A $2D$ quantum antiferromagnet with a four-fold degenerate valence-bond-solid ground state

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We study the competition between antiferromagnetic order and valence bond solid formation in a two-dimensional frustrated spin-1/2 model. The J_1-J_2 model on the square lattice is further frustrated by introducing products of three-spin projectors which stabilize four dimer-product states as degenerate ground states. These four states are reminiscent of the dimerized singlet ground state of the Shastry-Sutherland model. Using exact diagonalizations, we study the evolution of the ground state by varying the ratio of interactions. For a large range of parameters ($J_2 \gtrsim 0.25J_1$), our model shows a direct transition between the valence-bond-solid phase and the collinear antiferromagnetic phase. For small values of J_2, several intermediate phases appear which are also analyzed.

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I. INTRODUCTION

In the last several years, a large amount of work has been devoted to the study of quantum systems with frustrated magnetic interactions due to their propensity to present spin liquid states. These phases indeed present interesting low energy properties, starting with the absence of magnetic order even at zero temperature. Recently, a lot of attention has been focused particularly on the Valence Bond Solids (VBS), and more precisely on the transition between these states and a magnetically ordered phase. In this context, it has been suggested that this transition could be a second order transition that does not belong to the Landau-Ginsburg paradigm[1,2].

In a VBS state, spins are coupled in pairs forming singlet states, evocating valence bonds. These pairwise singlets are themselves arranged in a periodic pattern. Such states break the translation symmetry, and it is possible to define an order parameter quantifying the singlet long range order. The question on the nature of quantum phase transition that separates this state from a long-range ordered magnetic phase is very interesting as well as current. Supposing a second order transition, according to Landau-Wilson paradigm[3], the order parameter of both phases should vanish precisely at the transition. It seems more likely that the two parameters will not vanish exactly at the same point, leading either to a first order transition, or to two second order transitions separated by an intermediate phase. Considering spin-1/2 on square lattice, Senthil and coworkers have recently suggested that there could exist second order transitions that are not described by Landau-Ginsburg theory[1,2]. The transition could instead be described by means of fractional degrees of freedom, namely spinons. These spinons become deconfined at the transition point, called the deconfined quantum critical point.

A priori, the spin-1/2 J_1-J_2 antiferromagnetic model on the square lattice seems the simplest choice to investigate the relevance of this theory. In the two limiting cases where either J_1 or J_2 is very large compared to the other, the system presents antiferromagnetic order. In the former case it corresponds to the usual Néel order, and in the latter case the ground state has a collinear antiferromagnetic order, corresponding to the Néel order on two sublattices (that are obtained by connecting second neighbor sites). It is generally accepted that this model presents an intermediate spin disordered phase in the range $0.4 < J_2/J_1 < 0.6$, which might break the translational symmetry[4,5,6,7,8,9,10]. It is still unclear if the transition between the Néel state and the spin disordered phase is a deconfined quantum critical point or a simple first order transition, although recent works are in favor of a weak first order transition[11,12].

The difficulty in understanding the J_1-J_2 model comes from the intermediate phase whose nature is subject to discussions[13]. Some studies show a four-spin plaquette order[9,10], while others are in favor of a columnar dimer order[6,7,8]. In the latter case, not only the translational symmetry, but also the rotational symmetry is broken. However, if this phase develops dimer-dimer correlations, it is far away from being the simple direct product of dimer singlet wave functions. Indeed, if that was the case, the ground state would present a strong signal of long range order which is not observed for the intermediate phase of the J_1-J_2 model. Exact diagonalization studies show that the dimer-dimer correlation is rapidly decreasing with distance[6,14], and that, if the dimer-dimer order exists, it should be rather small[14]. The analysis of the properties of the phase transition is therefore not so easy in the J_1-J_2 model. Recently, a new model including ring exchange has been proposed[15] to explore the possibility of non Landau-Ginsburg phase transitions. It has however been shown that this model presents a first order transition[15].

In the present paper, we propose a new quantum spin-1/2 model with frustrated antiferromagnetic interactions. Interestingly, for a simple choice of the interaction parameters, this model has an exactly solvable singlet ground state. The exact ground state is a pure direct product of dimers, arranged in the same pattern as that in the ground state of the Shastry-Sutherland model[16]. However, unlike in the Shastry-Sutherland model, the Valence Bond Solid (VBS) ground state of our model presents a case of spontaneous symmetry breaking, and is four-fold degenerate[17,18]. Using exact diagonalization, we investigate the evolution of the ground state as the interaction parameters are varied away from the exactly solvable VBS case, particularly towards magnetically ordered phases. In the following section, we describe the model and discuss its main properties and its relation to the J_1-J_2 model. In section III-A, we study the competition between the VBS phase and the antiferromagnetic collinear phase. In section III-B, we analyze the competition with the usual Néel antiferromagnetic phase. Finally, the last section is devoted to conclusions and perspectives.

