

Ultrafast laser-induced magnetic relaxation in artificial spin ice driven by dipolar interactions

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It is of great interest to develop methods to rapidly and effectively control the magnetic configurations in artificial spin ices, which are arrangements of dipolar coupled nanomagnets that have a variety of fascinating collective magnetic phenomena associated with them. This is not only valuable in terms of acquiring fundamental understanding but is also important for future high-performance applications. Here, we demonstrate ultrafast control of magnetic relaxation in artificial square ice through femtosecond laser pulsed excitation, enabling rapid access to low-energy states via dipolar interactions. Time-resolved magneto-optical Kerr effect measurements reveal that, after laser-induced demagnetization, the magnetization recovers 60% of its original value within 40 picoseconds. During this brief time window, dipolar coupling drives a collective magnetic ordering. *Ex situ* magnetic force microscopy confirms the emergence of extended domains with the lowest-energy vertex configuration, characteristic of ground-state ordering, thus establishing ultrafast laser-driven relaxation as a route to attain the low-energy states. Through complementary energy barrier calculations and micromagnetic simulations incorporating Landau-Lifshitz-Bloch dynamics, we elucidate the underlying mechanism: transient ultrafast demagnetization followed by rapid remagnetization that enables a dipolar-driven collective rearrangement. Moreover, a tailored decreasing-fluence laser excitation protocol is shown to enhance ground-state ordering, consistently achieving over 92% ground-state vertex populations. This work opens the way to ultrafast and spatially selective control of magnetic states in artificial spin ice for spin-based computation and memory technologies, and highlights the critical interplay of thermal fluctuations, magnetostatic coupling, and transient magnetization dynamics.

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I. INTRODUCTION

The ability to manipulate the magnetic configuration of networks of nanomagnets is a key area of interest for advancing high-speed in-memory computation [1], nanomagnetic logic [2,3], neuromorphic and reservoir computing [4–6], and reprogrammable magnonic crystals [7–10]. Particularly appealing for such applications are artificial spin ices (ASIs), which are arrays of magnetically coupled single-domain nanomagnets arranged on the sites of different lattices. These provide a platform to explore various collective phe-

nomena, including magnetic frustration [11,12], relaxation processes [13], and phase transitions [14–16].

While the field has significantly expanded, now encompassing a range of different lattices and nanomagnet shapes [17], ASIs were originally conceived to mimic the behavior of their crystal spin ice counterparts: the rare-earth pyrochlores [18]. Here, stadium-shaped nanomagnets were arranged on the square or kagome lattices [11,19], referred to as artificial square ice or artificial kagome spin ice, respectively. The pronounced shape anisotropy associated with the elongated shape of the nanomagnets confines their overall magnetic moments – or macrospins – to point in one of two stable directions parallel to the nanomagnet long axis, resulting in Ising-like behavior. Importantly, in contrast to their crystal counterparts, ASIs allow direct real-space imaging of the magnetic configurations, enabling the observation of individual reversal events and collective dynamics with magnetic imaging techniques [19,20].

Control of the magnetic configuration, and hence energy, of ASIs is a key goal, with methods to allow magnetic relaxation being progressively refined [13,21–28]. With each advancement, the aim has been to achieve low-energy states

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more effectively, often as a means to access the ground state. The methods employed for this include demagnetization with a magnetic field [21,22], thermal annealing [23–26] and even patterning nanomagnets thin enough so that they are thermally active close to room temperature [13,20,27]. However, while these techniques are effective in terms of bringing the system to a lower energy configuration, they suffer from limitations in spatial control and operational speed [13,28]. To overcome these limitations, various light-induced methods for controlling the magnetization of the nanomagnet ensemble have been developed, including continuous-wave all-optical magnetic switching [29,30] and plasmon-assisted photoheating [1,31]. Although these techniques allow specific regions within an ASI to be addressed, they do not represent the fastest means of manipulating the magnetization, as they typically operate on time scales ranging from several nanoseconds to milliseconds. Achieving magnetization control on the sub-nanosecond time scale is particularly compelling, as it matches the characteristic time scale (GHz frequencies) of modern computing hardware, positioning ASIs as viable candidates for integration into high-speed, spin-based logic and memory technologies. In this context, laser-driven ultrafast demagnetization [32] with femtosecond pulses has emerged as one of the most promising methods for achieving magnetization control on sub-nanosecond time scales. However, so far, femtosecond laser experiments have been primarily employed to study ultrafast dynamics and all-optical switching in ferromagnetic and ferrimagnetic thin films, as well as in uncoupled magnetic nanostructures [33]. This raises the question of whether femtosecond-laser-induced magnetization dynamics could be harnessed to rapidly drive ASIs to their lowest energy state.

Here, we demonstrate that femtosecond laser excitation, as illustrated in Fig. 1(a), drives rapid magnetic relaxation of artificial square ices, enabling faster access to the ground state compared with existing methods [13,21–31]. Our results show that this relaxation process, governed by the magnetostatic (or dipolar) interactions between the nanomagnets, occurs on the same picosecond to a few 10's picosecond time scale as the magnetization recovery within each nanomagnet following a laser pulse. This is unexpected, as dipolar-driven collective relaxation is typically considered to be much slower [34]. Nevertheless, under these conditions, the system can reach a low-energy state in the exposed region. Our approach thus provides an order of magnitude increase in speed, while simultaneously offering spatial control over the resulting magnetic configuration, with local excitation of micrometer-scale regions, which is not possible with global thermal or magnetic-field-driven demagnetization protocols.

To better understand the mechanism of such fast relaxation of the artificial square ice, we employed a combination of Simplified and Improved String Method (SISM) energy-barrier calculations and Landau-Lifshitz-Bloch (LLB) micromagnetic simulations to model ultrafast demagnetization-remagnetization dynamics. These different simulation approaches each span different time and temperature scales, highlighting the critical relationship between the initial ultrafast demagnetization and the subsequent partial recovery of the magnetization within individual nanomagnets. We propose that the magnetization within nanomagnets must be

