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Spin-liquid versus dimer phases in an anisotropic J_1-J_2 frustrated square antiferromagnet

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The spin-1/2 J_1-J_2 antiferromagnet is a prototypical model for frustrated magnetism and one possible candidate for a realization of a spin-liquid phase. The generalization of this system on the anisotropic square lattice is given by the J_1-J_2-J_1'-J_2' Heisenberg model, which can be treated by a renormalization group analysis of coupled frustrated chains. The J_1-J_2-J_1'-J_2'-model shows an interesting interplay of Néel order, dimerization, and spin-liquid behavior. Extrapolating the analytical findings supported by numerical results indicates that for the isotropic model the phase at intermediate coupling strength $0.4J_1 \lesssim J_2 \lesssim 0.6J_1$ is not a spin liquid, but has instead a staggered dimer order.

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In the search of exotic quantum states and quantum phase transitions, frustrated antiferromagnetic systems have increasingly become the center of attention [1,2]. The competing interactions between spins potentially lead to a large entropy even at low temperatures, which together with quantum fluctuations may give rise to quantum phases with unconventional or topological order parameters [3–6]. In particular, the so-called spin-liquid state without long-range order of a conventional (local) order parameter has been much discussed in the literature ever since Anderson related this phase to high-temperature superconductivity [7]. A solid proof for a system which shows a spin-liquid ground state has long been elusive, due to inherent numerical and analytical problems in frustrated systems. Nonetheless, some good evidence for possible spin-liquid states has recently been presented for the Hubbard model on the honeycomb [8] and the anisotropic triangular lattice [9], as well as for the Heisenberg model on a kagome lattice [1,10–12] and on the J_1-J_2 frustrated square lattice [13,14].

In particular, the J_1-J_2 Heisenberg model has been treated with a vast array of theoretical methods [13–44], which is also the model originally considered by Anderson [7]. Early numerical works have shown an intermediate phase between ordinary Néel order for $J_2 \lesssim 0.4J_1$ and collinear Néel order for $J_2 \gtrsim 0.6J_1$ [16], but the underlying correlations in this phase remain hotly debated. Most works have predicted a plaquette or columnar dimer order as the most likely scenario [15,17–27], but more recent numerical works have again proposed a spin liquid [13,14]. Unfortunately, the issue may never be conclusively solved using numerical methods, since convergence with finite size and/or temperature of data from density matrix renormalization group (DMRG) or tensor matrix methods is very slow. In particular, it was shown recently for the related two-dimensional (2D) J-Q model that the finite size scaling on moderate lengths would lead to the prediction of a spin liquid, even though the system orders in the thermodynamic limit [45]. In general, the slow convergence is related to the observation that spin-liquid states are often very close in energy to competing states with dimer order [11,12].

In light of the disappointing numerical situation, analytical methods become very important, but the problem is difficult since series expansions [17,37], chain mean-field theories [46], spin wave theories [24], or coupled cluster methods [26,41,43] have to incorporate frustrating interactions, which tend to cancel each other. Therefore, the methods often become unstable or diverge near the most interesting phase transitions. Another very promising analytical approach is to couple one-dimensional (1D) systems [18,47–50]. However, even in this case the frustrating interchain interactions tend to cancel each other so that interesting new phases only come up in higher order treatments at intermediate interchain coupling strength. This is not necessarily a fundamental limitation, however, since the leading frustration effects can be taken into account within each chain if a different geometry is chosen as we will show here.

We now propose to study a more general J_1-J_2-J_1'-J_2' model, which takes into account the effects of frustration *within* each chain before they are coupled, but at the same time allows one to make exact analytic predictions in parts of the phase diagram. The model is given by the standard Heisenberg model on the 2D lattice

$$H = \sum_{\langle i,j \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j \tag{1}$$

with $J_{ij} > 0$ acquiring values according to the geometry of Fig. 1. Notice that the lattice vectors of the square lattice are tilted by 45° relative to the directions of the zigzag chains. One special property of this model is that it can be treated by a renormalization group (RG) treatment of quasi-one-dimensional (1D) chains which becomes exact in the limit of small interchain coupling J_1', J_2' while the in-chain frustration J_2 is now *finite* and can be fully taken into account without the need to go to higher order RG equations. In fact, it turns out that J_2 becomes the determining factor for the behavior of the phase diagram. We predict the intermediate phase to posses a *staggered* dimer order, which is in contrast to previous considered states of spin liquids [13,14], columnar dimer order, and plaquette dimer order [18,24,26].

