

Frustrated kagome-lattice bilayer quantum Heisenberg antiferromagnet

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We consider the $S = 1/2$ antiferromagnetic Heisenberg model on a frustrated kagome-lattice bilayer with strong nearest-neighbor interlayer coupling and examine its low-temperature magnetothermodynamics using a mapping onto a rhombi gas on the kagome lattice. Besides, we use finite-size numerics to illustrate the validity of the classical lattice-gas description. Among our findings there are (i) the absence of an order-disorder phase transition and (ii) the sensitivity of the specific heat at low temperatures to the shape of the system just below the saturation magnetic field even in the thermodynamic limit.

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I. FRUSTRATED BILAYER HEISENBERG ANTIFERROMAGNETS

Two spins one-half entangled in a singlet state are at the heart of quantum mechanics. In the theory of spin lattices, singlets emerging due to the antiferromagnetic Heisenberg interaction lead to various fascinating valence-bond solid or valence-bond liquid quantum states. The resonating valence-bond state proposed by P. W. Anderson as the ground state of spin liquids is a famous example of the latter kind of states [1]. Valence-bond solid states, on the other hand, imply a short-range pairing and the formation of localized static bonds obeying certain long-range order. On some lattices the antiferromagnetic Heisenberg interaction promotes the emergence of valence-bond crystal states as, for instance, in the Majumdar-Ghosh model [2,3]. Among such lattices one may also mention frustrated bilayer lattices which have been dealt with in a number of earlier publications, see Refs. [4–18].

Frustrated bilayer systems consist of two identical layers, a and b , with an intralayer interaction J_1 , a nearest-neighbor interlayer interaction J_2 , and a (frustrated) next-nearest-neighbor interlayer interaction J_x , compare Fig. 1 as an example. If $J_1 = J_x = J$, then one faces the fully frustrated case. For the fully frustrated case, a local singlet on a J_2 bond ($|\uparrow_a \downarrow_b\rangle - |\downarrow_b \uparrow_a\rangle)/\sqrt{2}$ in the environment of the remaining

polarized spins is an eigenstate of the Hamiltonian. Moreover, a product of such singlets surrounded by the polarized spins is also an eigenstate of the Hamiltonian. Furthermore, if $J_2 > J_{2c}(J)$ for some critical $J_{2c}(J)$, then these states are the lowest-energy eigenstates and therefore may dominate the low-temperature magnetothermodynamics of the frustrated quantum spin systems in question.

Since the localized eigenstates for the frustrated square-, honeycomb-, and triangular-lattice bilayers can be mapped onto spatial configurations of squares or hexagons on the square or hexagonal/triangular lattice, their contribution to thermodynamics is accounted for by means of classical statistical mechanics [7,13,16]. The most interesting prediction for these bilayers is an order-disorder phase transition related to singlet ordering: Just below the saturation magnetic field h_{sat} the ground state is the twofold (square and honeycomb bilayers) or threefold (triangular bilayer) degenerate gapped localized-singlet crystal state, which corresponds to an ordered pattern of singlets respecting hard-core rules. Such a (spontaneously chosen) ordered pattern persists up to some finite temperature $T_c(h)$ while further temperature increase drives the system into a disordered phase through a phase transition of Ising (square and honeycomb bilayers) or three-state Potts (triangular bilayer) universality class [7,13,16].

Similarly, the localized eigenstates for the frustrated kagome-lattice bilayer can be mapped onto rhombi on the kagome lattice. However, since the kagome lattice can be covered by hard rhombi (nearest-neighbor exclusion) in a huge number of ways which grows exponentially with lattice size, one cannot expect an order-disorder phase transition pertaining to singlet ordering. This difference between the already studied frustrated bilayers and the not yet examined frustrated kagome-lattice bilayer inspired us to take a closer look at the latter in this publication.

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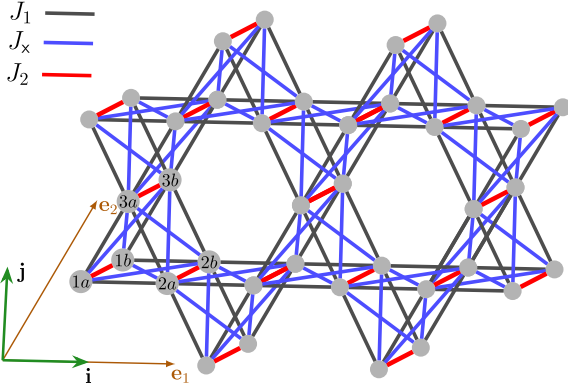


FIG. 1. Frustrated kagome-lattice bilayer. The lattice sites are denoted by $\mathbf{m}\alpha l$, where $\mathbf{m} = m_1\mathbf{e}_1 + m_2\mathbf{e}_2$, m_1, m_2 are integers, $\mathbf{e}_1 = 2\mathbf{i} = (2, 0)$, $\mathbf{e}_2 = \mathbf{i} + \sqrt{3}\mathbf{j} = (1, \sqrt{3})$ are the unit lattice vectors (here the triangle side has unit length), $\alpha = 1, 2, 3$ denote the sites of the unit cell within a layer, and $l = a, b$ is the layer label.

In the present paper, we consider the $S = 1/2$ isotropic Heisenberg Hamiltonian,

$$H = \sum_{\langle pq \rangle} J_{pq} \mathbf{s}_p \cdot \mathbf{s}_q - h \sum_p s_p^z, \quad (1)$$

where the first (second) sum runs over the edges (vertices) of the frustrated kagome-lattice bilayer shown in Fig. 1, all exchange interactions are antiferromagnetic $J_{pq} > 0$, and $h \geq 0$ is an external magnetic field. The total $S^z = \sum_p s_p^z$ commutes with H and the subspaces with different good quantum numbers S^z can be considered separately. As said above, we distinguish the intralayer interactions J_1 , the nearest-neighbor interlayer interactions J_2 , and the next-nearest-neighbor interlayer interactions J_x (cf. black, red, and blue edges in Fig. 1). We set $(J_1 + J_x)/2 = J = 1$ to fix the units. The goal of the present study is to examine thoroughly the ground-state and finite-temperature properties of the frustrated kagome-lattice bilayer quantum Heisenberg antiferromagnet following the treatment of Refs. [7, 13, 16].

The remainder of this paper is organized as follows. We begin with numerics for finite-size systems, Sec. II, focusing on the fully frustrated case $J_1 = J_x$. Then we turn to analytical studies in Sec. III. We discuss one-magnon and many-magnon eigenstates as well as a mapping onto classical lattice models and calculate the low-temperature magnetothermodynamics of the frustrated kagome bilayer. In Sec. IV, we present an effective low-energy theory for a slightly violated fully frustrated condition $J_1 \neq J_x$. Finally, we summarize our findings in Sec. V.

II. NUMERICAL CALCULATIONS FOR SMALL FINITE-SIZE LATTICES

In our numerical calculations, we consider several finite-size frustrated kagome-lattice bilayers, in particular, with $N = 36$ and $N = 42$ (i.e., with the number of sites in one layer $\mathcal{N} = 18$ and $\mathcal{N} = 21$). Periodic boundary conditions are imposed. Two shapes of the cluster with $N = 36$ are determined by the edge vectors $\mathbf{a}_1 = (6, 0) = 3\mathbf{e}_1$, $\mathbf{a}_2 = (2, 2\sqrt{3}) = 2\mathbf{e}_2$

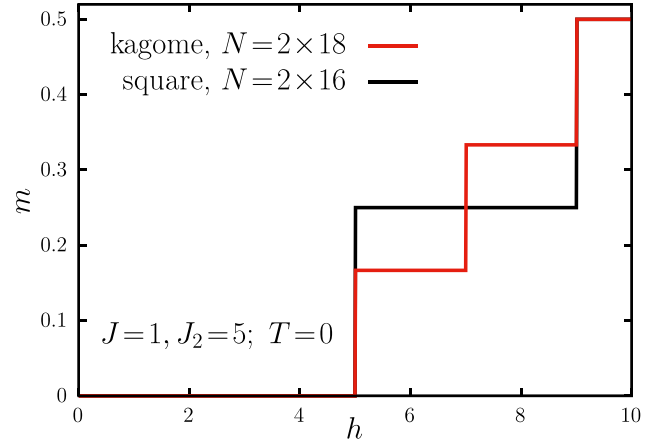


FIG. 2. Zero-temperature magnetization curve for the $S = 1/2$ fully frustrated kagome-lattice bilayer with $J = 1$, $J_2 = 5$, $N = 36$ (red curve). Black curve corresponds to the square-lattice counterpart, $N = 32$. There are no finite-size effects for this set of parameters.

($N = 36_a$) and $\mathbf{a}_1 = (6, 0)$, $\mathbf{a}_2 = (0, 2\sqrt{3})$ ($N = 36_b$); see Appendix A. The cluster with $N = 42$ is determined by the edge vectors $\mathbf{a}_1 = (7, -\sqrt{3})$, $\mathbf{a}_2 = (0, 2\sqrt{3})$. For ideal flat-band geometry $J_1 = J_x = 1$ we examine several $J_2 = 1, 2, 3, 5$; see Secs. II and III and Appendix A. Small deviations from the flat band geometry are represented by the set $J_1 = 1.1$, $J_x = 0.9$, $J_2 = 5$; see Sec. IV. We use full diagonalization for large S^z or the finite-temperature Lanczos method [19, 20] to calculate various ground-state characteristics (energy and degeneracy of low-lying levels, magnetization curve) and temperature dependencies of magnetization, susceptibility, entropy, and specific heat. Finite-size calculations play a role of “experiments,” which are then explained by characterizing the low-energy eigenstates and mapping onto lattice gases.

The typical ground-state magnetization curve for the large- J_2 regime shown in Fig. 2 as red curve has a 0-, a 1/3-, and a 2/3-magnetization plateau for $0 < h < h_1$, $h_1 < h < h_2$, and $h_2 < h < h_{\text{sat}}$, respectively. Here $h_1 = J_2$, $h_2 = J_2 + 2J$, and $h_{\text{sat}} = J_2 + 4J$. The plateau states are associated with the respective number of singlets in the unit cell. For example, the 2/3 plateau corresponds to one singlet and two polarized triplets, whereas the 1/3 plateau corresponds to two singlets and one polarized triplet within each unit cell. Interestingly, both these plateau states have a large degeneracy: For $S^z/(N/2) = 2/3$, $\mathcal{W}_{\text{GS}} = 17, 20, 31$, for $N = 36_a, 36_b, 42$, respectively. The ground-state magnetization curve for the square-lattice bilayer, black curve in Fig. 2, exhibits only a 0 and a 1/2 plateau for $0 < h < h_1$ and $h_1 < h < h_{\text{sat}}$. The former state corresponds to a crystal of singlets at all J_2 bonds. The latter state corresponds to a crystal state with checkerboard order of singlets which alternate with polarized triplets; such a state is only twofold degenerate, see Refs. [5, 7].

More numerical results for the ground states and excitations are reported in Appendix A. Most importantly, the data for low-energy eigenstates in the subspaces with $S^z = N/2 - 1, \dots, N/3$ clearly illustrate the emergence of the large- J_2 regime: For $J_2 > 2J$, the low-energy eigenstates have easily

understandable energy and degeneracy, see Sec. III below. Moreover, the ground-state degeneracy $\mathcal{W}_{\text{GS}}$ does not depend on J_2 , and $\mathcal{W}_{\text{GS}}$ for the $2/3$ plateau state is rather large and grows with increasing N (17, 20 or 31 for $N = 36_a, 36_b$ or $N = 42$). For $J_2 < 2J$ the ground state and the first excited state are of completely different nature without regularly varying energies and large degeneracies.

Numerical results for finite temperatures are reported in Sec. III. In particular, we present data for the temperature dependence of magnetization, susceptibility, entropy, and specific heat. Moreover, we present some of such data at zero and finite temperatures for the case $J_1 \neq J_x$ in Sec. IV.

