Dynamics of Thermally Active Magnetic Dipolar Plaquettes.

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The collective behavior of thermally active structures offers clues on the emergent degrees of freedom and the physical mechanisms that determine the low energy state of a variety of systems. Here, the thermally active dynamics of magnetic dipoles at square plaquettes is modeled in terms of Brownian oscillators in contact with a heat bath. Solution of the Langevin equation for a set of interacting $x-y$ dipoles allows the identification of the time scales and correlation length that reveal how interactions, temperature, damping and inertia may determine the frequency modes of edge and bulk magnetic mesospins in artificial dipolar systems.

**Introduction.** In the study of dynamical systems, temperature has proved to be an ally for the elucidation of new orders and matter phases. To capture the thermally active phenomenology, timing is crucial and therefore a prevalent challenge consists in the commensuration of the experimental frequencies with a system proper time scales [1]. The difficulty arises because often there are several time scales, and worst, one or few may be the result of intrinsic interactions [2]. A distinctive playground for thermal dynamics research is supplied by frustrated magnetic systems [3]. Here, the lack of compromise of the interacting particles of the system with a long range order may be due to a plethora of collective low energy configurations offered by the underlying lattice [4]. This scenario is further enhanced when temperature becomes involved. A prototypical example are spin ice materials [5], where dipolar interactions and weak antiferromagnetic superexchange result in an effective ferromagnetic coupling that in combination with single site anisotropy, yield a frustrated spin arrangement that mimics the geometric frustration in water ice [6]. Thermal excitations in spin ice are manifested as violations of the ice rule and are analogous to magnetic monopoles [7]. In addition, the study of the thermal relaxation process by means of a.c. magnetic susceptibility measurements [8] has revealed a monopole like dynamics mediated by the Coulomb interaction between charges [9]. In two dimensions, in the metamaterials arena, artificial dipolar systems [10] have become ideal settings for observing dynamical effects. In artificial spin ice (ASI) structures [11], the arrangement of moments product of elongated single-domain nanopatterned magnetic islands can lead to excited states with magnetic charges [12], analogous to the monopole excitations reported in rare-earth pyrochlores [13]. Recently, susceptibility measurements [14] of thermally active extended square ASI [15] revealed that magnetic fluctuations and excitation population depend on lattice spacing and interaction strength between islands [16]. A Vogel-Fulcher-Tammann law [17], has recently employed, with the purpose of extracting parameters related to the magnetostatic energies of ASI arrays directly from the susceptibility measurements [13]. Nevertheless the results showed that this approach fails to address the dynamics of thermal ASI arrays. The failure of this and other phenomenological models for describing the dynamic response from frequency measurements in systems as diverse as spin ices, spins glasses and superconductors [18, 19] is rooted in the ad-hoc time scale distributions used to complement models underlying Debye processes.

**Main results.** Here this problem is addressed by studying two simple dipolar systems. The first consists of a single square plaquette made out of four interacting inertial dipoles that rotate in the $x-y$ plane, see Fig. 1(a). The second is a small lattice made out of four of such plaquettes, see Fig. 1(c,d). The systems dynamics is modeled by a Langevin equation with gaussian thermal noise [20] and dipolar interactions. Inspection of the Langevin equation for small angular oscillations allows to identify the relevant time scales for the thermal relaxation dynam-

![FIG. 1. (color online) (a) Dipolar square plaquette in the vortex configuration. Geometrical parameters are shown. (b) Phase diagram of the four square plaquette lattice as a function of dimensionless temperature and lattice spacing. Red squares denote the occurrence of the magnetic configuration shown in (c) named $g_1$, while blue diamonds the magnetic configuration shown in (d) named $g_2$.](image-url)
ics and detect their manifestation in the time autocorrelation function \( C(s) \). They originate from the internal magnetic field due to dipolar interactions, from temperature, and from the intrinsic properties of the system such as inertia and damping. Further analysis of \( C(s) \) allows to identify a magnetic correlation length that defines the boundary between weakly and strongly interacting regimes in terms of physical and geometrical parameters. The approximated solution of \( C(s) \), valid for short times, is corroborated and complemented by molecular dynamics simulations. We find that the earliest dynamics of \( C(s) \) can be quantified in terms of three times scales consequence of the interplay between temperature, dipolar interactions and inertia. The second stage of the time autocorrelation function evolution can be described in terms of a fourth time scale product of dipolar interactions and damping.