Our starting point is the 1D model without interchain couplings $J_1' = J_2' = 0$, which is well understood in terms of a $(1+1)$-dimensional field theory Hamiltonian based on Abelian or non-Abelian bosonization [51]. In the continuum limit the spin operators are expressed in terms of field theory operators

$$\mathbf{S}(x) \approx \mathbf{J}(x) + (-1)^x \Omega \mathbf{n}(x), \tag{2}$$

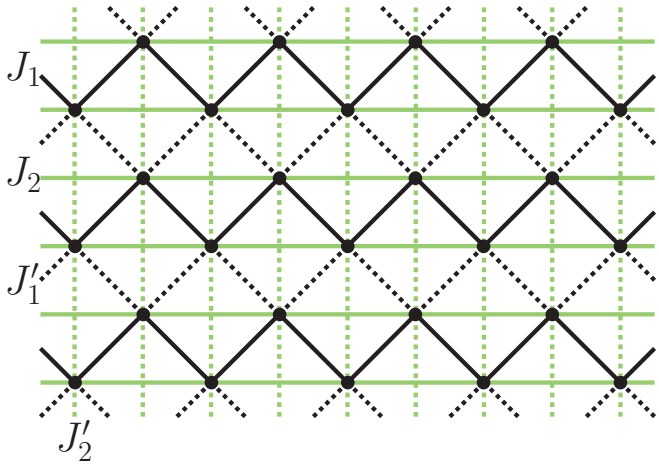

FIG. 1. (Color online) The generalized anisotropic J_1-J_2-J_1'-J_2' antiferromagnet.

where Ω is a nonuniversal constant of order one, $\mathbf{J} = \mathbf{J}_L + \mathbf{J}_R$ is the sum of the chiral SU(2) currents of the Wess-Zumino-Witten (WZW) model, and $\mathbf{n}$ is related to the fundamental field g of the WZW model via $\mathbf{n} = \mathrm{tr}\,\boldsymbol{\sigma} g$. The lattice constant is set to unity here. The conformally invariant fixed point Hamiltonian H_0 of the frustrated chain is perturbed by the backscattering marginal operator [52]

$$H = H_0 + 2\pi v \lambda_a \int dx\, \mathbf{J}_L \cdot \mathbf{J}_R, \quad \text{with}$$
$$H_0 = \frac{2\pi v}{3} \int dx [:\mathbf{J}_L \cdot \mathbf{J}_L: + :\mathbf{J}_R \cdot \mathbf{J}_R:], \tag{3}$$

where the dots denote normal ordering. The velocity is approximately $v = \frac{\pi J_1}{2} - 1.65 J_2$ if a linear dependence on J_2 is assumed [53], while to leading order [54] $\lambda_a \approx 1.723(J_2 - J_c)$ where $J_c \approx 0.241\,167 J_1$ [54–57].

The leading instabilities of the chains are a finite staggered field $(-1)^x \mathbf{S}(x) \propto \mathbf{n}$ with effective scaling dimension $d_n = 1/2 + \lambda_a/4$ and a dimerization $(-1)^x \mathbf{S}(x) \cdot \mathbf{S}(x+1) \propto \epsilon$, where $\epsilon = \mathrm{tr}\, g$ has effective scaling dimension $d_\epsilon = 1/2 - 3\lambda_a/4$ [52]. The marginal coupling λ_a itself renormalizes and becomes relevant for $\lambda_a > 0$, which leads to a dimerized phase for $J_c < J_2 < \infty$ [54–58]. Therefore, the in-chain frustration J_2 determines if the leading instability is dimerization for $J_2 > J_c$ or Néel order for $J_2 < J_c$. For $J_2 = J_c$ the system is a spin liquid in the form of a Luttinger liquid.

In the presence of interchain couplings J_1' and J_2' the field theories of neighboring chains become coupled and give rise to a richer operator content [59–61]. Any chain in the system is coupled to two neighboring chains, so it is natural to consider three copies of the field theory that are weakly coupled. For self-consistency all three copies must renormalize in the same way, which implies the following symmetric formulation of the perturbing Hamiltonian in terms of three non-Abelian field theories $\nu = 1,2,3$:

