

Dynamic freezing in a quasicrystal nanomagnet array

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Magnetic long-range order in quasiperiodic lattices has rarely been observed, either in natural materials or in artificially engineered systems. In this study, we investigate a nanomagnet array based on P1 Penrose tiling and demonstrate that, within the framework of the vertex model, the system is unfrustrated, making the emergence of long-range order on quasiperiodic lattice a promising possibility. Large domains of ordered states appear following a thermal annealing protocol. However, numerous excitations also persist. Through statistical analysis, we show that the system falls out of equilibrium during the cooling process, which we attribute to a trapping effect. We further show that this trapping arises from the complex energy landscape, which obstructs the kinetic pathways necessary for the system to relax these excitations. Our findings provide important insights into the mechanisms of dynamic freezing in artificial spin ice systems.

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I. INTRODUCTION

Two-dimensional arrays consisting of interacting nanomagnets, known as artificial spin ice, provide a powerful platform to investigate the frustrated magnetism [1–5]. The flexibility and tunability and the ability to probe spin configuration afforded by nanofabrication techniques offer unique opportunities to test predictions from spin models, leading to a profound understanding of the physics occurring in nature materials [6–23]. Based on these advantages, spin models with various geometries were tested, including periodic, aperiodic, and even disordered lattices [24–33]. The geometry-frustration-induced collective behaviors, especially the cooperative magnetic phenomena analogous to those in classic spin models, have become of keen interest [7,10,34,35].

An example of this is dynamical freezing, a phenomenon observed in spin ice compounds. It arises because the system follows the so-called ice rule constraint, which cannot guarantee long-range order at low temperatures [36,37]. As a result, a large number of degenerate low-energy states that satisfy the ice rule emerge. The large energy barriers separating these degenerate spin configurations prevent the system from reaching its true ground state. As the temperature decreases, the system

becomes trapped in one of these low-energy configurations: the spin-flip dynamics freeze [38]. A similar phenomenon also exists in a lithography patterned array [39]. However, such behavior may be obscured by the blocking temperature problem. Due to the shape anisotropy of the nanomagnets, the macrospins can become “frozen” at relative high temperatures before the dynamical freezing occurs [40]. Nevertheless, such phenomenon can be evidenced by Monte Carlo simulation with a single-spin-flip algorithm where the blocking temperature problem can be avoided. The long-range ordered (LRO) state approach remains challenging in such simulations due to dynamical freezing, unless the loop flip algorithm is introduced [41].

The dynamic freezing effect can also occur in unfrustrated systems, such as the square ice lattice. In this case, the blocking temperature problem is avoided due to the relatively high transition temperature. Here, spin freezing arises from the formation of domain walls that separate two degenerate ground states. Removing these domain walls through single spin flips is unlikely, as residual domain structures often remain after thermal annealing [4,11]. In quasiperiodic lattices, the interplay between aperiodic geometry and frustration further complicates relaxation dynamics, giving rise to spin-glass-like freezing [42–44]. These observations highlight the critical role of lattice geometry in determining dynamical freezing, both in natural and artificially designed systems.

In this article, we investigate an artificial spin ice based on P1 Penrose tiling and demonstrate that, within the vertex model framework, the system is unfrustrated, making long-range order formation in a quasiperiodic lattice feasible. Following thermal annealing, large domains of the ground

