Exotic phases of matter in quantum magnets

A tensor networks tale

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Citation for published version (APA):
The square lattice bilinear-biquadratic Heisenberg model

Infinite projected entangled pair states simulations of the nearest-neighbor spin-1 bilinear-biquadratic Heisenberg model on the square lattice reveal an emergent Haldane phase in between the previously predicted antiferromagnetic and 3-sublattice $120^\circ$ magnetically ordered phases. This intermediate phase preserves spin-rotation and lattice-translation symmetry but breaks lattice-rotation symmetry, and it can be adiabatically connected to the Haldane phase of decoupled spin-1 chains. Moreover, we find an unexpected partially magnetic partially nematic phase in between the antiferroquadrupolar and ferromagnetic regions. Furthermore, we describe all observed phases and discuss the nature of the phase transitions involved.

The material presented in this chapter is based on Refs. [1, 2].

3.1 Introduction

The search for novel states of matter in quantum many-body systems is one of the most active areas in condensed matter physics. A fascinating example is the ground state of the spin-1 antiferromagnetic Heisenberg chain which, unlike the spin-1/2 chain, exhibits an energy gap, exponentially decaying spin-spin correlations, and gapless edge excitations in case of open boundaries. Thanks to Hal-
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dane’s pioneering work and conjecture that such a gapped state emerges in integer
Heisenberg spin chains in general [133,134], this phase has been named after him.
The Haldane phase also extends to related spin-1 models, such as weakly-coupled
spin-1 Heisenberg chains [135–139] realized in several quasi one-dimensional materials
[140–145], or the spin-1 bilinear-biquadratic Heisenberg (BBH) chain for $\theta$
in between $-\pi/4$ and $\pi/4$; see Eq. (3.1). More recently, the Haldane phase has
been understood as a simple example of a symmetry protected topological (SPT)
phase [146–149].

In this chapter, we study the spin-1 bilinear-biquadratic Heisenberg model on the
two-dimensional square lattice: a model that consists of spin-1 particles, one per
lattice site, that interact only with their nearest neighbors. The nearest-neighbor
two-particle interaction is a combination of two types of competing interactions:
the ordinary bilinear Heisenberg coupling, and the biquadratic coupling, which is
the Heisenberg coupling squared. The corresponding coupling constants appearing
in the Hamiltonian are commonly parametrized by an angle $\theta$:

$$H = \sum_{i,j} \cos(\theta) S_i \cdot S_j + \sin(\theta) (S_i \cdot S_j)^2,$$

(3.1)

where $S_i = (S_i^x, S_i^y, S_i^z)$ is the vector of spin-matrices\(^1\) for the spin-1 particle on
site $i$,

\[
S_i^x = \frac{1}{\sqrt{2}} \begin{pmatrix}
0 & 1 & 0 \\
1 & 0 & 1 \\
0 & 1 & 0
\end{pmatrix}, \quad S_i^y = \frac{i}{\sqrt{2}} \begin{pmatrix}
0 & -1 & 0 \\
1 & 0 & -1 \\
0 & 1 & 0
\end{pmatrix}, \quad \text{and}, \quad S_i^z = \begin{pmatrix}
1 & 0 & 0 \\
0 & 0 & 0 \\
0 & 0 & -1
\end{pmatrix},
\]

and the sum goes over all nearest-neighbor pairs. The biquadratic term can appear
as a second order correction in the expansion of the exchange interaction, in which
case it is small compared to the bilinear term. However, it has been argued
that a significant biquadratic interaction may exist. For example, the behavior
of the magnetic susceptibility of the one-dimensional material LiVGe\(_2\)O\(_6\) can be
explained by a significant biquadratic interaction [150], a suggested underlying
microscopic mechanism of which can be found in Ref. [151].

The BBH model on the square lattice has been subject to a lot of interest in re-
cent years [152–160], for a number of reasons. For one, it was proposed that the
nematic phases—which involve the breaking of spin-rotational symmetry while
preserving time-reversal symmetry [39]—of the spin-1 BBH model on the trian-
gular lattice could be related to the unusual behavior of NiGa\(_2\)S\(_4\) [154,156,161]
and Ba\(_3\)NiSb\(_2\)O\(_9\) [159,162,163]\(^2\). Second, at $\theta = \pi/4$, the model is equivalent
to the SU(3) Heisenberg model which can be experimentally realized using ultra-
cold fermionic atoms in optical lattices [164–167]. The latter has been shown to

\(^1\)Setting $\hbar = 1$.

\(^2\)In the so-called 6H-B phase.
exhibit 3-sublattice order on the square and triangular lattices [168, 169], and an important question concerns the stability of this phase away from the SU(3) symmetric point. Previous studies on the square lattice based on linear flavor-wave theory [158], exact diagonalization [158], and series expansion [160] predicted a direct transition between the AFM and the 3-sublattice phase for \( \theta \approx 0.2\pi \). However, the accurate study of this parameter regime remains very challenging because Quantum Monte Carlo suffers from the negative sign problem [153]. Finally, the spin-1 BBH model is also interesting from a theoretical perspective, because it is the most general lattice-translation, lattice-rotation and spin-rotation-symmetric model with nearest-neighbor interactions\(^3\).

In this chapter, we show that in between the antiferromagnetic (AFM) and the 3-sublattice 120° magnetically ordered (AFM3) phases an intermediate quantum paramagnetic phase emerges which preserves lattice-translation and spin-rotation symmetry, but breaks lattice-rotation symmetry. We identify this intermediate phase as the Haldane phase by showing that it can be adiabatically connected to the Haldane phase of decoupled spin-1 chains. This result at first appears surprising in view of the fact that for \( \theta = 0 \) already a small inter chain coupling \( J_y > J_y^c = 0.0436 \) [138] is sufficient to destabilize the Haldane phase. However, we show that with increasing \( \theta \) the critical inter chain coupling \( J_y^c(\theta) \) separating the Haldane phase from the AFM phase dramatically increases, and eventually reaches the isotropic two-dimensional limit.

Furthermore, we provide a complete study of the ground state phase diagram. Contrary to previous studies, we find the occurrence of a half nematic half magnetic ‘\( m = 1/2 \)’ phase that was previously predicted to appear only in the presence of an external magnetic field [158]. Additionally, we describe all phases and determine the nature of the corresponding phase transitions. The ground state phase diagram of the spin-1 BBH model according to our computations, which in itself is the main result of this chapter, can be found in Fig. 3.7.

This chapter is organized as follows. Section 3.2 sets the stage by discussing some of the relevant properties of spin-1 particles—in particular in the context of spin-nematic order—as well as the symmetries of the Hamiltonian, followed by an overview of previous studies of the BBH model in Section 3.3. We then proceed to discuss our own findings in Section 3.4, which includes a simple update study of the entire system, an in-depth full-update investigation of the appearance of the Haldane phase incorporating a simple update study of the anisotropic spin-1 BBH model, a full update study of all occurring phase transitions, and an analysis of the partially magnetic partially nematic \( m = 1/2 \) phase. Finally, we conclude with Section 3.5.

