement and, therefore, quantum fluctuations are restricted to the clusters. Spins of different clusters are not entangled. Within this publication, the time-dependence is taken into account by solving the modified Gisin–Schrödinger equation [34–37]. Therefore, the t-CMFT can calculate the ferrimagnetic $J_1–J_2$ model’s ground state configuration and its dynamics. An essential aspect of the t-CMFT is that in the limit of a single spin cluster, the spin dynamics become semi-classical [38]. The reason is simple. Between the clusters, there is no entanglement; entanglement only occurs within the clusters. In the case of a single-spin cluster ($1 \times 1$), there no second spin the cluster spin can entangle with. This means there is no entanglement in the single-spin cluster. Moreover, because the clusters do not entangle, there is no entanglement at all. Without entanglement, the main criteria for quantum spin dynamics is gone. Therefore the phase diagram and the dynamics are (semi-) classical. This publication uses this circumstance to investigate the (semi-) classical and quantum-mechanical phase diagram of the frustrated two-dimensional ferrimagnetic $J_1–J_2$ model.

The publication is organized as follows. The next section describes the ferrimagnetic $J_1–J_2$ model and the t-CMFT. After this section, the results are presented: the calculated phase diagram within the semi-classical limit of the single-spin cluster, followed by the calculations using 2 × 2 clusters. The later calculation results in the quantum phase diagram. The publication finishes with a summary.

2. Model

The investigated system within this publication is the two-dimensional ferrimagnetic $J_1–J_2$ model. The lattice is simple cubic, and the spins arranged in a checkerboard-like configuration. The spin operators $\vec{\sigma}_n$ and $\vec{S}_n$ corresponding to the spin quantum numbers $S = 1/2$ and $S = 1$ describe the magnetic moments.

As in the case of classical micromagnetism, where the spin dynamics and the ground state configurations are investigated by solving the Landau–Lifshitz–Gilbert (LLG) equation, the zero-temperature spin dynamics and the phase diagram are investigated by solving the modified Gisin–Schrödinger equation

$$i\hbar(1 + \alpha^2) \frac{d}{dt} |\Psi\rangle = H |\Psi\rangle - i\alpha (H - \langle H \rangle) |\Psi\rangle.$$  

(1)

In the $\alpha \to 0$ limit, this differential equation becomes the known time-dependent Schrödinger equation. However, for $\alpha > 0$ this differential equation describes the evolution of the quantum state $|\Psi\rangle$ with energy dissipation and relaxation into an eigenstate of the Hamilton operator $H$. Thereby, the trajectory of the spin expectation value of a single spin $\langle \vec{S} \rangle$ is identical to the trajectory of a classical spin $\vec{S}$ solved by the LLG equation. Of course with entanglement, the trajectories differ.

The Hamilton operator of the ferrimagnetic system is given by

$$\hat{H} = J_1 \sum_{\langle i,m \rangle} \vec{\sigma}_i \cdot \vec{S}_m + J_2 \sum_{\langle \langle i,m \rangle \rangle} \left( \vec{\sigma}_i \cdot \vec{S}_m + \vec{S}_i \cdot \vec{\sigma}_m \right).$$  

(2)

The first sum describes the antiferromagnetic $J_1 > 0$ exchange interaction between nearest-neighbor spins. The second sum describes the antiferromagnetic $J_2 > 0$ exchange interaction between the next-nearest neighbor spins.

