

Stretched exponential magnetic relaxation dynamics in artificial square ice revealed through x-ray photon correlation spectroscopy

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X-ray photon correlation scattering measurements are undertaken on a thermally active artificial spin ice based on the square lattice, referred to as artificial square ice, to probe the fluctuation timescales as a function of temperature as the system passes through the paramagnetic-antiferromagnetic phase transition, which belongs to the two-dimensional Ising universality class. In the paramagnetic regime, a single exponential timescale is seen, whereas at and below the critical temperature, a stretched exponential decorrelation is observed, with the stretching exponent decreasing from unity down to below one-half as the temperature reduces. This trend is confirmed by kinetic Monte Carlo simulations of a simplified point-dipolar square ice system, and is in agreement with past theoretical work on the kinetic Ising model where stretched exponential relaxation due to equilibrium domain wall dynamics below the critical temperature was found.

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I. INTRODUCTION

Artificial spin systems are magnetic metamaterials that have attracted significant attention in the last two decades as they exhibit collective magnetic phenomena that can be tuned by design [1–10]. An artificial spin system is created by arranging nanosized islands of a ferromagnetic (FM) material on a substrate in close proximity using state-of-the-art lithography techniques. These nanomagnets typically have in-plane magnetization and an elongated shape that, due to magnetic shape anisotropy, orients the total magnetic moment in one of two directions parallel to the nanomagnet long axis, giving it an Ising-like degree of freedom. The bistable nanomagnets interact through the magnetic stray field (often approximated via the leading order point-dipole interaction) and, depending on the particular geometry of the patterned two-dimensional nanomagnet lattice, artificial spin systems can exhibit ice-like physics and/or phase transitions to long-range ordered states. This versatility and tunability by design enables direct comparison between experimentally observed configurations and predictions from Monte Carlo and kinetic Monte Carlo simulations of emergent magnetic behavior [11–18].

To probe the equilibrium statistical mechanics of artificial spin systems, the nanomagnets need to be superparamagnetic with their net moment (or macrospin) able to switch rapidly within the time frame of the experimental measurement. This allows the ensemble of nanomagnets to explore different configurations and potentially exhibit collective ordering driven by dipolar interactions, potentially leading to a phase transition. To achieve this, the blocking temperature of the thermally active nanomagnets must be below the temperature range associated with the transition. When frustration is present, this is difficult to achieve, as the energy associated with the critical temperature is often only a small fraction of the characteristic interaction energy. Routes to access the critical temperature involve decreasing the nanomagnet size and thickness [17,19] or introducing an interfacial Dzyaloshinskii–Moriya interaction [20].

Artificial square ice, with elongated nanomagnets arranged on a square lattice as shown in Fig. 1(a), has been an important focus of the community [1] with a number of experimental and theoretical papers concerning the phase transition [2,4,5,7,18]. It has been theoretically established that square ice exhibits a continuous transition at the Néel temperature T_N belonging to the two-dimensional Ising universality class (henceforth called Ising-2) from a paramagnetic to an antiferromagnetic (AFM) phase with the lowest-energy state characterized by an alternating array of clockwise and anticlockwise loops of head-to-tail moments on the square plaquettes of four nanomagnets [11]. The low temperature square-ice phase has been observed with imaging techniques including magnetic force microscopy [1] or synchrotron x-ray photoemission electron microscopy [13,21]. Examples of the

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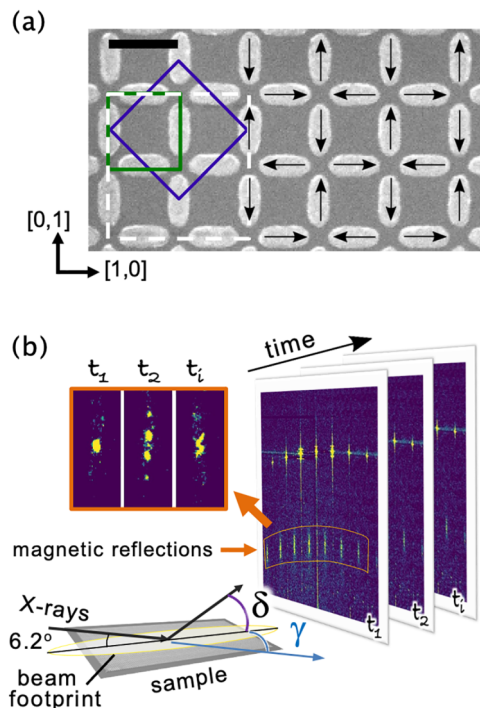


FIG. 1. (a) Scanning electron microscopy image of a region of an artificial square ice. The scale bar corresponds to 170 nm. The nanomagnets are 3 nm thick. Black arrows indicate the magnetic moments of the nanomagnets in one of the two possible ground state configurations. The area delimited by the continuous green square is the structural unit cell with $a = 170$ nm. The blue square corresponds to the smallest magnetic unit cell with edge $a\sqrt{2}$, which is associated with the low temperature AFM ordered state characterized by alternating handedness of loops of head-to-tail of magnetic moments on square plaquettes. To be consistent with previous reports [17], our indexation of the observed diffraction peaks corresponds to the structural unit cell. With this indexation, the magnetic peaks are described with half-integer Miller indexes, which reflect the enlarged magnetic unit cell outlined with a white dashed square. (b) Schematic of the XPCS experimental geometry where a coherent beam scatters off the sample. In this configuration, we are particularly sensitive to the magnetic correlations along the beam path. The incident angle of the beam (6.2°) creates an ellipsoidal beam footprint that illuminates a large fraction of the artificial spin ice. An example of an image on the detector with the structural peaks (top row) and magnetic peaks (bottom row, within orange box) is shown. An example of the intensity distribution of one of the magnetic peaks, recorded at three different times steps (t_1 , t_2 , t_i), is shown in the top-left corner of this panel.

observed AFM domain configurations characteristic of the square-ice phase, such as alternating clockwise and anticlockwise loop structures, can be found in Refs. [13,22,23].

Real-space imaging is inherently limited in its ability to probe fluctuations, particularly in terms of quantifying their temporal dynamics. In particular, the restricted field of view and long acquisition times hinder the collection of statistically significant information on fluctuation timescales near T_N . As a result, confirming the universality class of the phase transition based solely on real space imaging is challenging. In contrast, synchrotron x-ray scattering-based techniques offer

access to magnetic correlations in reciprocal space [24–27] and have proven valuable for characterizing the equilibrium properties of artificial spin systems [19,28]. Notably, with this technique, Sendetskyi *et al.* [17] confirmed the Ising-2 universality class in artificial square ice by analyzing critical scattering as a function of temperature in the vicinity of the AFM ordering peak, extracting static critical exponents consistent with Ising-2 behavior. While such scattering methods provide important insights into equilibrium thermodynamics, they generally do not resolve the fluctuation time scales of microscopic magnetic dynamics, such as macrospin reversals or domain boundary motion, which underlie the relaxation processes near and below T_N . In a paramagnetic phase, fluctuations are typically governed by a single, temperature-dependent relaxation time. In contrast, below T_N , systems in the Ising-2 universality class are expected to exhibit a more intricate magnetic dynamics due to domain wall dynamics [29–34]. Furthermore, the phenomenon of critical slowing down near T_N leads to a divergence of characteristic time scales, further complicating the dynamic response [35].