\(^3\)All higher powers of \( S_i \cdot S_j \) can be rewritten in terms of the bilinear and biquadratic terms.
3. The Square Lattice Bilinear-Biquadratic Heisenberg Model

3.2 The model

3.2.1 Spin-1 nematic order

Individual spin-1 particles can display more types of order than their spin-1/2 counterparts. For example, every spin-1/2 single-particle state—assuming it is a pure state, which we take all single-particle states in this subsection to be—has a fixed magnetization

\[ m = \sqrt{\langle S_x \rangle^2 + \langle S_y \rangle^2 + \langle S_z \rangle^2} \]

of exactly 1/2 (setting \( \hbar = 1 \)), and is fully described by its magnetic dipole moment \( \mathbf{S} = (S_x, S_y, S_z) \). This is a consequence of the fact that any spin-1/2 single-particle state can be obtained by applying a specific rotation to the up state in the z-basis (or any other reference state for that matter). By contrast, spin-1 particles can have a magnetization of anywhere between 0 and 1. Moreover, since spin-1 particles live in a higher-dimensional space, the dipole moment is not sufficient to fully describe a given spin-1 single-particle state, for which in addition also the quadrupole moment is required.

The quadrupole moment is measured by products of spin operators, which we can combine into the matrix \( T_{\alpha\beta} = S_\alpha S_\beta \), with \( \alpha, \beta \in \{x, y, z\} \). However, \( T \) contains more than just the quadrupole moment, as the anti-symmetric part of \( T \) is just the vector of spin operators itself due to the spin commutation relations. The remaining symmetric part of \( T \) can, because \( \text{Tr}(T) = S(S+1) = 2 \) is constant, be captured by five independent operators that can conveniently be organized into the vector

\[
Q := \begin{pmatrix}
(S_x)^2 - (S_y)^2 \\
\frac{1}{\sqrt{3}} \left[ 2(S_z)^2 - S(S+1) \right] \\
S_x S_y + S_y S_x \\
S_y S_z + S_z S_y \\
S_z S_x + S_x S_z
\end{pmatrix}
\]

(3.2)
of quadrupolar operators (following the conventions of Ref. [170]).

Typically, spin-1 single-particle states display both magnetic and quadrupolar order. However, when referring to a quadrupolar state, what we mean is a state that is solemnly described by \( Q \). In particular, we require that its magnetic dipole moment \( m \) is zero. An example of a quadrupolar state is the \( |0\rangle \) state in the \( S_z \)-basis. As can be checked directly: \( m = 0 \). Interestingly, even in the absence of magnetization, the \( |0\rangle \) state does break spin-rotation symmetry because \( \langle (S_z)^2 \rangle = 0 \), whereas the fluctuations in x and y-direction are non-zero: \( \langle (S_x)^2 \rangle = 1 = \langle (S_y)^2 \rangle \).

Classically, this state can be thought of as a dipole moment that fluctuates in the x-y plane in such a way that it is zero on average. We will picture it by a disc portraying the plane of fluctuations (see Fig. 3.1). A normal vector to the plane of fluctuations is called a director \( (\pm e_z \text{ in the case of } |0\rangle) \). Any quadrupolar single-particle state is fully described by the orientation of its directors.
3.2. The model

A convenient on-site basis for investigating quadrupolar order is the time-reversal invariant basis:

\[ |x\rangle = \frac{i}{\sqrt{2}} (|\uparrow\rangle - |\downarrow\rangle), \quad |y\rangle = \frac{1}{\sqrt{2}} (|\uparrow\rangle + |\downarrow\rangle), \quad |z\rangle = -i|0\rangle, \]

(3.3)

where \( \{|\uparrow\rangle, |0\rangle, |\downarrow\rangle\} \) is the standard \( S^z \)-eigenbasis. Invariance of the above vectors under the time-reversal operator \( T \), which is anti-unitary, follows immediately from the fact that \( T \) interchanges \( |\uparrow\rangle \) and \( |\downarrow\rangle \), and adds a sign to \( |0\rangle \): i.e. \( T |\uparrow\rangle = |\downarrow\rangle, T |\downarrow\rangle = |\uparrow\rangle \), and \( T |0\rangle = -|0\rangle \).

Any real linear combination of the above basis states \( \sum_{\alpha=x,y,z} u_\alpha |\alpha\rangle \) is also a quadrupolar state, with a director pointing along \( u \). More generally, an arbitrary spin-1 single-particle state can be expanded in the basis of (3.3) as \( |d\rangle = \sum_{\alpha=x,y,z} d_\alpha |\alpha\rangle \), where \( d \) has real and imaginary parts \( d = u + iv \). In terms of \( u \) and \( v \), the magnetic moment of \( |d\rangle \) is given by \( S = 2u \times v \). We can normalize the state and use global phase invariance to have \( u \) and \( v \) satisfy \( u^2 + v^2 = 1 \) and \( u \cdot v = 0 \). Assuming the last two equations hold, then so does the following: fully magnetized \( m = 1 \) states are precisely those for which \( u = v \), whereas non-magnetic \( m = 0 \) states correspond to \( u, v = (0,1) \) or \( (1,0) \). The latter case \( (m = 0) \) describes purely quadrupolar states with a director \( d \) pointing along the direction of whichever of \( u \) or \( v \) is non-zero. Whenever \( u \) and \( v \) are both non-zero but \textit{not} of equal magnitude, and consequently \( 0 < m < 1 \), the state is of mixed character: neither fully magnetic nor purely quadrupolar.

Because the basis of (3.3) is invariant under the anti-unitary time-reversal operator \( T \), we can immediately conclude that the time-reversal invariant single-particle states are precisely those for which the coefficients in the above basis do not change under \( T \), up to a global phase. Since \( T \) is anti-unitary, this means that the time-reversal invariant states are precisely those for which \( d \) is either completely real \( (v = 0) \), or purely imaginary \( (u = 0) \); i.e. \( |d\rangle \) is a quadrupolar state. (This is most obvious in the above gauge \( u \cdot v = 0 \), where \( u \) and \( v \) both being non-zero results in \( S \) being flipped under \( T \).) In other words, following Andreev’s definition [39] of a spin-nematic state as a state that breaks spin-rotational symmetry but preserves

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Figure 3.1: The \(|0\rangle\) quadrupolar state. The disc represents the magnetic dipole fluctuations in the \( x-y \) plane, and the dotted line represents a director perpendicular to the plane of fluctuations pointing along the \( z \)-axis.
time-reversal symmetry, we conclude that, for a single spin-1 particle, the notions of quadrupolar and spin-nematic are equivalent.

Comparing to nematic order in liquid crystals, which is the alignment of rod-like particles in a liquid, we observe that quadrupolar states have the exact same symmetry properties as rods. Indeed: two directors $\mathbf{d}$ and $-\mathbf{d}$ correspond to the same quantum state (they differ by an overall phase). Contrast this to magnetic states, where the magnetic moment is described by the magnetic dipole vector, which is certainly not equal to minus itself. More explicitly, the magnetic the $|\uparrow\rangle$ and the quadrupolar $|0\rangle$ states are invariant (the first up to a phase) under rotations about the $z$-axis, but, in addition, the $|0\rangle$ state is (up to a minus sign) also invariant under a $\pi$-rotation about any axis that lies in the $x-y$ plane. Finally, we speak of nematic order when neighboring directors order in space, similar to the spatial ordering of rods in liquid crystals.

### 3.2.2 High-symmetry points

It shall not come as a surprise to the reader that the biquadratic term $(\mathbf{S}_i \cdot \mathbf{S}_j)^2$ in the Hamiltonian (3.1), which involves on-site products of spin operators, is related to quadrupolar order. However, part of the biquadratic term also includes the ordinary spin matrices (the anti-symmetric part of $\mathbf{T}$ defined above), which are more naturally absorbed into the bilinear term. To better separate the two competing magnetic and quadrupolar interactions, let us rewrite the Hamiltonian in terms of $\mathbf{S}$ and $\mathbf{Q}$. Doing so yields

$$H = \sum_{\langle i,j \rangle} J_S(\theta) \mathbf{S}_i \cdot \mathbf{S}_j + J_Q(\theta) \mathbf{Q}_i \cdot \mathbf{Q}_j$$

(3.4)

up to an additive $\theta$-dependent constant of $4\sin(\theta)/3$ that is irrelevant for the remainder of this chapter. The spin and quadrupolar coupling constants are given by $J_S(\theta) = \cos(\theta) - \sin(\theta)/2$ and $J_Q(\theta) = \sin(\theta)/2$.

