

Prediction of stable nanoscale skyrmions in monolayer Fe_5GeTe_2

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Using first-principles calculations and atomistic spin simulations, we predict stable isolated skyrmions with a diameter below 10 nm in a monolayer of the two-dimensional van der Waals ferromagnet Fe_5GeTe_2 , a material of significant experimental interest. A very large Dzyaloshinskii-Moriya interaction (DMI) is observed due to the intrinsic broken inversion symmetry and strong spin-orbit coupling for monolayer Fe_5GeTe_2 . We show that the nearest-neighbor approximation, often used in literature, fails to describe the DMI. The strong DMI together with moderate in-plane magnetocrystalline anisotropy energy allows to stabilize nanoscale skyrmions in out-of-plane magnetic fields above ≈ 2 T. The energy barriers of skyrmions in monolayer Fe_5GeTe_2 are comparable to those of state-of-the-art transition-metal ultra-thin films. We further predict that these nanoscale skyrmions can be stable for hours at temperatures up to 20 K.

Magnetic skyrmions – localized, stable spin textures with intriguing topological and dynamical properties – have emerged as a promising avenue to realize next-generation spintronics devices [1–6]. During the last ten years, the main focus of the community so far has been on skyrmions in bulk systems [7–9] and interface-based systems of ultrathin films [10–14], and multilayers [15–17]. Recently, magnetic skyrmions were discovered in atomically thin two-dimensional (2D) van der Waals (vdW) materials, providing an ideal playground to push skyrmion technology to the single-layer limit [18, 19]. Stabilizing skyrmions in 2D magnets can avoid pinning by defects due to high-quality vdW interfaces and the possibility of easy control of magnetism via external stimuli.

The Dzyaloshinskii-Moriya interaction (DMI), which prefers a canting of the spins of adjacent magnetic atoms, is often recognized as the key ingredient in forming magnetic skyrmions. The DMI originates from spin-orbit coupling (SOC) and relies on broken inversion symmetry. However, most 2D magnets exhibit inversion symmetry, therefore, the DMI is suppressed. Several strategies have been proposed to achieve DMI by breaking inversion symmetry. These include the family of Janus vdW magnets [20–22], electric field [23], and 2D vdW heterostructures [24–26]. In particular, the Fe_nGeTe_2 family ($n = 3, 4, 5$) with high Curie temperature (T_c) near room temperature has been proposed as a promising candidate for magnetic skyrmions. Néel-type magnetic skyrmions are reported in Fe_3GeTe_2 heterostructures by experiments [24–27] and explained by *ab initio* theory [28, 29] in terms of the emergence of DMI at the interface. All-electrical skyrmion detection has also recently been proposed in tunnel junctions based on Fe_3GeTe_2 [30]. More recently, several experimental groups reported the observation of topological spin structures (i.e., skyrmions or merons) and magnetic bubbles in 2D vdW Fe_5GeTe_2 [31–36]. Additionally, Fe_5GeTe_2 exhibits high T_c above room temperature [37, 38],

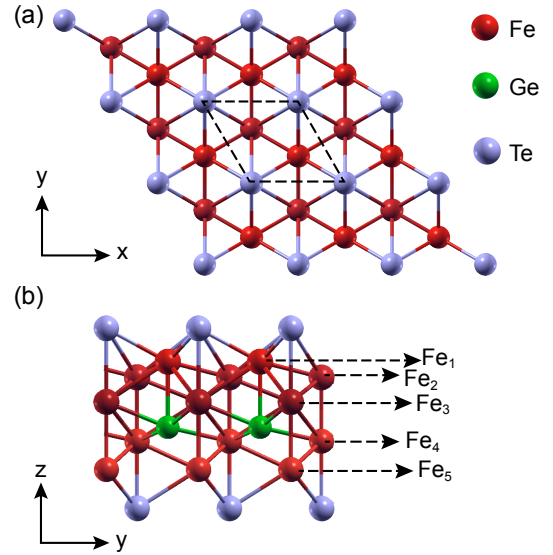


FIG. 1. (a) Top and (b) side views of the atomic structure of the Fe_5GeTe_2 monolayer. The black dashed lines draw up the 2D primitive cell.

which makes it promising for spintronics device applications. However, the quantification of individual skyrmions' stability and lifetime in Fe_5GeTe_2 , crucial for device applications, has been reported neither in experiments nor in theory.