$$\delta\mathcal{H} = 2\pi v \sum_{\nu=1}^{3} [\lambda_a \mathbf{J}_{\nu,L} \cdot \mathbf{J}_{\nu,R} + \lambda_n \mathbf{n}_\nu \cdot \mathbf{n}_{\nu+1} + \lambda_\epsilon \epsilon_\nu \epsilon_{\nu+1} + \lambda_b (\mathbf{J}_{\nu,L} \cdot \mathbf{J}_{\nu+1,R} + \text{e.c.}) + \lambda_c (\mathbf{J}_\nu \cdot \mathbf{n}_{\nu+1} - \text{e.c.})], \tag{4}$$

with e.c. denoting the operator after exchanging the chain index, and the bare couplings are

$$\lambda_n = \Omega^2 \frac{(J_2' - J_1')}{2\pi v}, \quad \lambda_b = \frac{J_1' + J_2'}{2\pi v}, \quad \lambda_c = \Omega \frac{J_1'}{2\pi v}, \quad \lambda_\epsilon = 0. \tag{5}$$

Letting the ultraviolet cutoff evolve according to $\Lambda(l) = \Lambda_0 e^{-l}$ and using the operator product expansion [62–65], we get the following coupled differential equations for the coupling constants [66]:

$$\dot{\lambda_a} = \lambda_a^2 + \lambda_\epsilon^2 - \lambda_n^2, \tag{6a}$$
$$\dot{\lambda_b} = \lambda_b^2 - \lambda_\epsilon \lambda_n + \lambda_n^2, \tag{6b}$$
$$\dot{\lambda_\epsilon} = \lambda_\epsilon + \tfrac{3}{2}\lambda_a \lambda_\epsilon - \tfrac{3}{2}\lambda_b \lambda_n - \tfrac{3}{2}\lambda_c^2, \tag{6c}$$
$$\dot{\lambda_n} = \lambda_n - \tfrac{1}{2}\lambda_a \lambda_n - \tfrac{1}{2}\lambda_b \lambda_\epsilon + \lambda_b \lambda_n - \lambda_c^2, \tag{6d}$$
$$\dot{\lambda_c} = \tfrac{1}{2}\lambda_c - \tfrac{1}{4}\lambda_a \lambda_c + \lambda_b \lambda_c + \tfrac{1}{2}\lambda_c \lambda_\epsilon - \lambda_c \lambda_n, \tag{6e}$$

where $\dot{\lambda} = d\lambda/dl$.

The system remains approximately scale invariant as long as $\Lambda(l)$ is the largest energy scale in the system. However, typically one of the coupling constants λ becomes of order unity at one point so that the RG equations break down. This defines a new intrinsic energy scale Δ, below which scale invariance is lost and no further renormalization is possible.

The couplings λ_n and λ_ϵ are the most relevant and likely dominate. A large negative $\lambda_n < 0$ corresponds to a regular Néel order, while for a large positive value $\lambda_n > 0$ the Néel order on each chain is collinear to the neighboring chains. A large $\lambda_\epsilon < 0$ indicates a staggered dimer order on the 2D lattice, while $\lambda_\epsilon > 0$ gives a herringbone dimer structure. Depending on the microscopic couplings, it is also possible for the system to acquire a dimerized or magnetically ordered ground state when one of the marginal couplings λ_a or λ_b, respectively, reaches first the strong coupling limit [67,68]. Finally, the relevant coupling constant λ_c with scaling dimension $d_c = 3/2$ may also become largest, a case where no simple order can be assigned to the 2D system and a spin-liquid phase is possible.

We now proceed to numerically integrate the RG equations (6) for different lattice parameters until one of the couplings becomes unity [56,68]. The constant Ω in the definition of the initial values in Eq. (5) must be of order unity according to bosonization, but cannot be derived exactly, so that we choose $\Omega = 1$ here. We have also tested other values for the cutoff and Ω and found that the resulting phase diagram remains qualitatively the same, while the quantitative positions of the phase transition lines may change slightly (see below).

There are two key parameters of physical interest in this model, namely, the frustration $f = \frac{J_2}{J_1} = \frac{J_2'}{J_1'}$ and the anisotropy $r = \frac{J_1'}{J_1} = \frac{J_2'}{J_2}$, which determine the value of all four couplings [69]. The isotropic 2D model is recovered for $r \to 1$. The resulting phase diagram as a function of r and f is shown in Fig. 2.

