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Spin-phonon induced magnetic order in magnetized Spin Ice systems

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Abstract. We study the behavior of spin ice pyrochlore systems above the well known [111] 1/3 plateau, under slight deviations of the direction of the external field. We model the relevant degrees of freedom by Ising spins on the kagome lattice. We propose the inclusion of lattice deformations, which imply phononic degrees of freedom in the adiabatic limit. We use analytical calculations to estimate how these new degrees of freedom affect the short and long range spin interactions in the presence of an external magnetic field. We then obtain the magnetization curves, explore the phases and the ground states of this system in the presence of magnetic field by Monte Carlo simulations. We discuss comparisons with experimental results.

1. Introduction

Spin ice systems are one of the most studied experimental realizations of magnetic frustration. Their ground state is highly degenerate and they present an effective finite entropy. Materials such as $Ho_2Ti_2O_7$ [1, 2] and $Dy_2Ti_2O_7$ [3, 4] have the magnetic rare earth ions forming a tetrahedral corner sharing lattice, the so called pyrochlore lattice. The intense crystal fields in this materials force their magnetic moments in the direction towards the center of the tetrahedron. Therefore it is possible to model them as local Ising variables. Due to frustration, the stable configuration is two magnetic moments pointing inside the tetrahedron, and two pointing out (the ice rule "2in2out"). This is analogous to the relative position of hydrogen and oxygen in water ice [5], thus the name spin ice.

Several models have been considered for this materials: nearest neighbor interaction, inclusion of long-range dipolar interactions (which lifts the degeneracy but keeps the 2in2out configuration) and a modified dipolar model that considers exchange interactions up to third nearest neighbors [6].

The response of this materials to a magnetic field along the [111] direction is well known. The [111] direction is parallel to one of the spins of the tetrahedron, the so-called "apical" spin. Before saturation, there is an intermediate plateau at magnetization 1/3 [7–10] where the apical spins are aligned with the external magnetic field and the spins still obey the ice rules. Since the apicals are fixed and the remaining degrees of freedom are in the kagome layers, this is often referred to as kagomé ice, where in each kagome cells two magnetic moments must be *in* and one *out* (the apical spin is already *out*, fulfilling the ice rules).

Experiments [11] have studied the magnetization of $Dy_2Ti_2O_7$ upon slight deviations of the magnetic field from the [111] direction. Results show an intermediate peak in the susceptibility between the 1/3 plateau and saturation when the field is tilted towards the [112] direction. This feature is absent when the inclination is towards the [110] direction.

We have already presented a toy model [12] with the idea of studying lattice deformations in the kagome plane in order to introduce new degrees of freedom that could reproduce the above mentioned features. In this work, we aim for a more realistic description and introduce lattice deformations in a kagome lattice where spins are in the same direction as in the pyrochlore lattice. An external magnetic field orthogonal to the kagome plane mimicks the field in the [111] direction. Deviations from this direction are obtained introducing in-plane components of the field.

2. Effective Model

We study a kagome plane with local Ising spins $\vec{S}_i = S\vec{s}_i = S\sigma_i\hat{e}_i$, where S is the spin magnitude, $\sigma_i = \pm 1$ (+1 indicating *in* and -1 *out*) and $\hat{e}_i$ are the local directions out of the plane towards the center of the tetrahedron formed by apical spins in the pyrochlore lattice. This means that $\vec{s}_i \cdot \vec{s}_j = -\frac{1}{3}\sigma_i\sigma_j$ and that for nearest neighbours $\vec{s}_i \cdot \hat{e}_{ij} = \pm\sqrt{\frac{2}{3}}$.

We consider nearest neighbor exchange interactions and long range dipolar interactions, as in the following Hamiltonian:

$$H = \sum_{\langle ij \rangle} J(r_{ij})\vec{s}_i \cdot \vec{s}_j + Da^3\frac{1}{2}\sum_{i\neq j}\frac{\vec{s}_i \cdot \vec{s}_j}{r_{ij}^3} - 3\frac{(\vec{s}_i \cdot \hat{r}_{ij})(\vec{s}_j \cdot \hat{r}_{ij})}{r_{ij}^3} - \vec{h}\sum_i \vec{s}_i \tag{1}$$

J_0 is the nearest neighour exchange interaction, D the dipolar constant, a the distance between nearest neighbors, $\vec{h}$ the external magnetic field, r_{ij} the distance between spins at sites i and j (i.e. $r_{ij} = |\vec{r}_j - \vec{r}_i|$) and $\hat{r}_{ij}$ is the unit vector that points from site i to site j.

