

# Role of the spin magnitude of the magnetic ion in determining the frustration and low-temperature properties of kagome lattices

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In view of the variety of low-temperature magnetic properties reported recently for kagome lattices with transition-metal ions in different oxidation states, we have investigated the low-energy spectrum and low-temperature thermodynamic properties of antiferromagnetic kagome lattices with varying magnitudes of site spins, employing quantum many-body Heisenberg models. The ground state and the low-lying excitation spectrum are found to depend strongly on the nature of the spin magnitude of the magnetic ions. The system remains highly frustrated if spins are half-odd-integer in magnitude, while the frustration is very weak or almost absent for integer spins or mixed-spin systems. In fact, for a mixed-spin kagome system with a certain magnitude, the whole system behaves as a classical magnet with a ferrimagnetic ground state without any frustration. These theoretical findings are consistent with a few experimental observations recently reported in the literature and would be of value in designing new kagome systems with unusual and interesting low-temperature magnetic properties. © 2005 American Institute of Physics.

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## I. INTRODUCTION

Studies of insulating magnets with geometrical frustration have generated much interest because of their unique low-energy characteristics.<sup>1</sup> Thus, the kagome lattice exhibits magnetic frustration due to the competition of the antiferromagnetic interactions between the neighboring spins.<sup>2</sup> Although the most common examples of frustrated systems are the triangular lattices, the kagome system offers new and interesting possibilities. The kagome lattice has indeed emerged to be a classical case of Heisenberg antiferromagnet which could represent a quantum spin liquid phase. Even the classical spin kagome system exhibits nontrivial ground-state degeneracy with both coplanar and noncoplanar states.<sup>3</sup> The kagome antiferromagnet thus offers the possibility of exploring both classical and quantum spin liquid ground states and the correlated electron problems.

While the triangular lattice has six nearest neighbors and the adjacent triangles share one side or two lattice points, the kagome lattice has four nearest neighbors with the adjacent triangles on the lattice sharing only one lattice point, thereby reducing the restriction from the neighboring spins. For a spin-1/2 kagome lattice, this leads to a quantum disordered ground state with a finite gap to spin excited state together with a large number of spin-zero excitations with little or no gap below the lowest magnetic excited state.<sup>4-6</sup> Interestingly, tunneling between the different quantum ground states is found to occur only for half-odd-integer kagome systems.<sup>7</sup> Despite the considerable volume of research on kagome systems, there has not been a systematic study of the role of magnetic ions in determining the low-temperature thermody-

amic properties. Most of the theoretical attempts are restricted to spin-1/2 kagome lattice, and their magnetic analogs with spin,  $s > 1/2$ , have received little or no attention.<sup>8,9</sup>

On the other hand, on the experimental front, there have been many efforts to synthesize and characterize different types of kagome structures in the last few years. Most of these systems belong to a family of jarosites. Jarosites are the prototype for a spin-frustrated magnetic structure because they are composed exclusively of kagome layers. Since jarosite-type materials are difficult to obtain in a pure form, new synthetic approaches have been employed to obtain kagome compounds. Redox-based techniques have been employed by Nocera *et al.* to prepare single-crystalline kagome samples with an almost 100% coverage of the magnetic ions.<sup>10(c)</sup> The magnetic properties of these materials have been investigated after establishing the structures by x-ray crystallography. It is found that jarosites and analogous materials with Fe<sup>3+</sup> ions exhibit characteristics of frustrated low-temperature antiferromagnetism and occasional long-range antiferromagnetic order in the ground state,<sup>2,10-13</sup> although they are not expected to exhibit such a long-range order. A typical situation is one where a kagome compound exhibits divergence between the zero-field-cooled and field-cooled magnetization data as in a frustrated system; occasionally showing spin-glass-like characteristics. Vanadium (V<sup>3+</sup>) jarosites exhibit an antiferromagnetic ordering of neighboring ferromagnetic kagome layers at low temperatures, characteristic of metamagnetic behavior.<sup>10</sup> Organically templated kagome compounds have been prepared by hydrothermal methods. Thus, an amine-templated mixed-valent iron sulphate with the kagome structure becomes ferrimagnetic at low temperatures, although it shows some evidence for magnetic frustration.<sup>14</sup> Interestingly, an organically tem-

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plated  $\text{Fe}^{2+}$  sulphate with the kagome structure also shows similar properties at low temperatures.<sup>15</sup> A kagome compound of  $\text{Co}^{2+}$ , however, shows magnetic properties comparable to those of the  $\text{Fe}^{3+}$  compounds.<sup>16</sup> The question therefore arises as to the cause for the marked differences in the magnetic properties of the kagome compounds with  $\text{Fe}^{3+}$  and those with  $\text{Fe}^{2+}$ , mixed-valent Fe and  $\text{Co}^{2+}$ , although all of them possess a common structure.

