Frustrated magnetism and resonating valence bond physics in 2D kagome-like magnets

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We determine the phase diagram and the low-energy physics of three Heisenberg antiferromagnets which, like the kagome lattice, are networks of corner-sharing triangles but which contain more short inequivalent resonance loops. We use a combination of exact diagonalization, analytical strong-coupling theories and resonating valence bond approaches. In one limit, the lattices effectively become bipartite, while at the opposite limit heavily frustrated nets emerge. In between, competing tunneling processes result in short-ranged spin correlations, the presence of a manifold of low-lying singlets and the stabilization of valence bond crystal and spin-nematic phases.

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Introduction – Highly frustrated antiferromagnets (AFMs) realize exotic collective states of matter in both experiment [1–5] and theory, ranging from valence bond crystals [6–13] to chiral [14, 15] and $Z_2$ topological spin liquids carrying fractionalized excitations [5, 16–19]. A long-established and very powerful theoretical starting point to capture such magnetically disordered quantum phases is the short-range resonating valence bond basis [20–25]. Its physical relevance is particularly clear in spin $S = 1/2$ Heisenberg AFMs that are built from triangular units. Pairing any two spins of a triangle into a valence bond (VB) – a quantum-mechanical singlet – minimizes the local energy. An essential point is that on lattices with corner-sharing triangles, such as for instance the kagome lattice, the VB covering is geometrically frustrated: a finite fraction of triangles, the defect triangles, must do without a singlet [26]. This frustration is due to the presence of closed loops, for instance the loop around a hexagon of the kagome lattice, cf. Fig. 1(a). The ensuing defect triangles are the source of non-trivial VB dynamics. They cause the coherent evolution from one dimer covering into another while the total number of VBs present on the lattice stays the same.

As initially shown by Zeng and Elser [27], the low-energy VB dynamics of the $S = 1/2$ kagome magnet is governed by tunneling events of defect triangles across the hexagon loops [12, 18, 28], while the actual ground state is extremely sensitive to the competition between different such tunneling processes [12]. This calls for a deeper study of the kind of exotic phases which arise from controlling and tuning the most relevant local tunneling amplitudes, while preserving the basic two-dimensional (2D) corner-sharing-triangle layout of the lattice.

In this Letter we consider the simplest generalizations to kagome-type lattices that naturally accommodate loops of different lengths, which in turn introduce another degree of tunneling competition. The three 2D triangle-based lattices that we consider are shown in Fig. 1(b–d). Each of the lattices features two sets of inequivalent sites, labelled A and B. On all lattices the A-sites are four-fold coordinated ($z_A = 4$) as sites on the kagome. The B-sites, however, have a coordination $z_B ≥ 4$. The inequivalent sites cause the presence of two types of short resonance loops: whereas the kagome lattice has one minimal loop of length six (denoted as L6 from here on), we now have loop lengths of L6-L4 (sorrel net [29]), L4-L4 and L4-L8 (squa-kagome net [30–34]).

The resulting $S = \frac{1}{2}$ AFM Heisenberg Hamiltonian $\mathcal{H} = \sum_{\langle ij \rangle} J_{ij} S_i \cdot S_j$ is characterized by two inequivalent nearest neighbor (NN) exchange parameters $J_{AA}$ and $J_{AB}$, their ratio $x = J_{AB}/J_{AA}$ setting the “coupling strength” of the model. We establish the phase diagrams of these Hamiltonians using a combination of exact diagonalization (ED), analytical strong-coupling theories, and resonating valence bond (RVB) approaches based on quantum dimer model (QDM) derivations. Away from intermediate coupling ($1 \lesssim x \lesssim 2$), we find a number of generic features, including Lieb ferrimagnetism, fractional magnetization plateaux, exactly and almost exactly localized modes, as well as physics governed by an effectively frustrating NN and next-NN exchange ($J_1$ and $J_2$, respectively), with $J_2 \gg J_1$. At intermediate coupling, strong frustration leads to short-ranged spin correlations and a manifold of low-lying singlets, some of which correspond to localized bound states of triplets.

