Quantum disorder in the spatially completely anisotropic triangular lattice

Philipp Hauke*

ICFO–Institut de Ciencies Fotòniques, Parc Mediterrani de la Tecnologia, Av. Carl Friedrich Gauss 3, 08860 Castelldefels, Spain and Institute for Quantum Optics and Quantum Information of the Austrian Academy of Sciences, 6020 Innsbruck, Austria

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Spin liquids are important for a fundamental understanding of quantum magnetism and may find applications in quantum computing, but it is still not clear under which general circumstances these exotic, quantum-disordered phases occur. To gather deeper insights, we analyze a generalization of the spatially anisotropic triangular lattice (SATL), the spatially completely anisotropic triangular lattice (SCATL), using Takahashi’s modified spin-wave theory, complemented by exact diagonalizations. For Heisenberg interactions, calculations on the SATL predict quantum disorder for several materials in which experiments have found magnetic long-range order. In the SCATL, the corresponding parameter values yield ordered ground states and this discrepancy disappears. We also study the model with \(XY\) interactions, which can be implemented with current technology in ultracold atoms in optical lattices. This allows to estimate the stability of quantum-disordered phases towards imperfect implementations of the coupling parameters. For both kinds of interactions, we find indications for extended quantum-disordered phases. In part of the parameter space of the \(XY\) model, exact diagonalization hints at a gapped state with chiral long-range order. In both models, our results suggest that two gapped nonmagnetic regions, identified as distinct in the SATL, could actually be continuously connected via the additional anisotropy of the SCATL. Further, we find that different kinds of order are always separated by disordered phases. We propose that this is a quite universal feature of two-dimensional frustrated antiferromagnets with continuous symmetry in the couplings. The SCATL may therefore—besides being relevant for experiments—yield fundamental insights into quantum-disordered phases.

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I. INTRODUCTION

Understanding magnetically disordered quantum materials is of fundamental interest, e.g., for layered magnetic insulators/metals in which magnetism is disrupted by charge doping, leading to dramatic phenomena such as high-temperature superconductivity.\(^1\)\(^-\)\(^3\) Also, disordered quantum phases can have excitations that are fractionalized even in two dimensions,\(^4\) which may be useful for quantum computation.\(^5\)

However, classical order is typically quite resilient, especially in two or three dimensions.\(^6\)\(^-\)\(^10\) To disrupt the classical order and reach quantum-disordered phases, it is assumed that frustration could be a crucial ingredient.\(^4\)\(^,\)\(^11\)

Indeed, an increasing number of theoretical studies has identified quantum spin liquids in frustrated quantum antiferromagnets. Still, however, it is not clear under which general circumstances these disordered quantum phases emerge.\(^9\)\(^,\)\(^10\)\(^,\)\(^12\)\(^,\)\(^13\)

A quite general Hamiltonian describing frustrated quantum antiferromagnets is the XXZ model,

\[
\mathcal{H}_S = \sum_{\langle i,j \rangle} J_{ij} \left( S_i^x S_j^x + S_i^y S_j^y + \lambda S_i^z S_j^z \right),
\]

where \(S_i^\alpha\) are spin operators acting on site \(i\) (here, we are interested in the extreme quantum limit \(S = 1/2\)), and \(J_{ij}\) are the couplings between two spins. We will focus on the cases \(\lambda = 1\), corresponding to Heisenberg interactions, relevant to magnetic organic salts, and \(\lambda = 0\), corresponding to \(XY\) interactions, relevant to quantum simulations using ultracold atoms or trapped ions.

To get further insight into where nonmagnetic, quantum-disordered phases appear, we study in this work a generalized model with highly tunable frustration that has, to the best of our knowledge, never been analyzed before, despite its relevance to experiment. Namely, we consider a two-dimensional, triangular lattice where the nearest-neighbor (NN) couplings \(J_{ij}\) along all three lattice directions are different, the spatially completely anisotropic triangular lattice (SCATL). It is depicted in the left side of Fig. 1. For simplicity, we will work throughout this paper in the associated square lattice (right side of Fig. 1), where the vectors connecting NN sites are \(\mathbf{r}_1 = (1,1)\), \(\mathbf{r}_2 = (0,1)\), and \(\mathbf{r}_3 = (-1,0)\). and define \(J_{\mathbf{r}_1} \equiv J\), \(J_{\mathbf{r}_2} \equiv J',\) and \(J_{\mathbf{r}_3} \equiv J''\). This model generalizes the spatially anisotropic triangular lattice (SATL), which has two of the couplings equal. Similar to the SATL, the SCATL can be tuned between vanishing and strong frustration.

This specific model may help to understand the nonmagnetic phases occurring in related systems. For example, in previous works on the SATL,\(^14\)\(^-\)\(^20\) square-lattice \(J_1 J_2 J_3\) models,\(^20\)\(^-\)\(^32\) or frustrated honeycomb lattices,\(^33\)\(^-\)\(^36\) it was found that magnetically disordered regions of quantum-mechanical origin appear at the transition between different types of long-range order (LRO). In particular, spiral and collinear LROs seem to oppose being directly connected. In this paper, we want to examine if such a behavior remains true in the more general SCATL, to further substantiate if this is a general characteristic of two-dimensional frustrated spin systems.

A related advantage of the chosen geometry is the possibility, given by the additional anisotropy, to approach the nonmagnetic phases from different angles, possibly revealing crucial information not only about their location in parameter space, but also about their nature. Indeed, the results presented in this paper suggest that two putative gapped spin liquids, previously claimed to exist as two distinct phases in the SATL (both for Heisenberg and \(XY\) interactions), might actually
be continuously connected via the additional anisotropy of the SCATL. Studying the persistence and characteristics of the putative spin-liquid phases with respect to this additional anisotropy is the first main aim of the present paper.

The second main aim is related to experimental findings in magnetic organic salts governed by Heisenberg spin models. While the SATL has found considerable attention in recent years, to our knowledge, the ground-state phase diagram of the Heisenberg SCATL has never been thoroughly investigated. Recent first-principles calculations, however, show that some magnetic materials, such as the organic salts Me₄–Et₄Pn[Pd(dmit)₂][abbreviated Pn-n], TMTTF, or BaAg₂Cu[VO₄]₂, which are well described by weakly coupled 2D triangular lattices, can have considerable anisotropies between all three intra-plane couplings. Typically, to locate the material within the well-studied SATL model, the two closer bond strengths are averaged. However, this places materials such as Sb-0 and As-2, which are experimentally investigated, far from the SATL model. Previously, we have shown that this improves the putative spin-liquid phases with respect to this additional anisotropy between the couplings. To this end, we investigate the couplings \( J_{1,2,3} \) of the SCATL, which can all be mutually different. The shown geometry is the one used in the ED of the 15-site system, chosen for maximal symmetry between all three couplings.

FIG. 1. (Color online) Geometry of the SCATL. The spins (gray bullets) are coupled to NNs along the lattice vectors \( \mathbf{r}_{1,2,3} \) by the couplings \( J_{1,2,3} \equiv J, J_{1,2,3} \equiv J', \) and \( J_{1,2,3} \equiv J'', \) which can all be mutually different. (Right) Associated square lattice. The shown geometry is the one used in the ED of the 15-site system, chosen for maximal symmetry between all three couplings.

