

From Néel long-range order to spin liquids in the multiple-spin exchange model

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The phase diagram of the multiple-spin exchange model on the triangular lattice is studied using exact diagonalizations. The two-spin (J_2) and four-spin (J_4) exchanges have been taken into account for 12-, 16-, 19-, 21-, 24-, and 27-site samples in the parameter region $J_4=0-0.25$ (for a fixed $J_2=1$). It is found that the three-sublattice Néel-ordered state built up by the pure two-spin exchange can be destroyed by the four-spin exchange, forming a spin-liquid state. The different data suggest that the phase diagram in this range of parameters exhibits two phases. The pure J_2 phase is a three-sublattice Néel-ordered phase; a small J_4 drives it into a spin-liquid state with a spin gap filled of a large number of singlets. This spin-liquid phase is not of the same generic kind as the phase studied by Misguich *et al.* [Phys. Rev. B **60**, 1064 (1999)]. It is observed on the finite-size samples that the spin-liquid phase, as the Néel-ordered phase, exhibits a magnetization plateau at $m=1/3$, and for $J_4>0.15$ a second plateau at $m=1/2$. These two plateaus are associated, respectively, with the semiclassical orderings *uud* and *uuud*.

I. INTRODUCTION

The two-dimensional triangular lattice antiferromagnet (2D-TLA) was first proposed to be a candidate for the disordered (or spin-liquid) ground state of the spin- $\frac{1}{2}$ Heisenberg model (Anderson and co-worker in the 1970's (Refs. 1 and 2). Different approaches failed to support this conjecture, but favor a ground state with Néel long-range order (LRO).³⁻⁵ Nevertheless, the lattice frustration on the 2D-TLA has attracted a great deal interest by theorists, providing a challenge for exotic antiferromagnets. Recently, the multiple-spin exchange model has been extensively studied as an alternative to the Heisenberg model, showing a rich structure of ground states.⁶⁻⁸ In this model, the ground state can be ferromagnetic (FM), antiferromagnetic (AFM) with Néel LRO, or a spin-liquid (SL). A prospective phase diagram has been given by Misguich *et al.*, who considered two-, four-, and five-spin exchange interactions on the 2D-TLA.⁸ They found that a large enough four-spin exchange interaction drives the FM phase into a SL phase. They did not study how the AFM Néel LRO is destroyed by the four-spin exchange interaction, and how the transition between Néel LRO and the short-range resonating valence bond (RVB) phase takes place. This question is the main object of this paper.

Unhappily there is no exact method allowing the study of the zero-temperature phases of such frustrated systems. For a finite system, however, one can always, in principle, represent the eigenstates in the complete basis of spin configurations. This allows one to have the exact ground states of small size systems through numerical computations. The huge number of spin configurations (2^N) becomes a great obstacle in the way of numerical simulations. On the most recent computers, the largest sample that may be handled in exact diagonalizations has 6×6 sites. On the triangular lattice, the quantum Monte Carlo method is plagued by the well-known sign problem, but a new technique called stochastic reconfiguration allows handling samples up to 12×12 sites.⁹ All these calculations point to Néel LRO, with a

sublattice magnetization of the order of 40% of the saturated value.⁵ Series expansions give a reduced (20%) but nonzero sublattice magnetization.¹⁰

On the other hand, in the case of short-range correlations, the situation is more straightforward, as soon as the available sizes are of the order of, or larger than, the correlation length. This is fortunately the case in the SL phase found in the J_2-J_4 model ($J_2\leq 0$, $J_4>0$) by Misguich *et al.*^{8,7}

In this work, we use exact diagonalizations to obtain the exact eigenenergies versus wave vectors and total spin for 12-, 16-, 19-, 21-, 24-, and 27-site samples of the J_2-J_4 model ($J_2=1$, $J_4>0$) on the 2D-TLA. In the classical limit, the AFM ground state of the 2D-TLA can be described as a three-sublattice structure, with spins of different sublattices making angles of $2\pi/3$. Periodic boundary conditions are compatible with the three-sublattice structure for samples with 12, 21, 24, and 27 sites, but not for the 16- and 19-site samples. Therefore, we use twisted boundary conditions for the 16- and 19-site samples⁴ and periodic boundary conditions for the 12-, 21-, 24-, and 27-site ones.

