Response to Prof. László Udvardi

May 29, 2025

1 Question

As stated in reference [1], $ZrCl_3$ was a potential candidate for the physical realization of the SU(4) spin liquid. It was later revealed that the system undergoes dimerization [2], thus invalidating the description of its electronic structure as a spin liquid. What does dimerization mean in this case? Is it similar to Peierls instability? If we describe the system using a Hubbard model, is it possible to determine a range of parameters where this instability occurs?

Answer

Yes, the dimerization happens precisely as in the Peierls instability of a 1D chain. The authors of Ref. [2] calculated the parameters of the Hubbard model which characterize α-$ZrCl_3$, but they have not calculated the range of the parameters, in which this dimerization happens. I have not found such calculations in the articles citing Ref. [2] either.

The authors of Ref. [2] argue that the original article [3] claiming that the Zr atoms in α-$ZrCl_3$ form layers of almost perfect honeycomb lattices, and that the Cl atoms create an octahedral environment for the Zr atoms seems to be wrong. Therefore, they tried to establish a probable candidate for the crystal structure of α-$ZrCl_3$ using Density Functional Theory. Their results suggest that the honeycomb layers of Zr atoms are distorted in a way, that pairs of Zr atoms are closer to each other than to the rest of surrounding Zr atoms, as shown in Fig. 1.

Let me cite the appropriate parts of their article: "We optimized cell volume, its shape and atomic positions in the structural relaxation, which was performed until the total energy change from one ionic iteration to another was larger than 0.1 meV... Surprisingly the lowest in energy turned out to be not a uniform structure (all Zr-Zr bonds are the same), but the dimerized one with dimers being parallel to each other... the dimerization is a result of the formation of

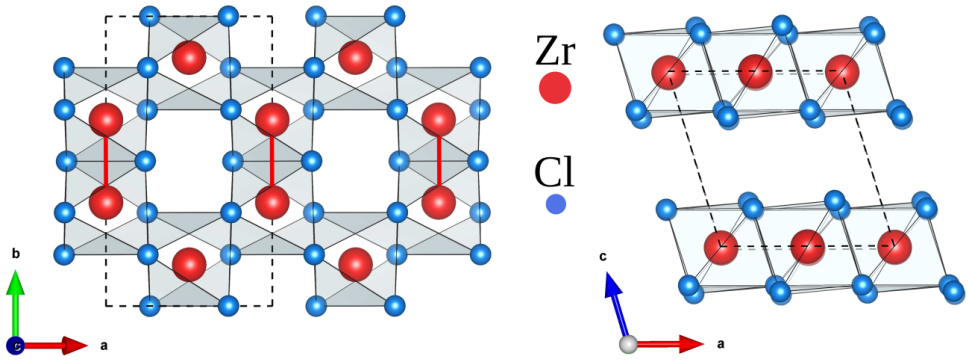

Figure 1: The left panel shows the distorted hexagon of Zr (red) atoms, so that the Zr atoms which are connected with a red line are closer to each other than to the other surrounding Zr atoms. These figures were taken from Ref. [3] (Fig. [1.]).

strong molecular bonding (or orbital order) between two t_{2g} orbitals looking towards each other. Stabilization of a single (per site) electron at the particular orbital will kill SU(4) invariance of the spin-orbital model.

It is worth noting that while α-$RuCl_3$ has a structure with nearly regular hexagons (and this is exactly what was obtained for this compound in our calculations), it is known to dimerize under a tiny pressure of 0.2 GPa [15]. Furthermore, it dimerizes exactly in the same structure, featured by parallel orientation of the Ru-Ru dimers, as α-$ZrCl_3$ in our calculations at the ambient pressure."

Let's suppose that their calculations are correct, and α-$ZrCl_3$ indeed dimerizes at ambient pressure. The question arises whether this dimerization also happen at lower pressure. Furthermore, the authors of Ref. [1] claimed that in the class of materials $\alpha - MX_3$, with M = Ti, Zr, Hf, etc. and X = F, Cl, Br, etc. all materials are good candidates to realize the antiferromagnetic SU(4) Heisenberg model. Thus, perhaps another material of this family will be a better realization of the model.