III. ANALYTICAL AND COMPUTER-AIDED CALCULATIONS

A. Flat bands and localized states

We begin this section with the one-magnon spectrum of the system at hand to show that this lattice supports completely dispersionless (flat) magnon bands. To this end, we proceed as follows. First, set for a while $h = 0$ in Eq. (1) and represent the Hamiltonian in the form: $H = \sum_{l=a,b} H_l + H_{ab}$, where H_l is the kagome-layer Hamiltonian and H_{ab} denotes the interlayer coupling. Second, for the one-magnon spectrum calculations, we have to replace $s_p \cdot s_q \rightarrow (s_p^+ s_q^- + s_q^+ s_p^-)/2 - s_p^+ s_p^-/2 - s_q^+ s_q^-/2 + 1/4$ (here the term $s_p^+ s_p^- s_q^+ s_q^-$ is omitted as irrelevant one). Third, as usual, to convert from the $\mathbf{m}$ -space to the $\mathbf{k}$ -space, we introduce

$$\begin{aligned} s_{\mathbf{k}\alpha l}^\pm &= \frac{1}{\sqrt{\mathcal{N}}} \sum_{\mathbf{m}} e^{\mp i\mathbf{k} \cdot \mathbf{m}} s_{\mathbf{m}\alpha l}^\pm, \\ s_{\mathbf{m}\alpha l}^\pm &= \frac{1}{\sqrt{\mathcal{N}}} \sum_{\mathbf{k}} e^{\pm i\mathbf{k} \cdot \mathbf{m}} s_{\mathbf{k}\alpha l}^\pm, \end{aligned} \quad (2)$$

where $\mathbf{k} = (k_1, k_2)$ acquires $\mathcal{N}/3$ values: $k_{1,2} = 2\pi z_{1,2}/\mathcal{L}_{1,2}$, $z_{1,2}$ obtains $\mathcal{L}_{1,2}$ integer values, $\mathcal{L}_1 \mathcal{L}_2 = \mathcal{N}$, $\mathcal{N} = N/2$, and $k_1 = k_x$, $k_2 = (k_x - \sqrt{3}k_y)/2$. Fourth, it is convenient to use matrix notations. As a result, the Hamiltonian (1) in one-magnon space can be cast into

$$\begin{aligned} H_{\text{lm}} &= \left(J_1 + J_x + \frac{J_2}{4} \right) \mathcal{N} \\ &+ \sum_{\mathbf{k}} \begin{pmatrix} s_{\mathbf{k}1a}^- & s_{\mathbf{k}2a}^- & s_{\mathbf{k}3a}^- & s_{\mathbf{k}1b}^- & s_{\mathbf{k}2b}^- & s_{\mathbf{k}3b}^- \end{pmatrix} \mathbf{H} \begin{pmatrix} s_{\mathbf{k}1a}^+ \\ s_{\mathbf{k}2a}^+ \\ s_{\mathbf{k}3a}^+ \\ s_{\mathbf{k}1b}^+ \\ s_{\mathbf{k}2b}^+ \\ s_{\mathbf{k}3b}^+ \end{pmatrix}, \end{aligned} \quad (3)$$

where the first term is the ferromagnetic-state energy, and

$$\begin{aligned} \mathbf{H} &= \begin{pmatrix} \mathbf{A} & \mathbf{B} \\ \mathbf{B} & \mathbf{A} \end{pmatrix}, \\ \mathbf{A} &= -\left(2J_1 + 2J_x + \frac{J_2}{2} \right) \mathbf{1} + J_1 \mathbf{K}, \quad \mathbf{B} = \frac{J_2}{2} \mathbf{1} + J_x \mathbf{K}, \\ \mathbf{K} &= \begin{pmatrix} 0 & \frac{1+e^{-ik_1}}{2} & \frac{1+e^{-ik_2}}{2} \\ \frac{1+e^{ik_1}}{2} & 0 & \frac{1+e^{i(k_1-k_2)}}{2} \\ \frac{1+e^{ik_2}}{2} & \frac{1+e^{-i(k_1-k_2)}}{2} & 0 \end{pmatrix}. \end{aligned} \quad (4)$$

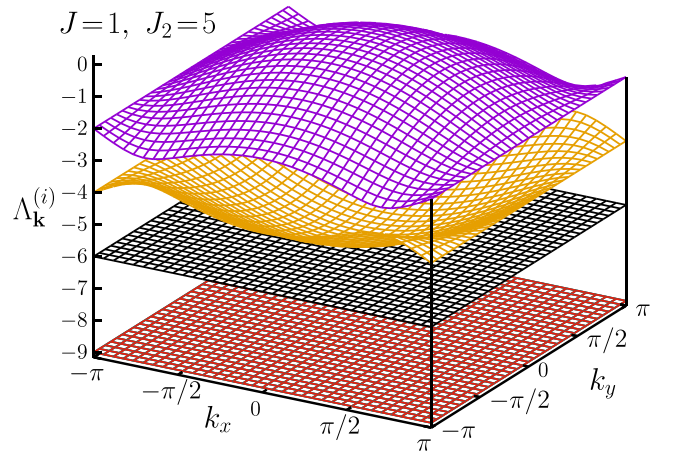


FIG. 3. One-magnon spectrum $\Lambda_{\mathbf{k}}^{(1)}, \dots, \Lambda_{\mathbf{k}}^{(6)}$ given by Eq. (5) with $J_1 = J_x = J = 1$, $J_2 = 5$. The lowest-energy flat band is three-fold degenerate; setting $J_1 = 1.1 \neq J_x = 0.9$ lifts the degeneracy and makes two of the three lowest-energy bands slightly dispersive, see Eq. (5).

The eigenvalues of matrix $\mathbf{K}$ (4) are as follows: $\kappa_1 = -1$, $\kappa_{2,3} = (1 \mp \sqrt{3 + 2\gamma_{\mathbf{k}}})/2$, where $\gamma_{\mathbf{k}} = \cos k_1 + \cos k_2 + \cos(k_1 - k_2)$. Now we may use the formulas for determinants of block matrices [21,22]. Since $\mathbf{A}$ and $\mathbf{B}$ commute, the sought eigenvalues of $\mathbf{H}$ follow from the equations: $\det(\mathbf{A} + \mathbf{B} - \Lambda_{\mathbf{k}}^{(i)} \mathbf{1}) = 0$, $i = 1, 2, 3$ and $\det(\mathbf{A} - \mathbf{B} - \Lambda_{\mathbf{k}}^{(i)} \mathbf{1}) = 0$, $i = 4, 5, 6$. As a result,

$$\begin{aligned} \Lambda_{\mathbf{k}}^{(1)} &= -3(J_1 + J_x), \\ \Lambda_{\mathbf{k}}^{(2,3)} &= -\frac{J_1 + J_x}{2} (3 \pm \sqrt{3 + 2\gamma_{\mathbf{k}}}), \\ \Lambda_{\mathbf{k}}^{(4)} &= -(3J_1 + J_x + J_2), \\ \Lambda_{\mathbf{k}}^{(5,6)} &= -\left(\frac{3J_1 + 5J_x}{2} + J_2 \right) \mp \frac{J_1 - J_x}{2} \sqrt{3 + 2\gamma_{\mathbf{k}}}. \end{aligned} \quad (5)$$

In the presence of a magnetic field one has to add $-h\mathcal{N}$ to the ferromagnetic-state energy and replace $\Lambda_{\mathbf{k}}^{(i)}$ by $\Lambda_{\mathbf{k}}^{(i)} + h$. Note that $\Lambda_{\mathbf{k}}^{(1,2,3)}$ depend on $J_1 + J_x = 2J$ only and do not depend on J_2 . For the fully frustrated case $J_1 = J_x = J$, there are four flat bands: $\Lambda_{\mathbf{k}}^{(1)} = -6J$ and $\Lambda_{\mathbf{k}}^{(4,5,6)} = -(4J + J_2)$. Furthermore, $\Lambda_{\mathbf{k}}^{(2,3)} = -J(3 \pm \sqrt{3 + 2\gamma_{\mathbf{k}}})$ remain dispersive and $\Lambda_{\mathbf{k}=0}^{(2)}$ touches the flat band $\Lambda_{\mathbf{k}}^{(1)}$. The one-magnon spectrum is illustrated in Fig. 3. Evidently, Eq. (5) is in perfect correspondence with the numerical results for $S^z = 17$ ($N = 36$); see Appendix A.

By inspection, the flat-band states with the energy $\Lambda_{\mathbf{k}}^{(1)}$ have the form

$$\propto [(e^{ik_2} - e^{ik_1})t_{\mathbf{k}1}^- + (1 - e^{ik_2})t_{\mathbf{k}2}^- + (e^{ik_1} - 1)t_{\mathbf{k}3}^-] |\uparrow \dots \uparrow\rangle \quad (6)$$

with $t_{\mathbf{k}\alpha}^- = s_{\mathbf{k}\alpha a}^- + s_{\mathbf{k}\alpha b}^-$. In the $\mathbf{m}$ -space, these are the hard-hexagon states [23–27] associated with the two nearest hexagons from different layers. Furthermore, the flat-band states with the energies $\Lambda_{\mathbf{k}}^{(4,5,6)}$ to be denoted further by $\epsilon_0 = -(4J + J_2)$ are the singlets localized on the J_2 bonds,

$$\frac{1}{\sqrt{2}} (|\uparrow_{\mathbf{m}aa}\rangle |\downarrow_{\mathbf{m}ab}\rangle - |\downarrow_{\mathbf{m}aa}\rangle |\uparrow_{\mathbf{m}ab}\rangle), \quad (7)$$

in the environment of all other fully polarized sites. In the present study, we are interested in the case $J_2 > 2J$ (the large- J_2 regime), when the localized singlets, i.e., the localized magnons from the flat bands $\Lambda_{\mathbf{k}}^{(4,5,6)}$, are the lowest-energy one-magnon states. Importantly, the local nature of the one-magnon flat-band states paves the way to the construction of many-magnon ground states in the subspaces with $\mathcal{N}/3 \leq S^z < \mathcal{N} - 1$.

Another route to determine and characterize the eigenstates of the frustrated bilayer Heisenberg Hamiltonian is to introduce the total spin operator on the J_2 bonds, i.e., $\mathbf{t}_{\mathbf{m}\alpha} = \mathbf{s}_{\mathbf{m}\alpha a} + \mathbf{s}_{\mathbf{m}\alpha b}$ [28]. Moreover, we also introduce another spin operator on the J_2 bonds: $\mathbf{d}_{\mathbf{m}\alpha} = \mathbf{s}_{\mathbf{m}\alpha a} - \mathbf{s}_{\mathbf{m}\alpha b}$. One can easily convince oneself that the Hamiltonian (1) in terms of these operators becomes as follows:

$$H = \sum_{\mathbf{m}\alpha} \left[-ht_{\mathbf{m}\alpha}^z + \frac{J_2}{2} \left(t_{\mathbf{m}\alpha}^2 - \frac{3}{2} \right) \right] + \sum_{(\mathbf{m}\alpha, \mathbf{n}\beta)} \left(\frac{J_1 + J_x}{2} \mathbf{t}_{\mathbf{m}\alpha} \cdot \mathbf{t}_{\mathbf{n}\beta} + \frac{J_1 - J_x}{2} \mathbf{d}_{\mathbf{m}\alpha} \cdot \mathbf{d}_{\mathbf{n}\beta} \right). \quad (8)$$

For the fully frustrated case $J_1 = J_x = J$, the last term in Eq. (8) drops out and the Hamiltonian depends only on the total spin operators on the J_2 bonds $\mathbf{t}_{\mathbf{m}\alpha}$. Since $\mathbf{t}_{\mathbf{m}\alpha}^2$ commutes with the Hamiltonian for all $\mathbf{m}\alpha$ (local integrals of motion), the Hamiltonian eigenstates can be labeled by the set of $\mathcal{N}$ good quantum numbers $\mathbf{t}^2 = t(t+1)$ assigned to each J_2 bond. Obviously, t may acquire only two values, $t = 0$ (singlet) or $t = 1$ (triplet). In the latter case, when $t = 1$ for all $\mathbf{m}\alpha$, one arrives at the $S = 1$ kagome-lattice Heisenberg antiferromagnet, which is among the reference models of frustrated quantum magnetism. This model represents the low-energy physics of the $S = 1/2$ kagome-lattice bilayer for the ferromagnetic interlayer interaction $J_2 < 0$. In the present study, however, we focus on the case of large enough antiferromagnetic interlayer interaction $J_2 > 2J > 0$ and $h \geq 0$. Then localized singlets and (polarized) triplets dominate the low-temperature properties and one arrives at classical statistical mechanics problems.