It is caused by the fact that the matrix corresponding to $H$ increases very fast with an increasing number of spins; the treatment of the ferrimagnet by solving the modified Gisin–Schrödinger equation is only possible in the case of a small number of spins, e.g., $N = 20$. To overcome that limit, the system is divided into smaller parts that can be treated precisely. These are the clusters in the t-CMFT. The interactions within these clusters are described by the Hamilton operator $H_0$, in the following referred to as $H_0$. $H_1$ describes the connection between the clusters

$$H_1 = J_1 \sum_{\langle i,m \rangle} \left( \vec{\sigma}_i \cdot \langle \vec{S}_m \rangle + \vec{S}_i \cdot \langle \vec{\sigma}_m \rangle \right)$$

$$+ J_2 \sum_{\langle \langle i,m \rangle \rangle} \left( \vec{\sigma}_i \cdot \langle \vec{\sigma}_m \rangle + \vec{S}_i \cdot \langle \vec{S}_m \rangle \right).$$  

(3)

This Hamilton operator contains the same exchange interactions as $H_0$. However, now the interacting spins belong to different clusters. Moreover, the spins inside the cluster do not see the neighboring clusters’ spins but their effective fields (mean-field interaction). Finally, the Hamilton operator $H$, which is taken into account within the t-CMFT, is given by $H = H_0 + H_1$.

The clusters used within the t-CMFT are single-spin ($1 \times 1$) and quadratic $2 \times 2$ clusters. Larger $n \times m$ clusters, with $n, m \in \mathbb{Z}$, are in principle possible. However, the $2 \times 2$ clusters give already a fair description without using a supercomputer [30]. In the described ferrimagnet, one $2 \times 2$ cluster contains two $S = 1/2$ spins and two $S = 1$ spins ordered in a checkerboard structure. The single-spin cluster contains only one spin belonging to the $S = 1/2$ or the $S = 1$ sublattice. Additionally, it should be clear that in the single spin limit, the Hamilton operator $H_0$ is obsolete because there is no second spin within the cluster. In this limit, $H_1$ is still valid and describes the exchange interactions between the single-spin clusters. However, as already mentioned, $H_1$ does not allow entanglement. Also mentioned, the spins inside the clusters do not see the other clusters’ spins, only their effective fields. This means especially that the spins do not see each other. Each spin only experiences an effective field. The two facts, no entanglement, and the spins only experience effective fields are similar to the classical description. Therefore, we have to expect a classical behavior in the limit of single-spin clusters.

The calculations have been performed for quadratic systems with three by three $2 \times 2$ clusters. This is in total eighteen
S = 1/2 and eighteen S = 1 spins. In the case of single-spin clusters, the system has the same dimension, 36 spins, where 18 spins belong to the S = 1/2 and 18 to the S = 1 sublattice. For the 2 × 2 and the single-spin cluster description, periodic boundary conditions have been considered. The calculations themselves have been performed as follows. Starting from an initial configuration given by the quantum state |Ψ⟩, the system relaxes into a stable configuration. This configuration is not necessarily the ground state. However, with the knowledge of the phase diagram of the antiferromagnetic J₁−J₂ model, some of the configurations can be guessed, and the corresponding |Ψ⟩ can be chosen.

3. Results

3.1. Semi-classical phase diagram

This subsection presents the results of the semi-classical calculations using single spin 1 × 1 clusters. To characterize the phase diagram, the energy per atom and the magnetization have been calculated. Moreover, the corresponding quantum states have been analyzed.

Figure 1 shows the energy per atom plotted over the ratio J₂/J₁. The system’s three ground states highlighted in different colors. This figure can be seen as the phase diagram of the system. The filled symbols correspond to the ground state energies and the open symbols to excited states. Not surprisingly, the ground state for J₂ = 0 is the Néel state (see figure 2(a)). The energy of the Néel state increases linearly with increasing J₂. The reason is that with J₂/J₁ ̸= 0, the system is frustrated. The Néel state is the ground state until J₂/J₁ = 0.25. For larger J₂ values, the Néel state is still stable but no longer the ground state. At J₂/J₁ = 0.25, the ferrimagnet reacts to the frustration and starts canting. Between J₂/J₁ = 0.25 and J₂/J₁ = 0.5 the canting state is the ground state. Within this state, the S = 1 spin rotate clockwise, respectively, counter-clockwise against the S = 1/2 spins (see figure 2(b)). The pitch angles θ can be calculated by minimizing the system energy. As a result of such a calculation, one gets θ = acos(J₁/4J₂). From this formula, it becomes clear that there can be no canting for J₂/J₁ < 0.25. For J₂/J₁ < 0.25, one gets x = J₁/4J₂ > 1. However, the arcus cosine acos(x) is only defined for x values between −1 and 1.