It is well known that frustrations in a magnetic lattice result in the spin-disordered singlet ground state.<sup>6</sup> It is therefore unexpected that kagome systems with frustrations show a long-range magnetic order or a finite ground-state magnetization, as found in few of the kagome systems experimentally.<sup>10,14,15</sup> These studies also reveal that the nature of the ground state depends strongly on the transition-metal species. While the spin-disordered ground state is theoretically predicted for kagome systems with unique spin sites, for the  $S=1/2$  or  $d^1$  configurations,<sup>8</sup> magnetic interactions between two inequivalent magnetic centers mediated by topological frustrations may, however, lead to completely different situations. In the present work, we consider a finite-size kagome cluster with unique and two different types of metal ions in the unit cell. The exchange pathways are restricted only to the single-layer spins with electrons residing in  $3d$  orbitals with localized magnetic moments. In all cases, we assume that the magnetic ions interact antiferromagnetically with their nearest neighbors described by isotropic Heisenberg Hamiltonian.

## II. GROUND-STATE AND LOW-LYING EXCITATIONS

Since the kagome structure shares one spin between two adjacent triangles, as the basic unit, let us consider a two-triangle structure with five spins, where one spin is common to both the triangles. For a general understanding, we consider spins  $s_1$  at the edges of the two triangles and spin  $s_2$  as the corner shared, so that there are four  $s_1$  spins and one  $s_2$  spin in this five-spin system. It is easy to solve this system exactly if we assume that the site spins interact via nearest-neighbor Heisenberg exchange Hamiltonian. We will consider the cases with  $s_1=s_2$  and  $s_1 \neq s_2$  separately within this simple exchange Hamiltonian. For  $s_1=s_2=1/2$ , the ground state is sixfold degenerate, with each doublet configuration of the shared spin giving rise to threefold degenerate states for each triangle. The ground-state magnetization is finite in each lattice point due to strong frustration. For  $s_1=s_2=1$ , the ground state is threefold degenerate and is a singlet. There are three ways one can produce a singlet structure, and in each case, apart from the total magnetization, the magnetization in each of the edges as well as in the corner spin vanishes separately. Next we consider the cases with  $s_1 \neq s_2$ . There are two possibilities; each can be either an integer or a half-odd-integer spin. For  $s_1=1/2$  and  $s_2=1$ , the ground state is a singlet and is threefold degenerate. The states are similar to the  $s_1=s_2=1$  case, although in this case, antiferromagnetic coupling in each edge is between the two spin-1/2 sites. For  $s_1=1/2$  and  $s_2=3/2$ , the ground state is a doublet, with double degeneracy. Both the edges together form a triplet, which couples with the  $S_z=1/2$  component of the spin  $3/2$ .

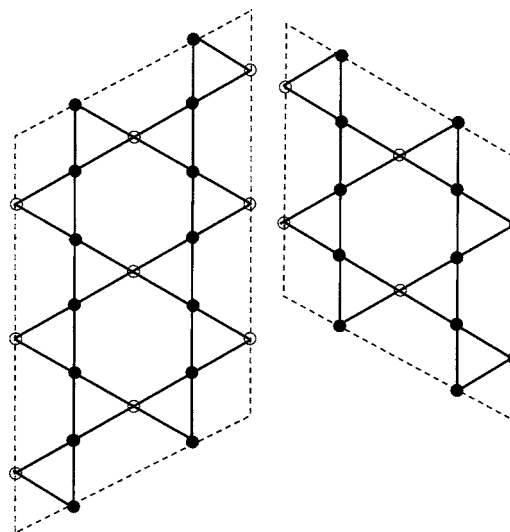


FIG. 1. Schematics of 12-site and 18-site kagome lattices. The solid lines represent the antiferromagnetic exchange interactions,  $J$ . The dashed line defines the periodic boundary conditions. The filled and empty circles represent the site spins  $s_1$  and  $s_2$  respectively.

For  $s_1=1/2$  and  $s_2=2$ , the ground state is a singlet and unique. Each site magnetization is zero, whether it is an edge or a corner-shared spin. For the case when  $s_1=1/2$  and  $s_2=5/2$ , the ground state is a doublet and has the same features as with  $s_1=1/2$  and  $s_2=3/2$ . However, for  $s_1=1/2$  and  $s_2=3$ , the ground state has a finite magnetization and it is a triplet. In the ground state, each of the edge spin-1/2 sites forms a triplet and the corner spin 2 is free with magnetization exactly equals two. This is a ferrimagnetic state, wherein the frustration in the corner-shared structure is completely removed. By increasing the value of  $s_2$  further and keeping  $s_1=1/2$  fixed, we get the same ferrimagnetic state with ground-state spin  $s_G=s_2-4s_1$ .

