Skyrmions and magnetic bubbles in spin-orbit coupled metallic magnets

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Motivated by the observation of Skyrmion-like magnetic textures in two-dimensional (2D) itinerant ferromagnets Fe_nGeTe_2 ($n \ge 3$), we develop a microscopic model combining itinerant magnetism and spin-orbit coupling on a triangular lattice. The ground state of the model in the absence of magnetic field consists of filamentary magnetic domain walls revealing a striking similarity with our magnetic force microscopy experiments on Fe_3GeTe_2 . In the presence of magnetic field, these filaments were found to break into large-size magnetic bubbles in our experiments. We identify uniaxial magnetic anisotropy as an important parameter in the model that interpolates between magnetic Skyrmions and ferromagnetic bubbles. Consequently, our work uncovers new topological magnetic textures that merge properties of Skyrmions and ferromagnetic bubbles.

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I. INTRODUCTION

The topologically stable magnetic structures, such as the Skyrmions, are considered the building blocks of nextgeneration data storage and processing devices [1-8]. Consequently, identifying suitable magnetic materials that host such unusual magnetization textures has become a rapidly emerging area of research [9-19]. One of the key requirements for applications is the manipulation of such textures via ultralow electrical currents, and therefore it is desirable to have such magnetic textures realized in metallic magnets [11,12,20-25]. The appearance of isolated Skyrmions as well as Skyrmion lattices has been reported in thin films of a variety of chiral metallic magnets [12-15,26-33]. While a common understanding of magnetic Skyrmions in metals relies heavily on either a suitable free-energy functional in a continuum or classical spin models on lattices [8,34–39], more recently the importance of microscopic Hamiltonian-based understanding of Skyrmion formation has been recognized [40-43]. The latter approach allows for a finer distinction between ferromagnetic bubbles and Skyrmions, thereby providing a tunable control on the stability of different topological textures.

Motivated by the recent observation of metallic ferromagnetism in the van der Waals (vdW) magnets Fe_nGeTe_2 with n = 3, 4, 5 [44–46], we extend and investigate a recently proposed microscopic model for the broad class of vdW magnets. The model combines the effects of itinerant magnetism, spinorbit coupling, and uniaxial anisotropy [40]. Starting from the microscopic electronic Hamiltonian on a triangular lattice, we derive an effective spin model. We explicitly demonstrate via large-scale Monte Carlo (MC) simulations, that the ground state of the model consists of filamentary domain wall structures which show agreement with the magnetic force microscopy (MFM) images on high-quality single crystals of Fe_3GeTe_2 . However, the MFM images in the presence of an external magnetic field deviate from the in-field calculations within the model. We show that the inclusion of a uniaxial anisotropy term in the Hamiltonian allows for a consistent description of the results at zero and finite magnetic fields. Furthermore, we identify the easy-axis anisotropy as an important tuning parameter for the relative stability of different types of topological structures in spin-orbit coupled itinerant magnets.

II. THEORETICAL MODEL

In metallic magnets that consist of large magnetic moments and spin-orbit coupling (SOC), a generic starting model is the ferromagnetic Kondo lattice model (FKLM) in the presence of a Rashba term. We consider the Hamiltonian on a triangular lattice as

$$H = -t \sum_{i,\gamma,\sigma} (c_{i,\sigma}^{\dagger} c_{i+\gamma,\sigma} + \text{H.c.}) - J_{\text{H}} \sum_{i} \mathbf{S}_{i} \cdot \mathbf{s}_{i}$$
$$-i\lambda \sum_{i,\gamma,\sigma\sigma'} c_{i\sigma}^{\dagger} [\tau \cdot (\hat{\gamma} \times \hat{\mathbf{z}})]_{\sigma\sigma'} c_{j\sigma'} - h_{z} \sum_{i} S_{i}^{z}. \quad (1)$$

