

## Tunable meron pair excitations and Berezinskii-Kosterlitz-Thouless phase transitions in the monolayer antiferromagnet MnPSe<sub>3</sub>

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(Received 14 July 2023; accepted 21 February 2024; published 11 March 2024)

Exploration of exotic quantum physics in magnetic systems of reduced dimensionality is among the central themes of condensed matter physics. Here we investigate the nature of magnetic couplings, topological excitations, and critical phenomena in monolayer MnPSe<sub>3</sub>, a representative two-dimensional antiferromagnet. We first establish, based on first-principles calculations, that the  $S = 5/2$  Mn<sup>2+</sup> ions possess single-ion anisotropy, favoring in-plane magnetization, and exhibit isotropic antiferromagnetic pairwise couplings. Next, our atomistic magnetism simulations identify antiferromagnetically coupled meron pairs to be the characteristic topological excitations, whose size, polarity, and dynamics can be delicately tuned by an external magnetic field. Significantly, a critical temperature of  $\sim 30$  K is indicated in both the susceptibility and specific heat, and detailed scaling analyses on the spin correlations and stiffness unambiguously attribute the critical behavior to the Berezinskii-Kosterlitz-Thouless universality class. Our findings thereby provide an ideal candidate antiferromagnetic counterpart of the recently identified monolayer XY ferromagnet of CrCl<sub>3</sub>, the former may harbor distinct topological excitations and multicriticality.

DOI: [10.1103/PhysRevB.109.L100403](https://doi.org/10.1103/PhysRevB.109.L100403)

**Introduction.** The discovery of two-dimensional (2D) crystals [1] opens the era of exploring novel quantum physics and critical phenomena in solid-state systems under reduced dimensionality, wherein the effects of different types of fluctuations become crucial [2–4]. Among these developments, the rise of monolayer magnetic materials provides fertile platforms for investigation of quantum magnetism in genuine 2D spin lattices. In monolayer ferromagnets, the development of long-range order [5–8] vitally relies on anisotropy in easy-axis magnetization. It has been further recognized that, with easy in-plane magnetization, topological defects in the form of vortex-antivortex pairs can be excited, which is the microscopic foundation for the celebrated Berezinskii-Kosterlitz-Thouless (BKT) phase transition [9–12] beyond the Landau scenario. As counterparts, antiferromagnets have also been thinned to the monolayer regime, with the transition-metal phosphorus trichalcogenides of  $TMPX_3$  ( $TM = \text{Cr, Mn, Fe, Co, Ni}$ , and  $X = \text{S, Se}$ ) [13–16] as the most extensively explored class [17]. This family of layer materials shares the commonality of hosting intralayer antiferromagnetism, favoring either the Néel or zigzag phase within the honeycomb lattice. These systems may provide not only ideal platforms for probing 2D antiferromagnetic (AFM) criticality and dynamics under various external fields [14,18–20], but also innovative technology potentials rooted in their inherent fast responses and non-stray-field characteristics [21,22].

The AFM coupling usually induces strong magnetic frustration and fluctuation to the system, capable of exciting or stabilizing various topological quasiparticles such as AFM skyrmions, in the presence [23–25] or even absence [26,27] of Dzyaloshinskii-Moriya (DM) interaction [28–30].

As compelling examples of novel physical phenomena harbored in the layer  $TMPX_3$  compounds, a line of exotic observations has been reported as the systems reduce their thickness towards the monolayer limit. In particular, some monolayer systems exhibit clear signature of suppressed long-range AFM order [15], and may thus serve as candidate arenas to exhibit BKT criticality. Here, the systems inherently possess internal sublattice degrees of freedom beyond their ferromagnetic (FM) counterparts [31–34], potentially enabling richer variety and greater tunability of the local topological excitations with external fields.

In this work, we perform a comprehensive multiscale simulation study of the nature of magnetic couplings, topological excitations, and critical phenomena in monolayer MnPSe<sub>3</sub>, which has been experimentally realized [14]. Based on first-principles calculations, we establish that the  $S = 5/2$  Mn<sup>2+</sup> ions possess single-ion anisotropy, favoring in-plane magnetization, and exhibit isotropic antiferromagnetic pairwise couplings. Next, our detailed atomistic magnetism simulations allow us to identify antiferromagnetically coupled meron pairs to be the characteristic topological excitations, whose size, polarity, and dynamics can be delicately tuned by an external magnetic field. Significantly, a critical temperature of  $\sim 30$  K is indicated in both the susceptibility and specific heat, and detailed scaling analyses on the spin correlations and stiffness unambiguously attribute the critical behavior to the

