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Effects of competing interactions on low-energy characteristics of a spin-1/2 cubic cluster

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Abstract

We consider a simple cubic magnetic cluster with spin-1/2 ions at its eight corners. The superexchange Hamiltonian employed in this paper involves nearest, next-nearest (nn), and next to next-nearest (nnn) neighbour interactions. These competing exchange interactions are found to generate a number of phases with ground states: (a) ferromagnetic; (b) nn-neighbour dimers; (c) regular antiferromagnetic; (d) nnn-neighbour dimers; and (e) nnn-neighbour forming triplets. We have also found fully polarized and partially polarized lowest spin excitations in the broken symmetry phases. The effects of the low-energy characteristics in various phases have been quantified by computing thermodynamic properties, such as magnetization and susceptibility.

1. Introduction

In recent years, the discovery that magnetic clusters such as Mn_{12} and Fe_8 show resonant magnetization tunnelling and quantum interference has paved the way for extensive research in magnetic clusters [1]. In an applied magnetic field the magnetization curves form hysteresis loops with unique 'staircase'-like structures at the transition points [2,3]. These clusters show finite magnetization at zero temperature in the ground state, which is essential to account for such quantum tunnelling. The tunnelling rate obtained experimentally depends on the applied magnetic field and the temperature. The clusters which are used for experimental studies have varying sizes. They could be dimers (two magnetic ions) or very large magnetic blocks, with negligible inter-cluster interactions [4]. Thus the net magnetization depends on the magnitude of the spin at each site, and the possible interaction pathways between the neighbouring spins. The purely isotropic antiferromagnetic Heisenberg one-dimensional spin system has a singlet ground state. However, depending on the coordination number of each spin, and the nature of the exchange interactions, a variety of quantum phases, such as ferromagnetic and frustrated nonmagnetic, can be generated.

These phases have been investigated extensively in the last three decades, since the Haldane conjecture, which differentiates between integer and half-odd-integer (hoi) spin chains [5]. While the hoi spin chains show quasi-long-range order (QLRO) and gapless excitations, the integer spin chains have short-range spin–liquid ground states with gapped excitations. On the other hand, competing exchange interactions in hoi spin chains introduce frustrations [6]. With the inclusion of the next-nearest (nn) neighbour antiferromagnetic exchange interactions (J_2), the system remains gapless (with ground-state QLRO) until a critical $J_{2c} = 0.2411$, but above this J_2, the system becomes continuously dimerized [7, 8]. These results have been experimentally verified [9], and are for the chains with antiferromagntic exchanges. When the nearest-neighbour (J_1) and J_2 exchanges have opposite signs, the ground state can be either antiferromagnetic (singlet state) or ferromagnetic (fully polarized), depending on the magnitude of $\alpha = J_2/J_1$ [10].

Due to the fact that inter-cluster hopping is negligible in magnetic clusters, the magnetization of the entire system is dependent only on the individual cluster magnetizations which in turn depend on the exchange interactions among the neighbouring spins. Thus it would be interesting to study the effects of competing interactions on molecular clusters. Although there have been a large number of studies on frustrated extended systems, frustrated clusters are relatively new and, to our knowledge, have not been explored seriously [11,12]. In this paper, we consider a simple cubic spin-1/2 cluster. In general, the exchange pathways are complex, and so the nature (sign and magnitude) of the exchange parameters are difficult to predict *a priori*. Clusters with high symmetry and small size are relatively easy to study, and hence a detailed insight into the energy spectrum can be obtained with a variety of exchange parameters, both in terms of sign and magnitude.

In the next section, we introduce the model Hamiltonian for a cubic spin-1/2 magnetic cluster which is used in our study. In section 3, we present the results of our computations of the ground state and low-lying excitations of the cubic system. We also calculate the thermodynamic properties such as magnetization and susceptibility at certain exchange parameter values where the system shows interesting low-energy characteristics. We conclude the paper with a summary of all our results.