For the L6-L4 net, the ED results reveal the presence of a VB crystal state which breaks the rotational symmetry, in agreement with the prediction from the corresponding QDM. In the two other lattices we find evidence for spin nematic physics. The behavior in the different coupling regimes are presented in detail in turn below.

Bi-partite regime – For $x \gg 1$ all three lattices become effectively bi-partite, leading to semiclassical collinear Néel states with, say, the A-spins pointing up and the B-spins down. Since $N_A \neq N_B$, these are Lieb ferrimagnets [35] with total moments $M \approx 1/2$, $3/5$, and $1/3$ for L6-L4, L4-L4, and L4-L8 respectively. Classically the onset of this phase is $x_{\text{ferri}} = 2$, while our ED results show...
a slightly smaller $x_{\text{ferr}}$ in all lattices, reflecting a
general preference of quantum fluctuations for collinear spin
arrangements. In addition, in presence of a magnetic
field $B$, these phases survive in a large $x-B$ region of the
phase diagram, see Fig. 2. Quantum fluctuations reduce
the local spin lengths, but in such a way that the total
moment $M$ is conserved. This means that the deviation
d$s$ of the spin lengths from their classical $s = \frac{1}{2}$ value
satisfies the “conservation law” $dS_B/dS_A = N_A/N_B$ \[?\].
Our ED results give $(S_A,S_B) \approx (0.4545,0.3635)$,
(0.472049,0.388196), and (0.4084,0.3168) for L6-L4 (32
sites), L4-L4 (20 sites), and L4-L8 (30 sites) respectively.

**Strong-coupling regime** – The opposite limit, $x \ll 1$, is
much richer. Here the A-spins form isolated compound
objects (hexamers for L6-L4, tetramers for L4-L4 and L4-
L8) with a total spin $S_{AA} = 0$, while the B-spins are free
to point up or down, defining a highly degenerate mani-
fold. An infinitesimal $J_{AB}$ mediates an effective coupling
between the B-spins through the virtual excitations of the
A-spins out of their $S_{AA} = 0$ state. Degenerate pertur-
bration theory shows that the leading exchange between
the B-spins is not a NN exchange $J_1$ but a next-NN coupling $J_2$,
which in conjunction with the underlying topology of the B-spins,
gives rise to rich frustration effects. This is related to a destructive interference mechanism
which cancels out exactly (L4-L4 and L4-L8) or
almost exactly (L6-L4) the leading contribution to $J_1$. A
very similar mechanism appears in the strong dimer
limit of the Cairo pentagonal AFM \[37\]. Here this mechanism
is too make explained in the nearest neighbor VB (NNVB) picture of the $S_{AA} = 0$ objects. For the L6-
L4 net, one obtains a $J_2$-$J_3$-$J_4$ model on the honeycomb
lattice \[38-41\] with $J_2 = 0.115037x^2 + 0.10576478x^3 +
O(x^4), J_3 = 0.0384319x^2 + 0.03071212x^3 + O(x^4), J_4 = 2 \left(-0.019216x^2 + 0.0553166527x^3\right) + O(x^4)$. The relative
strengths of these couplings suggests that the GS lies in the
spiral “phase-III” region of Refs. \[38, 39\].

The L4-L4 and L4-L8 nets share the same $S_{AA} = 0$
plaquette, and so we expect very similar couplings.
Indeed, an expansion up to third order in $x$ gives an effective
$J_2$-$J_1$ model on the square (L4-L4) or the checker-
board (L4-L8) lattice with $J_2 = \frac{1}{2}x^2 + \frac{1}{4}x^3 + O(x^4), J_1 = f \frac{1}{2}x^3 + O(x^4)$, where $f = 2$ (L4-L4) or 1 (L4-L8).
For the L4-L4 net, the finite $J_1$ appearing in third order
stabilizes the collinear striped phase among the one-
parameter family of states favored by $J_2$ alone \[42\], while
in L4-L8 the B-sites form a crossed-dimer VBC \[43-45\]
(in Fig. 2 this state is denoted by $p$-CD-VBC, where $p$
stands for the plaquette structure of the A-spins). Pushing
the expansion up to fourth-order gives a four-spin exchange
interaction term around each AA-plaquette, similar to
the case of the Cairo pentagonal lattice in the strong dimer
limit \[37\]. In the notation of Eq. (27) of Ref. \[37\], the
fourth-order amplitudes of these “plaquette” terms are
\[K_h = K_v = -\frac{7}{4}x^4, K_x = +\frac{3}{4}x^4\]
Since both $K_h$
and $K_v$ are negative, they also favor a collinear phase
and thus one expects this phase to survive as long as $J_2$
remains the dominant coupling.