Thermal relaxation of a lattice made out of four square plaquettes allows the manifestation of two metastable magnetic configurations. Magnetization plateaux in the plaquettes is described in terms of the angle \( \alpha_i \) and intensity \( m_0 \). The magnetic moment \( m_i = m_0 \hat{m}_i \) at position \( \mathbf{r}_i \) has unit vector \( \hat{m}_i = \frac{\mathbf{r}_i - \mathbf{r}_k}{|\mathbf{r}_i - \mathbf{r}_k|} \) with \( \mathbf{r}_i \) the position of the center of dipole \( i \), \( \hat{e}_{ik} = (\mathbf{r}_i - \mathbf{r}_k)/|\mathbf{r}_i - \mathbf{r}_k| \), \( \gamma = \frac{\mu_0 m_0^2}{4 \pi} \) ([Nm\(^4\)]), \( \mu_0 \) the magnetic permeability. The magnetic moment \( m_i = m_0 \hat{m}_i \), has unit vector \( \hat{m}_i = (\cos \alpha_i, \sin \alpha_i) \) and intensity \( m_0 \) ([m\(^2\) A]). Dipoles of length 2\( a \) are located at the vertices of square plaquettes as shown in Fig. 1. Their rotation in the \( x - y \) plane is described in terms of the angle \( \alpha_i \) chosen respect to their equilibrium position, and the distance between the centers of two nearest neighbor dipoles is \( \sqrt{2}(a+\Delta) \). Tuning \( \Delta \) changes the strength of the dipolar interactions.

Here we address the thermal dynamics of dipolar lattices and for that effect we study the square plaquette of Fig. 1(a) and a small square lattice made out of four of such plaquettes, Fig. 1(c,d). Each dipole is modeled as a Brownian oscillator in contact with a heat bath. The Langevin equation that determines the dynamics of the angular variable \( \alpha_i \) is:

\[
I \frac{d^2 \alpha_i}{dt^2} = \sqrt{2 \eta k_B T \xi(t)} - \eta \frac{d \alpha_i}{dt} - K_i \alpha_i, \tag{2}
\]

where \( I \) ([Kg m\(^2\)]) is the inertia moment of each dipole, and \( \eta \) ([Kg m\(^3\) s\(^{-1}\)]) is a damping coefficient that accounts for the viscous rotation. Thermal fluctuations due to the coupling of the magnet with the heat bath are modeled by a \( \delta \)-correlated Gaussian noise \( \xi(t) \) of zero mean and unit intensity: \( \langle \xi(t) \rangle = 0 \), \( \langle \xi(t) \xi(t') \rangle = \delta(t - t') \).

The units of \( \xi(t) \) are \([1/\sqrt{s}]\). \( T \) denotes temperature and \( k_B \) is the Boltzmann constant. The last term \( K_i \alpha_i \) deserves special attention. It accounts for the torque along the \( z \) direction, on dipole \( m_i \) due to the net internal magnetic field \( h_i \) originated by all other dipoles at \( \mathbf{r}_i \). Such a torque is \( \sum_{j \neq i} (m_i \times h_j)^z = m_0 \sum_{j \neq i} (h_j^z \cos \alpha_i - h_j^|| \sin \alpha_i) \). Assuming that 1) dipoles deviate slightly from their equilibrium positions and 2) at a given position the total internal fields perpendicular and parallel to \( m_i \) are such that \( |h_i^z| \ll |h_i^|| \) (this assumption will be justified in the next sections), yields \( (m_i \times h_i)^z \approx m_0 h_i^|| \cos \alpha_i \), and \( K_i = m_0 h_i^|| \), where the precise form of the internal field at position \( \mathbf{r}_i \), depends on the geometry of the system.