II. MODEL: THE MULTIPLE-SPIN EXCHANGE HAMILTONIAN

The Hamiltonian of the multiple-spin exchange model is given by

$$H = \sum_n (-1)^n J_n (P_n + P_n^{-1}), \quad J_n > 0, \quad n \geq 2, \quad (1)$$

where J_n are the n -spin exchange tunneling probabilities (exchange coefficients), and P_n and P_n^{-1} are the n -spin exchange operators and their inverse operators, respectively. The alternative sign in the summation over n in Eq. (1) comes from the permutation of fermions. In general, the exchange coefficients decrease with increasing n . The two-spin exchange term gives exactly the Heisenberg Hamiltonian up to a constant, since one has

$$P_2 = 2\mathbf{S}_i \cdot \mathbf{S}_j + \frac{1}{2}, \quad (2)$$

where \mathbf{S}_i and \mathbf{S}_j are spins localized at sites i and j , respectively. The three-spin exchange operator is exactly equivalent to a sum of two-spin exchange operators.⁸ Thus, the three-spin exchange term of Eq. (1) can be absorbed into the two-spin exchange term, as long as J_2 is replaced by the effective two-spin exchange coefficient $J_2^{\text{eff}} = J_2 - 2J_3$. Therefore, except for the two-spin exchange, the next most important term is the four-spin exchange. A pure positive two-spin exchange (i.e., the Heisenberg Hamiltonian) on the 2D-TLA gives an AFM phase with Néel LRO, and, as shown in Ref. 8, a pure four-spin exchange gives a SL phase. In this paper we use the specific properties of the spectra of these different kinds of phases to study the transition from one phase to the other, when the relative weight of the four-spin exchange J_4 increases relatively to the antiferromagnetic two-spin coupling. In the following, all the energies are measured in units of $J_2^{\text{eff}} = 1$.

Little is known of this region of the phase diagram. Previous works are based on a classical approximation,⁶ semi-classical spin-wave calculations¹¹ or mean-field Schwinger-boson results.¹² The classical result predicts a transition from the three-sublattice Néel state to a four-sublattice tetrahedral state at $J_4 = 0.24$. Both quantum approaches indicate that the four-spin exchange strongly enhances fluctuations in the three-sublattice Néel phase. Kubo *et al.*¹¹ found that the sublattice magnetization vanishes for $J_4 > 0.17$. In the Schwinger-boson approach,¹² the Néel state is destroyed when $J_4 > 0.25$. These two techniques have a general tendency to underestimate the effects of quantum fluctuations on ordered phases. The exact diagonalization analysis presented here indeed shows that the Néel long-ranged order disappears for a smaller value of J_4 (the critical value is estimated to be in the interval $J_4^C \sim 0.07-0.1$).

III. CRITERION TO DISCRIMINATE BETWEEN NÉEL LRO AND A SL PHASE: POSSIBILITY OF THE SPIN GAP

A. Finite-size energy spectrum of the Néel LRO phase

In the classical limit, an N -site 2D-TLA sample with Néel LRO is characterized by a three-sublattice structure with spin $N/6$ on each sublattice. Coupling of these three $N/6$ spins gives total spin S with $\min\{2S+1, N/2-S+1\}$ degeneracy.⁴ In an isotropic antiferromagnet (such as the collinear AFM which has equal spin susceptibilities and spin-wave velocities) the finite-size total energy depends on the total spin S (to first order in $1/N$) as

$$E_S = E_0 + \frac{1}{2N\chi} S(S+1), \quad (3)$$

where $E_0 = N\epsilon_0$ is the energy of the ground state in the thermodynamic limit and χ is the isotropic magnetic susceptibility of the sample. In the anisotropic case this equation should be rewritten:

$$E_S = E_0 + \frac{1}{2N\chi_{\perp}} S(S+1) + \frac{1}{2N} \left(\frac{1}{\chi_{\parallel}} - \frac{1}{\chi_{\perp}} \right) S_3^2, \quad (4)$$

where S_3 is the component of the total spin S on the internal symmetry axis of the spin system, and χ_{\parallel} and χ_{\perp} are the magnetic susceptibilities on the internal symmetry axis and on the perpendicular plane, respectively. In the broken symmetry picture the symmetry axis is perpendicular to the plane of the spins and χ_{\parallel} (χ_{\perp}) measures the spin fluctuations orthogonal to (in) the spin plane. Equation (3) [Eq. (4)] is the dynamical equation of a rigid rotator (of a quantum top).