2 Question

In Figure 3.2 of Chapter Three, the fifth column shows the stability of the Dirac spin liquid as a function of next-nearest-neighbor and ring exchange couplings for different mean-field hopping structures. In the figure, the region where the spin liquid state is stable becomes smaller and smaller. Is there an explanation for this trend?

Answer

I have included the two Figures about stability from my dissertation, shown in Figs. 2 and 3 (the Figure in the question is the latter). In the white regions of the local stability figures, the Monte Carlo error did not allow us to conclude about the stability of the DSL, so it is difficult to compare the areas of the regions of stability (the DSL might be stable in the complete white area). The error is much smaller in the last two rows of Fig. 3, because the contour lines in the $\Delta\langle\mathcal{P}_{1\mathrm{st}}\rangle$ and $\Delta\langle\mathcal{P}_{\triangle} + \mathcal{P}_{\triangle}^{-1}\rangle$ are almost perfect spheres, so it was much easier to fit an ellipsoid, then on the rows above.

At the mean-field level, the smallest region of stability of the DSL is expected for those ansatze, which can open a gap at the Fermi energy (just as in the Peierls instability). Among the ansatze discussed in my dissertation, only the T_{1g} (first two rows in Fig. 3) and T_{2g} (first two columns in 2) can open a gap at the Fermi energy. However, the Gutzwiller projector can change the energies in an unexpected way. For example, at the mean-field level the David star T_{1g} has lower energy than the DSL, but the Gutzwiller projector makes the DSL lower in energy. Therefore, the mean-field arguments are not very strong.

3 Question

Figure 6.7 shows the decay of spin correlation on a logarithmic scale for SU(4) and SU(6) spin liquids. In both cases, the exponent falls between three and four. Is there any explanation for this?

Answer

Yes, at the mean-field level, the correlation function of every two-dimensional Dirac spin liquid decays as r^{-4} [4, 5]. As the SU(N) symmetry is raised, the fluctuations beyond the mean-field approximation are expected to become weaker, and the power of the algebraic correlations