II. CLASSICAL PHASE DIAGRAM

In this section, we discuss the classical phase diagram of the SCATL, which can serve to estimate which ordered phases to expect, and which allows to appreciate the changes induced by quantum fluctuations. To obtain the classical solution, we replace the spin operators in Hamiltonian (1) by classical rotors, which—without loss of generality—lie in the \( xy \) plane. The in-plane ordering vector \( \mathbf{Q}_{\parallel} = (Q_{\parallel x}, Q_{\parallel y}) \) is the vector which minimizes the Fourier transform of the coupling strengths. It fixes the direction of each spin (up to a global phase) as

\[
S = S(\cos(Q_{\parallel x} \cdot r_i), \sin(Q_{\parallel y} \cdot r_i), 0).
\]

We find

\[
Q_{\parallel x} = \begin{cases} \pi & \text{for } -\frac{J}{2J} < \frac{J'}{2J} + \frac{J''}{2J} < 1, \\ \arccos\left(\frac{-J}{2J} + \frac{J'}{2J} + \frac{J''}{2J}\right) & \text{else} \end{cases}
\]

\[
Q_{\parallel y} = \begin{cases} \pi & \text{for } -\frac{J}{2J} < \frac{J'}{2J} + \frac{J''}{2J} < 1, \\ \arccos\left(\frac{-J}{2J} + \frac{J'}{2J} + \frac{J''}{2J}\right) & \text{else} \end{cases}
\]

The classical phase diagram of the SCATL, plotted in Fig. 2, contains several Néel-ordered phases and an extended spiral-ordered phase. The Néel phases spread around the square-lattice limits \( (J'/J, J''/J) = (1,0) \) with \( Q_{\parallel x} = (0,\pi), \) \((J'/J, J''/J) = (0,1) \) with \( Q_{\parallel x} = (\pi,0), \) and \((J'/J, J''/J) \gg 1 \) with \( Q_{\parallel x} = (\pi,\pi). \) The spiral phase, with continuously varying ordering vector, connects smoothly to the Néel phases, and occupies the extended region between them. In particular, it extends all the way to \( J'/J = J''/J = 0 \) [and, symmetrically, to \((J'/J = 1, J''/J \to \infty) \) and \((J'/J = 1, J''/J \to \infty) \), where the system decouples into an ensemble of 1D chains. 

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The ordering vector evidences three Néel-ordered phases, an extended spiral-ordered phase, and limits where the system decouples into an ensemble of independent chains, as indicated by the labels in the left panel. The thick red lines denote transitions between different kinds of order, and along the dashed black lines the system is in the SATL limit.

III. THE QUANTUM SCATL WITH HEISENBERG INTERACTIONS

We now analyze the quantum SCATL with Heisenberg interactions, which is relevant for the interpretation of recent experiments on magnetic organic salts.

A. Known results in limiting cases

First, we discuss well-known limiting cases of the Heisenberg quantum SCATL, including results on the SATL. This helps us to assess which phases to expect. For $J'/J, J''/J \gg 1$, $(J'/J, J''/J) = (1, 0)$, and $(J'/J, J''/J) = (0, 1)$, one recovers the square lattice limit. Here, Néel order persists also in the quantum case.

Similarly, in the isotropic triangular lattice, $J' = J'' = J$, spiral LRO survives quantum fluctuations. The limits $(J' = J'' = 0)$, $(J' \to \infty \text{ with } J'' = \text{const})$, and $(J'' \to \infty \text{ with } J' = \text{const})$ correspond to ensembles of uncoupled, critical Heisenberg chains, with algebraic correlations along individual chains but no correlations between them.

For $J' = J'' \equiv \alpha J$ (or, equivalently, $J' = J$ or $J'' = J$), one recovers the SATL, which is realized in a variety of $S = 1/2$ compounds, e.g., Cs$_2$CuCl$_4$ and $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$. The model may display spin-liquid phases, although their extent and nature is still under intensive debate.

In the rest of this section, we review the main features of the SATL phase diagram as found in the literature, proceeding from large to small $\alpha \equiv J'/J = J''/J$ (for comparison, Fig. 3 reproduces the MSWT phase diagram from Ref. 20).

It is commonly accepted that order-by-disorder effects due to quantum fluctuations stabilize the Néel phase considerably over the classical model, moving the point where Néel order disappears downwards from the classical value $\alpha = 2$ to values between $\alpha \approx 1.1$ and 1.67, depending on the method used. Further, several methods predict that quantum fluctuations spread the transition point between the Néel and the spiral phase into a quantum-disordered phase.

In the following, we term this predicted disordered region “large-$\alpha$ quantum-disordered phase” (QDP). One of the main aims of this paper is to study if it is a more general feature of frustrated quantum antiferromagnets that a quantum-disordered phase intervenes in transitions between commensurate and incommensurate order.

Similar behavior has been found in a variety of quantum spin models, including $J_1-J_2-J_3$-models on the square lattice, and frustrated honeycomb models with Heisenberg or $XY$ interactions. In fact, since quantum phase transitions are driven by quantum fluctuations, one might expect that—if anywhere—a complete restructuring of the ground state in the presence of quantum-mechanical configuration may occur preferentially close to a quantum critical point. Indeed, a similar effect occurs in classical statistical physics. Assume that there is a transition between a commensurate and an incommensurate phase that both show LRO. As a first notable observation, close to this transition, the thermal phase transition to a disordered state will typically happen at lower temperature than far away from it. Beyond the thermal phase transition, the disordered phase will show short-range order of the type corresponding to the adjacent long-range ordered phase. The transition between the two different kinds of short-range order is called a disorder point. Interestingly, the correlation length associated with the two kinds of short-range order has a minimum just at this point. Hence—similar to what is found in the above-mentioned quantum models—thermal fluctuations tend to suppress order most effectively at a commensurate-incommensurate transition.

At the low-$\alpha$ side of the spiral phase, previous works predict a disordered phase, which could appear for as large $\alpha$ as $\approx 0.8-0.9$. Our previous MSWT results suggest $\alpha \approx 0.65$. In the following, we term this predicted disordered region “small-$\alpha$ QDP”. It may be associated to a spread of the gapless spin liquid of the isolated chains ($J' = 0$) to finite coupling, possibly followed by a gapped spin liquid. This double nature of the disordered region is still under debate, since some works only find a gapless spin liquid. Consistent between these methods is the prediction that quantum fluctuations disrupt ordering tendencies between the chains even for relatively large interchain couplings. But consent about the physics in this region seems far from

FIG. 2. (Color online) Classical phase diagram of the SCATL. The ordering vector evidences three Néel-ordered phases, an extended spiral-ordered phase, and limits where the system decouples into an ensemble of independent chains, as indicated by the labels in the left panel. The thick red lines denote transitions between different kinds of order, and along the dashed black lines the system is in the SATL limit.

FIG. 3. (Color online) The MSWT phase diagram of the quantum Heisenberg SATL (from Ref. 20) contains Néel order (which is considerably more stable than in the classical model), spiral order (which is destabilized by quantum fluctuations), and two putative spin-liquid (SL) phases. These are found through the breakdown of the theory and a disappearing spin stiffness, which indicates a gapped disordered phase. In the purely 1D limit, MSWT recovers convergence and produces the 1D critical state. We include sketches of classical states, where blue arrows indicate the directions of the classical rotors, namely, the 1D state at $J'/J = 0$, the spiral state at $J'/J = 1$, and the 2D-Néel state at $\alpha \geq 2$. The limits $(J'/J, J''/J) = (1, 0)$, $(J'/J, J''/J) = (0, 1)$, and $(J''/J \to \infty \text{ with } J' = \text{const})$ correspond to ensembles of decoupled, critical Heisenberg chains, with algebraic correlations along individual chains but no correlations between them.
reached. For example, recent DMRG studies entirely question the existence of the small-\(\alpha\) spin liquid(s).\(^{49}\) And a recent renormalization-group analysis\(^ {40}\) found collinear AFM long-range order in the region \(\alpha \approx 0.3\) (see also\(^ {45}\)), and, above that value, spiral order. The weakness of the spiral order found in that study leaves, however, the possibility that the true quantum ground state hosts a disordered phase in the parameter range \(0.3–0.5\).

**B. Quantum-mechanical phase diagram**

From the discussion of the classical phase diagram and the limiting cases, we have the necessary background to tackle the quantum-mechanical ground-state phase diagram of the Heisenberg SCATL. To compute it, we use the MSWT supplemented with ordering-vector optimization, working directly in the thermodynamic limit. Since this method is described in detail in our previous articles,\(^ {20,41}\) we delegate the technical aspects to the Appendix, and only summarize here the main idea. The starting point is a classical state, which one describes with quantum fluctuations in a second-order spin-wave expansion. This yields a bosonic Hamiltonian, the ground state of which is found self-consistently by minimizing its mean-field free energy. For this, quartic terms, i.e., interactions between spin waves, are decoupled via Wick’s theorem. Additionally, we employ Takahashi’s modification of vanishing magnetization. This constricts the average number of spin-wave excitations to a physical value, in contrast to conventional spin-wave theory, where the spin-wave excitations grow completely unchecked. This modification has proven a crucial improvement to describe low-dimensional systems with weak order tendencies.