II. MODEL

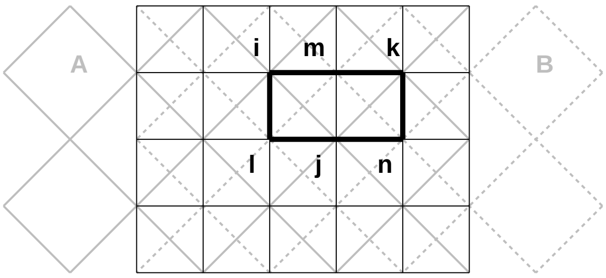

FIG. 1: Representation of a 3 by 2 horizontal rectangular plaquette. A vertical 2 by 3 plaquette can be obtained by a 90° rotation of the plaquette. Gray plain and dashed lines represent the nearest-neighor bonds of the A and B sub-lattices.

Consider a system of spin-1/2 on the square lattice. The model we consider contains only two and four spins interactions which can be conveniently described using a six-site rectangular plaquette (see Fig. 1). It is also convenient to distinguish between the two sublattices A and B of the square lattice. The six sites of a plaquette thus contain 3 sites belonging to the A sublattice and other 3 sites belonging to the B sublattice. For each set of three spins, we consider the spin projector on the quartet (spin 3/2) state:

$$P^A_{i,j,k} = |\vec{S}_i + \vec{S}_j + \vec{S}_k|^2 - \frac{3}{4} \tag{1}$$

$$P^B_{l,m,n} = |\vec{S}_l + \vec{S}_m + \vec{S}_n|^2 - \frac{3}{4} \tag{2}$$

where A and B refer to the two sublattices, and i, j, k, l, m, n are the sites of the plaquettes as depicted in Fig 1. The interaction we consider is obtained by taking the product of the two projectors of a plaquette:

$$H_0 = \sum_{[i,j,k,l,m,n]} \frac{1}{4}\, P^A_{i,j,k} P^B_{l,m,n} \tag{3}$$

where the sum runs over all horizontal and vertical plaquettes, and $[i, j, k, l, m, n]$ are the sites of one plaquette.

One should notice that the value of a projector is always positive except when the three spins are in a doublet state (that is when it becomes zero). The latter condition is fulfilled, in particular, if any two of the three spins form a singlet. These two spins could either be two first neighbor or two second neighbor spins of a sub-lattice, and correspond respectively to two second or two third neighbor spins of the original square lattice. Since the interaction in the model is the product of two projectors, it is only necessary to cancel one of these projectors to minimize the corresponding term. For this purpose, it is thus sufficient to have one dimer on the plaquette, either between second or third neighbor spins. Therefore, it is obvious that if a valence bond configuration has one such dimer on every plaquette, it will form a zero energy ground state of H_0.

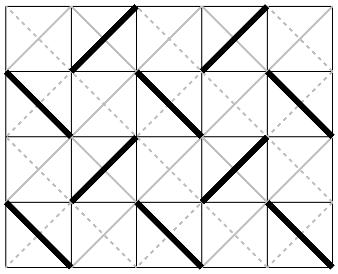

FIG. 2: Representation of one of the four ground states of model H_0. Black lines indicate the pairs of spins that form singlet states. The three other ground states can be obtained by translation of the dimer pattern by lattice vectors.

The idea of stabilizing valence bond states by spin projectors is also contained in the Majumdar-Ghosh model on the linear chain[19]. Recently, Batista and Trugman studied a model with such projectors on four-site square plaquettes[15], but their Hamiltonian has a highly degenerate ground state since a great number of dimer patterns minimize all projectors. In the present model, the ground state is obtained when dimers are arranged in the pattern given in Fig. 2 and is much less degenerate (see also the discussion in the Appendix). This arrangement evokes the Shastry-Sutherland model (and we will further refer to this phase as the SS-VBS phase), but contrary to the SS model, our Hamiltonian does not break the translational symmetry of the square lattice. The ground state presents a spontaneous symmetry breaking, and is four-fold degenerate. The other three of SS-VBS states can be obtained by translation of the dimer pattern by lattice vectors. Interestingly, the four SS-VBS states are also exact zero energy eigenstates (although not the ground state) of the nearest-neighbor Heisenberg model on the square lattice, which motivated us to seek for a model on the square lattice with four SS-VBS states as the exact ground state[20].

The spin projectors are two-spin operators which can be rewritten in terms of the exchange couplings between the first and second neighbor spins of a sublattice:

$$P^A_{i,j,k} = 2(\vec{S}_i \cdot \vec{S}_j + \vec{S}_i \cdot \vec{S}_k + \vec{S}_j \cdot \vec{S}_k) + \frac{3}{2} \quad (4)$$

Therefore, the product of two projectors generates two- and four-spin operators in H_0. As the projector is invariant under rotation and involves spins on a given sublattice, it will commute with the total spin of this sublattice, and obviously, with the total spin of the other sublattice. H_0 will thus conserve the total spin on each sublattice. This extra symmetry may introduce a further factor two in the degeneracy of excited states.