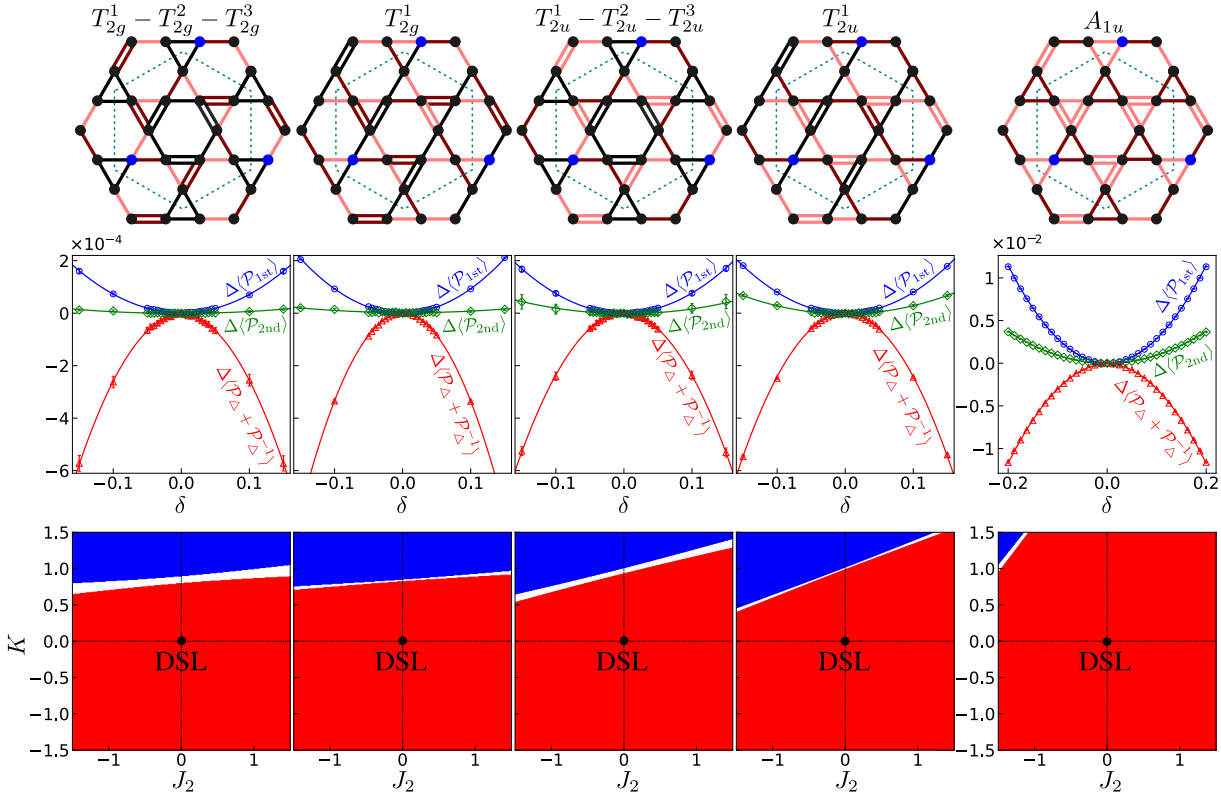

Figure 2: In the first row, we show the hopping structure of the real perturbations of the Dirac spin liquid having a single free parameter δ. Different shades represent different absolute values of the hoppings. The empty bonds stand for positive hoppings, while the solid bonds for negative hoppings (each ansatz has a $\pi_{\hexagon}\pi_{\triangle}\pi_{\triangledown}$ flux structure, just as the DSL). The black bonds have absolute value 1, the dark red hoppings $1+\delta$, and the light reds $1-\delta$. In the middle row, the red points show the $\Delta\langle\mathcal{P}_\triangle + \mathcal{P}_\triangle^{-1}\rangle$, the blue points $\Delta\langle\mathcal{P}_{1\text{st}}\rangle$, and the green points $\Delta\langle\mathcal{P}_{2\text{nd}}\rangle$, while the solid lines are the fitted parabolas. The bottom row shows the local stability of these ansätze, as a function of K and J_2, fixing $J_1 = 1$. The DSL is the lowest energy state in the red region, and the perturbation wins in the blue region. All these results were calculated for a cluster of 192 sites with APBC.