Consider, for example, the eigenstates with only one $t = 0$ and all others are polarized triplets, cf. Eq. (7). Such eigenstates correspond to the flat-band states with the energy $\epsilon_0 = -(4J + J_2)$. Indeed, each such a state has the energy $E_{\text{FM}} - 4J - J_2$, where $E_{\text{FM}} = (2J + J_2/4)\mathcal{N}$ is the energy of the ferromagnetic state (polarized triplets at all J_2 bonds), cf. the first term in Eq. (3).

Because of the local character of the singlet state, there is a class of many-magnon states, which are located on J_2 bonds sufficiently far from each other. More precisely, this class of states contain $n = 2, \dots, n_{\text{max}}$, $n_{\text{max}} = \mathcal{N}/3$ J_2 bonds with $t = 0$ (the rest $\mathcal{N} - n$ J_2 bonds carry polarized triplets) and satisfy the geometrical restriction: The singlets are not neighbors (hard-core rule), see Fig. 4 (top). Alternatively, such many-magnon states can be viewed as hard-core dimers on an auxiliary honeycomb lattice, see Fig. 4 (top). The energy of hard-rhombi states is $E_{\text{FM}} + n\epsilon_0$. And such states are the ground states in the subspaces with $S^z = \mathcal{N} - 1, \dots, \mathcal{N} - n_{\text{max}}$, $n_{\text{max}} = \mathcal{N}/3$ if $J_2 > 2J$. This picture perfectly

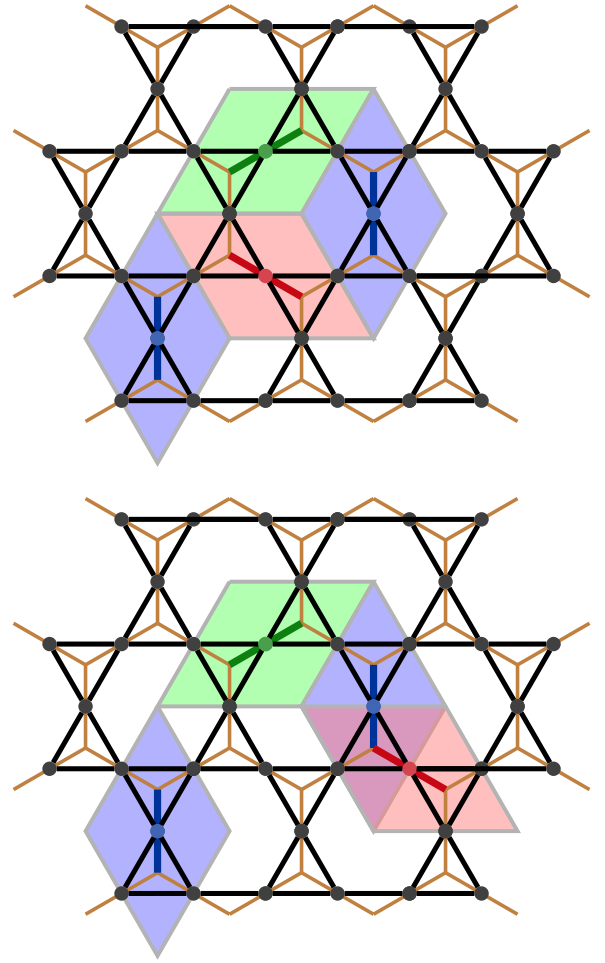


FIG. 4. Top: Pictorial representation of many-magnon ground states for $S^z = \mathcal{N} - 1, \dots, 2\mathcal{N}/3$. Localized singlets (denoted by colored disks) obey hard-rhombi rule on the underlying kagome lattice (thick black) and therefore can be presented as hard-core rhombi. They also can be viewed as hard-core dimers on an auxiliary honeycomb lattice (thin brown). Bottom: Partially overlapping (soft) rhombi represent other localized eigenstates of the fully frustrated Hamiltonian (8).

agrees with numerical data for $S^z = 17, \dots, 12$ ($N = 36$); see Appendix A.

There are two more classes of localized states which are eigenstates of the fully frustrated Hamiltonian (8). That is, (i) the states with $n = 2, \dots, 2n_{\text{max}}$ singlets on the J_2 bonds forming patterns with one (or more) pair(s) which are neighbors [see Fig. 4 (bottom)] and (ii) the states with $n = 3, \dots, 3n_{\text{max}}$ singlets on the J_2 bonds forming patterns with one (or more) triple(s) of singlets which are neighbors. These states can be visualized as containing partially overlapping two rhombi [see Fig. 4 (bottom)] or partially overlapping three rhombi. The energy penalty for each overlap is J .

For sufficiently large J_2/J the three classes of localized eigenstates described above (their total number is $2^{\mathcal{N}}$) are the ground states and low-lying excited states of the model at hand. Again, this picture of overlapping rhombi is confirmed by numerical data for $N = 36$; see Appendix A.

TABLE I. Counting the number of n hard-rhombi spatial configurations on periodic kagome lattices of $\mathcal{N}$ sites. For $\mathcal{N} = 18$ two clusters are determined by the edge vectors $\mathbf{a}_1 = (6, 0)$, $\mathbf{a}_2 = (2, 2\sqrt{3})$ ($\mathcal{N} = 18_a$) and $\mathbf{a}_1 = (6, 0)$, $\mathbf{a}_2 = (0, 2\sqrt{3})$ ($\mathcal{N} = 18_b$). For $\mathcal{N} = 48$ two clusters are determined by the edge vectors $\mathbf{a}_1 = m\mathbf{e}_1$, $\mathbf{a}_2 = n\mathbf{e}_2$, $m = n = 4$ ($\mathcal{N} = 48_a$) and $\mathbf{a}_1 = m\mathbf{e}_1$, $\mathbf{a}_2 = -\mathbf{e}_1 + n\mathbf{e}_2$, $m = n = 4$ ($\mathcal{N} = 48_b$).

$\mathcal{N} = 18$		
n	$g_{18_a}(n), \mathcal{N} = 18_a$	$g_{18_b}(n), \mathcal{N} = 18_b$
1	18	18
2	117	117
3	336	336
4	417	420
5	186	192
6	17	20
$\mathcal{N} = 48$		
n	$g_{48_a}(n), \mathcal{N} = 48_a$	$g_{48_b}(n), \mathcal{N} = 48_b$
1	48	48
2	1032	1032
3	13 136	13 136
4	110 244	110 244
5	643 056	643 056
6	2 677 896	2 677 896
7	8 052 432	8 052 432
8	17 486 550	17 486 558
9	27 158 096	27 158 224
10	29 567 736	29 568 536
11	21 843 888	21 846 384
12	10 417 700	10 421 844
13	2 969 616	2 973 264
14	446 136	447 704
15	27 952	28 208
16	417	417

B. Mapping onto classical lattice gases

The hard-rhombi picture is a good starting point to count the eigenstates described above. In Table I we report the number of spatial configurations of n hard rhombi on the periodic $\mathcal{N}$ -site kagome lattice of two shapes, $g_{\mathcal{N}_a}(n)$ and $g_{\mathcal{N}_b}(n)$. These numbers match perfectly the ground-state degeneracy $\mathcal{W}_{\text{GS}}(S^z)$ for the initial frustrated quantum spin system (1) of $N = 36_a$ sites and $N = 36_b$ sites (see Appendix A). However, for hard rhombi we can examine larger systems, see the results for $\mathcal{N} = 48$ in Table I, whereas the corresponding initial frustrated quantum spin system of $N = 96$ sites is far beyond reachable sizes by numerics. More results on hard rhombi are collected in Appendix B.

Hard rhombi on the kagome lattice can be treated similarly to other lattice gases of hard-core objects [29–32]. More specifically, one can introduce the grand-canonical partition function

$$\Xi(z, \mathcal{N}) = \sum_{n=0}^{n_{\text{max}}} z^n g_{\mathcal{N}}(n), \quad (9)$$

where $z = e^{\mu/T}$ is the activity and $g_{\mathcal{N}}(n)$ is the canonical partition function of n hard rhombi on the kagome lattice

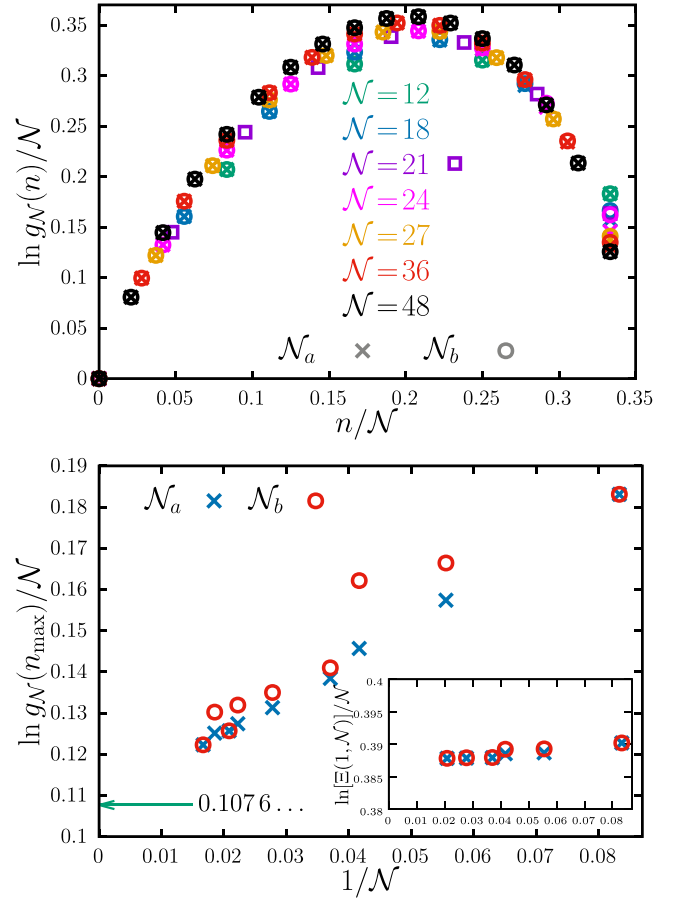


FIG. 5. Hard-rhombi description of the kagome-lattice bilayer. $\mathcal{N}$ denotes the number of kagome-lattice sites; n denotes the number of hard rhombi. Top: $\ln g_{\mathcal{N}}(n)/\mathcal{N}$ versus $n/\mathcal{N}$ for various periodic clusters. Bottom: $s_0(\mathcal{N}/3, \mathcal{N})$ (10) versus $1/\mathcal{N}$; we get in the thermodynamic limit, linear fit, $0.1069\dots$ ($\mathcal{N}_a$) and $0.1106\dots$ ($\mathcal{N}_b$). Inset in the bottom panel: $\ln \kappa(1)$ (11) versus $1/\mathcal{N}$; we get in the thermodynamic limit, linear fit, $0.3867\dots$ ($\mathcal{N}_a$) and $0.3869\dots$ ($\mathcal{N}_b$).

of $\mathcal{N}$ sites or, in other words, the number of allowed spatial configurations.

Clearly, $g_{\mathcal{N}}(n)$ which is related to the ground-state degeneracy yields the residual ground-state entropy through the relation

$$s_0(n, \mathcal{N}) = \frac{\ln g_{\mathcal{N}}(n)}{\mathcal{N}}. \quad (10)$$

In the upper panel of Fig. 5 we report the dependence of the residual ground-state entropy s_0 on the density of hard rhombi on the kagome lattice $n/\mathcal{N}$.