It is interesting to notice that this Hamiltonian is somehow related to the J_1-J_2 model. Let us assume that a state can be expressed as a direct product of the wave-function of spins on A sublattice $|\Psi_A\rangle$ and of spins on B sublattice $|\Psi_B\rangle$:

$$|\Psi\rangle = |\Psi_A\rangle \otimes |\Psi_B\rangle \quad (5)$$

In that case, it is possible to define a Hamiltonian for one sublattice. Let us, for instance, choose the A sublattice:

$$H_A = \sum_{[i,j,k,l,m,n]} \frac{1}{4} P^A_{i,j,k} \langle \Psi_B | P^B_{l,m,n} | \Psi_B \rangle \quad (6)$$

This Hamiltonian corresponds, for the A sublattice, to a model with first and second neighbor interactions whose amplitudes are modulated by the local spin state of the B sub-lattice. It is easy to show that, in the case where the mean value of the projector on B sublattice is homogeneous, the Hamiltonian can be expressed as:

$$H_A = \langle P^B \rangle \sum_{<i,j>_A} \vec{S}_i \cdot \vec{S}_j + \frac{1}{2} \langle P^B \rangle \sum_{\ll i,j \gg_A} \vec{S}_i \cdot \vec{S}_j \quad (7)$$

where $< i,j >_A$ are the first neighbor couples of spins of A sublattice, and $\ll i,j \gg_A$ the second neighbor ones, $\langle P^B \rangle$ is the uniform value of $\langle \Psi_B | P^B_{l,m,n} | \Psi_B \rangle$. This Hamiltonian is precisely the J_1-J_2 model on A sublattice at the $J_2/J_1 = 0.5$ point, which is located in the intermediate spin liquid phase of the model. Clearly, if we consider the ground state of our model, we do not expect the mean value of the P^B projector to be homogeneous. Nevertheless, this comparison introduces a simple picture of the model, in which the local spin correlations self-consistently modulate the J_1-J_2 model on each sub-lattice. This process allows the system to stabilize a pure direct product of dimers as ground state. This valence bond ground state is different in nature from the one expected in the intermediate phase of J_1-J_2 model, since the dimer order on each sub-lattice is not columnar but staggered[21] (see Fig. 2).

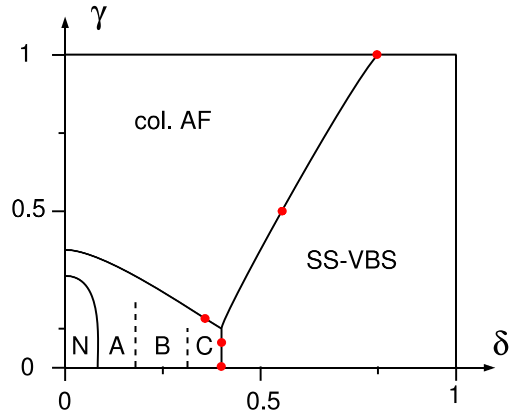

FIG. 3: (color online) Sketch of the phase diagram of the model (8). Phase transitions obtained by exact diagonalizations are indicated by full dots. The lines are guide to the eyes. The nature of the different phases is described in the following sections.

The existence of this exact ground state makes this model an interesting candidate to explore the transition between the VBS phase and the antiferromagnetically ordered state. In order to drive the system into such phases, we consider the following Hamiltonian:

$$\begin{aligned} H &= J(1-\gamma)(1-\delta) \sum_{<i,j>} \vec{S}_i \cdot \vec{S}_j \\ &+ J\,\gamma(1-\delta) \sum_{\ll i,j \gg} \vec{S}_i \cdot \vec{S}_j + J\,\delta\,H_0 \end{aligned} \quad (8)$$

where $< i,j >$ and $\ll i,j \gg$ are respectively first and second neighbor spins of the original (square) lattice, γ and δ are dimensionless parameters, and J is the energy scale (assumed to be positive). When δ is equal to 1, one just retrieves the H_0 model, while when $\delta = 0$ is zero, the Hamiltonian is simply the J_1-J_2 model, with γ being equal to $J_2/(J_1 + J_2)$. A sketch of the phase diagram of the Hamiltonian (8) is shown in Fig. 3. It contains three phases where the order has been unambiguously identified denoted by N (antiferromagnetic (π,π) Néel order), col. AF (collinear $(\pi,0)/(0,\pi)$ order with Néel order on the two sublattices) and SS-VBS (Valence-Bond order with Shastry-Sutherland arrangement). Between these phases, there is a region where correlations change very significantly, defining possible phase transitions between three phases denoted by A, B and C. These phases are discussed in the last part of the next section.