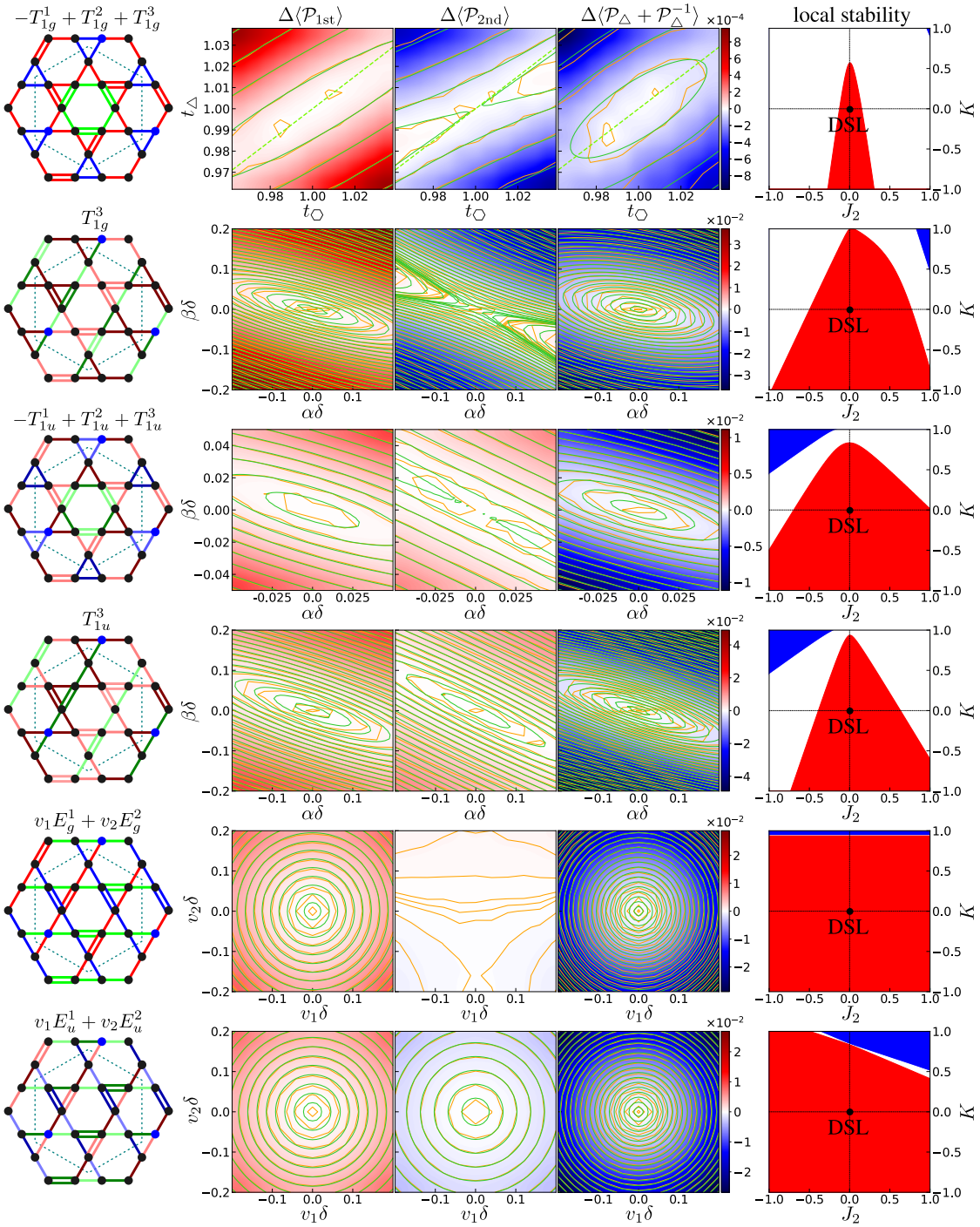

Figure 3: In the first column, we show the hopping structure of the real perturbations of the Dirac spin liquid having two free parameters. Different colors represent different absolute values of the hoppings for the denoted irreducible representation. The empty bonds stand for positive hoppings, while the solid bonds for negative hoppings (each ansatz has a $\pi_\hexagon\pi_\triangle\pi_\triangledown$ flux structure, just as the DSL). The second column shows the $\Delta\langle\mathcal{P}_{1st}\rangle$, the third $\Delta\langle\mathcal{P}_{2nd}\rangle$, and the fourth $\Delta\langle\mathcal{P}_\triangle+\mathcal{P}_\triangle^{-1}\rangle$, calculated by VMC for a cluster of 192 sites with APBC, the $\delta=0$ is the DSL. The contours of the fitted ellipsoids are shown in light green, while the contours of the data are orange. The fifth column shows the local stability of these ansätze, as a function of K and J_2, keeping $J_1=1$. The DSL has the lowest energy in the red region and the perturbed ansatz of the given row in the blue region.

should approach -4. Since SU(4) and SU(6) are already high symmetries, we expect powers not very different from -4.

4 Question

In Chapter Five, the author defines the Heisenberg chain with the following Hamiltonian operator:

$$H = J \sum_{i=1}^{N_s} \sum_{a=1}^{8} T_i^a T_{i+1}^a \tag{1}$$

In the SU(2) case, if the scalar coupling J is replaced with a tensorial coupling, the resulting Dzyaloshinskii–Moriya interaction can significantly influence the ground state. Can we expect a substantial change if we extend the model to the following form:

$$H = \sum_{i=1}^{N_s} \sum_{a,b=1}^{8} J_{a,b} T_i^a T_{i+1}^b ? \tag{2}$$

Answer

Yes, we expect relevant changes.