At J₂/J₁ = 0.5 a spin-flop transition occurs, and the ground state becomes the collinear configuration (see figure 2(c)). J₂/J₁ = 0.5 is also the value in the classical antiferromagnetic J₁−J₂ model where the ground state changes from Néel to the row-wise antiferromagnet, which is a special form of the collinear state. Before the spin-flop transition, the spins within the two sublattices, S = 1/2 and S = 1, show parallel alignment. After the spin-flop transition, the spins in the sublattices are antiferromagnetically ordered. Therefore, the contribution of the nearest neighbor exchange Σ_jn ⟨Ψ|J_j n·Sn|Ψ⟩ to the energy E = ⟨Ψ|H|Ψ⟩ is zero. As a result, the two sublattices decouple and can be twisted against each other without a change of energy.

Figure 3 shows the pitch angles found during the calculations. As mentioned, for J₂/J₁ ≤ 0.25, the ground state is the collinear Néel state. Therefore, all θ are zero. In between J₂/J₁ = 0.25 and J₂/J₁ = 0.5, the pitch angles increase as a result of the canting. Please note that the S = 1 spins rotate clockwise, respectively, counter-clockwise against the S = 1/2 spins, but the S = 1/2 spins do not twist. Therefore, the angles between two S = 1/2 spins θ+1 −1 remain zero until J₂/J₁ = 0.5, while the angles between different spins θ+1 −1 change follow the law θ+1 −1 = acos(J₁/4J₂). As a result of the clockwise, respectively, counter-clockwise rotation, the angles θ+1,1 between two S = 1 spins are two times the pitch angle θ+1 −1. At J₂/J₁ = 0.5, the system undergoes a spin-flop transition. As a result, all the spins within the two sublattices are antiparallel. This means the pitch angles θ+1 −1 and θ−1,1 are both equal to 180°. The pitch angle between the two sublattices θ−1,1 has no further meaning anymore. Indeed, the simulations show random θ−1,1 values (not shown in figure 3).

Figure 1. Energy per atom as a function of J₂/J₁. Filled symbols refer to ground state energies, open symbols excited states.

Figure 2. Ground states: (a) Néel state, (b) canting state, and (c) collinear state. The circles as markings for the lattice sites refer to the S = 1/2 spins, the squares to the S = 1 spins. Excited state: (d) S1-Néel state.
Even if the results are equal to what we can expect from a classical description of the ferrimagnet, the calculations are entirely quantum-mechanical. The reason has already been mentioned in the introduction. Nevertheless, a few more comments. In the single-spin cluster calculation, the Hamilton operator \( H \) contains only the mean-field contribution \( H_1 \). This enables us to write \( H \) as a Zeeman term Hamilton operator

\[
\hat{H} = \sum_n \vec{\sigma}_n \cdot \vec{B}_n + \sum_k \vec{S}_k \cdot \vec{B}_k = \sum_n \hat{h}_n^{(1)} + \sum_k \hat{h}_k^{(1)},
\]

with

\[
\vec{B}_n = J_1 \sum_m (\vec{S}_m) + J_2 \sum_m (\vec{\sigma}_m)
\]

and

\[
\vec{B}_k = J_1 \sum_m (\vec{\sigma}_m) + J_2 \sum_m (\vec{S}_m).
\]