2. Model Hamiltonian

We consider a simple cubic cluster with spin-1/2 ions at its eight corners. A sketch of this cluster is shown in figure 1. The Hamiltonian for this system can be explicitly written as

$$H = J_1 H_1 + J_2 H_2 + J_3 H_3 \tag{1}$$

where

$$\begin{aligned} H_1 &= \vec{S}_1 \cdot \vec{S}_2 + \vec{S}_1 \cdot \vec{S}_4 + \vec{S}_1 \cdot \vec{S}_6 + \vec{S}_2 \cdot \vec{S}_3 + \vec{S}_2 \cdot \vec{S}_5 + \vec{S}_3 \cdot \vec{S}_4 \\ &\quad + \vec{S}_3 \cdot \vec{S}_8 + \vec{S}_4 \cdot \vec{S}_7 + \vec{S}_5 \cdot \vec{S}_6 + \vec{S}_7 \cdot \vec{S}_8 + \vec{S}_5 \cdot \vec{S}_8 + \vec{S}_6 \cdot \vec{S}_7 \\ H_2 &= \vec{S}_1 \cdot \vec{S}_3 + \vec{S}_1 \cdot \vec{S}_5 + \vec{S}_1 \cdot \vec{S}_7 + \vec{S}_2 \cdot \vec{S}_4 + \vec{S}_2 \cdot \vec{S}_6 + \vec{S}_2 \cdot \vec{S}_8 \\ &\quad + \vec{S}_3 \cdot \vec{S}_5 + \vec{S}_3 \cdot \vec{S}_7 + \vec{S}_4 \cdot \vec{S}_6 + \vec{S}_4 \cdot \vec{S}_8 + \vec{S}_5 \cdot \vec{S}_7 + \vec{S}_6 \cdot \vec{S}_8 \\ H_3 &= \vec{S}_1 \cdot \vec{S}_8 + \vec{S}_2 \cdot \vec{S}_7 + \vec{S}_3 \cdot \vec{S}_6 + \vec{S}_4 \cdot \vec{S}_5. \end{aligned} \tag{2}$$

In the above expression, J_1, J_2 and J_3 are the nearest, next-nearest (nn) and next to next-nearest (nnn) neighbour interactions, respectively. $\vec{S}_i$ is the spin vector located at site i. We have considered the case where $S_i = 1/2$. The total Hamiltonian (H) commutes with the total spin operator, $S^2 = (\sum_{i=1}^{8} \vec{S}_i)^2$, and the z-component of the total spin, S^z_{tot}. However, as the size of the system is small (eight sites), we have used only S^z_{tot} as a good quantum number. For the cubic cluster, out of the total 256 basis states, 70 and 56 states are found to be in $S^z_{\mathrm{tot}} = 0$ and 1 subspaces, respectively.

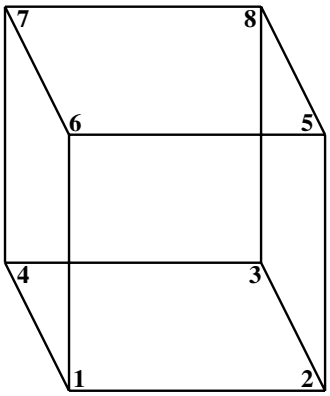

Figure 1. Schematic diagram of a cubic cluster.

To solve this Hamiltonian, we have used the exact diagonalization scheme. Since we are dealing with a relatively small number of basis states in each $S^z_{\rm tot}$ subspace, we can obtain all the eigenvalues and eigenfunctions (which are a linear combination of the basis states). The standard Davidson algorithm has been used to diagonalize the full Hamiltonian. To correctly assign the total spin quantum numbers to each of the $S^z_{\rm tot}$ states, we compute the eigenvectors of all the states in $S^z_{\rm tot} = 0$ subspace, and operate by the S^2 operator. To determine the phase of the ground state, we have computed equal time two-spin correlation functions. Due to the high symmetry of the cubic cluster, there are only three unique two-spin correlation functions, namely, $\langle \vec{S}_1 \cdot \vec{S}_2 \rangle$, $\langle \vec{S}_1 \cdot \vec{S}_3 \rangle$ and $\langle \vec{S}_1 \cdot \vec{S}_8 \rangle$.

3. Results and discussion

3.1. Ground state and low-lying excitations

We have computed the properties of the cubic cluster in the ground and excited states for a range of exchange parameter values. All three exchanges (J_1, J_2, J_3) can in principle be varied. However, we set one of these parameters, J_1, to be either +1 or −1, so that we can now vary only two parameters, namely, the ratios $\alpha = J_2/J_1$ and $\gamma = J_3/J_1$. In most of the limiting cases, exact results can be obtained. For example, $\gamma = 0$ and α positive (negative) ∞, with J_1 positive, results in a decoupled system with antiferromagnetic (ferromagnetic) couplings only between the nn neighbours. The interest, however, is in the realistic parameter regime where these phases or other new phases appear[1].