We should remark that quadrupolar order is formally captured by the symmetric part of $\mathbf{T}$, and the set of operators in (3.2) is just one of many possible sets of operators that can be chosen to describe quadrupolar order. However, using the set of operators in (3.2) does unveil the at first sight hidden points of higher symmetry that the BBH Hamiltonian possesses. When expressing all three spin matrices and all five quadrupolar operators given by (3.2) in terms of the time-reversal invariant basis of (3.3): we observe that the spin matrices become equal to the three imaginary Gell-Mann matrices, whereas the above five quadrupolar operators equal the remaining five real Gell-Mann matrices. Recall that the Gell-Mann matrices are the generators of SU(3). Consequently, whenever the spin coupling constant equals the quadrupolar coupling constant, i.e. $J_S(\theta) = J_Q(\theta) = J$, the Hamiltonian reduces to an equal-weighted product over all Gell-Mann matrices:
3.3 Previous studies

\[ H = J \sum_{(i,j)} \lambda_i \cdot \lambda_j, \] which is manifestly SU(3) invariant. This occurs for \( \theta = \pi/4 \) and \( \theta = 5\pi/4 \). Additionally, when \( J_S(\theta) = -J_Q(\theta) = J \), which happens for \( \theta = \pi/2 \) and \( \theta = 3\pi/2 \), all matrices again have equal weight, except that the imaginary matrices come with a relative extra minus sign. Since the square lattice is bipartite, we can compensate for this extra sign by taking the antifundamental representation \( \tilde{\lambda} = -\lambda^* \) of SU(3) on one sublattice. The Hamiltonian then takes the form \( H = J \sum_{(i,j)} \lambda_i \cdot \tilde{\lambda}_j \) (where \( i \) is in the A-sublattice, and \( j \) in the B-sublattice), which is also SU(3) symmetric.

At the SU(3)-symmetric points, there is a larger set of operators commuting with the Hamiltonian, which means that the ground state degeneracy increases. Moreover, because the SU(3)-symmetric points are exactly the points at which the coupling constants are equal in magnitude (\( |J_S| = |J_Q| \)), the regions in between are precisely those for which one of the two coupling constants dominates the other. Hence, we can naively expect to have four different phases separated by the SU(3)-symmetric points, with magnetic or quadrupolar order depending on which of the two coupling constants is larger in magnitude.

3.3 Previous studies

In 1988 Papanicolaou [152] conducted a product-state analysis of the spin-1 BBH model, a concise summary of which can be found in [158]. Because the square lattice is bipartite, finding the product ground state reduces to a two-body problem (one product state per sublattice). Papanicolaou’s results agree with our analysis above; he found four different phases separated by the SU(3) points: the familiar ferromagnetic (FM) and antiferromagnetic (AFM) phases, and in between a ferroquadrupolar (FQ) phase wherein neighboring sites have aligned directors, and a phase he called semi-ordered (see Fig. 3.2). The latter has a degenerate product ground state: minimizing the two-particle energy leads to the condition that one sublattice state has to be quadrupolar, whereas the other sublattice state can be either quadrupolar with a director perpendicular to that of the first sublattice, or magnetic with a magnetic moment aligned with the director of the first sublattice, or a combination of the two.

As an interesting side note, let us mention that exactly at each SU(3) point both product ground states left and right of the SU(3) point in question are simultaneous ground states of the system, and can be rotated into one another through some element of SU(3).

\(^4\)Similar to SU(2) invariance of \( S_i \cdot S_j \), where \( S \) is the vector of (a half times) the Pauli matrices (the generators of the Lie algebra of SU(2)), the inner product \( \lambda_i \cdot \lambda_j \) over the generators of the Lie algebra of SU(3) is invariant under SU(3) rotations. Note that SU(2) and SU(3) act on their respective Lie algebras through the adjoint representation.
The square lattice bilinear-biquadratic Heisenberg model

3. **The square lattice bilinear-biquadratic Heisenberg model**

![Phase Diagram](image)

**Figure 3.2:** The product ground state phase diagram according to Papanicolaou [152]. In counter-clockwise order starting at $\theta = 0$ we have the antiferromagnetic (AFM), semi-ordered (SO), ferromagnetic (FM) and ferroquadrupolar (FQ) phases. The SU(3)-symmetric points are marked with black dots.

The product ground state degeneracy in the top right octant of the phase diagram means that we can expect quantum fluctuations to play an important role there. Moreover, the types of ground states found open up a lot of possibilities for interesting types of order\(^5\); for example: the fact that neighboring directors can be perpendicular in three different ways (e.g., pointing in x, y and z direction, respectively), allows for the possibility of a three-sublattice ground state (Fig. 3.3), which is surprising for a model with nearest-neighbor interactions on a bipartite lattice.

The energy per site for the product ground state is plotted in Fig. 3.4: it shows clear kinks at the SU(3)-symmetric points, suggesting that the phase transitions are all of first order.

The lower half $-\pi \leq \theta \leq 0$ of the phase diagram is free of the sign problem and has been studied using quantum Monte Carlo in 2002 by Harada and Kawashima [153], who found that the actual ground state phase diagram agrees with the product ground state phase diagram. Moreover, they observed jumps in the quadrupolar order parameter at the SU(3) points $\theta = 5\pi/4$ and $3\pi/2$, which agrees with the product state analysis and confirms that the phase transitions from the antiferromagnetic to the ferroquadrupolar and from the ferroquadrupolar to the ferromagnetic phases are both of first order.

\(^5\)E.g., any three-color node coloring of the square lattice corresponds to a ground state, where each of the three colors stands for a quadrupole aligned in one of three mutually perpendicular directions; or, there are also fancier orderings, such as a two-sublattice ground state with $z$-aligned directors on the A-sublattice, and on the B-sublattice a linear combination of a $z$-magnetized state and any quadrupole with a director that lies in the x-y plane.
Figure 3.3: Different types of product ground states in semi-ordered phase ($\pi/4 \leq \theta \leq \pi/2$). Left: three-sublattice order with neighboring directors being perpendicular (antiferroquadrupolar 'AFQ3' state—discs with differing colors are quadrupolar states viewed at different angles); right: two-sublattice order with directors aligning parallel to neighboring magnetic moments (half nematic half magnetic 'm = 1/2' state). Note that the color coding is only meant to highlight which particles are in the same quantum state.

Figure 3.4: The product ground state energy. The kinks in the energy per site suggest that the phase transitions at the SU(3)-symmetric points are of first order.

The Monte Carlo study was followed up by an exact-diagonalization and linear-flavor-wave study at the SU(3)-symmetric point $\theta = \pi/4$ by Tóth et al. [168] in 2010, who concluded that the ground state is actually a three-sublattice state that is favored over the two-sublattice state because it has a lower zero-point energy (the energy of the wave-excitations on top of the classical ground state averaged over the Brillouin-zone). The three-sublattice order of the ground state at the SU(3) point was confirmed by DMRG and iPEPS simulations by Bauer et al. [169].