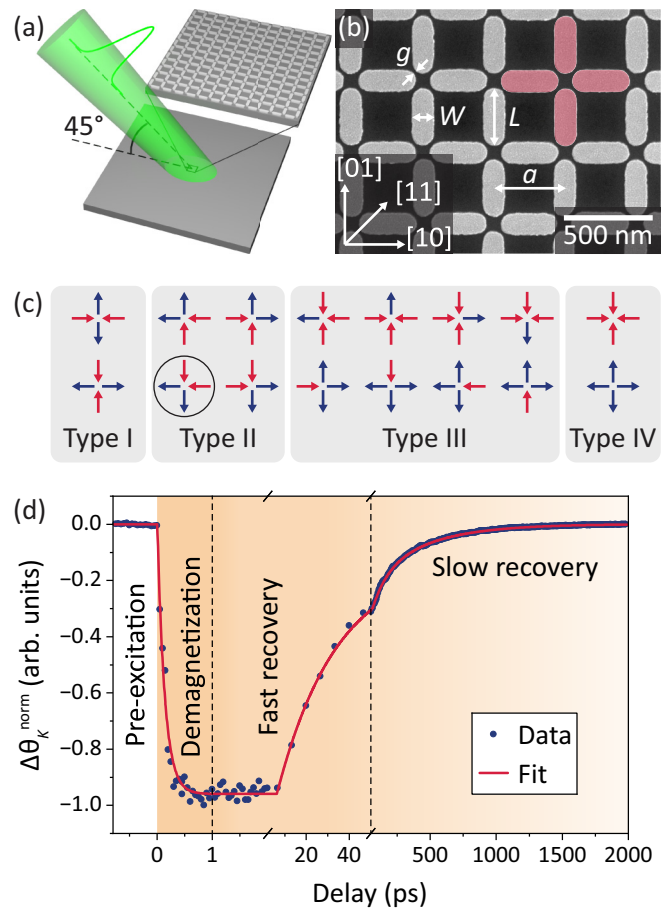


FIG. 1. Applying ultrafast pulsed laser excitation to an artificial square ice. (a) Schematic of the pulsed laser experimental geometry and sample layout. (b) Scanning electron microscope image of the artificial square ice ($L = 300$ nm, $W = 100$ nm, $a = 400$ nm, $g \approx 15$ nm), with a four-nanomagnet vertex highlighted in red. (c) Vertex types (Type I–IV) ordered in terms of increasing dipolar energy. Red (blue) arrows indicate macrospins pointing towards (away from) the vertex. The initial state of the artificial square ice used in the laser excitation experiments has all vertices in the Type II vertex configuration highlighted with a black circle. (d) Time-resolved changes in Kerr rotation representing the temporal evolution of the magnetization of the artificial square ice after pulsed laser excitation. The signal is normalized such that the Kerr rotation is zero at equilibrium and -1 at the point of maximum demagnetization. The vertical dashed lines demarcate the characteristic temporal regions after laser excitation: demagnetization, fast recovery, and slow recovery. The measurements were performed under an in-plane magnetic field along both the $[11]$ and $[\bar{1}\bar{1}]$ directions, enabling background subtraction and isolation of the magnetic signal.

sufficiently reduced on a short enough time scale to allow macrospin reorientation, while also recovering sufficiently quickly to re-establish dipolar interactions between nanomagnets in the artificial square ice. This transient regime, in which thermal fluctuations and magnetostatic interactions coexist, facilitates collective relaxation into low-energy configurations, as directly evidenced by the experimental observation of long-range magnetic order.

II. SAMPLE PREPARATION AND EXPERIMENTAL SETUP

Our samples consist of $200\text{ }\mu\text{m} \times 200\text{ }\mu\text{m}$ arrays of 8 nm-thick permalloy ($\text{Ni}_{80}\text{Fe}_{20}$) nanomagnets on a silicon (100) substrate, covered with a native oxide layer ($\sim 1\text{--}2\text{ nm}$ -thick) [Fig. 1(b)]. To fabricate the nanomagnets, permalloy thin films were thermally evaporated at a base pressure of $\sim 6 \times 10^{-6}$ mbars and patterned with electron beam lithography combined with lift-off. The nanomagnets have a length $L = 300\text{ nm}$ and width $W = 100\text{ nm}$, and are arranged on a square lattice with lattice constant $a = 400\text{ nm}$. Each nanomagnet is capped with a 2 nm-thick aluminum layer to prevent oxidation, and the smallest gap between neighboring elements is $g \approx 15\text{ nm}$.

First, time-resolved longitudinal magneto-optical Kerr effect (TR-MOKE) measurements were performed using an ytterbium-doped fiber laser in a pump-probe configuration, with a 200 kHz repetition rate, 1030 nm fundamental wavelength and a nominal pulse duration of 80 fs. These measurements were used to record the time evolution of the magnetization dynamics of an 8 nm-thick permalloy thin film following ultrafast laser excitation in a magnetic field [Fig. 1(d)]. S-polarized probe pulses were at the fundamental wavelength of 1030 nm, while the P-polarized pump (excitation) pulses with a wavelength of 515 nm were obtained via second-harmonic generation using a barium borate crystal. During TR-MOKE measurements, the average power of the pump beam was 5 mW (0.88 mJ/cm^2), while the average power of the probe beam was $300\text{ }\mu\text{W}$ (0.05 mJ/cm^2). Orthogonal polarizations of the pump and probe beams were used to minimize optical interference and enable selective detection of the Kerr rotation signal. To effectively distinguish the magnetic signal from the background noise, we performed these measurements with a magnetic field of $H = 500\text{ Oe}$ applied along both the $[11]$ and $[\bar{1}\bar{1}]$ directions for every data point. The two measurements were then subtracted to isolate the purely magnetic contribution and suppress non-magnetic (optical and thermal) background effects. The Kerr signal shown in Fig. 1(d) represents the normalized change in Kerr rotation measured following laser excitation, $\Delta\theta_K^{\text{norm}}(t) = \frac{\theta_K(t) - \theta_K^{\text{eq}}}{\theta_K^{\text{eq}} - \theta_K^{\text{min}}}$, where $\theta_K(t)$ is the Kerr rotation measured at a pump-probe delay t , θ_K^{eq} is the equilibrium Kerr signal before excitation (i.e., for $t < 0$), and θ_K^{min} is the minimum value of the Kerr signal reached after excitation, corresponding to the maximum degree of demagnetization. This normalization ensures that the signal is zero before excitation and reaches -1 at the point of maximum demagnetization. Although the longitudinal Kerr rotation is not a direct measure of magnetization in absolute units, it is, to first approximation, linearly proportional to the in-plane component of the magnetization in the probed volume. At equilibrium, the Kerr signal is set to zero to remove the static offset and highlight dynamic changes induced by the pump pulse. As shown in Fig. 1(d), the TR-MOKE signal exhibits a three-stage response: an ultrafast demagnetization on the sub-picosecond time scale, followed by a fast partial recovery and then a slower complete recovery with characteristic time constants of $\sim 30\text{ ps}$ and $\sim 300\text{ ps}$, respectively, obtained by fitting the recovery curve with a biexponential function.

This initial TR-MOKE characterization of the magnetization dynamics in a magnetic field is performed at low laser

fluence where the cumulative sample heating is minimal and switching of the magnetization is not expected. This serves as a reference point that allows the effects of laser exposure on the magnetic configuration of the ASIs to be interpreted. The ASIs were exposed with pump pulses with a pulse duration of 100 fs at the sample position, an incidence angle of 45° , a full width at half maximum laser spot size of $85\text{ }\mu\text{m}$, and a laser fluence between 6.1 mJ/cm^2 and 9.7 mJ/cm^2 (corresponding to an average laser power between 35 mW and 55 mW), in the absence of an external magnetic field, as illustrated in Fig. 1(a). All experiments were performed under ambient conditions.