Let us first analyse the system with varying α, setting $\gamma = 0$. There are four cases, which we consider separately: (i) J_1 positive and α positive; (ii) J_1 positive, α negative; (iii) J_1 negative, α positive; and (iv) J_1 negative, α negative. For all the above mentioned cases $|J_1| = 1$.

Before we proceed with the above cases, let us consider the Hamiltonian when $\gamma = 0$. With this condition, the total Hamiltonian can be written as

$$H = \pm(\vec{S}_{1357} \cdot \vec{S}_{2468} - H_3 + \alpha/2(S^2_{1357} + S^2_{2468}) - 3\alpha) \tag{3}$$

where $\vec{S}_{ijkl} = (\vec{S}_i + \vec{S}_j + \vec{S}_k + \vec{S}_l)$, and $S^2_{ijkl} = S_{ijkl}(S_{ijkl} + 1)$. The first term in the above equation consists of nearest-neighbour and nnn-neighbour interaction terms. The H_3 term is to eliminate the extra nnn-neighbour interaction terms. The third term takes into account the

[1] The word 'phase' is used only for convenience, to distinguish regions with different two spin–spin correlation functions. Our model has no *true* phase transitions in its proper sense.

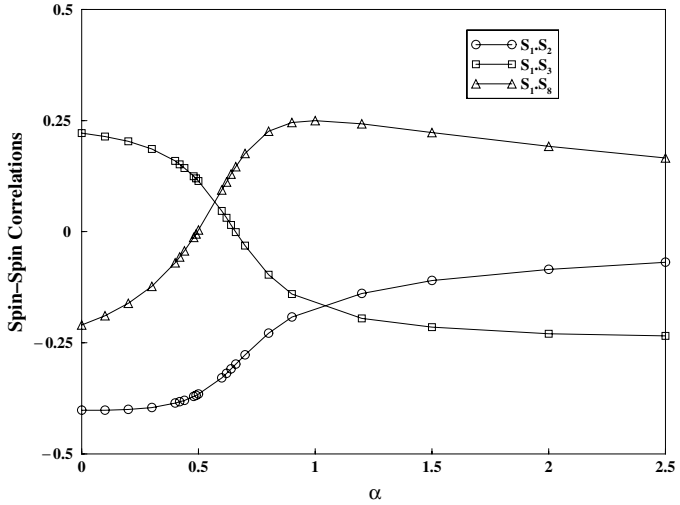

Figure 2. Nearest-neighbour $\langle S_1 S_2 \rangle$, nn-neighbour $\langle S_1 S_3 \rangle$, and nnn-neighbour $\langle S_1 S_8 \rangle$ correlation functions as a function of α.

nn-neighbour interactions, with an additional $\frac{1}{2}\sum_i S_i^2$, which is 3 for an eight site spin-1/2 cube. The $\pm$ sign is to take into account the sign of J_1.

We consider case (i) with both J_1 and α antiferromagnetic (positive). Then, for $\alpha = 1$, the above Hamiltonian (equation (3)) simplifies to $H = -(H_3 + 3)$. We have considered a singlet ground state as is the case for an antiferromagnet. Thus, in this case, the Hamiltonian of the system depends only on the nnn-neighbour interactions (H_3). For a minimum energy configuration, H_3 should be a positive maximum. H_3 contains four interaction terms, and so the maximum positive value would be 1, if each term contributes its largest possible value, 1/4. This condition emphasizes that each of the nnn neighbours—(1, 8), (2, 7), (3, 6), (4, 5)—form triplets for $\alpha = 1$ with positive J_1, while the entire system remains in the singlet ground state. We plot the spin–spin correlations for case (i), as a function of α in figure 2. As can be seen from the figure, $\langle \vec{S}_1 \cdot \vec{S}_2 \rangle$ correlations are always antiferromagnetic, but the correlation strength decreases with the increase in α. $\langle \vec{S}_1 \cdot \vec{S}_3 \rangle$ is ferromagnetic at $\alpha = 0$ and continues to remain so until $\alpha = 2/3$, where it becomes antiferromagnetic. $\langle \vec{S}_1 \cdot \vec{S}_8 \rangle$ correlation is -0.21 at $\alpha = 0$, but changes sign at $\alpha = 1/2$. At $\alpha = 1$, $\langle \vec{S}_1 \cdot \vec{S}_8 \rangle$ shows exact triplet correlations, 1/4, as predicted above. The spin gap is found to increase with the increase in α.