We have also calculated the ground states with varying  $s_1$  and keeping  $s_2$  fixed at a smaller value. At no  $s_1$  value do we get a ferrimagnetic state. Even when  $s_1$  is very large compared to  $s_2$ , the competing interactions in each triangle give rise to the smallest spin state possible, either a singlet or a doublet. Thus, the key to obtain a ferrimagnet in a frustrated corner-shared triangle system would be to ensure that the corner-shared spin is large compared to the edge spins. As we will show below, this is true even for the highly complicated kagome structure.

For a detailed theoretical study, we have considered 12- and 18-site kagome lattices. They are shown in Fig. 1. Since experimentally there are examples of mixed-spin jarosite systems, we have considered a number of mixed spin together with unique site spin kagome systems. In both cases, the integer as well as the half-odd-integer spins are included. The Hamiltonians correspond to antiferromagnetic exchanges between the spins connected by the solid lines (see Fig. 1). While the filled circles correspond to the  $s_1$  site spin, the empty circles have site spins  $s_2$ . The site spins,  $s_1$  and  $s_2$ , can be the same or different with integer ( $I$ ) or half-odd-integer (HOI) values. Specifically, we consider the following four cases based on the above results for two-triangle systems: (a)  $s_2=$ HOI with  $s_1=s_2$ , (b)  $s_1 \neq s_2$  but  $s_2 < 2s_1$ , (c)

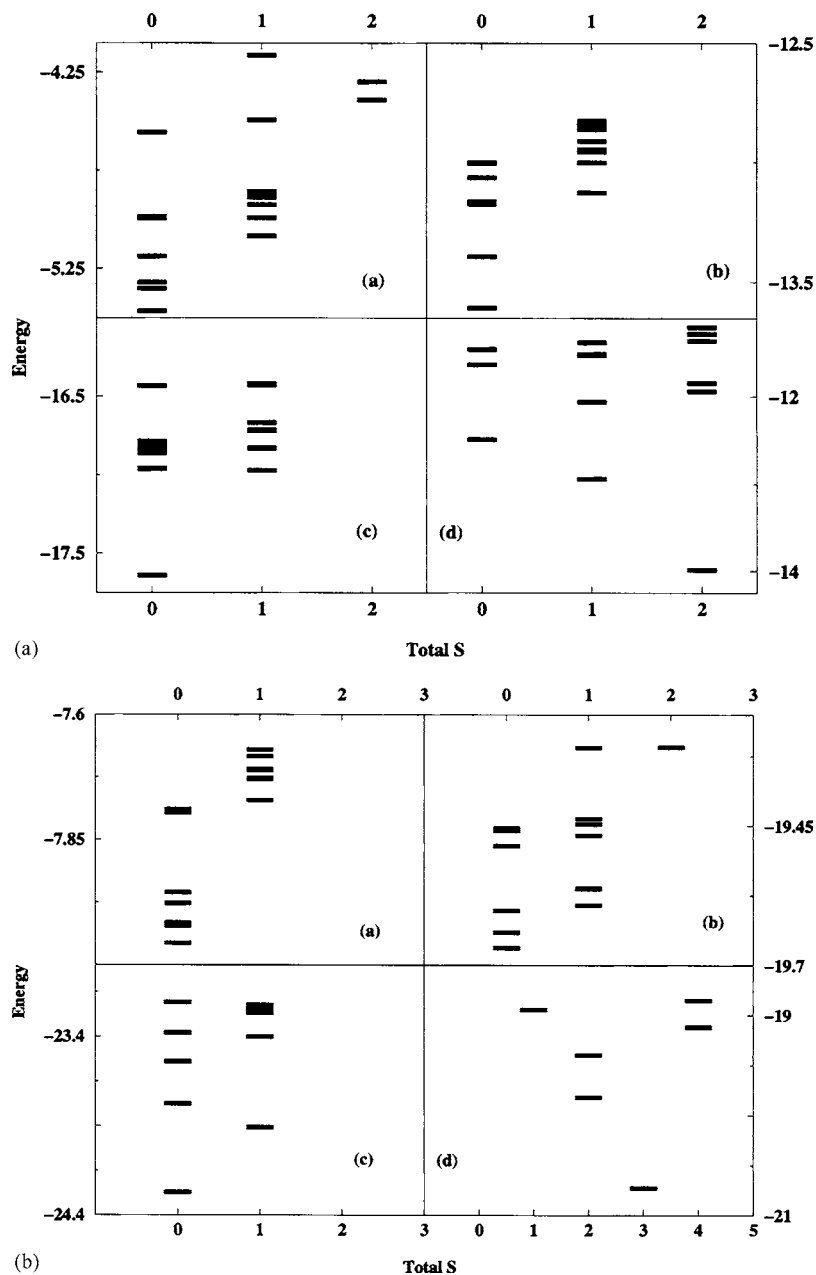


FIG. 2. (a) Energy levels as a function of total spin  $S$ , for the  $(s_1, s_2)$  systems, for a 12-site cluster. (a)  $(1/2, 1/2)$ , (b)  $(1, 1/2)$ , (c)  $(1, 1)$ , and (d)  $(1/2, 3/2)$ . (b) Energy levels as a function of total spin  $S$ , for the  $(s_1, s_2)$  systems, for an 18-site cluster. (a)  $(1/2, 1/2)$ , (b)  $(1, 1/2)$ , (c)  $(1, 1)$ , and (d)  $(1/2, 3/2)$ .