Equations (3) and (4) show that the slopes of the total energy versus $S(S+1)$ and S_3^2 approach zero as $1/N$ does when $N \rightarrow \infty$. Here S_3 is an internal quantum number dynamically generated, which is not under control in a finite-size study. But the total spin S is a good quantum number and the N^{-1} scaling of the $S(S+1)$ dependence of the total energy versus sample size is interesting because it is more rapid than the scaling law of the order parameter (which goes as $N^{-1/2}$).^{4,13,14}

B. Finite-size scaling in the SL phase

In a SL phase, contrary to the Néel LRO phase, the spin gap (i.e., the difference in total energy between ground states in the $S=1$ sector and in the $S=0$ sector) does not collapse to zero in the thermodynamic limit. The finite-size scaling law in this second situation is not known exactly, insofar as the ‘‘massive’’ phase is not characterized precisely. Heuristically, we expect the finite-size spin gap to decrease exponentially to a finite value $\Delta(\infty)$ with the characteristic length ξ of the spin-spin correlations. For samples of linear size L smaller than the correlation length and in the crossover regime, there are not enough quantum fluctuations to destroy the sublattice magnetization and the system probably behaves as it were classical (i.e., with a spin gap decreasing as N^{-1}). The following heuristic law might be used to interpolate between the two behaviors:

$$\Delta(L) = \Delta(\infty) + \frac{\beta}{L^2} \exp(-L/\xi). \quad (5)$$

C. Quantum critical regime

The use of this heuristic law [Eq. (5)] encounters a severe difficulty as soon as the disordered system approaches a quantum critical point: in such a situation the correlation length ξ diverges, the gap closes to zero, and on a finite-size sample it is impossible to discriminate between such a situation and isotropic Néel LRO [Eq. (3)].

D. Numerical results

In view of this difficulty we have done a finite-size scaling of the spin gap by using the simplest linear $1/N$ behavior which probably gives a lower bound of the gap in a SL outside critical points. The physical reason is that we expect the finite-size corrections of the gap value to be smaller in a system with a finite correlation length (no long-range order) than in a LRO Néel phase where it vanishes as $1/N$.¹⁵ The results extrapolated to $N \rightarrow \infty$ are shown in Fig. 1. Strictly speaking the spin gap never extrapolates to zero except for a pure J_2 where it is equal to zero within its error bar. (At $J_4 = 0$, the data of Bernu *et al.*⁴ for $N=36$ are added to the

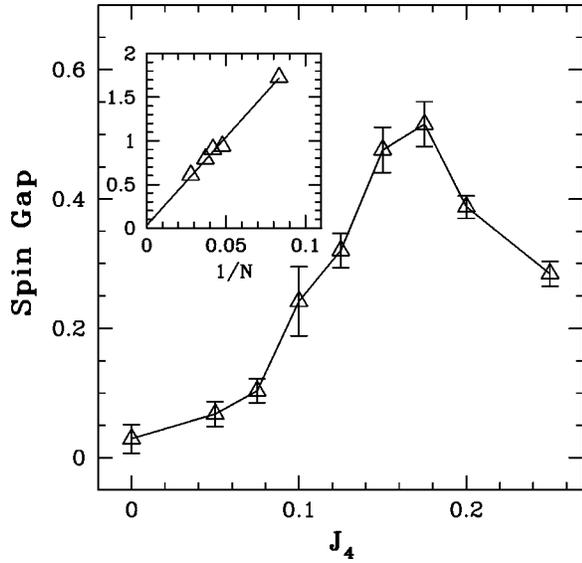


FIG. 1. Extrapolation $N \rightarrow \infty$ of the spin gaps of 12-, 21-, 24-, and 27-site samples for different J_4 . The inset shows the extrapolation at $J_4=0$. The spin gap for $N=36$ is evaluated from data of Ref. 4. Since not all symmetry sectors were investigated for $S=1$ and $N=36$, the energy of the lowest triplet is not known exactly. Therefore we estimated the spin gap by $\Delta = \frac{1}{6}[E(S=3) - E(S=0)]$, as in Ref. 5. Lines are guides for the eye.

