Spin-Dot interactions in Artificial Spin Ice: population inversion as an entropic effect

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Abstract – In the present Letter we discuss the origin of the vertex population inversion observed experimentally in the mediated Artificial Square Ice [46]. An interaction modifier is a disc-shaped magnetic nanoisland which is placed at the center of a vertex to mediate the interaction between the nearby islands. We show that the inversion is of entropic origin, and can be explained via the renormalization of the vertex configuration energies due to local interaction between the nanoislands and the dot. We show in a simple model with mixed Heisenberg and Ising spins (a spin-dot interaction) that as a function of the island size, entropic effects become important. Because of the renormalization of the vertex energies, we observe a level crossing between Type I and Type II vertices which is similar to the observed experimental results. We also discuss possible implications of spin-dot interactions in the eight- and sixteen-vertex models phase diagrams.

Introduction. – The last years have seen the use of a variety of interacting magnetic nanostructures [1–3] in different geometries, and the introduction of artificial materials whose behavior is similar to the one of spin ice. Today, because of the experimental understanding of these materials, the interest is shifting from reproducing the behavior of known statistical physics models, to novel ones. In particular, various new phenomena have been investigated [4, 10–13], ranging from topological order in materials [11, 20], memory in materials [21, 22], disordered systems and slow relaxation [23], novel resistive switching [24, 25], and embedding logic circuits in the magnetic substrate just to mention a few. The level of manipulation obtained of each island is remarkable [1, 16, 31–33], which now suggests the study of new types of interactions for novel models [34, 35].

A well known example of possible application of artificial spin ice (ASI) is the possibility of having monopole like charges in spin ices without a string tension [36–42]. In the case of ASI [2], however, the key problem is that nanoislands have an asymmetric interaction, due to the dipolar nature of the exchange couplings. In the approximation of nearest-neighbor only dipolar-like interactions, the energy of an artificial square ice can be approximated by an energy associated to the interaction between parallel and perpendicular islands $\epsilon_\perp > \epsilon_\parallel > 0$, which are associated with the distance between the islands [43].

Let us now briefly explain how the energy landscape is obtained below the Curie temperature $T_c$. Magnetic islands acquire an Ising-like spin because of the typical geometrical elongation of the nanoislands. The artificial square ice is thus described by the following Hamiltonian based on Ising-like variables, but lying on the plane:

$$H_{ASI} = -\sum_v \left[ \epsilon_\parallel \sum_{\langle i,j \rangle_v} s_x s_x + \sum_{\langle i,j \rangle_v} s_y s_y' + \epsilon_\perp \sum_{\langle i,j \rangle_v} s_x s_y' \right]$$

(1)

where $s_- = s_x \hat{x}$ and $s_\parallel = s_y \hat{y}$. Alternatively, one can describe these type of models in terms of the four type of vertex configurations (their energies) only, as in Fig. 1. For the square spin ice, it has been noted that vertices have four increasing energies parametrized by $\epsilon_\perp$ and $\epsilon_\parallel$, with a nomenclature Type I, ..., Type IV respectively. The vertex energies are $\epsilon_I = -4\epsilon_\perp + 2\epsilon_\parallel$, $\epsilon_{II} = -2\epsilon_\parallel$, $\epsilon_{III} = 0$, $\epsilon_{IV} = 4\epsilon_\perp + 2\epsilon_\parallel$, where $\epsilon_I < \epsilon_{II} < \epsilon_{III} < \epsilon_{IV}$. The vertex population in the ground state is determined by this energy hierarchy. In units of the temperature for $\kappa = 1$, we can use $\epsilon_\perp \approx 0.38675$ and $\epsilon_\parallel \approx 0.2735$ for realistic phase diagrams, as noted in [44]. Following the discussion above, in order to have...
monopole-like excitations freely to move in the material, it would be desirable that vertices of Type I and Type II have the same energy. Recently, ways to overcome this difficulty have been proposed in the literature, using for instance 3-d materials (by raising two of the four islands at a vertex) or via intermediate interactions. In the case of the recent paper, it has been proposed to introduce a “dot”, a disc-shaped island at each vertex of an artificial square ice, also called interaction modifier. In this paper we focus on this type of mediated interactions.

Interestingly, it has been experimentally observed that as the size of the dot island increases, the low temperature population of each vertex configuration in the ground state can swap, which we report in Fig. 2. When the size of the dot-island increases, vertices of Type I invert the population with vertices of Type II. It has thus been observed that there is an optimal dot size such that the energy of the two configurations are the same. We introduce a simple model to explain this phenomenology, based on the idea that the dot island “renormalizes” the energy configuration of the vertices.

