

# Quantum phase diagram of ferromagnetically coupled singlet dimers on a distorted Shastry-Sutherland lattice

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Frustrated spin-1/2 model with all-antiferromagnetic exchange on Shastry-Sutherland lattice (SSL) has been extensively explored, and in this work we study the quantum phase diagram of isotropic Heisenberg spin-1/2 model on the SSL with ferromagnetic exchange interactions along leg, and antiferromagnetic couplings along the diagonal bonds ( $J$ ). Using density matrix renormalization group (DMRG) and exact diagonalization calculations, we identify four distinct ground-state (gs) phases for this frustrated spin-1/2 model: stripe ( $\pi, 0$ ), stripe ( $0, \pi$ ), ferromagnetic, and spiral-dimer phases. In the spiral-dimer phase the gs can exhibit dimer formation along the diagonal bonds with or without spins spiral along the  $x$  axis or  $y$  axis in different parameter regime and these states are connected through crossovers. There is a continuous transition between stripe with a long-range spin-spin correlations and spiral-dimer phase with a short-range correlations. An effective spin parameter is introduced to accurately determine the phase boundaries. Our DMRG results differ significantly from previous Schwinger boson mean-field theory predictions. The spiral phase occurs in a narrow range of the parameter space and shows short-range correlations in the system.

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## I. INTRODUCTION

Frustration in quantum magnetic system leads to emergence of fascinating quantum phases such as dimer [1], noncollinear spin wave [2], spin-liquid [3,4], nontrivial topological phase [5,6] in the ground state (gs). Many of these quantum phases have been realized in real materials and have potential application in spintronics and quantum computation devices. Over the past few decades, these systems have been synthesized in different dimensions, for example, in one-dimensional geometry:  $\text{LiCuSbO}_4$  [7],  $\text{LiCuVO}_4$  [8], and  $\text{Rb}_2\text{Cu}_2\text{Mo}_3\text{O}_{12}$  [9,10]; in ladderlike geometry:  $\text{SrCu}_2\text{O}_3$  [11] and  $(\text{VO})_2\text{P}_2\text{O}_7$  [12,13]; in two-dimensional systems such as  $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$  (Herbertsmithite) [14–16]. In order to understand their physical properties different models have been proposed such as Heisenberg spin-1/2  $J_1$ - $J_2$  model for one-dimensional (1D) spin chain [1,17–19], Heisenberg antiferromagnetic spin-1/2 model on Shastry-Sutherland lattice (SSL) [20], on square [21–23], triangular [24], Kagóme lattice [15], etc.

The two-dimensional magnetic systems are most often realized in layered materials [25] and competing exchanges can lead to magnetic frustration in two dimensions, which is critical

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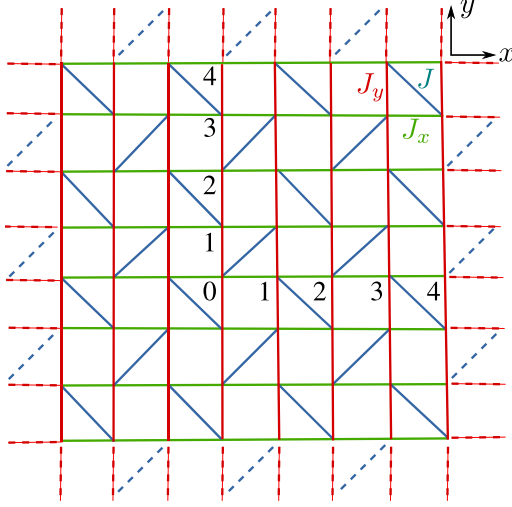


FIG. 1. Figure shows a schematic diagram of SSL. The coupling constants along  $x$  axis ( $J_x$ ) and along  $y$  axis ( $J_y$ ) are ferromagnetic (FM) and the diagonal bond couplings  $J$  are antiferromagnetic (AFM) in nature. The site index 0 is the reference site and the numbers along  $x$  axis and  $y$  axis represent the distance ( $r$ ) from the reference site.

dimension as suggested by Mermin-Wagner theorem [26]. These systems can form the square [27], the triangular [24], the kagomé [15] and the SSL [28–30], etc. Here, we are interested in the SSL, which is shown in Fig. 1 and it is a unique 2D lattice composed of two types of squares. The lattice is formed by arranging these squares in a periodic pattern, with dimers (pairs of spins) positioned along the diagonals of the squares. This distinctive arrangement gives rise to intriguing magnetic behaviors, making it a significant area of research [31–34].

An isotropic antiferromagnetic Heisenberg spin-1/2 model on the SSL with leg exchange  $J_x$ , rung exchange  $J_y$ , and diagonal exchange  $J$  can host a quantum spin-liquid gs in certain parameter regimes, as shown by Shastry and Sutherland [20]. Mihara *et al.* [35] determined the critical value of the quantum phase transition from the dimer state to the Néel ordered state. They also showed that the  $\text{SrCu}_2(\text{BO}_3)_2$  exhibits the dimer gs, however, exchange parameters are close to the transition point, which leads to an unusual temperature dependence of the susceptibility.

In small and large  $J/J_y$  limit, gs of the system is showing Néel and dimer phase, respectively, with  $J_x = J_y$ . Koga and Kawakami [36] predicted a new spin-gap phase, which is adiabatically connected to the plaquette-singlet phase existing between the known dimer phase and the magnetically ordered phase. Later, by using projected entangled-pair states (PEPS) technique, the existence of a plaquette phase between Néel and dimer region has been confirmed by Corboz *et al.* [37]. The first experimental identification of the conjectured plaquette singlet intermediate phase in  $\text{SrCu}_2(\text{BO}_3)_2$  has been observed in Ref. [29]. In 2021 Yang *et al.* [38] predicted a spin liquid phase. On the other hand, the topological properties of this model has been explored by Ronquillo and Peterson [39] through the calculation of entanglement entropy.