The annihilation (creation) operators $c_{i\sigma}$ ($c_{i\sigma}^{\dagger}$) satisfy the usual Fermion algebra. $J_{\rm H}$ (λ) denotes the strength of Kondo (Rashba) coupling, *t* is the nearest-neighbor hopping parameter on triangular lattice. τ is a vector operator with the three Pauli matrices as components. $\mathbf{s}_i(\mathbf{S}_i)$ denotes the electronic spin operator (localized classical spin) at site *i*. Assuming the lattice constant to be unity, $\hat{\gamma} \in \{\mathbf{a}_1, \mathbf{a}_2, \mathbf{a}_3\}$ are the primitive vectors of the triangular Bravais lattice with \mathbf{a}_1 =(1,0), \mathbf{a}_2 =(1/2, $\sqrt{3}/2$) and $\mathbf{a}_3 = (-1/2, \sqrt{3}/2)$. The last term in Eq. (1) represents the Zeeman coupling of local moments to

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an external magnetic field of strength h_z . t = 1 sets the basic energy unit in the model. Presence of Skyrmion Hall effect in the vdW magnets indicates that the strong coupling limit is more relevant as compared to the weak-coupling Ruderman-Kittel-Kasuya-Yoshida (RKKY) limit [47–50]. This leads to the Rashba double-exchange (RDE) Hamiltonian on a triangular lattice [40]

$$H_{\rm RDE} = \sum_{\langle ij \rangle, \gamma} \left[g_{ij}^{\gamma} d_i^{\dagger} d_j + \text{H.c.} \right] - h_z \sum_i S_i^z, \qquad (2)$$

where, $d_i(d_i^{\dagger})$ annihilates (creates) an electron at site *i* with spin parallel to the localized spin. Site $j = i + \gamma$ is the nearest neighbor (nn) of site *i* along one of the three symmetry directions on the triangular lattice. The projected hopping g_{ij}^{γ} depend on the orientations of the local moments \mathbf{S}_i and \mathbf{S}_j . The tight-binding t_{ij}^{γ} and Rashba λ_{ij}^{γ} contributions to $g_{ij}^{\gamma} =$ $t_{ii}^{\gamma} + \lambda_{ii}^{\gamma}$ are given by [40]

$$t_{ij}^{\gamma} = -t \left[\cos\left(\frac{\theta_i}{2}\right) \cos\left(\frac{\theta_j}{2}\right) + \sin\left(\frac{\theta_i}{2}\right) \sin\left(\frac{\theta_j}{2}\right) e^{-i(\phi_i - \phi_j)} \right],$$

$$\lambda_{ij}^{\mathbf{a}_1} = \lambda_{ij}^x, \quad \lambda_{ij}^{\mathbf{a}_{2/3}} = \pm \frac{1}{2} \lambda_{ij}^x + \frac{\sqrt{3}}{2} \lambda_{ij}^y,$$

$$\lambda_{ij}^x = \lambda \left[\sin\left(\frac{\theta_i}{2}\right) \cos\left(\frac{\theta_j}{2}\right) e^{-i\phi_i} - \cos\left(\frac{\theta_i}{2}\right) \sin\left(\frac{\theta_j}{2}\right) e^{i\phi_j} \right],$$

$$\lambda_{ij}^y = i\lambda \left[\sin\left(\frac{\theta_i}{2}\right) \cos\left(\frac{\theta_j}{2}\right) e^{-i\phi_i} + \cos\left(\frac{\theta_i}{2}\right) \sin\left(\frac{\theta_j}{2}\right) e^{i\phi_j} \right],$$

(3)

where θ_i (ϕ_i) is the polar (azimuthal) angle for localized moment \mathbf{S}_i .

The Hamiltonian Eq. (2) describes a modified tight-binding model where the hopping integrals are dependent on the configuration of classical spins. Therefore, the energy of the system depends on the classical spin configurations. This dependence can be formally written as an effective spin Hamiltonian by following a procedure well known for doubleexchange models [40,51]. For the present case of Rashba coupling on a triangular lattice, we obtain

$$H_{\text{eff}} = -\sum_{\langle ij \rangle, \gamma} D_{ij}^{\gamma} f_{ij}^{\gamma} - h_z \sum_i S_i^z,$$

$$\sqrt{2} f_{ij}^{\gamma} = [t^2 (1 + \mathbf{S}_i \cdot \mathbf{S}_j) + 2t\lambda \hat{\gamma'} \cdot (\mathbf{S}_i \times \mathbf{S}_j) + \lambda^2 (1 - \mathbf{S}_i \cdot \mathbf{S}_j + 2(\hat{\gamma'} \cdot \mathbf{S}_i)(\hat{\gamma'} \cdot \mathbf{S}_j))]^{1/2},$$

$$D_{ij}^{\gamma} = \langle [e^{ih_{ij}^{\gamma}} d_i^{\dagger} d_j + \text{H.c.}] \rangle_{gs}.$$
(4)