Diagonal $J_{a,b}$

Let me start with the simplest case, when the matrix $J_{a,b}$ is diagonal $J_{a,b} = J_a \delta_{a,b}$, but not the identity. If $J_1 = J_2 = J_3 = J$ and $J_4 = J_5 = J_6 = J_7 = J_8 = J'$, but $J \neq J'$, then we recover the SU(2) symmetric bilinear biquadratic Hamiltonian in the spin-1 irrep

$$\mathcal{H} = \cos\theta \sum_{i=1}^{N_s} \mathbf{S}_i \cdot \mathbf{S}_{i+1} + \sin\theta \sum_{i=1}^{N_s} (\mathbf{S}_i \cdot \mathbf{S}_{i+1})^2. \tag{3}$$

discussed in the fourth answer of the response to Prof. Oroszlány.

The most striking change can be achieved if we set every $J_a = 0$ except $J_3 \neq 0$, for which we get the Ising model $J_3 \sum_i S_i^z S_{i+1}^z$, which no longer has a continuous spin rotation symmetry, so the Mermin Wagner theorem does not prevent the ground state to be ordered. For $J_3 > 0$, the two degenerate ground states are antiferromagnetically ordered $|\uparrow,\downarrow,\uparrow \ldots\rangle$, $|\downarrow,\uparrow,\downarrow \ldots\rangle$, and the spectrum is gapped.

Off-diagonal $J_{a,b}$ in the SU(2) case

In the SU(2) case, the spin-orbit coupling in the underlying Hubbard model results in off-diagonal $J_{a,b}$

$$J_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{D}_{i,j} \cdot (\mathbf{S}_i \times \mathbf{S}_j) + \mathbf{S}_i \cdot \mathbf{\Gamma}_{i,j} \cdot \mathbf{S}_j \tag{4}$$

via the superexchange mechanism, as discussed by Moria in Ref. [6]. The Dzyaloshinskii–Moriya (DM) interaction

$$\mathbf{D}_{i,j} \cdot (\mathbf{S}_i \times \mathbf{S}_j) \equiv \sum_{a,b,c=1}^{3} D_{i,j}^a \varepsilon_{a,b,c} S_i^b S_j^c \tag{5}$$

is antisymmetric due to the Levi-Civita symbol $\varepsilon_{a,b,c}$. Since the $\mathbf{D}_{i,j}$ vector chooses a preferred direction, the DM interaction breaks down the continuous SU(2) global spin rotation symmetry to a U(1) symmetry.

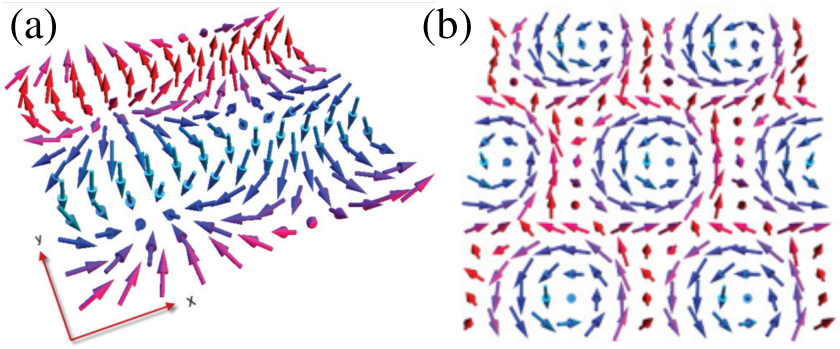

Figure 4: (a) Double periodic spiral order (b) skyrmion crystal. Both figures were taken from Ref. [8] (Figs. [2.] and [3.], respectively).