A further study by Tóth et al. [158] in 2012 based on exact diagonalization and linear-flavor-wave theory examined the question whether the above-mentioned three-sublattice phase extends beyond the SU(3) point $\theta = \pi/4$. They found that, in the semi-ordered phase, the ground state is the three-sublattice anti-ferroquadrupolar (AFQ3) state with perpendicular directors on neighboring sites.
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(the analogous product state is depicted in Fig 3.3: left). Interestingly, Tóth et al. also found that the three-sublattice pattern extends into the antiferromagnetic regime, where, for $0.2\pi \lesssim \theta < \pi/4$, instead of ordinary antiferromagnetism, a $120^\circ$ magnetic order emerges (Fig. 3.5). This result was thereafter confirmed by series expansion [160].

Figure 3.5: Magnetic order in the $120^\circ$ magnetically ordered phase: the spins align in a plane and orient themselves at relative angles of $120^\circ$.

3.4 iPEPS results

3.4.1 Overview

To get a first rough picture of the ground state phase diagram, and also as a consistency check, we initialize $D = 1, 2, 3, \ldots, 6$ simple update simulations for $\theta$ at 40 evenly spaced points covering all of $[0, 2\pi]$ using 2x2 and 3x3 unit cells (Fig. 3.6; only $D = 1, 2$ and 6 shown). The top plot of Fig. 3.6 shows the energy per site, which is seen to decrease as the bond dimension increases, except in the FM region $\pi/2 < \theta < 5\pi/4$, where the ground state can be represented by a product state (i.e. $D = 1$). Moreover, we observe a transition from a two-sublattice to a three-sublattice ground state around $\theta \approx 0.2\pi$, with the $0.2\pi$ three-sublattice simulations showing $120^\circ$ magnetic order observed in the AFM3 phase.

The bottom plot of Fig. 3.6 shows the magnetization per site (for $D = 6$ simulations), which is of order one in the FM and AFM phases, and zero in the quadrupolar phases, as expected. Contrary to magnetic order, quadrupolar order cannot be captured by a single order parameter, because it is described by a matrix: the traceless symmetric part of $S^\alpha S^\beta$, denoted by $Q^{\alpha\beta}$. The convention used for the quadrupolar operators in the vector $Q$ given by Eq. (3.2) has a preferred choice of direction in spin-space, as can be seen from the first two components of $Q$ which represent the diagonal part of $Q^{\alpha\beta}$. However, when measuring quadrupolar order, we prefer to work with invariants of $Q^{\alpha\beta}$ such as its eigenvalues, or equivalently, the matrix invariants $I_Q = \text{tr}(Q) = 0$, $II_Q = \frac{1}{2} (\text{tr}(Q)^2 - \text{tr}(Q^2)) = -\frac{1}{2} \text{tr}(Q^2)$ and

$\text{I.e. the matrix } Q^{\alpha\beta} = S^\alpha S^\beta + S^\beta S^\alpha - \frac{2}{3} S(S + 1) \delta^{\alpha\beta} \text{ that has one-third of the trace subtracted from each diagonal element).}$
3.4. iPEPS results

![Image of graph](image)

**Figure 3.6:** Top: energy per site for \( D = 1, 2, 6 \) simple update simulations using 2x2 and 3x3 unit cells (color online); for each value of \( \theta \) only the lowest (in energy) of the two is shown. The black dots correspond to a 1x1 unit cell state (energies for 2x2 and 3x3 simulations coincide). Bottom: magnetization per site \( m \) and the \( II_Q \) and \( III_Q \) tensor invariants of the \( Q \)-matrix per site for the \( D = 6 \) simulations. \( III_Q \) is identically zero (not shown).

\( III_Q = \det(Q) \) rather than the individual components of \( Q \). Fig. 3.6 shows the two non-zero matrix invariants \( II_Q \) and \( III_Q \), which are clearly larger in magnitude in the quadrupolar phases than they are in the magnetized phases\(^7\). Moreover, the determinant \( III_Q \) changes sign when going from a nematic to a magnetic state.

The \( D = 1, 2, \ldots, 6 \) simple update simulations reproduce the AFM, AFQ3, FM and FQ phases separated by the SU(3) points, and hint at the existence of the AFM3 phase\(^8\) found by Tóth et al. [158] in between the AFM and AFQ3 phases. Because the lower half \(-\pi < \theta < 0\) of the phase diagram has already been established

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\(^7\)For a typical spectrum of \( Q \), see Fig [14] in the appendix.

\(^8\)The iPEPS simulations at \( \theta = 0.2\pi \) show that the state with the 3x3 unit cell is competitive in energy, and all of the corresponding 3x3-unit-cell simulations display the AFM3 magnetization pattern.
by quantum Monte Carlo [153], and in the top left quadrant $\pi/2 < \theta < \pi$ we only found ferromagnetic translationally invariant product states that have the same energy independent of $D$ or unit cell size, the only section of the phase diagram that remains to be thoroughly investigated is the top right quadrant $0 < \theta < \pi/2$. In order to obtain more accurate data on this interesting region, we will use finer-spaced higher-$D$ full update simulations rather than the less accurate simple update simulations shown in Fig. 3.6.

The $0 < \theta < \pi/4$ part of the top right quadrant containing the magnetic and Haldane phases will be thoroughly investigated in Section 3.4.2. The predominantly quadrupolar part $\pi/4 < \theta < \pi/2$ of the top right quadrant, which also contains the partially nematic partially magnetic `$m = 1/2$' phase, will be discussed in Section 3.4.3 together with the remaining phases that occur in the square lattice spin-1 BBH model. Combining all of our full update iPEPS results, we arrive at the phase diagram shown in Fig. 3.7. Note that the Haldane, AFM3 and $m = 1/2$ phases are not visible in the simple update plots in Fig. 3.6 because the $\theta$-grid used is too coarse, and, in case of the Haldane phase, the bond dimension $D$ is too low. Finally, Section 3.4.4 investigates the nature of all occurring phase transitions—also shown in Fig. 3.7.

**Figure 3.7:** The phase diagram according to our iPEPS data. All phase transitions are of first order (solid lines), except possibly the AFM to Haldane phase transition, which is a weak first or second order phase transition (dotted line).
3.4. iPEPS results

The analysis presented below involves simulations with a bond dimension as high as \( D = 10 \) (\( D = 16 \) for simulations in the Haldane phase), followed by a \( D \to \infty \) extrapolation where necessary. As a consequence, the expectation values may differ slightly from those shown in Fig. 3.6 which are only meant to give a rough overview of the type of order that can be expected.

3.4.2 The extended Haldane phase

We first discuss the well-established limits of the phase diagram in the range \( \theta \in [0, \pi/4] \).

AFM phase

For \( \theta = 0 \) the model reduces to the spin-1 Heisenberg model where the ground state exhibits antiferromagnetic order. Unlike for \( \theta > 0 \), Quantum Monte Carlo has no negative sign problem in this limit, and the sublattice magnetization \( m = \sqrt{\langle S_x \rangle^2 + \langle S_y \rangle^2 + \langle S_z \rangle^2} \) in the thermodynamic limit has been accurately determined: \( m = 0.805(2) \) \cite{138}. Our iPEPS result extrapolated to the infinite \( D \) limit (see Fig. 3.8), \( m = 0.802(7) \), is in agreement with this value. States in the AFM phase are U(1)-spin-rotation symmetric around the axis of magnetization.

AFM3 phase

For \( \theta = \pi/4 \) the model is equivalent to the \( SU(3) \) Heisenberg model (with the fundamental representation on each site), for which a three-sublattice ordered state has been predicted by several methods \cite{168, 169}, including previous iPEPS simulations. For \( \theta \) slightly below \( \pi/4 \) this order corresponds to a 120° order formed by the spins on three sublattices as displayed in Fig. 3.5. The magnetization per site varies from \( m \approx 0.4 \) at \( \theta = 0.22\pi \) (Fig. 3.10) to \( m \approx 0.1 - 0.3 \) near \( \theta = \pi/4 \) (Fig. 3.12). AFM3 states have no residual spin-rotation symmetry.