In the above, f_{ij}^{γ} (h_{ij}^{γ}) is the modulus (argument) of complex number g_{ij}^{γ} , and $\langle \hat{O} \rangle_{gs}$ denotes expectation values of operator \hat{O} in the ground state. Assuming constant coupling parameters has been shown to be a good approximation for studying ground-state phases of H_{eff} [40], therefore we set $D_{ij}^{\gamma} \equiv D_0 = 1/\sqrt{2}$ to study H_{eff} . We perform Monte Carlo (MC) simulations on Hamiltonian Eq. (4) via the standard Markov chain MC using Metropolis algorithm. We begin the simulations at high temperature and reduce temperature in small steps, using 10^5 MC steps for equilibration at each temperature. The magnetic field value is kept zero during

this cooling process. Finally, at low but finite temperature we increase magnetic field in step-wise manner. Once again 10⁵ MC steps for equilibration are used at each magnetic field value, followed by an equal number of steps for recording averages of observables. Most importantly, the MC approach allows access to full spatial details that can be used not only to identify unusual magnetization textures but also to compare with spatially resolved experimental data as will be discussed later. Therefore, our analysis focuses on the typical spin configurations obtained at low temperatures and their evolution with applied magnetic field. We also compute the spin structure factor $S(\mathbf{q})$ in order to identify the presence of ordered magnetic phases. The spin structure factor (SSF) is given by $S(\mathbf{q}) = \frac{1}{N^2} \langle \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j \exp[-i(\mathbf{r}_i - \mathbf{r}_j) \cdot \mathbf{q}] \rangle$, where N is the number of sites, and the angular bracket denotes averaging over MC configurations.

The low-temperature magnetic states, with varying strengths of SOC, are shown in Fig. 1. For small to intermediate values of λ/t , we find the filamentary domain structure of spins [see Figs. 1(a)-1(b)]. The thickness of these domains decreases with the increasing strength of SOC. For a sufficiently large lattice size, we find that the filaments are oriented along all possible directions, and this is more clearly reflected in the structure factor plots where a circular pattern in the $S(\mathbf{q})$ is obtained [see Figs. 1(d)-1(e)]. This freedom of the domains to orient freely in any direction is a consequence of the anisotropic Dzyaloshinskii-Moriya (DM) interaction encoded in the linear λ term in the f_{ij} in Hamiltonian Eq. (4). For larger values of λ/t , we obtain a spin-spiral state with a single-ordering wavevector [see Figs. 1(c) and 1(f)]. The origin of this state is related to the importance of the λ^2 term in the effective Hamiltonian. Note that the λ^2 term prefers Ising-like interactions of different components along different directions, leading to a classical Kitaev model with degenerate ground states. Therefore, there are three states, related by the rotational symmetry of the triangular lattice, that can be realized as ground states. In terms of the SSF, the symmetry-related states will display a SSF that is already rotated by $2\pi/3$ and $4\pi/3$ with respect to Fig. 1(f). Note that the λ^2 term resembles a truncated dipolar interaction term. Therefore, similar magnetic domains are likely to be present in any simulation that includes dipolar interactions [52].

III. EXPERIMENTS

A. Crystal growth and characterization

Now, we focus on the experimentally observed magnetic domain structures on Fe_3GeTe_2 where the characteristic features of the model, namely the existence of ferromagnetism [53], metallicity [44], and high spin-orbit coupling [54] exist naturally. Fe_3GeTe_2 is a van der Waals layered material exhibiting a hexagonal lattice structure with spacegroup $P6_3$ /mmc. It is an itinerant ferromagnet with a Curie temperature ranging from 220-230 K in bulk [53,55]. Highquality single crystals of Fe_3GeTe_2 , synthesized by chemical vapor transport, were used for our magnetic force microscopic (MFM) measurements. A stoichiometric mixture of the ingredients elements [Fe (3N), Ge (3N), and Te (3N)] in the powder form was sealed in an evacuated quartz tube along with I₂ as