In this Letter, we predict the formation of nanoscale skyrmions in the atom-thick vdW Fe_5GeTe_2 monolayer based on first-principles calculations and atomistic spin simulations. The diameters of these skyrmions are below 10 nm, which is technologically desirable for improving the controllability and integrability of skyrmion-based functional devices. However, such small skyrmions have not yet been observed in 2D vdW magnets. The origin of these nanoscale skyrmions is attributed to strong DMI together with moderate in-plane magnetocrystalline anisotropy energy (MAE) and weak exchange frustration. Furthermore, the calculated energy barriers of skyrmion collapse for Fe_5GeTe_2 monolayer are ~ 80

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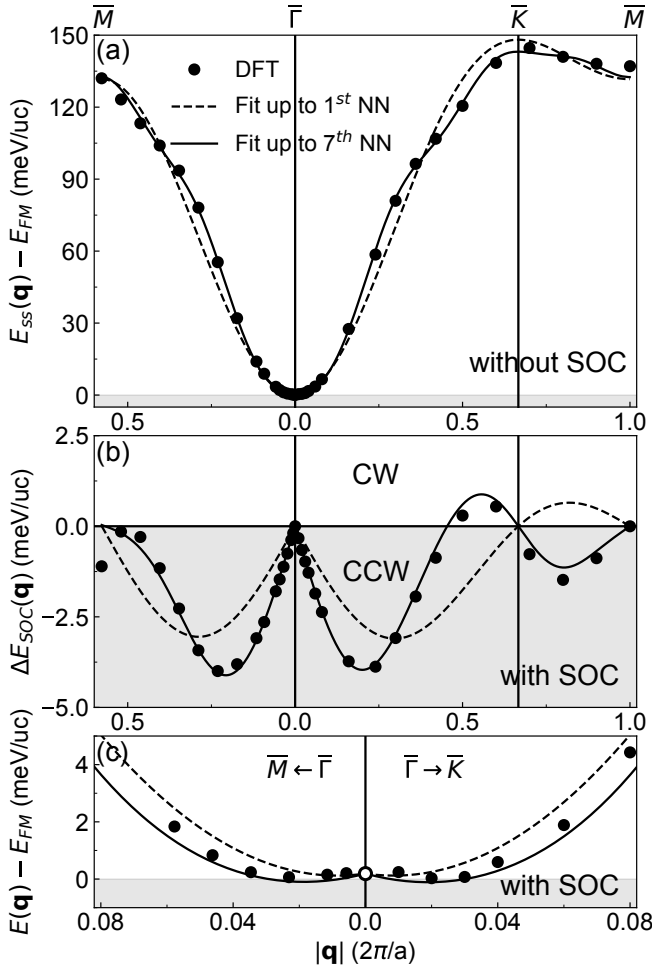


FIG. 2. (a) Energy dispersion of flat spin spirals (E_{SS}) for Fe_5GeTe_2 along the high symmetry path $\bar{M}-\bar{\Gamma}-\bar{K}-\bar{M}$ without SOC. The symbols represent the DFT calculations in scalar-relativistic approximation, while the dashed and solid lines are the fits to the Heisenberg exchange interaction up to the first nearest neighbor (NN) and up to the seventh NN, respectively. (b) Energy contribution of cycloidal spin spirals due to SOC (ΔE_{SOC}), also known as DMI contribution. All energies are measured with respect to the FM state (E_{FM}) at the $\bar{\Gamma}$ point. Note that positive and negative energies represent a preference for clockwise (CW) and counterclockwise (CCW) spin configurations. (c) Zoom around the FM state ($\bar{\Gamma}$ point), including the Heisenberg exchange, the DMI, and the MAE, i.e. $E(\mathbf{q}) = E_{SS}(\mathbf{q}) + \Delta E_{SOC}(\mathbf{q}) + K/2$. The DMI leads to a CCW rotational sense, and the MAE is responsible for the constant energy shift ($K/2$) of the spin spirals with respect to the FM state.

meV at a moderate magnetic field of about 2 T. This substantial energy barrier is comparable to that of ultrathin films [39–41], which serve as prototype systems for hosting nanoscale skyrmions. Finally, we also calculate explicitly skyrmion lifetime as a function of magnetic field and temperature. Our results demonstrate that Fe_5GeTe_2 monolayer is an excellent candidate to experimentally observe nanoscale skyrmions in a 2D vdW magnet.

Our first-principles calculations were performed using the

FLEUR code [42] based on the full-potential linearized augmented plane wave method (see Supplemental Material for computational details [43]). We have calculated the energy dispersions $E(\mathbf{q})$ of flat spin spiral states [44, 45] for the Fe_5GeTe_2 monolayer. A magnetic moment \mathbf{m}_i at atom position \mathbf{R}_i for a flat spin spiral is given by $\mathbf{m}_i = M[\cos(\mathbf{q} \cdot \mathbf{R}_i), \sin(\mathbf{q} \cdot \mathbf{R}_i), 0]$, with M denoting the size of the magnetic moment.

