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On thermalization of magnetic nano-arrays at fabrication

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Abstract. We propose a model to predict and control the statistical ensemble of magnetic degrees of freedom in artificial spin ice (ASI) during thermalized adiabatic growth (Wang et al 2006 Nature 439 303; Morgan et al Nature Phys. 7 75). We predict that as-grown arrays are controlled by the temperature at fabrication and by their lattice constant, and that they can be described by an effective temperature. If the geometry is conducive to a phase transition, then the lowest-temperature phase is accessed in arrays of lattice constant smaller than a critical value, which depends on the temperature at deposition. Alternatively, for arrays of equal lattice constant, there is a temperature threshold at deposition and the lowest-temperature phase is accessed for fabrication temperatures larger rather than smaller than this temperature threshold. Finally, we show how to define and control the effective temperature of the as-grown array and how to measure critical exponents directly. We discuss the role of kinetics at the critical point, and applications to experiments, in particular to as-grown thermalized square ASI and to magnetic monopole crystallization in as-grown honeycomb ASI.
1. Introduction: artificial spin ice (ASI) and its ground state(s)

1.1. Artificial spin ice
The study of frustration, degeneracy and entropy in artificial materials that can be designed to desired specifications is a new trend in both magnetic \([1, 3, 4]\) and colloidal systems \([5, 6]\). Artificial spin ice (ASI) is a two-dimensional (2D) array of elongated, magnetically interacting, single-domain, permalloy nano-islands whose shape anisotropy defines Ising-like spins arranged along the sides of a regular lattice \([1]\). Unlike naturally occurring magnetically frustrated spin ice pyrochlores \([7, 8]\), ASI allows direct imaging of its microstate, thus providing a precise experimental benchmark to theoretical treatments. Since its introduction, ASI has been employed successfully to study frustration \([1]\), and the extension of thermodynamics to granular systems governed by nontrivial interactions \([9–11]\), topological defects \([2]\) and information encoding \([12]\); it has also become a preferred ground for the direct imaging of a new, striking fractionalization phenomenon: magnetic monopoles \([13–15]\).

1.2. Magneto-fluidization and real thermalization
The dimensions of the islands that compose ASI vary somewhat between the different physical realizations. The choice of Morgan et al \([2]\) is rather typical (and not much different from the
ASI of Wang et al [1]): 280 × 85 nm$^2$ for the surface, with a height of 16 nm, arranged on a square lattice of lattice constant $a = 400$ nm, which returns magnetic interactions of the order of $10^4$–$10^5$ K. Clearly, thermal fluctuations cannot induce ‘spin’ flips, and the material is static at room temperature. Therefore, early proponents, including the author, approached ASI as a complex granular material that could be externally driven via magneto-fluidization [10, 11]. The application of a rotating and time-decreasing magnetic field to ASI proved successful in lowering its energy and in returning a controlled variety of statistical ensembles whose detail can be remarkably predicted in terms of an effective temperature [11]. However, for the square ASI the protocol failed to realize—or even approach—its nondegenerate ground state.

In 2010, in a novel approach to achieve ASI’s lowest-energy state, Morgan et al successfully attained what seems to be ASI thermalization during fabrication [2]. They grew square ASI via permalloy evaporation as very thin films on a pre-patterned substrate of silicon, and observed, through magnetic force microscopy, the formation of large crystallites of ground-state domains, separated by domain boundaries and containing only sparse topological defects. Magnetic monopoles in square ASI are energy excitations on top of the ground state; hence the interest in an approach that can reach that ground state. Obviously, control over the microstate of the as-grown ASI would be highly desirable. In this paper, we propose ways to achieve that control during fabrication.

2. Adiabatic growth

2.1. Assumptions

We model the thermalization at growth for ASI of different geometries under the following assumptions:

- The growth is adiabatically slow: at each instant the array is in thermal equilibrium.
- At each instant during deposition, the height $h$ of each island is almost the same across the array.
- As ASI grows, it crosses an energy region in which the magnetic interactions are of the order of the thermal energy or smaller.
- At each stage of growth, there is a blocking temperature below which the system freezes on a time scale commensurate with the deposition rate.