Typically, one uses the classical ground state as the reference state. However, in many models quantum fluctuations considerably shift the type of predominant order. Therefore, we find the ordering vector giving the best classical reference state by including it in the self-consistent optimization. This has proven crucial to capture, e.g., the stabilization of the Néel phase by quantum fluctuations. As has been proposed in Refs. \(^ {20}\) and \(^ {41}\), the breakdown of the theory strongly suggests that at mean-field level \textit{no} semiclassical reference state yields a good description of the quantum ground state. This is then interpreted as an indication of quantum-disordered behavior in the true ground state. This will be an important aspect for the interpretation of the quantum phase diagram.

We compare these MSWT results to exact diagonalization (ED) of a 15-site lattice, as depicted in Fig. 1. The geometry is chosen for its symmetry between \(J, J’, J”\) bonds. It is important to leave the boundaries open to allow for incommensurate ordering vectors.

**1. MSWT and ED results: ordering vector and order parameter**

In this section, we give a first overview over the phase diagram, as obtained from the ordering vector \(\mathbf{Q}\) and the order parameter \(M\), followed in the next two sections by more detailed analyses. In MSWT, the ordering vector and order parameter are the direct results of the optimization [see Appendix, Eqs. \((A8)\) and \((A11)\)]. In ED, they can be extracted from the static structure factor,

\[
S(k) = \frac{1}{N^2} \sum_{i,j} e^{i k \cdot (r_i - r_j)} \langle S_i \cdot S_j \rangle. \tag{3}
\]

Its peak lies at the ordering vector \(\mathbf{Q}_{\text{ED}}\), and the square root of its height, \(\sqrt{S(\mathbf{Q}_{\text{ED}})} \equiv M_{\text{ED}}\), approaches in the thermodynamic limit the order parameter \(M\).

As seen in the MSWT and ED ordering vectors, presented in Fig. 4, quantum fluctuations stabilize the Néel phases compared to the classical case, as already observed in the SATL (see Sec. III A). In the central region around \(J' \approx J'' \approx J\), a broad range of incommensurate ordering vectors indicates spiral order. The finite MSWT order parameter (see Fig. 5, left panel) shows that in these phases indeed LRO survives quantum fluctuations. (Note that the self-consistent MSWT calculations become relatively unstable for small order parameters, which results in ragged phase boundaries.)

In the Néel phases, the ED order parameter (see Fig. 5, right panel) is maximal, giving support to the assumption that here LRO persists. However, it is much smaller in the spiral phase than the MSWT value, a discrepancy already found in the SATL.\(^ {20}\) (This could be due to third-order corrections to

![FIG. 4. (Color online) Quantum-mechanical phase diagram of the SCATL, ordering vector. (Top) MSWT data. Quantum fluctuations stabilize the Néel phase. Around \(J' \approx J'' \approx J\), a part of the classical spiral phase survives quantum fluctuations (labels in the upper left panel). The solid symbols (upper right panel) denote some experimental materials (see Table I). (For clarity, we show only symbols in the lower right part of the figure, excluding points symmetric under exchange of \(J, J',\) and \(J'')\). (Bottom) ED data for \(N = 15\) sites. Already for this small system, it can be appreciated that (compared to the classical case) the Néel phase grows at the expense of spiral order.)](image-url)
FIG. 5. (Color online) Quantum-mechanical phase diagram, order parameter. ED results qualitatively confirm MSWT. In particular, the order parameter for both methods decreases rapidly upon approaching the MSWT breakdown regions.

the spin-wave expansion, which our approach neglects, and which can become important in spiral configurations.57)

Between the ordered phases, we find a broad region where MSWT breaks down, indicating as usual20,41 that these regions do not allow a description in terms of an ordered, semiclassical state. Therefore it appears that it is a quite universal feature of frustrated quantum antiferromagnets that spiral- and collinearly ordered phases are always separated by quantum disordered phases. This is the first main result of this paper.

The strong decrease of the MSWT and ED order parameters (see Fig. 5) upon approaching this region gives support to this interpretation (which we will further corroborate in the next two sections). Note also that both the ED and MSWT order parameter seem to disappear more smoothly when approaching the putative 1D-like QDP (consider, e.g., in the range $2 \lesssim J'/J \lesssim 3$, $J''/J \rightarrow 1$). Upon approaching the putative large-$\alpha$ QDP dividing spiral from N´eel LRO, on the other hand, for ED, the order parameter decreases sharply (consider, e.g., the line $J'/J = 1$, $J''/J \rightarrow 1$). Here, for MSWT, the breakdown occurs abruptly at finite order parameters. This could point at a difference in the type of phase transition upon approaching the large-$\alpha$ QDP and the disordered region at the decoupled-chains limit.

The second main result of our paper concerns experimental measurements of ground-state behavior of some materials, taken from Ref. 37 (see also the reviews Refs. 58 and 59) as well as from Refs. 60 (Cs$_2$CuCl$_4$) and 61 (Cs$_2$CuBr$_4$). For reference, they are presented in Table I and included as solid symbols in the upper right panel of Fig. 4. We mark magnetically disordered materials (spin liquids, VBS: valence-bond solid, RVB: resonating valence-bond states) within MSWT, symmetrizing the couplings puts Sb-0 just at the border to the breakdown region (which should be a lower limit for a disordered phase in the true ground state) and As-2 within it. The appearance of AFM N´eel LRO in these experiments may find, therefore, a simple explanation in the full anisotropy of the SCATL. This second main result of our paper shows how crucial the full anisotropy is for the interpretation of experimental data. The rest of this section is devoted to fleshing these main findings out.

2. Supporting observables from MSWT: spin stiffness and spin-wave velocities

In Refs. 20 and 41, the spin-stiffness tensor, which characterizes the stiffness of the magnetic order under change of the ordering vector, has proven a valuable consistency check of our MSWT calculations. Its components are

$$\rho_{\alpha\beta} = \frac{d^2F}{dQ_\alpha dQ_\beta}, \quad (4)$$

where $F$ is the free energy. Even if the order parameter is finite, a small spin stiffness suggests that further quantum fluctuations than taken into account within MSWT could disrupt the remaining order.94

Since for our purposes an upper bound for the spin stiffness is sufficient, we take the partial derivative in Eq. (4). The exact spin stiffness can be computed via the total derivative. To this, within the self-consistent MSWT calculations, one first has to find the optimal ordering vector. Then, one renurs the self-consistent MSWT equations for several fixed, slightly nonoptimal ordering vectors, yielding slightly larger energies. The spin stiffness can be derived by fitting a quadratic form to the resulting energy landscape. In the self-consistent iteration, the mean fields characterizing the MSWT state can adjust to a changed ordering vector. If one neglects the anisotropy between $J'$ and $J''$, taking as usual the mean of both couplings, they would lie at the position of the empty squares at $(J'/J,J''/J) = (1.57,1)$ [equivalent to $(J'/J,J''/J) = (0.64,0.64)$] and $(J'/J,J''/J) = (1.50,1)$ [equivalent to $(J'/J,J''/J) = (0.67,0.67)$], inside a phase where many methods15,16,18,19 predict disorder; specifically, within MSWT, symmetrizing the couplings puts Sb-0 just at the border to the breakdown region (which should be a lower limit for a disordered phase in the true ground state) and As-2 within it. The appearance of AFM N´eel LRO in these experiments may find, therefore, a simple explanation in the full anisotropy of the SCATL. This second main result of our paper shows how crucial the full anisotropy is for the interpretation of experimental data. The rest of this section is devoted to fleshing these main findings out.