$s_2=1$  with  $s_1=s_2$ , and (d)  $s_2>2s_1$ . The periodic boundary conditions of the finite lattice systems ensure that these are small clusters of infinite two-dimensional kagome structure. Note that our Hamiltonian corresponds to an ideal kagome lattice with localized site spins interacting via isotropic nearest-neighbor superexchange pathways. In real systems, there may exist many other energy scales due to competing exchange pathways, anisotropy, and disorder depending on the specificity of the systems. However, since most of these systems consist of  $3d$  transition-metal ions, often the dominant energy scale is set by the superexchange processes between nearest-neighbor localized moments. In view of this, we believe that our model calculations will serve as a guideline for a general understanding of quantum fluctuation effects in these systems.

We have diagonalized the Hamiltonian matrix corresponding to the nearest-neighbor Heisenberg model for the above four systems with varying site spin  $(s_1, s_2)$  values for

both lattice sizes. Low-energy spectrum in terms of total spin quantum number ( $S$ ) can be obtained with Hamiltonian matrix in a smallest  $s_{\text{tot}}^z$  sector, since every total spin has a projection in the smallest possible  $s_{\text{tot}}^z$  value. For both 12- and 18-site systems, after diagonalizing Hamiltonian matrix in the  $s_{\text{tot}}^z=0$  sector and operating by the  $S^2$  operator on each eigenvector, we obtain the total spin value corresponding to every eigenenergy. In Fig. 2(a), we compare and contrast the energy-level diagram as a function of total spin,  $S$ , for these four cases for the 12-site system. The same for the 18-site system is shown in Fig. 2(b). Although we present results only for some fixed system sizes ( $N=12$  and  $18$ ), for smaller spin values [ $s_1=s_2=1/2$ ,  $s_1=s_2=1$ , and  $(s_1, s_2)=(1/2, 1)$ ] we have verified that the low-energy spectrum remains qualitatively similar even for system sizes up to  $N=21$ . For example, for  $(1/2, 1/2)$  kagome, the ground-state energies per site for  $(N)$  sites are  $-0.454(12)$ ,  $-0.440(15)$ ,  $-0.447(18)$ ,

and  $-0.437(21)$ . Similarly, for  $(1,1)$  and  $(1/2,1)$  they are  $-1.468(12)$  and  $-0.824(12)$ ,  $-1.62(15)$  and  $-0.81(15)$ ,  $-1.34(18)$  and  $-0.801(18)$ , and  $-1.39(21)$  and  $-0.79(21)$ . Interestingly, the ground state energy per site extrapolated to  $N \rightarrow \infty$  for spin-1/2 kagome lattice was reported to be  $-0.434$  by Zeng and Elser.<sup>5(c)</sup> This shows that, for such small system sizes, the low-lying energy states are quite clearly defined, even for the smallest  $(s_1, s_2)$  values.

For cases (a) and (b) [in both Figs. 2(a) and 2(b)], we find that the ground state is a nondegenerate singlet. The lowest excitations are to the singlet states for both cases although there are more numbers of excited singlet states very close to the ground state for case (a) than for case (b). Moreover, there exists a finite gap between the ground and the lowest magnetic states in both cases. In case (c), with  $s_1 = s_2 = I$ , the ground state is a nondegenerate singlet and the lowest excited state is a magnetic triplet state. The low-energy excitation spectrum consists of alternate singlet and triplet states and the system behaves more like a regular one-dimensional antiferromagnetic chain. In fact, the ground state consists of a resonating valence bond (RVB) state with nearest-neighbor spins forming alternating singlet combinations and the lowest excitation involves energy cost (therefore the finite gap) due to disappearance of one of the singlets.<sup>9</sup> The situation becomes completely different for case (d). The ground state in this case is a magnetic state with ground-state spin  $s_2 - 2s_1$  per triangle. This is quite surprising since ferrimagnetic ordering generally does not seem to exist with strong frustration.<sup>17</sup> In the ground state, the site magnetizations for both the spin  $s_1$  and spin  $s_2$  deviate from their ideal values of  $s_1$  and  $s_2$ . However, the reduction occurs in such a way that, effectively, in each triangle the magnetization is  $s_2 - 2s_1$ , the classical Néel configuration value. This implies that there is no frustration-induced quantum fluctuations in any triangles, while it is present in the site spins. In fact, for these systems ( $s_2 > 2s_1$ ), the couplings between  $s_2$  and  $s_1$  spins become strongly antiferromagnetic forcing the  $s_1 - s_1$  couplings to be ferromagnetic, resulting in the ground-state spin  $S_G = s_2 - 2s_1$ . Note that, even for the basic unit cell (five spin system), we found the same nature of the resulting couplings for cases  $s_2 > 4s_1$ . Similar to the low-dimensional mixed-spin ferrimagnet,<sup>18</sup> the lowest excitation in case (d) is to the  $S_G - 1$  state. However, while this excitation is gapless in a one-dimensional mixed-spin ferrimagnet, in our case (d), the lowest excitation to the  $S_G - 1$  state has a gap of  $\approx J$  for any  $(s_1, s_2)$  with  $s_2 > 2s_1$  ( $J$  is the antiferromagnetic exchange constant). Moreover, the next higher excitation in this case is not to the  $S_G + 1$  state, as is the case for the mixed-spin one-dimensional ferrimagnet.<sup>18</sup>