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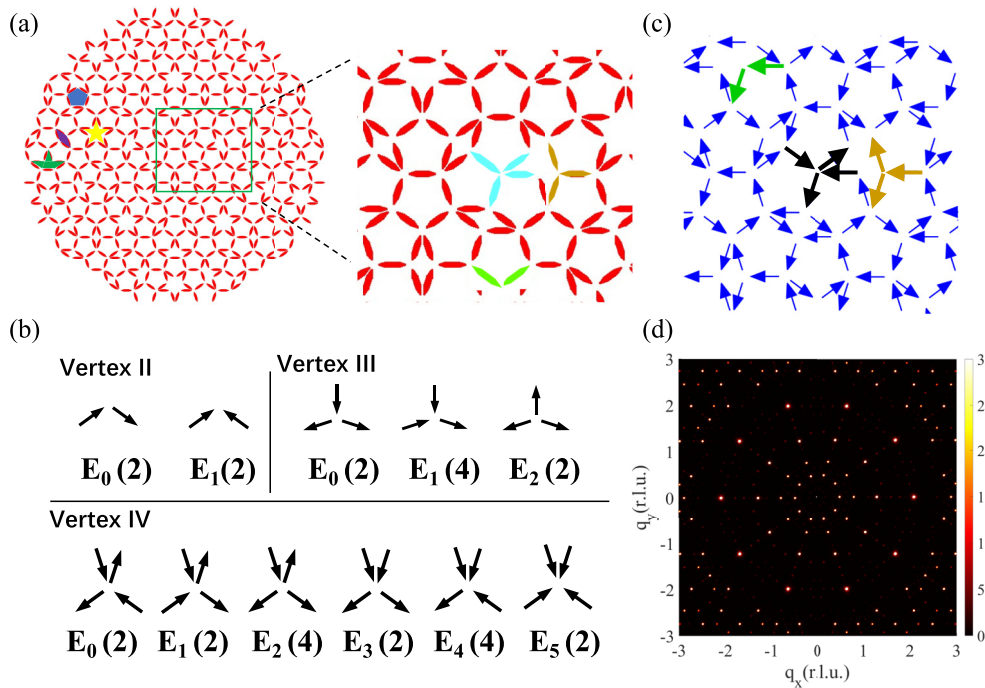


FIG. 1. (a) The structure of the P1 Penrose tiling. The four basic units—pentagon, boat, star, and diamond—are highlighted in different colors. The highlighted portion is magnified on the right, with the three basic types of vertices colored green, brown, and cyan, respectively. (b) The energy landscape of the three vertex types, labeled as vertices II, III, and IV, respectively, with their energy degeneracies indicated in brackets. (c) The ground state of the P1 Penrose tiling, constructed by the lowest energy state of the three vertex types. (d) The calculated magnetic structure factor based on the ground state of the P1 Penrose tiling.

state emerge, as confirmed by sharp Bragg peaks in the magnetic structure factor. However, excessive excitations persist even in Monte Carlo simulations. The statistical analysis reveals that, for certain types of vertices, out-of-equilibrium freezing occurs, leading to the retention of these excitations during cooling. By examining all possible single-spin-flip processes in the low-energy regime, we attribute this behavior to trapping effects arising from high kinetic barriers. We further show that the inclusion of paired-spin-flip dynamics in simulations overcomes these constraints, enabling the system to fully access its ordered ground state.

II. P1 PENROSE TILING MODEL

The P1 Penrose tiling comprises four prototiles: pentagon, star, boat, and diamond [Fig. 1(a)]. For analytical purposes, the pattern is reformulated in terms of three vertex types (labeled as vertices II, III, and IV according to the number of islands in each vertex) as highlighted in the magnified section of the image. To study the energy landscape of the system, we first calculated the magnetostatic energy of the vertices at the nearest-neighbor level. The energy calculation was performed using the oommf micromagnetic simulation package [45]. In Fig. 1(b), we enumerate all possible moment configurations for each vertex type and label them with the relative energy levels. A moment configuration of the ground state is illustrated in Fig. 1(c), where all vertices in the E_0 state are placed together compatibly. The ground state of the P1 Penrose tiling thus possesses twofold degeneracy, i.e., the spins are unfrustrated in the vertex model framework.

The magnetic structure factor, calculated based on the ground-state spin configuration, shows clear Bragg peaks associated with tenfold rotational symmetry [see Fig. 1(d)], confirming the quasi-periodic nature of the magnetic order.

III. EXPERIMENTAL AND SIMULATION OF THERMAL ANNEALING RESULTS

We fabricated the artificial quasicrystals using the P1 Penrose tiling. The patterns were realized with electron beam lithography (EBL) followed by electron beam evaporation of permalloy ($\text{Ni}_{80}\text{Fe}_{20}$) on a silicon nitride substrate (see Methods section for details). A scanning electron microscopy (SEM) image of the fabricated sample is illustrated in Fig. 2(a), where the islands are designed in a shuttle shape to save space. We fabricated the samples with lattice spacings ranging from 550 to 700 nm, which allow us to alter the interaction energies between nanomagnets. The island size is fixed at 450 nm in length, 80 nm in width, and 25 nm in thickness. All islands are placed along the edges of the pattern. Magnetic force microscopy (MFM) images confirm that all islands are single domain with moments pointing along the long axis of the islands, as shown in Fig. 2(b). A thermal annealing protocol was performed to bring the system into the low energy state (details in the Methods section). The efficiency of the annealing protocol is confirmed by a square pattern subjected to the same annealing conditions, where fully ordered ground states were observed.