The resulting magnetic configurations of the artificial square ices were subsequently imaged via *ex situ* magnetic force microscopy (MFM).

III. ULTRAFAST LASER-INDUCED MAGNETIC RELAXATION IN ARTIFICIAL SQUARE ICE

A convenient framework to describe the magnetic state of an artificial square ice is to consider the vertex populations. In the artificial square ice, four neighboring nanomagnets surround a lattice vertex in a crosslike arrangement, which is highlighted in red in Fig. 1(b). Four distinct vertex types, Type I to Type IV [Fig. 1(c)], with increasing dipolar energy, can be identified based on the orientation of the macrospins of the constituent nanomagnets. Although the total energy of the system arises from the full set of dipolar interactions, the relative populations of the different vertex types serve as an effective proxy for the energy of the system: the more low-energy vertex configurations that are present in the system, the closer the system is to its ground state. The lower energy Type I and Type II vertex configurations are both characterized by two macrospins pointing towards the center of a vertex and two pointing away, following the so-called “ice rule”, which is similar to that observed in a bulk crystal spin ice [18]. However, Type I vertex configurations have a lower energy than Type II vertex configurations because the dipolar interactions between the nanomagnets at a vertex depend on their relative orientations and separation, which are different for nearest-neighbor orthogonal pairs of nanomagnets and the parallel pairs of nanomagnets across the vertex [11,35]. As a consequence, the ground state of an artificial square ice consists of an alternating tiling of the two Type I vertex configurations, and this ground state is doubly degenerate. An array in which all vertices adopt the same Type II vertex configuration can be easily obtained by applying a magnetic field along any of the four $\langle 11 \rangle$ directions of the artificial square ice. Type III vertex configurations have a higher dipolar energy than Type II vertex configurations, with three macrospins pointing in and one pointing out, or vice versa. Type III vertex configurations typically appear as defects within the chains of Type II vertices that form at the boundaries between ground state domains [36]. Finally, Type IV vertex configurations are the highest in energy, with all the macrospins pointing in or all pointing out, but are usually not observed.

To monitor the extent to which laser excitation drives artificial square ice into low-energy configurations, the $200\text{ }\mu\text{m} \times 200\text{ }\mu\text{m}$ nanomagnet arrays are prepared in a well-defined configuration. This is achieved by applying a magnetic field,

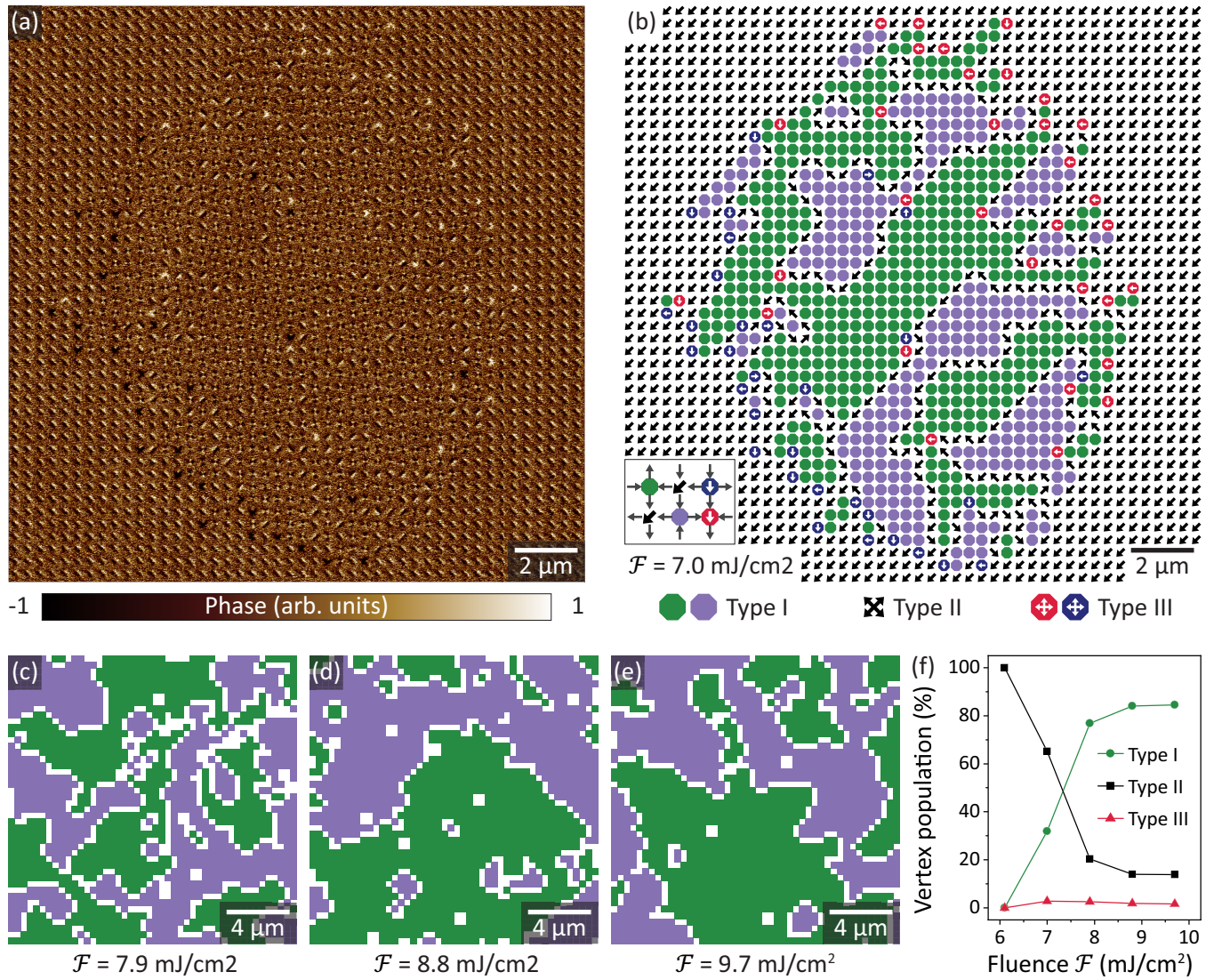


FIG. 2. Vertex configurations and populations in artificial square ice following laser excitation with a constant fluence over time. (a) MFM image and (b) corresponding map of the vertex configurations following laser exposure with a laser fluence of 7.0 mJ/cm². The different vertex types are color coded with symbols as indicated. Simplified maps of the vertex configurations are provided for the magnetic configurations of the artificial square ices exposed to laser fluences of (c) 7.9 mJ/cm², (d) 8.8 mJ/cm² and (e) 9.7 mJ/cm². Here, Type I symbols are replaced with squares of the same color to highlight the Type I domains; Type II and Type III symbols are omitted. (f) Populations of Type I, Type II, and Type III vertices as a function of laser fluence extracted from the maps of the vertex configurations. For each data point, the vertex population is given as a percentage of the total number of vertices within the 20 μm × 20 μm area scanned by MFM, i.e. including vertices that remain in the initial Type II vertex configuration outside the region where the local fluence exceeds the switching threshold, as seen for a laser fluence of 7.0 mJ/cm².