To directly probe fluctuation timescales, x-ray photon correlation spectroscopy (XPCS) [36,37] can be employed. XPCS utilizes coherent x-rays to enhance the visibility of speckle patterns—granular intensity fluctuations that arise from the interference of scattered waves—enabling the study of temporal and spatial correlations in a wide variety of systems, which might have disordered, glassy, or magnetically ordered phases exhibiting dynamic fluctuations [36,38,39]. From extended observation of such fluctuating speckle patterns, one can extract intensity-intensity correlations and momentum-transfer-dependent relaxation time scales. In terms of the evolution of fluctuations on atomic scales, XPCS has been extensively applied to determine the dynamics associated with aging phenomena in colloidal gels and glasses [37], as well as with transport in bulk-metallic glasses [40]. It has also been used to study the equilibrium dynamics near phase transitions [41–44], as well as the dynamics of antiferromagnetic domain walls in chromium [36]. Finally, XPCS has also been applied to artificial square ice [45–47]. Here, a particularly intriguing result for magnetic fluctuations within the low-temperature ordered phase suggested some form of ballistic spin transport below T_N characterized by a *compressed* exponential function [47], which is in contrast to theory and simulations that predict a *stretched* exponential function [29–31].

In this work, we employ XPCS experiments in combination with kinetic Monte Carlo simulations to probe the dynamics of an artificial square ice above and below the critical temperature, T_N , called the Néel temperature. As in Ref. [47], we find a simple exponential behavior in the high-temperature regime above T_N , reflecting a single characteristic timescale for paramagnetic fluctuations. This behavior is in contrast to that found below T_N , where we observe more complex fluctuations characterized by a stretched-exponential with a stretching exponent value close to that theoretically predicted for the kinetic Ising-2 model [29–31].

The paper is organized as follows: in Sec. II, we outline the sample fabrication and describe the XPCS technique, including the theory and the experimental measurements. In Sec. III, we present the experimental results, which are discussed and

compared to simulations in Sec. IV with a brief conclusion in Sec. V. In Appendix A we give a detailed description of the Monte Carlo simulations using a simplified dipolar square-ice model and, in Appendix B, we provide additional diffraction data as a function of time at temperatures below and above T_N .

II. EXPERIMENTAL METHODS

A. Sample fabrication

Arrays of nanomagnets arranged in the prototypical square geometry were fabricated using electron beam lithography. A 70 nm-thick polymethylmethacrylate (PMMA) layer was spin-coated on a Si(100) substrate. The patterns were exposed in the resist using a Vistec EBPG electron beam writer operated at 100 keV accelerating voltage. After development, a 3 nm-thick Permalloy ($\text{Ni}_{80}\text{Fe}_{20}$) film was deposited by thermal evaporation and subsequently capped with approximately 3 nm of Al to prevent oxidation. The remaining resist and unwanted metallic material were removed by ultrasound-assisted lift-off in acetone.

The nanomagnets had a stadium shape with a length of 120 nm and a width of 50 nm, as shown in Fig. 1(a). With a lattice parameter of $a = 170$ nm and 252×108 unit cells [one unit cell is indicated by the dark green square in Fig. 1(a)], the resulting dimensions of the artificial square ice were $42.8 \times 18.4 \mu\text{m}^2$. The past work of Sendetskyi *et al.* [17] suggests that, with the above nanomagnet dimensions, T_N will be approximately 360 K and the blocking temperature will be well below room temperature. Thus, on reducing the temperature, the system will be thermally active during and below the transition to long-range order until the blocking temperature is reached.

B. X-ray photon correlation spectroscopy

To probe the evolution of the magnetic domains, we performed resonant coherent XPCS measurements at the 23-ID-1 beamline of the National Synchrotron Light Source II. The x-ray beam was sent through a $10 \mu\text{m}$ pinhole placed roughly 6 mm upstream of the sample, providing a transversely coherent illumination of $\sim 15 \times 15 \mu\text{m}^2$ at the sample surface. The longitudinal coherence length is limited by the beamline optics and is on the order of $2 \mu\text{m}$. The energy of the coherent beam was set to 707 eV, corresponding to the Fe L_3 resonant absorption edge to ensure magnetic sensitivity. The sample was mounted on a liquid helium flow cryostat part of the TARDIS scattering chamber (referred to as a diffractometer in the context of x-ray experiments) and oriented so that the x-ray beam was incident at a glancing angle of $\theta = 6.2^\circ$ with the geometry illustrated in Fig. 1(b). At such a small incidence angle, the projection of the beam along the scattering plane was elongated, resulting in a beam size of $\sim 90 \times 15 \mu\text{m}^2$ on the sample surface.

With all diffractometer angles equal to zero, the $[0,1]$ lattice direction [indicated in Fig. 1(a)] was parallel to the x-ray propagation direction. During the experiment, the center of the detector was positioned close to the specular (zeroth order) reflection. The location of the scattered intensity on the detector, shown in Figs. 2 and 3, can be described by two angles: δ (vertical direction) and γ (horizontal direction)

[Fig. 1(b)]. When $\gamma = 0$, δ represents the total scattering angle 2θ . In the images retrieved from the 2D detector, we refer to the solid angle subtended by the detector using the angles δ and γ , which define the angular coordinates of each detector pixel relative to the direct beam. While this angular mapping approximates the actual geometry, it does not fully account for the curvature of reciprocal space because of the detector's flat geometry. However, this approximation introduces only a small correction and does not affect the interpretation of the experimental data within the angular range of interest.

Diffraction from the artificial square ice yields several Bragg peaks visible in the field of view of the camera [see Fig. 1(b)]. Because of the chosen experimental geometry, the detector covers an area in the reciprocal space that is about five times more extended in the horizontal direction than in the vertical one. This distortion arises from the nonlinear relationship between the angular coordinates measured by the detector and the momentum transfer components in reciprocal space. This leads to an elongation of the scattered signal along the vertical direction, which corresponds to the angle δ . The peaks at $[\frac{1}{2}, \frac{1}{2}]$, and multiples thereof, indicate the doubling of the structural unit cell along both in-plane directions through the emergence of long-range magnetic order, with the magnetic unit cell indicated by the dashed white box in Fig. 1(a). These magnetic peaks are spatially clearly distinct from the Bragg peaks originating from the underlying structural nanomagnet lattice [48], and we have shown the emergence of such peaks due to long-range magnetic ordering in a previous work [28].

Speckle patterns were collected at six different temperatures between 337 and 364 K on heating the sample [see Fig. 2(a)] using synchrotron x-rays with polarization perpendicular to the scattering plane (σ polarization). Each time the temperature was changed, we waited 30 minutes to ensure that any temperature gradients across the cryostat were minimized and did not cause a drift in the position of the sample during XPCS measurements. The scattered x-rays were collected using a fast CCD detector [49] with a readout rate of 10 Hz (i.e., an acquisition time of 100 ms) and a $30 \times 30 \mu\text{m}^2$ pixel size, placed at a distance of 340 mm from the sample. At each temperature the measurement time t was 500 seconds. Thus, at each temperature, 5000 speckle patterns were recorded (a selection of frames for different temperatures are shown in the Appendix B).