Spin-1/2 layered copper oxyhalides  $(\text{CuX})\text{A}_{n-1}\text{B}_n\text{O}_{3n+1}$  are frustrated 2D magnets among the layered magnetic materials. These materials exhibit many interesting features, which can be induced by only tuning the composition [40,41]. The  $\text{CuX}$  layers are surrounded by nonmagnetic layers. The anion orbitals are involved in the exchange pathways, but the exchange of the cation orbitals maintains the homogeneity of the magnetic layer. However, exchange interaction can be tuned by changing the lattice parameters, electrostatic fields, and crystal-field splittings [42]. Tuning the composition of anions and cations can drive the system beyond quantum critical points. Experimental results indicate that the gs can be tuned to have a collective singlet with a spin gap

in (CuCl)LaNb<sub>2</sub>O<sub>7</sub> [40,43–45], a collinear stripe magnetic order in (CuBr)LaNb<sub>2</sub>O<sub>7</sub> [41], and a magnetization-1/3 plateau in (CuBr)Sr<sub>2</sub>Nb<sub>3</sub>O<sub>10</sub> [46].

(CuCl)LaNb<sub>2</sub>O<sub>7</sub> was initially predicted to be a  $S = 1/2$  frustrated square lattice [40], however, from the band structure calculations, it was revealed that the simplest model for this material can be best described as strong antiferromagnetic (AFM) exchange interaction between fourth neighbors forming a strong singlet dimer and these dimers are coupled with each other by ferromagnetic (FM) interactions, i.e., this model looks like the Shastry-Sutherland model (SSM) with FM exchange  $J_x$  and  $J_y$  along square and AFM diagonal exchange  $J$  [47]. This model was theoretically studied by using mean-field and exact diagonalization (ED) methods [48], and the study showed the existence of different exotic phases in the exchange parameter space. In the intermediate value of FM interaction limit, they reported two types of stripe phases,  $(0, \pi)$  and  $(\pi, 0)$ , which are separated by a noncollinear spiral phase.

For  $J_x, J_y < J/2$ , the dimer singlet stabilizes, while FM gs is stable for large  $J_x$  and  $J_y$  [48]. We notice that the phase boundaries as well as the different exotic phases calculated by using ED are not consistent with that of obtained from mean-field calculation, for example, noncollinear phase does not exist in ED results, but this phase appears in the mean-field calculations. Therefore, it is very exciting to explore such an interesting model with a more sophisticated numerical tool such as density matrix renormalization group (DMRG) method, which can give more accurate results through large lattice sizes.

In this work, we investigate the ferromagnetically coupled  $S = 1/2$  dimers on the SSL using the DMRG method and construct the quantum phase diagram of this model using spin correlation, gs energy variation, and effective spin calculations. In the  $J_x$ – $J_y$  parameter space, the gs exhibits four distinct phases. For large  $J_x$  or  $J_y$ , the system stabilizes stripe-ordered phases characterized by ordering wave vectors  $(0, \pi)$  and  $(\pi, 0)$ , respectively. When both  $J_x$  and  $J_y$  are large, the gs spins align ferromagnetically. In the regime of moderate  $J_x$  and  $J_y$ , diagonal singlet dimers interact weakly via FM exchange, introducing frustration and giving rise to a spiral-dimer phase. This phase hosts three competing states: a singlet dimer state and spin-spiral states propagating along either the  $x$  or  $y$  direction. These spin-spiral and dimer states may coexist or be connected through crossover regions. A perfectly formed diagonal singlet dimer state is realized along the line  $J_x = J_y$ . The most challenging part is to construct the phase boundaries between spiral-dimer phase and stripe phase as in this case, the system goes from short-range spin-spin correlations to long-range spin correlations.

The results are organized as follows. In Sec. II, the model of the ferromagnetically coupled SSL and numerical methods are discussed. In Sec. III, all the numerical results have been shown. In this section, different phases and the quantum phase diagram have been discussed. In Sec. IV, we will discuss how different phase boundaries are obtained.

## II. MODEL HAMILTONIAN AND NUMERICAL METHODS

We consider a Heisenberg spin  $S = 1/2$  model on SSL where diagonal exchange interaction  $J = 1$  is AFM type and defines as the energy scale of the system. The strength of exchange interaction along the  $x$  axis and  $y$  axis on the square is represented by  $J_x$  and  $J_y$ , respectively, and both of these are of FM type. The arrangement of the exchange interactions have been shown in Fig. 1. Here, we call this model as SSM and it can be written as

$$H = -J_x \sum_{\langle ij \rangle_x} \mathbf{S}_i \cdot \mathbf{S}_j - J_y \sum_{\langle ij \rangle_y} \mathbf{S}_i \cdot \mathbf{S}_j + J \sum_{\langle ij \rangle_d} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (1)$$

where the first sum is for nearest-neighbor (NN) bonds along the  $x$  axis, the second sum is for NN bonds along the  $y$  axis, and the last sum runs for NN diagonal bonds in the square.