As shown in Ref. [7], the fermionic half-filled Hubbard model at strong on-site repulsion $U/t \gg 1$ in the presence of spin-orbit coupling leads to the tilted antiferromagnetic Heisenberg model

$$H = \sum_{\langle j,l \rangle} \frac{4t_{jl}^2}{U} \mathbf{S}_j^T \mathcal{J}_{j,l} \mathbf{S}_l, \tag{6}$$

where $\mathbf{S}_l$ is a column vector containing S_l^x, S_l^y, and S_l^z. Furthermore,

$$\mathcal{J}_{j,l}\mathbf{S}_l = \cos(2\theta_{j,l})\mathbf{S}_l + \sin(2\theta_{j,l})(\mathbf{S}_l \times \mathbf{n}_{j,l}) + 2\sin^2(\theta_{j,l})\mathbf{n}_{j,l}(\mathbf{n}_{j,l} \cdot \mathbf{S}_l), \tag{7}$$

where the angles $\theta_{j,l}$ and the axis $\mathbf{n}_{j,l}$ depend on the hoppings and the spin-orbit interaction in the underlying Hubbard model. These three terms correspond to the three terms in Eq. (4), and the tilted Heisenberg model (6) is equivalent to the proposed new Hamiltonian with off-diagonal exchanges (2). The first two terms in Eq. (7) describe an SO(3) rotation of the spin operator. In Ref. [8] the authors treat a similar, but ferromagnetic tiled Heisenberg model, where they keep only the first two terms in Eq. (7), describing an SO(3) rotation of the spin operator. They obtain spiral, and skyrmion crystal solutions on the square lattice, shown in Figs. 4(a) and (b), respectively. The form of the tilted Heisenberg model provides an intuition for these magnetic orders, since if the $J_{a,b}$ acts as an SO(3) rotation and we interpret the spin operators classically, then the interaction is like $\mathbf{S}_j^T \mathcal{J}_{j,l} \mathbf{S}_l = \mathbf{S}_j \mathbf{S}_l'$, where $\mathbf{S}_l'$ was "rotated".

Off-diagonal exchanges in the SU(3) case

The $\varepsilon_{a,b,c}$ appear in the commutation relations of the SU(2) Lie algebra $[S^a, S^b] = i\hbar \sum_{c=1}^{3} \varepsilon_{a,b,c} S^c$.

In the SU(3) case, the most similar interaction to the antisymmetric DM interaction was introduced in Ref. [9], by replacing the $\varepsilon_{a,b,c}$ in the DM interaction (5) with the structure constants $f_{a,b,c}$ appearing in the commutation relations of the SU(N) Lie algebra $[T^a, T^b] = i\hbar \sum_{c=1}^{8} f_{a,b,c} T^c$, since the $f_{a,b,c}$ are antisymmetric in the exchange of any two indices. Thus, the generalized DM interaction can be written as

$$\mathbf{D}_{i,j} \cdot (\mathbf{T}_i \times \mathbf{T}_j) \equiv \sum_{a,b,c=1}^{8} D_{i,j}^a f_{a,b,c} T_i^b T_j^c. \tag{8}$$

This interaction was claimed to create a spiral order in the one-dimensional *ferromagnetic* SU(3) Heisenberg chain, though they explicitly broke SU(3) symmetry and restricted the energy minimalization to a polarized subspace.

The starting point of Ref. [9] is the Bose-Hubbard model with threefold pseudospin degree of freedom, describing three component bosonic quantum gases in optical lattices. The effective

model at one boson per site is the ferromagnetic SU(3) Heisenberg model, and the generalized DM interaction (8) was derived from the spin-orbit coupling of the underlying Hubbard model.

Similarly to the SU(2) cases, the SU(3) ferromagnetic Heisenberg model in the fundamental representation on the square lattice was shown to form CP^2 skyrmion crystals in Refs. [10, 11].

I found no research articles considering the generalized DM interaction for the *antiferromagnetic* SU(3) Heisenberg model. However, the generalized DM interaction breaks the global SU(3) spin rotation symmetry, since the $\mathbf{D}$ vectors choose a preferred direction. Therefore, I would expect the introduction of the generalized DM interaction to cause severe changes to the quantum spin liquid ground state of the antiferromagnetic Heisenberg model.