To determine the interactions of magnetic moments for the Fe_5GeTe_2 monolayer, we adopt the following atomistic spin Hamiltonian, which is fitted from spin spiral calculations without and with SOC

$$H = - \sum_{ij} J_{ij}(\mathbf{m}_i \cdot \mathbf{m}_j) - \sum_{ij} \mathbf{D}_{ij} \cdot (\mathbf{m}_i \times \mathbf{m}_j) + \sum_i K(m_i^z)^2 - \sum_i M(\mathbf{m}_i \cdot \mathbf{B}) \quad (1)$$

where \mathbf{m}_i and \mathbf{m}_j are normalized magnetic moments at position \mathbf{R}_i and \mathbf{R}_j respectively. The four magnetic interaction terms correspond to the Heisenberg isotropic exchange, the DMI, the MAE, and the external magnetic field, and they are characterized by the interaction constants J_{ij} , \mathbf{D}_{ij} , and K , and B , respectively. Note that our spin model is adapted to a collective 2D model by treating five Fe layers of Fe_5GeTe_2 as a whole system, similar to a monolayer system. All magnetic interaction parameters are measured in meV/unit cell (uc).

We consider a Fe_5GeTe_2 monolayer where one Fe_1 is situated above Ge as shown in Fig. 1(a), the so-called UUU configuration. The calculated lattice constant is about 3.96 Å, which agrees well with previous results [46]. We are aware that there are two possible configurations for monolayer Fe_5GeTe_2 , namely UUU and UDU configurations [47, 48]. In this work, we focus on the UUU configuration since skyrmionic spin structures are experimentally observed in this configuration [31, 32]. For both spin channels, Fe_5GeTe_2 exhibits a metallic property (See Fig. S1 in Supplemental Material [43]). The calculated spin moments are $-0.21\mu_B$, $2.21\mu_B$, $1.70\mu_B$, $1.28\mu_B$, and $2.41\mu_B$ for the Fe_1 , Fe_2 , Fe_3 , Fe_4 , and Fe_5 atoms, respectively. Note that the calculated spin moments are, in general, in good agreement with previous *ab initio* results, as summarized in detail in Table S2 in Supplemental Material [43]. It is important to note that the spin moment on Fe_1 is significantly quenched, which is also in good agreement with dynamical mean-field (DMFT) results [48].

We focus first on spin spiral calculations without SOC along the $\bar{M}-\bar{\Gamma}-\bar{K}-\bar{M}$ high-symmetry direction (Fig. 2(a)). From the fitted parameters shown in Table I, we find the Heisenberg exchange to be largely dominated by the nearest neighbor contribution. This is also clearly seen from the fitted curve using only the nearest neighbor (NN) term (see dotted line in Fig. 2(a)). On the other hand, exchange constants up to seventh neighbors (Table I) are necessary to fit the DFT results accurately (solid line in Fig. 2(a)). Without SOC, the energy dispersion shows a minimum at the $\bar{\Gamma}$ point, which represents the ferromagnetic (FM) state. The energy difference between the $\bar{\Gamma}$ and the \bar{M} point (row-wise antiferromagnetic (AFM) state) is found to be about 137 meV for five Fe atoms. This

	J_1/J_{eff}	J_2	J_3	J_4	J_5	J_6	J_7	D_1/D_{eff}	D_2	D_3	D_4	D_5	D_6	D_7	K	m_s
Full model	14.803	0.898	0.466	0.916	0.231	-0.443	-0.599	-0.659	-0.523	-0.083	0.020	-0.008	-0.021	-0.008	0.380	7.306
Effective model	16.457	-	-	-	-	-	-	-0.880	-	-	-	-	-	-	0.380	7.306

TABLE I. Full model: Shell-resolved Heisenberg exchange constants (J_i) and DMI constants (D_i) obtained by fitting the energy contribution to spin spirals without and with SOC, and spin moments (m_s) from DFT calculations as presented in Fig. 2 for the Fe_5GeTe_2 monolayer. Effective model: Parameters of the effective NN exchange and DMI model obtained by fitting the DFT results. A positive (negative) sign represents FM (AFM) coupling. A positive (negative) sign of D_i denotes a preference for CW (CCW) rotating cycloidal spin spirals. The Fe_5GeTe_2 monolayer favors in-plane MAE (i.e., $K > 0$). Note that all parameters are treated as a collective 2D spin model in a hexagonal symmetry (i.e., five Fe atoms in the supercell are treated as a whole). The magnetic moments are given in $\mu_B/\text{unit cell}$ (uc), and other parameters in meV/uc.

energy difference is much smaller than that in the Fe_3GeTe_2 or Fe_4GeTe_2 monolayers (see Fig. S2 in Supplemental Material [43]), leading to much smaller J_1 values (see Table I in Supplemental Material [43]). We also note that the spin moment variation is mainly from Fe_1 and Fe_4 atoms (Fig. S3a in Supplemental Material [43]). Additionally, we have also carefully checked the effect of variation of spin moments for our spin model by calculating conical spin spirals (see Section II, Fig. S3b, and Fig. S4 in Supplemental Material [43].)