We follow the procedure presented in [12]: we introduce first order lattice deformations in each site as $\vec{r}_i \approx \vec{r}_i^{\,0} + \vec{u}_i$. We calculated how this changes the exchange and dipolar terms of the Hamiltonian doing a first order Taylor expansion. We treat deformations in the standard Einstein phonon model [13], taking deformations with elastic constant K, noting that bond phonon model [14] leads to trivial results in this case.

As in [12], these deformations can be integrated out as Gaussian function, provided it be sharp enough, which gives rise to effective interactions. Since we only consider deformations for nearest neighbour bonds, we get these effective interactions up to third-nearest neighbours. We include dipolar interactions up to that order and get an effective third-nearest neighbour Ising model under a magnetic field, shown below

$$H_{\text{eff}} = J_0\left(j_1^{eff}\sum_{\langle i,j\rangle}\sigma_i\sigma_j + j_2^{eff}\sum_{\langle\langle i,j\rangle\rangle}\sigma_i\sigma_j + j_{3d}^{eff}\sum_{\langle\langle\langle i,j\rangle\rangle\rangle}\sigma_i\sigma_j + j_{3\ not\ d}^{eff}\sum_{\langle\langle\langle i,j\rangle\rangle\rangle}\sigma_i\sigma_j - \vec{h}\cdot\sum_i \vec{s}_i\right) \tag{2}$$

where

$$j_1^{eff} = \left(-\frac{1}{3} + \frac{5}{3}d - \delta\right);\ j_2^{eff} = \left(\frac{-d}{3\sqrt{3}} + \delta\right);\ j_{3d}^{eff} = \frac{d}{8};\ j_{3\ not\ d}^{eff} = \frac{-d}{8} + 2\delta \tag{3}$$

where δ is the contribution arising from the fluctuation of the phonons, $\delta = \frac{1}{4k}\left(\frac{\lambda}{3} - 5d\right)^2$, with $d = \frac{D}{J_0}$, $k = \frac{Ka^2}{J_0}$, $\lambda = \frac{\alpha}{J_0}$. Notice that as in [12] lattice deformations in nearest neighbor bonds produce only effective third neighbor couplings along the bonds and not across the hexagon diagonals. The expression for δ and the values of the couplings differ from [12] due to the fact that the spins have an out of plane component.

For our simulations, we use for $Di_2Ti_2O_7$, $d = D/J_0 = 1.44K/3.72K = 0.38$ [15] and adimensionalize choosing $J_0 = 1$. To get from (2) to Kelvin, one must simply multiply by $J_0 = 3.72K$. For example, in our simulations we chose $\delta = 0,\ 0.01,\ 0.02...$, which in Kelvin are $\delta = 0,\ 0.0372\text{K},\ 0.0744\text{K}....$ This non zero values of delta, calculating $\lambda \approx 20$ from [16], imply $k \sim 500, 250$. This value of k fullfills the requirement of a sharp Gaussian in the integration of the phonon variables.

3. Monte Carlo Simulations and Discussion

We simulated arrays with $3L^2$ sites, where L^2 is the number of cells, $L = 8, 12, 24$. We used a simple Monte Carlo algorithm combined with the annealing technique, lowering the temperature as $T_{i+1} = 0.9T_i$, doing 40 steps from $T_0 = 0.3(1.116\text{K})$ up to $T_f = 0.00443426(0.016\text{K})$.

We chose δ from 0 to 0.1 in steps of 0.01, and introduced an external magnetic field $\vec{h}$ fixing the h_z (perpendicular to the kagome plane, [111] direction) component and calculating the h_y component in the kagome plane ([11$\bar{2}$] direction) as $h_y = \tan\theta h_z$, where θ is deviation from the [111] direction. Positive θ means that the h_y component of the field favours that the spin that is in this direction is *out*, as the [112] direction. We worked with both positive and negative θ, mimicking inclination of the magnetic field in [111] towards [112] and [110].

We calculated the magnetization per site of the tetrahedron associated to the kagome cell in the $\hat{z}$ direction, $\langle M_z\rangle$ as $\langle M_z\rangle = \frac{1+\frac{1}{3}\sum_i \sigma_i}{4}$.