We have used the ED method to solve the model Hamiltonian for a system size up to 32 sites and for the larger system sizes, the DMRG method [49–52] is used. The DMRG method is a state-of-the-art numerical technique to solve the quantum many-body model Hamiltonian in low dimensions, and it is based on the systematic truncation of irrelevant degrees of freedom while growing the

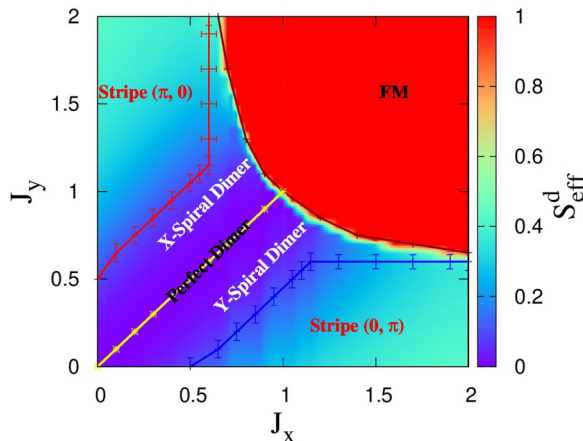


FIG. 2. Figure shows the quantum phase diagram of the SSM. The phase diagram comprises four phases: two stripe order phases with ordering wave vector  $(0, \pi)$  and  $(\pi, 0)$ , spiral-dimer phase, and ferromagnetic phase. The color in the phase diagram represents magnitude of effective spin along diagonal dimer bond  $S_{\text{eff}}^d$  for  $6 \times 4$  lattice system.

system sizes [49,50]. This method is a state-of-the-art numerical method to solve the many-body Hamiltonian and to get accurate low-lying eigenvalues and wave functions.

In this work, a modified DMRG method has been used [53] in which four new sites are added at every step in order to reduce uses of the multiple times renormalization of the operators in constructing the superblock [53]. We retain up to  $m = 900$  block states, which are the eigenvectors of the system block density matrix corresponding to the largest eigenvalues and this chosen value of  $m$  is able to keep the truncation error to less than  $\sim 10^{-8}$ . We also used 10–12 finite sweeps in order to obtain more improved convergence and to optimize the wave function. We use cylindrical geometry of the SSL with a periodic boundary along the width of the lattice and open boundary along the length. The largest system size ( $N$ ) used in our study is up to  $12 \times 8$  (length  $\times$  width) in case of eight-leg geometry and  $32 \times 4$  for four-leg geometry. We have also analyzed the convergence of energy with the various values of  $m$  in the Supplemental Material [54]. The per-site energies for  $12 \times 4$  system are shown for  $m = 256, 512$ , and  $900$ , whereas for  $12 \times 8$  system, it is shown for  $m = 256, 512$ , and  $700$  in Table II of the Supplemental Material [54]. We have noticed that  $m = 512$  is sufficient for accuracy up to fifth decimal place for  $12 \times 4$  and fourth for  $12 \times 8$  system. The convergence of spin-spin correlation function as function of  $m$  is shown in the Table III and IV of the Supplemental Material [54] and these are accurate up to four decimal places for  $N = 12 \times 4$  and  $12 \times 8$ .

### III. RESULTS

Competing FM interaction along  $x$  and  $y$  direction and AFM exchange interaction along the diagonal direction shown in Fig. 1, induces frustration in the system and gives rise to many exotic quantum phases. To understand these phases we calculate gs energy, spin-spin correlations, and effective spin by using the ED and the DMRG methods. In this section, different quantum phases and the phase diagram of the model Hamiltonian in Eq. (1) are discussed.

#### Quantum phases and phase diagram

The quantum phase diagram in  $J_x$ - $J_y$  parameter space is shown in Fig. 2 and the ferromagnetic  $J_x$  and  $J_y$  parameters are restricted between 0 and 2. We identify four different quantum phases

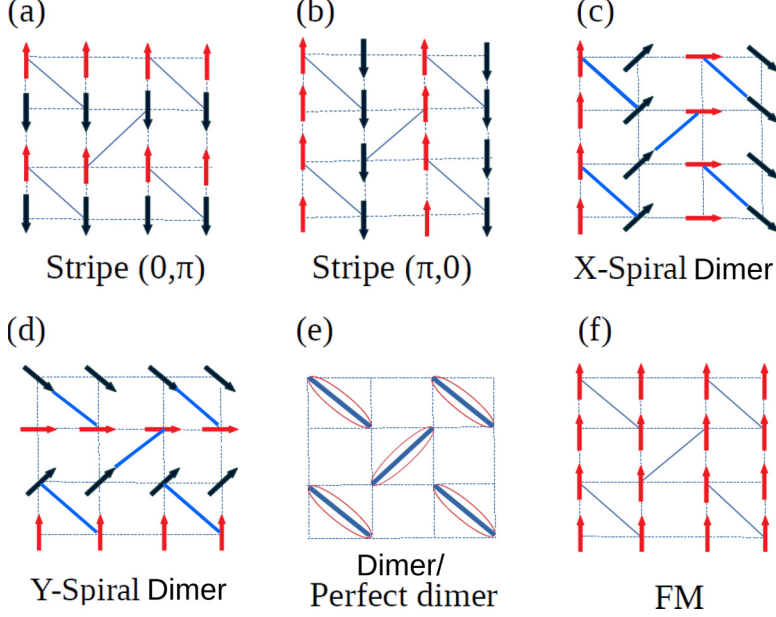


FIG. 3. Figure shows a schematic diagram of spin arrangement in different phases or states in the ferromagnetically coupled dimers on the SSL, where arrows represent the spin alignments and the ellipses are the dimers (defined in the main text). (a) and (b) are two stripe phases with ordering wave vector  $(0, \pi)$  and  $(\pi, 0)$ , respectively. (c) and (d) are two noncollinear spiral-dimer phases with strong diagonal bond, the spiral dimer along  $x$  and  $y$  directions. (e) and (f) are the dimer/perfect dimer and FM phase, respectively.

based on the spin arrangements which are obtained from the spin-spin correlations,  $C(r)$ . All these quantum phases are described below.