The case of $n = n_{\text{max}}$ (full covering) deserves more discussion. In Table II we report the results for $g_{\mathcal{N}}(n_{\text{max}})$, $n_{\text{max}} = \mathcal{N}/3$ for periodic lattices up to $\mathcal{N} = 60$ and in the lower panel of Fig. 5 we use these data to plot the residual ground-state entropy $s_0(n_{\text{max}}, \mathcal{N}) = [\ln g_{\mathcal{N}}(n_{\text{max}})]/\mathcal{N}$ (i.e., for $n/\mathcal{N} = n_{\text{max}}/\mathcal{N} = 1/3$) as a function of $1/\mathcal{N}$ to illustrate what happens in the limit $\mathcal{N} \rightarrow \infty$. The simplest linear fit results in, depending on the specific boundary conditions, $0.1069\dots$ or $0.1106\dots$. Both numbers for the estimate

TABLE II. Numbers of spatial configurations of $n_{\max} = \mathcal{N}/3$ hard rhombi on the kagome lattice of $\mathcal{N}$ sites for two clusters with edge vectors $\mathbf{a}_1 = m\mathbf{e}_1$, $\mathbf{a}_2 = n\mathbf{e}_2$, $mn = \mathcal{N}/3$ ($\mathcal{N}_a$) and $\mathbf{a}_1 = m\mathbf{e}_1$, $\mathbf{a}_2 = -\mathbf{e}_1 + n\mathbf{e}_2$, $mn = \mathcal{N}/3$ ($\mathcal{N}_b$).

$n_{\max}$	$g_{\mathcal{N}_a}(n_{\max}), \mathcal{N}_a$	$g_{\mathcal{N}_b}(n_{\max}), \mathcal{N}_b$
$\mathcal{N} = 12$	9	9
$\mathcal{N} = 18$	17	20
$\mathcal{N} = 24$	33	49
$\mathcal{N} = 27$	42	45
$\mathcal{N} = 36$	113	129
$\mathcal{N} = 45$	309	379
$\mathcal{N} = 48$	417	417
$\mathcal{N} = 54$	860	1133
$\mathcal{N} = 60$	1537	1537

of $s_0(\mathcal{N}/3, \mathcal{N})$ in the thermodynamic limit are close to 0.1076..., and this issue is discussed in the next paragraph.

From another perspective, as is evident from Fig. 4, the kagome-lattice hard-rhombi problem can be mapped onto the honeycomb-lattice hard-dimer problem with the relation $N = 2\mathcal{N}/3$ between the numbers of honeycomb-lattice sites N and kagome-lattice sites $\mathcal{N}$ and the number of dimers $0, 1, \dots, N/2$ corresponds to the number of rhombi $0, 1, \dots, \mathcal{N}/3$. Moreover, it is known that the number of dimer coverings (close-packed dimers) W on the infinite periodic honeycomb lattice with $N \rightarrow \infty$ sites is $\ln W/(N/2) = 0.3230\dots$ [33]. Since $W = g_{\mathcal{N}}(n_{\max})$, the limiting value of $s_0(n_{\max}, \mathcal{N})$ is three times smaller than 0.3230... in agreement with the lower panel of Fig. 5.

Another function $\kappa(z)$, which has been intensively investigated for lattice gases of hard-core objects, is defined as

$$\ln \kappa(z) = \frac{\ln \Xi(z, \mathcal{N})}{\mathcal{N}}. \quad (11)$$

Its values at $z = 1$, i.e., $\ln \kappa(1) = \ln[\sum_{n=0}^{n_{\max}} g_{\mathcal{N}}(n)]/\mathcal{N}$ were estimated for several models in the past. Namely, for hard squares on the square lattice $\ln \kappa(1) = 0.4074\dots$, for hard hexagons on the honeycomb lattice $\ln \kappa(1) = 0.4359\dots$, for hard hexagons on the triangular lattice $\ln \kappa(1) = 0.3332\dots$, see, e.g., Ref. [34]. As can be seen from the inset in the lower panel of Fig. 5, for hard rhombi on the kagome lattice we obtain $\ln \kappa(1) \approx 0.387$. As a result, we conclude that within the hard-rhombi picture we take into account $\kappa(1)^{\mathcal{N}} \approx e^{0.387\mathcal{N}}$ states, whereas the soft-rhombi picture accounts for $2^{\mathcal{N}} \approx e^{0.693\mathcal{N}}$ states. Although these localized states constitute only a part of the total $2^{2\mathcal{N}} \approx e^{1.386\mathcal{N}}$ states for the initial model (1), they dominate the low-temperature magnetothermodynamics if $J_2 > 2J$.

C. Low-temperature magnetothermodynamics

After counting the hard-rhombi states, we can go one step further and elaborate their contribution to observable properties. In the presence of an external magnetic field $h > 0$, the energy of the n hard-rhombi state is $E_n(h) = E_{\text{FM}} - \mathcal{N}h + n(\epsilon_0 + h)$. The number of hard-rhombi states $g_{\mathcal{N}}(n)$ is equal to the number of allowed spatial configurations of n hard rhombi on the $\mathcal{N}$ -site kagome lattice, that is, to their canonical

partition function. As a result, the contribution of localized singlets satisfying the hard-core rule to the partition function of the frustrated quantum spin system $Z_{\text{lm}}(T, h, N)$ is related to the grand-canonical partition function of hard-core rhombi on kagome lattice $\Xi(z, \mathcal{N})$ (9)

$$\begin{aligned} Z_{\text{lm}}(T, h, N) &= \sum_{n=0}^{n_{\max}} g_{\mathcal{N}}(n) e^{-\frac{E_n(h)}{T}} \\ &= e^{-\frac{E_{\text{FM}} - \mathcal{N}h}{T}} \sum_{n=0}^{n_{\max}} z^n g_{\mathcal{N}}(n) = e^{-\frac{E_{\text{FM}} - \mathcal{N}h}{T}} \Xi(z, \mathcal{N}), \\ z &= e^{\frac{\mu}{T}}, \quad \mu = h_{\text{sat}} - h, \quad h_{\text{sat}} = -\epsilon_0 = 4J + J_2. \end{aligned} \quad (12)$$

This is a dominant contribution at low temperatures and high fields.

It is convenient to represent the grand-canonical partition function of hard-core rhombi on a kagome lattice $\Xi(z, \mathcal{N})$, which enters Eq. (12), in terms of the on-site occupation numbers $n_i = 0, 1$:

$$\Xi(z, \mathcal{N}) = \sum_{n_1=0,1} \dots \sum_{n_{\mathcal{N}}=0,1} z^{n_1 + \dots + n_{\mathcal{N}}} e^{-\frac{V \sum_{\langle ij \rangle} n_i n_j}{T}} \quad (13)$$

(the sum in the exponent runs over all nearest neighbors on the kagome lattice). Sending here $V \rightarrow +\infty$ (infinite repulsion), we restrict the allowed configurations to the hard-rhombi patterns. Finite repulsion V accounts for overlapping rhombi states and setting $V = J$ we reproduce correctly the energy of these states. Clearly, in Eq. (13) we face the grand-canonical partition function of a classical lattice-gas model on the kagome lattice defined by the Hamiltonian

$$\begin{aligned} \mathcal{H}(\{n_i\}) &= -\mu \sum_i n_i + V \sum_{\langle ij \rangle} n_i n_j, \\ \mu &= h_{\text{sat}} - h, \quad h_{\text{sat}} = 4J + J_2 \end{aligned} \quad (14)$$

with infinite nearest-neighbor repulsion $V \rightarrow \infty$ (hard-core rhombi) or $V = J$ (partially overlapping or soft-core rhombi). The lattice-gas model of rhombi (14) can be cast into the antiferromagnetic Ising model in a field after the change $n_i = 1/2 - T_i^z$:

$$\mathcal{H}(\{T_i^z\}) = \frac{V - \mu}{2} \mathcal{N} + (\mu - 2V) \sum_i T_i^z + V \sum_{\langle ij \rangle} T_i^z T_j^z. \quad (15)$$

Kagome-lattice Ising models were examined in the early 1950s [35,36].

It is worth stressing that the lattice-gas model (and the Ising model) possesses a symmetry with respect to the change of variables n_i to $\bar{n}_i = 1 - n_i$ (equivalently T_i^z to $-T_i^z$): The thermodynamic quantities for $h_{\text{sat}} - h$ are straightforwardly related to those for $h - h_1$. Another symmetry occurs at $h_2 = J_2 + 2J_1$: Again, the thermodynamic quantities for $h_2 + \Delta h$ are related to those for $h_2 - \Delta h$.

Now we use the classical lattice models to corroborate the suggested description of low-lying Hamiltonian eigenstates by comparison with numerics. More precisely, we perform direct calculations for the lattice models given in Eq. (14) or Eq. (15) with $V \rightarrow \infty$ and $V = J$, to obtain various quantities and compare them to the finite-lattice results for the initial

quantum spin model (1). In particular, we consider the magnetization and the susceptibility per site of the initial model (1)

$$\begin{aligned} m(T, h) &= \frac{1}{2} + \frac{T}{2\mathcal{N}} \frac{\partial \ln \Xi(T, \mu, \mathcal{N})}{\partial h}, \\ \chi(T, h) &= \frac{\partial m(T, h)}{\partial h} \end{aligned} \quad (16)$$

along with the entropy and the specific heat per site of the initial model (1)

$$\begin{aligned} s(T, h) &= \frac{\ln \Xi(T, \mu, \mathcal{N})}{2\mathcal{N}} + \frac{T}{2\mathcal{N}} \frac{\partial \ln \Xi(T, \mu, \mathcal{N})}{\partial T}, \\ c(T, h) &= T \frac{\partial s(T, h)}{\partial T}, \end{aligned} \quad (17)$$

where $\Xi(T, \mu, \mathcal{N})$ is the grand-canonical partition function of a lattice gas.

Let us begin with the temperature dependencies around saturation, see the left column in Fig. 6. According to numerics for $N = 36_b$ (circles), $m(T)$ for $h > h_{\text{sat}}$ decreases as T grows, while $m(T)$ for $h \lesssim h_{\text{sat}}$ slightly increases as T deviates from zero and then decreases as T grows further. This is in accordance with the expected smearing out of the ground-state magnetization jump at the saturation field h_{sat} . The hard-rhombi description (thin curves) works perfectly well in the low-temperature region but fails above $T \approx 0.2$ and cannot reproduce vanishing magnetization to zero in the high-temperature limit. Instead, as is obvious from Eq. (16), the hard-rhombi prediction in the high-temperature limit tends to a nonzero value, which is related to the averaged density of hard rhombi $\langle n \rangle / \mathcal{N}$ at $z=1$ [according to Table I, $(\langle n \rangle / \mathcal{N})|_{z=1} \approx 0.202$ for $\mathcal{N} = 18$]. Soft rhombi (thick curves) provide a correct description up to $T \approx 1$ but for higher temperatures tends again to a nonzero value, which is related to the averaged density of soft rhombi $\langle n \rangle / \mathcal{N}$ at $T \rightarrow \infty$, $(\langle n \rangle / \mathcal{N})|_{T \rightarrow \infty} = 1/2$. The next panel in the left column of Fig. 6 concerns the temperature dependence of the uniform susceptibility. In the high-temperature limit it obeys the Curie law: $\chi(T \rightarrow \infty) \rightarrow 1/(4T)$; in the temperature range $T < 10$ this regime is not achieved yet (circles). Hard- and soft-rhombi correctly reproduce the low-temperature behavior as well as the $1/T$ dependence at high temperatures, however, with a constant smaller than $1/4$. Furthermore, the temperature profiles of entropy have clear interpretation. For $h > h_{\text{sat}}$, $s(T=0)=0$, but for $h_2 < h < h_{\text{sat}}$, $s(T=0) = \ln \mathcal{W}_{\text{GS}} / \mathcal{N}$, $\mathcal{W}_{\text{GS}} = g_{\mathcal{N}}(\mathcal{N}/3)$, i.e., is ≈ 0.083 for $N = 36_b$. Rhombi pictures perfectly reproduce the low-temperature region. On the other hand, as $T \rightarrow \infty$, the entropy tends to $\ln 2$ (exact value), but to $\ln 2/2$ (soft rhombi) or $\ln \kappa(1)/2$ (hard rhombi). We finish the discussion with a comparison of the temperature dependence of the specific heat obtained for the initial and effective models, $N = 36_b$ and $\mathcal{N} = 18_b$, see the lowest panel in the left column in Fig. 6. The lower peak of $c(T)$, the position of which is controlled by the energy scale $|h_{\text{sat}} - h|$, perfectly matches the rhombi predictions. The main peak of $c(T)$ at $T \approx 4$ is beyond the capabilities of the rhombi description. Note that the main peak is also present at $h = 0$ (red circles in the bottom right panel, slightly below $T = 2$) and it comes from *all* possible, not only localized, spin-fliplike excitations:

Rhombi states in this case (solid red curve) account for only a part of all states relevant at these temperatures.