FIG. 1. Real-space view of spin configurations at low temperature (T/t = 0.01) for (a) $\lambda/t = 0.1$, (b) $\lambda/t = 0.5$, and (c) $\lambda/t = 1$. The color bar corresponds to the *z* component, and the arrows indicate the planar components of the spins. (d)–(f) The corresponding spin structure factors for the three values of the SOC strength λ/t .

the transport agent. The tube was kept in a two-zone furnace at a temperature gradient of 750° C/ 650° C. Platelike single crystals were obtained after two weeks. Figure 2(a) shows the optical microscope image of a single crystal of Fe₃GeTe₂. Figure 2(b) shows the experimental out-of-plane x-ray diffraction (XRD) results for the single crystal where sharp peaks were observed. The observed Bragg peaks can be indexed with (00*l*) peaks. Figure 2(c) shows the Laue diffraction pat-



FIG. 2. (a) Optical micrograph of single crystal of Fe_3GeTe_2 utilized in the measurements. (b) Out-of-plane x-ray diffraction pattern of Fe_3GeTe_2 at room temperature. (c) Laue diffraction pattern obtained on Fe_3GeTe_2 single crystal. (d) EDX spectra for Fe_3GeTe_2 .

tern of a Fe_3GeTe_2 crystal, confirming the sixfold symmetry of the hexagonal structure and high crystallinity of the grown sample. The chemical composition of the grown crystals was confirmed from atomic percentage ratios obtained from energy-dispersive x-ray (EDX) spectroscopy measurements within the instrumental limit [Figure 2(d)]. Further characterization details of the sample are reported elsewhere [56].

B. MFM results and their comparison with the theoretical model

We imaged the magnetic domains in Fe₃GeTe₂ by magnetic force microscopy (MFM) performed at different temperatures and under magnetic fields. In magnetic force microscopy (MFM), a silicon cantilever/tip coated with a magnetic coating (typically Co or Al) is used along with an interferometer-based detection system to probe the local magnetic properties of the sample by measuring the magnetic interactions between the tip and the sample. A lowtemperature compatible magnetic force microscope (Attocube LT-MFM), working down to 1.6 K, equipped with a singlemode fiber-based interferometer was used for performing the ferromagnetic domain imaging on the ferromagnet. The phase images presented in this work were obtained in dual-pass mode, also known as lift mode. In the single pass, the cantilever using a feedback loop maps the topography of the sample surface where the effect of van der Waals interactions dominates. Since the magnetic forces are long ranged as compared to the van der Waals forces, the effect of the topographic variations can be eliminated to obtain a pure magnetic signal by lifting the tip to a certain height above the sample. For image processing, Gwyddion software [57] was used.



FIG. 3. MFM dual pass phase images taken on a cleaved single crystal of Fe₃GeTe₂ where (a) was taken at 1.6 K in the zero-field cooled (ZFC) state. The filamentary domains can be seen clearly. (b)–(c) were imaged in the field-cooled state in the presence of the field of the MFM cantilever. Representative spin configurations at T/t = 0.01 and $\lambda/t = 0.5$ for (d) $h_z = 0.2$, (e) $h_z = 0.44$, and (f) $h_z = 0.84$. The color bar corresponds to the *z* component, and the arrows indicate the planar components of the spins.

At 1.6 K, under zero-field cooled (ZFC) condition, we found filamentary domains with a typical domain width of around 900 nm as shown in Fig. 3(a). The domain structure involves stripes and interconnects with striking similarities with the computationally obtained filamentary domains as shown in Fig. 1(b). Note that the domain width obtained in simulations is much smaller compared to what is observed in Fe₃GeTe₂ (FGT). However, we explicitly verify that simulations on larger lattices with smaller λ lead to wider domains as shown in Fig. 4. Therefore, the mechanism of the formation of such domains is correctly captured within our approach. It is important to note that, unlike stripe domains [58] which consist of wide, parallel stripes of alternating magnetic



FIG. 4. Real-space view of spin configurations in $N = 240^2$ system at (a) T/t = 0.01 and $\lambda/t = 0.1$, and (b) T/t = 0.005 and $\lambda/t = 0.05$. The color bar corresponds to the *z* component, and the arrows indicate the planar components of the spins.

orientations, or fractal domains [59,60] which exhibit intricate and irregular patterns, filamentary domain wall structures are characterized by their continuous nature. In certain magnetic materials, especially those with strong magnetic anisotropy and competing interactions, the formation of filamentary domain wall structures can be energetically favorable.