5 Question

What effect could the introduction of next-nearest-neighbor interactions have on the Heisenberg chain discussed in Chapter Five?

Answer

As a consequence of the second nearest neighbor interactions, the ground state becomes trimerized, similarly as in the SU(2) model, where the ground state becomes dimerized (Majumdar-Ghosh model).

Majumdar-Ghosh model in the spin-1/2 irrep of the SU(2) symmetric model

In the spin-1/2 representation of the antiferromagnetic SU(2) Heisenberg model with second-nearest-neighbor interactions

$$\mathcal{H}^{J_1-J_2} = J_1 \sum_{i=1}^{N_s} \mathbf{S}_i \cdot \mathbf{S}_{i+1} + J_2 \sum_{i=1}^{N_s} \mathbf{S}_i \cdot \mathbf{S}_{i+2} \tag{9}$$

for weak $0 \leq J_2 < 0.2411 J_1$ (and $J_1 > 0$) the ground state is a quantum spin liquid with fractionalized spin-1/2 gapless excitations [12]. However, for $J_2 > 0.2411 J_1$ the ground state becomes dimerized [12]. At $J_2 = J_1/2$ the two degenerate ground states were constructed exactly by Majumdar and Ghosh [13] as direct products of singlet states formed from nearest neighbor spins $|\mathrm{GS}_1\rangle = [12][34]\dots[N_s-1, N_s]$ and $|\mathrm{GS}_2\rangle = [23][45]\dots[N_s, 1]$ (where $[ij] \equiv \frac{1}{\sqrt{2}}(|\uparrow_i\downarrow_j\rangle - |\downarrow_i\uparrow_j\rangle)$, N_s is even, and we imposed periodic boundary condition). This can be understood by rewriting the Hamiltonian as

$$\begin{aligned}\mathcal{H}^{J_1-J_2} &= J_1 \sum_{i=1}^{N_s} \mathbf{S}_i \cdot \mathbf{S}_{i+1} + J_2 \sum_{i=1}^{N_s} \mathbf{S}_i \cdot \mathbf{S}_{i+2} = J_1/2 \sum_{i=1}^{N_s} (\mathbf{S}_i \cdot \mathbf{S}_{i+1} + \mathbf{S}_{i+1} \cdot \mathbf{S}_{i+2} + \mathbf{S}_i \cdot \mathbf{S}_{i+2}) \\ &= \frac{J_1}{4} \sum_{i=1}^{N_s} \left((\mathbf{S}_i + \mathbf{S}_{i+1} + \mathbf{S}_{i+2})^2 - \frac{9}{4} \right)\end{aligned} \tag{10}$$

as shown in Refs. [14, 15]. The dimerized states $|\mathrm{GS}_1\rangle$ and $|\mathrm{GS}_2\rangle$ have the smallest possible eigenvalue, because they are eigenstates of all $(\mathbf{S}_i + \mathbf{S}_{i+1} + \mathbf{S}_{i+2})^2$ with the smallest possible eigenvalue 3/4.

Trimerization in the SU(3) model

In the fundamental representation of SU(3) (one fermion per site in the underlying Hubbard model) three neighboring SU(3) spins are needed to form a singlet state. Denoting the basis on