which is significantly higher than the coercive field of the nanomagnets, along the $[\bar{1}\bar{1}]$ direction. The field is then removed, so that the macrospins of the horizontal (vertical) nanomagnets shown in Fig. 1(b) point left (down). As a result, all vertices are in the same Type II configuration indicated with a black circle in Fig. 1(c). Subsequently, different copies of the same artificial square ice were exposed to a train of 100 fs laser pulses for 1 s at a repetition rate of 200 kHz (i.e., one pulse every 5 μs, with the laser effectively extinguished between laser pulses) with a given laser fluence. Each artificial square ice was exposed to a different, but constant, laser fluence, which was increased from 6.1 mJ/cm² to 9.7 mJ/cm²

in steps of 0.9 mJ/cm². Subsequently, the magnetic configuration of each artificial square ice was imaged using MFM over an area of 20 μm × 20 μm, which contains approximately 5000 nanomagnets. An example of a resulting MFM image is provided in Fig. 2(a), where each nanomagnet is associated with a bright and a dark contrast that result from the stray field associated with opposite magnetic poles at the ends of the nanomagnets. This contrast allows us to determine the orientation of the macrospin assigned to each nanomagnet. From this, the vertex configurations of the region are mapped as shown in Fig. 2(b), with each vertex type assigned a different color and symbol. Here, the two degenerate ground state

domains consisting of ordered Type I vertices are indicated by green and purple dots. Vertices in a Type II configuration are identified by a black arrow that indicates the direction of the net magnetic moment. Similarly, the net magnetic moment of vertices in a Type III configuration is indicated with white arrows, enclosed in red and blue dots to denote positive (three-in, one-out) and negative (one-in, three-out) magnetic charges, respectively. Here we are referring to the effective magnetic charge defined in the dumbbell model, where each nanomagnet is treated as a dumbbell with opposite charges at its ends [19,37]. The magnetic charge at a vertex is then the net charge associated with the four dumbbell charges residing at the vertex.

As mentioned above, we began by applying trains of laser pulses of different but constant fluence. At a fluence of 6.1 mJ/cm^2 , we observed no change in the artificial square ice. However, when we employed a fluence of 7.0 mJ/cm^2 , we saw reversal of macrospins in the central region of the area scanned with MFM, while the vertices near the edges remained in their initial Type II configuration [see Fig. 2(a)]. Since the laser pulses have a Gaussian energy distribution over the sample surface, reversal was only observed in the central region where the laser fluence was higher than the threshold required to reverse the macrospins within the nanomagnets. In the central region, we observe the laser-induced formation of Type I domains, reflected by the green and purple regions in the vertex configuration map in Fig. 2(b). The size of these domains remains relatively small, spanning only a few vertices in width. We ascribe this observation to the fact that the laser fluence is only slightly above the switching energy threshold, providing just enough energy to induce local switching but not sufficient energy to drive the formation of extended Type I domains. The vertex configuration map in Fig. 2(b) also illustrates how Type II vertex chains serve as boundaries separating Type I domains. Finally, we also observe vertices in a Type III configuration. These are primarily located at the interfaces between the unswitched Type II region and the newly formed Type I domains. Although these exposures were performed using 1 second-long pulse trains at 200 kHz to ensure stable excitation conditions, control experiments confirmed that magnetization reversal and Type I domain formation also occurs with a single femtosecond laser pulse, provided that the fluence is sufficiently high (details of the single-pulse experiments are given in Sec. I of the Supplemental Material [38]).

As the laser fluence was increased to 7.9 mJ/cm^2 , we observed that the magnetic moments in the entire $20 \mu\text{m} \times 20 \mu\text{m}$ area scanned with MFM received sufficient energy to reverse. This is shown in Fig. 2(c), where Type I symbols are replaced with squares of the same color to highlight domains with opposite nanomagnet polarization, while Type II and Type III symbols are omitted. In particular, the region with Type I domains increased in size, indicating that more nanomagnets could undergo reversal, and a lower-energy magnetic configuration was reached. Increasing the fluence to 8.8 mJ/cm^2 [Fig. 2(d)] promotes the formation of even larger Type I domains, bringing the system closer to its ground state, while a higher fluence of 9.7 mJ/cm^2 [Fig. 2(e)] did not appear to give a further significant change in the domain sizes. In order to quantify this, we determined the populations

of different vertex types, which allows us to gauge the degree of order in the system, and track their evolution with laser fluence, as shown in Fig. 2(f). Here, the vertex population for each of the three observed vertex types is given as a percentage of the total number of vertices within the $20 \mu\text{m} \times 20 \mu\text{m}$ area scanned by MFM. As the laser fluence is increased, the Type I vertex population increases, until it reaches approximately 80% at a fluence of 8.8 mJ/cm^2 . Increasing the laser fluence still further produces no substantial increase in the Type I vertex population. The percentage of Type II vertices, found in the domain boundaries, is significantly reduced to 20%, while the limited number of Type III vertices observed at low fluences remain close to zero. These vertex populations are comparable to those achieved through thermal annealing protocols, where similar Type I vertex populations are observed under optimal conditions [23,26]. However, we have achieved this on a significantly faster time scale.

In addition to the vertex population analysis discussed above, further inspection of the macrospin configurations reveals additional features. At low fluence [Fig. 2(b)], we observe that the domain boundaries (chains of Type II vertices) separating ground state domains exhibit a preferred orientation. This is reflected in the statistics of the four (*a priori* equivalent) Type II vertex configurations within these boundaries: 60% of Type II vertices have a net magnetic moment pointing to the bottom-left, compared to 22% having a net magnetic moment point to the top-left, 14% to the bottom-right, and 6% to the top-right. The orientation of the net magnetic moment is directly linked to the orientation of the domain boundaries. Therefore, the predominance of Type II vertices with a magnetic moment pointing to the bottom-left results in domain boundaries that tend to run from top-right to bottom-left, i.e., they retain a memory of the initial macrospin configuration.