<table>
<thead>
<tr>
<th>Material</th>
<th>$(J'/J, J''/J)$</th>
<th>State</th>
</tr>
</thead>
<tbody>
<tr>
<td>N-3</td>
<td>(10, 9.1)</td>
<td>AFM</td>
</tr>
<tr>
<td>P-2</td>
<td>(2.63, 1.89)</td>
<td>AFM</td>
</tr>
<tr>
<td>Sb-2</td>
<td>(2.08, 1.79)</td>
<td>CO</td>
</tr>
<tr>
<td>Sb-1</td>
<td>(1.72, 1.49)</td>
<td>SL</td>
</tr>
<tr>
<td>κ-CN</td>
<td>(1.41, 1.41)</td>
<td>SL</td>
</tr>
<tr>
<td>P-1</td>
<td>(1.32, 1.32)</td>
<td>VBS</td>
</tr>
<tr>
<td>Cs</td>
<td>(1.16, 1.16)</td>
<td>CO</td>
</tr>
<tr>
<td>Sb-0</td>
<td>(0.74, 0.60)</td>
<td>AFM</td>
</tr>
<tr>
<td>Cs$_2$CuBr$_4$</td>
<td>(0.74, 0.74)</td>
<td>AFM</td>
</tr>
<tr>
<td>As-2</td>
<td>(0.69, 0.58)</td>
<td>AFM</td>
</tr>
<tr>
<td>Cs$_2$CuCl$_4$</td>
<td>(0.34, 0.34)</td>
<td>RVB</td>
</tr>
</tbody>
</table>

TABLE I. Some relevant materials for which the ground state has been measured in experiment, together with the coupling strengths, and the state they are found to be in (from Refs. 37, 60, 61 and references therein). AFM: antiferromagnetic LRO, CO: charge-ordered, SL: spin-liquid, VBS: valence-bond solid, RVB: resonating valence-bond states.
putative small-\(\alpha\) QDP, not only the total inter-chain, but also the total intrachain spin stiffness decreases strongly. Since such a behavior is not consistent with algebraic correlations along the chains, this can be interpreted as an indication of a gapped nonmagnetic region. The partial spin stiffness computed in Ref. 20, on the other hand, only vanishes in the interchain direction. Hence it may not be able to distinguish gapped from gapless spin liquids. However, it still seems to adequately capture the location of disordered regions.

In Fig. 6, upper panel, we show the determinant of the spin-stiffness tensor, \(\det(\rho)\), normalized to the coupling strengths \(1 + J' + J''\). As we should expect, \(\det(\rho)\) decreases upon approaching the phase transitions, especially from the Néel-ordered side. At large \(J' (J'')\), this decrease is due to a softening of the stiffness in \(x\) (\(y\)) direction, and at small \((J'/J, J''/J)\) in the direction perpendicular to \(\tau_1\) (as has also been found in Ref. 20).

Another indicator for approaching disordered phases is given by the spin-wave velocities \(v_{x,y}\), which can be connected to the spin stiffness via the susceptibility.\(^{62}\) Since the spin-wave velocities are defined as the leading order of an expansion of the spin-wave dispersion relation, Eq. (A7), around small \(|k|\), i.e.,

\[
\begin{align}
    v_x &= \lim_{k_x \to 0} \frac{\omega_k}{k_x} \bigg|_{k_y = 0}, \\
    v_y &= \lim_{k_y \to 0} \frac{\omega_k}{k_y} \bigg|_{k_x = 0},
\end{align}
\]

they can be measured directly from the spin-wave dispersion, allowing an experimental check of our findings.

As seen in Fig. 6, lower panels, close to the 1D breakdown region, they, too, soften in the direction perpendicular to the dominating coupling. On the other hand, when approaching the putative large-\(\alpha\) QDP, the spiral from the Néel phase, both spin-wave velocities remain finite. This is another (besides the different behavior of the order parameter) indication that the large-\(\alpha\) disordered phase could be qualitatively different from the disordered state found in the limit of decoupled chains.

3. Supporting observables from ED: energy derivative, gap, and chiral correlations

The ED observables investigated in Sec. III B1 allowed to interpret the predominant ordering behavior, but did not yield clear evidence if within ED really quantum phase transitions exist, and if yes, where. The second derivative of the ED ground-state energy per spin, plotted in Fig. 7, can provide such an indicator. In the thermodynamic limit, it diverges at a quantum phase transition.

Indeed, there are clear peaks at lines similar to where in MSWT the Néel order breaks down. Also, a peak appears around \((J', J'') = (1, 1)\). This might be a precursor of a quantum phase transition away from the spiral state, and to an intermediate phase, possibly to the quantum disordered region that is supposed to exist in this system.

We get further support for this phase diagram from the ED energy gap between ground and first excited state (see Fig. 8).

In the well-known limiting cases of the SCATL, it behaves as expected: there is no singlet gap close to the decoupled-chains limits, since the system is then in a critical phase. In the Néel ordered phases, there is a large gap that separates the ground state from closely spaced excitations, which in larger lattices become the spin waves, collapsing slowly towards the ground state.\(^{10}\) This is consistent with the considerable size dependence found for these parameter regions, as can be seen in the right panels of Fig. 8, where we plot cuts of the gap per spin, \(\Delta E^{\text{ED}}\), at fixed \(J''/J = 1, 2, 3\) for triangular systems similar to the one in Fig. 1 with \(N = 6, 10, 15\).

On the contrary, there is no gap in the spiral phase, because there are two degenerate ground states with opposite chirality.\(^{35}\) We find that this vanishing of the gap depends strongly on the system geometry, but it occurs consistently for all triangular systems considered.

Interestingly, the gapless spiral phase is surrounded by a region where the gap attains considerable values. The very

FIG. 6. (Color online) (Top) The partial spin stiffness decreases upon approaching the MSWT breakdown region, suggesting the disruption of magnetic LRO. (Bottom) The spin-wave velocities perpendicular to the dominating coupling strength soften in the 1D limits. Differences in the spin-wave velocities might allow to measure the anisotropy of the SCATL. All quantities are normalized to the coupling strengths \(1 + J' + J''\).

FIG. 7. (Color online) Second derivative of ED ground-state energy per spin for \(N = 15\). For clarity, we plot the logarithm after a shift to values larger one, \(L(\partial^2 E^{\text{ED}}/\partial J''^2)\), where \(L(x) = \ln[1 + \max(x) - x]\), and \(J'' = J'\) or \(J''\). Strong peaks mark the transitions from the Néel phases. An additional peak around \((J', J'') = (1, 1)\) might be an indication of an additional phase, separating the Néel phases from the spiral one.

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small dependence on system size for this parameter region indicates that this is stable towards the thermodynamic limit. A finite gap is not consistent with a spiral-ordered phase. On the other hand, the predominant order in this region is different. Scanning along $J'/J$, within MSWT, it is smallest around $J'' = 2J$, while for ED it decreases monotonously with increasing $J''$. As the right panels in Fig. 8 indicate, this discrepancy could be due to finite-size effects. Indeed, we find in ED that for $J''/J = 2$ the transition point at $J'/J = 1.5$ shows an appreciable size dependence, while the transition points for $J''/J = 3$ do not. Therefore the lateral extent of the possibly nonmagnetic region at around $J'/J = 2$ could decrease with $N$, making the MSWT and ED pictures consistent.

From the gap, it seems that there is support for an extended gapped phase separating spiral and Néel LRO. Still, it would be desirable to exclude for this region spiral LRO in the thermodynamic limit. For this, we now study where chiral correlations persist. The vector chirality is defined as

$$\kappa_{i,j,k} = \frac{2}{3\sqrt{3}} (S_i \times S_j + S_j \times S_k + S_k \times S_i)_z ,$$

where the sites $[i,j,k]$ are located counterclockwise on a triangle. For the small systems used in our ED, we generalize the chiral correlations to $\Psi^- = \frac{4}{N_A} \sum_c \sum_a s_a \kappa_a \sum_a s_a \kappa_a$ ,

where $s_a$ runs over all triangles, while $c$ runs only over the central ones to reduce boundary effects. The factors $s_a \kappa_a$ weight $\kappa_a$ with $a = (+)$ sign if the triangle points upwards (downwards). The prefactor, where $N_A$ is the number of summands, is chosen such that the chiral correlation has the same theoretical maximum of 9/4 as the usual definition for large lattices.