To better visualize the ordering in the annealed states, we mapped the macrospins observed in the MFM images using

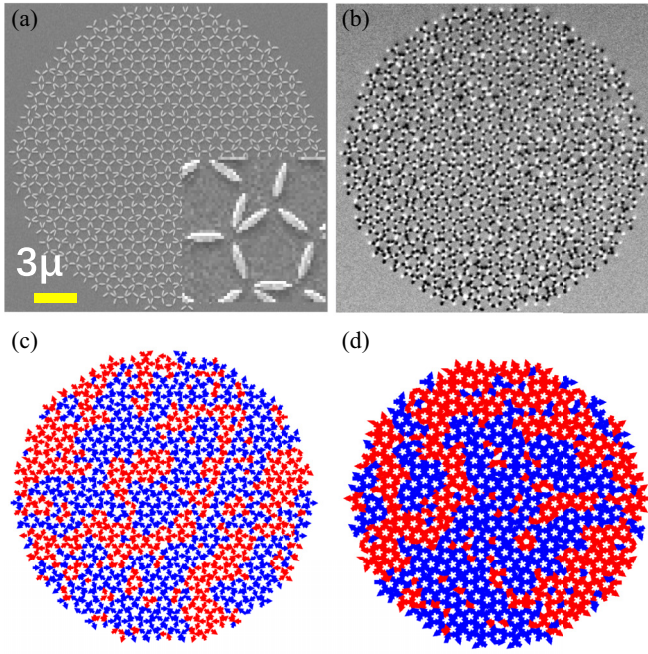


FIG. 2. (a) SEM image of an artificially designed P1 Penrose tiling array with a lattice constant of 550 nm. (b) MFM image of the same array in (a) after thermal annealing. (c) Color map of the MFM image shown in (b), where vertices are represented by red and blue lines corresponding to two degenerate states. (d) Color map of Monte Carlo simulation results, using the same mapping method as in (b).

different colors. As shown in Fig. 2(c), a color map of the MFM image of the P1 Penrose tiling array with a lattice constant of 550 nm is presented. The red and blue lines represent the macrospins belonging to the two degenerate spin configurations. Large crystallites of the ground state were obtained, characterized by sizable ground-state domains. Such crystallization surpasses what has been achieved in natural quasicrystals, where magnetic long-range order is rarely observed due to the complex form of frustration [43,44]. This highlights the advantages of the artificial systems. However, a large amount of domain walls remains in the annealed states. One might attribute this to the relatively weak dipolar interactions between the islands, as the lattice spacing in the P1 Penrose tiling can only be reduced to 550 nm due to fabrication constraints. To rule out this extrinsic constraint, we compared the experimental results with Monte Carlo simulations where all interactions between islands were considered. A single-spin-flip algorithm along with a simulated annealing procedure was employed (see details in the Methods section). Snapshots of spin configurations at the lowest temperature were analyzed using the same color-mapping method as in the experiment. As shown in Fig. 2(d), numbers of domain walls persisted, which indicates that the fully ground state cannot be accessed via coupling strength enhancement.