Intermediate paramagnetic phase

In Refs. \cite{158, 160} a direct transition between the AFM state and the 120° magnetically state has been predicted to occur around \( \theta \approx 0.2\pi \) based on exact diagonalization, linear flavor-wave theory and series expansion. We first attempt to reproduce this result with iPEPS by determining the critical value \( \theta_c \) for which the energies of the two states—distinguished by different unit cells—intersect. To do so, we initialize simulations from deep within the AFM (120° magnetically ordered) phase, and slowly increase (decrease) \( \theta \) up to the point where the energies...
cross. The resulting critical value $\theta_c \approx 0.21\pi$ (for $D = 10$) is close to the previous prediction. However, from a systematic analysis of the AFM order parameter shown in Fig. 3.8 we find that the AFM order actually vanishes long before $\theta_c$, i.e. that the AFM phase is only stable up to $\theta = 0.189(2)\pi$. This indicates the presence of an intermediate non-magnetic phase in between the AFM and 120° magnetically ordered phases\(^9\).

We next explore the region around $\theta = 0.2\pi$ in more detail. When starting from different random initial states with a two-sublattice ansatz, we observe a competition between a weakly magnetized state and a non-magnetized state which breaks lattice rotational symmetry but preserves SU(2) and translational symmetry\(^{10}\). This non-magnetized state can also be found by restricting the simulation to a 1-site unit cell\(^{11}\); it exhibits the lowest variational energy for large $D$.

The rotational symmetry breaking manifests itself in energy differences in $x$ and $y$—direction, reminiscent of coupled one-dimensional chains. Since the ground state of the BBH chain for $\theta \in (-\pi/4, \pi/4)$ is in the Haldane phase, the question naturally arises whether the intermediate two-dimensional phase could possibly be adiabatically connected to the Haldane phase by continuously decreasing the $y$-coupling to zero. A first hint that this picture is correct comes from the observation that, when initializing the iPEPS as a product of chains in the one-dimensional

\(^9\)In Ref. [160] a consistent value of $\theta_c = 0.190\pi$ was predicted for the vanishing of the AFM order based on fitted series expansion data, from which the authors concluded that there is a continuous or a very weak first order transition between the AFM and the 120° magnetically ordered phase.

\(^{10}\)We have also checked for potential valence-bond solid states in unit cells up to $4 \times 4$ but could not find stable solutions.

\(^{11}\)A special full-update imaginary-time evolution algorithm has been used in this case which will be discussed elsewhere.
3.4. iPEPS results

Haldane phase, the simulation converges to the same non-magnetized state as with randomly initialized tensors. In order to confirm the above hypothesis, we study the stability of the Haldane phase in the anisotropic BBH model in the following.

The anisotropic model

We introduce different coupling strengths in \(x\) and \(y\)-direction and study the phase diagram in the \((\theta, J_y)\) plane (setting \(J_x = 1\) in the following). For \(J_y = 0\) the model simply reduces to independent spin-1 chains, which are known to lie in the Haldane phase (for \(\theta \in (-\pi/4, \pi/4)\)). The goal is now to determine the critical coupling \(J_y^c(\theta)\) separating the Haldane phase from the AFM phase (or the 120° magnetically ordered phase for large \(\theta\)), for different values of \(\theta\). In order to obtain an estimate of the critical value of \(J_y^c(\theta)\) for fixed \(\theta\), we initialize the iPEPS in the Haldane phase and in the AFM or 120° magnetically ordered phase respectively, run simulations for different values of \(J_y\), and determine the value \(J_y^c(\theta)\) for which the energies of the two states intersect using a fixed bond dimension \(D = 10\) and the simple update optimization (see data in the Appendix 3.A.1). The resulting phase diagram is shown in Fig. 3.9.

![Figure 3.9: The phase diagram of the anisotropic bilinear-biquadratic spin-1 model. The phase boundaries were estimated based on \(D = 10\) simple update simulations, see Appendix 3.A.1.](image)

We note that this approach provides only an approximate phase boundary, in contrast to the extrapolated full update simulations used in the isotropic case. However, it is computationally much more efficient, which becomes significant when probing the extended two-dimensional parameter space \((\theta, J_y)\). Moreover, a comparison with Monte Carlo and full update results (see below) indicates that the relative error on the phase boundary is only a few percent, which is accurate enough for our current purpose.

For \(\theta = 0\) we find a critical value \(J_y^c(0) = 0.042\) which is close to the Quantum Monte Carlo result 0.0436 from Ref. [138] (see also Refs. [135–137, 139]). This
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value lies far away from the isotropic two-dimensional limit \( J_y = 1 \). However, we find that \( J_c^y(\theta) \) monotonously increases with \( \theta \)—as shown in the phase diagram in Fig. 3.9—and that, beyond \( \theta_c = 0.200\pi \) no phase transition occurs, demonstrating that the one-dimensional Haldane phase can indeed be adiabatically connected to the isotropic two-dimensional limit. Finally, for \( 0.213\pi \leq \theta < \pi/4 \) we find a transition value \( J_c^y(\theta) \) between the Haldane and the 120° magnetically ordered phase which decreases with increasing \( \theta \).

Comparing to the full update results (Figs. 3.8 and 3.10 below), which predict the two transitions to be at 0.189(2)\( \pi \) and 0.217(4)\( \pi \) respectively, we see that the simple update underestimates the extent of the Haldane phase at the isotropic point. Moreover, it does so by a margin of at most 0.01\( \pi \) (and by much less for \( \theta = 0 \)), indicating that the continuous path that connects the intermediate two-dimensional phase to the one-dimensional Haldane phase persists also when taking the error margin on the phase boundary into account.

**Transition from Haldane to 120° magnetically ordered phase**

We next focus again on the isotropic two-dimensional case (\( J_y = 1 \)) and accurately determine the transition from the Haldane to the 120° magnetically ordered phase by pushing the simulations up to \( D = 16 \) (Haldane state) and \( D = 10 \) (120° magnetically ordered state) using the full update optimization, and compare the energies of the two states in the infinite \( D \) limit. Figure 3.10(a) shows the energies

![Figure 3.10: (a) Energy per site (full update) of the Haldane and three-sublattice (3-SL) AFM3 states for two different values of \( \theta \), plotted as a function of the truncation error \( w \). (b) Local magnetic moment \( m \) (triangles) of the three-sublattice state, and the difference in bond energies \( \Delta E \) in x and y-direction in the Haldane state (squares), plotted as a function of \( 1/D \). Note that \( m \) and \( \Delta E \) are zero for the Haldane and three-sublattice states respectively.](image)

extrapolated in the so-called truncation error \( w \) (see Ref. [132] for details). For \( \theta = \)}
0.21\(\pi\) the state in the Haldane phase is clearly lower than the 120\(^\circ\) magnetically ordered state, whereas for \(\theta = 0.22\pi\) the opposite is true. By linear interpolation of the energies, taking into account the extrapolation error, we find a critical value of \(\theta_c = 0.217(4)\pi\).

Finally, the squares in Fig. 3.10(b) show the difference in bond energies \(\Delta E = E_y - E_x\) in \(x\) and \(y\)-direction of the Haldane state for two values of \(\theta\). In the infinite \(D\) limit \(\Delta E\) tends to a finite value, e.g. \(\Delta E = 0.07(1)\) for \(\theta = 0.21\pi\), which shows that the rotational symmetry in the Haldane phase is indeed spontaneously broken.