*Figures 3(a), 3(b), stripe  $(0, \pi)$  and stripe  $(\pi, 0)$ .* The stripe  $(0, \pi)$  and the stripe  $(\pi, 0)$  phases are the compensated magnetic states, i.e., total magnetic moment ( $S^z = \sum_i s_i^z$ ) of the gs is zero. In the stripe  $(0, \pi)$  phase, the gs spins are aligned ferromagnetically along the  $x$  axis and antiferromagnetically along the  $y$  axis, while in the stripe  $(\pi, 0)$ , they align ferromagnetically along the  $y$  axis and antiferromagnetically along the  $x$  axis, as shown schematically in Figs. 3(a) and 3(b) respectively.

Two types of AFM correlations are observed within these phases. Short-range AFM ordering develops along the  $x$  or  $y$  direction for moderate  $J_x$  and large  $J_y$ , or for moderate  $J_y$  and large  $J_x$ , respectively. This short-range AFM correlation emerges in the intermediate  $J_x$  or  $J_y$  regime due to frustration in the system, analogous to FM chains coupled through competing FM and AFM exchanges. These results suggest that in this parameter regime, the gs is not a true stripe phase characterized by long-range order, rather a state with spin arrangements resembling stripe order and only short-range AFM correlations.

In contrast, the true stripe  $(0, \pi)$  phase is characterized by long-range AFM ordering along the antiferromagnetic direction for  $J_x > 2.39$  at small  $J_y$ . For a true stripe phase, long-range order sets in along both the AFM and FM directions, which requires weak or absent frustration. Consistent with this expectation, we find that long-range AFM order in the stripe  $(0, \pi)$  phase exists for  $J_y = 0$  when  $J_x > 2.39$ . Similarly, for the stripe  $(\pi, 0)$  phase, long-range AFM order is observed for  $J_x = 0$  and  $J_y > 2.39$ . For small finite values of  $J_x$  or  $J_y$  long-range AFM order is observed for larger values of  $J_y > 2.39$  or  $J_x > 2.39$ .

*Figures 3(c)–3(e), X-spiral-dimer, Y-spiral-dimer, and dimer states.* In this model, the competition among the exchange interactions  $J_x$ ,  $J_y$ , and  $J$  introduces frustration, which stabilizes three

TABLE I. Properties of various quantities like spin-spin correlations along the diagonal bond  $C^d(r=1)$ , nearest-neighbor spin correlations along the  $x$  direction  $C^x(r=1)$ , nearest-neighbor spin correlations along the  $y$  direction  $C^y(r=1)$ , effective spin of diagonal dimer ( $S_{\text{eff}}^d$ ) and effective spin along ferromagnetically ordered leg ( $S_{\text{eff}}^L$ ) in different exotic phases or states.

| Phases or states  | $C^d(r=1)$ | $C^x(r=1)$ | $C^y(r=1)$ | $S_{\text{eff}}^d$ per site | $S_{\text{eff}}^L$ per site |
|-------------------|------------|------------|------------|-----------------------------|-----------------------------|
| Perfect dimer     | 0.75       | 0          | 0          | 0                           | 0                           |
| Dimer             | large      | small      | small      | $\approx 0$                 | $\approx 0$                 |
| Stripe $(0, \pi)$ | finite     | finite     | finite     | $> 0$                       | $> 0$ (along $x$ )          |
| Stripe $(\pi, 0)$ | finite     | finite     | finite     | $> 0$                       | $> 0$ (along $y$ )          |
| X-spiral dimer    | finite     | finite     | finite     | $> 0$                       | $> 0$ (along $y$ )          |
| Y-spiral dimer    | finite     | finite     | finite     | $> 0$                       | $> 0$ (along $x$ )          |
| FM                | 0.25       | 0.25       | 0.25       | 0.5                         | 0.5                         |

distinct states depending on the parameter regime: a perfect dimer state, X-spiral-dimer state, and Y-spiral dimer state. In the X- and Y-spiral-dimer states, the spins form spiral patterns along the  $x$  and  $y$  directions, respectively, while coexisting with diagonal spin dimers, as illustrated in Figs. 3(c) and 3(d). Spins perpendicular to the spiral direction are ferromagnetically aligned.

The coexistence of dimer and spiral order is analogous to that found in the one-dimensional  $J_1$ - $J_2$  model [55,56]. These states are smoothly connected through crossover regions. For  $|J_x - J_y| \ll J$ , the diagonal exchange  $J$  dominates, leading to the formation of strong diagonal singlet dimers. In this phase, spin-spin correlations along the  $x$  and  $y$  directions remain short ranged and decay exponentially. A perfect singlet dimer state is realized only along the line  $J_x = J_y$ , a schematic of which is shown in Fig. 3(e). The spiral-dimer and perfect dimer phases are also connected via a crossover.

To visualize the strength of the singlet dimers in the parameter space, we calculated the effective spin  $S_{\text{eff}}^d$ , which is the sum of two spins  $S_1$  and  $S_2$  sitting at a diagonal bond and  $S_{\text{eff}}^d$  can be written in terms of  $S_1$  and  $S_2$  as

$$(S^d)_{\text{eff}}^2 = (S_1^2 + S_2^2 + 2\mathbf{S}_1 \cdot \mathbf{S}_2), \quad (2)$$

where,  $S = \sqrt{s(s+1)}$ . Color gradient in the phase diagram shows the variation of the  $S_{\text{eff}}^d$ , which changes with interaction strengths. It is exactly zero for the perfect dimer region at the  $J_x = J_y$  and increases as we go deeper into the stripe phases. It has highest values in the FM phase  $S_{\text{eff}}^d = 1$ , i.e., the spin dimer forms a triplet state.

Figure 3(f), FM. For a large value of  $J_x$  and  $J_y$  the ferromagnetic exchanges are dominant compared to the AFM diagonal exchange and it leads to FM spin arrangement as shown schematically in Fig. 3(f). In the FM phase region correlation function  $C(r)$  is positive for all the values of  $r$ .