We now consider the systems at strong coupling in
the presence of a magnetic field $B$. Given the pertur-

ative scale of their effective couplings, the B-spins
will be quickly polarized by the field, and so these magnets
show successive plateaux at the level-crossing fields of the
AA-hexamers (for L6-L4 ) or tetramers (for L4-L4
and L4-L8) from $S_{AA} = 0$ up to $L_{AA}/2$, see Fig. 2. At
saturation, there are exact magnetization jumps which are
related to the existence of localized magnons on the
AA-loops. Such magnons are generic for corner-sharing-
triangle lattices \[33, 34, 46-48\]. Here they are present
for all $x$, but become the lowest excitations above the
fully polarized state only below a characteristic value of
\[x_c = \frac{3}{5} \text{ for L6-L4, } x_c = \frac{2}{5} \text{ for L4-L4, and } x_c = 1 \text{ for L4-L8, see Fig. 2. Besides the exact jumps at saturation, Fig. 2}
shows almost exact jumps for the transitions between the

remaining plateaux. This feature stems from the corresponding excitations being large objects (an AA-hexamer or AA-tetramer) that therefore have very small tunneling amplitudes and that are thus very localized.

Intermediate coupling – This is the most challenging regime as correlations are expected to be strong as in the kagome [49–51]. Indeed, our ED data show that the spin-spin correlations become short-ranged. We also observe an accumulation of many low-lying singlets below the lowest magnetic excitations around \( x \approx 1 \) (see Fig. 3(a) for the 32-site L6-L4 case). Specifically, we find 49, 15, and 27 singlets respectively for the 32-site L6-L4, 30-site L4-L4, and 30-site L4-L8 net (the latter is also discussed in [33]). From a strong coupling perspective, many of these singlets can in fact be understood as bound states of two triplets, one residing on AA-plaquettes and the other on their surrounding BB-plaquettes (hexagon for L6-L4, square for L4-L4 and L4-L8). The AA-triplet costs a lot of energy at \( x = 0 \) but the binding energy with a BB-triplet scales linearly with \( x \) as can be directly seen in the ED spectra and can be readily found by first-order perturbation theory and equivalent operators. Importantly, as discussed above the AA-triplets are almost localized and the same is expected for the bound singlets. One therefore expects an extensive number of low-energy singlets in the intermediate region [7].

Now, one or more of the low energy singlets approach quickly and remain close to (or even cross) the GS in a wide region above \( x = 1 \), which is evidence for phase transitions. Examining the GS level-crossings and the symmetry of the low-lying states suggests one intermediate phase in L6-L4 and L4-L4, and at least two intermediate phases in L4-L8, see Fig. 2. To determine the nature of the new phase(s) we turn to the NNVB basis.

We first check that the asymmetric distribution of the GS energy on the two types of bonds agrees qualitatively with the corresponding distribution over the NNVB basis states without resonances, see Table I. To include the effect of resonances we proceed in analogy with the kagome system [12, 27, 28], and cast the problem in terms of tunneling events of defect triangles across the two resonance loops. The resonances around a hexagon at \( x = 1 \) are known from the kagome [12, 27, 28], but need to be re-evaluated for \( x \neq 1 \). The processes around a square involve co-tunneling of zero, one, or two defect triangles, while on an octagon they involve up to four defect triangles. We have evaluated all resonance amplitudes using a method that essentially corresponds to the infinite order overlap expansion of [12]. The central result is that the amplitudes are strongest for the smallest square loop, particularly so for the so-called “perfect resonance” process. Therefore one can find good candidate states by requiring that the defect triangles resonate around the smallest loops. For the L6-L4 net this is achieved by the translationally invariant VBC state of Fig. 3(b) which breaks the six-fold rotational symmetry down to two-fold. Inspecting the 32-site ED data of Fig. 3(a), we find three translationally invariant singlets with angular momenta \( l = 0 \) and \( l = \pm 2\pi/3 \), which remain very close in energy in a wide region above \( x = 1 \). In conjunction with the GS connected dimer-dimer correlations shown in Fig. 3(c), they are strong evidence that the VBC state of Fig. 3(b) is indeed stabilized in the thermodynamic limit.