\( \vec{B}_n \) and \( \vec{B}_k \) can be seen as effective fields acting on one spin only. This means the spins only see the effective fields and not the other spins, similar to the description using the LLG equation. \( \hat{h}_n^{(1)} \) and \( \hat{h}_k^{(1)} \) are single spin Hamilton operators. With these Hamilton operators, the modified Gisin–Schrödinger equation becomes a set of independent differential equations

\[
\mathrm{i} \hbar (1 + \alpha^2) \frac{d}{dt} |\psi_n^{(h)}\rangle = \hat{h}_n^{(0)} |\psi_n^{(0)}\rangle - \mathrm{i} \alpha (\hat{H} - \langle \hat{h}_n^{(0)} \rangle) |\psi_n^{(0)}\rangle, \tag{4}
\]

one differential equation for each single-spin cluster state \( |\psi_n^{(h)}\rangle; \eta = \frac{1}{2}, \) respectively, \( \eta = 1 \). The quantum state of the whole system is given as product state of these quantum states \( |\Psi\rangle = \bigotimes_n |\psi_n^{(h)}\rangle \). The cluster states \( |\psi_n^{(h)}\rangle \) itself are the spin coherent states

\[
|\psi_n^{(h)}\rangle = \cos \frac{\theta_n}{2} |\uparrow\rangle + \mathrm{e}^{\mathrm{i} \phi_n} \sin \frac{\theta_n}{2} |\downarrow\rangle.
\]

in the case of \( S = 1/2 \), respectively, for \( S = 1 \)

\[
|\psi_n^{(1)}\rangle = \cos \frac{\theta_n}{2} |\uparrow\rangle + \sqrt{2} \mathrm{e}^{\mathrm{i} \phi_n} \cos \frac{\theta_n}{2} |0\rangle + \mathrm{e}^{2\mathrm{i} \phi_n} \sin \frac{\theta_n}{2} |\downarrow\rangle.
\]

The angles \( \theta_n \) and \( \phi_n \) within these quantum states refer to the spherical coordinates at lattice site \( n \), and are related to the pitch angles shown before.

In the case of \( 2 \times 2 \) clusters, the system quantum state \( |\Psi\rangle \) is also a product state

\[
|\Psi\rangle = \bigotimes_n |\psi_n\rangle. \tag{5}
\]

However, the \( |\psi_n\rangle \) now characterize the \( 2 \times 2 \) clusters and are no longer single spin states.

\[
|\psi_n\rangle = \sum_r a_r^{(n)} |r\rangle \tag{6}
\]

describes the \( 2 \times 2 \) cluster states \( |\psi_n\rangle \). The \( a_r^{(n)} = \langle r | \psi_n \rangle \) are the expansion coefficients and \( |r\rangle \) a complete set of orthogonal basis states, e.g., the Zeeman basis. In the description using \( 2 \times 2 \) clusters, the basis states are given as \( |r\rangle = |m_2 m_1 m_1 m_1\rangle \). Therefore, \( m_2 \) and \( m_1 \) are magnetic quantum numbers, where \( m_1 \) is either spin up \( \uparrow \) or down \( \downarrow \) and \( m_1 \) up \( \uparrow \), zero \( 0 \), or down \( \downarrow \).

### 3.2. Quantum phase diagram

This subsection presents the results of the calculations using quadratic four spin clusters (\( 2 \times 2 \) spin clusters). Describing the \( 2 \times 2 \) spin clusters in a full quantum mechanical manner allows us to expect a quantum mechanical phase diagram. When increasing the cluster size from a single spin to \( 2 \times 2 \), the Hamilton operator \( H_0 \) is no longer obsolete, and the dominating contribution. Therefore, we can expect entanglement and, as a result, quantum fluctuations. The result will be a quantum mechanical phase diagram.