The temperature dependencies of m , χ , s , and c for lower fields are shown in the right column of Fig. 6. The reported comparisons demonstrate that the soft-rhombi description works even at $h = 0$ as long as the temperature does not exceed ≈ 0.45 ; the hard-rhombi picture (not shown here) in general fails further away from h_{sat} .

Overall, the comparison between hard/soft-rhombi predictions and exact numerics reported in Fig. 6 indicates the region of validity of the effective-model description which obviously concerns sufficiently low temperatures only. In this region of validity, such a description opens new possibilities to examine the frustrated quantum spin model (1) by means of the classical statistical mechanics. Besides direct calculations for $\mathcal{N}$ about 60 (see Appendix B), classical Monte Carlo simulations for much larger $\mathcal{N}$ can be used, too.

D. Do the bulk properties depend on the form?

We conclude this section with an interesting consequence of the established dimer representation for the low-energy physics of the frustrated quantum spin model (1). Usually it is implied that the bulk properties are insensitive to the boundary conditions in the limit of a large system. Remarkably, in 1961 P. Kasteleyn [37], while studying dimer arrangements on a square lattice, expressed doubts on the independence of the bulk free energy on boundary conditions (for more recent publications see Refs. [38,39]). More relevant in the present context is the paper by V. Elser [40], who considered the dimer problem on the honeycomb lattice with boundary and demonstrated explicitly that the bulk entropy per dimer in the thermodynamic limit depends on the shape of the boundary.

To be precise, for periodic (toroidal) boundary conditions the bulk entropy per dimer $\ln W/(N/2)$ has been known since the 1950s, and it equals $0.3230\dots$ [33]. Elser studied the dimer problem on a general hexagon defined by the sides k , l , and m , see Fig. 7. In particular, using MacMahon's results for the combinatorial problem of "plane partitions," he provides the limiting value of the entropy per dimer $\ln W/(N/2) = S_{klm}/(N/2)$ as $N \rightarrow \infty$:

$$\begin{aligned} \frac{S_{klm}}{N/2} &\sim s(x, y, z), \\ \frac{N}{2} &= kl + lm + mk, \\ s(x, y, z) &= \frac{1}{2(xy+yz+zx)} [x^2 \ln x - (1-x)^2 \ln(1-x) \\ &\quad + y^2 \ln y - (1-y)^2 \ln(1-y) \\ &\quad + z^2 \ln z - (1-z)^2 \ln(1-z)], \\ x &= \frac{k}{n}, \quad y = \frac{l}{n}, \quad z = \frac{m}{n}, \quad n = k + l + m. \end{aligned} \quad (18)$$

This formula demonstrates how the bulk entropy per dimer depends on the shape of the boundary. Moreover, the maximum of the entropy per dimer $s(x, y, z)$ is obtained when the boundary is a regular hexagon, i.e., $s(1/3, 1/3, 1/3) = (3/2) \ln 3 - 2 \ln 2 \approx 0.262$ which is smaller than the value for

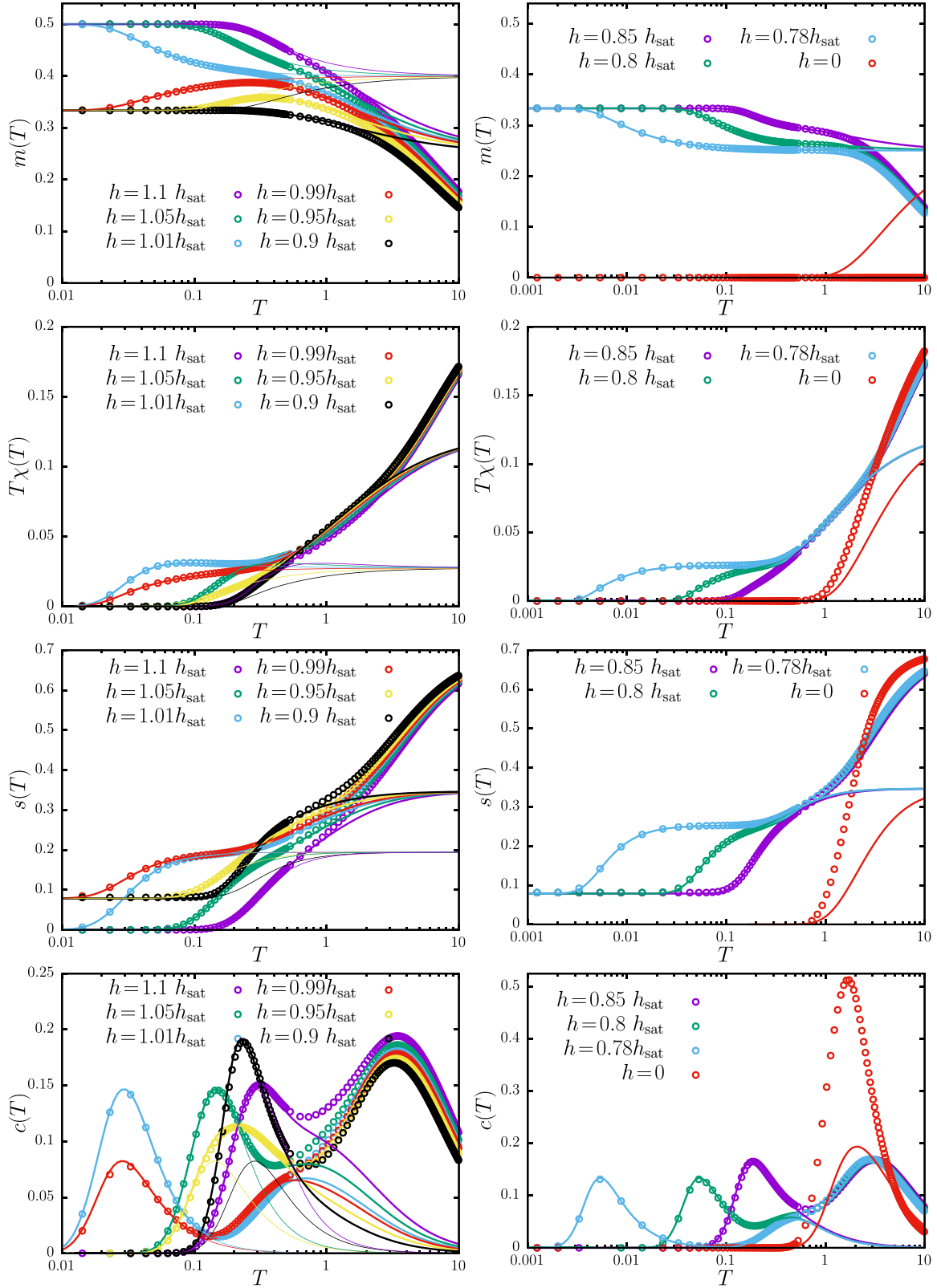


FIG. 6. Hard- and soft-core rhombi predictions (thin and thick curves) against finite-temperature Lanczos method data (circles) for thermodynamic characteristics, $J = 1$, $J_2 = 5$, $N = 36_b$. From top to bottom: Magnetization m , susceptibility χ , entropy s , and specific heat c per site of the initial model (1). Left column: High fields $h = 9.9, 9.45, 9.09, 8.91, 8.55, 8.1$. Right column: Moderate fields $h = 7.65, 7.2, 7.02$ and $h = 0$. Hard-core rhombi work up to $T \approx 0.15$; soft-core rhombi work up to $T \approx 0.45$.

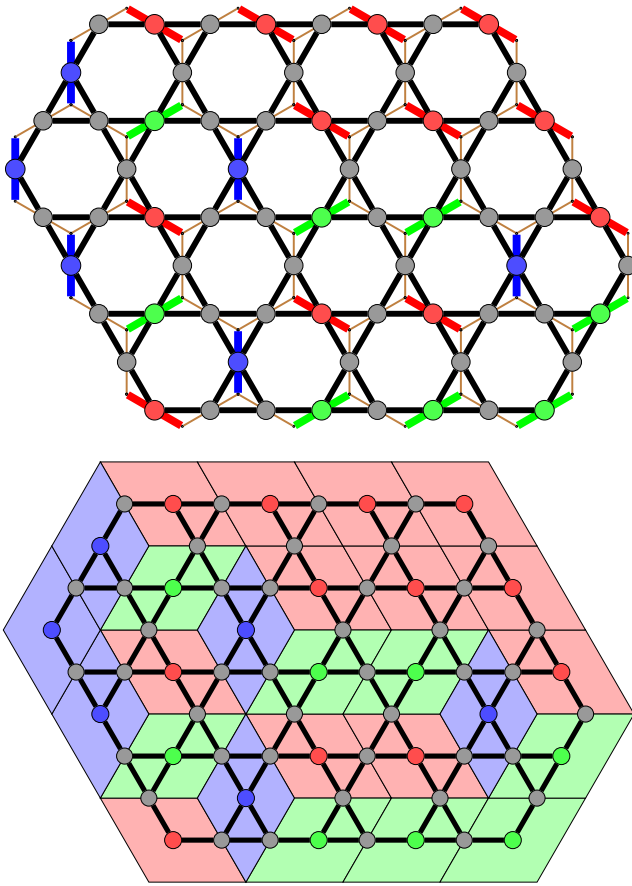


FIG. 7. General hexagon with the sides $k = 4$, $l = 3$, and $m = 2$ considered for calculating the bulk entropy for the dimer problem on the honeycomb lattice by Elser [40] (top) and the corresponding rhombi on the kagome lattice (bottom).

periodic boundary conditions. And the entropy per dimer goes to zero if, e.g., $x = 1$, $y = z = 0$.

Since the dimer problem on the honeycomb lattice is equivalent to the hard-rhombi covering of the kagome lattice and the residual ground-state entropy for the $2/3$ plateau state $s_0(N/3, N)$ (10) is three times smaller than $s(x, y, z)$, see above, we conclude that for $h_2 < h < h_{\text{sat}}$ the residual ground-state entropy (17) $s(T = 0, h \approx h_{\text{sat}}) = s(x, y, z)/6$ depends on the shape of thermodynamically large open-boundary (hexagonal) system, see Eq. (18). On the other hand, $s(T) = \ln 2 - \int_T^\infty dT c(T)/T$ and, as a result, we have the following sum rule:

$$\int_0^\infty dT \frac{c(T)}{T} = \ln 2 - s(T = 0). \quad (19)$$

Since $s(T=0)$ on the right-hand side of Eq. (19) depends on the shape of the thermodynamically large open-boundary (hexagonal) system, $c(T)$ in the left-hand side of Eq. (19) should also depend on the shape.

It is not easy to check a dependence of $c(T)$ on the form of the thermodynamically large system having access only to rather small systems. In particular, the finite-temperature Lanczos method deals with $N = 36, 42$ (i.e., $\mathcal{N} = 18, 21$) only, while direct calculations for hard rhombi are restricted

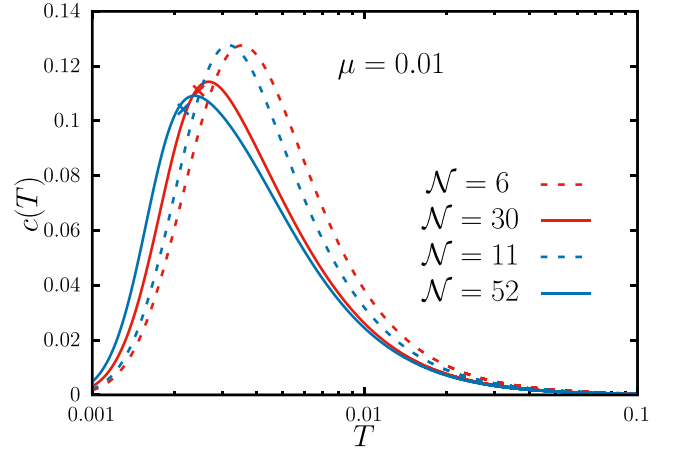


FIG. 8. Temperature dependence of the specific heat ($\mu = h_{\text{sat}} - h = 0.01$) for two open hexagonal systems with $k:l:m = 1:1:1$ (red) and $k:l:m = 2:1:1$ (blue). $c(T)$ for $\mathcal{N} = 6, 11$ (dashed curves) and for $\mathcal{N} = 30, 52$ (solid curves). The position of the peak is linearly extrapolated for $1/\mathcal{N} \rightarrow 0$ (crosses).

to $\mathcal{N}$ about 60. In Fig. 8 we report preliminary results for two hexagon shapes: $1:1:1$ (red curves) and $2:1:1$ (blue curves) as they follow from direct calculations for hard-rhombi systems with $\mu = h_{\text{sat}} - h = 0.01$ of sizes $\mathcal{N} = 6, 30$ (red dashed and solid curves) and $\mathcal{N} = 11, 52$ (blue dashed and solid curves). Moreover, we extrapolate the peak of $c(T)$ as $1/\mathcal{N} \rightarrow 0$ (linear extrapolation), which yields the red and blue crosses in Fig. 8. Our findings agree with the expectation about a shape-dependent $c(T)$. However, extensive numerical studies for larger systems are required to better understand this issue.