Spin-Dot as an Ising-Heisenberg interaction. –

The presence of the island at each vertex changes the energy landscape of the model. We propose to describe this type of interaction with the introduction of extra degrees of freedom at each vertex in the standard approach to understand Artificial Square Ice.

The disc island lacks any breaking of the horizontal and vertical symmetry. We thus find it is reasonable to assume that below the Curie temperature such island acquires a two dimensional Heisenberg type spin, as shown in Fig. 3. Since a disc island sits at each vertex, we suggest to consider the additional interaction between the ASI nanoislands with the dot as mixed spin-Heisenberg interaction of the type

\[
H_{sd} = J \sum_v \left( \sum_{\langle i,j \rangle_v} \mathbf{s}_i \cdot \mathbf{\sigma}_j \right),
\]

where \(\mathbf{\sigma}_j\) is now a two dimensional Heisenberg spin, \(\sigma = \cos \theta \hat{x} + \sin \theta \hat{y}\). Here, the energy \(J\) effectively can be ascribed to the physical dimension of the dot island. These type of models are known in the literature, and various type of decorations and mappings are possible for arbitrary lattice configurations. However, in the present paper we will see that integrating out the local degrees of freedom will be sufficient to explain the observed phenomenology.

The total Hamiltonian is thus given by \(H = H_{ASI} + H_{sd}\) which we now study. The advantage of using the Hamiltonian above is that the spin-Heisenberg interaction can be exactly integrated out. The partition function is thus now generalized by a

\[
Z = \sum_{\{n_k=\pm1\}} \prod_k w_n^{n_k},
\]

with \(\beta\) representing the inverse temperature. Since the interaction in \(H_{sd}\) are between two different type of spins, the interaction graph is bipartite, and thus we can integrate out the degrees of freedom of the disc at each vertex. This fact can also be seen by noticing that the energy is the sum of elements on the vertices only. Because of this, the partition function can be written as \(Z = \sum_{\{n_k=0\}} \prod_k w_n^{n_k} \), where for \(J = 0\), \(w_j = e^{-\beta \epsilon_j}\), and \(N\) is consistent with the constraints. Because of the spin-dot interaction, however the corrected weights can be written as \(w_n^c = w_n v_j(\beta)\), where we now study the entropic contribution \(v_j\).

The spin-dot interactions are interesting on their own, as we have a mixing between Ising and Heisenberg spins,
and some comments on this matter will be made at the end of this Letter. First, we note that the integral over each dot island can be performed exactly. In fact, the lattice is bipartite, which implies that integrating away the dot island implies an effective interaction between the nano-islands. As in the case of the one dimensional model, we can now write

\[ Z_{sd} = \int_{0}^{2\pi} d\theta v e^{-\beta H_{sd}(\theta_x)} = \int_{0}^{2\pi} d\theta v e^{\beta J(\theta_x + \theta_y) \cos \theta + \beta J(\theta_x + \theta_y) \sin \theta} = 2\pi I_0(\beta J \sqrt{4 + 2\eta(s_x + 2s_u s_d)}) \] (4)

where \( I_0(x) \) is the Modified Bessel function of the first kind. We see that this term can be re-inserted again inside the energies of the 16-vertex model, but now with an asymmetric energy. We find at finite temperature corrections to the energy levels of the form \( \epsilon_{1}^{\beta} = \epsilon_{1}, \epsilon_{11}^{\beta} = \epsilon_{11} - \beta^{-1} \log I_0(\sqrt{8} \beta J), \epsilon_{1II}^{\beta} = \epsilon_{1II} - \beta^{-1} \log I_0(2\beta J), \epsilon_{1V}^{\beta} = \epsilon_{IV} \). We see immediately that since for \( x \to 0, I_0(x) \to 1 \), the correction of the dot islands in each vertex is expected not to be important. This is also observed in the experimental results \[46\]. It is interesting to note that the entropic measure is asymmetric, as it affects vertex configuration differently; this will have a role below. However, if \( J \neq 0 \) and sufficiently large, then at low temperatures we get modifications of the energetics for the vertex.

For \( \beta \gg 1 \), we have \( I_0(x) \approx \frac{x}{\sqrt{2\pi}} + O\left(\frac{1}{x^2}\right) \), from which we have \( \delta \epsilon = \beta^{-1} \log I_0(\beta J x) = \beta^{-1} (\beta J x - \frac{1}{2} \log(\beta J x)) \), which can be approximated as \( \delta \epsilon \approx J x \) at temperatures close to zero.