Before we get to the phase diagram itself, there is one crucial point that has to be mentioned first. The relaxation term of the modified Gisin–Schrödinger equation (second term on the right-hand side of equation (1)) tends to relax towards a quantum state, which is an eigenstate of the given Hamiltonian. This state is stable. However, there is no warranty that the found quantum state is a ground state. Moreover, when starting with a single basis state \( |r\rangle \) of the Zeeman basis, the relaxation process will end with a quantum state described only by basis states belonging to the same sub-Hilbert space as the initial basis state. The problem is that all collinear states can be described by a minimal basis set, meaning just by basis vectors belonging to one of the sub-Hilbert space. This statement is no longer valid for non-collinear states. For the description of non-collinear quantum states, all basis vectors of the Zeeman basis are needed. The critical point is that non-collinear states are described by superpositions of basis states belonging to different sub-Hilbert spaces. Therefore, to get the correct phase diagram, we have to ensure that all basis states are included in the calculation.

As for the semi-classical phase diagram, we characterize the system by its energy and the magnetization. Figure 4 shows the energy per atom as a function of \( J_2/J_1 \). Concerning the semi-classical description, the ground states are the same. For small \( J_2/J_1 \) values, the Néel state is the ground state. In the intermediate regime \( 0.27 \leq J_2/J_1 \leq 0.5 \), a non-collinear phase, the canted state similar to the semi-classical phase diagram, occurs. For \( 0.5 \leq J_2/J_1 \), the collinear state

![Figure 3. Pitch angles θ over J2/J1. θ(J2/J1 = 0.5) refers to the heavy side step function.](image)
is the ground state. The energies are different concerning the semi-classical phase diagram. However, the ground states and the types of phase transitions between these phases (all of the second-order) are the same. Also, the critical values where the phase transitions occur are nearly identical. This is in agreement with the former study of Ivanov et al [19]. However, there is one difference between the former study and this one. Ivanov et al predict another ground state, the S1-Néel state. This state is characterized by the fact that the magnetization of the $S = 1/2$ sublattice is gone, and the remaining spins of the $S = 1$ sublattice show Néel order (see figure 2(d)). In Ivanov et al’s calculations, this phase is the ground state for a small range of $J_2/J_1$ values: $0.46 \leq J_2/J_1 \leq 0.5$. Within this study, the S1-Néel is always a excited state, also in the small range predicted by Ivanov et al. Within that range, the energies of the canted state and the S1-Néel state are very close.

The second physical value which we use to characterize the system is the magnetization. Figure 5 shows the absolute values of the magnetization as a function of $J_2/J_1$. While in the semi-classical calculation, the absolute values are constant, we see a reduction of the magnetization towards $J_2/J_1 = 0.5$ due to quantum fluctuations. However, the magnetization does not drop down to zero. From former studies of the antiferromagnetic $J_1$–$J_2$ model, it is clear that the CMFT, in comparison to other methods, overestimates the magnetization. The reason is the confinement of quantum fluctuations inside the clusters. There is no correlation between the clusters. An increase of the cluster size will increase the fluctuations’ correlation length and let the magnetization decrease. On the other hand, an increase in cluster size increases the numerical effort, especially the requested memory, to store the vast matrices. On the other hand, the overestimation of the magnetization is the only problem and not critical. As already mentioned, Ren et al [30] have shown that the CMFT can be used to reproduce the correct phase diagram of the antiferromagnetic $J_1$–$J_2$ model. All phases, the phase transitions, and the critical values are in agreement with other calculations [31–33]. Only the magnetization is compared to other methods, e.g., QMC or ED, too high.

Zero magnetization can be seen for the S1-Néel and the QSL state. Both states belong to the excited states. As already said in the S1-Néel state case, the magnetization of the $S = 1/2$ sublattice is gone, while the $S = 1$ sublattice still shows magnetic order. In the case of the QSL state, also this order is gone. This state shows no magnetic order at all. When looking at the quantum states, respectively, the von Neumann entropy, it seems that this state is similar to the RVB state in the antiferromagnetic $J_1$–$J_2$ model.