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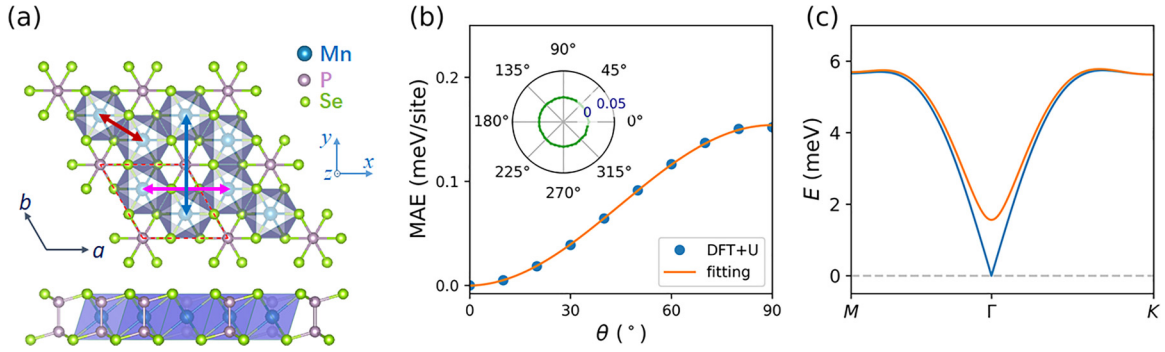


FIG. 1. (a) Top and side views of atomic structure of the  $\text{MnPSe}_3$  monolayer. In the top view, the exemplified first, second, and third Mn-Mn pairs are respectively denoted by red, cyan, and blue dual-headed arrows. (b) MAE upon rotating the Néel vector of the AFM state from the in-plane ( $0^\circ$ ) to out-of-plane ( $90^\circ$ ) direction. The curve is a parabolic fitting using  $\sin\theta$ . The inset shows the MAE evolution (green circle) upon rotating the Néel vector within the  $xy$  plane. (c) The gapped and gapless spin excitation spectra calculated from an in-plane Néel configuration.

Berezinskii-Kosterlitz-Thouless universality class. Furthermore, by elevating the strength of the out-of-plane magnetic field, we can convert the AFM meron pairs into a new type of topological quasiparticles with FM-coupled core and AFM-coupled in-plane vorticity, with uncompensated topological charge. Collectively, our results provide valuable atomic-scale insights into the AFM-BKT physics harbored by the easy-plane  $\text{TMPX}_3$  monolayers.

*First-principles establishment of the spin Hamiltonian.* Figure 1(a) schematically shows the crystal structure of monolayer  $\text{MnPSe}_3$ , composed of a honeycomb lattice formed by edge-sharing  $\text{MnSe}_6$  octahedra and P dimers at the hollow sites. It is noted that recently monolayer  $\text{MnPSe}_3$  has been experimentally synthesized, with its Néel vector and universality class demonstrated to be switchable to the Ising-type via in-plane strain [14]. We here focus on the equilibrium case subject to the XY regime, which has not yet been explicitly characterized in the monolayer limit.

We perform first-principles calculations [35] (see also Refs. [29,36–53] in the Supplemental Material) to obtain the equilibrium lattice structure and predominant exchange couplings of the  $\text{MnPSe}_3$  monolayer. The rotationally invariant DFT+U approach [39] is applied to account for the strong onsite Coulomb repulsion of Mn-3d electrons, and spin-orbit coupling is included to resolve the magnetic anisotropy. The magnetic ground state within the collinear regime is verified to be Néel-type AFM, energetically more favorable than the FM, stripy-AFM, and zigzag-AFM phases by 66.0, 33.2, and 24.2 meV/Mn respectively, consistent with previous studies [54,55]. Figure 1(b) shows the magnetocrystalline anisotropy energy (MAE) upon rotating the Néel vector from in-plane to out-of-plane directions. The calculated MAE is 0.15 meV/Mn preferring in-plane magnetization, suggesting that the monolayer system is an intrinsic XY-like magnet.

Given the highly localized nature of Mn-3d electrons, the magnetic interactions within the  $\text{MnPSe}_3$  monolayer can be described by a generic bilinear Hamiltonian

$$H = - \sum_i \mathbf{S}_i \mathbf{A}_i \mathbf{S}_i - \sum_{i<j} \mathbf{S}_i \mathbf{J}_{ij} \mathbf{S}_j \quad (1)$$