Figures 1 and 2 show the magnetization $\langle M_z\rangle$ as a function of h for different values of the parameter of the deformations, δ, and three different inclinations of the magnetic field ($\theta = 0°, 5°, 10°$). In both cases, the system shows a magnetization plateau at 1/3 (where we checked that the configuration satisfies the 2in1out rule). At $\delta = 0$ (no deformations) there is no intermediate plateau. Such a plateau appears at $\langle M_z\rangle = 2/3$ for $\theta > 0$ and $\delta > 0$. This feature is not seen for negative values of θ. This behaviour holds for $\delta \leq 0.04$. For larger δ, more intermediate plateaux appear, which are not compatible with the experimental results mentioned before. We point out that at $\delta > 0.04$, $j_{3\ not\ d}^{eff}$ becomes positive.

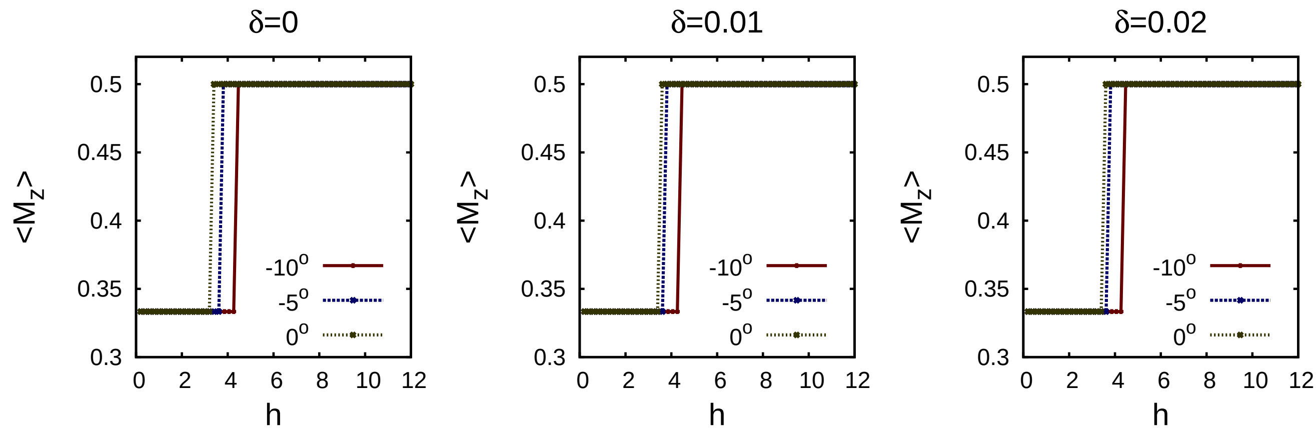

Figure 1. $\langle M_z\rangle$ as a function of h for $\delta = 0, 0.01, 0.02$ and $\theta \leq 0$ at the lowest simulated temperature ($T_f = 0.00443426 \equiv 0.016\text{K}$) for $L = 12$.

The experimental results in [11] do not include magnetization curves. However, they do include susceptibility data. For the [110] inclination, which corresponds in our work with negative θ, there is a single peak in the susceptibility presented in figure 3(a) of [11], the peak where there is a spin flip to the three-in, one-out state in pyrochlore. Our work is consistent with

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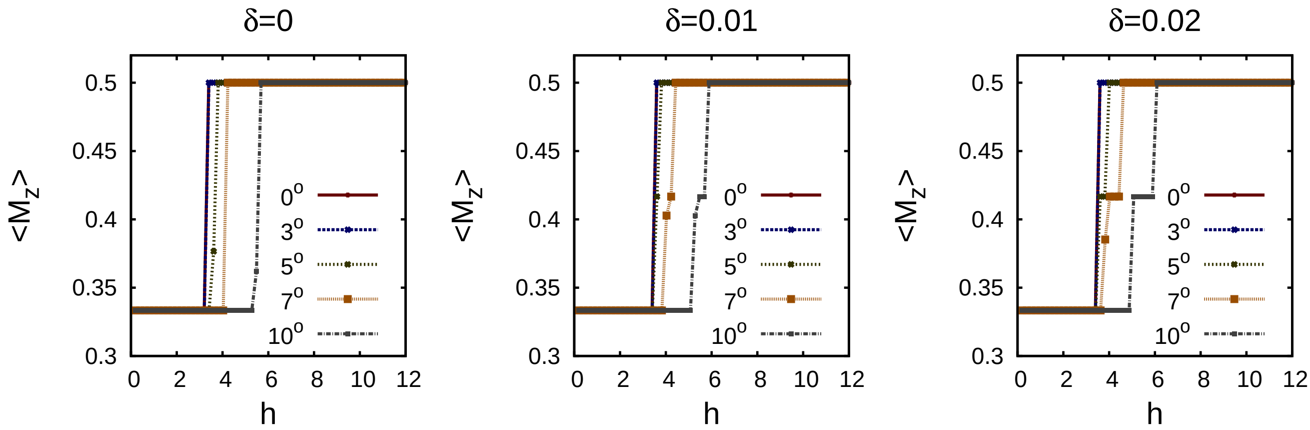

Figure 2. $\langle M_z\rangle$ as a function of h for $\delta = 0, 0.01, 0.02$ and $\theta \geq 0$ at the lowest simulated temperature ($T_f = 0.00443426 \equiv 0.016$K) for $L = 12$.

this: for negative θ there is no intermediate plateau before saturation, even when deformations are considered.