This behavior can be understood by considering the way in which ground state domains form. We can assume that there is a distribution of switching energy barriers across the array of nanomagnets. At low fluence, only those nanomagnets with the lowest energy barriers can switch to give vertices with the lowest-energy Type I configurations that act as seeds for the local reorganization of the surrounding macrospins. Since this distribution can be considered uniform throughout the lattice, Type I domains can nucleate at random positions and expand until they meet other Type I domains. When two ground state domains with opposite chirality meet, the resulting Type II domain boundary tends to preserve the original orientation of net moments at the vertices, because reversing the associated macrospins would require additional energy. This also explains why very few Type II vertices (6%) are found with a net magnetic moment pointing to the top-right: obtaining this Type II configuration from the initial Type II configuration would require switching of all four macrospins at a vertex, whereas the other Type II configurations only require two macrospins to switch. At higher fluences [Figs. 2(c)–2(e)], this directionality is no longer observed because the macrospins in all nanomagnets are more likely to overcome their energy barriers and switch freely. In this regime, the domain boundaries can evolve without retaining a memory of the initial macrospin orientation.

In the low-fluence regime [Fig. 2(b)], since the effective excitation area is relatively small, there is a boundary between the nanomagnets that switch within the laser footprint and those that remain in their initial configuration outside it. As already mentioned, along this boundary, we observe an accumulation of Type III charges. Interestingly, the positive charges predominantly at the top-right of the laser spot and negative charges at the bottom-left. Assuming that Type IV vertices cannot form, this is the only configuration that can arise when a Type I domain nucleates, expands, and encounters a region that is still in the initial state. These defects therefore emerge purely as a result of the lattice topology, so that opposite charges naturally appear on opposite sides of the laser spot, with their exact location on the boundary depending on the orientation of the net magnetic moment in the initial configuration. Therefore, this separation of charges is another residual imprint of the initial configuration. At higher fluences [Figs. 2(c)–2(e)], this charge accumulation is no longer visible because the area in which the nanomagnets switch increases, so that the boundary between switched and unswitched regions lies outside the MFM scan area.

In summary, when sufficient energy is delivered to the artificial square ice via a femtosecond laser pulse, a significant macrospin dynamics is triggered that allows the system to reorganize into a lower-energy state. Given the ultrashort laser-sample interaction time (<200 fs) and the fact that magnetization recovery in the nanomagnets completes within 2 ns, as seen in Fig. 1(d), our results raise the question of when and how macrospin ordering occurs. Notably, the TR-MOKE measurements shown in Fig. 1(d) indicate that the system recovers approximately 60% of its original magnetization within just 40 ps following laser-induced demagnetization. This suggests that the macrospin rearrangement, which is governed by dipolar interactions, occurs during the fast magnetization recovery, i.e., on picosecond to a few 10's picosecond time scale.

Ideally, time-resolved imaging experiments [39] could be used to track the evolution of the artificial square ice configuration following laser excitation. However, real-space time-resolved imaging with techniques such as free-electron laser-based time-resolved x-ray magnetic circular dichroism-photoemission electron microscopy is highly challenging. In particular, these methods suffer from limited magnetic contrast and low signal strength under typical stroboscopic conditions, which would be exacerbated by the fact that the nanomagnets have in-plane magnetization and are only 8-nm thick.

Given the above constraints, we turned to an alternative approach: by systematically modifying the laser excitation protocol, we sought indirect insight into the underlying relaxation dynamics. For this, a protocol based on exciting the system with a series of laser pulses with a gradually decreasing fluence over time (total exposure duration of 1 s), from 9.7 mJ/cm^2 down to 6.1 mJ/cm^2 in steps of $13.2 \text{ } \mu\text{J/cm}^2$ (corresponding to 273 steps with ~ 3.7 ms per step), was implemented and this was found to significantly improve the degree of magnetic ordering compared to exposures with a constant laser fluence. While we did not systematically explore other protocols or step sizes, this fine decrement was chosen to ensure a gradual reduction in the energy imparted

by the laser, allowing controlled ordering of the magnetic moments. Conceptually, this strategy resembles simulated annealing [40], in which a gradual reduction in energy input enables the system to escape local energy minima and progressively evolve towards more ordered configurations. The protocol was found to be successful based on its consistent ability to produce large Type I domains with minimal defects across multiple samples. To assess the effectiveness and reproducibility of this approach, we applied this laser excitation protocol to four different but nominally identical artificial square ices (Samples I to IV). The resulting vertex configuration map for one of these (Sample I) is shown in Fig. 3(a), where large Type I domains can be seen, while Fig. 3(b) displays the populations of Type I, II, and III vertices in each of the four samples following this decreasing fluence protocol. Reflecting the large ground state domains seen in Fig. 3(a), we achieve an average Type I vertex population of $92.4 \pm 0.6\%$.

To understand how large Type I domains form in artificial square ice with this decreasing-fluence laser excitation protocol, we first need to acknowledge that there is a distribution of the energy barriers for nanomagnet switching [19]. This can arise from defects and other variations in the microstructure of the permalloy introduced during fabrication. At the highest laser fluences, all macrospins can reorient. However, the probability that the magnetic moment of a given nanomagnet will switch is strongly dependent on its energy barrier. This means that, as the laser fluence, and thus the energy delivered per pulse, decreases, the nanomagnets with the highest energy barriers will become effectively blocked with their switching probability drastically reduced. These nanomagnets can then act as nucleation sites for the formation of Type I domains. Here, the stray field generated by such nanomagnets with “frozen” macrospins will bias the orientation of the macrospins in neighboring nanomagnets to form energetically favorable vertex configurations, so that small Type I clusters form that will grow in size. Crucially, once a Type I domain forms, its magnetostatic environment stabilizes it. This means that, as the Type I domain grows, the probability that nanomagnets will switch back into a higher-energy state becomes infinitesimally small, as the energy barrier for such transitions increases. In contrast, nanomagnets at the domain boundaries can still undergo reversal, promoting further expansion of the Type I domain into adjacent higher-energy regions. This self-reinforcing process continues until expanding Type I domains collide, forming boundaries composed of Type II vertex configurations between them. If there is sufficient time, these Type II boundaries can then shrink and annihilate through the motion of Type III vertices [36]. The formation of large, well-ordered Type I domains shown in Fig. 3(a), therefore suggests a collective relaxation mechanism governed not only by stochastic nucleation but also by the local energetics that drive the growth and coalescence of Type I domains. A similar seed-driven relaxation mechanism, in which early remagnetizing nanomagnets bias the switching of their neighbors through dipolar fields, has been proposed in thermally quenched ASIs [41]. In our case, the gradually decreasing laser fluence plays an analogous role by allowing the elements that remagnetize first to nucleate extended Type I domains that stabilize as the system progressively freezes.