Lower critical line. The lower critical line $f_{c_1}(r)$ between the *Néel* and the *staggered dimer* order is determined by the competition between the relevant coupling $\lambda_n < 0$ and the marginally relevant coupling $\lambda_a > 0$. The relevant coupling

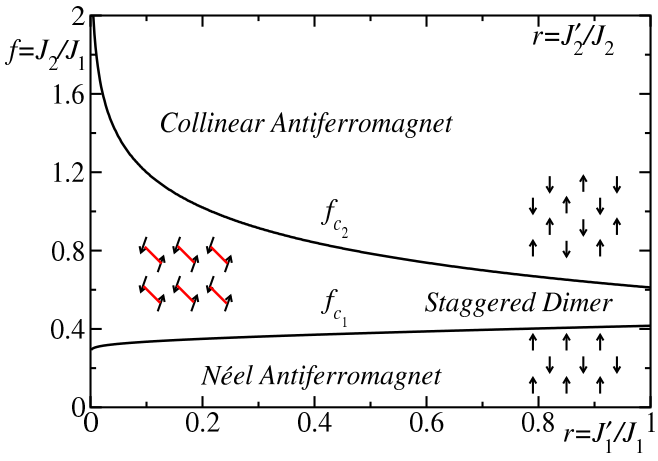

FIG. 2. (Color online) The phase diagram of the anisotropic J_1-J_2-J_1'-J_2' model from the RG analysis as a function of the anisotropy r and the frustration f. The analytical functions in Eqs. (7) and (10) are indistinguishable on this scale.

λ_ϵ is initially zero and remains so small that it does not play a role here. For a different choice of a strongly reduced value for $\Omega = 0.5$ the critical line is shifted down by 10%, so that Néel order is slightly suppressed for $\Omega < 1$ (and slightly stabilized for $\Omega > 1$).

If we completely ignore all coupling constants except λ_n and λ_a in Eqs. (6), the lower critical line $f_{c_1}(r)$ can be analytically determined to be

$$f_{c_1}(r) = J_c/J_1 + \frac{a_1}{b_1 - \ln r}, \tag{7}$$

which is the critical value of frustration at which the breakdown energy scales for dimer order $\Delta_a = \Lambda_0(1 - 1/\lambda_a)$ and Néel order $\Delta_n = \Lambda_0\lambda_n$ become equal. In terms of the initial couplings we have $a_1 \approx 1/1.723$ and $b_1 \approx 1 + \ln[2\pi v/(J_1 - J_c)\Omega^2]$ from Eq. (5). Remarkably, this analytical form remains a rather accurate estimate for the fully integrated RG flow even in the presence of all other couplings, albeit with fitted values of $a_1 = 0.459$ and $b_1 = 2.629$. In the limit $r \to 1$, we predict the critical point $f_{c_1}(1) \to 0.416$ for the transition to a staggered dimerized phase in the 2D J_1-J_2 square antiferromagnet.

Upper critical line. While a single chain remains in the dimer phase for $J_c < J_2 < \infty$ [56,58], in the J_1-J_2-J_1'-J_2' model a large J_2 can destroy the dimer phase [70]. This behavior defines an upper critical line $f_{c_2}(r)$, with $f_{c_2}(0) \to \infty$, beyond which the system is driven out of the dimer phase. The RG equations (6) *cannot* capture this behavior since it occurs for large deviations from $J_2 = J_c$. However, it is possible to consider the limit $J_2 \gg J_1$ as a new fixed point, where (unfrustrated) chains along the horizontal J_2 direction in Fig. 1 are weakly coupled by all other couplings. The low-energy physics is determined by the competition of a coupling with J_2' to every second chain (rectangular pattern) and a coupling J_1 or J_1' which couples even and odd chains (zigzag pattern) [48,56,58,71]. Here the weaker coupling $J_1' < J_1$ can never dominate over J_1 in the course of the RG flow and can safely be ignored. In the limit of small J_1 the coupled chain system above is now described by six copies of the field theory which are coupled self-consistently corresponding to the horizontal chains in Fig. 1 without in-chain frustration:

$$\begin{aligned}\delta\mathcal{H}' = 2\pi v \sum_{\nu=1}^{6} [&\lambda_a \mathbf{J}_{\nu,L}\cdot\mathbf{J}_{\nu,R} + \lambda_b(\mathbf{J}_{\nu,L}\cdot\mathbf{J}_{\nu+2,R} + \text{e.c.}) \\ &+ \lambda_\epsilon\epsilon_\nu\epsilon_{\nu+2} + \lambda_n\mathbf{n}_\nu\cdot\mathbf{n}_{\nu+2} + \lambda_v\epsilon_\nu\partial_x\epsilon_{\nu+1} \\ &+ \lambda_u(\mathbf{J}_{\nu,L}\cdot\mathbf{J}_{\nu+1,R} + \text{e.c.}) + \lambda_w\mathbf{n}_\nu\cdot\partial_x\mathbf{n}_{\nu+1}].\end{aligned} \tag{8}$$