### 3.4.3 Description of the remaining phases

Next, we continue our iPEPS investigation of the phase diagram by providing a description of all occurring phases in order of increasing \(\theta\), starting at \(\theta = \pi/4\) up to \(\theta = 2\pi\).

#### AFQ3 phase

Similar to the AFM3 phase, the three-sublattice antiferroquadrupolar phase is also described by a 3x3 unit cell, but now the magnetization per site is near zero at \(\pi/4\) (Fig. 3.12\(^{12}\)), and exactly zero from about \(\theta \approx 0.27\pi\) up to the \(m = 1/2\) phase (see Fig. 3.13 for \(\theta = 0.487\pi\)). At the product state level, in addition to being time-reversal symmetric, the AFQ3 state has a residual spin-rotation symmetry given by \(\pi\)-rotations around the (mutually perpendicular) axes of nematic polarization (see also Fig. 3.3).

#### \(m=1/2\) phase

Next, let us focus on the extent of the three-sublattice AFQ3 phase in the region \(\pi/4 < \theta < \pi/2\). Recall that, at the product state level, there is a multitude of distinct types of ground states, two of which are depicted in Fig. 3.3. Tóth et al. [158] predicted that the three-sublattice (AFQ3) state is the ground state for \(\pi/4 < \theta < \pi/2\). However, their product state analysis in this same region shows that the addition of an infinitesimal external magnetic field favors the two-sublattice half nematic half magnetic \(m = 1/2\) state as the ground state. Augmented by linear-flavor-wave theory, they continue to show that the AFQ3 phase extends into the small but non-zero magnetic field \(h > 0\) region for \(\pi/4 < \theta < \pi/2\),

\(^{12}\) Possibly, the magnetization is not exactly zero at \(\pi/4\) and \(5\pi/4\) because of the higher SU(3) symmetry that allows magnetic and quadrupolar states to be rotated into each other at the cost of no energy, which weakens the hysteresis effect and prevents the quadrupolar simulation from remaining completely nematic at the phase transition.
but that, as $\theta$ approaches $\pi/2$ from below, the phase boundary between the $m = 1/2$ and AFQ3 phases moves down to $h \to 0$ as $\theta \to \pi/2$. Since their exact diagonalization results for the AFQ3 phase with non-zero magnetic field do not extrapolate well to infinite system size, and linear-flavor-wave theory is semi-classical, it is not clear what happens close to $\theta = \pi/2$.

We have thoroughly investigated the parameter range of $0.48\pi < \theta < \pi/2$ using iPEPS. To get an idea of what types of states to expect, we initialized simulations using eleven different types of unit cells (to also allow for possibilities such as stripe order, as well as the more conventional types of order that can be represented by unit cells up to size $4x4$) and evolved them in imaginary time using the simple update algorithm up to bond dimension $D = 6$. We then picked the minimal unit cell for each type of state obtained (e.g. the $4x2$ state displayed the same pattern as the $2x2$ state, so we discarded the former), and evolved those in imaginary time using the full update algorithm up to $D = 8$. This left two competitive states: the three-sublattice AFQ3 state described by a $3x3$ unit cell iPEPS, and the two-sublattice $m = 1/2$ state described by a $2x2$ unit cell iPEPS. Interestingly, the $m = 1/2$ state turned out to have a lower energy than the AFQ3 state for $0.49\pi \lesssim \theta < \pi/2$. Finally, we ramped up the bond dimension to $D = 10$ to nail down the precise location of the phase transition between the AFQ3 and $m = 1/2$ phases.

As can be seen in Fig. 3.11, for $\theta = 0.487\pi$, the AFQ3 simulation is lower in energy than the $m = 1/2$ simulation, whereas for $\theta = 0.490\pi$ the AFQ3 simulation is higher in energy. Taking the error bars from the $D \to \infty$ extrapolation into account, we can conclude that—in contrast to previous predictions—the AFQ3 phase does not persist all the way up to the SU(3)-symmetric point $\pi/2$, but remains stable only up to $\theta = 0.4886(7)\pi$, after which the system transitions into the $m = 1/2$ phase.

In case the reader is wondering why the $m = 1/2$ phase occupies such a small portion of the phase diagram, it is insightful to remark that the size in $\theta$-space is not a physical quantity, as $[\cos(\theta), \sin(\theta)]$ is but one of the many possible parameterizations of the coupling constants of the Hamiltonian in Eq. (3.1). Additionally, there is a reason for the $m = 1/2$ phase to only appear close to $\pi/2$. The actual ground state is similar but not equal to the product state from Fig. 3.3 (right), and on the quadrupolar sublattice a small magnetic moment parallel to that of the magnetized sublattice can be detected. This is only energetically favorable when the spin coupling parameter $J_S(\theta)$ in the Hamiltonian expressed in terms of $S$ and $Q$—see Eq. (3.4)—becomes of a magnitude that is similar to that of the quadrupolar coupling parameter $J_Q(\theta)$, which only happens around $0.49\pi$ where $J_S(\theta)$ decreases rapidly from 0 towards $-1/2$.

\textsuperscript{13}Specifically, we have initialized simple update simulations using $2x2$, $3x2$, $3x3$, $4x2$, $4x3$, $4x4$, $5x2$, $5x3$, $6x2$, $7x2$, and $8x2$ sized unit cells.
As its name suggests, the magnetization per site of $m = 1/2$ states is exactly 1/2 throughout the entire phase (e.g., see Figs. 3.13 and 3.14 for $\theta$ close to the transition points). It has a residual U(1)-spin-rotation symmetry around the axis of magnetization. As $\theta$ approaches $\pi/4$, the $m = 1/2$ state gradually turns into a product state (see Section 3.4.4).

**FM phase**

The familiar ferromagnetic phase is described by a translationally invariant product state represented by a 1x1 unit cell. It has a residual U(1)-spin-rotation symmetry around the axis of magnetization, and a magnetization per site of exactly $m = 1$ (see Figs. 3.6, 3.14 and 3.15).

**FQ phase**

The ferroquadrupolar phase is also translationally invariant, and can be represented by a 1x1 unit cell. Its magnetization is zero or very close to zero throughout the phase (see Figs. 3.15 and 3.16 for the transition points, and also Footnote 12). At the product state level, it is symmetric under time-reversal, residual U(1)-spin-rotations around the axis of nematic order, and $\pi$-rotations around any axis.
perpendicular to the axis of nematic order. Close to the FM phase, states in the FQ phase are product states, but, as we approach the AFM phase, the FQ states become gradually more and more entangled (see Sections 3.4.4 and 3.4.4).

**AFM phase**

Finally, the last phase appearing in the phase diagram before we come full circle is the familiar antiferromagnetic phase discussed in Section 3.4.2. The magnetization per site varies throughout the phase from \( m \approx 0.5 - 0.6 \) close to the FQ phase (Fig. 3.16) to \( m \approx 0.8 \) around \( \theta = 0 \), after which it monotonically decreases to zero or almost zero when approaching the Haldane phase (Fig. 3.8).

### 3.4.4 Nature of the phase transitions

In the following section, we will discuss the nature of the phase transitions in the phase diagram shown in Fig. 3.7.

From the previous two sections, we gather that all neighboring phases break different symmetries, which suggests that all phase transitions are either first order, or unconventional second order transitions (see e.g. [24,171]). To support this hypothesis—and where possible distinguish between the two options—we will next look for kinks in the energy per site, or jumps in typical order parameters such as the magnetization or the Q-matrix invariants per site due to varying \( \theta \).