2.2. Blocking temperature

We approach the problem from the point of view of superparamagnetism [16] in which the nano-islands are treated as single-domain magnets, and can randomly flip the direction of their magnetization at temperatures larger than a blocking temperature. Single-domain nano-islands have a volume-dependent, and therefore a height-dependent, energy barrier for spin flipping. We can therefore introduce a height-dependent blocking temperature $T_b(h)$, where $h$ is the height of the islands, and assume that when $T > T_b$ no spin flip takes place (see below for a discussion on the kinetics at stopping). $T_b$ can be computed via micromagnetic simulations for islands of any particular shape and dimensions, but in general we postulate

$$T_b(h) = \tau_1 Ah,$$

(1)
where $h$ is the height of the island and $\tau_1$ (a temperature per unit volume) only slightly depends on the area $A$, as cooperative internal relaxations soften the magnetic reversal. Since $\tau_1 \propto M^2$, $\tau_1$ has a slight dependence on temperature through the density of magnetization $M$ from magnon contribution, which for permalloy we can neglect (the introduction of that dependence on the following is trivial). As thermalization takes place at small $h$, of the order of a few nanometers, we take $T_b(h)$ linear in $h$.\(^1\)

As deposition increases it reaches a blocking point, after which the blocking temperature $T_b(h)$ is larger than the temperature $T$ at which deposition is performed, or $T_b \geq T$. The system then freezes in a thermodynamic state dependent on the deposition temperature $T$. From (1) we find that

$$h^*(T) = \frac{T}{\tau_1 A},$$

for the blocking height, or the (average) height of the islands when dynamics stops, for deposition performed at a temperature $T$. Obviously, $h^*$ increases with $T$: larger temperature at deposition extends the dynamical range of ASI, delaying its freeze to higher depositions. (In the following, we will denote the value of observables at the blocking point by a star symbol.)

### 2.3. Effective temperature

We have assumed the magnetic degrees of freedom of the array to be in thermal equilibrium when the blocking temperature $T_b$ crosses the deposition temperature $T$ and dynamics stops. Since the array freezes into a definite thermodynamic state at the blocking point, we can introduce an effective temperature $T_{\text{eff}}$ as the temperature the observed ensemble would have in order to be Gibbsian in the as-grown energetics.

In treatments of superparamagnetism, interactions between magnetic nano-islands are often neglected\[17, 18\], yet their role in determining the statistical ensemble of ASI is obviously fundamental. Let $E(h)$ be any relevant energy emerging from inter-island interactions in an array whose islands have height $h$. Given the dipolar nature of the interaction, we assume that $E(h)$ scales as

$$E(h) = \epsilon(l/a) \frac{h^2}{a^3} A^2,$$

where $a$ is the lattice constant, $l$ is the length of an island, and we have assumed that $l \gg \sqrt{A}$, which corresponds to strong anisotropy on the nano-island. Clearly, $\epsilon(l/a)$ tends to a constant in the limit of $l/a \to 0$, the ideal dipole approximation, and is, in general, proportional to the square of the density of magnetization.

If $H$ is the final height of the islands when deposition is complete and $h^*$ the blocking height, then our assumptions allow us to define $T_{\text{eff}}$ through the equation

$$\frac{E(H)}{T_{\text{eff}}} = \frac{E(h^*)}{T},$$

which holds for any thermodynamically relevant energy $E$: indeed if the system is in equilibrium at the stopping point, its thermodynamic ensemble is completely controlled by quantities like $E(h^*)/T$, and does not change after $h$ exceeds the blocking height $h^*$ and while it grows from $h^*$\(^2\)

\(^1\) Deviations from linearity would include a negative correction $\propto -h^2$ to account for increased internal relaxation during spin moment reversal in a taller nano-island, and would not qualitatively affect our treatment.
to $H$. From (3), the ratio between the energies is simply $E(h^*)/E(H) = h^{*2}/H^2$ and therefore one has that $T_{\text{eff}} = TH^2/h^{*2}(T)^2$. From (2) we then find our first result

$$T_{\text{eff}} = \frac{\tau_1^2 A^2 H^2}{T}.$$  \hspace{1cm} (5)

Equation (5) shows an interesting fact: the effective temperature is lowered when the deposition temperature is raised. This result is only apparently counterintuitive, since larger temperatures during fabrication extend the dynamic region of ASI during growth, as already noted.

The effective temperature in (5) does not depend upon the lattice constant but only on properties of the single islands, such as area, height and density of magnetization via $\tau_1$ (as also in the case of the effective thermodynamics for the ac demagnetization [9, 11], where an effective temperature describes instead a stochastic process out of equilibrium). This does not imply that arrays of different lattice constants fabricated at the same temperature would belong to the same statistical ensemble: the system is controlled by ratios of energy over temperature of the kind $E(H)/T \sim a^{-3}$, which increase at smaller lattice constant. Therefore arrays of smaller lattice constant $a$ belong to thermodynamic ensembles closer to the ground state. A less natural yet more general definition of effective temperature that might be more useful for experiments performed by varying the lattice constant will be given below.