III. NUMERICAL RESULTS

We study the evolution of eigenstates of this model by an exact diagonalization technique based on the Lanczos algorithm. Finite clusters of $N = 16$, 20 and 32 sites were used. Larger clusters were not accessible because of the complexity of the interaction.

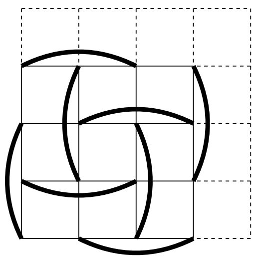

FIG. 4: Representation of one of the two additional ground states for the 16 site cluster. Full lines connect the 16 sites of the cluster, dashed lines symbolize the boundary conditions. The other additional ground state can be obtained by translation of the dimer pattern by lattice vectors.

The first noticeable result concerns the $\delta = 1$ case, when $H = JH_0$. We checked numerically that the four SS ground states presented in previous section are the only ground states of the model. This is always true except for the 16 site cluster, for which two more ground states are also present, as shown in Fig. 4. These two states are ground states only for the 16 site cluster, due to the very short loops which wrap around the boundaries of the cluster (see also the Appendix). Indeed, in this case a third neighbor dimer between the sites (x, y) and $(x, y+2)$ also represents a dimer between the sites $(x, y+2)$ and $(x, y+4)$ because of the periodic boundary conditions. This is no longer true for larger clusters and by extension in the thermodynamic limit.

In the following two subsections we map out the phase diagram of the model, with emphasis on two lines: In subsection III A we study first the case $\gamma = 1$, which connects to the case of $J_1 = 0$, $J_2 = J$ once $\delta = 0$. In subsection III B the case $\gamma = 0$ is presented, which connects the unfrustrated square lattice Heisenberg antiferromagnet to the fourfold dimerized ground state.

A. SS-VBS versus Collinear Order ($\gamma = 1$)

In the present section, we consider the case where $\gamma = 1$. According to the previous discussion on the relation between our model and the J_1-J_2 model, J_2 corresponds to a first neighbor interaction on each sublattice which, for large enough values, will lead to a Néel state on each sublattice.

For $\delta = 1$, the SS-VBS states are exact ground states even for finite clusters. Note that these four states are nonorthogonal on a finite cluster: they have a finite overlap that decreases exponentially with the cluster size[22]. However, since they are linearly independent, the ground state is indeed four-fold degenerate on a finite cluster for $\delta = 1$. This degeneracy is lifted however by the J_2 interactions that appear for $\delta < 1$. Fig. 5 represents the energy differences between the ground state and the lowest state of some symmetry sectors, obtained for the

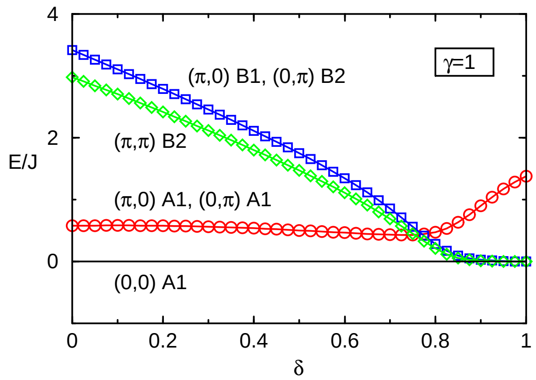

FIG. 5: (color online) Energy differences between the ground state and some of the lower states of the 32 site cluster. The symmetry of the different states is indicated on the figure. States with $(\pi, 0) - A_1$ and $(0, \pi) - A_1$ symmetry are triplet states, others are singlet states.

32 site cluster. The energy differences between the four lowest singlet states stay relatively small in the range $0.8 < \delta \leq 1$, while the spin gap progressively decreases. Below 0.8 the energy difference between singlet states rapidly increases, while the spin gap remains approximatively constant suggesting that the system is in the antiferromagnetic phase that will present a zero spin gap in the thermodynamic limit.

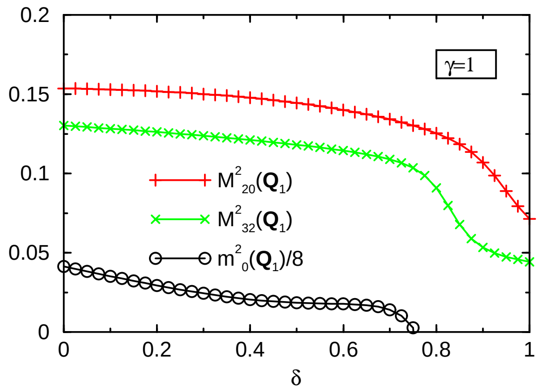

FIG. 6: (color online) $M_N(\vec{Q}_1)^2$ square magnetic susceptibility for 20 and 32 sites clusters and its extrapolation to thermodynamic limit $M_\infty(\vec{Q}_1)^2 = m_0^2(\vec{Q}_1)/8$.