We have also analyzed the pitch angle $\theta$ between the $S = 1/2$ and $S = 1$ spins to characterize the canted state. As in the case of the semi-classical calculations, the $S = 1/2$ spins do not change their direction while the $S = 1$ spins rotate in opposite directions (see figure 2(b)). Figure 6 shows the pitch angle $\theta$ as a function of $J_2/J_1$ in the range of the canted state. The symbols correspond to the quantum mechanical $2 \times 2$ spin cluster calculation results, while the dashed line corresponds to the semi-classical single spin cluster calculation.

As mentioned at the beginning of this subsection, when dealing with collinear states and using the Zeeman basis, we can restrict us to the use of basis vectors of one subspace of the Hilbert space. For instance, the Néel state $\uparrow\downarrow\uparrow\downarrow$ corresponds to
the \( m = +1/2 - 1 + 1/2 - 1 = -1 \) subspace, while the row-wise antiferromagnetic state (the collinear state with \( \theta_{S1} = 0° \) or \( 180° \)) \( \uparrow\uparrow\downarrow\downarrow \) corresponds to the \( m = +1/2 + 1 - 1/2 - 1 = 0 \) subspace. The eigenstates of the Néel, respectively, row-wise antiferromagnet, are described by all basis states of the corresponding subspaces \( m = -1 \), respectively, \( m = 0 \). The magnetic order of these eigenstates is collinear and in the \( \pm z \) direction. Non-collinear states and a magnetic order not in the direction of the quantization axis, the \( \pm z \)-direction, requests superpositions of all basis states \( |r⟩ \). The latter point becomes clear if we think about the description of a single \( S = 1/2 \) spin using the Bloch sphere. As long as the spin is oriented in \( \pm z \)-direction the spin up \( |\uparrow⟩ \) or down \( |\downarrow⟩ \) state is enough. Any other orientation requests a superposition of \( |\uparrow⟩ \) and \( |\downarrow⟩ \). In principle, it is always recommended to ensure that the complete set of basis vectors. As already mentioned, when starting with a single basis state, e.g., with \( |\uparrow\uparrow\downarrow\downarrow⟩ \), we will probably find one or more eigenstates. In that case, the eigenstates: row-wise antiferromagnet (the collinear state with \( \theta_{S1} = 0° \) or \( 180° \)), S1-Néel state, and the QSL state. However, we will not see a non-collinear state as the canted state. This is an important point to keep in mind.

Nevertheless, the restriction to the basis states of one subspace can also be helpful. In that case, we restrict the number of basis states, respectively, the matrices’ size. That can help understand the underlying quantum states or calculate physical quantities like the von Neumann entropy \( S \).