where  $\mathbf{S}_i$  is the spin operator of the  $i$ th  $\text{Mn}^{2+}$  ion with  $|\mathbf{S}_i| = 5/2$ ,  $\mathbf{A}_i$  and  $\mathbf{J}_{ij}$  are  $3 \times 3$  tensors quantifying the single-ion anisotropy (SIA) and two-ion exchange couplings up to the third-nearest-neighbor pairs, respectively. The threefold rotational symmetry reduces  $\mathbf{A}_i$  to a scalar  $A_i = A_{i,zz} - A_{i,xx}$ , whose positive (negative) value favors out-of-plane (in-plane) magnetization. The inversion symmetry prohibits the existence of DM interactions [28,29] between the first- (1NN) and third-nearest-neighbor (3NN) pairs because the bisecting points of these pairs are located right at the inversion centers of the monolayer, while the second-nearest-neighbor (2NN) pair allow finite DMI with the DM vector perpendicular to the twofold rotation axis bisecting the bond, following Moriya's rule [29,30]. Other symmetry-allowed components of SIA and exchange couplings can be computed by using the well-developed four-state method [41–45]. By mapping the DFT+U total energies of a series of noncollinear magnetic configurations [46] to the spin Hamiltonian in Eq. (1), this approach enables a full anatomy of bilinear exchanges including non-Heisenberg terms that might influence the spin ground state or dynamics [56,57]. For the antisymmetric part of exchange couplings (DM interactions), the magnitude for the symmetry-allowed 2NN pairs is  $|\mathbf{D}| = 8.28 \mu\text{eV}$ , rather small and should have negligible effect on the spin dynamics; the 1NN and 3NN DM interactions are even smaller by one order of magnitude and can be viewed as numerical zero as required by symmetry (bond center at the inversion center of the monolayer). More details can be found in the Supplemental Material [35]. For the symmetric parts, all off-diagonal components of  $\mathbf{J}_{ij}$  are calculated to be smaller than  $1 \mu\text{eV}$ , while the diagonal components have strengths larger by several orders of magnitude (Table I), and are all AFM

TABLE I. Exchange couplings of the monolayer  $\text{MnPSe}_3$  (in meV).

Exchange pair	$J_{xx}$	$J_{yy}$	$J_{zz}$
1NN	-0.582	-0.586	-0.586
2NN	-0.059	-0.058	-0.062
3NN	-0.332	-0.332	-0.332

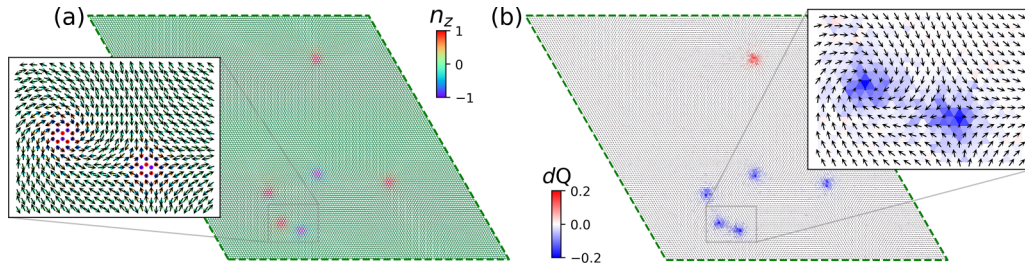


FIG. 2. Snapshots from an LLG simulation ( $L = 120$ ) at 0.5 K, showing (a) magnetization of the antiferromagnetically coupled sublattices containing topological excitations, and (b) topological charge distribution on one sublattice. The insets are zoomed-in views of a meronic vortex/antivortex pair. The color coded  $n_z$  and  $dQ$ , respectively, represent the out-of-plane component of the spin vectors and the topological charge.

exhibiting negligible exchange anisotropy. Interestingly, the coupling strengths of the 2NN pairs are even smaller than those of 3NN, mainly due to the weaker hybridization with anion  $p$  orbitals on the corresponding superexchange paths, as has been elucidated based on collinear magnetic calculations [54]. The only nonvanishing SIA parameter is calculated to be  $A_i = -0.035$  meV preferring in-plane magnetization, which accounts for the microscopic origin of MAE. As a running summary, our calculations characterize monolayer MnPSe<sub>3</sub> as an XY-type antiferromagnet, whose spin Hamiltonian can be effectively reduced to the form of an easy-plane SIA term plus AFM Heisenberg exchange couplings,

$$H = - \sum_i A_i S_{i,z}^2 - \sum_{i<j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j. \quad (2)$$

Based on Eq. (2) parameterized as Table I, we can calculate the magnon spectrum of monolayer MnPSe<sub>3</sub> with an in-plane Néel ordered state, invoking the linear spin wave theory [58]. As presented in Fig. 1(c), the SIA lifts the degeneracy of the two branches, with the splitting more pronounced near the  $\Gamma$  point. The gapless lower mode indicates the absence of finite-temperature long-range order in monolayer MnPSe<sub>3</sub>, consistent with 2D easy-plane anisotropy. In the language of symmetry breaking, this gapless magnon, with linear dispersion near the  $\Gamma$  point, signals the Goldstone mode associated with the broken U(1) symmetry. Such Goldstone mode is typically destructive to finite-temperature long-range order because it can introduce long-wavelength fluctuations to the system without energy cost.