In order to further investigate the transition, we calculate the $\vec{Q}$ dependent magnetic susceptibility for each cluster[25]:

$$M_N^2(\vec{Q}) = \frac{1}{N(N+2)} \sum_{i,j} \langle \vec{S}_i \vec{S}_j \rangle e^{i\vec{Q}(\vec{r}_j - \vec{r}_i)} \tag{9}$$

where $\vec{r}_i$ denotes the position of i^{th} spin, $\langle\rangle$ the mean value in the ground state and N the number of sites in the cluster. The evolution of the $\vec{Q}_1 = (\pi, 0)$ magnetic susceptibility relevant for antiferromagnetic collinear order is presented in Fig. 6. The extrapolation to the thermodynamic limit of the corresponding sublattice magnetization[4] has been performed using the finite size scaling predicted by non-linear sigma model studies[23,24]:

$$M_N^2(\vec{Q}_1) = \frac{1}{8} m_0^2(\vec{Q}_1) + \frac{\text{const.}}{\sqrt{N}} \tag{10}$$

The fitted value of $m_0(\vec{Q}_1)$ is also shown in Fig. 6. The extrapolated magnetization stays large up to $\delta \simeq 0.7$, which confirms that the collinear phase is stable in this range of parameter. It then rapidly drops, and vanishes around $\delta \approx 0.75$.

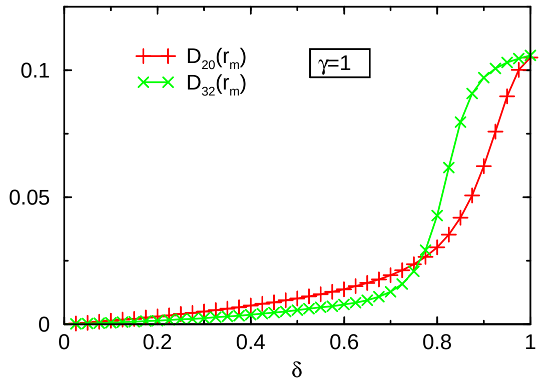

FIG. 7: (color online) Dimer-dimer correlation D_m obtained for the 20 and 32 sites cluster. This value correspond for one cluster to the correlation between the two dimers of the Shastry-Sutherland pattern separated by the largest distance.

The VBS phase, which is expected for larger values of δ, is characterized by long range dimer-dimer correlation. This long range correlation can be considered as the order parameter of this symmetry-breaking phase. In order to determine the stability of the phase, we compute the following dimer-dimer correlations in the ground state:

$$D_N(\vec{r}) = \langle (\vec{S}_{\vec{0}} \cdot \vec{S}_{\vec{r}_1})(\vec{S}_{\vec{r}} \cdot \vec{S}_{\vec{r}+\vec{r}_2}) \rangle - \langle \vec{S}_{\vec{0}} \vec{S}_{\vec{r}_1} \rangle \langle \vec{S}_{\vec{0}} \vec{S}_{\vec{r}_2} \rangle \tag{11}$$

where $\vec{0}$ stands for the origin, and $\vec{r}_1$ and $\vec{r}_2$ can be either equal to $(1,1)$, $(1,-1)$, $(-1,1)$ or to $(-1,-1)$. As expected, for $\delta \sim 1$, values of $D(\vec{r})$ that corresponds to the dimers of the Shastry-Sutherland pattern are quite large. For a given cluster, and close to $\delta = 1$, fluctuations of these values are very small, of the order of a few percent of the average value of these correlations. Since we are interested in the value of $D(\vec{r})$ for r going to infinity, we only considered among these correlations the one obtained for the largest r value ($D_N(\vec{r}_m)$). These correlations, shown in Fig. 7, are quite small in the antiferromagnetic phase, and rapidly increase at around $\delta \sim 0.8$. Interestingly enough, the curves cross at $\delta = 0.76$, very close to the point were the antiferromagnetic order vanishes. This behaviour is consistent with a first order transition, with an order parameter scaling down to zero with the cluster size below a critical value, and scaling up to a finite value above. However, with only two sizes available (the 16 site cluster turns out to be rather pathological with essentially δ independent correlations), this information should be taken with care, and a definitive identification of the nature of the phase transition requires further investigation.

B. Competition with Néel order ($\gamma = 0$)

We now consider the case of $\gamma = 0$, which corresponds to the competition between the first neighbor coupling and the six sites plaquette interaction. The interest in the model obtained for $\gamma = 0$ comes from the fact that the SS-VBS states remain eigenstates for all values of δ with an energy equal to zero. It follows that the transition at which the valence bond state vanishes is necessarily a level crossing and is therefore first order.

It should be emphasized here that, in the previous section, both SS-VBS and the collinear order phases presented antiferromagnetic correlations between second neighbor spins. Therefore, a direct transition between them seems to intuitively exist as if the spin-liquid phase (SS-VBS) knows, through the spin-spin correlations present in it, to which ordered phase it must go. This is no more the case for the Néel order which is stabilized by the first neighbor coupling, and presents antiferromagnetic correlations between first neighbor spins, and ferromagnetic correlations between second neighbor spins. According to this simple consideration, a direct transition between the SS-VBS phase and the Néel phase seems therefore unlikely.