Case (ii) does not lend itself to an extended study because the ferromagnetic interactions between the nn neighbours increases the probability of antiparallel spin orientations of the nearest-neighbour spins.

The most interesting case occurs when J_1 is negative (cases (iii) and (iv)). In this case, for positive α, the system remains in a ferromagnetic ground state. The Hamiltonian for the $\alpha = 1$ point can be analysed with the help of only four two-point correlations, since at this point the Hamiltonian turns out to be $H = -7 + H_3$ (ground-state energy is -6). However, as α becomes negative, the system enters into an antiferromagnet ground state. This phase transition occurs precisely (up to fourth decimal place) at $\alpha = -1/3$.

The above transition is shown in figure 3. The spin–spin correlations, $\langle \vec{S}_1 \cdot \vec{S}_2 \rangle$, $\langle \vec{S}_1 \cdot \vec{S}_3 \rangle$ and $\langle \vec{S}_1 \cdot \vec{S}_8 \rangle$, as a function of α, indicate that all three correlations are 1/4 when $\alpha \geqslant -1/3$. In the antiferromagnetic phase, the spin–spin correlations show interesting behaviour as a function of $|\alpha|$. We find that $\langle \vec{S}_1 \cdot \vec{S}_2 \rangle$ decays slowly to zero, while the other two correlations

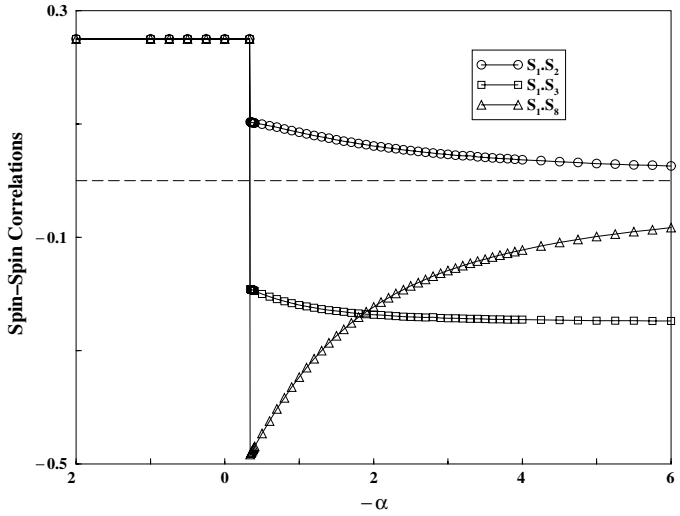

Figure 3. α-dependence of the correlation functions, with $J_1 < 0$.

become negative immediately as $|\alpha|$ increases from 1/3. $\langle \vec{S}_1 \cdot \vec{S}_8 \rangle$ eventually tends to zero, as we increase $|\alpha|$. $\langle \vec{S}_1 \cdot \vec{S}_3 \rangle$ approaches a finite limiting value, $-1/4$, as $|\alpha|$ tends to ∞. This limiting nn-neighbour correlation corresponds to an equal contribution from each interaction in a triangular structure formed by J_2 interactions. The ground state of a triangle is degenerate with a pseudo-spin symmetry corresponding to the clockwise and anticlockwise directions. This phase corresponds to a broken symmetry dimerized phase, wherein the nn neighbours form singlet dimers.

So far we have considered the cases with $J_3 = 0$. The situation becomes very interesting as we turn on γ. With nonzero γ (and $J_1 < 0$), the Hamiltonian (see equations (1) and (3)) immediately becomes

$$H = -\vec{S}_{1357} \cdot \vec{S}_{2468} + (1-\gamma)H_3 - \alpha/2(S^2_{1357} + S^2_{2468}) + 3\alpha. \tag{4}$$

In terms of the total spin operator, the above Hamiltonian simplifies to

$$H = -1/2S^2 - (\alpha - 1)/2(S^2_{1357} + S^2_{2468}) + (1-\gamma)H_3 + 3\alpha. \tag{5}$$