With XPCS, the intensity-intensity temporal correlation function, $\langle I(p, t)I(p, t + \Delta t) \rangle_t$, is probed. This is derived from the fluctuating speckle pattern within a reciprocal space region that is most sensitive to the dynamics under investigation. For artificial square ice, the region of interest is centered on one of the $[\frac{1}{2}, \frac{1}{2}]$ -family AFM peaks, as indicated by the white box in Fig. 3(a). From the measured intensities $I(p, t)$, the XPCS intensity-intensity temporal correlation function $g^{(2)}(\mathbf{q}, \Delta t) - 1$ can be obtained by averaging over both the measurement time t and pixels p in the region of interest

$$\begin{aligned} g^{(2)}(\mathbf{q}, \Delta t) - 1 &= \frac{\langle \langle I(p, t)I(p, t + \Delta t) \rangle_t \rangle_p}{\langle \langle I(p, t) \rangle_t^2 \rangle_p} - 1 \\ &= [g^{(1)}(\mathbf{q}, \Delta t)]^2. \end{aligned} \quad (1)$$

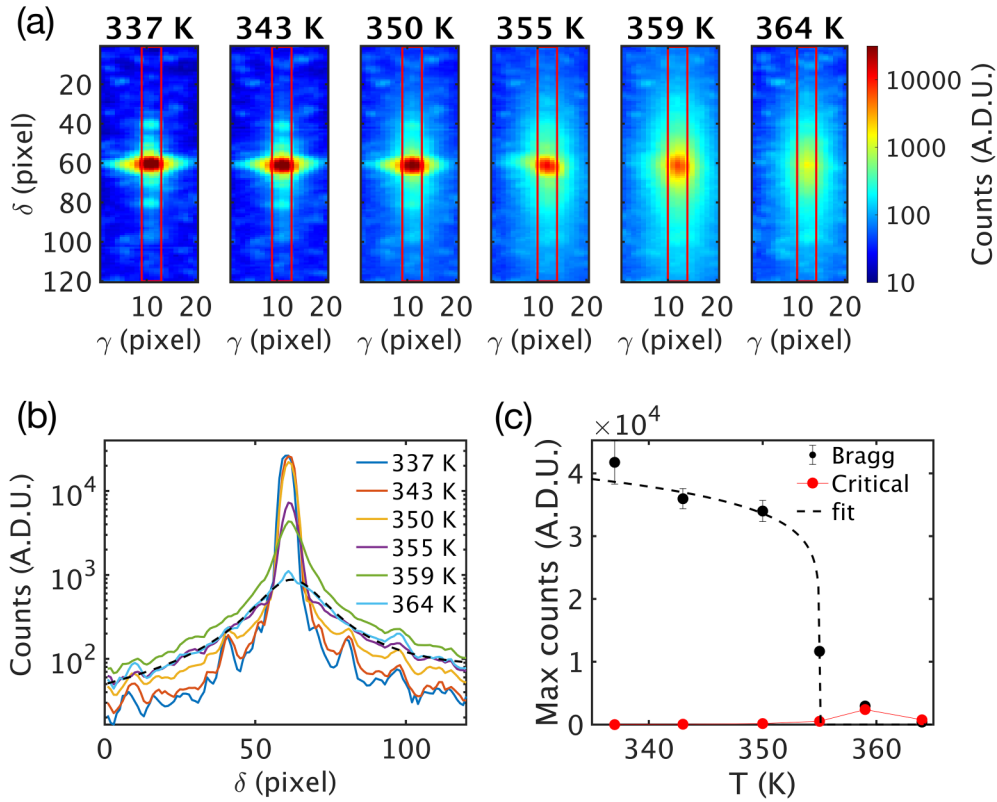


FIG. 2. (a) Time-averaged intensity of one of the magnetic $[\frac{1}{2}, \frac{1}{2}]$ diffraction peaks at different temperatures across the phase transition. The intensity corresponds to the number of counts in analog-to-digital units (A.D.U.). Red rectangles indicate regions that were used to obtain the peak intensity profiles shown in panel b. (b) Intensity profiles along δ of the time-averaged magnetic peak for each temperature. The side peaks adjacent to the main peak are attributed to the finite lateral extent of the illuminated patterned area. The experimental peak profile was fitted with the sum of a Voigt function and a Lorentzian function. For the Voigt function, we kept its full width at half maximum (FWHM) fixed to the value obtained for the lowest measured temperature. From the Lorentzian width, the correlation length of the antiferromagnetic domains is estimated to be $\simeq 1 \mu\text{m}$ in the vicinity of T_N . (c) Temperature dependence of the amplitude of the magnetic Bragg peaks (Voigt function) and critical scattering peaks (Lorentzian function) obtained by the fitting procedure. Due to the scarcity of the data points, only an approximate value of the Néel temperature T_N could be determined, which we estimated to occur between 355 and 360 K.

The quantity $g^{(1)}(\mathbf{q}, \Delta t)$ represents the normalized two-particle temporal correlation function at scattering vector $\mathbf{q}$ and time interval Δt . It is related to the dynamics of the system through $[g^{(1)}(\mathbf{q}, \Delta t)]^2 = f_S |F(\mathbf{q}, \Delta t)|^2$, where the function $F(\mathbf{q}, \Delta t)$ describes how density fluctuations at a given length scale (set by the scattering vector $\mathbf{q}$) evolve over a time interval Δt , and is referred to as the intermediate scattering function [50]. The intermediate scattering function is essentially the spatial Fourier transform of the Van Hove spatiotemporal correlation function [50–52], which characterizes how a collection of spins (macrospins in artificial spin ice) evolve in reciprocal space as a function of time. The constant f_S is the so-called contrast or Siegert factor, whose value is dictated by the x-ray coherence properties, scattering geometry and x-ray penetration depth [53,54].

An important step in gauging the reliability of XPCS data is assessing the level of baseline noise, which can obscure or mimic genuine dynamic signals. Even in the absence of intrinsic sample dynamics, small intensity fluctuations may still appear due to experimental artifacts such as mechanical vibrations, detector noise, or beam instability. These fluctuations manifest themselves as residual signal in the autocorrelation

function and must be carefully characterized to avoid misinterpreting the noise as dynamics. This is typically carried out by comparing $g^{(2)}(\mathbf{q}, \Delta t)$ values from the region of interest to those from a control region where no dynamics are expected, such as a charge peak. Our estimates suggest that this baseline noise leads to intensity variations of approximately 8% of the mean intensity value, which translates to a $g^{(2)}(\mathbf{q}, 0) - 1$ value of approximately 0.005.

III. EXPERIMENTAL RESULTS

To confirm that the transition temperature T_N lies within the considered temperature range between 337 and 364 K, we averaged the 5000 speckle patterns taken at each temperature to obtain the peak that would result from a standard x-ray resonant magnetic scattering experiment. Such average scattering patterns for different temperatures are shown in Fig. 2(a). Scattering distribution profiles were extracted from the red-outlined region of interest by averaging the γ -line profiles taken at every δ increment corresponding to the pixel width, and these profiles were fitted with a linear combination of a Voigt [55,56] and a Lorentzian peak profile [Fig. 2(b)].