**AFM to Haldane: \( \theta = 0.189(2)\pi \)**

At \( \theta = 0.189(2)\pi \), we have a transition between the antiferromagnetic and Haldane phases. In the AFM phase, we observed that the magnetization per site goes to zero when approaching the Haldane phase (see Fig. 3.8(b)), suggesting a second order phase transition, which is unconventional considering the fact that both states break different lattice translation and rotation, as well as different spin-rotation symmetries. Moreover, we did not find clear hysteresis behavior, which supports the claim that this transition is second order. However, due to the error bars in the magnetization close to the transition, from our simulations we were not able to conclude with certainty whether the phase transition is a second or a weak first order transition; all we can say for sure is that this is not a clear first order transition as no jump in magnetization or kink in the energy were observed.

We later on became aware of the work in Ref. [172] that first described the AFM to Haldane transition in the large N limit. Note that, here, “N” refers to the the symmetry group SU(N) of the Hamiltonian—which in our case is SU(2)—not to the total number of particles. At large distances, both the AFM and Haldane phases can be effectively described by the O(3) non-linear sigma model, a
continuous field theory of the continuum limit \( n \) of the staggered magnetization 
\((-1)^{\pm y} S(x,y)\) [24]. It is possible to express the order parameter field \( n \) in terms 
of a confined two-component complex scalar field \( z = (z_1, z_2) \), as follows
\[
 n = z^\dagger \sigma z, \tag{3.5}
\]
where \( \sigma \) is the vector of Pauli matrices, and \(|z|^2 = |z_1|^2 + |z_2|^2 = 1 \) because 
\( n \) has unit length. There is an additional \( U(1) \) gauge redundancy associated to 
Eq. 3.5. Indeed, mapping \( z \mapsto \exp[i\gamma(x,y)]z \) leaves \( n \) invariant for any \( U(1) \) 
gauge field \( \exp[i\gamma(x,y)] \). Consequently, \( z \) parametrizes \( CP^1 \). At the critical point, 
the \textit{fractionalized spinon fields} \( z_1 \) and \( z_2 \) deconfine\(^{14}\); and therefore, the above
phenomenon is understood as \textit{deconfined criticality} described by a \( CP^1 \) model [24].

**Haldane to AFM3:** \( \theta = 0.217(4)\pi \)

The Haldane to the three-sublattice 120° magnetically ordered phase transition 
at \( \theta = 0.217(4)\pi \) displays hysteresis effects around the transition point, which 
allows us to simulate both phases on both sides of the phase transition. Additionally, 
the sublattice magnetization is strictly positive throughout the AFM3 phase 
(see Fig. 3.10(b))—even for \( \theta = 0.2\pi \) where the AFM3 state is no longer the 
lowest energy state—implying that the magnetization does not go to zero when 
approaching the transition from above. Since the magnetization is zero in the 
Haldane phase, it jumps to zero at the transition, showing that the transition is 
clearly of first order.

**AFM3 to AFQ3:** \( \theta = \pi/4 \)

At \( \theta = \pi/4 \), we have a transition from the three-sublattice 120° magnetically 
ordered to the three-sublattice antiferroquadrupolar phase. Precisely at the symmetric point \( \theta = \pi/4 \), both the 120° magnetically ordered state and the 
antiferroquadrupolar state are ground states of the system. Hence, we can simulate 
both at the phase transition by initializing the simulations from deep within the 
120° magnetically ordered and quadrupolar phases respectively, and then moving 
towards the phase transition by initializing each simulation from the previous one. 
The resulting data at the critical point is shown in Fig. 3.12.

The left plot of Fig. 3.12 shows a subtle kink in the energy per site, which we 
observe for each fixed bond dimension simulation for \( D = 2,3,\ldots,8 \) (only \( D = 
4,6,8 \) shown), supporting the occurrence of a first order transition. The right 
plot shows the magnetization per site exactly \textit{at} the phase transition (where we

\(^{14}\)Meaning that, at the critical point, the norm \(|z|\) is allowed to fluctuate, and monopoles 
(skyrmions: topological defects in the \( n \)-field) become irrelevant, and therefore the skyrmion 
number is asymptotically conserved (in the large \( N \) limit).
increased $D$ up to 10). Despite the fact that the magnetization data points do not fit on a perfect straight line, a rough extrapolation of $D \to \infty$ shows that the magnetic AFM3 state at the transition has a magnetization of at least 0.1, whereas the quadrupolar AFQ3 state has a magnetization of around zero (see also Footnote 12), which agrees with the above observation that this is a first order transition.

Because the magnetization does not extrapolate very nicely to $D \to \infty$, we have also investigated the behavior of the eigenvalues of the quadrupole matrix (Appendix 3.A.2: Fig. 3.18 right), where a jump in spectrum can be observed. Additionally, we have also observed the occurrence of hysteresis (Appendix 3.A.2: Fig. 3.18 left). All of the above taken together, combined with the fact that, at the product state level, the two phases break different symmetries, lead us to conclude that this must be a first order phase transition.

**AFQ3 to $m=1/2$: $\theta = 0.4886(7)\pi$**

Using the same simulations as the ones shown in Fig. 3.11 (but now only for $\theta = 0.487\pi$ and $\theta = 0.490\pi$ to prevent the figure from getting too cluttered), we see that the magnetization on both sides of the phase transition for the $m = 1/2$ states is exactly one-half, whereas it is exactly zero for the antiferroquadrupolar states (Fig. 3.13). Thus, at the transition from AFQ3 to $m = 1/2$ at $\theta = 0.4886(7)\pi$, the magnetization jumps from zero to one-half, which means that the transition
is of first order. This notion is confirmed by the right plot of Fig. 3.11, which shows that, at the intersection, the energy per site graphs for the AFQ3 and $m = 1/2$ phases have different slopes, indicating a kink in the energy per site at the transition.

$m = 1/2$ to FM: $\theta = \pi/2$

In this case, the transition is between two magnetized phases: the half-magnetized
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$m = 1/2$ phase and the ferromagnetic phase. States in the ferromagnetic phase are product states, which in our simulations can be seen by the fact that the energy does not improve as the bond dimension increases. Hence, any fixed $D$ simulation will reproduce the exact ground state ($D = 2$ shown in Fig. 3.14).

As $\theta$ approaches $\pi/2$ from below, the $m = 1/2$ state also turns into a product state, which can be seen from the fact that the the energy per site graphs for different values of $D$ all converge to the same value at $\theta = \pi/2$ (Fig. 3.14 left). Hence, at the phase transition, no $D \to \infty$ extrapolation is necessary, as the $D = 2$ results already give the exact ground state.

The left plot of Fig. 3.14 displays a clear kink in the energy, demonstrating that this is a first order transition. Moreover, Fig. 3.14 right shows that the $m = 1/2$ phase has a magnetization per site of exactly $1/2$, whereas the ferromagnetic phase is fully magnetized, confirming that the transition is indeed of first order.

The remaining two critical points at $5\pi/4$ and $3\pi/2$ have previously been investigated using quantum Monte Carlo simulations [153]. Harada and Kawashima demonstrated that $(S_z)^2 - 2/3$, which they used as the quadrupolar order parameter, exhibits a jump at $\theta = 5\pi/4$ and at $\theta = 3\pi/2$. This indicates that both transitions are of first order; a conclusion that also follows from our simulations, which we present below for completeness. Because the jump in the quadrupole moment has already been established, we will focus on the energy and magnetization in the following, but remark that we also observe clear jumps in the spectrum of the $Q$-matrix.

**FM to FQ:** $\theta = 5\pi/4$

As before, we are dealing with product states on both sides of the phase transition. Thus, we can use any fixed $D$ simulations to investigate the nature of the ferromagnetic to ferroquadrupolar phase transition ($D = 2$ shown). Both the kink in energy and the jump in magnetization displayed in Fig. 3.15 confirm that the phase transition at $\theta = 5\pi/4$ is indeed of first order.