Let us analyse the situation with $\alpha = 1$ (which has a ferromagnetic ground state at $\gamma = 0$). In this limit, with nonzero γ, the Hamiltonian reduces to

$$H = -1/2S^2 + (1-\gamma)H_3 + 3. \tag{6}$$

In the ferromagnetic ground state, the total energy is $-7 + (1-\gamma)$, as $\langle H_3 \rangle = 1$ in a ferromagnet. We can expect a phase transition from this fully polarized state to a nonmagnetic state. To obtain the transition point, we equate the Hamiltonian for two different phases: the ferromagnetic phase with $H = -6 - \gamma$, and the antiferromagnetic phase for which $H = (1-\gamma)H_3 + 3$. Along the $\alpha = 1$ line, the Hamiltonian depends only on H_3. The lowest-energy antiferromagnetic configuration occurs when $H_3 = -3$. Equating these two equations, we obtain the transition point to be at $\gamma = -1.5$. Below this transition point, $\gamma < -1.5, \alpha = 1$, the ground state corresponds to a dimerized phase where nnn neigbours form singlet dimers. The lowest excitation from this antiferromagnetic phase is not to a triplet state, but rather to a fully polarized ferromagnetic state. However, the triplet excitation (one of nnn-neighbour singlet forming triplet) stabilizes after a certain γ. This γ is obtained by equating the triplet

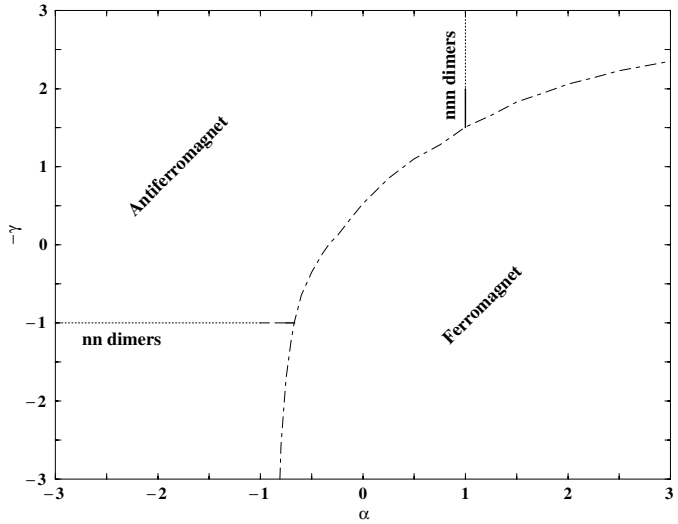

Figure 4. $J_1 < 0$ phase diagram in α–γ plane. The dot-dashed curve divides the ferromagnetic and nonmagnetic ground-state phases. The other two lines represent phases with exact broken symmetry singlet ground state, with the lowest spin excitation: ferromagnetic (solid line), quintet (long dashed) and triplet (dot).

excitation with the ferromagnetic state, i.e. $-1 + (1 - \gamma)(-2) + 3 = -7 + (1 - \gamma)$, which readily gives $\gamma = -2$. In the phase diagram (figure 4), we have marked this with the dotted line. Note that the ground state is same all along the line $\alpha = 1$ ($\gamma < -1.5$), however the low-lying excitations above and below $\gamma = -2$ are completely different. This difference is reflected in the low-temperature behaviour, outlined in the next section.

Another exact point with a broken symmetry phase can be obtained by setting $\gamma = 1$. This condition reduces the Hamiltonian to