In the light of the above findings, it is useful to recall some of the main experimental observations reported recently for kagome compounds. While the  $\text{Fe}^{3+}$  ( $S=5/2$ ) and  $\text{Cr}^{3+}$  ( $S=3/2$ ) compounds show low-temperature antiferromagnetism, kagome compounds with  $\text{Fe}^{2+}$  ( $S=2$ ) show ferromagnetism.<sup>15,19-21</sup> Interestingly, a  $\text{Co}^{2+}$  ( $S=3/2$ ) kagome compound is found to be similar to the  $\text{Fe}^{3+}$  compound. Although our model Hamiltonian can not be exactly termed as a perfect model for the above experimental systems, the prediction of the model, however, confirms that

within the nearest-neighbor superexchange processes, the variation of magnetic ions plays a dominant role in governing the nature of the low-energy spectrum. Note that partially polarized magnetization or long-range magnetic order in the ground state may arise due to Dzyaloshinsky-Moriya (DM) interactions<sup>22</sup> determined by the magnetic anisotropy-induced canted spin moments within a nonplanar geometry. DM interactions, however, most often depend on the ground-state magnetic structure set by the Heisenberg superexchange interactions, since the DM exchange integral is much smaller compared to the superexchange integral. However, DM-type interactions are generally present in magnetic systems without local inversion centers, which is reported to be the case for  $\text{Fe(III)}$  kagome systems.<sup>21</sup> On the other hand,  $\text{V(III)}$  kagome layer is magnetically polarized due to geometrically favored ferromagnetic exchange couplings.<sup>10</sup> In fact, the interplay between geometric factors (magnetic anisotropy, lattice disorder, and interplanar coupling) and restrictive exchange processes in such a geometry governs the detailed nature of the low-energy physics of kagome systems.

### III. LOW-TEMPERATURE PROPERTIES

Since the low-energy spectrum has a marked difference between the unique spins as well as between mixed-spin kagome systems, it would be interesting to study their low-temperature thermodynamics. Except for case (d) with  $s_2 > 2s_1$ , the ground state of all the other systems is a singlet state. While case (a) has a large number of gapless singlets, this number reduces for case (b), although in both cases singlets are the low-energy states. For case (c), there is a finite gap to the low-energy levels which consists of magnetic as well as nonmagnetic states. Case (d) is a ferrimagnetic case with an energy gap to the state with spin one unit less than the ground-state magnetization. Given such a large variation in the low-energy excitation spectrum, the thermodynamic characteristics are expected to show marked differences.

To calculate the thermodynamic properties of these systems, we set up the Hamiltonian matrices for all the  $s_{\text{tot}}^z$  sectors for each  $(s_1, s_2)$  system. The matrices turn out to be very large for large  $s_1$  and  $s_2$ , even for a 12-site kagome lattice. However, we use the parity symmetry for the  $s_{\text{tot}}^z = 0$  state which has the largest dimension. Matrices for the other  $s_{\text{tot}}^z$  sectors are quite tractable for the 12-site cluster. We can diagonalize these matrices completely to obtain all the eigenvalues in each of the  $s_{\text{tot}}^z$  sectors. Note that we have used proper periodic boundary conditions to minimize the finite-size effects. However, due to the periodic boundary conditions, the Hamiltonian matrices are not very sparse. Below we report the thermodynamic properties of a periodic 12-site kagome cluster for four cases with different site spin magnitudes as discussed above [(a)–(d)].