The addition of an external torque to Eq. (2), due to a uniform magnetic field will be studied at the end of the paper.

FIG. 2. (color online) (a-b) \( C(s) \) (from numerics) of a square plaquette relaxing into the vortex configuration (a) with \( \Delta = 1.7 \) for several values of \( T \) and (b) with \( T = 10^{-2} \) for several values of \( \Delta \). The shape of \( C(s) \) is linked to time and energy scales. (c) numerical (in red and cyan) and analytical (in blue and black) results of \( C(s) \) of a square plaquette with \( \epsilon_1 = 10^{-1} \) for several values of \( \Delta \). (d) analytical results comparing the early evolution of \( C(s) \) for several values of \( \epsilon_1 \). (e) \( C(s) \) comparing the relaxation of edge (red) and bulk (blue) dipoles at the sites of the lattice in the gs with \( \Delta = 2.2, T = 6 \times 10^{-2} \).
Time scales and time correlation function. Eq. (2) allows to identify four meaningful times scales. The relaxation time of the angular velocity from the inertial and damping contributions set $\tau_1 \equiv \frac{\tau}{\eta}$. The angular relaxation time from the damping and the internal field set $\tau_2 \equiv \frac{\tau}{\mu b}$. The time scale given by the rate between inertia and the internal dipolar fields set $\tau_3 \equiv \sqrt{\frac{\tau}{\mu b}}$. Finally $\tau_{th} \equiv \frac{n}{kT}$ weighs thermal up to damping energies. Here, the minimum time scale is set by $\tau_3$. Notice that $\tau_2$ and $\tau_3$ depend on the interaction between dipoles, which, for fixed $m_0$, is determined by $\Delta$ and the magnetic configuration. Because $K_i^2 \approx \frac{\mu b m_i}{\Delta^3}$, $\tau_2 \approx \frac{\eta \tau}{\mu b m_0}$ and $\tau_3 \approx \left(\frac{\tau}{\mu b m_0}\right)^{1/2} \Delta^{3/2}$ showing that the system dynamics is certainly affected by $\Delta$. Consider Eq. (2) in its dimensionless form:

$$\frac{d^2 \alpha_s}{ds^2} = 2 \sqrt{2} \xi_s(s) - \frac{d \alpha_s}{ds} - \tau_1 \alpha_s,$$

where $s \equiv \frac{t}{\tau_1}$ is the dimensionless time and $\tau_1 \tau_2 = \frac{1}{\eta^2}$. The rescaled gaussian noise has the same statistics as $\xi(t)$ but now $\xi(s)$ has no units. $\xi_0 = \frac{1}{\tau_1 \tau_2} = \frac{\eta b \tau}{\mu b m_0}$ is the rescaled thermal noise. Time $\tau_1 \ll \tau_2$ in Eq. (3) causes that $\frac{\xi_0^2}{\eta^2} \ll \Delta^3$ become a suitable criterion to define a dipolar array as a weakly interacting system (or a strongly damped one) in terms of its lattice constant.

As a non interacting limit consider the thermal relaxation of an isolated dipole right after an initial weak perturbation has taken it away from equilibrium. Its dimensionless Langevin equation reads:

$$\frac{d^2 \alpha_s}{ds^2} = 2 \sqrt{2} \xi_s(s) - \frac{d \alpha_s}{ds},$$

that corresponds to an Ornstein-Uhlenbeck process [21] with mean square rotation $\langle \delta \alpha(s)^2 \rangle = \epsilon_0(s - 1 + e^{-\tau_1 s})$, where $\delta \alpha_s(s) = (\alpha(s) - \alpha(0))$. The thermal relaxation of the dipole can be captured through the autocorrelation function,

$$C(s) = \langle \hat{\mathbf{m}}(s) \cdot \hat{\mathbf{m}}(0) \rangle = \Re(e^{i \delta \alpha(s)}),$$

(4)