The properties of quantum phases, spin arrangements, and nature of order are summarized in Table I.

#### IV. QUANTUM PHASE BOUNDARIES

We calculate per-site gs energy and spin-spin correlations in order to find out the phase boundaries. In some parameter regimes, the system undergoes a transition from the short-range correlations in the spiral-dimer phase to the long-range FM correlations in the stripe phase along a particular direction and calculating the phase boundary becomes challenging, therefore, we can calculate the effective total spin  $S_{\text{eff}}$  of each leg along the ordering direction.

We first analyze per-site energy in the gs,  $\varepsilon_{\text{gs}}$  for the model in Eq. (1) on cylindrical geometry with periodic boundary condition (PBC) for  $4 \times 6$  and  $6 \times 4$  systems and in Fig. 4,  $\varepsilon_{\text{gs}}$  is shown as a function of  $J_y$  with fixed  $J_x = 0.9$  for two systems. The discontinuity in  $\varepsilon_{\text{gs}}$  at  $J_x = 1.1$  indicates

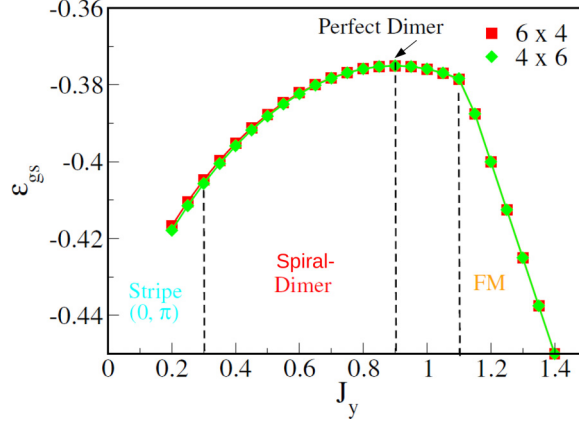


FIG. 4. Figure shows gs energy per site ( $\varepsilon_{gs}$ ) versus  $J_y$  plot at  $J_x = 0.9$  for  $6 \times 4$  and  $4 \times 6$  lattice sites of the SSL. Around  $J_y = 1.1$ , we show a kink in the energy curve indicating a sharp phase transition from spiral-dimer to FM phase. Around  $J_y = 0.3$ , the system transits from stripe  $(0, \pi)$  to spiral-dimer phase. (Shaded lines are only for eye guide of the transition points).

a first-order phase transition from FM to spiral-dimer phase. The maxima of  $\varepsilon_{gs}$  at  $J_x = J_y = 0.9$  indicates the perfect dimer line, and both of the systems give the similar phase boundary, i.e., it shows extremely weak finite-size effect. However, other phase boundaries can not be determined using  $\varepsilon_{gs}$ .

The isotropic nature of spin exchange in the Hamiltonian in Eq. (1) preserves the  $SU(2)$  symmetry and isotropic spin-spin correlation is expected in a singlet state. We can write the spin-spin correlation,  $C(r)$  as,

$$C(r) = \langle \mathbf{S}_i \cdot \mathbf{S}_{i+r} \rangle = 3 \langle S_i^z S_{i+r}^z \rangle, \quad (3)$$

where  $S_i^z$  is the  $z$  component of the spin operator at the reference site  $i$ . Distance between spin sites along the  $x$  and  $y$  axis are shown in Fig. 1.

We first analyze  $C(r)$  of the stripe  $(0, \pi)$  phase, which shows two types of AFM correlation: short- and long-range along  $y$  axis and long-range FM ordering along  $x$  axis in the gs. Let us consider for limiting case  $J_y = 0$ , the geometry of this system is a brick wall, see in Fig S1(c) of the Supplemental Material [54], and for  $J_x \gg J$ , this system shows a long-range order in both directions; FM ordering along  $x$  axis and AFM along  $y$  axis.

The transition from short-range to long-range order, the Néel phase can be characterized by using most distant spin-spin correlation  $C(r = L_y/2) = \langle \mathbf{S}_i \cdot \mathbf{S}_{i+L_y/2} \rangle$  [57] and we calculate the correlations on a cylindrical geometry for system size  $4 \times L_y$  and  $8 \times L_y$ . The correlations in the gs for both system sizes show weak finite-size effect. In Fig. 5(a), we extrapolate the most distant spin-spin correlations  $C(r = L_y/2)$  along  $y$  axis as function of inverse system size  $1/L_y$  for different values of  $J_x$  at  $J_y = 0$ , and notice that the extrapolated correlations  $C(r = L_y/2)$  is finite only for  $J_x > 2.39$ . The finite value of  $C(r = L_y = \infty)$  indicates long-range Néel order in the gs. The extrapolated value of  $C(r = L_y/2 = \infty)$  is shown in Fig. 5(b) and it is nonzero for  $J_x > 2.39$ , which indicates the transition point  $J_x^c = J_x = 2.39$  for  $J_y = 0$ . For finite value of  $J_y$ , the transition point  $J_x > J_x^c$  as the gs tends to get frustrated even for higher  $J_x$ . Therefore, this phase shows the long-range order for small finite  $J_y$  at higher value of  $J_x^c$ . Similar type of AFM long-range order appears along the  $x$  axis in stripe  $(\pi, 0)$  phase  $J_y \gg J \gg J_x$ . For  $J_{x/y} < J_{x/y}^c$  the gs only show the striplike spin arrangement not the true stripe phase. However, since the crossover boundary  $J_{x/y}^c$  separating short-range and long-range AFM correlations in the stripe phase is extracted from finite-size calculations, its precise value is subject to finite-size effects and would require simulations on larger system widths for a more accurate determination.