As can be seen from the ED results of the $N = 15$ lattice (see Fig. 9, left panel), the chiral correlations are relatively small in the Néel phases and largest in the spiral phase around $(J'/J, J''/J) \approx (1,1)$. However, at this lattice size, there are still appreciable chiral correlations in the rest of the parameter regime. In particular, in the 1D limit, they are only a little smaller than in the spiral phase. Therefore we also plot in Fig. 9, right panel, an extrapolation to large lattices by $\Psi_-(N) = \Psi_-(N = \infty) + \frac{\Delta \Psi_-(N)}{\Delta N} \frac{1}{N}$, where we use the known form for the leading finite-size behavior but also include subleading corrections due to the small systems under consideration (our data comes from lattices with $N = 7, 10, 12, 15, 18$, all chosen to have an equal number of $J$, $J'$, and $J''$, as sketched at the bottom of Fig. 9). This shows a clear trend, namely that the chiral correlations only survive in a small region around $(J'/J, J''/J) = (1,1)$, roughly where the vanishing gap indicated the spiral phase. This would mean that outside this region there is no spiral LRO.
With this, we have several independent observations from ED indicating the existence of a magnetically disordered phase surrounding the spiral phase: the increase of the gap when leaving the central region around \((J'/J, J''/J) = (1,1)\) and the disappearance of chiral LRO for large lattices both suggest that there is no spiral LRO in this region. On the other hand, the predominant order is at incommensurate ordering vectors, indicating that this phase is also not Néel ordered. Therefore it seems natural to assume that this region could host a nonmagnetic phase, possibly gapped far away from the 1D limit and gapless close to it, consistent with MSWT.

C. MSWT spin-wave dispersion relations

Finally, to connect to experiment, we provide the spin-wave dispersion relations \(\omega_k\), as computed from MSWT, Eq. (A7). In Fig. 10, we show parameters corresponding to a point from the spiral phase and the magnetically ordered materials listed in Table I. We also provide (where applicable) a comparison to the dispersion relation which would result if two of the couplings were equal. These comparisons can be seen more quantitatively in the cuts (c.i-iii) shown in the lowest row of Fig. 10. For the point from the spiral phase (a.i), the symmetrization (b.i) does not significantly change the dispersion relation, but for P-2 and, especially, for Sb-0, the differences are considerable. The latter in particular changes even qualitatively since a symmetrization would put it instead of into a Néel phase into a spiral phase. These differences seem significant enough to be measurable in experiment. Such a measurement could allow to quantify the actual magnitude of coupling anisotropies.

IV. THE QUANTUM SCATL WITH XY INTERACTIONS

Frustrated magnetic models, such as the Heisenberg SCATL, are fundamental to our understanding of condensed-matter physics, but solving them theoretically is a difficult task. This difficulty is mainly due to the exponential complexity of quantum-mechanical Hilbert space. In the 1980’s, Feynman first formulated the idea to solve these models by simulation on another, dedicated quantum device. Measurements on such a device, now known as a quantum simulator, would reveal properties of a quantum model that are otherwise difficult to access. As a clean and well-controllable quantum-simulator platform for strongly correlated lattice systems, including magnetic models, ultracold atoms in optical lattices have reached now very sophisticated levels (see, e.g., Refs. 67–69

![Diagram](https://example.com/diagram.png)
and references therein). In the following, we discuss how a model similar to the Heisenberg SCATL, the XY SCATL, can be realized in such a setup. We will show that its ground-state properties are very similar to the Heisenberg model, thus allowing a useful quantum simulation of phenomena occurring in magnetic materials.

**A. Frustrated XY models with ultracold atoms in optical lattices**

At low temperatures, ultracold bosonic atoms in optical lattices are typically well described by the Bose-Hubbard Hamiltonian

\[ \mathcal{H}_{\text{BH}} = \sum_{\langle i,j \rangle} \frac{t_{ij}}{2} (b_i^\dagger b_j + \text{H.c.}) + \frac{U}{2} \sum_i n_i (n_i - 1), \]

where \( b_i, b_i^\dagger \) are bosonic operators, \( n_i = b_i^\dagger b_i \), and \( \langle i,j \rangle \) represents pairs of nearest-neighbor (NN) sites. Frustration in the intersite hopping is characterized by AFM hopping amplitudes with positive sign, \( t_{ij} = |t_{ij}| \). These can be realized, for instance, by coupling the bosons to an artificial magnetic field, or by a periodical shaking of the optical lattice.

In the following, we will focus on the limit of infinite repulsion \( U \to \infty \) and half-filling \( \langle n_i \rangle = 1/2 \), under which the Bose-Hubbard model maps to the \( S = 1/2 \) XY Hamiltonian, given by Eq. (1) with \( \lambda = 0 \). For consistency with the literature on Bose–Hubbard models, we denote the spin-spin couplings by \( t \)’s instead of \( J \)’s. XY models describe the behavior of a number of systems: they can be regarded as the limiting case of AFM Hamiltonians with planar anisotropy in the couplings, which describe frustrated AFM materials, and they govern the physics of planarly trapped ions loaded into an optical lattice or Cooper pairs in arrays of ultrasmall Josephson junctions.

Recently, periodic driving of an optical lattice has allow to experimentally realize the XY SATL in an ultracold-atoms setup. As described in Ref. 75, the bare tunneling matrix elements \( -|t_{ij}| \), connecting vertices of a triangular lattice such as depicted in Fig. 1, are dressed by the periodic driving with a factor \( \langle e^{i\alpha t} \rangle \). Here, the double brackets denote time averaging and \( A \cdot r_{ij} \) is the projection of the periodic driving orbit, \( A \), on the vector \( r_{ij} \) connecting sites \( i \) and \( j \). For example, if \( A \) is chosen as a simple sine-wave, after the time averaging the bare tunneling becomes multiplied by a Bessel function, the argument of which is determined by the driving amplitude. If the driving amplitude is such that the Bessel-function attains negative values, the sign of the tunneling is flipped (\( -|t_{ij}| \to |t_{ij}| \)), thus creating AFM interactions leading to geometrical frustration. In Ref. 76, this has been used to realize the AFM SATL, although that experiment was at weak \( U \), which corresponds to the classical limit. However, expectations are high that soon also the regime of strong repulsion \( U \to \infty \) may be reached. In that regime, the XY SATL is particularly interesting, because of predictions for several spin-liquid phases similar to the Heisenberg SATL.

Another implementation of the XY SATL within reach of current technology is in trapped-ion setups. Here, the bosonic particles governed by Hamiltonian (8) are provided by the vibrational modes of the ion crystal. The tunneling matrix elements \( t_{ij} \) can be tuned via the preferred direction of vibration of the ions, and superposing an optical lattice allows to create anharmonicities that play the role of a strong on-site repulsion \( U \). Proof-of-principle experiments using trapped ions to simulate spin models without and also with frustration let it seem realistic that in the near future the SATL with a mesoscopic number of sites can be implemented.

Ultracold-atom experiments as in Ref. 76 and trapped-ions implementations after the proposal can easily be generalized to the SCATL with its three different tunneling matrix elements (see Fig. 1). For this, one simply has to choose an elliptical driving orbit or a preferred direction of ion vibration, respectively, which is not parallel to any of the sides of the triangular plaquettes. Besides its fundamental interest for studying the occurrence of quantum-disordered phases, the SATL is relevant for characterizing their sensitivity with respect to imperfect driving, i.e., driving which does not create two perfectly equal couplings.

In the rest of this section, we discuss the ground-state phase diagram of the quantum XY SCATL. Our analysis proceeds along the same lines as for the Heisenberg SCATL, and, interestingly, the results are qualitatively extremely similar.

**B. Known results in limiting cases**

In this section, we discuss well-known limiting cases of the quantum XY SCATL, including results on the SATL, in order to assess which phases to expect. The qualitative behavior is very similar to the Heisenberg model, but the decreased symmetry in the XY case mitigates the effects of quantum fluctuation. Again, in the square-lattice limit, \( t'/t, t''/t = (1,0), (t'/t, t''/t) = (0,1) \), and \( t'/t, t''/t \gg 1 \), Néel LRO persists in the quantum case. Similarly, in the isotropic triangular lattice, \( t' = t'' = t \), spiral LRO survives quantum fluctuations. The limits \( t'/t = 0, (t'' \to \infty \text{ with } t'' = \text{const}) \), and \( t''/t \to \infty \text{ with } t' = \text{const} \) correspond to ensembles of decoupled, critical XY-chains, with algebraic correlations along individual chains but no correlations between them.