Because $\delta \alpha(s)$ is linear in the noise and $\xi(s)$ has a Gaussian distribution, $\delta \alpha(s)$ is also Gaussian with a zero mean and a second moment $\langle (\delta \alpha(s))^2 \rangle$ [22]. For a gaussian variable $x$, with a mean $\mu_x$ and a variance $\sigma_x$, $\langle e^{iA} \rangle = e^{iA \mu_x - \frac{A^2 \sigma_x^2}{2}}$ and therefore $C(s) = e^{-\frac{(\delta \alpha(s))^2}{2}} = e^{-\alpha(s-1+e^{-\tau_1 s})}$. For short times $t \ll \tau_1$, $C(s) \sim e^{-\epsilon_1 s^2}$, while for long times $t \gg \tau_1$, $C(s) \sim e^{-\epsilon_0 \sigma^2}$. Under restricted temperature conditions, the early relaxation may be slowed down by increasing the damping to inertia quotient.

Thermally active relaxation and interactions. In the macrospin approximation, the four dipoles in Fig. 1(a) may represent the unit cell of a square lattice of mesospins. After thermal relaxation we find that the system settles into the magnetic vortex configuration shown in Fig. 1(a) (or its time reversal). Consider, the dipole highlighted in red in Fig. 1. We denote its magnetic moment $\mathbf{m}_1$. The torque along the z direction on $\mathbf{m}_1$ due to the other three magnets is $(\mathbf{m}_1 \times \mathbf{h}_1) = m_0 h_1 \cos(\beta_1 + \alpha_1) - h \sin(\beta_1 + \alpha_1))$, where $\beta_1$ is its equilibrium angle and $\alpha_1$ is a small angular deviation. In the vortex configuration of Fig. 1(a), $\beta_1 = n \pi$ (n integer), $h_1$ cancels out and $|h_1| = m_0 \mu_0 \Lambda$ with $\Lambda = \frac{(1+6\sqrt{2}}{8\sqrt{\alpha + \Delta^2}}$ a geometrical factor due to the lattice. The square plaquette has four oscillation modes. In the lowest energy mode, parallel dipoles oscillate in phase and small deviations out of the equilibrium barely change $\Lambda$. Thus $\sin(\beta_1 + \alpha_1) \sim \alpha_1$ along with $\frac{h_1}{h_1}((\alpha_1) \rightarrow 0$ produce that at the mean field level $|\langle \mathbf{m}_1 \times \mathbf{h}_1 \rangle|^2 = \Lambda m_0^2 \mu_0 \Lambda_1 = \kappa^2(\Delta) \alpha_1$. Symmetry ensures that $\Lambda$ and the mean field torque $\kappa^2(\Delta)$ are equivalent for all the dipoles in the square plaquette.

Eq. (2) can be solved by constructing the green function that verifies $i \partial_t \mathcal{G} + \eta \mathcal{G} + \kappa \mathcal{G} = \delta(t - t')$ as shown in the supplemental information [23]. Indeed, we can use $\mathcal{G}$ to find the mean square oscillations of the dipoles for small angular deviations:

$$\langle \delta \alpha(s)^2 \rangle = \epsilon_1 - \epsilon_1 \frac{\Lambda}{kT} \left(1 - \cos(\zeta s) + \zeta \sin(\zeta s) + \zeta^2 \right) e^{-s},$$

where $\epsilon_1 = \frac{\tau}{kT}$ and $\zeta^2 + 1 = 4 \beta_1^2$. For $\tau_1 \geq \frac{\tau}{kT}$, and for short times $t \leq \tau_3$, the time autocorrelation function becomes,

$$C(s) = e^{-\frac{s^2}{2} + \frac{1}{4} \left[1 - \cos(\zeta s) + \zeta \sin(\zeta s) + \zeta^2 \right] e^{-s}},$$

(5)

which in the limit of weak interactions yields $C(s) = e^{-\frac{s^2}{2} + \frac{1}{4} \zeta^2} e^{-s}$ and the single dipole limit is recovered.