3. Crystallization

3.1. The critical lattice constant

ASI of certain geometries are expected to undergo interesting phase transitions. In particular, the square lattice should crystallize into an antiferromagnetic tiling. Let $T_c$ be any critical temperature for a specific ASI geometry. It could be the temperature for crystallization of square ASI into its antiferromagnetic ground state. Or it could be the critical temperature of the ‘Ice II’ phase predicted for honeycomb ASI via numerical works [19, 20] and correspond to the crystallization of magnetic monopoles of opposite charge on neighboring vertices, much like a NaCl ionic crystal. (For specificity, from now on we will talk of ‘crystallization’ in the general sense whenever we allude to ASI undergoing any phase transition.)

$T_c$ is a particular case of (3) and for an array of lattice constant $a$, comprising islands of surface $A$ and height $h$, it reads

$$T_c(h) = \frac{\tau_2^2 h^2}{a^3} A^2.$$ \hspace{1cm} (6)

The same considerations exposed above for $\epsilon(l/a)$ apply now to $\tau_2(l/a)$, which like $\tau_1$ has the dimension of a temperature per unit volume and is proportional to the square of the density of magnetization of the material, $\tau_2 \propto M^2$.

Like $T_b$, $T_c$ also depends on the height $h$ of the islands, although quadratically rather than linearly. Figure 1(a) plots $T_c$ and $T_b$ versus $h$ for different lattice constants $a$. One can see that for large $a$, $T_c^* = T_c(h^*) < T$: therefore, when the islands stop flipping at $h^*$, the critical temperature $T_c^* = T_c(h^*)$ is lower than the experimental temperature $T$, and no crystallization has yet occurred. Instead the opposite happens for very small lattice constants. There exists therefore a temperature-dependent critical lattice constant $a_c(T)$ such that for lattice constants
Figure 1. Left: arrays of different lattice constants grown at the same fabrication temperature show an ordered microstate when the lattice constant is smaller than a critical value $a_c$. In the figure we plot the blocking temperature $T_b(h)$ and the critical temperatures $T_c(h)$ for arrays of three different values of the lattice constant ($a > a_c$, $a < a_c$, $a = a_c$ (this last one in red)) as a function of the height of the nano-islands $h$. $h^*$ is the stopping height, after which $T_b > T$ and therefore freezing-in starts. When $a > a_c$, $T_c^* > T_b = T$ at the blocking point, and therefore no crystallization is attained. When $a < a_c$, $T_c^* = T_b = T$ at the blocking point, and therefore crystallization is attained. $a_c$ is determined by the intersection $T = T_b = T_c$. For definiteness we have chosen $T = 350$ K, $\tau_1 = 6.5 \times 10^{-3}$ K nm$^{-3}$, $\tau_2 = 10$ K nm$^{-3}$, which return $h^* = 3.5$ nm and $a_c = 435$ nm. Right: during the fabrication of arrays of equal lattice constant the microstate can be controlled by varying the temperature $T$ at deposition. The figure shows the inverted temperature behavior, in which crystallization happens for deposition temperatures higher than the temperature threshold $\bar{T}$, defined by the intersection between blocking temperature $T_b[h]$ and critical temperature $T_c[h]$, or $T_b(\bar{h}) = T_c(\bar{h}) = \bar{T}$. When $T > \bar{T}$ the critical temperature for condensation (black dots) is larger than the deposition temperature (red dots) when the spin freeze-in, at the intersection of $T_c$ and $T$, and therefore the as-grown array shows a crystallized microstate. The opposite happens for $T < \bar{T}$. For $T \sim \bar{T}$ then $T_c \sim T_b$ and kinetics effects must be taken into account. For definiteness we have chosen $\bar{T}$ to coincide with room temperature.

$a < a_c$ one expects crystallization, whereas for $a > a_c$ none is expected. From figure 1(a) the critical lattice constant $a_c$ is found by equating $T_b^* = T_c^* = T$. Via (1)–(6) one finds

$$a_c(T) = \sqrt{\frac{\tau_2 T}{\tau_1}}.$$  

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Clearly, $a_c$ depends on the deposition temperature $T$ and larger temperature during deposition allows for larger arrays to reach crystallization. In the case of square ASI, arrays of lattice constant lower than $a_c$ are grown in a crystallized microstate, whereas arrays of larger lattice constant should show thermal disorder, while still being described by a Gibbsonian distribution, in terms of an effective temperature.