Effectively, since the interaction with the dot can be reabsorbed in a change of the energy levels, the entropic change can also be interpreted as a simple form of renormalization. If we add these correction to the energy of each vertex, we get low temperature renormalized value for the vertices configurations of the form

\[
\begin{align*}
\epsilon_{1}^{\beta} &= \epsilon_{1}, \\
\epsilon_{11}^{\beta} &= \epsilon_{11} - \sqrt{8} J, \\
\epsilon_{1II}^{\beta} &= \epsilon_{1II} - 2 J, \\
\epsilon_{1V}^{\beta} &= \epsilon_{IV}.
\end{align*}
\] (5)

Given the formulae in eqns. \[5\], we can now obtain the main result of this paper; the level inversion can be observed as a function \( J \) in Fig. 4 (top). The optimal interaction strength \( J^* \) such that \( \epsilon_{1} = \epsilon_{11} \) can immediately obtained, being

\[
J^* = \frac{\epsilon_{1II} - \epsilon_{1}}{\sqrt{8}}.
\] (6)

If we fix \( J = J^* \), then we can see how the effective energy of the vertices change as a function of the temperature. This is shown in Fig. 4 (bottom). It is important to note that we fixed \( J^* \) so that \( \epsilon_{1} = \epsilon_{11} \) at \( T = 0 \). However, one could choose \( J^*(\beta) \) so that the effective energies are the same at a finite temperature.

**Implications for ice models: paramagnetic-ferromagnetic transitions.** – Let us now briefly comment on the non-triviality of such interactions for the phase properties of ice models. Let us now consider a reduction of the model to the case of vertices of only Type I, Type II and Type IV, which is the 8-vertex model, which can be solved exactly \[48\]. For energies \( a = b = \exp(-\beta \epsilon_{11}), c = \exp(-\beta \epsilon_{1}), d = \exp(-\beta \epsilon_{IV}) \), let us define for the case without dot, the following order parameter \( \Delta = \frac{a^2+b^2-c^2-d^2}{2(ab+cd)} \). It is known that if \( \Delta > 1 \), the system is in a ferromagnetic phase, if \( \Delta < -1 \), the system is in an anti-ferromagnetic phase, and if \( -1 < \Delta < 1 \), the system is in a paramagnetic phase. Let us now consider again the case \( \epsilon_{1} \approx 0.38675 \) and \( \epsilon_{11} \approx 0.2735 \). In Fig. 5 we plot \( \Delta \) for various values of \( J \). We can see that for \( J = 0 \), at \( T = \infty \) the system is in a paramagnetic phase, and for \( T \to 0 \) the system undergoes a transition to the ferromagnetic phase. However, for \( J \neq 0 \) such picture changes if replace \( \epsilon_{k} \to \epsilon_{1k}^{\beta} \), and for \( J > J_c \approx 0.213 \) the system remains in a paramagnetic phase.

Unfortunately, a similar precise analysis for the sixteen vertex model cannot be done, as the model is not inte-
Since $\Delta_{16}$ is not an exact result, the result above should only be indicative of the importance of the interaction modifiers.

**Conclusion.** – In the present Letter we provided a simple explanation for the population inversion observed in the Artificial Square Ice, based . In conclusion, we have shown analytically that interaction modifiers can be a powerful mechanism to change the energy landscapes of ice models and provided a way to study these effects in detail using a spin-dot interaction model. The approach we have provided in this paper is, in its simplicity, extremely powerful. In fact, it allows to understand the typical renormalization of the energy of each vertex in artificial spin ice because of the local interaction, and it can help to shape the critical behavior of artificial nanomagnets. Our analysis also suggests that, similar to observed experimentally, there is an optimal size $J^*$ for the dot island such that monopoles in artificial square ice have no string tension, but that this depends on the targeted temperature at which the degeneration between the islands is required. Also, it is interesting to note that while here we were interested in the properties of the spin-like variables, nothing stops us from obtaining an effective model for the dot islands. It can be seen after a rapid calculation that a high-temperature effective (continuous) model is of the form $H_{eff} \approx -\beta^2 J^2(\nabla \theta)^2 + O((\nabla \theta)^4)$, where $\theta$ is now a continuous parameter. Thus, it might be possible that also the dot islands have coherent vortex-like behavior induced by a Berezinskii–Kosterlitz–Thouless transition 

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