In order to gain a further understanding of the magnetic state of Fe₃GeTe₂, we performed MFM imaging in the fieldcooled (FC) states of the crystal. We first field cooled the crystal under the magnetic field of the MFM cantilever and then performed MFM imaging at 1.6 K. Under this condition, as shown in Fig. 3(b), we found that circular domains (the dark circles) have formed within the stripes with the magnetization pointing opposite to the direction of the tip magnetization, along with rows of circular domains with the magnetization aligned in the opposite direction (the bright circles). This feature was reported earlier in Fe₃GeTe₂ and was related to high perpendicular magnetic anisotropy [61,62]. Interestingly, at several points, under the field, the stripes have also started breaking up into fragments. To investigate the microscopic structure in further detail, we imaged a smaller area [see Fig. 3(c)] under the same condition where we find an assembly of bright circular domains with a background hosting the fragmented stripes. Though the circular domains have a close resemblance with typical Skyrmions, their size is relatively larger (\sim 500 µm) and may be called "magnetic bubbles." These differ from the calculated domain structures within our model. In the model, the zero-field cooled protocol was followed with the temperature reduced to T/t = 0.01 in the absence of a magnetic field followed by the increase of field



FIG. 5. Representative spin configurations at T/t = 0.01 and $\lambda/t = 0.1$ for (a) $h_z = 0.008$, (b) $h_z = 0.024$, and (c) $h_z = 0.032$. The color bar corresponds to the *z* component, and the arrows indicate the planar components of the spins. (d)–(f) The corresponding spin -structure factor for the three values of the magnetic field h_z .

in a step-wise manner until a fully saturated ferromagnetic state is obtained. Within the model, for intermediate values of the SOC strength, which is relevant for Fe_3GeTe_2 as noted earlier, the filamentary domains are seen to gradually break into well-defined Skyrmions upon increasing the magnetic field strength [see Figs. 3(d)–3(f)].

For small SOC, the filamentary domains are wide and the presence of a magnetic field leads to the generation of isolated large Skyrmions [see Figs. 5(a)-5(c)]. The typical size of skyrmions obtained here can be estimated from the inverse of the radius of the circular pattern in SSF [see Figs. 5(d)-5(e)]. Since the Skyrmions do not form any pattern, their presence is difficult to infer from the SSF. Indeed, the SSF shown in Fig. 5(f) indicates a ferromagnetic phase via a single peak at the Γ point. For strong SOC, the zero-field state is threefold degenerate stripe state. This leads to the formation of a densely packed hexagonal pattern of Skyrmions at finite fields [see Figs. 6(a)-6(c)]. As expected, the SSF [Figs. 6(d)-6(f)] is more useful for identifying the formation of Skyrmions in this case as they organize into a lattice. The physics in the formation of filamentary domains in different SOC limit is similar, except for the domain width. In search for a deeper understanding of the difference between the experiment and the model, we studied the effect of a higher external field. For that, the crystal was first warmed up to 300 K and was then field cooled under an applied magnetic field of 1.2 kOe. Subsequently, the magnetic domains were imaged at zero applied field. As shown in Fig. 7(a), in a given area, we found that the stripes of bright domains with circular dark regions have broken into parts and formed individual circular domains with darker central region with brighter perimeters. There are regions where two such domains are seen to be connected where the breaking-up process remained incomplete. While

such circular domains are in majority, flat bright domains are also seen in the same region. In another region, where the surface is slightly tilted with respect to the MFM tip, the domains were imaged from an oblique direction which revealed a conical-like shape of the domains [see Fig. 7(b)]. This means, in Fig. 7(d), the ringlike domains are merely the top view of such conical-shaped domains. We then moved to an area where the bright flat domains are more in number, in order to investigate the distribution of their size and shape. As shown in Fig. 7(c), the flat domains appear to be fragments of the stripes in Fig. 3(a). The domains have condensed into different shapes including nearly circular and elongated ones. All these observations differed significantly from the results of the model [see Figs. 3(d)-3(f)].