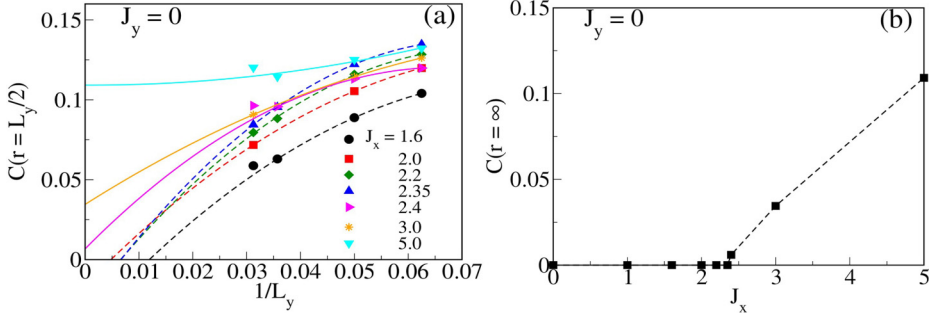


FIG. 5. The most distant spin-spin correlations  $C(r = L_y/2)$  along the  $y$  axis in the stripe  $(0, \pi)$  phase for  $J_y = 0$  and various values of  $J_x$  are extrapolated with  $L_y$  for a system size of  $4 \times L_y$  in (a). The extrapolated values of the correlations  $C(r = L_y/2 = \infty)$  are shown in (b). The transition from short-range spin correlations to long-range correlations along the  $y$  direction occurs at  $J_x \approx 2.39$ .

In Figs. 6(a) and 6(b) correlations along  $x$  and  $y$  axis are shown for three different phases; stripe  $(0, \pi)$  for  $(J_x = 2, J_y = 0.2)$ , perfect dimer  $(J_x = J_y = 0.2)$ , and stripe  $(\pi, 0)$  ( $J_x = 0.2, J_y = 2$ ), respectively. In Fig. 6(a) for stripe  $(0, \pi)$  ( $J_x = 2, J_y = 0.2$ ),  $C(r)$  along the  $x$  axis (represented with circle) shows a FM arrangement of the spins, whereas along the  $y$  axis, it is antiferromagnetically aligned and decays exponentially. Similarly, for stripe  $(\pi, 0)$  ( $J_x = 0.2, J_y = 2$ ), nature of correlation reverses and along  $y$  axis,  $C(r)$  (represented with squares) is long-range FM order and AFM short-range order along  $x$  axis. In the perfect dimer phase,  $C(r)$  is represented with diamond and is exactly zero in both the direction.

In Fig. 7,  $C(r)$  are shown for  $J_x = 0.7$  for three values of  $J_y = 1.4, 1.5$ , and  $1.6$  in the  $X$ -spiral-dimer phase, we notice that the correlation function decays exponentially and the spin arrangement are noncollinear in nature along  $x$  direction.

However, for  $|J_x - J_y| \ll J$ , the gs exhibits weakly interacting dimer phase as diagonal nearest-neighbor correlation  $C^d(r = 1)$  is strong, and this phase can be divided into two distinct regimes based on the spin arrangements along the  $x$  and  $y$  axes. However, in both cases, the gs does not exhibit any long-range magnetic order. In this limit, the correlations in both directions are short range. Spins are ferromagnetically aligned in  $x/y$  direction whereas antiferromagnetically aligned along  $y/x$  direction. Arrangement of the spins in the gs is similar to that in stripe phase except there is no ordering in any direction and have dominant diagonal dimer. For strong  $J_x > J/2$  or  $J_y > J/2$

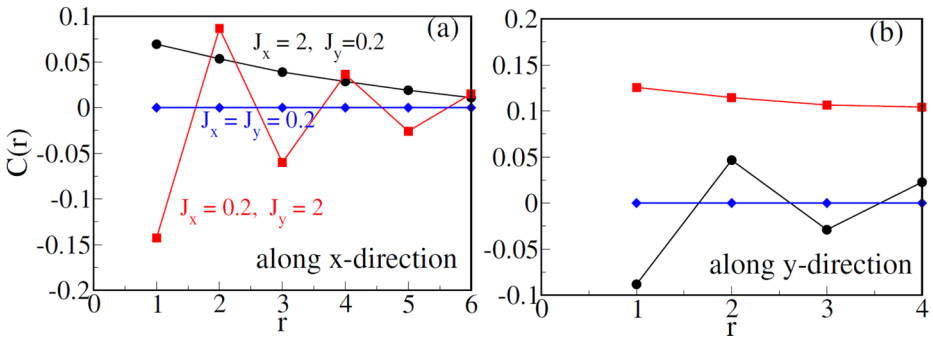


FIG. 6. Figure shows the variation of spin-spin correlations  $C(r)$  with distance ( $r$ ) for  $12 \times 8$  system size. Spin-spin correlations are calculated for different interaction limit. (a)  $C(r)$  are calculated along  $x$  direction. (b)  $C(r)$  are calculated along  $y$  direction.

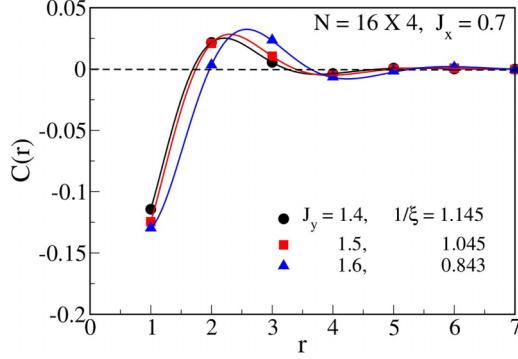


FIG. 7. Figure shows  $C(r)$  along length ( $x$  direction) for different  $J_y$  at  $J_x = 0.7$ . The solid curves represent respective fits with exponential decaying function in Eq. (4) for  $N = 16 \times 4$  and  $\xi$  is the correlation length.

and  $|J_x - J_y| > 0$ , gs is spiral-dimer phase as shown in Fig. 7. These two types of gs are connected through crossover.