The canonical partition function  $Z$  for the cyclic system can be written as

$$Z = \sum_j \exp[-\beta(E_j - B(S_{\text{tot}}^z)_j)], \quad (1)$$

where the sum is over all the energy levels of the system in all the  $s_{\text{tot}}^z$  sectors. Here,  $E_j$  is the energy of the state  $j$ ,  $B$  is

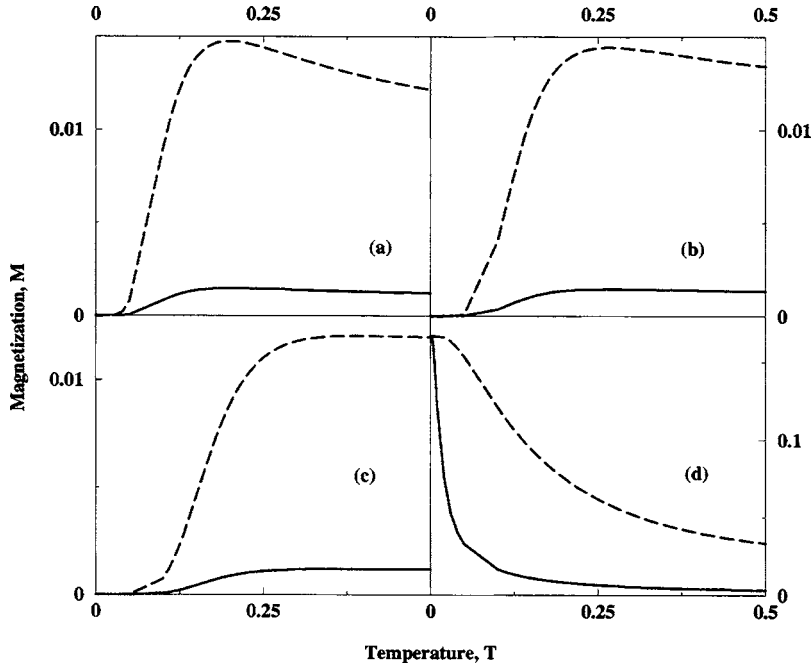


FIG. 3. Magnetization per site vs temperature,  $T$  for (a)  $(1/2, 1/2)$ , (b)  $(1, 1/2)$ , (c)  $(1, 1)$ , and (d)  $(1/2, 3/2)$  systems, with a small magnetic field (solid line) and strong field (dashed line).

the strength of the magnetic field in units of  $J/g\mu_B$  ( $g$  is the gyromagnetic ratio and  $\mu_B$  is the Bohr magneton) along the  $z$  direction, and  $\beta = J/k_B T$  with  $k_B$  and  $T$  being the Boltzmann constant and temperature, respectively. The field-induced magnetization,  $\langle M \rangle$ , can be defined as

$$\langle M \rangle = \frac{\sum_j (S_{\text{tot}j}^z) \exp[-\beta(E_j - B(S_{\text{tot}j}^z))]}{Z}. \quad (2)$$

The magnetic susceptibility,  $\chi$ , can then be related to the fluctuation in magnetization,

$$\chi = \beta[\langle M^2 \rangle - \langle M \rangle^2]. \quad (3)$$

Similarly the specific heat,  $C$ , is related to the fluctuation in energy as given by

$$C = \frac{\beta}{T}[\langle E^2 \rangle - \langle E \rangle^2]. \quad (4)$$

We present the dependence of the magnetization on temperature for different magnetic-field strengths for all the four systems in Fig. 3. Apart from the case (d), in the other three systems, the magnetization at low magnetic field shows a small increase at low temperatures and shows paramagnetic behavior at high temperatures. However, as the field strength is increased, the magnetization shows a large increase with temperature at low temperatures. This is because, as the field strength is increased, the gap to magnetic states with nonzero spin decreases as the Zeeman coupling to these states becomes large. Since for both cases (a) and (b) there are low-energy singlet excited states, accessing the states with finite magnetization requires finite  $k_B T$ . In terms of the excited magnetic states, both the systems have similar energy scales and the magnetization therefore is more or less similar. In case (c), the triplet is the lowest excitation with a finite gap and so the initial increase in magnetization is similar to cases (a) and (b). However, for the mixed-spin system case (d), with a ferrimagnetic ground state, the magnetization is non-

zero at zero temperature and decreases as the temperature is increased, with a larger decrease at a higher field strength. This behavior can be understood from the type of spin excitations present in the system. The lowest-energy excitation at low magnetic fields is to a state with spin  $s$  less than  $s_G$ . Therefore, the magnetization initially decreases at low temperatures.