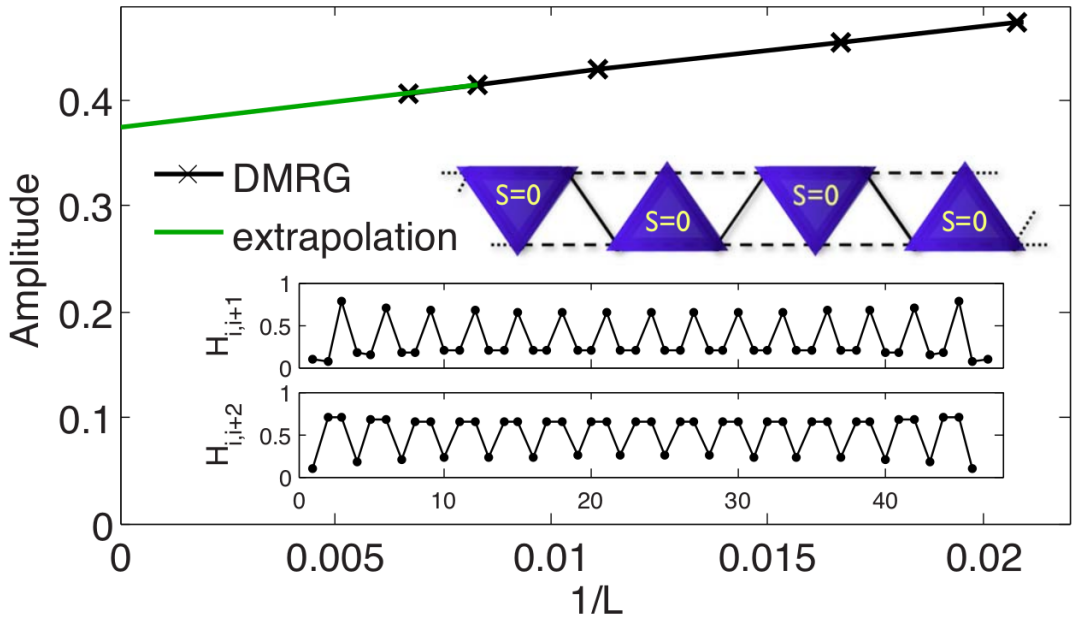

Figure 5: The important part of this figure is the inset, where $H_{i,i+1} \equiv \sum_{a=1}^{8} \langle \mathrm{GS}|T_i^a T_{i+1}^a|\mathrm{GS}\rangle$ and $H_{i,i+2} \equiv \sum_{a=1}^{8} \langle \mathrm{GS}|T_i^a T_{i+2}^a|\mathrm{GS}\rangle$. Both these quantities are smaller (corresponding to lower energy), when both sites i and $i+1$ (or i and $i+2$) are inside a trimer. Quoting the article [16]: "This pattern is formed because the two bonds belonging to a trimer have a lower energy than the bonds connecting two trimers." Furthermore, this oscillation amplitude does not vanish at infinite distances (long range trimer order), the extrapolated value is called a trimer order parameter. This figure was taken from Ref. [16] (Fig. [3.]).

every site by $|a\rangle$, $|b\rangle$, $|c\rangle$, we can write an SU(3) singlet state as $[ijk] \propto |a\rangle_i|b\rangle_j|c\rangle_k + |b\rangle_i|c\rangle_j|a\rangle_k + |c\rangle_i|a\rangle_j|b\rangle_k - |c\rangle_i|b\rangle_j|a\rangle_k - |b\rangle_i|a\rangle_j|c\rangle_k - |a\rangle_i|c\rangle_j|b\rangle_k$. If the sites i, j, and k are three neighboring sites, then we can call such a singlet state a trimer. One would think that the product of such trimer states $[123][456]\ldots[N_s-2, N_s-1, N_s]$ is an exact ground state for some J_2, but this turns out not to be true. For sufficiently large $J_2 \geq 0.5$ the ground state becomes similar to the trimerized state, in the sense that it shows similar correlations, but not exactly equal to it, based on a DMRG study [16]. The "similar" correlations are shown in Fig. 5.

According to Ref. [17], the trimerized state is an exact ground state of the following Hamiltonian,

$$\sum_{i=1}^{N_s} \left(\sum_{a=1}^{8} (T_i^a + T_{i+1}^a + T_{i+2}^a + T_{i+3}^a)^2 - \frac{4}{3} \right) \left(\sum_{a=1}^{8} (T_i^a + T_{i+1}^a + T_{i+2}^a + T_{i+3}^a)^2 - \frac{10}{3} \right), \qquad (11)$$

where after doing the multiplications we get terms like $T_i^a T_{i+3}^a$, corresponding to third nearest neighbor interaction. The authors of Ref. [17] also construct exact eigenstates for higher SU(N) symmetries, and other irreducible representations.

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