IV. BEYOND THE FULLY FRUSTRATED LIMIT: NUMERICS VERSUS EFFECTIVE THEORY

In this section, we examine what happens around the fully frustrated case, when the ideal flat-band condition $J_1 = J_x$ is slightly violated. Numerics for the magnetization curve and the temperature dependence of the specific heat for $J_1 = 1.1$, $J_x = 0.9$ are reported in Fig. 9 (circles). Next we discuss an effective theory explaining these data, see Refs. [14,41].

In the regime when J_2 is the dominating coupling, one may elaborate a strong-coupling approach and obtain an effective theory which is valid for $J_1 \neq J_x$, too. Within this approach, one starts from noninteracting singlets at the magnetic field $h_0 = J_2$, which are governed by the Hamiltonian $H_{\text{main}} = \sum_{\mathbf{m}\alpha} [J_2(\mathbf{t}_{\mathbf{m}\alpha}^2 - 3/2)/2 - h_0 \mathbf{t}_{\mathbf{m}\alpha}^z]$, cf. Eq. (8). The remaining Hamiltonian, i.e., $V = H - H_{\text{main}}$, is treated as a perturbation. Clearly, the ground state of H_{main} is $2^{\mathcal{N}}$ -fold degenerate and forms the model space defined by the projector $P = \otimes_{\mathbf{m}\alpha} (|u\rangle\langle u| + |d\rangle\langle d|)_{\mathbf{m}\alpha}$, where $|u\rangle = |\uparrow_a \uparrow_b\rangle$ and $|d\rangle = (|\uparrow_a \downarrow_b\rangle - |\downarrow_a \uparrow_b\rangle)/\sqrt{2}$. Switching on $V \neq 0$ lifts the degeneracy in the model space, and an effective Hamiltonian H_{eff} , which acts in this space and describes the low-energy properties of H around the limit $V = 0$, can be constructed perturbatively [42]. Calculating the first term in the expansion $H_{\text{eff}} = P(H_{\text{main}} + V)P + \mathcal{O}(V^2)$ and introducing the pseudospin one-half operators $T^z = (|u\rangle\langle u| - |d\rangle\langle d|)/2$,

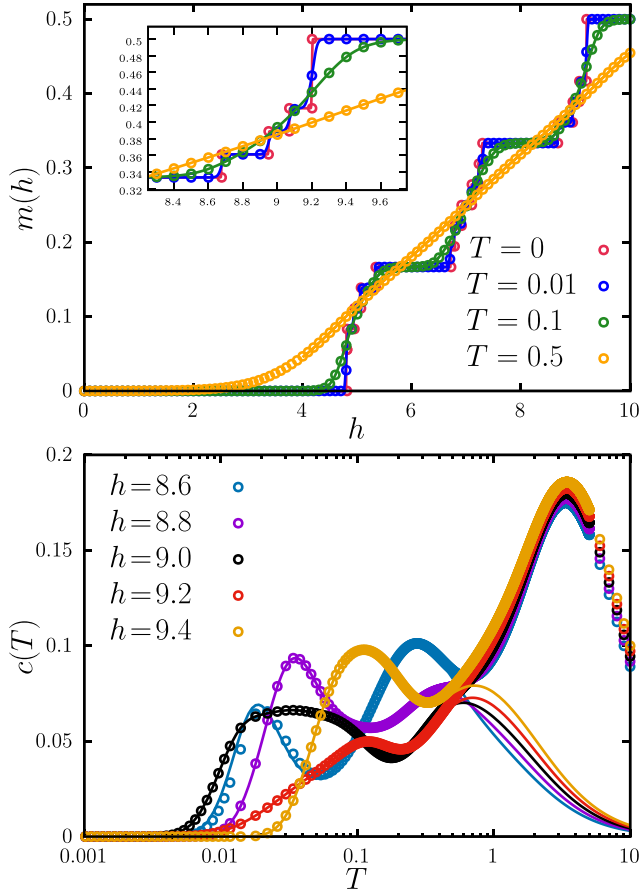


FIG. 9. Top: Magnetization curve $m(h)$ for the $S = 1/2$ frustrated kagome-lattice bilayer with $J_1 = 1.1$, $J_x = 0.9$, $J_2 = 5$, $N = 36_a$; $T = 0, 0.01, 0.1, 0.5$. Inset is a zoom-in of the region around $h = 9$. Bottom: Specific heat $c(T)$ for the same system at $h = 8.6, 8.8, 9.0, 9.2, 9.4$. In both panels along with numerics for the 36-site initial model (1) (circles) we also show the results of the effective theory (20) with $\mathcal{N} = 18_a$ (solid curves).

$T^+ = |u\rangle\langle d|$, $T^- = |d\rangle\langle u|$, one gets $P(t^x, t^y, t^z)P = (0, 0, 1/2 + T^z)$, $P(d^x, d^y, d^z)P = (-\sqrt{2}T^x, -\sqrt{2}T^y, 0)$ and finally arrives at the $S = 1/2$ XXZ model in a field on the kagome lattice. That is, the effective Hamiltonian reads:

$$\begin{aligned}
 H_{\text{eff}} &= \mathcal{CN} - \hbar \sum_m T_m^z \\
 &+ \sum_{\langle mn \rangle} [J^z T_m^z T_n^z + \mathbf{J}(T_m^x T_n^x + T_m^y T_n^y)], \\
 \mathcal{C} &= -\frac{\hbar}{2} \frac{J_2}{4} + \frac{J_1 + J_x}{4}, \quad \hbar = h - J_2 - J_1 - J_x, \\
 J^z &= \frac{J_1 + J_x}{2}, \quad \mathbf{J} = J_1 - J_x.
 \end{aligned} \tag{20}$$

Here the first sum ($m = \mathbf{m}\alpha$) runs over the vertices of the kagome lattice and the second sum ($\langle mn \rangle = \langle \mathbf{m}\alpha \mathbf{n}\beta \rangle$) runs over the edges of the kagome lattice. The ideal flat-band geometry $J_1 - J_x = 0$ corresponds to the kagome-lattice Ising model ($\mathbf{J} = 0$) and is simply another representation of the lattice-gas model discussed above, cf. Eq. (15). In general, we

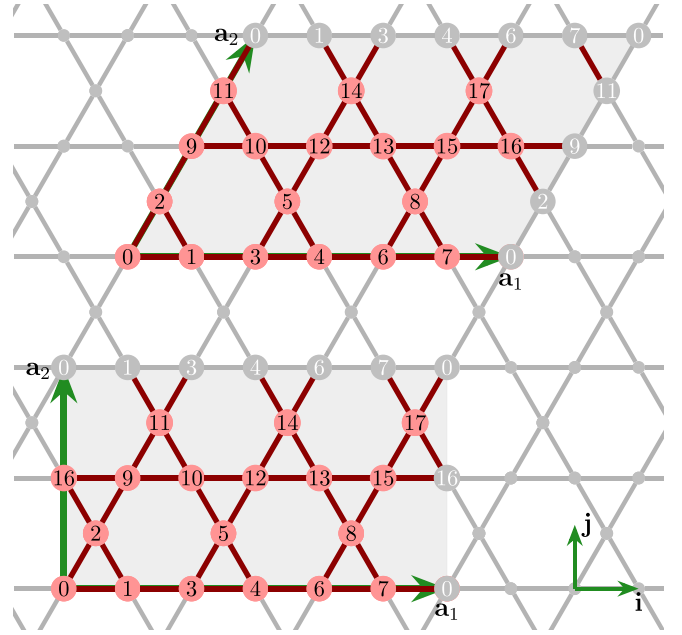


FIG. 10. Two types of periodic boundary conditions for $N = 36$ ($\mathcal{N} = 18$), i.e., (top) determined by the edge vectors $\mathbf{a}_1 = (6, 0)$, $\mathbf{a}_2 = (2, 2\sqrt{3})$ ($N = 36a$ or $\mathcal{N} = 18a$) and (bottom) determined by the edge vectors $\mathbf{a}_1 = (6, 0)$, $\mathbf{a}_2 = (0, 2\sqrt{3})$ ($N = 36b$ or $\mathcal{N} = 18b$).

TABLE III. Numerics for the kagome-lattice bilayer of $N = 36_a$ sites [periodic boundary conditions, the edge vectors are $\mathbf{a}_1 = (6, 0)$ and $\mathbf{a}_2 = (2, 2\sqrt{3})$], $J = 1$, and (from top to bottom) $J_2 = 5, 3, 2$, and 1. We present the energy E_{GS} and degeneracy $\mathcal{W}_{\text{GS}}$ of the ground state in various subspaces $12 \leq S^z \leq 17$. We also present the energy $E_{1\text{min}}$ and degeneracy $\mathcal{W}_{1\text{min}}$ of the first excited state as well as the energy gap $E_{1\text{min}} - E_{\text{GS}}$ in various subspaces $12 \leq S^z \leq 17$.

S^z	E_{GS}	$\mathcal{W}_{\text{GS}}$	$E_{1\text{min}}$	$\mathcal{W}_{1\text{min}}$	$E_{1\text{min}} - E_{\text{GS}}$
17	49.5	18	52.5	7	3
16	40.5	117	41.5	36	1
15	31.5	336	32.5	396	1
14	22.5	417	23.5	1356	1
13	13.5	186	14.5	1572	1
12	4.5	17	5.5	456	1
17	42.5	18	43.5	7	1
16	35.5	117	36.5	108	1
15	28.5	336	29.5	585	1
14	21.5	417	22.5	1456	1
13	14.5	186	15.5	1584	1
12	7.5	17	8.5	456	1
17	39	25	40.267 9...	2	1.267 9...
16	33	199	33.279 0...	3	0.279 0...
15	27	544	27.063 6...	12	0.063 6...
14	21	523	21.106 9...	24	0.106 9...
13	15	198	15.224 4...	48	0.224 4...
12	9	17	9.620 8...	24	0.620 8...
17	34.5	7	35.5	18	1
16	28.5	10	28.779 0...	3	0.279 0...
15	22.5	1	22.650 9...	1	0.150 9...
14	17.028 5...	1	17.260 8...	3	0.232 3...
13	11.950 5...	1	12.185 4...	3	0.234 9...
12	7.257 1...	1	7.431 9...	1	0.174 8...

TABLE IV. Numerics for the kagome-lattice bilayer of $N = 36_b$ sites [periodic boundary conditions, the edge vectors $\mathbf{a}_1 = (6, 0)$ and $\mathbf{a}_2 = (0, 2\sqrt{3})$], $J = 1$, and (from top to bottom) $J_2 = 5, 3, 2$, and 1 . We present the same quantities as in Table III.