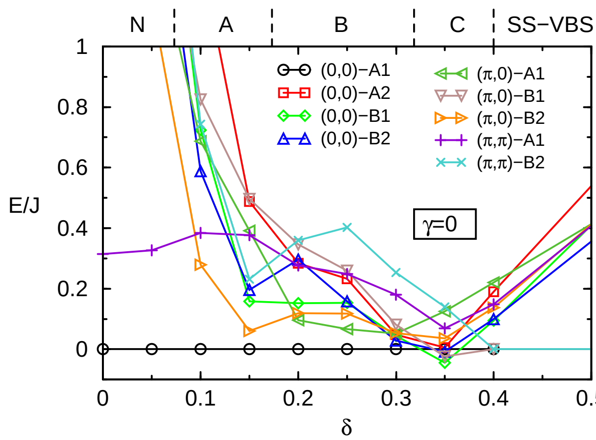

FIG. 8: (color online) Energy differences between the lower fully symmetric state with $\vec{k} = (0,0)$ momenta and some of the lowest states of the 32 site cluster. The symmetry of the different states is indicated on the figure.

We start the discussion by presenting in Fig. 8 the evolution of the energies of some of the lowest eigenstates obtained for the 32-site cluster, taking the lowest fully symmetric $\vec{k} = (0,0)$ level as the energy reference. Based on this figure and Fig. 10, we are lead to identify three different phases, tentatively labeled "A", "B" and "C" in addition to the well characterized (π, π) Néel phase at $\delta = 0$ and the fourfold degenerate Shastry-Sutherland states for $\delta \gtrsim 0.4$. We discuss each of these three phases in the following.

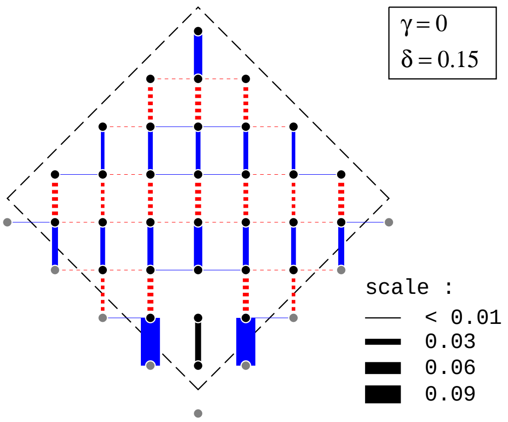

FIG. 9: (color online) Correlations between first neighbor spin dimers on the 32 sites cluster for $\gamma = 0$ and $\delta = 0.15$. Positive values are represented by (blue) plain lines, and negative values by (red) dashed lines. The thickness is proportional to the amplitude of the correlation as depicted on the figure. The (black) dimer in the lowest corner of the cluster is the reference dimer.

Phase "A" – Starting at small values of δ, we note that the plaquette interactions favor antiferromagnetic correlations between second neighbor spins. Therefore one could expect the evolution of the system to be similar to the J_1-J_2 model for small values of J_2. The symmetry of the four lowest states near $\delta = 0.15$ with momenta $(0, 0)$ (two states), $(\pi, 0)$ and $(0, \pi)$, are indeed compatible with the hypothesis of a four-fold degenerate ground state with a translational symmetry breaking. Fig. 9 shows the correlations defined in Eqn. (11), this time between dimers made of first neighbor spins obtained for $\delta = 0.15$. These correlations are relatively large, and show a columnar ordering of the dimers. Nevertheless, one should note that, as for the J_1-J_2 model, it is difficult to determine whether this phase presents columnar dimer order or plaquette order in the thermodynamic limit. So we believe the phase "A" is formed of some sort of valence bond solid with dimers on nearest-neighbor sites.

Phase "C" – At the other end of the δ axis, starting from large values of δ, one can see that the ground state stays exactly fourfold degenerate down to $\delta \simeq 0.4$, below which a level crossing occurs. Near $\delta = 0.35$ many levels are very close in energy. Some of these states are even found to be lower in energy than the fully symmetric $\vec{k} = (0, 0)$ state – which is again the ground state for $\delta \leq 0.3$, but this may well be a finite size effect on this particular sample.

In the case $\gamma = 1$ studied in section III A, we encountered a direct transition between a collinear $(\pi, 0)$ Néel ordered phase and the Shastry-Sutherland type VBS state. In order to test this scenario here, we determined the static spin structure factors for different momenta in Fig. 10. Indeed the $(\pi, 0)$ components are strongest around $\delta \sim 0.35$. In order to shed further light on the presence of magnetic long range order we study the evolution of the collinear magnetic order as a function of γ for a fixed value of $\delta = 0.35$. Using samples of 20 and 32 sites we obtain in Fig. 11 a finite size scaling which shows that the magnetic order is lost at a finite value of $\gamma \sim 0.1$, i.e. the point $\delta = 0.35$, $\gamma = 0$ does not sustain magnetic long range order.