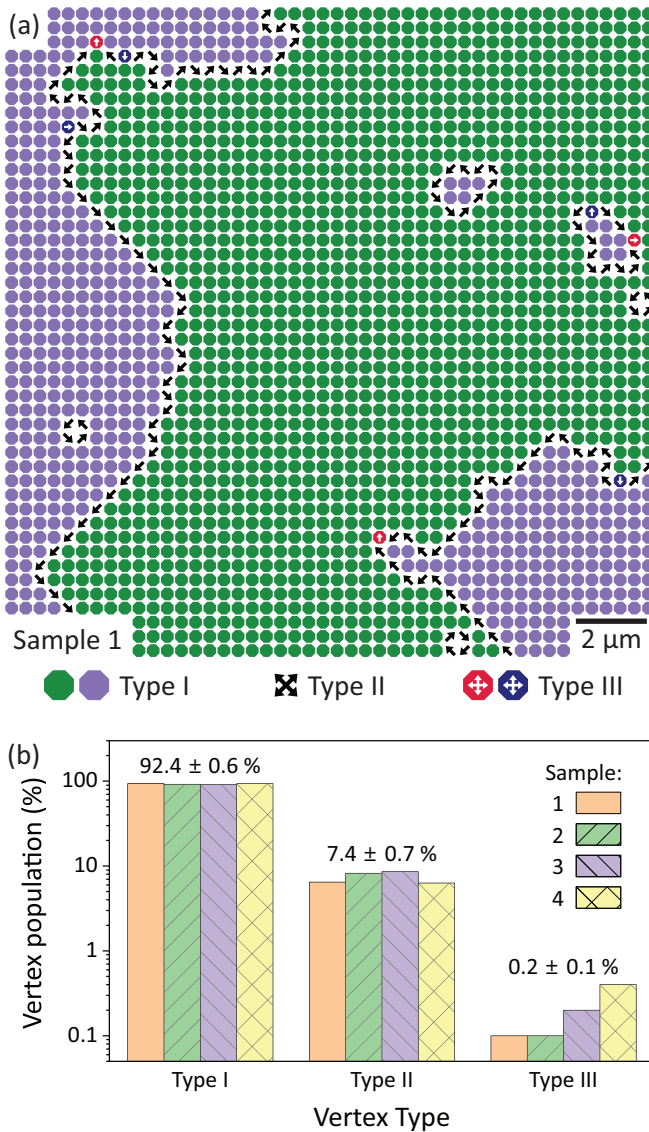


FIG. 3. Vertex configurations and populations following laser excitation with a series of laser pulses of gradually decreasing fluence. Here four different but nominally identical artificial square ices were exposed to a laser of decreasing fluence over time (total exposure duration of 1 s) going from 9.7 mJ/cm² down to 6.1 mJ/cm² in steps of 13.2 nJ/cm². (a) Map of the vertex configurations of an artificial square ice (Sample 1). (b) Type I, Type II, and Type III vertex populations extracted from the MFM images for nominally identical Samples 1 to 4. The average vertex population along with its standard deviation for each vertex type is given above the graphs and the corresponding simplified vertex configuration maps are given in Fig. S1 [38].

IV. MICROMAGNETIC SIMULATIONS OF MAGNETIZATION DYNAMICS IN ARTIFICIAL SQUARE ICE

We now clarify how long-range magnetic order can emerge following ultrafast laser excitation within the brief time window in which the magnetization within individual nanomagnets is still recovering. Our observations suggest that dipolar interactions play a key role during the magnetization

recovery that occurs over 10's of picoseconds following the ultrafast (sub-picosecond) quenching of the magnetization. Yet, it remains unclear how these interactions become effective so rapidly. We therefore performed numerical simulations designed to reproduce the physical conditions under which nanomagnet relaxation occurs and to shed light on the mechanisms governing their fast collective response. In a first step, we calculated the energy barrier for a transition from a Type II to a Type I vertex configuration as a function of temperature [Fig. 4(a)], using the SISM [42–45]. To account for temperature effects, the saturation magnetization and exchange stiffness were rescaled according to the temperature [42].

A detailed view of the entire transition pathway, comprising the evolution of the energy profile along the discretized trajectory at different temperatures, is provided in Fig. S2 [38], while details of the modeling of the magnetization reversal and associated magnetic parameters are given in Sec. II of the Supplemental Material [38]. For this, we modeled the simultaneous reversal of two nanomagnets to give a direct transition from the initial Type II to the final Type I vertex configuration (Fig. S3), which is characterized by a single energy barrier with no intermediate local minima. This corresponds to a minimum energy path in which both nanomagnets switch together without forming metastable states such as Type III vertex configurations. This contrasts with the mechanism that can occur during thermal annealing where the transition from a Type II to a Type I vertex configuration would proceed through sequential switching of two nanomagnets per vertex, so that it passes through an intermediate Type III vertex configuration and involves two distinct energy barriers [3]. Our choice of reversal mechanism is guided by the experimental observation that magnetization reversal can be triggered by a single ultrashort laser pulse. While the exact sequence of switching events remains unclear, assuming simultaneous reversal offers a reasonable approximation within the ultrafast time scale under consideration.

Our calculations reveal that, although the energy barrier separating vertices in Type II and Type I configurations decreases with increasing temperature [Fig. 4(a)], it is still at ~ 20 eV per nanomagnet at 750 K, which is still several orders of magnitude above $k_B T$ (~ 66 meV at this temperature). Achieving a significant probability of thermally activated switching would therefore require the system to remain at such a high temperature for much longer than is typically achieved through ultrafast excitation with femtosecond lasers. This rules out thermally activated switching of single-domain nanomagnets (i.e., the superparamagnetic regime) as the dominant mechanism during a single-pulse laser excitation. Instead, our results support the interpretation that magnetization reversal occurs due to ultrafast laser-induced demagnetization, followed by rapid magnetization recovery and dipolar-driven reorientation, a process that takes place only when the system is transiently driven extremely close to the Curie temperature (770 K). It is plausible that this condition is met during the short time window following laser excitation – at the start of the fast remagnetization [Fig. 1(d)] – when the electron temperature remains very close to the Curie temperature. As we show later in Fig. 4(b), our simulations indicated that this occurs on the sub-picosecond time scale. We thus believe that, while excitation with the ultrafast