present results; see Fig. 1.) Nevertheless, these data already show three distinct ranges for the parameter J_4 : for very small J_4 (below 0.075) Néel LRO is plausible but should be confirmed by another approach. For J_4 larger than 0.1 a gap certainly opens rapidly with increasing J_4 and then decreases for $J_4 > 0.175$. The spin gap criterion cannot give more insight into the phase diagram. We will now move to the analysis of the symmetries of the low-lying levels of the spectra to characterize more precisely these three phases.

IV. SYMMETRIES OF THE LOW LYING LEVELS IN A NÉEL-ORDERED PHASE

A. Theoretical background

First, we show the low-energy spectrum of the pure Heisenberg model on the 21-site sample (Fig. 2). In order to emphasize the low-energy structure we have displayed the low-energy spectrum minus a rigid rotator energy $\alpha S(S+1)$.

Let us first concentrate on the lowest part of the energy spectrum in each S sector (solid and open triangles in the figure). This family of levels forms on a finite-size lattice the quantum counterpart of the semiclassical Néel state. These specific states are in the trivial representation of the invariance group of the three-sublattice Néel ordered solution.⁴ To be definite, (i) the Néel ground state breaks the one-step translation but is invariant in a three-step translation: as a consequence the only wave vectors appearing in this family of QDJS's (for quasidegenerate joint states defined by Bernu *et al.*⁴) are, respectively, the center $\mathbf{k}=(0,0)$ and corners $\pm \mathbf{k}_0$ of the Brillouin zone.

(ii) These QDJS's belong specifically to the trivial representation of C_{3v} as the Néel state itself (i.e., they are invariant in a $2\pi/3$ rotation and in a reflection symmetry).

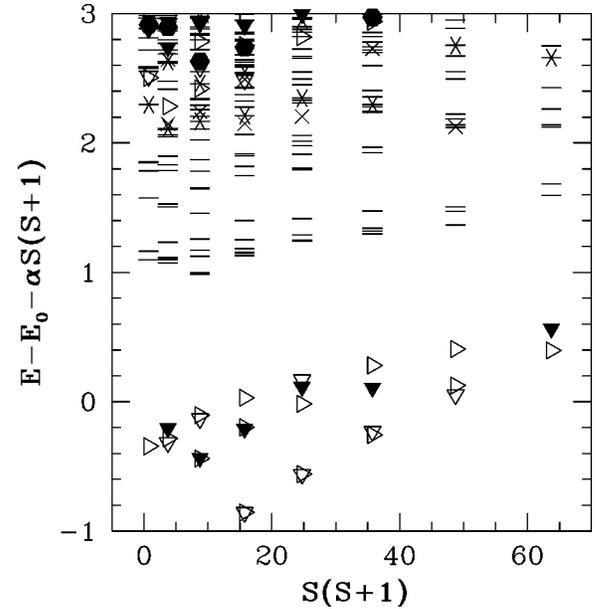


FIG. 2. Spectrum of the Heisenberg model ($J_4=0$) for the 21-site sample. ∇ : levels with $\mathbf{k}=(0,0)$ and symmetries $R_{2\pi/3}=1$ and $R_\pi=1$ (R_θ is the phase factor obtained in a θ rotation about the origin). \blacktriangledown : levels with $\mathbf{k}=(0,0)$ and symmetries $R_{2\pi/3}=1$, $R_\pi=-1$. \triangleright : levels with \mathbf{k}_0 (the corners of the Brillouin zone) and symmetries $R_{2\pi/3}=1$. Dashes: levels with wave vectors inside the Brillouin zone.

(iii) As the π rotation symmetry of the lattice is broken in this particular ground state, these QDJS's appear either in the odd or even representation of the twofold rotation group (see Ref. 4 for more details).