*BKT physics revealed by atomistic spin dynamics simulations.* We now proceed to study the magnetic properties of monolayer MnPSe<sub>3</sub> based on the established spin Hamiltonian. The large magnetic moments of Mn<sup>2+</sup> ions ( $5 \mu_B$ ) far away from the quantum limit can be treated elastically as classical spins, and their dynamics are investigated by numerically integrating [52] the Landau-Lifshitz-Gilbert (LLG) equation [49,50] (see Supplemental Material, Note S1 [35] for details). The system, simulated by an  $L \times L$  supercell, is cooled down from the paramagnetic state to various target temperatures and then equilibrated for subsequent analyses.

A representative snapshot, taken at  $t = 30$  ps from a simulated cooling process lasting for 200 ps with  $L = 120$ , is shown in Fig. 2(a), revealing the emergence of antiferromagnetically coupled vortex/antivortex pairs. Indeed, such topological defects can be observed at the very beginning

of the cooling process from a paramagnetic initial state ( $t \sim 2$  ps), and all annihilate after  $t > 125$  ps, leaving the system with zero topological charge while still presenting spin superfluidity behavior. Different from the archetypal 2D XY model [9,10] wherein spins near the vortex/antivortex cores have energetically unfavored large swirling angles, here the three-component spins of Heisenberg feature can accommodate out-of-plane vortex/antivortex cores, efficiently lowering the energy cost to excite topological defects [31]. In our simulations under fixed temperature of 0.5 K, the energy difference between a configuration containing 20 pairs of merons and another configuration with all merons annihilated is 0.223 meV/site. The topological charges of the emerging AFM vortices/antivortices can be calculated separately within the two sublattices [59] and should yield opposite values. As spatially resolved in Fig. 2(b) within one of the two sublattices, the topological charges peak at the core region of vortex/antivortex, and their integration over a single topological defect yields  $-1/2$  ( $1/2$ ), characterizing the excitations as merons (antimerons) commensurate with the in-plane magnetized background. A meron can be viewed as half a skyrmion, a more celebrated topological defect living in the out-of-plane magnetization background [60]. A pair of merons (insets of Fig. 2) restores the integer topological charge of  $\pm 1$ , forming a bimeron. Being the characteristic topological excitations in BKT phase [61,62], merons in various systems can exhibit distinct dynamics and decaying rate. As ubiquitously seen in BKT systems, the vortexlike and antivortexlike meronic quasiparticles with opposite vorticity experience an attractive force, governing their motion towards each other and collapse even at low temperature. Consequently, the lifetime of vortex pairs in our simulated AFM system is typically no longer than 125 ps, as estimated by our topological charge analysis and statistics on consecutive snapshots extracted from atomistic spin dynamics simulations. The faster dynamics of AFM bimerons compared to their FM counterparts [63–65] is akin to skyrmionic systems [59].

Going beyond the real-space visualization of topological defects, we carry out systematic analysis of the thermodynamic properties in the spirit of ensemble average to gain further insights into the criticality of the AFM monolayer. Simulations are performed with a much longer time scale of  $\sim 10^5$  ps at each target temperature to gather substantial equilibrium snapshot configurations for statistics. By varying the target temperature, we trace the evolution of thermodynamic observables including the normalized staggered