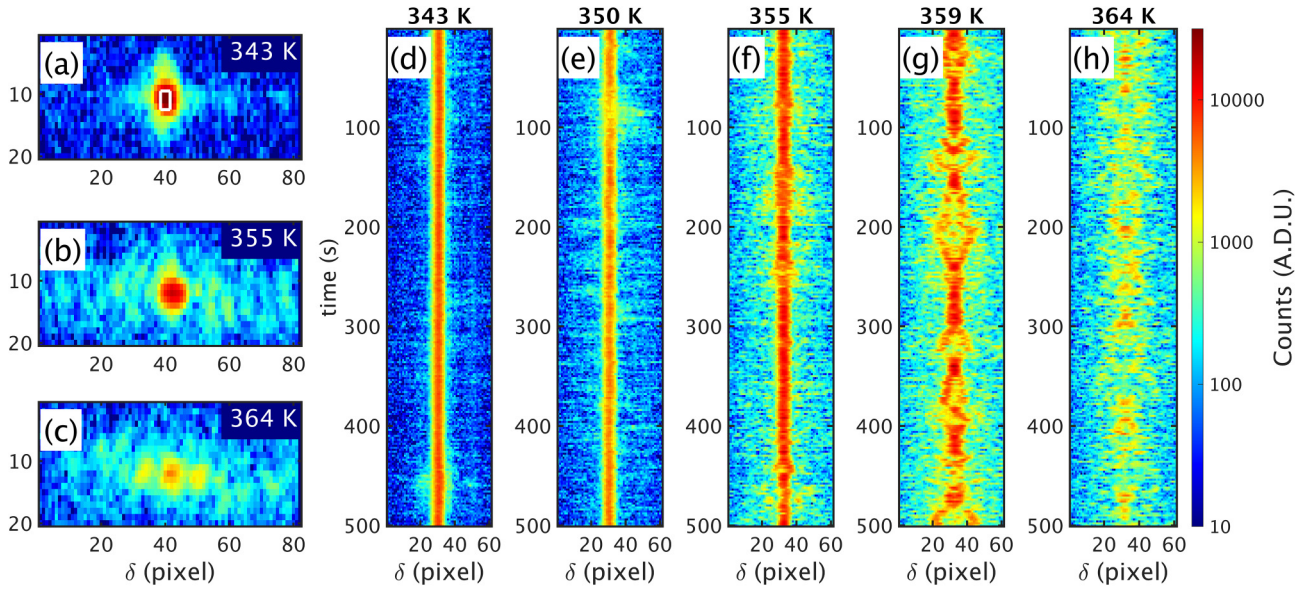


FIG. 3. (a)–(c) Speckle patterns observed for a selected AFM Bragg peak at three different temperatures. At 343 K, a well-defined AFM peak is observed. The region of interest used for the $g^{(2)} - 1$ calculation is highlighted with a white box in (a) and has the same size and location for all temperatures. In (b) and (c), we observe an increase in the number of speckles with increasing temperature. (d)–(h) Waterfall plots showing the time evolution of the scattering intensity—with the associated variation of the speckle size and positions—for selected temperatures. Each horizontal line represents a cut through the center of the AFM peak, corresponding to the intensity vs pixel position δ for the corresponding time t . At the two highest temperatures, there is a marked change in the variation in the intensity distribution from one line to the next, indicating fast fluctuations.

The amplitudes of the Voigt and Lorentzian peaks correspond to the Bragg and critical scattering contributions, respectively, and their temperature dependence is shown in Fig. 2(c). On decreasing the temperature, the AFM Bragg peak intensity shows a sharp increase at a temperature of 355 K (black dots), indicating the onset of the long range order. The corresponding Ising-2 transition temperature T_N is determined to lie in the range 355–359 K, as further supported by the peak observed in the critical scattering component (red dots), centered around 359 K. It is worth noting that a more accurate value of T_N and the scaling exponents associated with the critical scattering can be obtained from dedicated synchrotron x-ray measurements, as demonstrated in Ref. [17]. Here, we instead focus on the magnetic dynamics associated with the intensity fluctuations within the speckle patterns.

Examples of individual scattering patterns recorded over 100 ms are displayed for three temperatures below, at, and above the estimated T_N in Figs. 3(a), 3(b), and 3(c), respectively. The observed patterns arise from coherent magnetic x-ray scattering by magnetic domains and domain boundaries present in the sample. Since we are using collimated coherent radiation, interference between x-rays scattered from different magnetic domains within the sample produces a speckle pattern at the detector plane. This speckle pattern encodes the spatial distribution of the domains and, when tracked over time, reveals their evolution over time. As the temperature is increased, the thermal fluctuations increase, with the domain boundaries moving and the domains themselves evolve in shape and number over time, leading to corresponding changes in the speckle number, size, and position over time. At 343 K, Fig. 3(a), there is a relatively confined peak centered at $[\frac{1}{2}, \frac{1}{2}]$ reflecting a well-developed long-range AFM order

with one large ground state domain. At 355 K, close to T_N , the emergence of a richer speckle pattern reflects the disruption of the AFM domain into several smaller ones with different shapes and sizes [Fig. 3(b)]. Finally, at 364 K [Fig. 3(c)], the speckle pattern appears over a broader region of reciprocal space, reflecting the complex interference arising from a larger ensemble of magnetic domains that are, on average, smaller and more heterogeneous in size than at lower temperatures.

The temporal evolution of the speckle patterns can be visualized by plotting their time-dependent line profiles as waterfall plots [Figs. 3(d)–3(h)]. At the lowest measured temperatures, 337 K (not shown) and 343 K [Fig. 3(d)], the speckle pattern shows only minimal variation from line to line in the waterfall plot, with intensity fluctuations that are comparable to the estimated baseline noise.

This indicates that the magnetic configuration is largely static on the timescale of the measurement. In contrast, when temperatures approach the transition temperature T_N , the number of speckles, as well as their average distance from the $[\frac{1}{2}, \frac{1}{2}]$ peak position increase [see Figs. 3(e) and 3(f)], indicating that magnetic fluctuations become strong enough to visibly impact the speckle pattern, with the increase in structure beyond the central diffraction peak indicating more scattering centers in the form of multiples domains. At the highest considered temperatures above T_N [Figs. 3(g) and 3(h)], disperse speckles covering several pixels appear intermittently, varying in number, shape, and position over time. This temporal variation indicates strong fluctuations; nonetheless, each frame exhibits a well-defined speckle pattern consistent with residual short-range antiferromagnetic order, originating from small, spatially confined domains. It

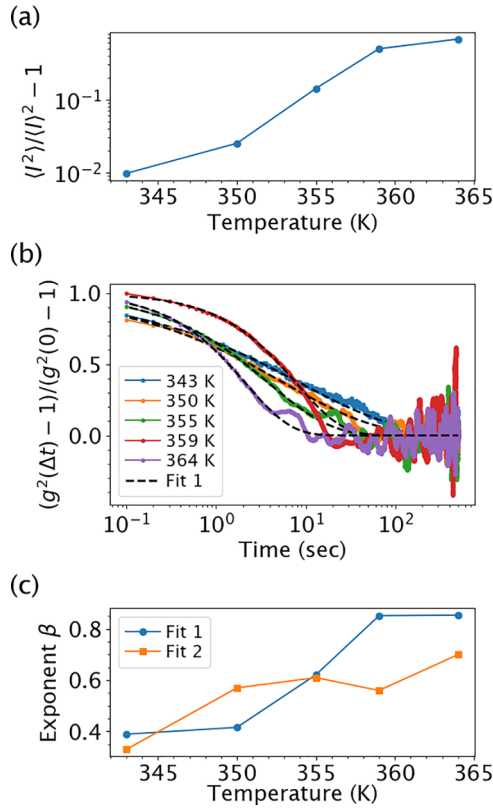


FIG. 4. (a) Contrast factors for the considered temperatures given by Eq. (2). (b) Normalized experimental $g^{(2)}(\mathbf{q}, \Delta t) - 1$, determined from speckle patterns at different temperatures using the reciprocal space region-of-interest centered around the selected AFM Bragg peak indicated with the white box in Fig. 3(a). The dashed black lines are fits to the normalized experimental $g^{(2)}(\mathbf{q}, \Delta t) - 1$ using the stretched exponential function given by Eq. (3). (c) Fitted values of the stretching exponent β as a function of temperature. Results from Fit 1 and Fit 2 are shown as blue circles and orange squares, respectively.

should be noted here that the speckle size on the detector is set primarily by the illuminated footprint and beam coherence, and therefore is not a useful indication of the domain size.