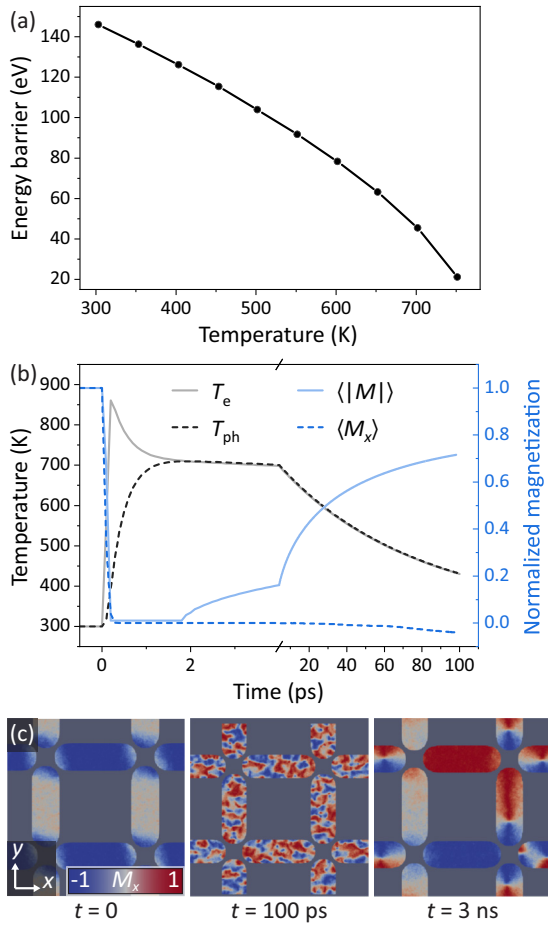


FIG. 4. Micromagnetic simulations of energy barrier and ultrafast magnetization dynamics. (a) Calculated energy barrier to switching from a Type II to a Type I vertex configuration as a function of temperature, using the SISM. The minimum energy path was computed for a simultaneous reversal of two nanomagnets. The full trajectory of the energy landscape as a function of step number and temperature is shown in Fig. S2 [38]. (b) Temporal evolution of the electron temperature T_e , phonon temperature T_{ph} , average magnetization modulus $\langle |M| \rangle$, and average component of the magnetization along the x direction $\langle M_x \rangle$ obtained from LLB simulations following excitation of the artificial square ice shown in (c). The artificial square ice is initially in a Type II vertex configuration, and is excited with a 60 fs laser pulse at high fluence (7 mJ/cm^2). $\langle |M| \rangle$ is normalized to its equilibrium value at 300 K and $\langle M_x \rangle$ is normalized to its initial value at $t = 0$. The quantity $\langle |M| \rangle$ reflects the average local magnetization amplitude and tracks the evolution of the spin temperature, while $\langle M_x \rangle$ corresponds to the net magnetization along the x direction and is sensitive to the global magnetic order. It can be seen that $\langle M_x \rangle$ does not recover after the pulse, indicating a transition from Type II to Type I vertex configurations. The corresponding results for the low-fluence case, in which $\langle M_x \rangle$ recovers and the vertices remain in the Type II configuration, are shown in Fig. S4 [38]. (c) Snapshots from LLB simulations of the magnetization configuration at different time delays following ultrafast demagnetization, illustrating the emergence of multidomain vortex-like structures within the nanomagnets. The simulated region is $0.8 \mu\text{m} \times 0.8 \mu\text{m}$ corresponding to two unit cells of the artificial square ice. The full sequence of magnetization snapshots from the LLB simulations, including additional intermediate time steps, is presented in Fig. S5 [38].

laser initially results in magnetic disorder, subsequent partial recovery of the magnetization creates favorable conditions for dipolar interactions to drive the system toward a lower-energy configuration.

To complement the SISM analysis, we employed LLB micromagnetic simulations [46–48] coupled to the two-temperature model [49,50], which are specifically designed to capture ultrafast demagnetization dynamics and incorporate longitudinal relaxation, i.e., the temperature-dependent changes in the modulus of the magnetization vector $|M|$, in contrast to standard micromagnetic models where the $|M|$ remains fixed. Because LLB simulations allow $|M|$ to vary with temperature and time, they are appropriate for modelling the magnetization dynamics following ultrafast laser excitation of artificial square ice.

The simulations were performed with a time step of 1 femtosecond, using magnetic parameters consistent with those adopted in the SISM calculations (Supplemental Material, Sec. II [38]), and assuming a pulse duration of 60 fs. The heating effect of the laser excitation is introduced via the two-temperature model, using parameters listed in Sec. III of the Supplemental Material [38]. These parameters define the temporal evolution of the electron and phonon temperatures following the laser excitation and are selected to qualitatively reproduce the conditions under which magnetization reversal is expected to occur. However, since the actual laser absorption in the experimental samples is not precisely known, the simulated laser fluence is not intended to correspond quantitatively to the experimental values, and should be regarded as a fitting parameter that ensures demagnetization in the simulations. As can be seen in Fig. 4(b), under these conditions, the two-temperature model shows a rapid rise in electron temperature T_e – on the order of a hundred femtoseconds – when the laser pulse transfers energy to the magnetic material, followed by thermalization between the phonon and electron baths. The magnetization dynamics are coupled to the electron temperature via the spin-orbit interaction. However, because spin relaxation has a finite time scale (~ 100 fs), the spin system does not immediately follow the electron temperature. As a result, the average magnetization modulus $\langle |M| \rangle$, which reflects the spin temperature, lags behind the electron temperature on a sub-picosecond time scale.

To explore ultrafast ASI dynamics further, we used the same LLB code to simulate the temporal and spatial evolution of the magnetization for two unit cells of artificial square ice with periodic boundary conditions [Fig. 4(c)]. Further details about the simulation procedure and parameters are provided in Sec. III of the Supplemental Material [38]. Starting from a state in which all vertices are in a Type II configuration at $t = 0$, following ultrafast laser excitation, each nanomagnet undergoes full demagnetization, which is achievable only when the electron temperature significantly exceeds the Curie temperature. As a result, each nanomagnet is in a disordered state, with fluctuating, uncorrelated spins on the atomic level, and therefore no net magnetization. During the subsequent recovery, magnetic domains nucleate within the nanomagnets themselves, and subsequently grow and coalesce, reaching about 30 nm in size at $t = 100$ ps. On this time scale, the intrinsic magnetization is almost fully recovered, and magnetostatic interactions become active. At a later time,

$t = 3$ ns, the magnetic configuration within the nanomagnets is either single-domain or hosts a vortex-like structure, which is a metastable configuration. As the system evolves, each vertex may relax into a low-energy Type I configuration via dipolar interactions. However, due to the high computational cost of LLB simulations, we are limited to small systems and cannot run enough statistically independent realizations to assess whether the system consistently relaxes into a lower-energy state, as experimentally observed.

To better understand the role of magnetic order during the relaxation process, we monitor both the average magnetization modulus, $\langle |M| \rangle$, and the average component of the magnetization along the x direction, $\langle M_x \rangle$ [see coordinate axes in Fig. 4(c)]. While $\langle |M| \rangle$ tracks the evolution of the spin temperature and the degree of local demagnetization, $\langle M_x \rangle$ is sensitive to the net orientation of the macrospins along the x axis and thus reflects the global magnetic configuration. This distinction is crucial in artificial square ice, where the Type II state exhibits a net magnetization along x (ferromagnetic-like), whereas the Type I ground state has zero net magnetization (antiferromagnetic-like). Therefore, tracking $\langle M_x \rangle$ allows us to distinguish whether the system returns to the original Type II state or evolves towards a Type I configuration. One concern is that, relying exclusively on $\langle M_x \rangle$ may lead to ambiguities, as certain Type III and Type IV vertex configurations also exhibit vanishing $\langle M_x \rangle$ and might be mistakenly identified as Type I. In practice, this issue does not arise in the LLB simulations, as these high-energy configurations are never observed during the recovery process. This is likely to be because the simulations are restricted to only two unit cells of artificial square ice with periodic boundary conditions, which significantly limits the number of accessible vertex configurations.