To analyze this crossover, we calculated correlations for a system size  $N = 6 \times 4$  system. Owing to the short-range nature of correlations, finite-size effects are negligible. The diagonal correlation  $C^d(r = 1) \approx 0.75$  confirms strong dimerization, while  $C^x(r)$  changes from antiferromagnetic below  $J_x^c$  to spiral for  $J_x > J_x^c$ , as summarized in Table I in the Supplemental Material [54]. This boundary can be determined by the change in the spin arrangement along the AFM leg direction and determination of the boundary is difficult due to short-range nature of the correlation.

For larger  $J_y$  in X-spiral-dimer phase, the correlations  $C(r > 1)$  acquire finite values and exhibit short-range spiral behavior along the  $x$  axis, indicating the coexistence of spiral correlations and diagonal dimers in this parameter regime as shown in Fig. 7 for  $J_y = 1.4, 1.5$ , and  $1.6$ . The spiral nature of gs spins can be determined by fitting the  $C(r)$  with the following equation

$$C(r) = a_0 \cos(Q_x r + c) e^{-\frac{r}{\xi}}, \quad (4)$$

where  $\xi$  is the correlation length,  $a_0$  and  $c$  are constants.

The spin alignment is spiral, but, an accurate calculation of pitch angle is difficult due to short-range nature of the spin-spin correlations. We compute the pitch angle  $Q_x$  in stripe  $(\pi, 0)$  phase along the  $x$  axis as a function of  $J_y$  for  $J_x = 0.7, J = 1$  as shown in Fig. S4 of Supplemental Material [54]. We notice that  $Q_x/\pi$  shows very weak variation and it is approximately 0.55. At the perfect dimer line, each singlet dimer in the gs behaves as an effective spin-zero entity, while in the ferromagnetic state,  $Q_x$  vanishes. Transition from spiral-dimer to the FM phase is first order, therefore,  $Q_x$  changes sharply at the spiral dimer to FM phase boundary. The width of spiral-dimer phase shrinks as go for larger value of  $J_y$  for given  $J_x$  (and vice versa), and detecting spiral phases is quite difficult. The correlation length  $\xi$  decreases as we go towards the perfect dimer line. In Fig. 7,  $1/\xi$  are listed for three values of  $J_y = 1.4, 1.5$ , and  $1.6$  at  $J_x = 0.7$  and the corresponding values of  $\xi$  are 0.87, 0.96, and 1.19, respectively. The spiral nature emerges for large  $J_x$  and  $J_y$  starting from the proximity of the perfect dimer line.

In the phase diagram Fig. 2, we only focus on a part of stripe phase which have long-range FM order and short-range AFM, and on moving toward the perfect dimer line system goes to spiral-dimer phase. Determination of the phase boundary between spiral-dimer and the stripe phases is a difficult. The stripe  $(0, \pi)$  phase shows a long-range FM order along chains but AFM aligned spins with short correlation along the  $y$  axis. To characterize the FM order along the chain, we define the total spin along a FM chain as

$$S_L^2 = \left( \sum_i S_i \right)_x^2 = \sum_i S_i^2 + \sum_{ij} \mathbf{S}_i \cdot \mathbf{S}_j. \quad (5)$$

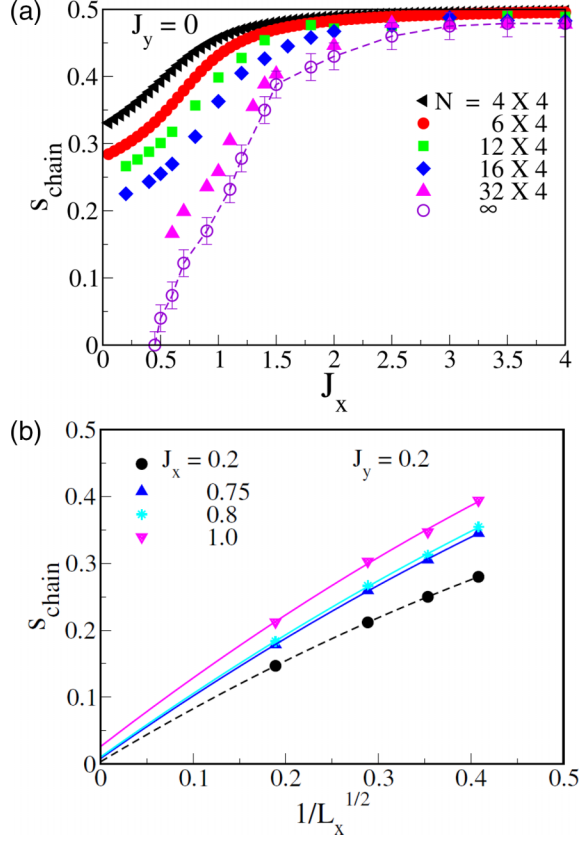


FIG. 8. (a) shows effective spin per site ( $s_{\text{chain}}$ ) along the leg versus  $J_x$  for  $N = 4 \times 4$ ,  $6 \times 4$ ,  $12 \times 4$ ,  $16 \times 4$  and  $32 \times 4$ . The violet color curve shows the extrapolated values of  $s_{\text{chain}}$  in thermodynamic limit and it goes from 0 to 0.5. (b) shows the extrapolated value of effective spin per site ( $s_{\text{chain}}$ ) along the leg for  $J_y = 0.2$ . Black dashed line is our reference line, which meets at zero.  $J_x = 0.75$  is considered as the transition point from  $s_{\text{chain}} = 0$  to some finite value.