### 3.2. Inverted temperature behavior

Alternatively, arrays of identical lattice constant can be grown at different deposition temperatures. Then figure 1(b) shows that if we call $\bar{T}$ the temperature at which the curves of $T_c(h)$ and $T_b(h)$ intersect, or $\bar{T} = T_c(\bar{h}) = T_b(\bar{h})$, the growth of crystallized arrays corresponds to a deposition temperature $T > \bar{T}$. Indeed, when the deposition temperature is higher, or $T > \bar{T}$, the critical temperature at the blocking point is larger than the temperature at which the experiment is performed, or $T^* > T$, and the system has already undergone crystallization when dynamics freeze. Conversely, growth at temperature $T < \bar{T}$ results in a thermally disordered array at the moment in which dynamics stop. Effectively, $\bar{T}$ represents a critical temperature, corresponding to $T_c(\bar{h})$ but, interestingly, the system crystallizes above rather than below that critical temperature. To avoid confusion with the height-dependent critical temperature of the arrays, we call $\bar{T}$ the temperature threshold at fabrication.

This inverted temperature behavior is a consequence of the larger dynamical regime afforded by larger fabrication temperature. $\bar{T}$ can be computed easily as

$$\bar{T} = \frac{\tau_1^2 a^3}{\tau_2},$$

and is a function of the specific material and the geometry, and is proportional to the square of the density of magnetization, $\bar{T} \propto M^2$. If $T_c^*(T)$ is the value of the critical temperature for the array at the blocking point for different values of the fabrication temperature $T$, from (2), (6) and (8) we have

$$\frac{T}{T_c^*(T)} = \frac{\bar{T}}{T},$$

which shows that theoretically the ground state can only be achieved for infinite temperature at deposition. In practice, crystallization detectable by the limited size of magnetic force microscopy (MFM) images can be typically achieved for reasonable values of $(T_c - T)/T$, typically larger than 0.1. From (9),

$$\frac{T_c^*(T) - T}{T} = \frac{T - \bar{T}}{T}.$$

Equations (9) and (10) show again that larger temperature at fabrication leads to a more ordered condensate. Equation (10) is useful in relating a quantity fundamental for diverging observables at the phase transition to the temperature at fabrication, therefore allowing for direct extraction of critical indices, using the technique introduced by Lammert et al to directly extract entropy from ASI [12].

### 3.3. Critical point and slow dynamics

We have shown that different phases can be obtained for fabrication temperature $T$ larger or smaller than the temperature threshold $\bar{T}$ in (8). But what happens when $T \sim \bar{T}$ and therefore...
(figure 1(b)) the blocking temperature is close to the critical temperature, or \( T_b \sim T^* \)? The dynamical response of the array slows down right when the system undergoes transition, and phenomena similar to those due to rapid cooling might be expected. Clearly, it is now time for a few kinetic considerations.

There are three characteristic rates in our problem. One is the rate of thermally induced magnetization reversal for each island, which in the limit of noninteracting islands is given by the Néel–Arrhenius law, \( v = v_0 \exp(-E_b/kT) \), where \( E_b \) is the energy barrier for magnetization reversal, and \( v_0 \) is the Arrhenius pre-factor (typically \( v_0 \sim 10^{10} - 10^{12} \text{ s}^{-1} \) [21]). Then there is the relaxation time for lattice equilibration, whose rate we call \( v_e \). Equilibrium is regained by flipping a certain number of spins (per unit area) and it is then reasonable to take \( v_e \propto v \), or

\[
v_e = v_{e,0} \exp[-E_b(h)/kT]. \tag{11}\]

Finally, there is the deposition rate \( v_h \), given by the number of layers deposited in one unit time.

For each subsequent layer, the change in the energetics of the system corresponds to a small deviation from equilibrium. But as deposition increases, so does the time \( v_e^{-1} \) needed for the system to respond and re-equilibrate into a new state. Eventually, the array response is too slow to catch up with the change in energetics, and the process converges to a state close to the equilibrium ensemble. If that state is far from a phase transition, we can follow the general approach in superparamagnetism and approximate it to an equilibrium state whose temperature is given by \( v_e(T_b) = v_h \). With that choice, \( T_b \) in (1) is related to the coercive energy barrier \( E_b \) via

\[
T_b(h) = \frac{E_b(h)}{\ln \left( \frac{v_{e,0}}{v_h} \right)}. \tag{12}\]

\( E_b \) is generally taken to be independent of the temperature \( T \) and proportional to the volume. Equation (1) is therefore justified with \( \tau_1 \propto M^2/\ln(v_{e,0}/v_h) \), where \( M \) is the density of magnetization of the material. The typical deposition rate in experiments is \( v_h \sim 10^{-1} \text{ s}^{-1} \), much smaller than \( v_{e,0} \propto v_0 \sim 10^{10} - 10^{12} \text{ s}^{-1} \). Therefore, from an experimental perspective, changing the deposition rate even by a few orders of magnitude has a negligible effect on \( \tau_1 \) and therefore on our predictions above.