In Fig. 4(b), we show the temporal evolution at high fluence, where the system does not recover its initial $\langle M_x \rangle$ value, indicating a transition from Type II to Type I vertex configurations. In contrast, for low fluence, as shown in Fig. S4 [38], $\langle M_x \rangle$ recovers after the laser pulse, consistent with a reversible excitation of the Type II configuration and high energy barriers. These two scenarios highlight that it is only when the energy delivered to the electronic system is sufficient to raise its temperature above the Curie temperature and fully demagnetize the nanomagnets that the system can access new microstates and, potentially, undergo a reconfiguration of its magnetic state. Note that, in the LLB simulations, although the intrinsic magnetization recovers on an ultrafast time scale, some elements form vortex-like states, which require a longer time – on the order of nanoseconds – to become single domain.

As mentioned above, from the LLB simulations, we observe that a single laser pulse can induce a transition from Type II to Type I vertex configurations. However, preliminary results indicate that the final outcome may not be fully deterministic, since simulations under identical conditions can yield different magnetic states due to thermal fluctuations and the influence of local dipolar fields. While LLB simulations are limited to single-pulse events and small system sizes, theoretical considerations based on kinetic Monte Carlo [51,52] and master equation [53] approaches suggest that even large energy barriers can be overcome via the cumulative effect

of many laser pulses. This supports the interpretation that multiple excitations are needed to reliably drive the system towards its ground state, with dipolar interactions playing a key role in guiding the relaxation.

Our results demonstrate that laser excitation enables the system to overcome significant energy barriers, and that long-range magnetic order can emerge within sub-nanosecond time scales, with the decisive macrospin reorientation occurring during the early stages of magnetization recovery. These observations suggest that the relaxation process may proceed via two distinct pathways. In the first scenario, the nanomagnets transiently demagnetize and recover their magnetization, temporarily remaining in a multidomain or vortex-like state during the recovery. As the magnetization begins to recover, each nanomagnet may develop a net magnetic moment oriented along its long axis, even if the internal magnetization texture is not yet fully uniform. This partial recovery means that the average magnetization of each nanomagnet can still be thermally reconfigurable. In this case, dipolar fields from neighboring elements can influence the orientation of these average magnetic moments, allowing the system to collectively evolve towards an ordered configuration before the nanomagnets become magnetically frozen. This mechanism suggests that full monodomain formation is not strictly required to achieve the ground state, as long as thermal fluctuations and dipolar interactions act in concert during the recovery. However, in this scenario, as observed in the micromagnetic simulations, multidomain or vortex-like configurations will tend to inhibit dipolar-driven reorganization and increase the likelihood of the system being trapped in higher-energy configurations.

Another, perhaps more plausible, relaxation pathway is one in which the nanomagnets recover into quasi-macrospin states with a sufficiently uniform magnetization but also sufficiently reduced energy barriers – due to the high temperature giving a reduction in the saturation magnetization – allowing thermally activated switching within the transient time window. In this regime, the system may follow an Arrhenius-like behavior, where the effective switching time is governed by the exponential dependence on the energy barrier, provided that the attempt rate $\tau_0 \approx 10^{-12}$ s [20,54,55] and the temperature remains sufficiently close to the Curie temperature. Since the system remains near the Curie temperature for a very short time, on the order of 1-2 picoseconds, the probability of such a reversal is very low. However, if a large enough fraction of nanomagnets undergoes thermally induced macrospin reversal before the system cools down, the dipolar interactions can guide the ensemble into a low-energy Type I configuration at nearly every vertex. This process should be more efficient with multiple laser pulses. We note here that the sequential freezing process, described earlier in the context of the experimental results (Sec. III) and arising from the distribution of energy barriers, is likely to play a role in both of these relaxation pathways, particularly in the multipulse regime where the system undergoes progressive reordering. This interpretation is consistent with seed-driven relaxation pathways previously discussed in the context of thermally driven quenching of the magnetization in artificial spin ices [41], and provides a microscopic rationale for the emergence of extended low-energy domains during the decreasing-fluence protocol.

Crucially, both mechanisms rely on a delicate interplay between the degree of demagnetization, the rate of remagnetization, and the strength of dipolar interactions. Despite the simplifications of the LLB model and the limited size of our simulations, the robustness and reproducibility of the experimental results suggest that this ultrafast access to the ground state is not coincidental but reflects an intrinsic and effective pathway for energy minimization.

V. CONCLUSIONS

In summary, we have shown that femtosecond laser excitation can drive magnetic relaxation in artificial square ices during the fast magnetization recovery, so on the picosecond to a few 10's picosecond time scale following ultrafast demagnetization, enabling the system to access low-energy configurations and reach the ground state. While our main experiments employ pulse trains to ensure stable excitation conditions, control measurements confirm that a single ultrashort laser pulse is sufficient to trigger magnetization reversal and initiate the relaxation process toward a more ordered magnetic configuration, provided the laser fluence is high enough. The magnetic relaxation relies on the delicate interplay between ultrafast demagnetization and the partial recovery of the magnetization, allowing dipolar interactions to drive collective ordering. By applying a decreasing-fluence laser excitation protocol, we enhanced the effectiveness of the relaxation, achieving a population of the lowest-energy Type I vertices exceeding 92%. We have thus demonstrated that the transient high-temperature regime provides favorable conditions for dipolar-driven reordering, offering a rapid and spatially selective route for controlling magnetic configurations in ASIs, thus offering a promising platform for ultrafast spin-based computing [5].

Although LLB micromagnetic simulations reproduce the transient magnetization dynamics well, they suggest that relaxation to the lowest-energy state after excitation with a

single laser pulse may not be fully deterministic, resulting from the interplay of thermal fluctuations and other contributions to the magnetic energy. Unfortunately, it is not viable to model the cumulative effects of multiple laser pulse excitations, as the simulations cannot realistically account for the cumulative effects of multiple laser pulses, since each excitation is simulated independently from a prescribed initial magnetic configuration. In addition, the high computational cost limits the feasibility of simulations over long time scales, especially for large ensembles. This highlights current limitations in modeling the subtle interplay between demagnetization, thermal fluctuations, and dipolar coupling on longer time scales. Bridging this gap will require multiscale approaches including atomistic spin dynamics [47,56] and kinetic Monte Carlo simulations [13] to fully capture the mechanisms behind ultrafast relaxation and ordering in arrays of coupled nanomagnets, as exemplified by recent multiscale modeling of magnon dynamics and spin-current-driven reordering in ferrimagnets [57].

Finally, while our work has focused on artificial square ice, the ultrafast relaxation mechanisms revealed here may also play a role in other ASI geometries, including both two-dimensional systems [17] and recently developed three-dimensional systems [58], offering exciting prospects for future investigations.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [59].

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