$$H = -1/2S^2 - (\alpha - 1)/2[S^2_{1357} + S^2_{2468}] + 3\alpha. \tag{7}$$

In this case the Hamiltonian depends only on the nn-neighbour interactions and not on H_3. Along the $\gamma = 1$ line, the total energy for a ferromagnetic ground state is $-10 - (\alpha - 1)6 + 3\alpha$. On the other hand, the lowest energy for an antiferromagnetic state is when nn neighbours form singlet dimers, such that $S^2_{1357} = S^2_{2468} = 0$. Equating the energy expressions for these two states, $-4 - 3\alpha = 3\alpha$, we obtain the transition to be at $\alpha = -2/3$. In the antiferromagnetic phase, the ground state is exact with nn neighbours forming singlets. Each of the four-spin terms in the square brackets in equation (7) form a tetrahedral configuration, which can accommodate only two singlets. The lowest energy for the $S = 1$ configuration (with one of the singlets forming a triplet) is $-1 - (\alpha - 1) + 3\alpha$. Quartet ($S = 2$) formation from two triplets in any one of the two tetrahedra (with singlets in other tetrahedra) costs the same energy, $-3 - 2(\alpha - 1) + 3\alpha$, as that of one triplet from each tetrahedra. Equating the energy expressions for the ferromagnetic, $S = 1$ and 2 states, we obtain lowest-energy excitations: (i) ferromagnetic for $-3/4 < \alpha < -2/3$, (ii) $S = 2$ when $-1 < \alpha \leqslant -3/4$, and (iii) $S = 1$ for $\alpha \leqslant -1$. Thus, in the $\gamma = 1$ line, although the ground state is antiferromagnetic, there are three different lowest spin excitations: (i) ferromagnetic; (ii) $S = 1$; and (iii) $S = 2$. In figure 4, we have shown these excitation characteristics in the $\gamma = 1$ line.

These transitions are exact only along the lines, $\alpha = 1$ and $\gamma = 1$, since in these cases the Hamiltonian is found to depend only on the nnn neighbours and nn neighbours, respectively.

Also, these transitions can be visualized from the limiting values: nnn-neighbour dimers for $\gamma \to -\infty$, and nn-neighbour dimers for $\alpha \to -\infty$. However, in the realistic regime, for other values of α and γ, we obtain the ground-state phases numerically. We have obtained the boundary lines dividing the ferromagnetic, nnn-neighbour dimers, nn-neighbour dimers and the regular antiferromagnetic phases. Figure 4 shows the full phase diagram, with J_1 negative. Note that, even for $J_1 > 0$, these dimer ground states can be obtained by equating the corresponding Hamiltonian expressions, similarly as discussed for $J_1 < 0$.

3.2. Thermodynamic properties

In this section we present the thermodynamic properties of the cubic cluster at various points in the parameter-space phase diagram. We have computed the magnetization and susceptibility as a function of the temperature T and an applied magnetic field B [13, 14].

The canonical partition function, Z, at a temperature, T, and for an external magnetic field, B, can be written as

$$Z = \sum_k \mathrm{e}^{-\beta(E_k - BS^z_{\mathrm{tot},k})} \tag{8}$$

where the sum is over all the energy states of the cubic cluster in all S^z_{tot} sectors. E_k and $S^z_{\mathrm{tot},k}$ denote the energy and z-component of the total spin respectively, of the state k. B is the magnetic field strength in units of $J_1/g\mu_{\mathrm{B}}$, (g is the gyromagnetic ratio and μ_{B} is the Bohr magneton) along the z-direction, and $\beta = 1/k_{\mathrm{B}}T$, with k_{B} and T being the Boltzmann constant and temperature respectively. The field-induced magnetization $\langle M \rangle$ can be written as

$$\langle M \rangle = \frac{\sum_k S^z_{\mathrm{tot},k} \mathrm{e}^{-\beta(E_k - BS^z_{\mathrm{tot},k})}}{Z}. \tag{9}$$

The magnetic susceptibility, defined as $\delta\langle M \rangle/\delta B$, is related to the fluctuation in magnetization

$$\chi = \frac{\beta[\langle M^2 \rangle - \langle M \rangle^2]}{T} \tag{10}$$

where

$$\langle M^2 \rangle = \frac{\sum_k (S^z_{\mathrm{tot},k})^2 \mathrm{e}^{-\beta(E_k - BS^z_{\mathrm{tot},k})}}{Z}. \tag{11}$$

We present our calculations of the thermodynamic properties in four different phases, namely

(a) ferromagnetic ground state at $\alpha = 0$ and $\gamma = 0$;
(b) nnn-neighbour dimer ground state with the lowest excitation (i) ferromagnetic at $\alpha = 1$ and $\gamma = -1.75$, and (ii) triplet at $\alpha = 1$ and $\gamma = -2.5$;
(c) regular antiferromagnetic ground state at $\alpha = -1$ and $\gamma = -1$;
(d) nn-neighbour dimer ground state with lowest excitation (i) ferromagnetic at $\gamma = 1$ and $\alpha = -0.7$, (ii) quartet at $\gamma = 1$ and $\alpha = -0.9$, and (iii) triplet at $\gamma = 1$ and $\alpha = -1.25$.

We have considered these points since these are the representatives of various ground-state and low-lying excitation characteristics possible in the full phase diagram shown in figure 4.