The XY SCATL (recovered for \( t' = t'' \equiv \alpha t \), or, equivalently, \( t' = t \) or \( t'' = t \)), has previously been studied with entangled-pair states (PEPS) calculations and MSWT (as well as ED). For comparison, Fig. 11 reproduces the MSWT phase diagram found in Ref. 41.

As with Heisenberg interactions, quantum fluctuations considerably stabilize the Néel order occurring at large \( \alpha \), moving the point where it disappears downwards to values between \( \alpha \approx 1.4 \) (PEPS, Ref. 17) and \( \alpha \approx 1.66 \) (MSWT, Ref. 41). Both methods predict that quantum fluctuations spread the transition point between the Néel and the spiral phase into a “large-\( \alpha \) quantum-disordered phase” (QDP), similar to what was found in the Heisenberg SATL.

At the low-\( \alpha \) side of the spiral phase, Ref. 17 predicts a “small-\( \alpha \) spin liquid,” consisting of a gapped disordered phase for as large \( \alpha \) as \( \approx 0.6 \), followed by a gapless disordered phase in the region \( 0 \lesssim \alpha \lesssim 0.4 \). Similarly, MSWT computations indicate a quantum-disordered phase below \( \alpha \approx 0.18 \), but the very small spin stiffness at already \( \alpha \approx 0.35 \) suggests that taking quantum fluctuations into account more completely than within MSWT could destabilize the weak magnetic order already at larger \( \alpha \).
FIG. 11. (Color online) The MSWT phase diagram of the quantum XY SATL (from Ref. 41) contains Néel order (which is considerably more stable than in the classical model), spiral order (which is destabilized by quantum fluctuations), and two putative spin-liquid (SL) phases. These are found through the breakdown of the theory and a disappearing spin stiffness, which indicates a gapped disordered phase. In the purely 1D limit, MSWT recovers the 1D critical state. We include sketches of classical states, where blue arrows indicate the directions of the classical rotors, namely, the 1D state at \( t'/t = 0 \), the spiral state at \( t'/t = 1 \), and the 2D-Néel state at \( \alpha \geq 2 \).

C. Quantum-mechanical phase diagram

As with Heisenberg interactions, we compute the quantum-mechanical phase diagram of the SATL using MSWT supplemented with ordering-vector optimization, working directly in the thermodynamic limit, and compare these results to exact diagonalization (ED) of a 15-site lattice, as depicted in Fig. 1.

1. MSWT and ED results

To obtain a first overview over the phase diagram, we study the ordering vector \( \mathbf{Q} \) and the order parameter \( M \). In ED, these can be extracted similar to Eq. (3) from the static structure factor, which for \( XY \) interactions we define as

\[
S(k) = \frac{1}{N^2} \sum_{i,j} e^{i(k \cdot r_i - r_j)} \langle S^x_i S^x_j + S^y_i S^y_j \rangle .
\]

The results are very similar to the Heisenberg case: the MSWT and ED ordering vectors, presented in Fig. 12, show that quantum fluctuations stabilize the Néel phases compared to the classical case, and in the central region around \( t' \approx t'' \approx t \), a broad range of incommensurate ordering vectors indicates spiral order. In the Néel phases, the ED order parameter (see Fig. 13, right panel) is maximal, hinting at strong ordering tendencies, and we find again a much smaller MSWT order parameter in the spiral phase than what is predicted by ED.

As with Heisenberg interactions, between the ordered phases, we find a broad region where MSWT breaks down, hinting at disordered ground states. The ED and MSWT order parameters decrease upon approaching this region, as does the spin stiffness, Eq. (4), as shown in the upper panel of Fig. 14. These findings lend support to the assumption that the MSWT breakdown region could host a magnetically disordered phase.

Similar to the Heisenberg SCATL, both the ED and MSWT order parameter seems to disappear relatively smoothly when approaching the putative 1D-like QDP (consider, e.g., in the range \( 2 \lesssim t'/t \lesssim 3, t''/t \rightarrow 1^- \)). On the other hand, when approaching the putative large-\( \alpha \) QDP dividing spiral from Néel LRO, the ED order parameter decreases more sharply (consider, e.g., the line \( t'/t = 1, t''/t \rightarrow 1^- \)), and for MSWT the breakdown occurs abruptly at finite order parameters. This points at a difference in the type of phase transition upon approaching the putative large-\( \alpha \) QDP and the disordered phase at the decoupled-chains limit. The spin-wave velocities, Eq. (III B2), give another hint in this direction. As seen in the lower panels of Fig. 14, close to the 1D breakdown region, they, too, soften in the direction perpendicular to the dominating coupling. On the other hand, when approaching the putative large-\( \alpha \) QDP dividing spiral from the Néel phase, both spin-wave velocities remain finite. This suggests that the large-\( \alpha \) disordered state could be qualitatively different from the disordered phase found in the limit of decoupled chains.

2. Supporting observables from ED: energy derivative, gap, and chiral correlations

To find stronger indications for the position of quantum phase transitions within ED, we consider the second derivative of the ED ground-state energy(see Fig. 15). We find clear peaks along lines similar to where in MSWT the Néel order breaks

FIG. 12. (Color online) Quantum-mechanical phase diagram of the SCATL, ordering vector. (Top) MSWT data. Quantum fluctuations stabilize the Néel phase. Around \( t' \approx t'' \approx t \), a part of the classical spiral phase survives quantum fluctuations (labels in the upper left panel). (Bottom) ED data for \( N = 15 \) sites. Already for this small system, it can be appreciated that the Néel phase grows at the expense of spiral order (compared to the classical case).
FIG. 14. (Color online) (Top) The partial spin stiffness decreases upon approaching the MSWT breakdown region, suggesting the disruption of magnetic LRO. (Bottom) The spin-wave velocities perpendicular to the dominating coupling strength soften in the 1D limits. Differences in the spin-wave velocities might allow to measure the anisotropy of the SCATL. All quantities are normalized to \(1 + t' + t''\).

down. Also, a peak appears around \((t', t'') = (1, 1)\). This might be interpreted as the precursor of a quantum phase transition away from the spiral state, possibly to the quantum-disordered region indicated by MSWT.

From the ED energy gap, Fig. 16, we get further support for the phase diagram extracted so far. It behaves similar to Sec. III B 3. There is no singlet gap close to the decoupled-chains limits where the system is critical. In the Néel ordered phases, a large gap separates the ground state from closely spaced excitations, which in larger lattices become the spin waves, collapsing slowly towards the ground state. This is consistent with the considerable size dependence in that parameter region (see right panels of Fig. 16). In the spiral phase, there is no gap due to the degeneracy of the two ground states with opposite chirality.

As for Heisenberg interactions, the gapless spiral phase is surrounded by a region with a relatively large gap that shows very small system-size dependence. It only closes upon approaching the Néel phases, indicating a quantum phase transition. Such a finite gap is not consistent with a spiral-ordered phase. On the other hand, the predominant order in this region is at incommensurate wave vectors, so that it can also not be due to Néel physics. Optimistically, these findings could therefore be interpreted as the precursors of a gapped QDP.

We now study which of these regions carry LRO of chiral correlations [see Eq. (7)]. Similarly to the Heisenberg case, the ED results on the \(N = 15\) lattice yield large chiral correlations in the spiral phase around \((r', t, t'') = (1, 1)\) (see Fig. 17, left panel). In the Néel phases the chiral correlations are small, but at this lattice size, they still reach appreciable strengths in the rest of the parameter regime. To study how they persist in the thermodynamic limit, we extrapolate these results to large lattices. For this, we use the known form for the leading finite-size behavior and include the next subleading correction: \(\Psi_-(N) = \Psi_-(N = \infty) + \frac{c_1}{N^{\gamma_1}} + \frac{c_2}{N^{\gamma_2}}\). The result, plotted in the right panel of Fig. 17, shows a clear trend, namely that the chiral correlations only survive in the central region around \((t'/t, t''/t) = (1, 1)\). This would mean that outside of this region, there is no spiral LRO. In particular, it disappears close to the decoupled chains and in the Néel phases. However, from this data, it appears that chiral order extends all the way from \((t'/t, t''/t) = (1, 1)\) to boundaries of the Néel phases, which would preclude a gapped spin liquid intervening between the spiral and the Néel phase. However, a gapped chiral phase without magnetic LRO but with chiral LRO would still be consistent with MSWT and previous results from PEPS. Such gapped chiral states constitute

FIG. 15. (Color online) Second derivative of ED ground-state energy per spin for the \(N = 15\) system. For clarity, we plot the logarithm after a shift to values larger one, \(L(\partial^2 E^{ED}/\partial t^m)\), where \(L(x) = \ln[1 + \max(x) - x]\) and \(t''\) is \(t'\) or \(t''\). Strong peaks clearly mark the phase transitions from the Néel phases. An additional peak around \((t', t'') = (1, 1)\) might be an indication of an additional phase, separating the Néel phases from the spiral one.