The temperature dependence of the observed speckle contrast factor is shown in Fig. 4(a). This quantity, not to be confused with the speckle contrast f_s , can be obtained from Eq. (1) for $\Delta t = 0$ and serves as a measure of the observed coherence or decorrelation of the fluctuations, and is proportional to the variance of the intensity fluctuations, which depends on both pixel position and time, as follows [37]:

$$g^{(2)}(\mathbf{q}, \Delta t = 0) - 1 = \frac{\langle I^2 \rangle_{p,t}}{\langle I \rangle_{p,t}^2} - 1. \quad (2)$$

This quantity approaches f_s in the limit of a frozen system giving static speckles and perfect coherence and decreases when dynamics decorrelate the speckles on short timescales. Indeed, we see that, as the temperature is reduced from the paramagnetic regime, the contrast factor rapidly dropped from ~ 0.6 in the paramagnetic regime at 365 K to ~ 0.01 with the onset of long-range magnetic order at 343 K. This decrease in the contrast factor corresponds to an increase in fluctuation

timescales, which is to be expected as long-range order sets in. The development of long-range magnetic order is also exemplified by the well-defined diffraction peak and fairly constant time trace in Figs. 3(a) and 3(d), respectively. In this low-temperature regime, the absence of measurable changes in the speckle pattern over time suggests that the fluctuation time scales extend well beyond that of the experimental measurement time of 500 seconds. As a result, the system does not explore different configurations during the measurement, and only a narrow subset of the most probable intensity values is sampled, leading to a small variance in the value of the scattering intensity, which results in $\langle I^2 \rangle \approx \langle I \rangle^2$ and thus a small speckle contrast, with $g^{(2)}(\mathbf{q}, 0) \sim 1$.

The normalized experimental correlation functions $(g^{(2)}(\Delta t) - 1)/(g^{(2)}(0) - 1)$ were derived at the different temperatures using Eq. (1) and are shown in Fig. 4(b). For the lowest temperature of 337 K (not shown), the speckle contrast was very close to the experimental noise level and the extracted correlation curve showed nonmonotonic undulations, so lack of convergence, at all time scales, which prevents further analysis. At temperatures between 343 and 364 K, the calculated intensity autocorrelation functions show consistent and stable behavior for time delays up to approximately 10 seconds, indicating reliable statistics within this time window. For time delays $\Delta t > 10$ seconds, the behavior becomes nonmonotonic, reflecting increased statistical noise. This is due to the limited total acquisition time (500 seconds), which reduces the number of available data pairs for larger Δt , thereby decreasing the statistical certainty of the correlation function. Much longer observations, at the timescale of several hours and beyond, would be required to improve the convergence of the correlation function—a timescale not possible because of the limited beamtime available.

Although the correlation function $g^{(2)}(\mathbf{q}, \Delta t) - 1$ does not converge well at longer times, it can still be meaningfully fitted with a stretched exponential function,

$$f(t) = \exp[-(t/\tau)^\beta], \quad (3)$$

giving reliable estimates of the stretching exponent β . This is justified by the mathematical property $\partial f / \partial \beta = -(\tau/\beta) \log(t/\tau) \partial f / \partial \tau$, which shows that the function is more sensitive to changes in β than changes in the relaxation time τ for times Δt much smaller than τ , where the data is more reliable. The use of the stretched exponential form [Eq. (3)] has the following physical interpretation: β quantifies the spread of relaxation rates, with $\beta = 1$ indicating a single timescale τ , while $\beta < 1$ signals a broad distribution typical of cooperative or hierarchical dynamics such as domain-wall fluctuations. In this latter case, τ reflects the longest timescales. In our experiment, the limited time window and increased statistical noise at large $\Delta \tau$ mean that τ may underestimate the true relaxation time, whereas β can still be extracted robustly from the well-converged short-time decay.

We consider two strategies to obtain β . The first focuses on the part of the decorrelation function which is monotonic (Fit 1), and the second focuses on only the data within the first second ($0 < \Delta t < 1$ sec) (Fit 2). Fit 1 is shown as a black dashed line for each temperature in Fig. 4(b),

revealing a reasonable agreement with experiments at shorter time intervals. Figure 4(c) displays the resulting optimal stretched exponents for the different temperatures. This shows that, for Fit 1 (blue circles), β decreases from close to unity to a lower temperature value of between 0.3 and 0.5 as the system transitions from paramagnetism to AFM long-range order. A similar trend is also observed for Fit 2 (orange squares), although the high temperature value is further from unity than for Fit 1. The two fitting strategies probe different parts of the dynamics: Fit 1 includes the full monotonic portion of the correlation function and thus can reflect a combination of fast and slower processes, while Fit 2 is restricted to $\Delta t < 1$ s, so it is dominated by the fastest dynamical components. This difference could be a possible explanation of the different forms of the $\beta(T)$ values. However, given the limited number of temperatures and the restricted Δt window, we rather refrain from overinterpreting the difference between the two $\beta(T)$ curves. The fitted values of the relaxation time parameter τ (not shown) all have the same order of magnitude (2–10 s), and show no systematic trend with temperature. This last aspect will be discussed further in the next section.

IV. DISCUSSION AND COMPARISON TO SIMULATION

We now turn to a comparison between the experimentally observed temperature dependence of the stretching exponent in our XPCS data and theoretical expectations derived from kinetic Monte Carlo simulations, aiming to assess the extent to which the model captures the underlying relaxation dynamics of the magnetic moment correlations. The observed temperature dependence of the stretching exponent seen in Fig. 4(c) is compatible with the known theoretical and numerical temporal behavior of the kinetic Ising-2 system [30,31]. Specifically, simulations have reported values of $\beta = 1$ above T_N down to $\beta = 1/2$ and $\beta = 1/3$ below T_N , which matches with our experimentally observed variation of β across the Néel transition in the measured temperature range. Indeed, the assertion that there is a stretched exponential behavior below the critical temperature T_N can be understood from the perspective of the spatiotemporal evolution of domains and their boundaries, which involves a continuum of relaxation timescales that depend sensitively on both system size and dimensionality (1D, 2D, 3D, etc.). In early theoretical work on the two-dimensional kinetic Ising system, a stretching exponent of either $\beta = 1/3$ [29] or $\beta = 1/2$ was predicted at temperatures below T_N [30,31]. Whilst $\beta = 1/2$ is considered to have a stronger theoretical grounding, with most simulations favoring this value, nonconservative dynamical simulations have revealed that both $\beta = 1/2$ and $\beta = 1/3$ exponents are possible [32–34]. Here “nonconservative” refers to simulations where local spin transitions do not preserve the global order parameter. This lack of clarity in the simulations concerning which exponent is correct might originate from the very strong dependence of the domain wall dynamics on the system size and shape (or aspect ratio) [57]. Finally, we note that recent theoretical work indicates a stretching exponent $\beta = 4/7$ at precisely T_N [35].