The dependence of magnetic susceptibility on temperature for different field strengths is shown in Fig. 4, for all the four cases. Except for case (d), all the other case systems have zero magnetic susceptibility at zero temperature and zero magnetic field, since the ground state is a singlet for these cases. The application of a magnetic field opens up a gap and the susceptibility goes to zero as  $T \rightarrow 0$  in all the cases. For a zero magnetic field, however, as the temperature is increased, the susceptibility increases rapidly. For case (a), the increase is linear at low  $T$ , but for cases (b) and (c) the increase is smaller and higher, respectively, than linear in  $T$ . This rise crucially depends on the magnetic states that are accessible at energy corresponding to the  $k_B T$  values. For cases (a) and (b), the susceptibility has two slopes at low temperatures. The slow rise is due to their energy-level structures wherein in between the magnetic triplet state, there exist a number of singlet levels. This number is larger for case (a) than for case (b). For the case (c), however, the low-energy levels consist of magnetic triplet states at small energy intervals and thus the rise in  $\chi$  is quite sharp. With a magnetic field,  $\chi$  rises and in each cases (a)–(c), the low  $T$  behavior reflects the respective position of the lowest-energy magnetic level. All the  $\chi$  curves with and without magnetic field should meet at the same point at high temperatures, since the high-temperature behavior of  $\chi$  should be independent of field strength and should saturate to the Curie-law value (not shown in the figure).

For a ferrimagnet at zero field, the zero-temperature value of  $\chi$  is infinite in the thermodynamic limit and it is finite and equal to the fluctuation in magnetization divided

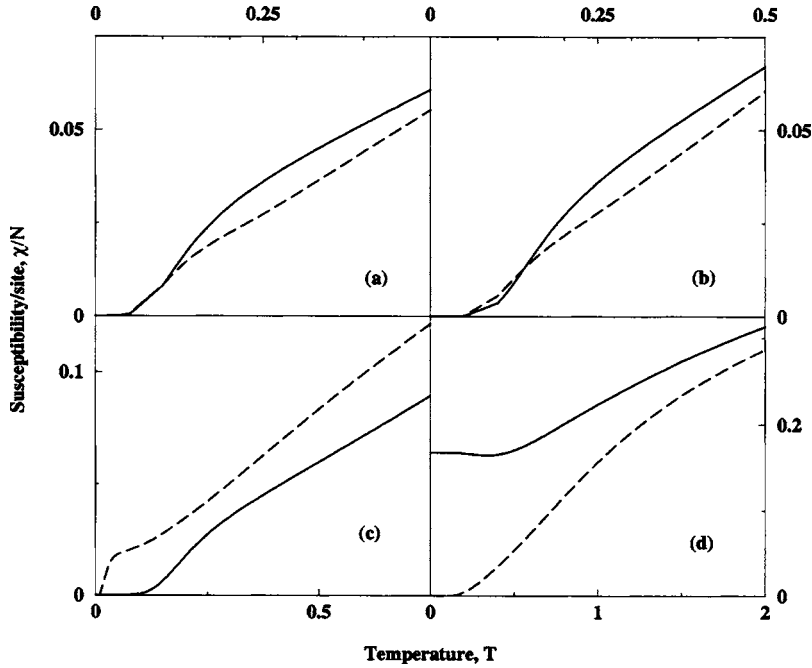


FIG. 4. Magnetic susceptibility per site vs temperature,  $T$  for (a)  $(1/2, 1/2)$ , (b)  $(1, 1/2)$ , (c)  $(1, 1)$ , and (d)  $(1/2, 3/2)$  systems, with a zero field (solid line) and with a small field (dashed line).

by the temperature for a finite system. As the temperature increases, the magnetization value decreases. This is because at low temperatures, the states with  $s_{\text{tot}}^z < s_G$  get populated. With an increase in  $T$ , the states with high spin get populated and  $\chi$  rises rapidly. Note that the low-temperature zero-field behavior of our system is quite different from the ferromagnetic system. In the latter, the spin wave analysis shows that the  $\chi$  increases as  $1/T^2$  at low temperatures.<sup>23</sup>

In a finite field, the behavior of  $\chi$  is quite different [Fig. 3(d)]. The  $\chi$  decays exponentially to zero for temperatures less than the gap due to the applied magnetic field. However, above a certain  $T$ , the finite field  $\chi$  follows the same path as that of zero-field  $\chi$ , since the states accessed at that temperature are the same with and without magnetic field. At high temperatures, the magnetic susceptibility corresponds to the Curie-law value for the  $(s_1, s_2)$  system.