(iv) The numbers and characteristics (quantum numbers) of the QDJS's in each S sector are precisely fixed by theory:⁴ for the 21-site spectrum displayed in Fig. 2 (as for all sizes that have been studied up to now) the numbers of low-lying levels and their quantum numbers correspond exactly to the above-mentioned theoretical predictions.

B. Numerical results

The dynamical law given by Eq. (4) is still imperfectly obeyed for the 21-site sample: in particular the generation of the internal symmetry is still imperfect but nevertheless the spectrum of a quantum top could already be anticipated.

Above these levels with specific properties, there appear eigenstates with wave vectors belonging to the inside of the Brillouin zone (simple dashes in Fig. 2). A group of such eigenstates with different total spin represents a magnon excitation of the Néel ground state. As the antiferromagnetic magnons have a linear dispersion law, the softest magnon energy scales as the smallest wave vector accommodated in the Brillouin zone of the finite-size sample. Thus, for sizes large enough, these levels collapse to the ground state as $1/\sqrt{N}$, more slowly than the QDJS's which collapse as $1/N$ to the thermodynamic Néel ground-state energy. That is the reason for the appearance in Fig. 2 of a quasigap between the QDJS's and the magnon excitations.

This hierarchy of low-lying levels is a very strong constraint on the finite-size samples spectra. It is perfect for

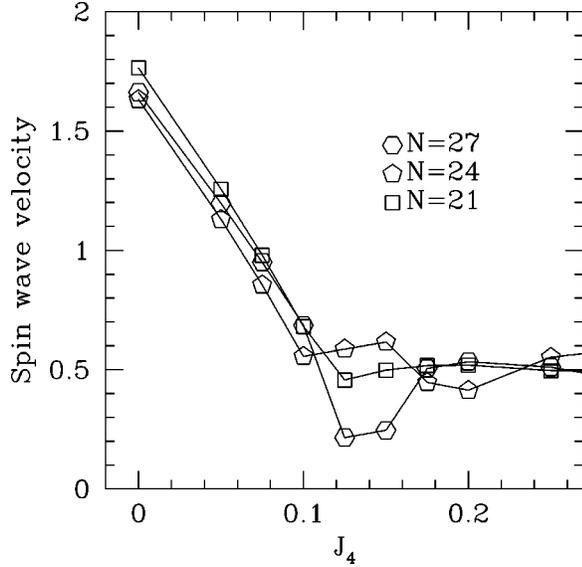


FIG. 3. Spin-wave velocities of 21-, 24- and 27-site samples. Lines are guides for the eye. These quantities are computed as the ratio of the first $\Delta S = 1$ excitation energy divided by the momentum of the corresponding excitation (according to $v = \Delta E / \Delta \mathbf{k}$). For $J_4 > 0.1$, these numbers do not correspond to well-defined physical excitations because of the strong perturbations of the spectra. Notice at $J_4 = 0.1$, Fig. 4 exhibits a huge number of low-energy singlet states below the spin gap and the system is therefore already in a SL phase.

sizes up to 27 and for J_4 smaller or equal to 0.075 and totally absent¹⁶ for J_4 larger or equal to 0.1.

(i) This result, associated with the spin-gap behavior, consistently proves that for J_4 larger or equal to 0.1 the system is in a spin-liquid state with rather short-range spin-spin correlations.

(ii) For $J_4 \leq 0.075$ the structure of the low-lying eigenlevels of the spectra are compatible with Néel LRO. But as discussed above, it is indeed impossible to precisely point to a quantum critical transition within this approach. In view of the spectra, we might speculate that the transition is second order and that it takes place between 0.07 and 0.1.

(iii) From $J_4 = 0$ to $J_4 \approx 0.1$ we see a softening of the spin-wave velocity consistent with the gradual decrease of the Néel LRO (Fig. 3). However, Fig. 4 shows that a large number of singlet states are already present at low energy when $J_4 = 0.1$. Therefore, the system is certainly no longer in the ordered phase at $J_4 \approx 0.1$. We can conclude from Fig. 3 that the spin-wave velocity does not vanish at the critical point (even if the precise location of the transition cannot be determined). This has been previously suggested by various analytical approaches.¹⁷⁻¹⁹ The sizes studied are nevertheless too small to check the prediction of Azaria *et al.*¹⁷ of an $O(4)$ symmetry of the effective field theory at the critical point.