When the condition $\tau_1 = \tau_2$, is met, the dynamics of the
plaque changes from a weak to a strong interacting regime. At zero temperature, this transition would occur for $\Delta^* = a + (a + \Delta)A^{1/3}\ell$. While $(a + \Delta)A^{1/3}$ collects the geometrical aspects of the lattice, $\ell = (\frac{\mu_0m_0^2}{\eta^2})^{1/3}$ sets a new length scale, which determines a magnetic correlation length on account of the magnetic degrees of freedom intrinsic properties, such as inertia, damping and the intensity of the magnetic moments. An array can be categorized in the strongly correlated regime when $\Delta \ll \ell$. While damping contributes to reduce $\ell$, inertial effects increase the correlation between magnets, which is also enhanced by increasing the intensity of their magnetic moments.

To test and complement these results, molecular dynamics simulations using the Verlet algorithm have been performed for several values of $\Delta$ and $T$ and for fixed $J$, $\eta$ and $m_0$. Details can be found in the supplementary information [23]. Numerical calculations along with the evaluation of Eq. (3) yield the results shown in Fig. 2 and Fig. 3. Hereinafter temperature $T$ is measured in units of $K^z(2a)$ and $\Delta$ in units of $a$. Fig. 2(a), shows the numerical solution of $C(s)$ for a lattice that relaxes from a disordered state with fixed interactions ($\Delta = 1.7$) at several $T$. The evolution of $C(s)$ is linked to the time scales of the system (and therefore gives hints on its interactions, inertia and damping). As depicted in Eq. (3) the dynamics is triggered by $\epsilon_1$, the ratio between thermal and dipolar interactions. Larger temperatures (red and blue curves) decrease $\tau_{th}$, increase the rescaled thermal noise $\epsilon_1$ and precipitate the departure of the initial magnetic configuration. The subsequent evolution, before $C(s)$ has reached its minimum, is controlled by inertia and interactions and lasts $t \sim \tau_3$. Next, the compromise between damping and interactions carries the system back to the equilibrium vortex configuration after a time $t \sim \tau_2$ has elapsed. In Fig. 2(a) $\tau_2$ and $\tau_3$ are the same for all curves because $\Delta$ remains constant. Fig. 2(b), shows the effect of variations of $\Delta$ for fixed values of temperature ($T = 10^{-1}$) in numerical simulations. Here, the increment of $\tau_3$ with the growing of $\Delta$ (and decrement of interactions) is apparent. Fig. 2(c) shows the agreement between the numerical solution of $C(s)$ and Eq. (5) for short times, $t < \tau_3$, when interactions are tuned and $\epsilon_1$ is kept constant. In each curve $\Delta$ has a different value and $T$ is adjusted in order to maintain the value of $\epsilon_1$ fixed. Finally Fig. 2(d) shows Eq. (5) for several values of $\epsilon_1$ confirming the numerical findings of Fig. 2(a).

Four plaquettes: Edges versus bulk. Aimed to compare the relaxation dynamics of edge and bulk magnets we study a small lattice made out of four square plaquettes of dipoles as shown in Fig. 1(c). From a disordered magnetic configuration, this cluster relaxes into two types of magnetic orders: the antiferromagnetic vortex state denoted $g_1$ and shown in Fig. 1(c) and the two vortex state named $g_2$, Fig. 1(d). Whether the outcome is $g_1$ or $g_2$ will depend on $\Delta$ and $T$ as summarized in the phase diagram of Fig. 1(b). This figure is result of the numerical solution of Eq. (2) for temperatures in the range $T \in (4 \times 10^{-2}, 1)$ and $\Delta \in (0.5, 2.2)$. The phase diagram shows that for $T \lesssim 10$, the $g_1$ (red points) is favored while for $\Delta \gtrsim 1.7$ the $g_2$ (blue crosses) is the preferred configuration at intermediate and large temperatures. Energetics dictates that at $T = 0$ the $g_1$ is slightly favorable for $\Delta < 1.2$ (see supplementary information for details [23]). However finite temperatures overcome this small difference and the two magnetic configurations become metastable states.