This description can break down when the blocking point is close to a critical point, since the change in the microstate during relaxation can, in principle, be dramatic. \( h_c \) is the height at which the system undergoes the phase transition (and is therefore defined implicitly as \( T_c(h_c) = T \)). When it is much smaller than the blocking height \( h^* \) the array undergoes the phase transition before freezing. But if \( T \sim \tilde{T} \), then \( h^* \sim h_c \sim \bar{h} \). There is a narrow window around the stopping height \( h^* \) in which the relaxation time of the system is one order of magnitude or less smaller than its value at \( h^* \). With our choice of \( T_b \) in (12), and with \( h^* \) from (2), we obtain from (11) the size of that window

\[
\frac{h^* - h}{h^*} < \frac{1}{\log_{10}(v_{e,0}/v_h)} \approx 0.1. \tag{13}\]

When \( h_c \) lies below that window, the system undergoes the phase transition. Naturally, the relative values of \( h_c \) and \( h^* \) cannot be chosen freely, but depend on the temperature at fabrication. From (1), (6) and (8) we obtain

\[
\frac{h^* - h}{h^*} = \frac{T - \tilde{T}}{2T}. \tag{14}\]
Therefore no kinetic concern should involve the phase transition when

\[
\frac{T}{\bar{T}} = \frac{\tau_2 T}{\tau_1^2 a^3} > \theta = \frac{\log_{10} \left( \frac{v_{e,0}/v_h}{10^{-2} v_{e,0}/v_h} \right)}{\log_{10} (10^{-2} v_{e,0}/v_h)} \simeq 1.2.
\] (15)

When instead \( T < \theta \bar{T} \), the system responds slowly at the deposition threshold for crystallization \( h_c \), which might correspond to super-cooling or to glassy behavior. Note that (9) and (15) imply that when kinetic effects are negligible we have \( \bar{T}/T_c^*(T) < \theta^{-1} \) and therefore no glassy behavior or lack of equilibration can be induced far below the critical point.

4. Implications for as-grown ASI

4.1. Effective critical temperature

The presence of an equilibrated ground state can be revealed by directly imaging the microstate of ASI, and our framework can be tested by mapping different microstates from arrays obtained at different deposition temperatures or of different geometry. It is therefore useful to relate the above considerations to measurable quantities in the as-grown samples.

As mentioned above, the as-grown array corresponds to a specific frozen-in thermodynamic state, and therefore it is useful to express our results in terms of the effective temperature. From (6) it follows that the critical temperature for an as-grown thermalized array with islands of height \( H \) is

\[
T_c^{\text{eff}} = \tau_2 \frac{H^2}{a^3} A^2,
\] (16)

which we call effective critical temperature. By direct substitution, it is easy to prove equations analogous to (9) and (10) in terms of the effective temperature:

\[
\frac{T_c^{\text{eff}}}{T} = \frac{T_c^{\text{eff}}}{T} = \frac{\tau_1^2 a^3}{\tau_2 T}
\] (17)

and

\[
\frac{T_c^{\text{eff}}(T) - T^{\text{eff}}}{T^{\text{eff}}} = \frac{T - \bar{T}}{\bar{T}}.
\] (18)

Again (17) shows how the effective temperature can be reduced by increasing \( T \) at fabrication, or by using lattices of smaller lattice constant. Figure 2 shows the behavior of \( T^{\text{eff}}/T_c^{\text{eff}} \) for different fabrication temperatures and lattice constants under reasonable assumptions for \( \tau_1, \tau_2 \). For definiteness we have chosen \( \tau_1 = 6.5 \times 10^{-3} \text{ K nm}^{-3} \), \( \tau_2 = 10 \text{ K nm}^{-3} \), to return \( h^* = 3.5 \text{ nm} \) and \( a_c = 435 \text{ nm} \) at a deposition temperature \( T = 350 \text{ K} \).

4.2. The most general effective temperature

The previous definition of effective temperature, as the temperature that the as-grown system would have in order to belong to the observed experimental ensemble at its as-grown energetics, is the most natural. It is also well suited for experiments in which the temperature at fabrication can be controlled. From an experimental point of view, it is interesting to introduce a more general effective temperature that can more directly take into account the changes in lattice constant.