IV. EVIDENCE OF OUT-OF-EQUILIBRIUM

To further understand the challenges in reaching the ground state in the P1 Penrose tiling, we examined the vertex energy

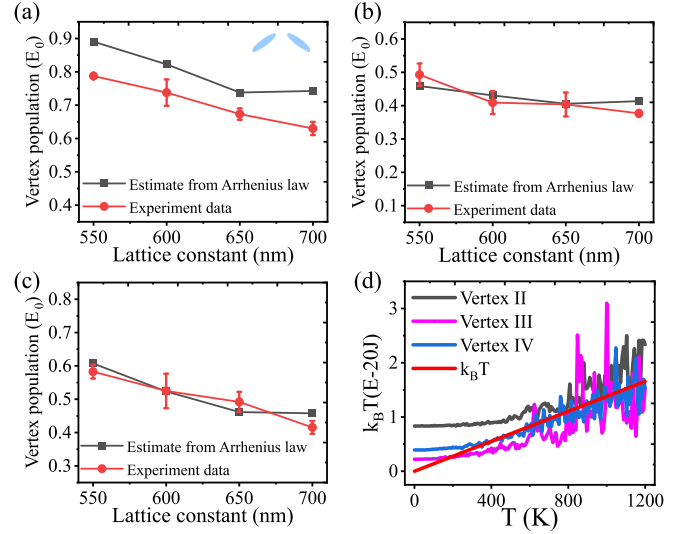


FIG. 3. (a)–(c) Averaged vertex energy populations for the three types of vertices (vertices II, III, and IV for a, b, and c, respectively) obtained from experimental data and compared with the calculated Boltzmann distribution at the blocking temperature. Data are averaged from three data sets. (d) Effective thermodynamic temperature $k_B T_{\text{eff}}$ deduced from the populations of different vertex types, compared with $k_B T$ at each temperature point of the Monte Carlo simulation.

distribution and compared it with the predicted distribution at equilibrium. Since the spin configurations arrested by the MFM images are those frozen at the blocking temperature T_B , the vertex population at equilibrium can be estimated based on the vertex energy at T_B . To achieve this, we estimate the blocking temperature T_B for the annealing process. This can be estimated using the expression [28,46]

$$k_B T_B \ln \frac{t_m}{t_0} = \frac{1}{2} \mu_0 M_S^2(T_B) DV. \quad (1)$$

Here, t_m is the measurement time, t_0 is the attempt time assumed to be 10^{-10} s, μ_0 is the permeability of free space, M_S is the saturation magnetization, D is the difference between the demagnetizing factors along the in-plane short and long axes of the nanomagnets [47], and V is the volume of a single nanoisland. This equation can be solved graphically combined with the temperature dependence of M_S , which can be measured from an unpatterned Py film with the same thickness as the artificial array [48]. The blocking temperature T_B was determined to be 696 K, and the saturate magnetization M_S at T_B is 88 kA/m. The vertex energy and population at T_B can be estimated using oommf simulation based on M_S at T_B . For vertices II–IV, the vertex population at E_0 , following the Boltzmann distribution at T_B , was calculated and compared with experiment results as a function of the lattice spacing, as illustrated in Figs. 3(a)–3(c). In contrast to vertices III and IV, the population of vertex II at T_B significantly deviates from the predicted state at equilibrium, which can be treated as evidence of out-of-equilibrium.

This out-of-equilibrium behavior is further confirmed by Monte Carlo simulation results, where the population of vertex II is found gradually deviated from the equilibrated