Insight into the present experimental observations can be further gained by directly calculating the intermediate scattering function $F(\mathbf{q}, \Delta t)$ of a model square-ice system with

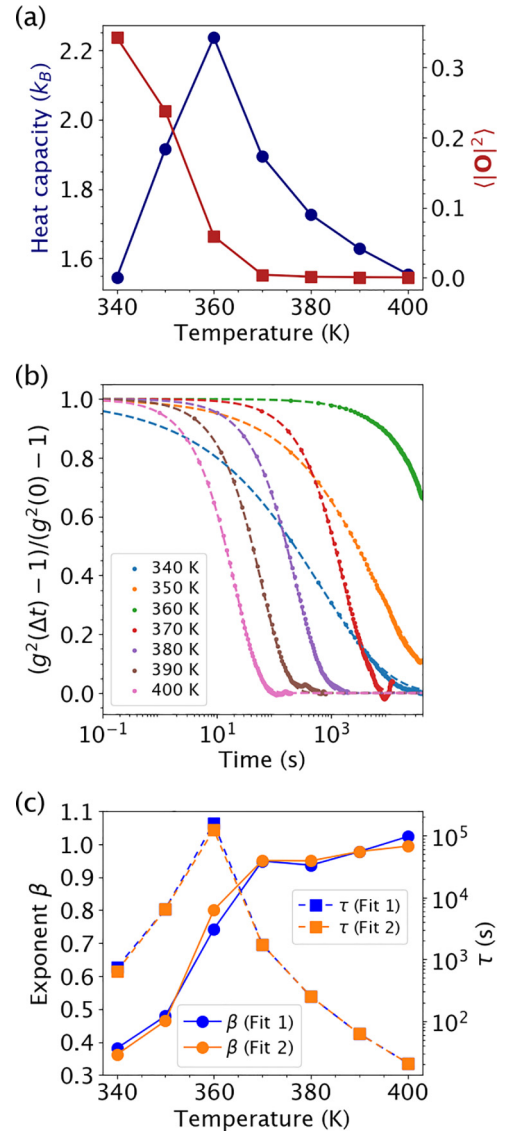


FIG. 5. Temperature dependent results from Monte Carlo simulations for a model square-ice system of the same size as in experiment, showing (a) heat capacity and AFM order parameter, and (b) kinetic Monte Carlo simulated magnetic decorrelation [taken as $\phi(t)$, see Eq. (5)]. (c) The corresponding best-fit stretched exponential parameters β and τ as a function of temperature.

dipolar-coupled nanomagnets. In past work, Monte Carlo methods revealed an Ising-2 phase transition for such a model [17] and we begin by repeating such equilibrium simulations for a system closely resembling the experimental one (details of the model system and how it is simulated are given in Appendix A). Our results are summarized in Fig. 5. In Fig. 5(a), we plot the equilibrium heat capacity and the static structure factor evaluated at the AFM wave vector $[\frac{1}{2}, \frac{1}{2}]$, or, equivalently, the intensive (per spin) quantity $\langle |\mathbf{O}|^2 \rangle / N$, where

$$\mathbf{O} = \sum_{i=1}^N \hat{\mathbf{m}}_i \exp(-i\mathbf{q}_{\text{AFM}} \cdot \mathbf{r}_i) \quad (4)$$

is the AFM order parameter of the system. Here $\mathbf{r}_i$ and $\hat{\mathbf{m}}_i$ are the position and orientation of the magnetic moment in the i th

nanomagnet, respectively. In Fig. 5(a), we see that the Monte Carlo simulations give a peak in the heat capacity at ~ 360 K corresponding to the Ising-2 transition from the paramagnetic phase to the low-temperature long-range ordered phase, which is also reflected in the corresponding increase in the AFM order parameter.

To simulate the temporal behavior of magnetic fluctuations, the kinetic Monte Carlo method is used to evolve the system stochastically as a function of time (see Appendix A). Whilst the entire XPCS experiment can be simulated to obtain $g^{(1)}(\mathbf{q}, \Delta t)$ via Eq. (1), it is more efficient to directly calculate the intermediate scattering function, $\langle \mathbf{O}^\dagger(t) \cdot \mathbf{O}(t + \Delta t) \rangle_t$, via the relaxation function

$$\phi(t) = \frac{\langle \mathbf{O}^\dagger(t) \cdot \mathbf{O}(t + \Delta t) \rangle_t - \langle \mathbf{O}^\dagger \rangle_t \cdot \langle \mathbf{O} \rangle_t}{\langle |\mathbf{O}|^2 \rangle_t - \langle \mathbf{O}^\dagger \rangle_t \cdot \langle \mathbf{O} \rangle_t}. \quad (5)$$

However, it is found that a well converged $\phi(t)$ can be only obtained using simulation timescales well beyond that of the 500 seconds probed by experiment. In Fig. 5(b), we show the time evolution of $\phi(t)$ for a range of temperatures, starting from Monte Carlo generated equilibrium spin configurations. We note that the total simulated time interval was several orders of magnitude longer than the experimental time scale of 500 seconds, where a time interval of 0.5 million seconds was simulated at the highest temperature of 400 K, and a time interval of 23 million seconds was simulated at the lowest temperature of 340 K. The dashed colored lines display the optimal stretched exponential fits for the corresponding temperatures and the stretching exponent β as a function of time is shown in Fig. 5(c). This was estimated using a fitting strategy similar to that employed for the experimental data (see Fig. 4 and the related discussion in Sec. III). Due to the better convergence achieved in the simulations, the optimal values of the relaxation time parameter τ can be extracted with confidence and are also shown in Fig. 5(c). Above T_N , the stretching exponent is close to unity indicating a single relaxation timescale for the paramagnetic regime whereas, below the critical temperature, the stretching exponent drops to the expected theoretical range of $\beta = 1/3$ to $1/2$. Therefore, we see that, for both the experimental and kinetic Monte Carlo simulation results, the artificial square ice, with its temperature-dependent relaxation behavior and the observed transition at T_N from stretched exponential to exponential dynamics, exhibits dynamics similar to those expected for a generic kinetic Ising-2 systems.

While there is an overall agreement between experiment and simulation in terms of relaxation dynamics reflected by the change in the critical exponent β as a function of temperature, a more detailed comparison is provided by considering the intermediate scattering function $\phi(t)$ across the temperature range. Although the precise value of the critical temperature is not identical in the simulations and experiment, the simulation nevertheless captures the essential features of the dynamics near the transition. This is particularly evident in the temperature-dependent behavior of $\phi(t)$, which displays a pronounced change in the decay behavior, with increasingly slower relaxation near the critical region. This is quantitatively captured by the fitted relaxation time τ from the Monte Carlo simulations shown in Fig. 5(c), which exhibits a clear peak near the critical temperature [corresponding to the peak in

the heat capacity shown in Fig. 5(a)]. The peak in τ , as seen in Fig. 5(c), reflects a divergence in relaxation timescales, a hallmark of critical slowing down, and highlights the progressive slowing down of dynamics as fluctuations become increasingly long-lived near the critical point. Such a broad distribution of timescales, spanning from approximately 1 to 10^5 s, is not observed in the experimental data shown in Fig. 4, where the fitted relaxation times τ (not shown) fall within the interval 2 to 10 s. In the case of the experiment, however, τ is not a reliable measure of the true relaxation timescale due to increasing noise and limited statistics at longer time intervals. As a result, the experimental fits are insensitive to the slow dynamics expected near the critical point, and thus do not capture the extended timescales associated with critical slowing down. This is because the fluctuations at large length-scales and timescales are extremely rare on the experimental timescale, and would therefore contribute negligibly to the contrast, given by Eq. (2), so that their signature is lost in the noise of the 500-second-long XPCS measurement.