The small lattice serves to compare the thermal dynamics of edge and bulk dipoles. Because of point symmetry, edge dipoles will sustain an anisotropic internal field. That causes that the edge of the sample be more susceptible to external perturbations than its bulk. As a mode of illustration consider the dipoles highlighted in red at the edge and bulk of the lattices shown Figs. 1(c) and d). The bulk magnet of Fig. 1(c) feels a net magnetic field parallel to its magnetic moment due to its nearest collinear dipole since at its position the field due to all other magnets cancels out. The edge dipole also feels the field due to its nearest collinear magnet, however this is attenuated by the contributions due to the other four parallel dipoles. Therefore, when the system is subject to a small perturbation, a magnet at the edge responds faster to the external torque than one at the bulk. A similar situation occurs for the dipoles of Figs. 1(d) in the $g_2$. This anisotropy in the magnetic field splits $K^z$ into $K^z_E$ and $K^z_B$ for edge and bulk states respectively affecting directly the thermal relaxation by shifting $C(s)$ of bulk dipoles respect to the one belonging to dipoles at the edge. Indeed, Fig. 2(e) shows in blue and red curves the time autocorrelation function of bulk and edge dipoles respectively in the case of $g_2$ ($\Delta = 2.2$ and $T = 6 \times 10^{-2}$). As expected, numerical simulations (main figure) show that dipoles at the edge loose memory faster than those at the bulk of the lattice. The inset, that corresponds to the analytical solution exposes the same distinction. Magnetization dynamics. Finally we investigate how thermal and dipolar fields define the response of these magnetic arrays under a uniform external magnetic field. To this effect consider a lattice that, after thermal relaxation, has set into the $g_1$, and a second one that has set into the $g_2$. The resulting magnetization along the $x$ direction, due to an external magnetic field applied along the $x$ axis, is shown in blue and red curves of Fig. 5 for the $g_1$ and $g_2$ respectively. The plateau at $m_z = \frac{1}{2}$ which is manifested in both curves deserves special attention. This feature is due to the anisotropy of the internal dipolar interactions between bulk and edges dipoles as discussed above. Indeed the insets of Fig. 5 show the dissimilar magnetization dynamics of bulk (top inset) and edge (lower inset) dipoles for lattices in the $g_1$ (in blue) and $g_2$ (in red). While dipoles at the edge
respond smoothly and coordinately to very small values of $b$ the four dipoles at the bulk of the lattices stay pinned longer until at $b \sim 1.8$ they suddenly rotate to follow the direction of the external field. Since they correspond to one fourth of the total, their action leaves a signature in the form of a plateaux in the magnetization loop. The width of the plateaux quantifies the differences between the internal field at the edge and at the bulk of the sample. The small shoulder at $b \sim 2$ in the red curves of the main figure and upper inset is due to the slightly delayed flip of one of the bulk dipoles in the less interacting but warmer lattice in the gs2 [23].

Conclusions. Simple dipolar systems have been used to elucidate the role of intrinsic interactions and intrinsic lattice features in the thermal relaxation and magnetization dynamics of mesospin lattices. We find that the early relaxation dynamics of the systems is determined by temperature, dipolar interactions and inertia while the long time relaxation is defined by the interplay between damping and magnetic couplings. Therefore, lattice intrinsic properties such as magnetic couplings, damping and inertial aspects are imprinted in the time scales that determine the evolution of the time autocorrelation function. The study of the Langevin dynamics allows to define a magnetic correlation length $\ell$ in terms of inertia, damping, magnetic intensity and the distance between spins. For the case of mesospins nanoarrays, $\ell$ could be a useful length scale to compare with the lattice constant, in order to determine whether or not internal correlations play a dominant role in the dynamics of the system at hand. For fixed interactions temperature allows the manifestation of magnetic configurations forbidden at zero temperature. Signatures of such metastability show up in the magnetization loops which also reveal qualitative differences in the dynamics of edge and bulk states, which we find are due to the anisotropy of the internal dipolar magnetic fields.

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