Now, it is interesting to see what happens if we start the calculations with the basis state \( |\uparrow\uparrow\downarrow\downarrow⟩ \) and differ \( J_2/J_1 \). If \( J_2/J_1 = 1 \), this basis state will be dominating, and the corresponding eigenstate will show a row-wise antiferromagnetic order similar to this basis state. When lowering \( J_2/J_1 \), the eigenstate will change and undergoes two second-order phase transitions where the magnetic order will disappear. Figure 7 shows the magnetization of the three collinear eigenstates of the \( m = 0 \) subspace: row-wise antiferromagnet, S1-Néel state, and a non-magnetic QSL state. The stepwise reduction of the magnetization from row-wise antiferromagnet (ultimately ordered, even with reduced magnetization), over the S1-Néel state (magnetization of the \( S = 1/2 \) sublattice is zero) towards the QSL state with no magnetization is interesting. All these calculations are performed at zero temperature. Therefore, the reduction of magnetization is a follow of quantum fluctuations. To quantify entanglement’s strength leading to the quantum fluctuations, we calculate the von Neumann entropy. Figure 8 shows the normalized von Neumann entropy \( S/S_{max} \) as a function of \( J_2/J_1 \) for the \( S = 1/2 \) and the \( S = 1 \) sublattice. The von Neumann entropy is a measure for the strength of entanglement. In this case, the von Neumann entropy gives the strength of entanglement between the spins inside one cluster. The von Neumann entropy is calculated as follows: using the cluster quantum state \( |\psi_{n}⟩ \) of the \( n \)th cluster, the corresponding density matrix \( \rho_n = |\psi_{n}⟩⟨\psi_{n}| \) is formed. In the next step, all spins except one spin, either one of the two \( S = 1/2 \) or one of the two \( S = 1 \) spins in the \( 2 \times 2 \) cluster have been traced out. The results of this process are the reduced density matrices for a single \( S = 1/2 \) or \( S = 1 \) spin. This calculation has been done for all four spins within the cluster. However, caused by the system’s translational symmetry, all spins of the \( S = 1/2 \) and all spins of the \( S = 1 \) sublattice give the same result for the entropy. With the reduced density matrices, the von Neumann entropies can be calculated using the eigenvalues \( \lambda_i \) of these matrices: \( S = - \sum \lambda_i \log_2 \lambda_i \). Thereby, \( S_{max} = \log_2(2) \) is the maximal entropy the \( S = 1/2 \) spins can reach. The maximum for the \( S = 1 \) spins is \( S_{max} = \log_2(3) \). Figure 8 shows that the \( S = 1/2 \) spins are maximally entangled, while the entanglement of the \( S = 1 \) spins is approximately \( 89\% \) of their possible entanglement. This means the QSL state is highly entangled. The same is true for the \( S = 1/2 \) spins in the S1-Néel state and the RVB state in the antiferromagnetic \( J_1-J_2 \) model. Therefore, the QSL is most likely a RVB state. Please note, the entanglement is constant for all \( J_2 \) in the case of the QSL state. The row-wise antiferromagnet, as the Néel state show entanglement, however drastically reduced concerning the S1-Néel state, respectively, the QSL state. In the row-wise antiferromagnet case, the entropy increases with decreasing \( J_2/J_1 \) (starting from \( J_2/J_1 = 1 \)). In the case of the Néel state, the entropy increases with increasing \( J_2/J_1 \). These results are not surprising because the magnetization is related to entanglement. With increasing entanglement (increasing entropy \( S \)), the magnetization decreases. Thereby, the states with zero magnetization have mostly \( S = S_{max} \) and the states which show magnetization values close to one, respectively one-half, have \( S < S_{max} \).

The quantum states used to calculate the von Neumann entropy \( S \) are shown in figure 9. More precisely, figure 9 shows the expansion coefficients \( \psi_{n}^{\mu} \) corresponding to the basis vectors \( |r⟩ \) which construct the eigenstates \( |\psi_{n}⟩ \). These are the basis states of the \( m = 0 \) subspace of the \( 2 \times 2 \) spin cluster. Therefore, these basis states are just a minimal set and not the complete set of basis vectors. As already said, the result is collinear eigenstates with a magnetic order in \( \pm z \)-direction. Nevertheless, this figure gives an interesting view of the quantum states. For instance, the row-wise antiferromagnetic state is dominantly characterized by the one basis state, which looks identical to the resulting magnetic order. This basis state is not
three values of the expansion coefficients $\alpha$, is described by ten basis states. However, there are only two QSL state, the $S$-Néel state, the other basis states are described by the basis vectors of the sub-Hilbert space. When starting with one basis state is dominating, the magnetic order gets more complex, as can be seen in the case of the $S$-Néel state. Finally, the QSL state, in the case of $2 \times 2$ spin clusters, is described by ten basis states. However, there are only three values of the expansion coefficients $\alpha^{(n)}$. Another feature of the QSL state is that this eigenstate does not change with changing $J_2/J_1$ values. This feature does not occur for all other eigenstates. The other eigenstates change with changing $J_2/J_1$. Both features, no dependence on $J_2/J_1$ and description with only three, are identical to the RVB state in the antiferromagnetic $J_1-J_2$ model case. Of course, this statement is only valid if the description is done using the time-dependent cluster mean-field method with $2 \times 2$ clusters.