We now discuss our findings with regards to previous XPCS experiments on artificial square ice [47], where a crossover from $\beta \sim 1$ to $\beta > 1$ was reported on cooling from the high temperature disordered phase to the AFM ordered phase. Such compressed exponential relaxation typically reflects avalanche-like, correlated rearrangements which drive a faster-than-exponential relaxation of the system [58], in contrast to the isolated, thermally activated events expected for $\beta \sim 1$. This markedly different behavior from our present findings might be due to a number of associated factors. For example, past Monte Carlo simulations suggested that the magnetic relaxation is strongly affected by the system size and shape as well as boundary effects [57]. Concerning the size, the system we have experimentally measured and simulated contains $2 \times 252 \times 108 = 54432$ macrospins (or Ising-like degrees of freedom), whereas in Ref. [47] a system of 2400–2500 nanomagnets is considered. In addition, the system experimentally investigated in Ref. [47] had an elongated shape optimized for the x-ray scattering experiment.

In order to elucidate the influence of the system size and shape on the magnetic relaxation dynamics, we have performed further Monte Carlo simulations on the following systems: I) a system with $2 \times 126 \times 54 = 13608$ macrospins, and two elongated systems with II) $2 \times 24 \times 100 = 5600$ and III) $2 \times 12 \times 100 = 2800$ macrospins. We found that System I shows a temperature dependence of the stretching exponent β similar (albeit, with slightly larger values) to that of our experimental system presented in Fig. 5(c). In contrast, Systems II and III display increasingly different behavior as the width of the system decreases. In Fig. 6 we display the fitted parameters τ and β obtained for System I as well as the elongated Systems II and III as a function of temperature. For the latter two systems, the temperature dependence of τ is qualitatively similar to that seen experimentally in Ref. [47], with no distinct signature of critical slowing down, having no decrease in τ with temperature and no peak in β that was seen for the artificial square ice with equal lateral dimensions in Fig. 5(c). Examining the temperature dependence of the stretching exponent β [blue and yellow lines in Fig. 6], we observe that decreasing the system width leads to a more modest reduction of β below T_N^{sim} . This suggests simpler

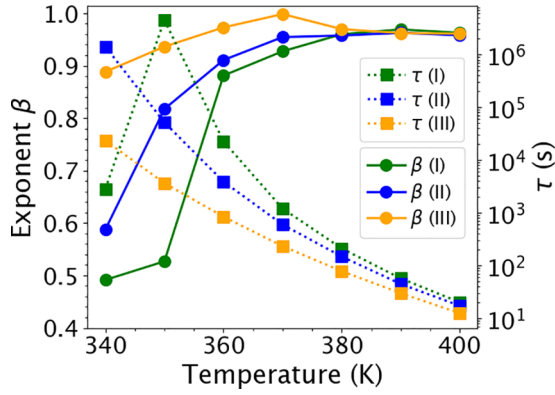


FIG. 6. Temperature dependence of the fitted stretched exponential parameters τ and β for the square ice Systems I, II, and III, comprising arrays of $2 \times 126 \times 54$ nanomagnets (same shape as in the experiment, but a factor of two smaller), and $2 \times 24 \times 100$, $2 \times 12 \times 100$ nanomagnets, respectively.

domain dynamics characterized by a narrower distribution of relaxation times compared to those in systems extended in both x and y directions. Whilst the narrowest system (System III) does exhibit a minor increase in β around T_N^{sim} , its behavior is still quite different from that reported by Ref. [47]. Thus, the observed discrepancies between our results and those of Ref. [47] cannot be attributed solely to system size. We speculate that, in the experimental system measured in Ref. [47], the evolution of domains and domain boundaries may be more constrained along one of the axes and more deterministic due to the ellipsoidal shape of the artificial square ice, potentially enhancing deterministic features in the dynamics. For such an elongated shape with a key-hole-like shape, certain regions might exhibit enhanced domain nucleation, leading to abrupt changes in domain boundary configurations that can be well described by a compressed exponential ($\beta > 1$). In addition, the magnetic dynamics may be influenced by the increased edge-to-area ratio in smaller or more elongated arrays, where boundary effects become more significant in determining the energetics and kinetics of domain evolution. These insights underscore the importance of considering the system geometry before attempting to generalize results across different, albeit similar, systems.

V. CONCLUDING REMARKS

We have employed x-ray photon correlation spectroscopy to investigate the temporal nature of thermally driven magnetic fluctuations in artificial square ice, a system undergoing a continuous Ising-2 phase transition to long-range antiferromagnetic order. In the paramagnetic regime, such fluctuations are characterized by exponential relaxation defining a single relaxation time-scale. For temperatures below the critical ordering temperature, T_N , a stretched exponential relaxation is found, with a stretching exponent close to that expected for the kinetic Ising-2 system [29–34] suggesting a fluctuation regime mediated by equilibrium domain wall dynamics. This result is confirmed by kinetic Monte Carlo simulations of a model dipolar interacting square-ice system, indicating that not only the statistical mechanics [17] but also the large-scale, slow dynamics (i.e., the long-wavelength behavior) is well described

by the Ising-2 model. The simulations also demonstrate that the current experimental timescale of 500 seconds would need to be increased by several orders of magnitude to probe the complete decorrelation of the domain wall dynamics as well as the signatures of critical slowing down at and around T_N .

Looking ahead, developments in high-coherence x-ray sources and detector technology, as well as novel approaches such as coherent correlation imaging [59] promise to further improve temporal and spatial resolution, potentially opening the door to resolve millisecond-scale collective dynamics such as monopole motion within ordered artificial spin ice phases. Such capabilities would deepen our understanding of emergent excitations and nonequilibrium behaviors in other artificial spin systems [8] as well as frustrated magnetic systems [60–63].

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DATA AVAILABILITY

The data supporting the findings of this article are openly available [64].

APPENDIX A: MODEL DIPOLAR HAMILTONIAN AND THE KINETIC MONTE CARLO METHOD

The artificial square ice can be well modeled by approximating the N nanomagnets as an array of point dipole moments, $\hat{\mathbf{m}}_i$ interacting via

$$E = -\frac{\mu_0 m^2}{4\pi} \sum_{i < j} \frac{1}{r_{ij}^3} [3(\hat{\mathbf{m}}_i \cdot \hat{\mathbf{r}}_{ij})(\hat{\mathbf{m}}_j \cdot \hat{\mathbf{r}}_{ij}) - \hat{\mathbf{m}}_i \cdot \hat{\mathbf{m}}_j]. \quad (\text{A1})$$

In the above, each $\hat{\mathbf{m}}_i$ can point in one of two directions, corresponding to magnetization aligned in one of two directions parallel to the long axis of the nanomagnet. The prefactor in Eq. (A1) may be written as the energy $\mu_0 m^2 / (4\pi a^3)$ when the distance in $1/r^3$ is taken in units of a . Here, a is usually taken as the nearest-neighbor distance of the lattice. For the simulations, the magnetic moment per nanomagnet, m , is derived from the product of a characteristic magnetization M and the known approximate volume of the nanomagnet V . For the present work, the magnetization is taken to be $M = 325 \times 10^3 \text{ Am}^{-1}$, a value that is similar to that used in past works [13,14,17], and the volume is $V = L \times W \times H = 120 \times 50 \times 3.0 \text{ nm}^3$. The array of nanomagnets contains $2 \times 252 \times 108 = 54\,432$ magnetic moments associated with the individual nanomagnets and open boundary conditions were used, reflecting the experimental geometry.