Looking at real-space spin correlations, the second neighbor correlation is much larger than all other spin correlations, so that the possibility of a dimer VBS state needs to be checked. We therefore also computed the real-space dimer-dimer correlations which are presented in Fig. 12-(a). They present an ordering reminiscent of the SS-VBS phase, although correlations are smaller than in the pure phase. For the purpose of comparison the same correlations in the pure SS-VBS phase are displayed in Fig. 12-(b). We recall at this stage that in this range of parameters, several states are very close in energy (see Fig. 8), all singlets. We also observe that they all present the same kind of dimer-dimer correlations. From our results it is difficult to determine if these dimer-dimer correlations are short ranged or long ranged. However it seems as though this phase could be best visualized by a condensation of singlet excitations above the SS ground states, and not by a simple level crossing into a new ground state of completely different character, as it seems to happen in the original Shastry-Sutherland model[26].

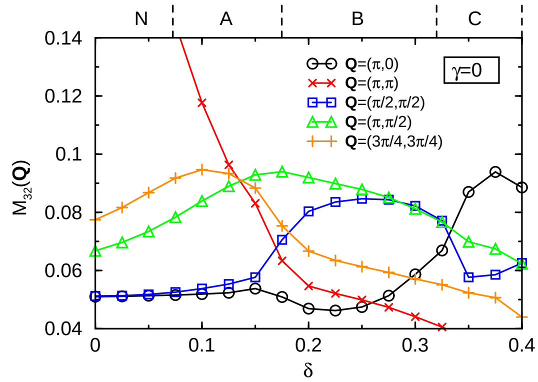

FIG. 10: (color online) Q dependent magnetic susceptibility obtained for the 32 sites cluster.

Phase "B" – Upon close inspection of Fig. 10 it is rather natural to suppose that there exists a third phase around $0.2 \lesssim \delta \lesssim 0.3$ sandwiched between the phases "A" and "C" which displays enhanced magnetic correlations for $Q = (\pi, \pi/2)$ and $Q = (\pi/2, \pi/2)$. Unfortunately the presence of correlations at these wavevectors renders the study of the system more difficult, since these momenta are not present on the 20 site cluster. It is therefore not possible to perform a finite size scaling of the $\vec{Q}$ dependent magnetic susceptibility. Nevertheless, one should note that there is also an enhancement of these components on the 16 site cluster. At that stage, it is difficult to characterize this phase "B" and to know if these large components correspond to a long range spin order, or to some more exotic phase.

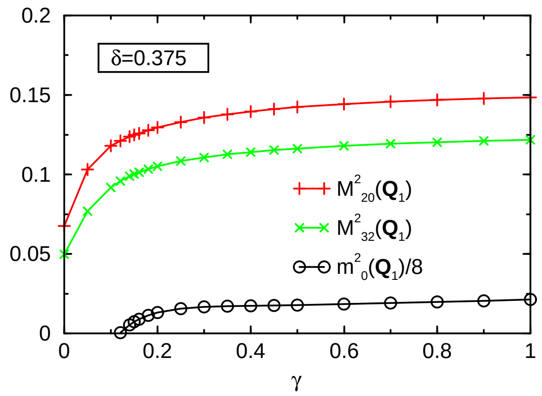

FIG. 11: (color online) $M_N^2(\vec{Q}_1)$ square magnetic susceptibility for 20 and 32 sites clusters and its extrapolation to thermodynamic limit $M_\infty(\vec{Q}_1)^2 = m_0^2(\vec{Q}_1)/8$, calculated for the lowest state with momenta $\vec{k} = (0,0)$ and highest symmetry.

IV. CONCLUSION

We introduced a new model with frustrated interactions which provides an interesting case of competition between antiferromagnetic orders and a valence bond solid order. We have shown that, for some values of the interaction parameters, the four fold degenerate VBS ground state is exactly a direct product of the dimer singlet wave functions. This model is thus an interesting candidate for investigating the possibility of a newly proposed scenario of quantum phase transition. It indeed presents, for a large range of parameters (namely $\gamma > 0.2$), a direct transition between the SS-VBS phase and a collinear antiferromagnetic phase. Further investigations are needed to determine if the transition is first order, of if it could correspond to the non-Landau-Ginsburg transition proposed by Senthil and coworkers[1,2].

For smaller values of the second neighbor interaction (i.e. small values of γ), the Néel and SS-VBS phases are separated by an intermediate region where different types of correlations dominate depending on the value of δ. It is likely that these correlations are the trace of exotic intermediate phases, but further work is clearly needed to fully characterize these phases and the nature of the transitions between them.