IV. ROLE OF EASY-AXIS ANISOTROPY

To account for the discrepancy discussed above, we note that the model discussed so far does not include an important feature of the material, i.e., the presence of easy-axis anisotropy. It is important to ask if the presence of easy-axis anisotropy can explain the presence of large ferromagnetic bubbles in Fe₃GeTe₂ and the domain structure at higher magnetic fields. In order to investigate this, we included an additional term $-A_u \sum_i (S_i^z)^2$ in the Hamiltonian Eq. (4). Positive values of A_u represent an easy-axis scenario relevant to the material. The correspondence between the magnetic field values used in the experiment and the simulations is an important one, and the best way to compare the parameters is to compare the $M/M_{\rm sat}$ ratio at the applied field values. We find that $M/M_{\text{sat}} \approx 0.2$ at 1.2 kOe. For $h_z = 0.028$ [corresponding to Fig. 7(c)], we find $M/M_{sat} = 0.15$. Furthermore, the value of anisotropy parameter $A_u = 0.08$ used in the



FIG. 6. Representative spin configurations at T/t = 0.01 and $\lambda/t = 1$ for (a) $h_z = 0.4$, (b) $h_z = 1.0$, and (c) $h_z = 1.4$. The color bar corresponds to the *z* component, and the arrows indicate the planar components of the spins. (d)–(f) The corresponding spin-structure factor for the three values of the magnetic field h_z .

simulations can be justified by comparing the ratio of the value of magnetization with the applied field oriented along the c axis to that oriented perpendicular to the c axis. We find that for the values of magnetic fields mentioned above,

 $M_z(h||z)/M_x(h||x) \approx 3$ in experiments [56] as well as the simulations.

We find that the anisotropy scale competes with the Dzyaloshinskii-Moriya (DM) terms and therefore, the



FIG. 7. (a)–(c) MFM dual pass images, recorded at different regions on the sample surface, in the field-cooled state (FC) where an external field of 1.2 kOe was applied. The lift height was kept constant at 30 nm for all the images. Representative spin configurations at T/t = 0.01, $\lambda/t = 0.1$, and $A_u = 0.08$ for (d) $h_z = 0.004$, (e) $h_z = 0.016$, and (f) $h_z = 0.028$.

tendency of spins to continuously tilt away from the single Skyrmion-core spin is suppressed. This leads to an expansion of the Skyrmion core and finite regions with ferromagnetic bubble character are stabilized [see Figs. 7(d)-7(f)]. These are consistent with the larger size "magnetic bubbles" that were seen experimentally under the field of the cantilever. Further, in the model, we find that upon increasing magnetic field strength the FM regions grow in size while remaining trapped inside self-enclosing domain walls. These are similar to the ringlike domain structures seen at 1.2 kOe. At even higher magnetic fields the model generates larger-size flat regions that show striking similarities with the flat regions seen experimentally. The variation of the experimental domain structure from point to point could be attributed to an inhomogeneity of the surface properties which might arise from an inhomogeneous distribution of the Fe vacancies in the system. Therefore, the results obtained in the simulations including an easy-axis anisotropy are now consistent with the evolution of the MFM images with applied magnetic field.

V. CONCLUSION

In conclusion, motivated by the demonstration of metallicity as well as ferromagnetism in the 2D materials Fe_nGeTe_2 ($n \ge 3$), we developed a model to understand the origin and the nature of topologically stable magnetic textures in a class of itinerant magnets. The model is generally applicable to spin-orbit coupled systems that display itinerant magnetism. Results of Monte Carlo simulations on our model display agreement with our experimental data on Fe_3GeTe_2 in identifying filamentary domain walls as the ground-state structure in the absence of magnetic field. In the presence of magnetic field the model

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predicts either isolated Skyrmions or Skyrmion lattice, depending on the strength of the SOC. Our MFM experiments, on the other hand, reveal large-size magnetic bubbles with certain additional features. These magnetic structures were understood within the model when an easy-axis anisotropy term, as relevant to the materials under discussion, was included.

Our combined experimental and theoretical investigation on a class of vdW itinerant magnets has allowed us to identify easy-axis anisotropy as an important parameter that can tune the nature of topological textures from Skyrmions to magnetic bubbles. This generalization of Skyrmion-like topological textures establishes a conceptually new framework for characterizing real-space imaging data of experiments on itinerant magnets.

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