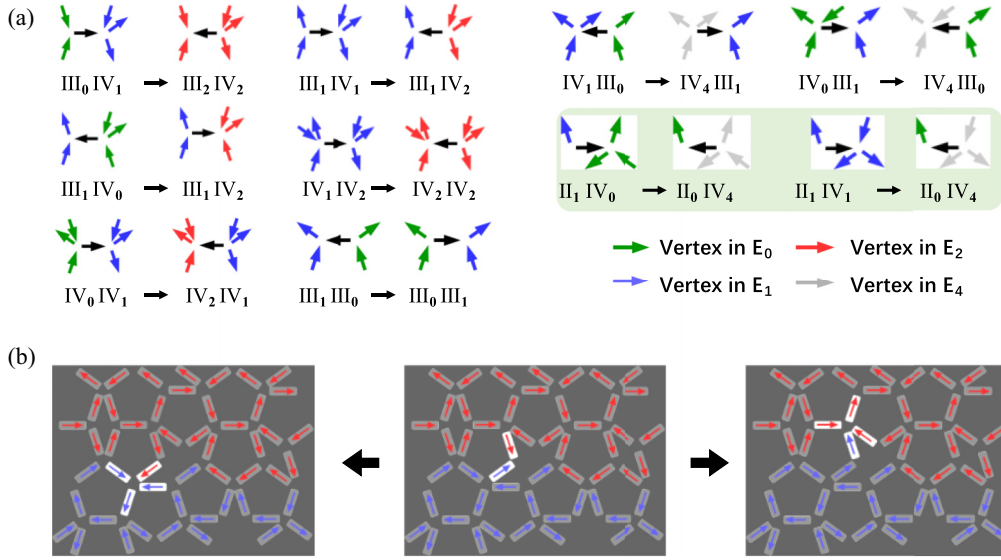


FIG. 4. (a) Kinetic processes of single-spin-flip induced spin configurations variations. The vertex-vertex spin configurations before and after a single spin flip (black arrow) are denoted by different colors. Vertex types are labeled using Roman numerals with subscripts indicating their energy levels. Kinetic processes involving vertex II are highlighted. (b) Schematic illustration of kinetic trapping involving vertex II. Any attempted single spin flip drives adjacent vertices into high energy states. High-energy spin configurations before and after the spin flip are highlighted.

states at low temperature. Here, a special defined quantity is measured to illustrate this behavior. For a system at equilibrium, the vertex population ratio can be estimated by $\frac{N(E_n)}{N(E_0)} = \frac{p_n}{p_0} \exp\left(\frac{E_0 - E_n}{k_B T}\right)$, where $N(E_i)$ represents the vertex population at E_i and p_i represents the degeneracy factor of E_i . We can rewrite this equation as $k_B T = (E_0 - E_n) / \ln\left(\frac{N(E_n)}{N(E_0)} \frac{p_0}{p_n}\right)$. The quantity $(E_0 - E_n) / \ln\left(\frac{N(E_n)}{N(E_0)} \frac{p_0}{p_n}\right)$ can be deemed as the “effective thermodynamic temperature,” which we denote as $k_B T_{\text{eff}}$. In the Monte Carlo simulation, since the coupling strength is set as a constant, the vertex energy remains invariant throughout the annealing process. Thus, $k_B T_{\text{eff}}$ can be deduced based on the measured $N(E_n)$ and $N(E_0)$. For each type of vertex, the $k_B T_{\text{eff}}$ should be the same and proportional to the real temperature T if the system is in an equilibrated state. For each temperature point in the simulation, we calculated the $k_B T_{\text{eff}}$ using $(E_0 - E_1) / \ln\left(\frac{N(E_1)}{N(E_0)} \frac{p_0}{p_1}\right)$ for each type of vertex and compared it with $k_B T$. The results are illustrated in Fig. 3(d). At high temperatures, the signal oscillates dramatically due to strong fluctuations but becomes stable as the system approaches lower temperatures. In contrast to vertices III and IV, the trend of the signal deduced from vertex II clearly deviates from the linear $k_B T$ as the system approaches lower temperatures, indicating that out-of-equilibrium happened. Additionally, the $k_B T_{\text{eff}}$ deduced from vertex II become nearly flat at low temperatures, implying that the ratio $N(E_1)/N(E_0)$ becomes invariant, i.e., vertex II is trapped at E_1 at low temperatures. In contrast, for the square pattern, a similar analysis shows that the $k_B T_{\text{eff}}$ deduced from different vertex types closely track the expected value $k_B T$ (see Fig. 2 in the Supplemental Material [48]). These findings are consistent with the analytical results in experiments, which suggests that the out-of-equilibrium behavior for vertex II is induced by intrinsic mechanisms. As we will show below, this phenomenon is a consequence of the dynamic freezing effect.