For $\theta > 0$, to compare with these experimental results, we plot the susceptibility as a function of h and three values of δ at a higher simulated temperature, $T = 0.06176734(0.23\text{K})$, in figure 3. It can be seen that without deformations ($\delta = 0$), there is only one peak, corresponding to saturation. This peak simply appears at a larger value of the magnetic field h as the inclination increases. This does not match the results from [11], where it is found that when the magnetic field is inclined in the [112] direction, the single peak is washed out, an in fact at larger inclination angles two peaks can be distinguished.

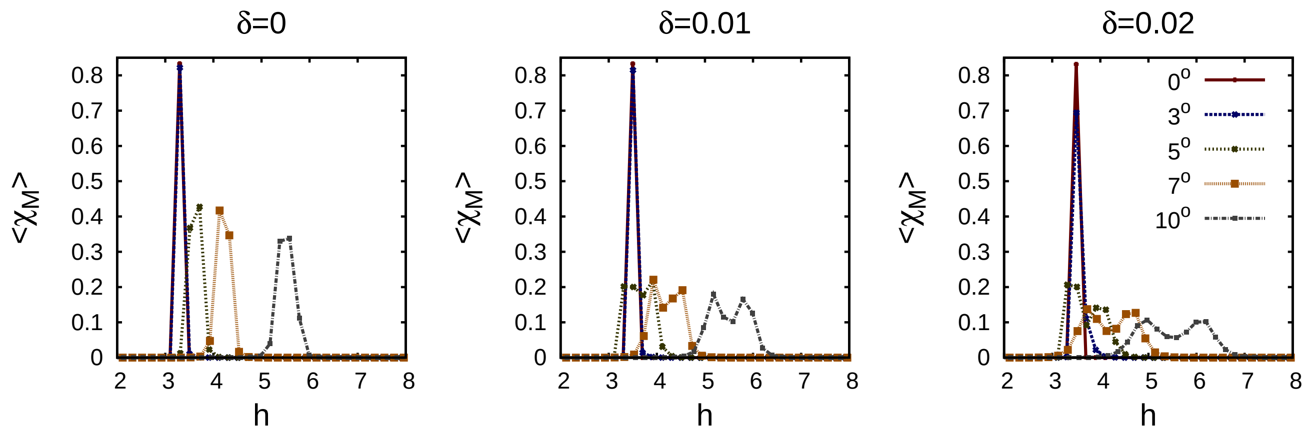

Figure 3. Susceptibility $\chi = \frac{\partial M_z}{\partial h}$ vs h for $L = 12$, $\delta = 0, 0.01, 0.02$ f, $\theta \geq 0$ at a higher temperature $T = 0.06176734 \equiv 0.23$K, to compare with figure 3 of [11]

In our work, as deformations are included, the single saturation peak becomes less defined. The middle panel of figure 3 shows that for the higher inclinations there seem to be two peaks. This is more clear in the right panel of figure 3, which correspons to a larger contribution from the deformations. Two peaks can be identified for $\theta = 7°, 10°$. Notice that the position of the first peak does not change with θ as much as that of the second peak. This is the same

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behaviour shown in figure 3(b) from [11], which only shows data up to $\theta = 7°$. It remains to be seen experimentally if these two peaks are observed at larger θ.

4. Outlook

Figure 3 suggests that the addition of elastic degrees of freedom, which change the magnetic moment couplings in a specific way, induce a second peak in the susceptibility when the magnetic field is slightly tilted from the direction perpendicular to the kagome plane. This describes quite well the experimental results in [11].

Encouraged by this fact, one should care about the role of phonons in spin ice at zero magnetization. In a possible scenario they could partially lift the large ground state degeneracy and explain the low temperature residual entropy.

Further work is being done in collaboration with Prof. Santiago Grigera and Prof. Rodolfo Borzi including simulations in pyrochlore and new experiments.

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