To determine the equilibrium thermodynamic properties of the above system as a function of temperature, ensemble Monte Carlo simulations were employed using, as trial moves, single nanomagnet magnetization reorientations. For the present work, the specific heat and the static structure factor $\langle |\mathbf{O}|^2 \rangle / N$ were calculated where, for the latter, $\langle \cdot \rangle$ refers to a converged thermal average.

To evolve the system as a function of time at a given temperature, the kinetic Monte Carlo (kMC) method is employed to determine dynamical quantities such as $\langle \mathbf{O}^\dagger(t) \cdot \mathbf{O}(t + \Delta t) \rangle_t$, where $\langle \cdot \rangle_t$ now refers to a converged temporal average. At a given moment in time, the transition rate for each possible magnetic moment reorientation, given by $\Gamma_k = \nu \exp(-E_k/k_B T)$, is calculated, where T is the temperature of the system. The barrier energy E_k for the k th magnetic moment reversal is given by $E_k = E_0 + (E_{k,\text{final}} - E_{\text{initial}})/2$.

Here E_0 is the intrinsic barrier energy of an isolated nanomagnet, and the second term is the configurational energy difference associated with the k th reorientation. The total reversal rate is then $\Gamma_{\text{total}} = \sum_k \Gamma_k$. The kMC iteration entails first considering the N possible spin reorientations—one for each spin in the system—each associated with a transition rate Γ_k . These possibilities are then sampled uniformly, with selection probabilities proportional to their respective rates. Once a transition has been selected and applied, the physical time is incremented stochastically by sampling the exponential distribution $\Gamma_{\text{total}} \exp(-t\Gamma_{\text{total}})$. The parameters used for the kinetic Monte Carlo simulations were $\nu = 10^9 \text{ s}^{-1}$ and $E_0 = 0.675 \text{ eV}$. The chosen prefactor value, ν , is what is typically taken for coherent reversal attempt rates and the barrier energy, E_0 , is comparable to that expected when taking shape anisotropy into account, again, assuming a coherent reversal.

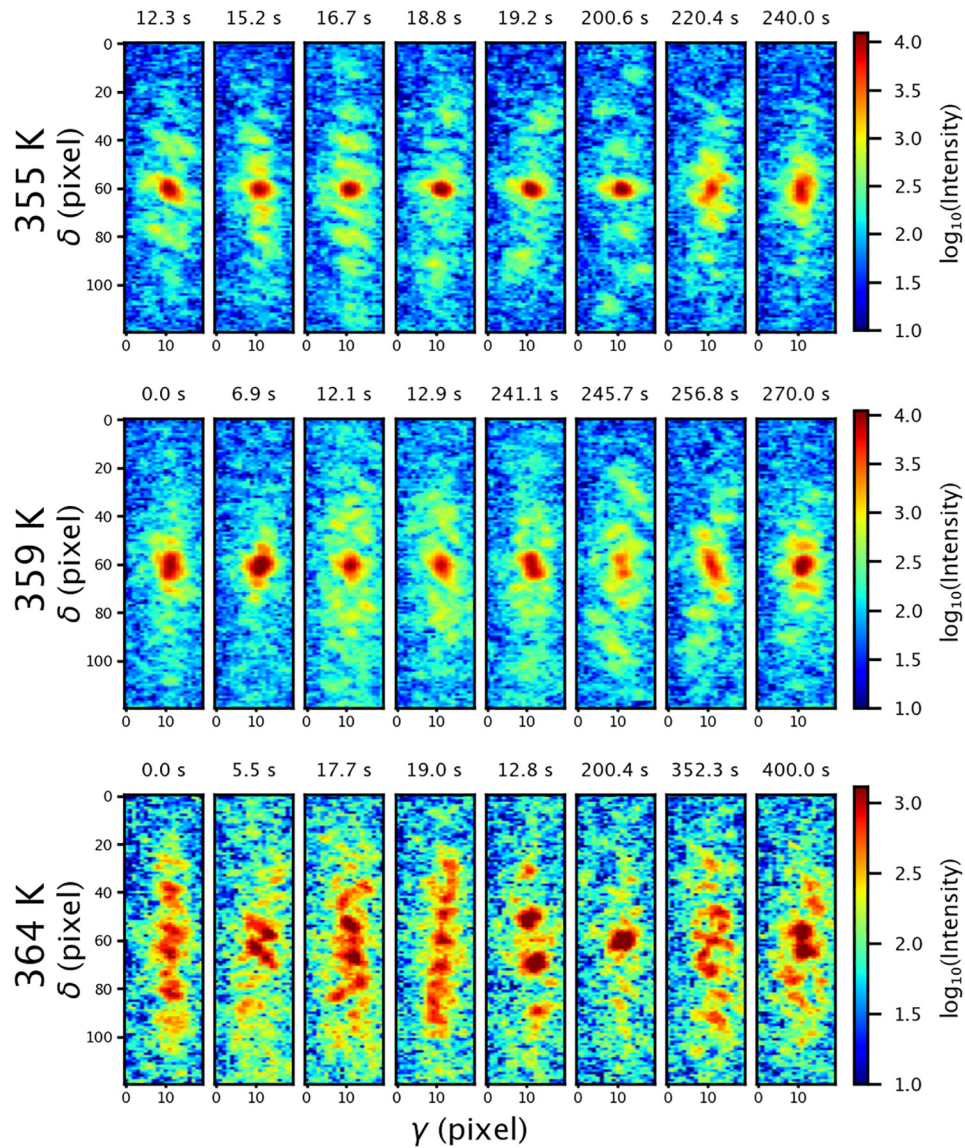


FIG. 7. Time series of speckle patterns recorded at three different temperatures near the Néel temperature T_N during XPCS measurements. Each row corresponds to a specific temperature (indicated on the left), and displays a sequence of snapshots acquired at different time points (in seconds, labeled above each image). The intensity is shown on a logarithmic color scale. These representative snapshots illustrate the temporal evolution and diversity of the speckle patterns observed in the vicinity of the magnetic phase transition.

For each chosen temperature, the starting configuration is obtained using standard site Monte Carlo simulations.

It should be noted that both the magnetization M and barrier energy E_0 are fine-tuned to obtain optimal quantitative agreement with the experimental timescales and temperature dependence. However, ultimately, such simplified models are at best expected to be qualitative. Indeed, the assumption of a simple Arrhenius model for thermally activated moment reversal, while capturing the macroscopic physics, is unlikely to be representative of the real microscopic reversal mechanisms.

APPENDIX B: REPRESENTATIVE SPECKLE PATTERNS

To illustrate the richness and variability of the scattering patterns observed in our XPCS experiments, we present in Fig. 7 a selection of representative speckle patterns acquired at temperatures close to the Néel temperature T_N . The observed variation in speckle patterns across snapshots reflects the succession of distinct, quasistatic magnetic configurations accessed over time, underscoring the complexity of the system's behavior near the magnetic transition.

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- [56] In general, separating the critical scattering from the Bragg contribution is challenging when their line shapes are not clearly distinguishable and deviate from simple Lorentzian or Gaussian forms. In our case, the peaks exhibit nontrivial shapes arising from finite-size effects and the system's dimensionality, which generates additional modulations in reciprocal space. Despite these caveats, we nevertheless attempted a separation in order to obtain a tentative estimate of the transition temperature.
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