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APPENDIX

The Hamiltonian H_0 (Eq. 3 of section II) has four zero energy SS-VBS singlet configurations forming an exact ground state. Proving (in a mathematically rigorous way) that these four SS-VBS states are the only states in the ground state and there exists no fifth state, is a non-trivial and hard task. We are not going to attempt it here. Historically, for the Majumdar-Ghosh model, which is considerably simpler as compared to H_0, it was already very hard to prove the exact two-fold degeneracy of the ground state (which was eventually shown by AKLT). However, it was much easier to show that there are two dimerized singlet configurations which form the exact ground state of the Majumdar-Ghosh model, and to argue that other dimer-singlet configurations, generated by the allowed variations, will not be the eigenstates. We will do a similar exercise for H_0, showing that the four SS-VBS state are the only allowed dimer configurations in the ground state.

The block Hamiltonian, $h_6 = P^A\ P^B$, of a six-site plaquette is the basic building block of H_0. Since P^A and P^B are the spin projectors, the lowest energy of h_6 is zero. This corresponds to either P^A or P^B or both becoming zero in a given spin configuration of the block. This happens when the three A sublattice spins in a six-site plaquette form a total spin=1/2 state, or the same thing happens for B sublattice spins or for both. One way, in which this can be achieved, is by forming exactly one singlet bond out of three A or B sublattice spins of a plaquette. Thus a simple rule emerges for constructing the dimerized ground state of H_0. If a dimer configuration on the full square lattice is such that, on every six-site plaquette, there exists at least one singlet bond (dimer) between only A or only B sublattice spins, then all the plaquette Hamiltonians can be simultaneously satisfied (that is, every h_6 is in its ground state), and such a configuration will be an exact zero energy ground state of H_0.

Now, the number of rectangular plaquettes is equal to two times the number of sites, hence four times the number of dimers of any dimer covering of the lattice. Since a dimer belongs at most to four rectangles, to satisfy all rectangles simultaneously, each dimer should belong exactly to four rectangles, and each rectangle should contain a single dimer. Since a dimer constructed from third neighbours belongs to only two rectangles, such dimers have to be rejected, and one should only use diagonal dimers.

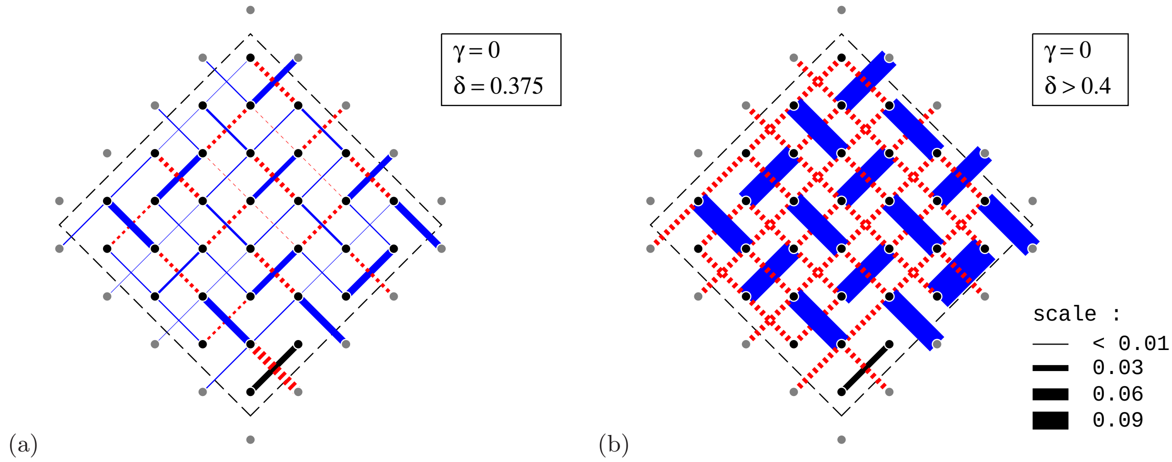

FIG. 12: (color online) Representation of correlations between second neighbor spin dimer on the 32 sites cluster calculated for the lowest state with momenta $\vec{k} = (0,0)$ and highest symmetry, (a) for $\gamma = 0$ and $\delta = 0.375$, (b) for $\gamma = 0$ and $\delta > 0.4$. Conventions are the same than on Fig. 9

Let us now consider one diagonal dimer. The remaining sites of the square plaquette on which this dimer sits have to be part of a dimer. But since two dimers cannot be on the same rectangular plaquette, the only possibility is that these dimers are perpendicular to the first one. This is precisely the prescription to construct a Shastry-Sutherland state. The freedom to chose the position and orientation of the first dimer leads to four different states. The exact diagonalization calculations on the 20 site and 32 site clusters presented in this paper support this assertion.

Note that with periodic boundary conditions of length 4, a dimer constructed from third neighbours satisfies four rectangles, which leads to two additional ground states on the 16-site cluster.

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