FIG. 16. (Color online) (Left) The singlet gap from ED gives support to the MSWT phase diagram: a finite gap separates in the Néel phases spin-wave excitations from the ground state. In the spiral phase, the ground state is doubly degenerate due to the ambiguity in choice of chirality. The finite gap surrounding the degenerate region could be a precursor of a gapped, disordered phase. At the quantum phase transitions to the Néel phases, the gap closes again. (Right) Cuts at fixed \(r''/t = 1, 2, 3\) for triangles with increasing \(N\) (from light to dark and thick to thin: 6, 10, 15). There is little size dependence in the central gapped phase \((r'' = 2, 3t, t' \approx t\) as well as \(r'' = t\) and \(t'/t \gtrsim 1.5\)).
The geometries used in the extrapolation are chosen for symmetry. LRO seems to extend all the way to a Néel ordered phase. However, in some parts of the phase diagram, chiral XY disordered phase. Already for small systems, it appears that chiral LRO only survive in a central region around \((t'/t, t''/t) = (1, 1)\), lending support to the appearance of an extended disordered phase. However, in some parts of the phase diagram, chiral LRO seems to extend all the way to a Néel ordered phase. The geometries used in the extrapolation are chosen for symmetry upon rotation by 60° and equal number of \(t, t', t''\) bonds.

an interesting class of quantum phases in their own right, with most occurrences being very subtle (see for example the \(J_1J_2\) chain or the chiral Mott insulators in Bose-Hubbard ladders). This result is the only remarkable quantitative difference between the phase diagrams of the \(XY\) and the Heisenberg SATL.

D. MSWT predictions for time-of-flight pictures

Finally, we wish to connect our predictions to experiment. A well-established experimental technique for ultracold atoms is time-of-flight (ToF) imaging of the atom momentum distribution,

\[
n_b(k) = \frac{1}{N} \sum_{i,j} e^{i k (r_i - r_j)} \langle b_i^\dagger b_j \rangle.
\]

Since the bosons of Hamiltonian (8) have infinite repulsion, \(U \to \infty\), this is equivalent to measuring the magnetic structure factor \(S(k)\), Eq. (9), for the \(XY\) spins.

Figure 18 presents ED predictions (for \(N = 15\)) for ToF images at various values of anisotropy. The uppermost row shows parameter values from the SATL. The other rows show from top to bottom results for increasing additional anisotropies in steps of 10%. The black lines denote the first Brillouin zone (first BZ). Commensurate 120° spiral order has peaks at the corners of the first BZ (as in the first panel of the second row), while peaks at the center of two opposing sides of the first BZ mark Néel order (as in the lower two panels of the second row). Incommensurate spiral order is characterized by peaks lying between these two limiting cases (as in the second panel of the second row). Close to the 1D limit, the peaks decrease in magnitude and smear out strongly along a straight line (as seen in the first panel of the first row). In large systems, disordered phases are characterized by a sub-extensive growth of the peak height.

As seen in the ToF pictures in Fig. 18, the additional anisotropy can clearly shift the system from one phase to a qualitatively different one. For example, the momentum distributions in the first row pass from an almost 1D-like spiral state to an adjacent Néel phase. Similar behavior is found for other values of \((t'/t, t''/t)\). Such ToF pictures, therefore, would allow to observe the influence of the additional anisotropy in experiment.

V. CONCLUSION

In conclusion, we have provided a thorough analysis of the ground-state phase diagram of the quantum SATL with Heisenberg interactions, which describes a variety of organic magnetic salts. We also discussed how a related model with \(XY\) couplings can be realized in ultracold-atoms and trapped-experiments, thus allowing the quantum simulation of an interesting magnetic model.

Using various observables from modified spin-wave theory supplemented with ordering-vector optimization, and supported by exact diagonalization data, we have found for both types of coupling that quantum fluctuations stabilize Néel order with respect to the classical phase diagram. Further, quantum fluctuations reduce the extent of the spiral phase, which seems to be entirely surrounded by a quantum-disordered region. This result, which constitutes our first main finding, is supported by the breakdown of MSWT, together with the strong decrease of the order parameter and the spin stiffness. While MSWT cannot be applied to studying this region, the fact that \(no\) semiclassical reference state describable by an ordering vector yields a stable solution is highly suggestive of a magnetically disordered phase of purely quantum origin. Hence, our results outline a very promising candidate region for such exotic states, meriting further research with more sophisticated theoretical methods or experimental setups.

The appearance of a quantum-disordered region derives support from ED data, with a finite gap and a strong decrease of the structure-factor peak. For ED, in this region the structure-factor peak is located at incommensurate wave vectors. In the Heisenberg model, chiral long-range correlations seem to vanish while they may remain finite for the \(XY\) model. This is the only fundamental difference that we found between the two types of interactions. It could hint at a gapped phase with chiral LRO appearing in the \(XY\) model on the SATL.

Our results suggest that collinear and spiral magnetic order are never directly connected, but instead always a nonmagnetic phase intervenes. This behavior has also been found in related models, both with Heisenbergs and \(XY\) interactions. It, therefore, seems to emerge as a quite universal feature of strongly frustrated two-dimensional systems with continuous symmetry in their spin couplings.

A complete encircling of the spiral phase by disordered phases could also naturally explain the succession of a gapped and a gapless quantum-disordered phase close to the decoupled-chains limit of the Heisenberg and the \(XY\) SATL. The gapless QDP would be continuously connected to the
limit of decoupled chains, while the additional anisotropy of the SCATL would adiabatically connect the gapped QDPs at small and large $\alpha$. This shows the great potential of the additional anisotropy to deliver new insights into the nature of these phases.

In this context, the behavior of the order parameter and the spin-wave velocities indicate that the transitions from the Néel phase to the putative disordered region close to the 1D limit could be qualitatively different from the one at larger $\alpha$.

Our second main finding is connected to experimental results for the Heisenberg SCATL: measurements on magnetic organic salts find magnetic LRO in materials which theoretical analyses on the SATL predict to be magnetically disordered. We show that this discrepancy finds a simple explanation in the additional anisotropy of the SCATL, which is neglected in the SATL. Taking it into account, we predict these material to lie in magnetically ordered phases, in accordance to experiment. Remarkably, they only come to lie in the Néel phase because of the stabilization of Néel order by quantum fluctuations. From the point of view of experiments in ultracold atoms, the persistence of the quantum-disordered phase under the additional anisotropy suggests that it can also be observed under slightly imperfect driving (i.e., a driving that is not completely symmetric under exchange of two bonds).

Finally, for Heisenberg interactions we provided spin-wave dispersion relations, and for $XY$ interactions predictions for the boson momentum distribution in time-of-flight pictures. A comparison to these might allow to probe the additional anisotropy experimentally.

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APPENDIX: MSWT FORMALISM

In this appendix, we shortly review the MSWT formalism. For more details for Heisenberg interactions, see Ref. 20, and for XY models, see Ref. 41.