Here, we drop the label of y axis, all the chains are equivalent and subscript  $x$  denotes the axis of the chain and  $S_i^2 = S_i(S_i + 1)$ .  $\mathbf{S}_i \cdot \mathbf{S}_j$  is spin-spin correlations. In thermodynamic limit,  $S_L$  per site can be denoted by  $s_{\text{chain}}$  and can take value from 0 to 0.5. In Fig. 8(a), we have shown the  $s_{\text{chain}}$  along the leg for  $N = 4 \times 4$ ,  $6 \times 4$ ,  $12 \times 4$ ,  $16 \times 4$ , and  $32 \times 4$ .

If  $L_x$  is the number of sites in each chain and  $s_{\text{chain}}$  is effective spin per site along leg, then in perfect dimer line, Eq. (5) reduces to,

$$L_x^2 s_{\text{chain}}^2 + L_x s_{\text{chain}} = 3L_x/4 = \alpha L_x, \quad (6)$$

where,  $\alpha = s(s + 1) = 3/4$  for spin-1/2 case. It should be mentioned that the second term in Eq. (5) vanishes as spin-spin correlations for perfect dimer line is zero along the leg. From Eq. (6) we get,

$$s_{\text{chain}} = 1/2L_x[-1 \pm \sqrt{(1 + 4\alpha L_x)}].$$

In large  $L_x$  limit,  $s_{\text{chain}}$  goes as  $1/\sqrt{L_x}$ . The extrapolation of the  $s_{\text{chain}}$  with  $1/\sqrt{L_x}$  are shown in Fig. 8(b) for various value of  $J_x$  and  $J_y = 0.2$ . In Fig. 8(a) we show the  $s_{\text{chain}}$  as function  $J_x$  for  $J_y = 0$  and the extrapolated value of the phase boundary is shown with circle. We have done these extrapolation for various values of  $J_y$  to determine the phase boundary.

## V. DISCUSSION AND CONCLUSION

In this work, we present a constructed quantum phase diagram for the spin-1/2 SSL with FM couplings using numerical methods: the exact diagonalization and the density matrix renormalization group method. The exchange interactions along the horizontal ( $J_x$ ) and vertical ( $J_y$ ) directions are FM, while the diagonal couplings ( $J$ ) are AFM. We identify four different quantum phases: stripe phase ( $\pi, 0$ ), stripe ( $0, \pi$ ), ferromagnetic, and spiral-dimer phase where dimer coexist with spin spiral either along  $x$  or  $y$  axis. Our analysis confirms the existence of spiral-dimer phases characterized by short-ranged spin correlation either along  $x$  or  $y$  axis and ferromagnetic arrangement of spins along perpendicular to direction of spiral. Singlet dimers form between the diagonal spins. These states are connected through the crossover.

We have also analyzed the phases in the limiting parameter regime  $J_x \ll J \ll J_y$  and find that, in the absence of frustration, the stripe phase exhibits long-range correlations along both lattice directions. However, in the competing parameter regime shown in the quantum phase diagram, the spin arrangements remain striplike, but the nature of magnetic order becomes anisotropic: ferromagnetic long-range order develops along the direction with dominant ferromagnetic exchange, whereas the antiferromagnetic order is only short ranged. The true stripe phase emerges only for  $J_{x/y}^c > 2.39$  when  $J_{y/x} = 0$ , or for even larger values of  $J_{x/y}^c > 2.39$  when  $J_{y/x} > 0$  in our studied systems with finite width, however, an accurate crossover boundary  $J^c$  requires further finite-size scaling.

Our findings show significant deviations from those predicted by Schwinger boson mean-field theory (SBMFT) in Ref. [48]. Although all of the quantum phases proposed by SBMFT are observed, the phase boundaries differ markedly. Our DMRG results are, however, consistent with those obtained via ED. We also find that correlations along the leg increase with  $|J_x - J_y|$  for  $J_x > 0.5$  or  $J_y > 0.5$ , indicating the presence of finite short-range correlations in the spiral phase near the dimer line, except at the  $J_x = J_y$  line, where only diagonal correlations remain finite. While SBMFT predicts noncollinear spin structures along both the  $x$  and  $y$  directions, these were not confirmed by ED; our DMRG results, however, detect both types of spiral spin alignment in small regions of the phase diagram, with short but finite correlation lengths and it coexists with diagonal dimer.

Tassel *et al.* reported that  $(\text{CuCl})\text{LaNb}_2\text{O}_7$  exhibits ferromagnetic couplings with  $J_y/J \sim 0.39$  and  $J_x/J \sim 0.38$  [47], placing it on the  $J_x = J_y$  dimer singlet line. In contrast, Tsirlin *et al.* reported differing exchange parameters based on two types of density functional theory (DFT) calculations [42]. Inelastic neutron scattering (INS) data on powder samples reveal a dynamical structure factor  $S(Q, \omega)$  with a peak around  $Q \sim 0.5 \text{ \AA}^{-1}$ , suggesting that this compound lies near the dimer phase. However, a detailed theoretical investigation is still required to fully understand its spin configuration.

In summary, we have numerically investigated the isotropic Heisenberg spin-1/2 model on a SSL and constructed a quantum phase diagram using the DMRG method. We calculated spin-spin correlation functions and pitch angles that can be directly compared with experimental INS data. The phase boundaries derived from our DMRG analysis differ from those predicted by mean-field theory and ED. The transition from the stripe to the spiral-dimer phase is continuous and it goes from long-range to short-range order. Except for the ferromagnetic and stripe phase, all identified phases are gapped. Our results suggest that real materials such as  $(\text{CuCl})\text{LaNb}_2\text{O}_7$  [47,58–60] should be reexamined in light of this study. One can tune magnetic field to explore the quantum phases.

## ACKNOWLEDGMENTS

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