The initial bare couplings λ_ϵ and λ_v vanish, $\lambda_a \approx -0.36$ [54], and

$$\lambda_u = \frac{2J_1}{2\pi v}, \quad \lambda_w = \Omega^2\frac{J_1}{2\pi v}, \quad \lambda_b = \frac{J_2'}{2\pi v}, \quad \lambda_n = \Omega^2\frac{J_2'}{2\pi v}.$$

The RG flow of this set of operators reads

$$\dot{\lambda}_a = \lambda_a^2 + \lambda_\epsilon^2 - \lambda_n^2 + \tfrac{1}{4}\left(\lambda_w^2 - \lambda_v^2\right), \tag{9a}$$

$$\dot{\lambda_b} = \lambda_b^2 - \lambda_\epsilon\lambda_n + \lambda_n^2, \tag{9b}$$

$$\dot{\lambda_\epsilon} = \lambda_\epsilon + \tfrac{3}{2}\lambda_a\lambda_\epsilon - \tfrac{3}{2}\lambda_b\lambda_n, \tag{9c}$$

$$\dot{\lambda_n} = \lambda_n - \tfrac{1}{2}\lambda_a\lambda_n - \tfrac{1}{2}\lambda_b\lambda_\epsilon + \lambda_b\lambda_n, \tag{9d}$$

$$\dot{\lambda}_u = \lambda_u^2 - \tfrac{1}{2}\lambda_v\lambda_w + \tfrac{1}{2}\lambda_w^2, \tag{9e}$$

$$\dot{\lambda}_v = \tfrac{3}{4}\lambda_a\lambda_v - \tfrac{3}{2}\lambda_u\lambda_w, \tag{9f}$$

$$\dot{\lambda}_w = -\tfrac{1}{4}\lambda_a\lambda_w - \tfrac{1}{2}\lambda_u\lambda_v + \lambda_u\lambda_w, \tag{9g}$$

where the rectangular $(\lambda_b,\lambda_\epsilon,\lambda_n)$ and the triangular $(\lambda_u,\lambda_v,\lambda_w)$ patterns renormalize almost independent of each other, connected only indirectly via the in-chain marginal coupling λ_a. In this case, the equations result in a competition between a *collinear* Néel order with dominant $\lambda_n > 0$ (rectangular pattern) and a dimer order with a dominant $\lambda_u > 0$ (zigzag pattern). While the collinear Néel order is different from the regular Néel order discussed above, the dimer order is actually continuously connected to the staggered dimer order in the opposite limit $J_2 \gtrsim J_c$ [48,56,59,67], as indicated in Fig. 2.

Again, ignoring all coupling constants except for λ_u and λ_n, we can find an analytic solution for the upper phase transition line $f_{c_2}(r)$:

$$f_{c_2}(r) = a_2 - b_2 \ln r, \tag{10}$$

with $a_2 = (1 + \ln 2\pi v/\Omega^2)/\pi v$ and $b_2 = 1/\pi v$. Remarkably, the integrated RG flow from all couplings is again fitted well by the same form with $a_2 = 0.606$ and $b_2 = 0.259$. Therefore, the critical point for transition from dimer to collinear Néel order in the 2D model is predicted to be $f_{c_2}(1) = 0.606$.

We now turn to an independent numerical check of the phase diagram of a minimal model of two zigzag chains with in-chain frustration J_2 and interchain coupling J_1' and J_2' using the DMRG algorithm [72]. This model can be used to study the stability of the different phases, but quantitative agreement with the full 2D J_1-J_2-J_1'-J_2' model should not be expected. To obtain the phase diagram we use the dimer order O_D parameter

$$O_D = \tfrac{1}{2}|[h_1(l) - h_1(l+1)] + [h_2(l) - h_2(l+1)]|, \tag{11}$$

with $h_\nu(l) = \langle\mathbf{S}_{\nu,l}\cdot\mathbf{S}_{\nu,l+1}\rangle$ and the two-rung entropy $\tilde{S} = -\mathrm{Tr}\,\tilde{\rho}\ln\tilde{\rho}$ in terms of the reduced density matrix $\tilde{\rho}$ of two successive rungs [59,73]. Possible anomalies in $\tilde{S}$ or its derivative with respect to some microscopic coupling $\tilde{S}'$ indicate a first- or second-order phase transition, respectively