In the absence of an external magnetic field, the magnetization of the system is zero at all temperatures, since the spins are randomly oriented. An applied magnetic field polarizes the system producing a finite magnetization either at zero or higher temperatures. The dependence of magnetizations on temperature for a magnetic field strength of $B = 0.01/g\mu_{\mathrm{B}}$ are shown in figure 5, for four different cases as mentioned above. For the ferromagnetic ground state (case (a)), the magnetization decreases rapidly from $1/2$, tending to zero at higher temperatures.

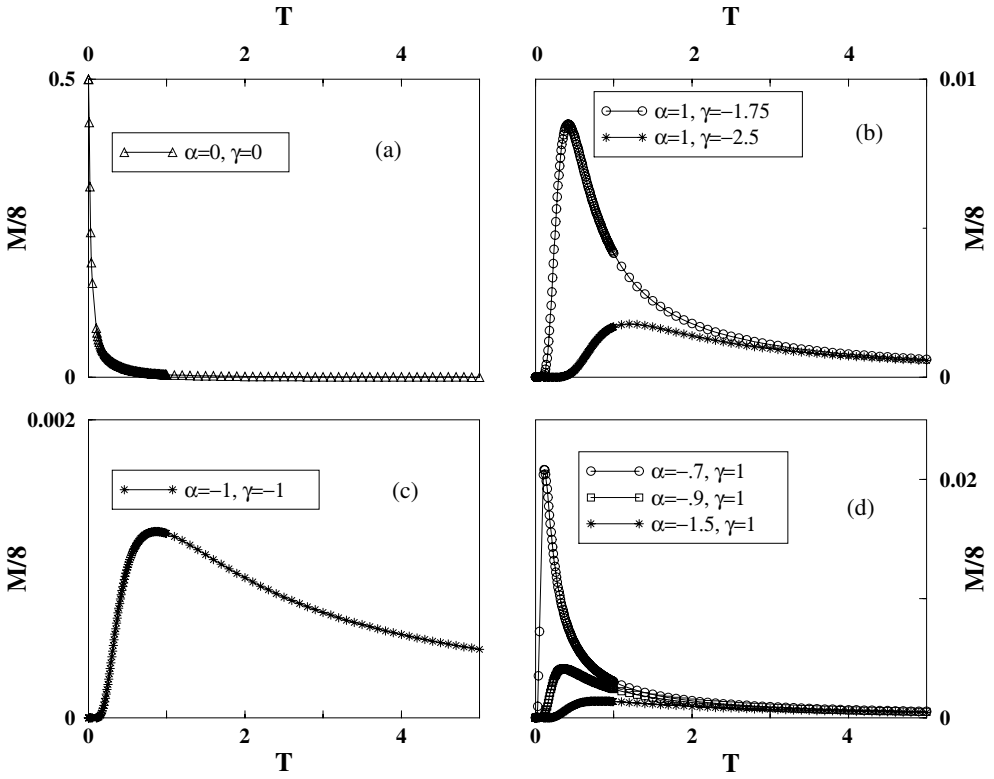

Figure 5. Magnetizations per site as a function of temperature, for seven different sets of parameters, defined in the text.

This is due to the effect of thermal vibrations, which destroy the alignment of the S_z components at high temperatures. For the $S = 0$ ground states (cases (b)–(d) in figure 5) the magnetization is zero at zero temperature, increases to a maximum at a characteristic temperature, but decreases with a further increase in temperature. The specific behaviour of these magnetization curves can be analysed with the help of the spin excitations present in the system. In cases (b,i) and (d,i), the lowest spin excitation at low temperature is to the highest spin state possible for the system ($S = 4$). Therefore, initially, the magnetization shows an abrupt increase, signalling that the spin excitation at this temperature is to the highest spin state. The subsequent drop in the magnetization is due to the higher-energy low-magnetization spin states (triplets etc) accessed with the increase in temperature. For other cases ((b,ii), (d,ii), (d,iii) and (c)), as the lowest spin excitation is to either a triplet, or a quintet state, the initial increase is comparatively smoother, but nonetheless depends explicitly on the excited state spin multiplets. At high temperatures in all cases, the magnetization averages out to be zero, but the temperature at which this paramagnetic behaviour sets in varies for different parameters.