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Figure 2. Left: plot of $T_{\text{eff}} / T_c$, the effective temperature of an as-grown array, measured in units of the critical effective temperature as a function of fabrication temperature $T$ in °C, for different values of the lattice constant $a$, from 500 to 300 nm, from (9). The dashed vertical line represents the temperature of 50 °C estimated in the experiments of [2]. For definiteness we have chosen $\tau_1 = 6.5 \times 10^{-3}$ K nm$^{-3}$, $\tau_2 = 10$ K nm$^{-3}$, which at $T = 350$ K return the blocking height $h^* = 3.5$ nm and the critical lattice constant $a_c = 435$ nm (dashed red line), likely close to the experimental case of [2]. Inset: the temperature threshold $\bar{T}$ for crystallization for arrays of different lattice constants. Right: for perspective, curves described on the left are drawn as cuts on the surface plot of $T_{\text{eff}} / T_c$ versus the fabrication temperature $T$ (°C) and the lattice constant $a$ of the array (nm).

In general, as explained above, the thermodynamic state is controlled by the quantities $E(h^*(T))/T$, where $E(h)$ is any relevant energy for the system of height $h$ and is given by (3). Therefore, from (2) and (3), and taking $\tau_1 \propto M^2$, $\epsilon \propto M^2$, where $M$ is the density of magnetization, we can introduce the most general choice of an effective temperature $\tilde{T}_{\text{eff}}$ as the one normalized to an energy scale independent of the variables of the problem, or

$$\tilde{T}_{\text{eff}} \propto M^2 a^3 T^{-1}. \quad (19)$$

Clearly, (17) and (18) hold for this alternative definition of effective temperature as well.

4.3. Disordered as-grown square ASI

Let $n_\alpha$ be the relative occurrence of a state of the system. Using (19) one can compute $n_\alpha$ as a function of different temperatures at fabrication, different lattice constants and for materials of different magnetizations, via the Gibbs distribution

$$n_\alpha = Z^{-1} \exp \left( \chi_\alpha T / M^2 a^3 \right), \quad (20)$$

where $\chi_\alpha$ is a constant and depends only on the scale-invariant geometry and the particular state $\alpha$. 

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Direct visualization of as-grown square ASI obtained via a slow and careful deposition of permalloy (see the introduction) [2] has revealed the formation of crystallites of long range order. From (7) one expects that a thermally disordered picture can be regained for larger lattice constants (lower temperatures would be impracticable, since \(h^*\) for those experiments seems already rather low, of the order of a few nanometers). Since thermal disorder might warrant an approach in terms of a gas of independent vertices [9, 11], \(n_\alpha\) in (20), where \(\alpha\) labels different vertex configurations [1], can be used to predict the relative abundance of different vertices in depositions performed at different lattice constants or temperatures.

4.4. Crystallites in as-grown square ASI

The presence of grain boundaries in ordered as-grown arrays has been ascribed to inherent disorder in inter-island interactions [22], a phenomenon also seen in numerical simulations of different systems at zero temperature [23, 24]. Recent numerical and experimental works on magneto-fluidization of square ASI have shown that disorder in the nonuniform energy barriers for magnetization reversal leads to nucleation sites for ground-state crystallites of opposite orientation, rendering a single-domain ground state unattainable [25]. In the case of as-grown thermalized ASI, our assumption of equal height of the islands at growth, although plausible, might neglect a disordered distribution of heights, which in turn could provide similar nonuniform energy barriers for magnetization reversal.

Even though the disorder-based mechanism for grain boundary formation is not well understood in this case, it is reasonable to assume that if indeed it is the disorder in inter-island interactions that is responsible for the observed fragmentation, then a smaller lattice constant would lead to larger crystallites. Indeed, the size of the crystallites at zero temperature is likely to be determined by the ratio between the energy cost of the grain boundaries and the energy variations in inter-island interactions due to quenched disorder in the size and shape of the nano-islands. As lower lattice constants increase the inter-island interaction without changing the energy disorder, it might lead to larger crystallites. A proper thermodynamics of quenched disorder in square ASI and its effect on crystallite formation could be employed within our framework to predict crystallite size in as-grown ASI, via the effective temperature in (19). If instead fragmentation is mostly a consequence of the disorder in the coercive barrier of the islands due to nonuniform height \(h\) during deposition, which reflects in nonuniform blocking temperatures, then our treatment would suggest that larger temperatures at deposition should return crystallites of larger size. In fact, a larger fabrication temperature \(T\) leads to larger stopping height \(h^*\) and therefore to a reduced relative magnitude of disorder in \(\Delta h/h^*\) at the blocking point.