When SOC is taken into account, the DMI arises due to broken inversion symmetry. The Fe_5GeTe_2 monolayer favors cycloidal spin spirals with a counter-clockwise (CCW) rotational sense, as seen from the calculated energy contribution to the dispersion due to SOC, $\Delta E_{\text{SOC}}(\mathbf{q})$ (Fig. 2(b)). If we apply the NN approximation, we obtain the effective DMI constant $D_{\text{eff}} = -0.88$ meV, and the corresponding micro-magnetic DMI is given by $D = \frac{3\sqrt{2}D_{\text{eff}}}{N_F a^2}$ (a and N_F are the lattice constant and the number of ferromagnetic layers). For the Fe_5GeTe_2 monolayer, the value is approximately 0.76 mJ/m², which is in reasonable agreement with the one calculated by the supercell approach (~ 0.48 – 0.67 mJ/m²) [31] based on the NN approximation. However, as clearly seen in Fig. 2(b), the dashed lines with an effective NN DMI fail to capture our DFT results beyond the regime of small $|\mathbf{q}|$. The inclusion of interactions up to the seventh NN is needed to reproduce the DFT data accurately, and this is also reflected in the fitted parameters presented in Table I. The second NN interaction is on the same order of magnitude compared to the first NN one.

Additionally, SOC also introduces MAE. We find the easy magnetization axis of the Fe_5GeTe_2 monolayer to be in-plane with a MAE of 0.38 meV/uc. As observed in Fig. 2(c), a spin spiral energy minimum of -0.1 meV/uc compared to the FM state occurs in the $\bar{\Gamma}\bar{K}$ direction, which corresponds to a spin spiral period of $\lambda = 2\pi/|\mathbf{q}| = 19.9$ nm.

Including all interactions, i.e. Heisenberg exchange, DMI, and MAE, results in the spin spiral energy dispersion shown in Fig. 2(c). The energy contribution from the MAE leads to an energy offset of $K/2$ for spin spirals with a long period, i.e. small value of $|\mathbf{q}|$, with respect to the FM state, as can be seen in a zoom of $E(\mathbf{q})$ around $\bar{\Gamma}$. Interestingly, the ground state within the NN approximation is the FM state, while it is the spin spiral state when we include interactions up to the seventh NN. It is worth emphasizing that the spin spiral curve becomes extremely flat near the $\bar{\Gamma}$ point. It has been demonstrated that a flat energy dispersion around $\bar{\Gamma}$ is beneficial for stabilizing nanoscale skyrmions in ultrathin films [14]. To

check the validity of our 2D collective spin model, we performed Monte-Carlo simulations (see section III in Supplemental Material [43]) with magnetic interaction parameters from the full model presented in Table I. We obtain the Curie temperature (T_c) of about 451 K (see Fig. S5 in Supplemental Material [43]), which is in reasonable agreement with the value of $T_c = 390$ K obtained in Ref. [47].

To check the possibility of stabilizing nanoscale magnetic skyrmions in Fe_5GeTe_2 , we performed atomistic spin simulations using the spin model described by Eq. (1) with the full set of DFT parameters. We apply atomistic spin-dynamics via the Landau–Lifshitz equation to obtain isolated magnetic skyrmions. Minimum energy paths between the initial skyrmion and final FM state were calculated using the geodesic nudged elastic band (GNEB) method [49]. The energy barriers stabilizing skyrmions against collapse were obtained from the saddle point along this path. Finally, we used harmonic transition-state theory to quantify skyrmion stability by calculating their lifetime [40, 50–52] (see Supplemental Material for computational details [43]).