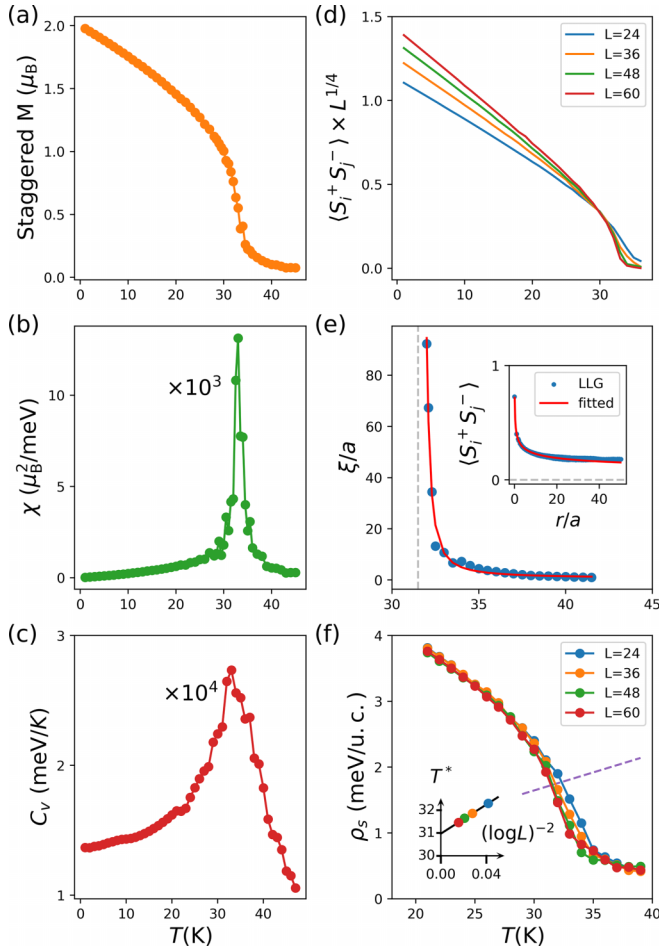


FIG. 3. Thermodynamic properties and scaling analyses of a monolayer MnPSe<sub>3</sub>. Temperature dependence of (a) staggered magnetization, (b) susceptibility, and (c) specific heat obtained from LLG simulations ( $L = 60$ ). (d) In-plane spin correlations between two most distant sites within the supercell, multiplied by  $L^{1/4}$  for finite-size scaling analysis. (e) Correlation length as a function of temperature, fitted by Eq. (4). Inset: in-plane spin correlation at  $T \sim 31.2$  K, fitted by the power-law decay  $A(r/a)^{-1/4}$  ( $a$  being the lattice constant). (f) Spin stiffness as a function of temperature. For a given simulation size  $L$ , the crossing point between the corresponding curve and the purple dashed line defined by the BKT universality relation  $\rho_s = 2k_B T/\pi$  yields a transition temperature  $T^*(L)$ . Inset: linear fitting using Eq. (5).

magnetization and its susceptibility, as well as the specific heat. As shown in Figs. 3(a)–3(c), these quantities coherently demonstrate that the system undergoes a magnetic transition at  $\sim 33$  K. Due to easy-plane anisotropy,  $M_z$  is quenched to zero at all temperatures. The broad peak of susceptibility characterizes the 2D XY behavior, with a slowly decaying tail below the critical temperature distinct from the vanishing susceptibility commonly observed in prototypical Ising phase transitions.

It has long been established, based on renormalization group analysis [66] or quantum Monte Carlo simulations [67] in some simplified models with nearest-neighbor couplings, that even very weak easy-plane anisotropy can enable a Heisenberg spin system to present BKT criticality. However,

for models associated with realistic materials, the interplay between multiple exchange couplings might mediate non-collinear long-range orders competing the BKT phase [68]. Even in the BKT regime, recent studies [69] elucidate that the involvements of long-range couplings can subtly alter the criticality. Analyses of the critical behavior based on the statistical results thus require extra caution. Typically, the  $T_{\text{BKT}}$  value estimated from specific heat and magnetic susceptibility may suffer from some quantitative uncertainty due to finite-size effect [70–72]. To access the true thermodynamic limit, reliably characterize BKT phase transition, and determine  $T_{\text{BKT}}$ , scaling analysis on the finite-size simulation results is necessary. Considering the in-plane spin components, for systems subject to the 2D XY universality class, the correlation function at  $T = T_{\text{BKT}}$  satisfies

$$\langle S_i^+ S_j^- \rangle \sim L^{-1/4}, \quad (3)$$

with  $S_i$  and  $S_j$  being a pair of most distant spins within the supercell in size of  $L$ , and  $S_i^\pm = S_i^x \pm iS_i^y$ . Given this scaling law, we can define a new scaling function  $\langle S_i^+ S_j^- \rangle L^{1/4}$ , which should predict the transition temperature independent of the supercell size. Figure 3(d) shows its temperature dependence, wherein the four curves corresponding to different  $L$  cross at the same transition temperature of 30.2 K, laying a slight correction on the value obtained from susceptibility and specific heat analyses.

Elevating the temperature across  $T_{\text{BKT}}$ , the spatial decay behavior of in-plane spin correlations switches from  $1/4$  power law to exponential. At the critical temperature, the correlation length  $\xi$  diverges. This universal property provides an alternative way to confirm the BKT nature of the phase transition by fitting the high-temperature sector of the  $\xi$ - $T$  curve, which satisfies

$$\xi \sim \exp[b(T - T_{\text{BKT}})^{-1/2}] \quad (4)$$

with  $b$  a system-specific parameter. Figure 3(e) shows such exponential fitting from which we attain  $T_{\text{BKT}} \sim 31.2$  K in nice consistency with scaling analysis. As a crosscheck from the low-temperature side, our power-law fitting of the in-plane spin correlation at  $T = 31.2$  K [inset of Fig. 3(e)] attests it as the lower bound of transition temperature.