V. LOW-ENERGY EXCITATIONS OF THE SL PHASE

Contrary to our expectations, the SL phase which appears immediately after the disappearance of the Néel-ordered phase is not the phase studied by Misguich *et al.*⁸ It is indeed a SL phase, with a gap and short-range spin-spin correla-

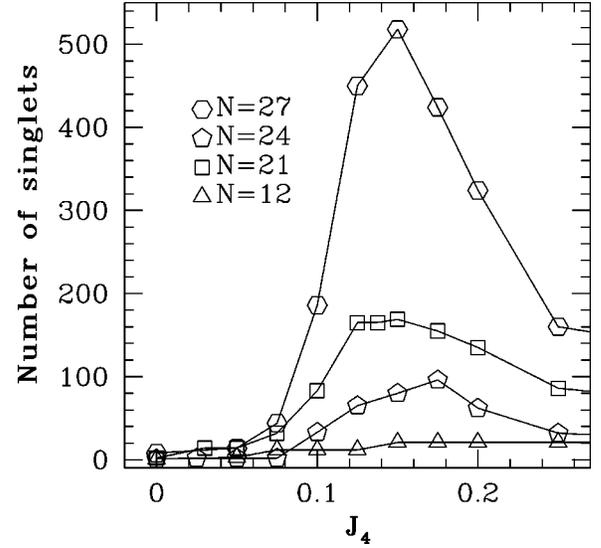


FIG. 4. Number of singlet states between the ground states of the $S = 0$ (or $1/2$) and $S = 1$ (or $3/2$) sectors. Lines are guides for the eye.

tions. But as might be seen in Fig. 4, this phase exhibits a very large number of singlets in the magnetic gap and seems in this respect similar to the spin-liquid phase of the Heisenberg model on the *kagomé* lattice.^{20,21} However, we are not aware of any exponential degeneracy [i.e., $\sim \exp(N)$] in the classical multiple-spin exchange (MSE) model,²² as is the case for the classical *kagomé* antiferromagnet.

We suspect that a much larger four-spin exchange parameter will be needed to recover the SL phase studied by Misguich *et al.* These data point to the existence of this phase in a finite range of parameters $0.075 \leq J_4 \leq 0.25$. However, one cannot disregard the hypothesis that these properties are in fact those of a critical point, with a critical region enlarged by finite-size effects. More work with different methods is needed to clarify this point.

VI. MAGNETIZATION PLATEAUS

In an external magnetic field B along the z axis, the total energy of the state with component S_z of the total spin is given by

$$E_B = E_S - S_z B. \quad (6)$$

The magnetization is determined by the minimum of E_B respective to S_z , which requires $\partial E_B / \partial S_z = 0$. Therefore, in the isotropic case with Néel LRO, one has

$$m = 2\chi B - 1/N, \quad (7)$$

where $m = 2S/N$ is the polarization relative to the saturated magnetization $N/2$ and χ is indeed the magnetic susceptibility.

We noticed that when the four-spin exchange interaction increases, deviation from Eq. (3) occurs about $S_z = N/6$ and $N/4$. This is in agreement with the earlier mean-field calculation of Kubo and Momoi⁶ who predicted magnetization plateaus at $m = 1/3$ and $m = 1/2$ in the $J_2 - J_4$ model. We