A fundamental assumption of spin-wave theory is that the ground state has LRO with ordering vector \( \mathbf{Q} \). Hence it is convenient to rotate the local reference system as

\[
S_i^+ = -\sin (\mathbf{Q} \cdot \mathbf{r}_i) S_i^0 + \cos (\mathbf{Q} \cdot \mathbf{r}_i) S_i^\xi, \tag{A1a}
\]

\[
S_i^- = \cos (\mathbf{Q} \cdot \mathbf{r}_i) S_i^0 + \sin (\mathbf{Q} \cdot \mathbf{r}_i) S_i^\xi, \tag{A1b}
\]

\[
S_i^\xi = -S_i^0. \tag{A1c}
\]

Then \( S_i^\xi \), which will be the quantization axis, lies parallel to the classical spin \( S_i = (\cos (\mathbf{Q} \cdot \mathbf{r}_i), \sin (\mathbf{Q} \cdot \mathbf{r}_i), 0) \). This defines the classical reference state. We do not make any assumption on the ordering vector \( \mathbf{Q} \). In particular, it may well differ from the one of the classical limit (\( \mathbf{Q}^{(c)} \)).

Spin waves around this reference state can be described by the Dyson-Maleev (DM) transformation, which maps the physical spins to interacting bosons,

\[
S_i^+ \rightarrow \frac{1}{\sqrt{2S}} (2S - a_i^\dagger a_i) a_i, \tag{A2a}
\]

\[
S_i^- \rightarrow \sqrt{2S} a_i^\dagger, \tag{A2b}
\]

\[
S_i^\xi = -S + a_i^\dagger a_i, \tag{A2c}
\]

where \( S_i^\xi = S_i^0 \pm i S_i^0 \).

This transformation maps the Hamiltonian (1) to the nonlinear bosonic Hamiltonian

\[
\mathcal{H} = \frac{1}{4} \sum_{\langle ij \rangle} J_{ij} \left[ 2S(a_i^\dagger a_j + a_i a_j^\dagger) - a_i^\dagger a_i^\dagger a_j a_j^\dagger - a_i^\dagger a_i a_j^\dagger a_j^\dagger \right] \times (\mathbf{Q} \cdot \mathbf{r}_{ij})
\]

\[
+ \left[ 2S(a_i^\dagger a_j^\dagger + a_i a_j) - a_i^\dagger a_i a_j^\dagger a_j - a_i^\dagger a_j a_i^\dagger a_j^\dagger \right] \times (\mathbf{Q} \cdot \mathbf{r}_{ij})
\]

\[
+ 4\left( S^2 - S(a_i^\dagger a_i^\dagger a_j a_j + a_i^\dagger a_i a_j^\dagger a_j^\dagger) \right) \cos (\mathbf{Q} \cdot \mathbf{r}_{ij}), \tag{A3}
\]

where \( a_i \) (\( a_i^\dagger \)) destroys (creates) a DM boson at site \( i \), and \( S \) is the length of the spin. Here, we neglected the kinematic constraint that restricts the DM-boson density \( n \) to the physical subspace \( n < 2S \). Moreover, we dropped terms with six boson operators, which are of order \( O[n/(2S)^3] \) and are negligible for \( n/(2S) < 1 \). Using Wick’s theorem and defining the correlators \( \langle a_i a_j^\dagger \rangle = F(r_{ij}) - \frac{1}{2} \delta_{ij} \) and \( \langle a_i a_j \rangle = \langle a_i^\dagger a_j^\dagger \rangle = G(r_{ij}) \), the expectation value \( E \equiv \langle \mathcal{H} \rangle \) can be written as

\[
E = \langle \mathcal{H} \rangle = \sum_{\langle ij \rangle} \frac{J_{ij}}{2} \left[ \left( S + \frac{1}{2} - F(0) \right) F(r_{ij}) - F(r_{ij}) \right]^2 \times [\lambda + \cos (\mathbf{Q} \cdot \mathbf{r}_{ij})] - \left[ S + \frac{1}{2} - F(0) \right] \times \left[ \lambda - \cos (\mathbf{Q} \cdot \mathbf{r}_{ij}) \right] \right]. \tag{A4}
\]

After Fourier transforming, \( a_k = \frac{1}{\sqrt{N}} \sum a_i e^{-i k \cdot r_i} \), and a subsequent Bogoliubov transformation, \( a_k = \cos \theta_k a_k - \sin \theta_k a_{-k}^\dagger \) and \( a_{-k}^\dagger = -\sin \theta_k a_k + \cos \theta_k a_{-k}^\dagger \), we minimize the free energy \( \mathcal{F} \) under the constraint of vanishing magnetization at each site, \( \langle a_i^\dagger a_i \rangle = S \), which is known as Takahashi’s modification. This yields a set of self-consistent equations,

\[
\tanh 2 \theta_k = \frac{A_k}{B_k} \tag{A5}
\]

with

\[
A_k = \frac{1}{N} \sum_{\langle ij \rangle} J_{ij} [\lambda - \cos (\mathbf{Q} \cdot \mathbf{r}_{ij})] G_{ij} e^{i k \cdot r_{ij}}, \tag{A6a}
\]

\[
B_k = \frac{1}{N} \sum_{\langle ij \rangle} J_{ij} [\lambda - \cos (\mathbf{Q} \cdot \mathbf{r}_{ij})] G_{ij}
\]

\[
- [\lambda + \cos (\mathbf{Q} \cdot \mathbf{r}_{ij})] F_{ij} (1 - e^{i k \cdot r_{ij}}) - \mu, \tag{A6b}
\]

where \( \mu \) is the Lagrange multiplier for Takahashi’s constraint. The spin-wave spectrum reads

\[
\omega_k = \sqrt{B_k^2 - A_k^2}. \tag{A7}
\]

At \( T = 0, \mu \) vanishes, which implies the disappearance of the gap at \( k = 0 \) that may exist for finite temperature. This is a necessary condition for magnetic LRO, and enables Bose condensation in the \( k = 0 \) mode. Separating out its contribution, \( \langle a_{k=0} a_{k=0}^\dagger \rangle / N = \langle a_{k=0} a_{k=0}^\dagger \rangle / N \equiv M \), \( A8 \)

which corresponds to the magnetic order parameter, one arrives at the zero-temperature equations

\[
F_{ij} = M + \frac{1}{2N} \sum_{k \neq 0} \frac{B_k}{\omega_k} \cos (k \cdot r_{ij}), \tag{A9a}
\]

\[
G_{ij} = M + \frac{1}{2N} \sum_{k \neq 0} \frac{A_k}{\omega_k} \cos (k \cdot r_{ij}), \tag{A9b}
\]

and the constraint of vanishing magnetization at each site becomes

\[
\sum_{k \neq 0} \frac{B_k}{\omega_k}. \tag{A10}
\]

It is not \textit{a priori} clear that the classical ordering vector \( \mathbf{Q}^{(c)} \) correctly describes the LRO in the quantum system. To account for a competition between LRO at different ordering vectors \( \mathbf{Q} \), we extend the MSWT procedure by optimizing the free energy \( \mathcal{F} \) with respect to the ordering vector \( \mathbf{Q} \). This yields two additional equations which must be added to the set of self-consistent equations,

\[
\frac{\partial \mathcal{F}}{\partial Q_x} = 0, \tag{A11a}
\]

\[
\frac{\partial \mathcal{F}}{\partial Q_y} = 0. \tag{A11b}
\]
The values of \( F_{ij} \) and \( G_{ij} \) can now be calculated by solving self-consistently Eqs. (A6)–(A11). Through Wick’s theorem the knowledge of the quantities \( F_{ij} \) and \( G_{ij} \) determines the expectation value of any observable.
92Although in the last material the physics is dominated by a superposition of AFM and ferromagnetic 1D chains.
93On the other hand, some magnetically disordered materials lie in ordered regions of the MSWT phase diagram. It is known, however, that MSWT overestimates ordered phases.
94Also, if the spin stiffness is small when approaching the MSWT breakdown region, we are led to assume that the breakdown is not caused by numerical problems, but that it is a physical effect, i.e., due to the lack of a description in terms of a semi-classically ordered reference state.
95In the spiral phase, there is a gap, similar to the spin-wave gap of the Néel phases, between the second and the third energy level. The smaller peaks around $(J'/J, J''/J) = (3, 1)$ and $(J'/J, J''/J) = (1, 3)$ result from the strong geometry dependence for the small lattices used. It can be understood that these peaks are artifacts, because it is highly implausible that the chiral LRO first disappears when increasing the one-dimensionality and then finds a revival.