S^z	E_{GS}	$\mathcal{W}_{\text{GS}}$	$E_{1\text{min}}$	$\mathcal{W}_{1\text{min}}$	$E_{1\text{min}} - E_{\text{GS}}$
17	49.5	18	52.5	7	3
16	40.5	117	41.5	36	1
15	31.5	336	32.5	396	1
14	22.5	420	23.5	1344	1
13	13.5	192	14.5	1560	1
12	4.5	20	5.5	456	1
17	42.5	18	43.5	7	1
16	35.5	117	36.5	108	1
15	28.5	336	29.5	594	1
14	21.5	420	22.5	1504	1
13	14.5	192	15.5	1590	1
12	7.5	20	8.5	456	1
17	39	25	40	2	1
16	33	199	33.198 0...	7	0.198 0...
15	27	566	27.092 6...	24	0.092 6...
14	21	595	21.117 0...	24	0.117 0...
13	15	222	15.215 9...	12	0.215 9...
12	9	20	9.585 7...	6	0.585 7...
17	34.5	7	35.5	20	1
16	28.5	10	28.698 0...	1	0.198 0...
15	22.5	2	22.678 4...	1	0.178 4...
14	16.939 9...	1	17.160 9...	2	0.220 9...
13	11.995 8...	1	12.064 9...	1	0.069 0...
12	7.239 1...	1	7.256 2...	1	0.017 0...

arrive at the $S = 1/2$ XXZ model with easy axis anisotropy, $|J| \ll J^z$, $J^z > 0$, in a field on the kagome lattice, for which many results are known to date, see, e.g., Ref. [43] and references therein.

The presented field and temperature dependencies in Fig. 9 show good agreement between exact numerics ($N = 36$) and effective-theory predictions ($\mathcal{N} = 18$) for the chosen set of parameters. Interestingly, effective model (20) is constructed perturbatively around the main part H_{main} defined above, i.e., near the limit of noninteracting dimers at the degenerate singlet-triplet point ($J_2 > 0$, $J_1 = J_x = J = 0$, $h = h_0 = J_2$), and therefore $h - h_0$ and J_1 , J_x are implied to be small in Eq. (20). Besides, T also cannot be too large, because of the number of states taking into account in Eq. (20): $2^{\mathcal{N}}$ instead

TABLE V. Counting the number of n hard-rhombi spatial configurations on the kagome lattice of $\mathcal{N} = 21$ sites.

n	$g_{21}(n)$
1	21
2	168
3	644
4	1225
5	1085
6	371
7	31

of 2^N , this becomes visible at $T \approx 0.5$ in Fig. 9, bottom. Nevertheless, the magnetization curve, Fig. 9, top, is reproduced by Eq. (20) quite well for all h and $T = 0 \dots 0.5$. Again, the effective description has an advantage for examining the frustrated quantum spin model (1): Although the spin model (20) remains a frustrated quantum spin system, it contains a two times smaller number of sites, $\mathcal{N} = N/2$. Also, as explained above in Sec. III C, the region of the stable dominant peak of $c(T)$ at $T \approx 4$ is outside the applicability of the description on the basis of Eq. (20).

V. SUMMARY

The present study continues the examination of the $S = 1/2$ Heisenberg model on frustrated bilayer lattices in the regime of strong antiferromagnetic interlayer coupling. In this paper, we studied the case of the kagome-lattice bilayer. Such a bilayer shows a prominent difference to other ones such as square-, honeycomb-, or triangular-lattice bilayers. While the latter bilayers for $h \lesssim h_{\text{sat}}$ exhibit a ground-state ordering of singlets which persists up to a finite temperature $T_c(h)$, the former one does not show a symmetry breaking, but rather a huge degeneracy of the ground state, which grows exponentially with lattice size. This degeneracy has been known in combinatorics for many years. Two features of the kagome-lattice bilayer at low temperatures and just below the saturation field are peculiar and interesting: There is no phase transition related to the singlets ordering, and the bulk specific heat may depend on the sample form.

Concerning the experimental side, one may mention several quantum Heisenberg antiferromagnets on frustrated bilayer lattices: $\text{Ba}_2\text{CoSi}_2\text{O}_6\text{Cl}_2$ [11,17] is a square-lattice bilayer, $\text{Bi}_3\text{Mn}_4\text{O}_{12}(\text{NO}_3)$ [44,45] is a honeycomb-lattice bilayer, and $\text{K}_2\text{Co}_2(\text{SeO}_3)_3$ [46] is a triangular-lattice bilayer. Unfortunately, we are not aware of a solid-state realization of the frustrated kagome-lattice bilayer quantum Heisenberg antiferromagnet yet.

Finally, let us mention several problems which in our opinion deserve further studies. As follows from the calculation of the entropy as a function of temperature and magnetic field (Sec. III C), the system at hand should exhibit an interesting magnetocaloric effect since the residual entropy remains a nonzero constant for the $2/3$ - and $1/3$ -magnetization plateaus [47–49]. In view of Elser’s formula (18), see Sec. III D, it is worth to examine the thermodynamics of the hexagonal-shaped systems in detail. To this end, one has to consider larger systems and adapt Monte-Carlo simulations accordingly. One may also consider other Archimedean lattice bilayers [50,51], which, however, might not provide new physics in comparison to the cases studied already in Refs. [7,13,16] and in the present paper.

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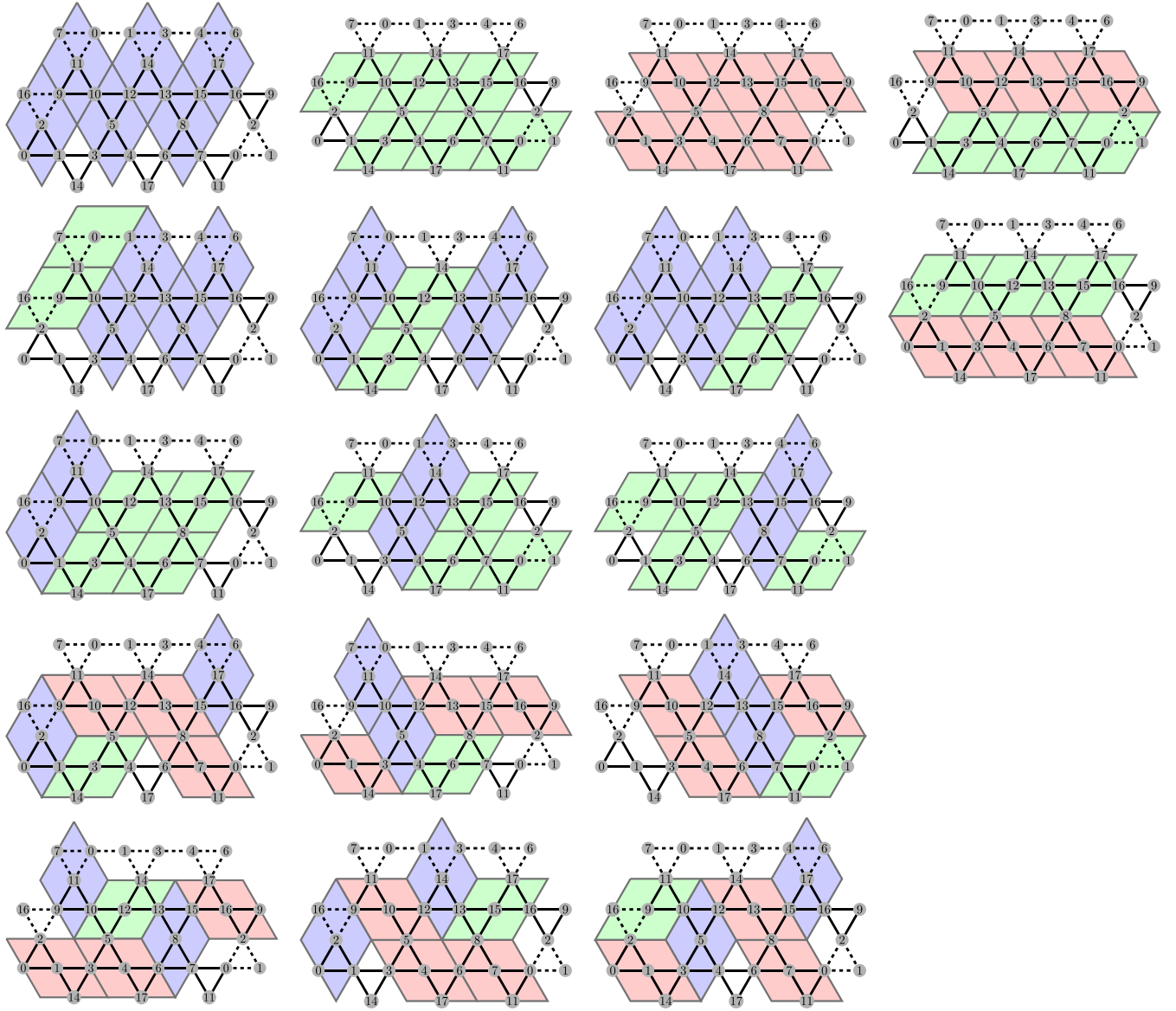


FIG. 11. Seventeen hard-rhombi coverings of the periodic kagome lattice of $N = 18_a$ sites. Numerics for the periodic spin system $N = 36_a$ gives for the localized-magnon crystal state the same degeneracy $\mathcal{W}_{18} = 17$; see Table III.

quantum spin models to explain the properties of magnets over wide temperature range”), which is funded by the European Union under Grant No. 871072. O.D. acknowledges the kind hospitality of the University of Bielefeld in October–December of 2023 (supported by Erasmus+ and DFG). O.D. acknowledges the hospitality of the MIPKs, Dresden at the Intercontinental Binodal Workshop Flat bands and high-order Van Hove singularities (27 May–7 June 2024) and thanks A. Honecker and K. Karlová for their hospitality at the Advanced quantum materials for magnetic cooling and quantum information science conference (Cergy-Pontoise, France, 19–21 February 2025). J.S. thanks the Max Planck Institute for the Physics of Complex Systems for the hospitality during a visit in April 2025. This work was supported by the Deutsche Forschungsgemeinschaft (DFG SCHN 615/28-1 and RI 615/25-1).

DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX A: NUMERICS FOR SMALL KAGOME BILAYERS

In this Appendix, we present more results obtained numerically for small lattices. In Fig. 10 we illustrate two types of the 36-site frustrated kagome-lattice bilayer which are used for numerical calculations in our study, cf. Tables III Table IV.

In Tables III and IV we report some characteristics of the ground state and low-lying states (energy and degeneracy) in the subspaces with total $S^z = N/2, \dots, N/3$