The temperature dependence of the  $C/T$ , where  $C$  is the specific heat per site, is shown in Fig. 5, for all the four systems. For case (a), the  $C/T$  shows a broad maxima with a humplike structure due to the presence of nonmagnetic (spin-zero) low-energy states above the ground state. The initial rise is nearly exponential since the singlets are almost gapless even for this small system size. For case (b), although there are low-energy singlet states, the  $C/T$  shows a peaked structure with a well-rounded maxima, since the singlets do not form a well-defined excitation spectrum in this case as in case (a). For case (c), the  $C/T$  is zero for a range of small  $T$  values as there are no states available in this small temperature range. It also shows a peaked structure, as generally observed for a regular quantum antiferromagnet with or without weak frustration. On the other hand, for case (d), the rise in  $C/T$  as a function of temperature is slow at low  $T$ , and

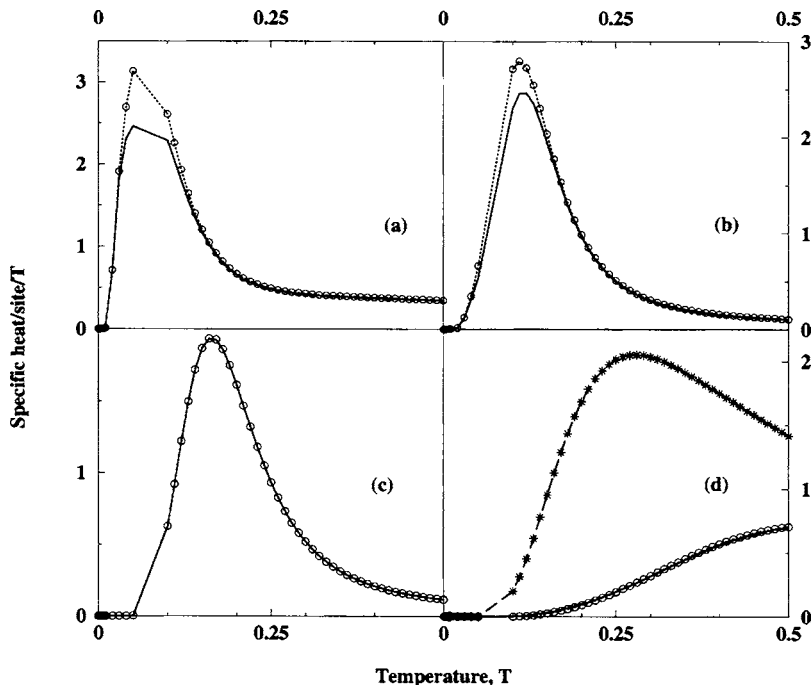


FIG. 5. Specific heat per site divided by temperature ( $C/T$ ) as a function of  $T$  for (a)  $(1/2, 1/2)$ , (b)  $(1, 1/2)$ , (c)  $(1, 1)$ , and (d)  $(1/2, 3/2)$  systems, with a zero field (solid line), with a small field (circles), and a very strong field (stars).

increases steadily with an increase in  $T$  without any maximum. For a finite but small magnetic field, there is a small increase in the hump/peak height for cases (a) and (b), at about the same temperature corresponding to the zero magnetic field, although the qualitative dependence is still the same. Note that, in the presence of a weak magnetic field, since the change in  $C/T$  curve is large in case (a), it amounts to a large change in magnetic contribution to the entropy ( $S$ ). Interestingly, this proves that the ground state for spin-1/2 kagome is a disordered spin liquid state. For cases (c) and (d), however, there is no change in the specific-heat structure at a small magnetic field. Only at a very high magnetic field does the  $C/T$  curve show a broad rounded maximum in case (d). This indicates that the higher-energy high-spin states are not accessible to within  $k_B T$  of the ground state at this small magnetic-field strength for these cases. For the system with a ferrimagnetic ground state it requires a strong magnetic field for polarization.

#### IV. SUMMARY

To conclude, we have analyzed the low-energy magnetic spectrum of the antiferromagnetic kagome lattice with variations in site spins extensively using quantum many-body formalism. The studies show that the role of the spin magnitudes of the magnetic ions is profound, giving rise to a variety of ground- and excited-state magnetic configurations. The system remains highly frustrated for half-odd-integer (HOI) spin systems. For an integer spin on the other hand, the frustration is very weak or nearly absent. For mixed-spin systems with  $s_2 > 2s_1$ , even though the site spins are quantum spins, the whole system behaves as a classical magnet with a ferrimagnetic ground state. While the  $s_1$  spins interact antiferromagnetically with strong quantum fluctuations, the competitive nature of interactions between the  $s_1$  spins and the  $s_2$  spins removes frustration in  $s_2$  if  $s_1 \neq s_2$  and completely of the whole lattice if  $s_2 > 2s_1$ . We conjecture that the key to generate a strong, weak, or zero frustration in kagome systems depends mainly on the magnitude and the relative position of the site spin within the nearest-neighbor Heisenberg model. Interestingly, experimental studies to date<sup>10,14–16,19</sup> show that ferri- or ferromagnetic order manifests itself at low temperatures only in those kagome compounds where the transition-metal ion has an integral spin. Although the results described here are applicable for a specialized isotropic uniform kagome lattice, it points towards the experimental challenges in realizing a number of ground states and overcoming barriers with a proper choice of synthetic methodology.

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