Our approach suggests that in addition to disorder, one might consider another source of fragmentation into subdomains. As explained above, when \(\bar{T} < T < \theta \bar{T}\), kinetic effects become important at crystallization. Real materials are known to crystallize into domains of different orientations when cooled at a fast rate, and the same phenomenon could be taking place in as-grown square ASI. We do not know the value of the temperature threshold \(\bar{T}\) [2], yet if the formation of crystallites is indeed a consequence of the proximity of the

\[\text{From our purely illustrative choice of } \tau_1 = 6.5 \times 10^{-3} \text{ K nm}^{-3}, \tau_2 = 10 \text{ K nm}^{-3}, \text{used in the figures, and } a = 400 \text{ nm}, \text{we get, from (8), } \bar{T} = 270 \text{ K. Since } T = 350 \text{ K, we have } T/\bar{T} = 1.3, \text{ which, according to (15), lies just above the kinetic window. Of course, a slightly different yet equally reasonable choice of those parameters returns a ratio of } T/\bar{T} \text{ corresponding to the region of slow dynamics, suggesting that kinetic effects might play a role.} \]
crystallization point to the blocking point, then from (8) and (15) an increase of say 30\% in the temperature at deposition, or a reduction of 10\% in the lattice constant, should take the critical point out of the kinetic window and therefore considerably change the size of crystallites.

4.5. Dynamical ASI

It would be interesting to fabricate more dynamical ASI which could then be equilibrated at different temperatures. Arrays responsive to thermal fluctuations might be obtained by playing with the magnetization of the material but also via deposition techniques, by keeping $H$, the final height of the islands, small, yet not too small. Figure 1(b) shows that when $H < \bar{h}$ the resulting as-grown ASI will never be able to approach the region of critical temperature which, depending on the application, might or might not be desirable: indeed when exposed to temperatures lower than $T_b(H)$, ASI would not respond, and since $T_c(H) < T_b(H)$ if $H < \bar{h}$, then the phase transition would be inaccessible to thermalization. $\bar{h}$ can be easily computed as

$$\bar{h} = \frac{\tau_1 a^3}{\tau_2 A}$$ (21)

and interestingly does not depend on the density of magnetization.

For square ASI, Morgan et al [2] show that crystallization is achieved and since they report a blocking height of a few nanometers, we can take $\bar{h} \sim 1$ nm for their system. Doubling the lattice constant would give, from (21), $\bar{h} \sim 10$ nm. Then an array of $a = 800$ nm and thickness $H \sim 5$ nm would then respond to external temperature, even at room temperature, while always being disordered. However, an array of $a = 400$ nm and thickness $H \sim 5$ nm would also equilibrate and respond to external temperature, while also accessing an ordered phase.

5. As-grown ASI and magnetic monopoles

Magnetic monopoles were introduced in naturally occurring spin ice pyrochlore to subsume the effect of long-range interactions in a simple description of low-energy excitations [13]. The role of their Coulomb-like charge is confirmed by the explanation of low-temperature behavior of spin ice in terms of a liquid–gas transition of monopoles.

Magnetic monopoles have been directly observed in honeycomb ASI. Yet while these topological excitations do correspond 'structurally' to the magnetic monopoles of spin ice, insofar as one can somehow formally attribute a ‘net magnetic charge’ to the excited vertices, the effects of their long-range interaction are still unclear. To rightfully deserve their name in ASI, monopoles must be shown to provide a similar low-energy description, amenable to thermodynamic treatment.

Unlike magneto-fluidized ASI, which returns higher-energy macro-states, thermalized as-grown ASI provides ordered states, in which magnetic monopoles could describe low-energy excitations. By controlling the temperature and lattice constant as explained in our approach, as-grown thermalized ASI can map microstate probabilities corresponding to different effective temperatures and provide a promising playground to test thermodynamic treatments of magnetic monopoles. This would finally assert their reality as point-like, long-range interacting excitations. Below we propose directions to achieve this goal within our framework.
5.1. Square ASI and monopole excitations

Morgan et al [2] witnessed the formation of local excitations inside ordered crystallites, computed their energy numerically via a point dipole model and showed that their relative frequencies follow a Gibbsian distribution, which further corroborates the idea that real thermalization is taking place during growth. They also pointed to particular defects in the form of monopole charges connected by energetically costly Dirac strings (or more properly Nambu strings [26, 27]) and noticed their tendency to form closed configurations with the string looping, rather than configurations with long open strings. They interpreted this as an effect of the monopole–antimonopole long-range magnetic attraction.