From the viewpoint of energetics, another quantity eligible for scaling analysis is the spin stiffness  $\rho_s$ . As a temperature-dependent thermodynamic quantity,  $\rho_s$  measures the free energy cost to achieve an infinitesimal phase twist, or gradient, to the spin system, which can characterize the whirling energy of thermally excited topological defects. The spin stiffness is finite at the low-temperature BKT phase, and vanishes above the transition temperature; critically, at  $T = T_{\text{BKT}}$ , it follows a universal relationship,  $\rho_s = 2k_B T/\pi$  [73]. In practice, the logarithmic finite-size correction to  $T_{\text{BKT}}$  has to be considered in the simulations, which reads [74]

$$T^*(L) = T_{\text{BKT}} + C(\ln L)^{-2}. \quad (5)$$

$T^*(L)$  is extracted from the crossing between the line representing the universal  $\rho_s = 2k_B T/\pi$  relationship and the  $\rho_s(T)$  curves from finite-size simulations, and  $C$  is a system-specific constant. Figure 3(f) presents such scaling analysis of  $\rho_s$ , from which we extrapolate the linearly fitted  $T^* - (\ln L)^{-2}$  correlation to the thermodynamic limit and estimate that

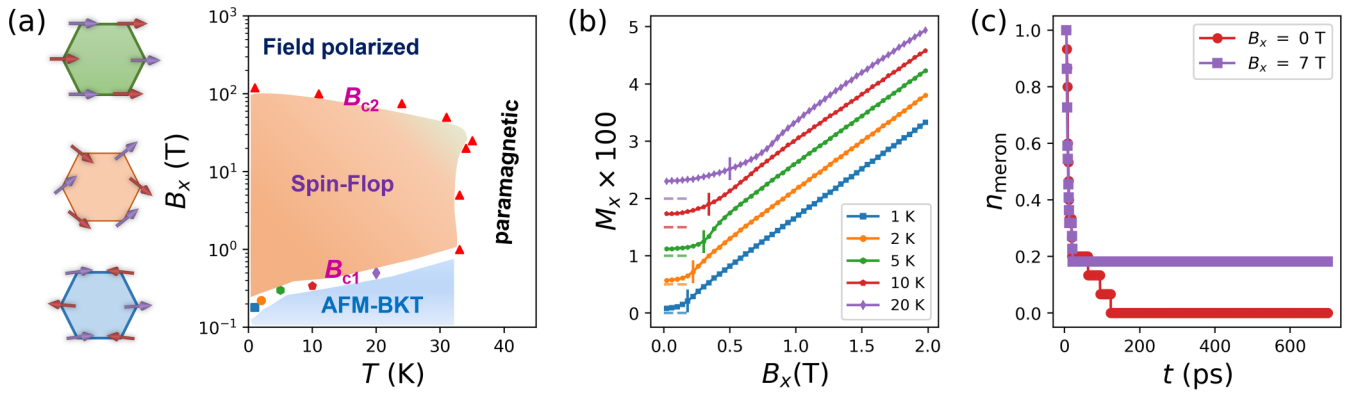


FIG. 4. (a) Schematic phase diagram of a monolayer MnPSe<sub>3</sub> under an in-plane magnetic field along  $x$ ,  $B_x$ . Panels on the left sketch the corresponding short-range spin alignments in the identified phases color highlighted in the diagram. (b) Magnetization along  $x$  as a function of  $B_x$  and temperature. The curves are vertically shifted for clarity, with the zeros indicated by the corresponding horizontal dashed lines. The upturning points denoted by the vertical bars determine the temperature-dependent  $B_{c1}$  marked in (a). (c) Evolution of the normalized meron density upon cooling the system to  $T = 0.5$  K without and with  $B_x$ .

$T_{\text{BKT}} \sim 31$  K, again in good accordance with previous analyses.

We have thus far confirmed intrinsic Néel-type AFM-BKT physics in monolayer MnPSe<sub>3</sub>, the underlying mechanism of which can be understood via analyzing the interplay between exchange couplings, single-ion anisotropy, and lattice geometry. In the  $J_1$ - $J_2$ - $J_3$ - $A$  model, All  $J_i$ 's are isotropic and negative, but due to the honeycomb geometry,  $J_1$  and  $J_3$  favor intersublattice AFM coupling, while  $J_2$  favors intrasublattice AFM coupling. The calculated  $J_1$  and  $J_3$  overwhelm  $J_2$  in magnitude, thus the Néel correlation prevails in the honeycomb lattice, coupling the two sublattice antiferromagnetically. The SIA term  $A$ , which favors in-plane magnetization and is isotropic within the XY plane, constitutes the only interaction to induce magnetic anisotropy and drives the system to the AFM-BKT regime with quasi-long-range order and meronic excitations at low temperature.

*Field-tunable phases and meron pairs.* Unlike ferromagnets, AFM systems encompassing staggered magnetized sublattices respond to external magnetic fields in a macroscopically less sensitive manner. Microscopically, the competition between the Zeeman interaction and AFM exchange couplings can be regarded as magnetic frustration, which might mediate richer physics in topological excitations and vortex dynamics. For uniaxially anisotropic Heisenberg antiferromagnets, applying a uniform magnetic field along the easy axis can switch it from the long-range AFM order to the spin-flop (SF) phase [75–77], the latter featuring FM (AFM) alignment of magnetic moments along (perpendicular) to the easy axis. It would be intriguing to exploit relevant physics in BKT systems with quasi-long-range order and topological excitations.