The temperature dependence of χT per site in zero magnetic field is shown in figure 6, for the same parameter sets mentioned above. In the absence of an external magnetic field, the zero temperature value of χT is equal to the average of the square of the magnetization in the ground state. For the ferromagnetic ground state ($S_G = 4$), $\chi T/N$, as $T \to 0$, is given by $S_G(S_G + 1)/3N$, which is 0.833 for this system. As the temperature increases, the product, χT, decreases because at higher temperatures, the states with $S < S_G$ become populated.

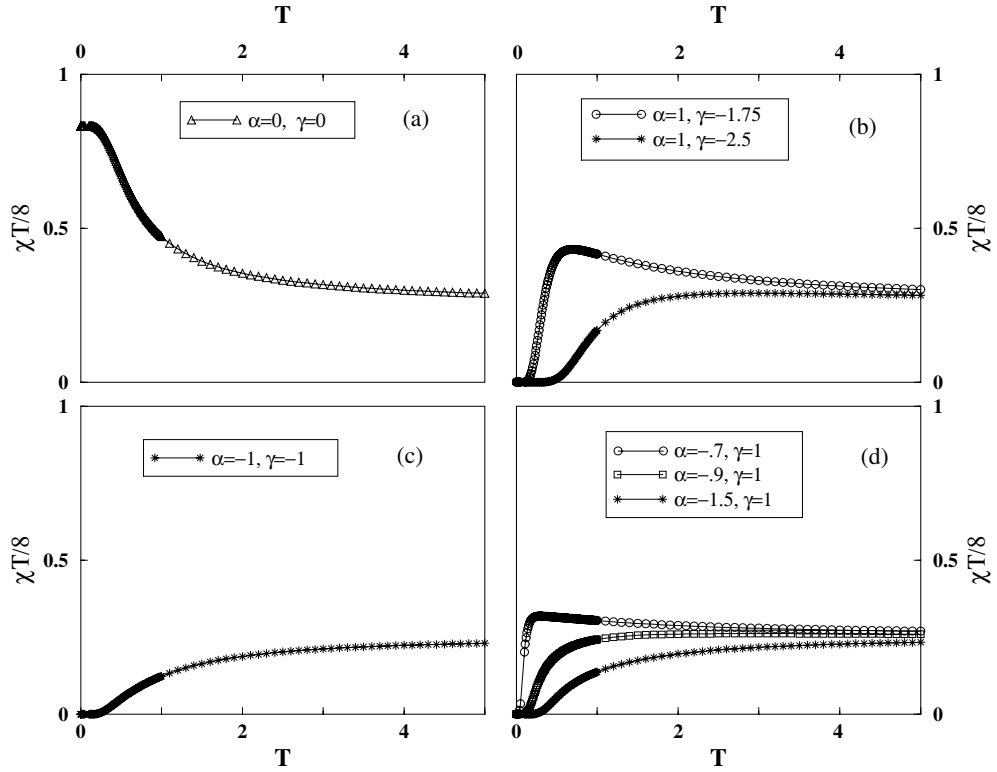

Figure 6. Temperature dependence of the product χT per site, for seven different sets of parameters, as in figure 5.

For cases with singlet ground state, the χT product is zero at zero temperature. However, at finite but low temperature ($T_{\min}$), the states with higher S values are accessed. This value of $T_{\min}$, however, depends on the lowest energy gap, and so is different for different parameters. The magnitude of the initial increment is directly proportional to the average of the square of the magnetization of the lowest excited spin state. The dependence of χT on temperature above $T_{\min}$ can be explained by the properties of higher-energy spin states which, in cases (b,i) and (d,i) ((b,ii), (c), (d,ii) and (d,iii)) have lower (higher) magnetizations than the lowest excited state. However, at very high temperatures, the spins are completely decoupled and the paramagnetic $\chi T/N$ product is $1/4$, which is the limiting value in each of the cases.

To summarize, we have considered a simple cubic spin-$1/2$ cluster, which undergoes various phase transformations subject to the changes in exchange pathways. There are as many as five different phases with ground state: (i) ferromagnetic; (ii) nnn-neighbour dimers; (iii) regular antiferromagnetic; (iv) nn-neighbour dimers; and (v) each of the nnn-neighbour forming triplets. The low-temperature thermodynamic properties such as susceptibility and magnetization indicate the transition between different spin states, quantifying the role of various low-lying excitations with the same ground state, at a number of points in the two-dimensional parameter space phase diagram.

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