4. Summary

The phase diagram of the quantum-mechanical ferrimagnetic $J_1-J_2$ model has been investigated with the use of the t-CMFT. The calculations have been performed for two different cluster sizes. The phase diagram is identical to the ferrimagnet’s classical description in the limit of a single-spin cluster ($1 \times 1$). The reason is the missing entanglement in that description. If larger clusters are taken to perform the calculation, the situation changes. The phase diagram is now that of a quantum spin system. The difference is the occurrence of quantum fluctuations, which causes a reduction of the magnetization. In the case of the $2 \times 2$ clusters calculation, entanglement is included. Here, we have to keep in mind that entanglement and therefore the quantum fluctuations only appear inside the clusters. There is no correlation between the clusters. This spatial confinement is a disadvantage of the CMFT. However, calculations of the phase diagram of the antiferromagnetic $J_1-J_2$ model have shown that the phases, and the phase transitions are well reproduced [30]. Even the critical $J_2$’s have the expected values. The only physical quantity which shows a discrepancy is the magnetization of the magnetic phases. Other methods propose a lower magnetization. A cluster size scaling could be performed to prove the accuracy further and see the infinite system’s values. However, an increase of the cluster size means an exponential increase of the numerical effort and requests adequate computation equipment.

A few words about the ground states. The ground states itself are identical to the semi-classical calculation. Moreover, the semi-classical phase diagram is identical to the classical phase diagram. The quantum mechanical phase diagram is different due to the occurrence of quantum fluctuations. The quantum phase diagram agrees with Ivanov et al’s presented phase diagram, except at one point. Ivanov et al predict another ground state, the $S$-Néel state. This state has also been found with the aid of the t-CMFT. However, in our study, the $S$-Néel state does not occur as a ground state. It occurs as an excited state for all $J_2/J_1$ values. Within the small range where Ivanov et al expect the $S$-Néel state as the ground state, the energy difference between the canted state and the $S$-Néel state is small. If the $S$-state is a ground state within the small range of $0.46 \leq J_2/J_1 \leq 0.5$, then one possible explanation of the discrepancy could be the underestimation of the quantum fluctuation. On the other hand, there is also the possibility that the $S$-Néel state is not a ground state.

The third aspect which has been discussed is the aspect of calculations using a minimal basis set, meaning just basis vectors of one sub-Hilbert space. When starting with one basis state the relaxation term of the modified Gisin–Schrödinger equation tends to relax to an eigenstate within the same subspace and lower in energy. This eigenstate is not necessarily the ground state. However, in that case, definitively an eigenstate with collinear magnetic order and a spin orientation in the $\pm z$-direction (quantization axis). Such calculations are useful. For example, the situation that happens if we start with the basis state $|\uparrow\uparrow\downarrow\rangle$ has been discussed. Depending on the $J_2/J_1$ value one of the three eigenstates: row-wise antiferromagnet, $S$-Néel state and the QSL state can be found. All these eigenstates are described by the basis vectors of the $m = 0$ subspace. It has been shown that with decreasing $J_2/J_1$ values, the magnetic order stepwise decreases, and at the same time, the entanglement inside the clusters increases. A look at the eigenstates

**Figure 8.** Normalized von Neumann entropy $S/S_{\text{max}}$ as a function of $J_2/J_1$ for an $S = 1/2$ and $S = 1$ sublattice spin.

**Figure 9.** Expansion coefficients $\alpha^{(n)}$ as a function of $J_2/J_1$ for the QSL state, the $S$-Néel state, and the row-wise antiferromagnet.
shows that quantum states with less entanglement, respectively quantum fluctuations are mostly described by a dominating basis state, while in the case of a completely entangled quantum state, all basis states are participating in a not negligible manner. About the QSL state in the phase diagram is to say that the behavior (only three different values of the expansion coefficients $a_1$, and a von Neumann entropy which does not change with changing $J_2$) is identical to the RVB state of the antiferromagnetic $J_1$–$J_2$ model.

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