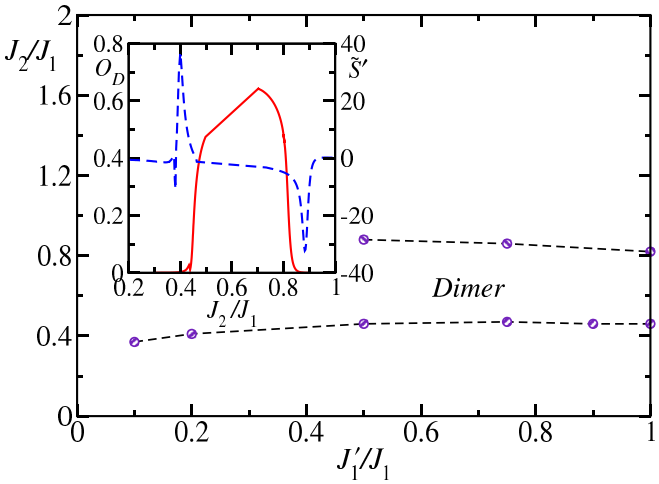

FIG. 3. (Color online) Numerical DMRG results for the phase diagram of two chains with in-chain frustration $f = J_2/J_1$ and interchain couplings $r = J_1'/J_1 = J_2'/J_2$. Inset: Dimer order parameter (red solid line) and the derivative of the two-rung entropy (blue dashed line) as a function of f for $J_1' = J_1$ and $L = 128$ rungs.

[74–77]. Measurements are taken close to the center of a system with open boundary conditions and we then apply finite size scaling as a function of both the number of DMRG states kept M and the size of the system L [78].

As shown in the inset of Fig. 3, the onset of the dimer order O_D coincides with the anomalies of the derivative $\tilde{S}' = d\tilde{S}/dJ_2$, indicating a second-order phase transition. The resulting phase diagram as a function of $r = J_1'/J_1$ and $f = J_2/J_1$ is shown in Fig. 3. In the intermediate region we find a nonzero-order parameter O_D corresponding to an in-chain dimerization and a *staggered* dimer arrangement in the 2D model. Note that Néel order in the lower part of the phase diagram corresponds to ferromagnetically correlated rungs of the zigzag chains and vice versa for the collinear order. The numerical data indicates that the intermediate phase is stable for all r.

In summary, we have considered the general anisotropic J_1-J_2-J_1'-J_2' model in Fig. 1 which can be tuned from $r = 0$ (decoupled chains) to $r = 1$, where it becomes the famous J_1-J_2 model [7,13–44], which is known to have an intermediate phase for $0.4 \lesssim J_2/J_1 \lesssim 0.6$ with strongly debated underlying correlations. One advantage of the J_1-J_2-J_1'-J_2' model is that the RG analysis becomes *exact* in the vicinity of two fixed points in the phase diagram, namely, close to $r \ll 1$, $f = J_c/J_1 = 0.241\,167$ and close to $r \ll 1 \ll f$. At those points a transition from a Néel order to a staggered dimer order and a transition from a staggered dimer order to Néel order can be rigorously shown. The phase transition lines behave asymptotically according to Eqs. (7) and (10), respectively. For larger values of r the RG solution becomes less reliable, but the predicted range of the staggered dimer phase $0.416 \lesssim J_2/J_1 \lesssim 0.606$ for $r = 1$ is in surprising agreement with previous studies. In principle, the staggered dimer phase could also be limited for larger r, which is in fact the case for the corresponding ladder model without frustrating interchain couplings $J_2' = 0$ [70]. However, in the J_1-J_2-J_1'-J_2' model there is a different symmetry for large and small values of f, so an intermediate phase must exist for all values of $r \leqslant 1$. The most likely scenario is therefore that the staggered dimer phase continuously extends from $r = 0$ to $r = 1$. Indeed, our numerical simulations and the RG equations do not indicate that the nature of this phase changes as a function of r. However, if the dimer phase does change it may do so only partially as a function of r and very recent numerical results indeed suggest that for $r = 1$ a spin liquid may exist *in part* of the phase diagram [15]. The RG approach presented here can only capture the leading instability between neighboring chains, but the results indicate that this appears to be sufficient to capture the essential physical behavior. For spin-liquid behavior, the dimer order would have to be destroyed by the emergence of long-range coherence perpendicular to the chains. In order to explore such an exotic possibility it would therefore be a promising future research direction to study a larger number of coupled chains in the J_1-J_2-J_1'-J_2' model with 2D numerical methods.

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