**FQ to AFM:** $\theta = 3\pi/2$

When increasing $\theta$ from $5\pi/4$ to $3\pi/2$, the ground state becomes more and more entangled while remaining in the ferroquadrupolar phase. Upon reaching $3\pi/2$ the ground state is no longer a product state, and therefore we require higher $D$ simulations to investigate the phase transition at $3\pi/2$.

As shown in Fig. 3.16, for each fixed value of $D$ there is a subtle but observable kink in the energy per site as a function of $\theta$. Additionally, the magnetization per site at the phase transition, which does not extrapolate very well to $D \to \infty$,
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**Figure 3.15:** Energy per site (left) and magnetization per site (right) for the ferromagnetic (FM) and ferroquadrupolar (FQ) states as a function of $\theta$. No extrapolation is needed because both phases are represented by product states. The kink in energy and jump in magnetization show that this transition is first order.

does appear to saturate in between 0.5 and 0.6 in the AFM phase, whereas it extrapolates to zero in the FQ phase, confirming that this transition is also of first order.

**Figure 3.16:** Energy per site as a function of $\theta$ for $D = 5, \ldots, 8$ (left) and magnetization per site for $D = 3, 4, \ldots, 10$ at $\theta = 3\pi/2$ (right) for the ferroquadrupolar (FQ) and antiferromagnetic (AFM) phases. The energy per site shows an observable kink for each $D$ plotted. Moreover, the magnetization per site is zero in the FQ phase, whereas it saturates in between 0.5 and 0.6 in the AFM phase, showing that the transition is first order.
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3.5 Conclusion

We have studied the ground state phase diagram of the spin-1 BBH model on the square lattice by means of infinite projected entangled pair states (iPEPS). Using low bond dimension simple update simulations, we were able to reproduce all phases that were previously predicted to occur [152, 158] in this model. We then turned our attention to the challenging top right quadrant of the phase diagram: the part that has the largest product ground state degeneracy, is beyond the reach of Monte Carlo, and had up to now only been studied on small systems with exact diagonalization [158] or by means of semi-classical approaches [158, 160].

In this interesting region, we found two new phases. The first is the paramagnetic extended Haldane phase that preserves spin-rotation and lattice-translation symmetry, but breaks lattice rotational symmetry and can be adiabatically connected to the one-dimensional Haldane phase. Second, we also encountered the $m = 1/2$ phase that had been known to exist in the presence of an external magnetic field [158], but, in contrast to previous predictions, our iPEPS data showed it also exists in a small $\theta$-window in the zero-external-field ground state phase diagram.

We concluded our analysis by investigating the nature of all phase transitions of the BBH model, mainly by looking at kinks in the energy and jumps in the magnetization per site as a function of $\theta$, while also taking symmetry considerations into account. We found clear or subtle kinks in the energy for all transitions but the AFM to Haldane transition. Similarly, the magnetization displayed slight to clear jumps except at the above-mentioned transition, leading us to conclude that all phase transitions are of first order, except possibly the AFM to Haldane transition, which we predict to be either of second or weak first order.

In addition to demonstrating that the spin-1 BBH model on the square lattice exhibits various exotic phases, such as several (partially) nematic phases ($m = 1/2$, FQ and AFQ3), three-sublattice ordering (AFM3 and AFQ3), and even a highly symmetric paramagnetic phase that only breaks lattice rotational symmetry (Haldane)—which are interesting in their own right from a theoretical point of view—we have now paved the way for our next investigation of the spin-1 BBH model on the numerically more challenging triangular lattice, presented in Chapter 4.

It is interesting to note that the AFM to Haldane transition, which was first predicted in Ref. [172], has also been encountered in a number of related studies [130, 173–176]. For the square lattice spin-1 $J_1$-$J_2$ model, DMRG results by Jiang et al. [173] showed that the two-dimensional Haldane phase appears in the ground state phase diagram. However, a recent DMRG and iPEPS study by Haghshenas et al. [130] of the same model suggests that the Haldane phase only appears as a result of the finite cylinder width used in DMRG, and that it is not present on the infinite square lattice. The same Haldane phase has also
been encountered in the ground state phase diagrams of the square lattice next-nearest-neighbor spin-1 BBH $J_1$-$J_2$-$K_1$-$K_2$ model by a DMRG study by Gong et al. [175], and the $J_1$-$J_2$-$K_1$ model by a combined exact diagonalization, DMRG and PEPS study by Chen et al. [176]. Moreover, a very recent paper by Lee and Kawashima [177] on the spin-1 BBH model on the star lattice also found a spin-liquid-like ground state in a parameter regime that encloses the region wherein we found the Haldane phase on the square lattice.

Our findings also provide an additional example of a nematic quantum paramagnet which in Ref. [174] was proposed to likely emerge in spin-1 systems with competing interactions, and suggested to be potentially relevant to understand the nematic phase in the iron-based superconductor FeSe. Finally, our results show that iPEPS is a competitive method for analyzing strongly correlated spin systems, especially where quantum Monte Carlo suffers from the sign problem.
3. Appendix

3.A.1 iPEPS simulations of the anisotropic model

In Fig. 3.17 we provide the iPEPS energies (for $D = 10$, simple update) for different cuts in the phase diagram of the anisotropic model, which were used to get an estimate of the phase boundary between the Haldane and AF phase, or Haldane and 3-sublattice phase, respectively. The phase transitions occur where

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig317.png}
\caption{iPEPS energies per site for $D = 10$ (simple update) as a function of $J_y$ (or $\theta/\pi$) for fixed values of $\theta/\pi$ (or $J_y$).}
\end{figure}
the energies of the states intersect. This data was used to plot the phase diagram of the anisotropic model shown in Fig. 3.9 in Section 3.4.2.

3.A.2 AFM3 to AFQ3 transition

We have included plots that provide extra evidence for the occurrence of a first order transition at $\theta = \pi/4$. For the other transitions, the jump in magnetization or kink in the energy is clear enough to draw conclusions from.

At $\theta = \pi/4$, hysteresis can be observed around the phase transition (Fig. 3.18 left): the quadrupolar phase specifically can be simulated at $\theta = 0.249\pi < \pi/4$ (where the ground state is in the magnetized phase) before jumping to a magnetized state at $\theta = 0.245\pi$.

![Figure 3.18](image)

**Figure 3.18:** Left: magnetization per site for $D = 4, 6, 8$ as a function of the coupling parameter $\theta$ for the 120° magnetically ordered (AFM3) and antiferroquadrupolar (AFQ3) phases—hysteresis can be observed. Right: eigenvalues of the $Q$-matrix for the AFM3 and AFQ3 states at the phase transition at $\theta = \pi/4$ for $D = 3, 4, \ldots, 10$. Extrapolating $D \to \infty$ shows a jump in the spectrum of the $Q$-matrix.

Also, the eigenvalues of the $Q$-matrix (Fig. 3.18 right) differ in both phases: a rough extrapolation of $D \to \infty$ shows that in the antiferroquadrupolar phase we have eigenvalues of approximately 0.15 (twice) and $-0.3$ once, whereas in the 120° magnetically ordered phase we have 0.15, 0 and $-0.15$, indicating a jump in the spectrum, which leads us to conclude that the transition is of first order.

Additionally, from the spectrum of the $Q$-matrix we obtain some extra information about the quadrupolar phase: because $Q$ has two identical eigenvalues, the nematic order is uniaxial (as opposed to biaxial).