V. SINGLE-SPIN-FLIP INDUCED FLUCTUATIONS

Since both experimental and simulation results reveal that vertex II deviated from equilibrium during the thermal annealing process, what makes it distinct from the other two vertex types? To address this, we examined the energy cost of all possible kinetic processes induced by single-spin-flip fluctuation in a low-energy background. We assume that only single-spin-flip dynamics are active at low temperatures [4,39,49]. Additionally, we assume the system resides in a background consisting exclusively of vertices with energies E_0 and E_1 . This assumption is consistent with the observation that only a small number of vertices occupy energy levels higher than E_1 in both experimental and simulation results (see Fig. 3 in the Supplemental Material [48]). All possible fluctuations induced by single spin flips in this background are illustrated in Fig. 4(a). For vertices III and IV, most relevant kinetic processes result in energy states E_0 , E_1 , and E_2 . In contrast, for vertex II [highlighted in the bottom panel of Fig. 4(a)], any attempt to transition from the E_1 to the E_0 state causes a neighboring vertex IV to transfer to its E_4 state. The energy cost of such processes is comparable to that of nucleating a type-IV vertex in a square pattern, as calculated based on oommf simulations (see Fig. 4 in the Supplemental Material [48]). Therefore, such fluctuations are prohibited. More importantly, as illustrated in Fig. 4(b), all vertex II types in the P1 Penrose tiling are surrounded by vertex IV, which means all kinetic “pathways” are prohibited. As a result, vertex type II becomes trapped at the E_1 energy level. Furthermore, because vertex type II is sparsely distributed throughout the structure, once trapped at E_1 , these vertex II excitations act as pinned “defects,” obstructing the flow of the domain wall. This mechanism sharply contrasts with square ice, where excitations can be expelled along with the propagation of the domain wall [50,51]. The ground-state domains in square ice thus can grow

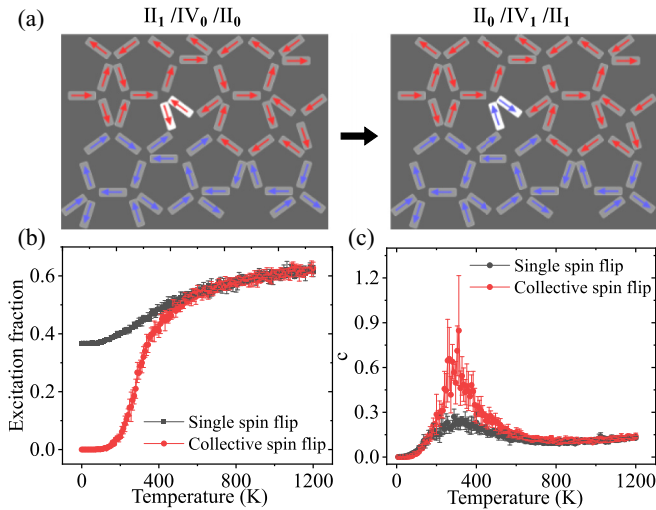


FIG. 5. (a) Demonstration of collective spin flips. Flipped spins are highlighted in bright color. Vertex states before and after the collective spin flips are indicated above the images. (b) and (c) The population of excitations and heat capacity c as a function of temperature T based on single-spin-flip and collective-spin-flip algorithms, respectively. The Monte Carlo step and sample size used for heat capacity calculation were the same as those used in the equilibrating procedure. The data are obtained from averaged five independent simulation results.

by shrinking neighboring domains at a small energy cost. However, such fluctuations are prohibited in the P1 Penrose tiling due to the presence of pinned defects, preventing further domain growth.

To further validate the effect of the kinetic constraint, we performed Monte Carlo simulations incorporating a collective-spin-flip algorithm to bypass the dynamic freezing effect. Unlike the loop-flip algorithm frequently used in kagome ice [41], we found that a collective spin flip can circumvent the formation of high-energy vertex IV states during cooling. Consequently, the population of excitations

is significantly suppressed after the collective-spin-flip algorithm is introduced [see Fig. 5(b)], and a phase transition or crossover emerges, indicated by the peak of the heat capacity [see Fig. 5(c)]. Compared to single spin flips, this strategy enables the system to reach the ideal ground state, providing strong evidence for the presence of kinetic constraints.

VI. CONCLUSION

We have demonstrated that in this article, for P1 Penrose tiling based artificial spin ice, due to complex energy landscape induced large energy barrier, the kinetic pathways to lower energy states are limited when fluctuations are mainly mediated via single spin flips. It provides a distinctive realization of dynamic freezing, presenting opportunities to explore new thermodynamic phenomena in artificially designed systems. Further exploration of this mechanism through direct experimental measurements in thermally active ultrathin samples is anticipated [9,52]. Moreover, recently developed techniques for engineering local structure to bypass dynamic freezing [12,39,53] may provide a route to achieving the fully ordered ground state of this model.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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