Given the easy-plane anisotropy, we first tackle the case of applying an in-plane field along the  $x$  axis ( $B_x$ ), which breaks the planar rotational symmetry of the Hamiltonian. By performing LLG simulations and tracking proper order parameters, we identify three distinct phases in addition to the trivial paramagnetic state. The corresponding short-range spin alignments and  $B_x$ - $T$  phase diagram are illustrated in

Fig. 4(a). The magnetization along  $x$  is invoked as the order parameter to trace the phase evolution. As shown in Fig. 4(b), the curves appear flat when  $B_x$  is small, and upturn after  $B_x$  exceeds a temperature-dependent critical field  $B_{c1}$ , signaling a field-induced transition from the AFM-BKT to SF phase, which persists up to the critical temperature (Fig. S1). Interestingly, the transition temperature from the AFM-BKT/SF phase to the paramagnetic state exhibits weak dependence on  $B_x$  [Figs. S2(a) and S2(b)]. Our scaling analyses on the squared staggered magnetization and spin stiffness coherently suggest that the BKT physics is fully suppressed at  $B_x = 20$  T, and the transition becomes Ising-type [Figs. S3(a) and S3(c)]. When  $B_x$  is further raised to surpass a temperature-dependent critical value of  $B_{c2}$ , the system enters a field-polarized phase [Fig. 4(a)], namely, all spins align with the field, and the staggered magnetization vanishes [Fig. S4(a)].

The topological defects provide a real-space signature to further distinguish the SF phase from the low-field AFM-BKT regime. Our simulations indicate that the meron excitation and dynamics below  $B_{c1}$  resemble the field-free case. On the contrary, in the SF phase under  $B_x > B_{c1}$ , the major spin components remain antiferromagnetically aligned, favoring two energetically degenerate directions perpendicular to the field ( $\pm y$  in the present case). Such in-plane anisotropy leads to formation of multidomain configurations and domain-wall-enriched meron dynamics. At  $B_x = 7$  T, the meron pairs are bounded to the Bloch-line-shaped domain and exhibit Néel-type geometry [Fig. S6(a)]. Compared to the field-free case, these domain-wall merons possess a substantially decreased mobility, and accordingly elongated lifetime. Figure 4(c) contrasts the temporal evolution of meron density in the AFM-BKT and the spin-flop regimes. In the zero-field case, all merons collapse within 125 ps, while the applied field  $B_x = 7$  T significantly increases the lifetime of a portion of merons pinning to the domain walls up to 500 ps. Such enhanced topological stability and frozen meron dynamics implies the formation of a meron glass state [78], which is observed in our simulations when  $B_x$  is in the interval from 5–10 T, thereby could be verified by the recently developed imaging technique [79]. Further increasing  $B_x$  to 20 T leads to severe

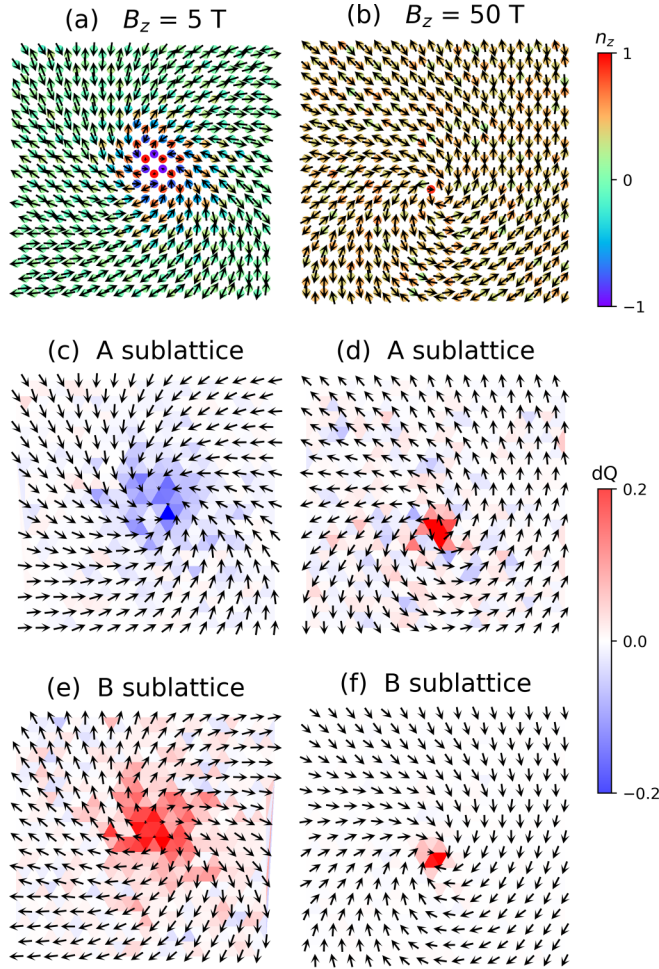


FIG. 5. Field-induced conversion of an AFM meron into a new type of FM-AFM hybrid meron. Snapshots from LLG simulations under an out-of-plane magnetic field (a), (c), (e)  $B_z = 5$  T and (b), (d), (f)  $B_z = 50$  T, corresponding to a typical AFM meron and a new FM-AFM hybrid meron, respectively. Color coding follows Fig. 2.

elongative distortion and eventual disappearance of the domain-wall merons [Fig. S6(b)].