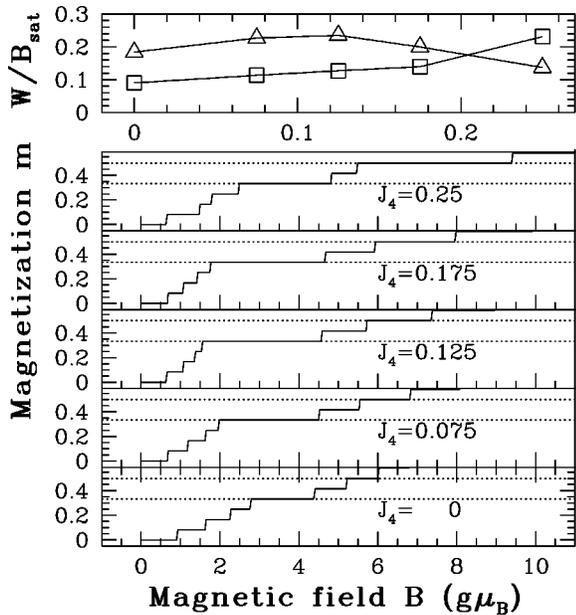


FIG. 5. Magnetization vs external magnetic field for different J_4 ($N=24$). The dotted lines label the $1/3$ and $1/2$ magnetization. The top panel shows the widths of the $1/3$ (∇) and $1/2$ (\square) plateaus vs J_4 .

present the magnetization curve of the 24-site sample in Fig. 5. A small plateau exists at $1/3$ magnetization for $J_4=0$, and its width first increases slowly as J_4 increases and then decreases from around $J_4=0.125$. This $1/3$ plateau has also been found in previous studies (see Ref. 24 and references therein) in the pure three-sublattice Néel-ordered system. The $m=1/2$ magnetization appears at about $J_4=0.1$ and its

width increases with J_4 . Finally, there still exists a plateau at about $m=1/2$ in samples with odd numbers of sites, but it distributes over the two closest positions to the $1/2$ magnetization. The $m=1/3$ and $m=1/2$ plateaus correspond to the classical uud and $uuud$ ordering of spins (see Momoi *et al.*²³ for the four-sublattice $uuud$ state). These phases are more “classical” than the zero-field phase, but yet show a decrease of the sublattice magnetization from the classical saturation values.

VII. CONCLUSION

In conclusion, we studied the transition between the Néel-ordered state and a spin-liquid state of the multiple-spin exchange model by means of the exact diagonalization method. The pure three-sublattice Néel-ordered phase is gradually destroyed by quantum fluctuations when increasing the four-spin exchange coupling. The spin-wave velocity decreases but apparently remains finite at the transition. The quantum-disordered phase tuned by the four-spin exchange coupling is different from the pure J_4 phase studied by Misguich *et al.* It exhibits low-energy singlet excitations, reminding one of *kagomé* SL. This result opens many interesting questions that cannot be answered in the present framework: is this phase a new generic SL phase or a finite-size manifestation of a quantum critical regime? Are these singlet excitations the “resonon” modes invoked by Rokhsar and Kivelson?²⁵ In agreement with previous studies,⁶ we find two magnetization plateaus at $1/3$ and $1/2$ of the full magnetization. These plateaus are associated with the semiclassical uud and $uuud$ ordering structures. A finite-size scaling on much larger sizes is needed to draw a definite conclusion on this magnetic phase diagram.

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¹⁵This assumption would be invalid at a critical point where the

uniform susceptibility vanishes: In such a case the gap might close as $1/\sqrt{N}$. The analysis of Sec. IV shows that the behavior of the system changes rather abruptly from a Néel-like spectrum to a spectrum with a very large number of low-lying singlets below the first $S=1$ state (and potentially a $T=0$ residual entropy in the singlet sector). This happens before any decrease to zero of the spin velocity or of the homogeneous spin susceptibility. So, if the true thermodynamical system has indeed a critical point between the two phases, the sizes we are looking at are too small to scrutinize the critical regime.

¹⁶An illustration of the striking qualitative differences between spectra in the Néel region and in the spin-liquid phase has been previously published: two $N=27$ spectra at $J_4=0$ and $J_4=0.1$ are displayed in Fig. 2 and Fig. 3 of Ref. 8.

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²²Momoi *et al.* (Ref. 23) found a ground-state degeneracy in the classical MSE model but this degeneracy only grows as the ex-

ponential of linear size of the system ($\sim \exp\sqrt{N}$). Moreover, this classical degeneracy was only found in a region where $J_2 < 0$ is *ferromagnetic* ($\frac{1}{4} \leq J_4 / |J_2| \leq \frac{3}{4}$).

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