It would be interesting to raise the effective temperature (19) by lowering the fabrication temperature, to see whether the change induces an opening of such loops and more separated monopoles, and if a description of their energies in terms of a Coulomb interaction can provide faithful predictions of their relative abundance via (20).

Mól et al., have predicted a monopole-unbinding transition [27–30], in which the entropy of the Dirac string overcomes its energy cost. This transition could also be investigated by fabricating ASI of different effective temperatures. Although numerical results predict the transition in a state of thermal disorder, which would prevent its direct observation in square ASI via magnetic force microscopy, that might be a consequence of the point dipole approximation employed by the authors. Indeed in a numerical work on honeycomb ASI, Möller and Moesner have shown that, as one would expect, monopole signature becomes less observable when the ratio $l/a$ tends to zero [19]. Also, even if impossible to spot by eye in an MFM image, a transition could be seen by extracting the entropy for different effective temperatures with the method illustrated by Lammert et al [12] and using it to compute the specific heat curve.

5.2. Honeycomb ASI and crystallization of monopoles

As mentioned above, our description of thermalization in as-grown ice is independent of the particular geometry and can be applied to honeycomb ASI as well. While as-grown square ASI could be the ideal candidate to study monopoles as sparse excitations, honeycomb$^3$ ASI is interesting in a different regard: it should reveal two phase transitions, recently explored numerically [19, 20]. At low temperature, a (pseudo) ice rule manifold appears, in which vertices manifest the 2-in/1-out or 2-out/1-in rule. Unlike the case of square ASI, in honeycomb ASI each low-energy vertex is endowed with a positive or negative magnetic charge. Therefore a new phase transition (which was predicted numerically although not yet observed experimentally) should bring it to a lower-energy configuration which the work [19] named Ice II, and which in practice corresponds to the crystallization of monopoles of opposite charges on nearest-neighbor vertices, therefore forming a triangular ionic crystal of monopoles. At even lower temperatures, an ordered phase emerges because of long-range dipolar interactions neglected in the monopole approximation: the loop state. The two critical temperatures for these

$^3$ A honeycomb ASI is made of nano-islands arranged along the edges of a honeycomb pattern. It can be modeled by dipoles on a kagome lattice or by monopoles on the vertices of a hexagonal lattice. Since both theoretical descriptions might apply to the same real material in different conditions, the author prefers to employ the more general nomenclature of ‘honeycomb ASI’ rather than the more particular ‘hexagonal ASI’ or ‘kagome ASI’ to demarcate the actual physical material used in experiments from the different possible theoretical models that might apply to it.
phases strongly depend on the ratio between the island length $l$ and the lattice constant $a$ [19], a property that can be exploited for intelligent fabrication.

Unlike the case of square ASI, magneto-fluidization [10, 11] successfully anneals the honeycomb ASI into its pseudo-ice manifold. Yet it fails to reveal any monopole crystallization. Magneto-annealed samples return an extracted entropy per spin of $\sim 0.75$ [12] rather than the $\sim 0.15$ expected at crystallization (taking the entropy of a random spin distribution to be 1), and extraction of nearest neighbors charge correlation from MFM images provides values $\sim 0.1$ rather than the crystallized value of 1. It would therefore be very interesting to attempt as-grown thermalization of honeycomb ASI to investigate monopole crystallization and the loop state by controlling its effective temperature in the way described above and by extracting its entropy and computing monopole–monopole correlations. Our predictions above apply, mutatis mutandis, to these two transitions as well, with a different choice of the constant $\tau_1$, $\tau_2$. Clearly, in the case of monopole crystallization, one expects a more complex dependence of those constants on the ratio $l/a$ as shown in numerical calculations [19]. In particular, in the case of $l \sim a$, because of the Coulomb interaction between magnetic monopoles, the dependence of the critical temperature for monopole crystallization will be $T_c(h) \sim h^2/a$ rather than the $\sim h^2/a^3$ of (6).

6. Conclusion

We have treated the fabrication of ASI by slow deposition as an adiabatic phenomenon and found that the probability of its microstate is described by an effective temperature which depends on the lattice constant of the arrays and the temperature at deposition. When a phase transition exists in ASI, there is a geometry-dependent temperature threshold such that the phase below the critical point can be achieved with fabrication temperatures above the temperature threshold. When the deposition temperature is close to the temperature threshold, kinetic effects due to the Néel–Arrhenius flipping dynamics are expected to play a role similar to fast cooling at the critical point. We have proposed how to use these considerations to study monopole-unbinding in square ASI and monopoles crystallization in honeycomb ASI.

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