Exertion of an out-of-plane magnetic field ( $B_z$ ) induces a more laconic physics. Because the rotational symmetry of the Hamiltonian is preserved, the transition temperature and critical behavior in such  $B_z$  induced spin-flop phase [Figs. S2(c) and S2(d); Figs. S3(b) and S3(d)] do not exhibit obvious difference from the field-free case. The in-plane staggered magnetization is gradually suppressed when  $B_z$  is increased, before vanishing to zero at  $B_z \sim 120$  T [Fig. S4(b)]. Figure S5 presents the phase diagram in the presence of  $B_z$ , illustrating the AFM phase falls into the spin-flop phase with arbitrary small  $B_z$ . Microscopically, the topological excitations in this circumstance are still merons presenting pairing behavior, but with shrinking core upon increasing  $B_z$  (Figs. S7 and S8). More importantly, a substantially large  $B_z$  can flip the out-of-plane polarizations of the meron cores within the two sublattices, from opposite [Fig. 5(a)] to aligned [Fig. 5(b)]; meanwhile, the in-plane whirling pattern remains intact albeit with smaller components under high field. Consequently, a drastic change to the topological charge distribution is

induced as compared to the low-field case [Figs. 5(c), 5(e) vs. 5(d), 5(f)]. The vortices in such spin-flop BKT phase, with field-modulated polarization and uncompensated topological charge, can be viewed as an intermediate regime between the pure AFM-BKT phase discussed above and the well-studied FM-BKT systems [31–34]. It would be intriguing to explore possible additional criticality associated with the meron core flipping, which is the subject of future works.

*Discussion and summary.* Since its conceptualization, the BKT phase transition as a profound framework to unify abundant critical phenomena by topology has been observed in a plethora of condensed matters in two dimensions, including superfluid [80], superconducting [81], and ultracold atom systems [82]. For decades, efforts to realize BKT transitions in magnetic systems have mainly focused on quasi-2D layer XY-type magnets [83–90], wherein the unavoidable interlayer couplings may mediate 3D long-range magnetic order that competes or even suppresses the BKT phase. The topological excitations revealed here in monolayer MnPSe<sub>3</sub> suggest it as a promising candidate to probe magnetic BKT criticality and study the field-tunable AFM vortex dynamics. Exempted from the complexity of interlayer coupling encountered in multilayer magnets [83–90], characterization of the intrinsic BKT order in monolayer systems are likely more transparent and definitive [31–34]. Here we note that, given the ultrathin nature of monolayer antiferromagnets, neutron scattering or nuclear magnetic resonance widely utilized for detecting BKT physics in multilayer magnets [87–89] might become inapplicable. Promising approaches for verifying the AFM-BKT criticality and visualizing the topological excitations may include the polarized Raman spectroscopy [15] and some real-space imaging techniques [14,16,79].

Summarizing, our work provides a comprehensive characterization of the anisotropic magnetic interactions, the type and dynamics of the AFM topological excitations, and tunability of the criticality in the monolayer MnPSe<sub>3</sub>. The insights from the present study can have broader implications beyond characterizing an intrinsic AFM-BKT order. The established phase diagram, the vortex dynamics harnessed by sub-Tesla  $B_x$ , and the newly identified meronic quasiparticles with flipped cores under  $B_z$ , collectively highlight the delicate interplay between the magnetic field and AFM coupling. Prospectively, it would also be intriguing to investigate how these fundamentally important responses evolve when chiral Dzyaloshinskii-Moriya interactions [91] are further introduced via proper symmetry reduction [92].

## ACKNOWLEDGMENTS

We acknowledge Professor Wei Qin and Dr. Xiaoyin Li for helpful discussions and valuable suggestions. This work was supported by National Natural Science Foundation of China (Grants No. 11904350, No. 12374458, and No. 11974323), the Anhui Provincial Natural Science Foundation (Grant No. 2008085QA30), the Innovation Program for Quantum Science and Technology (Grant No. 2021ZD0302800), the Strategic Priority Research Program of Chinese Academy of Sciences (Grant No. XDB0510200), and the Anhui Initiative in Quantum Information Technologies (Grant No. AHY170000).



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