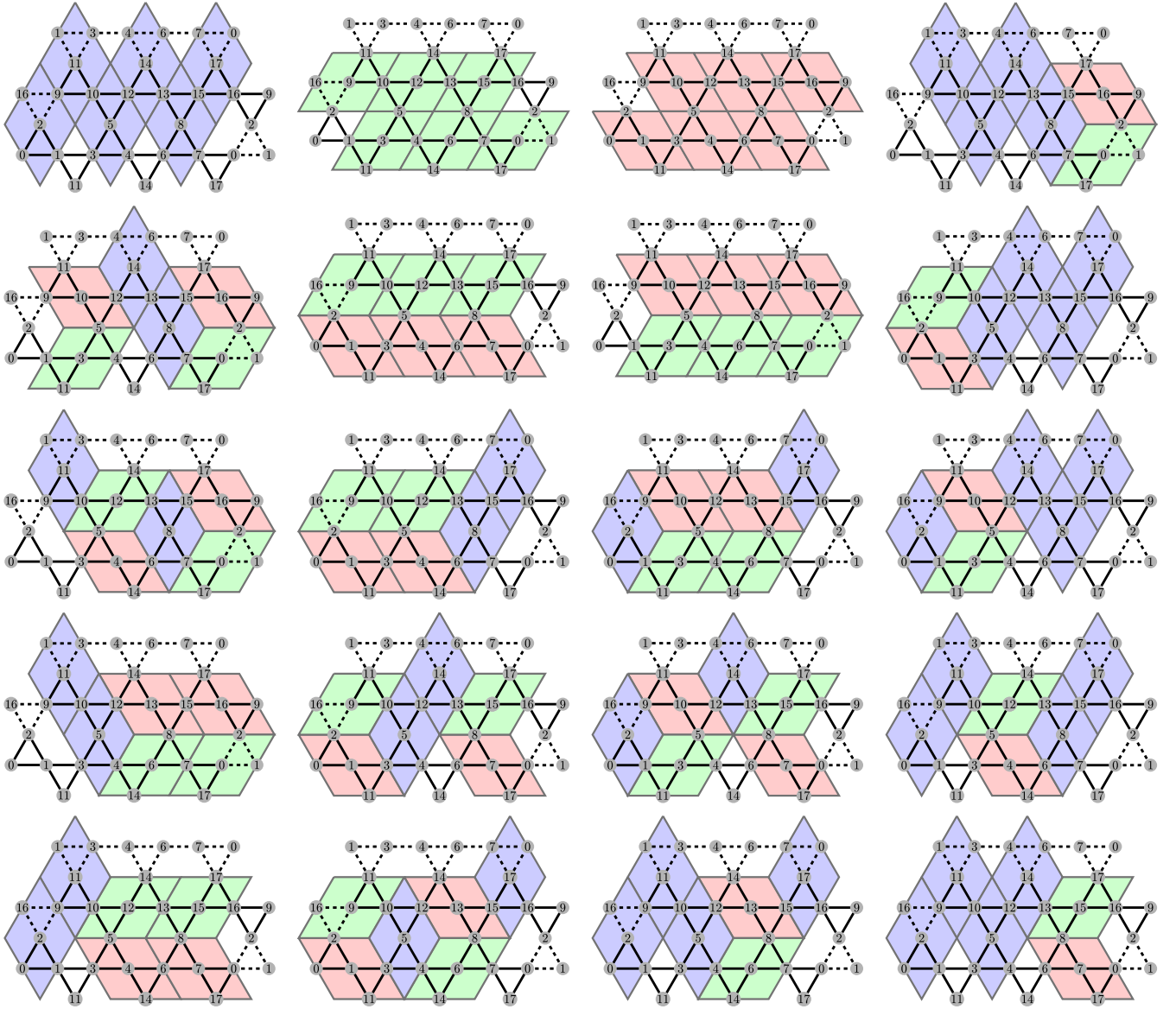


FIG. 12. Twenty hard-rhombi coverings of the periodic kagome lattice of $\mathcal{N} = 18_b$ sites. Numerics for the periodic spin system $N = 36_b$ gives for the localized-magnon crystal state the same degeneracy $\mathcal{W}_{18} = 20$; see Table IV.

($N=36$, two periodic clusters are examined) depending on J_2 to illustrate the emergence of the large- J_2 regime. If J_2 exceeds $2J$, then the low-energy states characteristics are well structured (easy understandable energy and degeneracy) and they do not depend on J_2 . Namely, E_{GS} fits to $E_{\text{FM}} + n\epsilon_0$ with $E_{\text{FM}} = (2J + J_2/4)\mathcal{N}$ and $\epsilon_0 = -(4J + J_2)$, whereas W_{GS} for $J_2 = 5$ and $J_2 = 3$ fits to the number of spatial configurations of $n = 1, \dots, 6$ hard rhombi on the kagome lattice of $\mathcal{N} = 18$ sites, see Table I. Besides, the ground-state degeneracy $\mathcal{W}_{\text{GS}}$ for $S^z = 12$ illustrates the degeneracy of the $2/3$ plateau state shown in Fig. 2. As can be seen in Tables III and IV, its value for $N = 36$ depends on the cluster shape and is rather large being 17 (Table III) or 20 (Table IV). For $N = 42$ the ground-state degeneracy for $S^z = 14$ is 31, i.e., it grows as N increases. $J_2 = 2J$ is a marginal case, exhibiting higher degeneracy of the ground state: The flat-band states (6) and (7) have the same energy. Smaller J_2 indicate completely different nature of the ground state: No regularly

structured energy and no large degeneracy are observed in that case.

The picture of overlapping rhombi is also confirmed by numerical data reported in Tables III and IV. For example, for $J_2 = 5$ the energy gap in the subspaces with $2 \leq n \leq 6$ is $J = 1$ and $\mathcal{W}_{\text{min}}$, according to the soft-rhombi calculations in Appendix B, corresponds to the number of ways to put one pair of neighboring singlets while the rest singlets are not neighbors (patterns with one pair of overlapping rhombi, see the lower panel in Fig. 4).

APPENDIX B: RHOMBI ON THE KAGOME LATTICE

In this Appendix, we provide more results about rhombi on the kagome lattice which complement those that have been reported in Sec. III.

First, in Figs. 11 and 12 we illustrate all spatial configurations of $n_{\text{max}} = 6$ hard rhombi on the $\mathcal{N} = 18$ kagome

TABLE VI. Counting the number of n hard-rhombi spatial configurations on the kagome lattice of $\mathcal{N}$ sites with two types of periodic boundary conditions imposed. These data are complementary to the ones in Table I.

$\mathcal{N} = 12$		
n	$g_{12_a}(n), \mathcal{N}_a$	$g_{12_b}(n), \mathcal{N}_b$
1	12	12
2	42	42
3	44	44
4	9	9
$\mathcal{N} = 24$		
n	$g_{24_a}(n), \mathcal{N}_a$	$g_{24_b}(n), \mathcal{N}_b$
1	24	24
2	228	228
3	1096	1096
4	2830	2834
5	3848	3880
6	2516	2588
7	632	696
8	33	49
$\mathcal{N} = 27$		
n	$g_{27_a}(n), \mathcal{N}_a$	$g_{27_b}(n), \mathcal{N}_b$
1	27	27
2	297	297
3	1719	1719
4	5643	5643
5	10 557	10 557
6	10 737	10 740
7	5319	5328
8	1026	1035
9	42	45
$\mathcal{N} = 36$		
n	$g_{36_a}(n), \mathcal{N}_a$	$g_{36_b}(n), \mathcal{N}_b$
1	36	36
2	558	558
3	4884	4884
4	26 613	26 613
5	93 888	93 888
6	216 232	216 240
7	320 304	320 400
8	293 637	294 045
9	155 364	156 116
10	42 006	42 606
11	4596	4788
12	113	129

lattice with two types of periodic boundary conditions, $\mathcal{N} = 18_a$ and $\mathcal{N} = 18_b$, see Fig. 10. These configurations may be found by direct search. Furthermore, they may be encoded by indicating the sites which are occupied by rhombi. Thus, in Fig. 11 we have (from left to right beginning from the top): 2,5,8,11,14,17; 0,3,6,9,12,15;

1,4,7,10,13,16; 0,3,6,10,13,16; 0,5,8,9,14,17; 2,3,8,11,12,17; 2,5,6,11,14,15; 1,4,7,9,12,15; 2,3,6,11,12,15; 0,5,6,9,14,15; 0,3,8,9,12,17; 2,3,7,10,13,17; 1,5,6,11,13,16; 0,4,8,10,14,16; 1,4,8,11,12,16; 2,4,7,10,14,15; and 1,5,7,9,13,17. Similarly, in Fig. 12 we have (from left to right beginning from the top): 2,5,8,11,14,17; 0,3,6,9,12,15; 1,4,7,10,13,16; 0,5,8,11,14,16; 0,3,8,10,14,16; 1,4,7,9,12,15; 0,3,6,10,13,16; 1,5,8,9,14,17; 0,4,8,11,12,16; 1,4,8,9,12,17; 2,3,6,10,13,17; 2,3,8,10,14,17; 0,5,6,11,13,16; 1,5,7,9,14,15; 2,3,7,10,14,15; 2,4,8,11,12,17; 2,4,7,11,12,15; 1,5,6,9,13,17; 2,5,6,11,13,17; and 2,5,7,11,14,15.

Next, we explain how we systematically count the spatial configurations of hard rhombi. A necessary condition to get an allowed spatial configuration is to put only one rhombus per unit cell. There are $\mathcal{N}/3$ unit cells, therefore, in the case of $1 < n \leq \mathcal{N}/3$ rhombi, we consider as the first step $\mathcal{C}_{\mathcal{N}/3}^n$ spatial configurations for which each rhombus is assigned to a specific unit cell. Since there are three sites in the unit cell, we consider as the second step 3^n ways to rearrange rhombus inside the unit cells. For each obtained in such a way spatial configuration of n rhombi on the $\mathcal{N}$ -site kagome lattice we verify the hard-core rule: If it holds, then the spatial configuration is accepted. Along these lines we obtain $g_{\mathcal{N}}(n)$ reported in Tables I and II. In Tables V and VI we report the results similar to what is given in Table I, but for other lattice sizes $\mathcal{N} = 21$ and $\mathcal{N} = 12, 24, 27, 36$.

Finally, we explain how we work with the soft rhombi configurations. It is convenient to cast Eq. (13) into

$$\Xi(z, \mathcal{N}) = \sum_{\Sigma_1=0}^{\mathcal{N}} \sum_{\Sigma_2=0}^{2\mathcal{N}} \mathcal{G}(\Sigma_1, \Sigma_2) z^{\Sigma_1} e^{-\frac{V\Sigma_2}{T}}. \quad (\text{B1})$$

To arrive at Eq. (B1), we denote $n_1 + \dots + n_{\mathcal{N}} = \Sigma_1$ and note that Σ_1 may acquire the values $0, 1, \dots, \mathcal{N}$, denote $\sum_{(ij)} n_i n_j = \Sigma_2$ and note that Σ_2 may acquire, in principle, the values $0, 1, \dots, 2\mathcal{N}$, and introduce $\mathcal{G}(\Sigma_1, \Sigma_2)$ which stands for the number of spatial configurations of $n = 0, 1, \dots, \mathcal{N}$ soft rhombi which yields $\Sigma_1 = n$ and Σ_2 [$\mathcal{G}(\Sigma_1, \Sigma_2)$ may be zero for some values of Σ_1, Σ_2]. Clearly, $\sum_{\Sigma_1=0}^{\mathcal{N}} \sum_{\Sigma_2=0}^{2\mathcal{N}} \mathcal{G}(\Sigma_1, \Sigma_2) = 2^{\mathcal{N}}$. In the hard-rhombi limit only the terms with $\Sigma_2 = 0$ in Eq. (B1) survive, $\Sigma_1 = n$ and $\mathcal{G}(\Sigma_1, 0) = g_{\mathcal{N}}(n)$. Furthermore, the number of patterns with one pair of overlapping rhombi is given by $\mathcal{G}(\Sigma_1, 1)$, $\Sigma_1 \geq 2$ (cf. the corresponding row in Table VII and $\mathcal{W}_{1\min} = 36, 396, 1356, 1572, 456$ in Table III). Finding $\mathcal{G}(\Sigma_1, \Sigma_2)$ is the most difficult task. For that we have to find the values of Σ_1 and Σ_2 for all $2^{\mathcal{N}}$ spatial configurations of $n = 0, \dots, \mathcal{N}$ soft rhombi on the $\mathcal{N}$ -site kagome lattice. In Table VII we report the output of such calculations for the case $\mathcal{N} = 18_a$ for illustration. The reported numbers satisfy $\sum_{\Sigma_1=0}^{18} \sum_{\Sigma_2=0}^{36} \mathcal{G}(\Sigma_1, \Sigma_2) = 2^{18}$. They also illustrate a “particle-hole” symmetry: Compare the columns for $\Sigma_1 = 0, \dots, 8$ and for $\Sigma_1 = 18, \dots, 10$ in Table VII.

TABLE VII. Nonzero $\mathcal{G}(\Sigma_1, \Sigma_2)$ for the kagome lattice of $\mathcal{N} = 18_a$ sites, see Eq. (B1). Along the row: $\Sigma_1 = 0, 1, \dots, 18$; along the column: $\Sigma_2 = 0, 1, \dots, 36$.

	0	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
0	1	18	117	336	417	186	17												
1			36	396	1356	1572	456												
2				72	972	3402	3192	444											
3				12	240	2160	6104	3588											
4					75	966	5001	8712	2190										
5						252	2604	8712	7548										
6						30	1004	6672	12048	3580									
7							168	2544	11256	8400									
8							18	1032	6849	14160	2190								
9								108	2880	11400	7548								
10								12	864	7284	12048	444							
11									120	2832	11256	3588							
12									3	844	6849	8712	17						
13										120	2880	8712	456						
14											864	6672	3192						
15											120	2544	6104						
16											3	1032	5001	186					
17												108	2604	1572					
18												12	1004	3402					
19													168	2160					
20													18	966	417				
21														252	1356				
22														30	972				
23															240				
24															75	336			
25																396			
26																72			
27																12			
28																	117		
29																	36		
30																			
31																			
32																		18	
33																			
34																			
35																			
36																			1

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