For the L4-L4 and L4-L8 nets, maximizing the number of “perfect squares” leads to the VBC states of Fig. 4. Comparing with the strong coupling limit, here a fraction of AA-singlets has turned into “square pinwheels”, that are analogous to the “hexagonal pinwheels” of the 36-site VBC candidate of the kagome [6–8, 12]. An independent ED study [52] of an isolated 8-site pinwheel cluster reveals that the “square pinwheels” are combinations of the above local bound singlets, which provides an alternative and intuitive picture for their condensation. The VBC state of L4-L8 (Fig. 4(b)) has been found pre-
in our largest clusters available (isolated square pinwheel cluster confirms this qualitative
finite negative (positive) value. Again, an ED study of the
L4-L8, see Fig. 5. The two VBC states maximizing the number of “perfect resonance”
processes on the AA-squares of L4-L4 and L4-L8. These processes favor $S_{AA} = 0$
singlets, indicated by the alternating solid (blue) and dashed (red) dimers. The
two-colored octagonal dimer loops represent the two “square pinwheels” that
dominantly minimize the energy exactly at $x = 1$. For $x > 1$ ($x < 1$) the two-fold
Ising degeneracy is lifted, selecting the even (odd) resonance of the two pinwheel states.

![FIG. 3. ED results for the 32-site L6-L4 cluster enclosed by the (green) parallelogram in (c), with periodic boundary
conditions. (a) Low-energy spectra as a function of $x$, labeled by linear and angular momenta quantum numbers. (b) The
VBC state that can be compared with (c) GS expectation values of the connected dimer-dimer correlations at $x = 1.2$
and sector “$2 \pi /3.Sxe$”. The empty rectangle denotes the reference bond.

naturally in Ref. [30] using a large-N theory. A qualitative
difference is that the Ising degeneracy associated with the
“square pinwheels” is lifted as soon as $x > 1$ ($< 1$), since
then the corresponding tunneling amplitude attains a fi-
nite negative (positive) value. Again, an ED study of the
isolated square pinwheel cluster confirms this qualitative
change at $x = 1$.

On the ED side, the situation in L4-L4 and L4-L8 is not
totally exclusive due to the small number of unit cells
in our largest clusters available ($N = 20, 30$ in L4-L4, and
$N = 24, 30$ in L4-L8). As mentioned above, the current
evidence points to one intermediate phase (phase X) in
L4-L4 and at least two intermediate phases (Ya and Yb)
in L4-L8, see Fig. 2. In particular, the ED data suggest
that phase X is not the pinwheel VBC state of Fig. 4(a),
but a spin nematic phase with d-wave symmetry.

![FIG. 4. The two VBC states maximizing the number of “per-
fected resonance” processes on the AA-squares of L4-L4 and
L4-L8. These processes favor $S_{AA} = 0$ singlets, indicated by
the alternating solid (blue) and dashed (red) dimers. The
two-colored octagonal dimer loops represent the two “square pinwheels” that
dominantly minimize the energy exactly at $x = 1$. For $x > 1$ ($x < 1$) the two-fold
Ising degeneracy is lifted, selecting the even (odd) resonance of the two pinwheel states.

![TABLE I. Basic elements of NN VB basis for each of the three
lattices. $D_{NNVB}$: Pauling estimate for the dimension of the
NN VB basis using a generalized arrow representation[53] with
two types of arrows[52], $n_{def-tr}$: percentage of defect triangles
(fixed for all NN VB states), $e_{AA}$ and $e_{AB}$: the GS expectation
values of $(S \cdot S)$ on AA and AB bonds, $e^{(0)}_{AA}$ and $e^{(0)}_{AA}$:
corresponding average energies over the NN VB basis states.

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