We create isolated skyrmions in the field-polarized background with an out-of-plane magnetization direction due to an applied magnetic field and fully relax these spin structures by solving the damped Landau–Lifshitz equation self-consistently. The in-plane FM state (denoted as FM_{\parallel}) exhibits slightly lower energy compared to the out-of-plane FM state, owing to the in-plane MAE of Fe_5GeTe_2 (see Fig. S6 in Supplemental Material [43]). At zero magnetic field, we do not observe the emergence of skyrmions. Instead, we observe labyrinth domains with chiral Néel domain walls, which hold the lowest energy state. As we gradually increase the magnetic field B perpendicular to the monolayer, the labyrinthine domain shrinks and eventually vanishes, giving rise to isolated magnetic skyrmions at a critical field of $B_c = 2.2$ T. Note, that for fields above $B = 2K/M \approx 1.8$ T applied perpendicular to the film the Zeeman energy exceeds the MAE [53].

Our atomistic spin simulations predict Néel-type magnetic skyrmions stabilized in the FM background (see inset of Fig. 3(a) for skyrmion profile and spin texture at $B = 2.2$ T). As expected, the skyrmion size decreases in Fe_5GeTe_2 with increasing magnetic field (Fig. 3(a)). Interestingly, nanoscale skyrmions occur with a radius below 6.4 nm at $B > 2.2$ T, and isolated skyrmions can be obtained. Note that the skyrmion radius is estimated using the Bocdanov- Θ profile [54] within a wide range of $B = 2.2 \sim 7.6$ T. At $B > 7.6$ T, skyrmions collapse into the FM state.

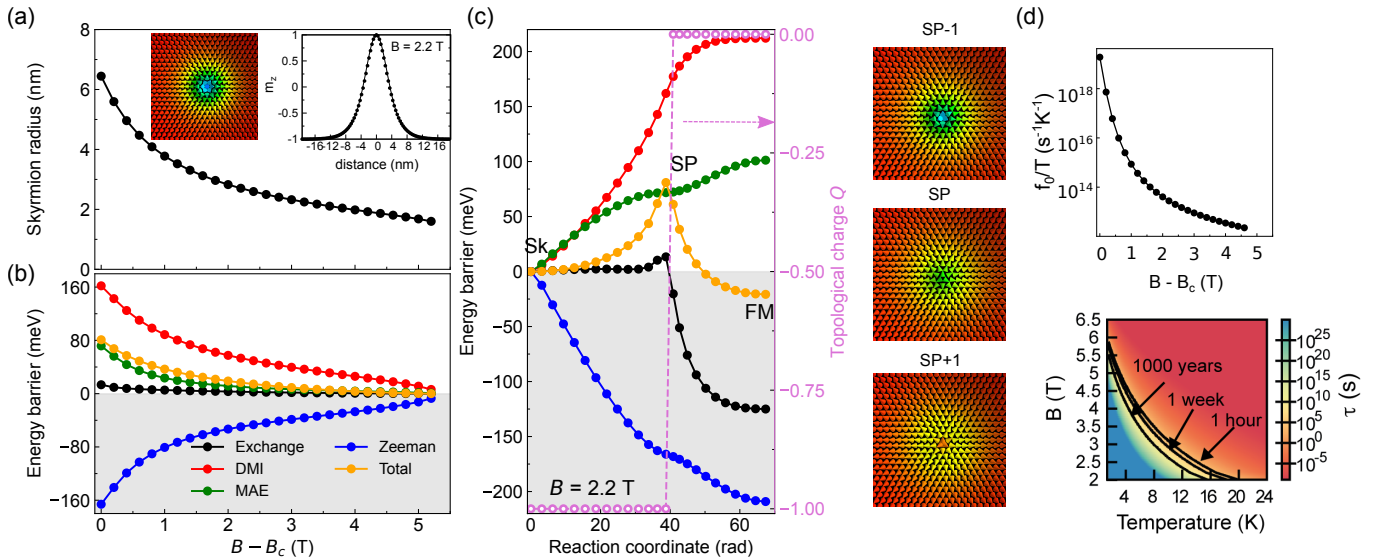


FIG. 3. Skymion radius and energy barrier for skymion collapse in the Fe_5GeTe_2 monolayer evaluated using magnetic interaction parameters from DFT. (a) Skymion radius as a function of $B - B_c$ where B_c is the critical field for stabilizing metastable magnetic skymions. The calculated value is $B_c = 2.2$ T for Fe_5GeTe_2 . The skymion profile and spin texture at $B = 2.2$ T are shown as insets. (b) Total energy barriers and their decomposition (see legend) of isolated skymions versus $B - B_c$. (c) Energy contributions from the different interactions (the same legend as in panel (b), left axis) are shown versus the reaction coordinate along the minimum energy path from the initial (skymion) state to the final (FM) state through the saddle point (SP). The energies are summed over all atoms of the simulation box and are given relative to the energies of the initial isolated skymion state. The topological charge (open circles, right axis) is plotted versus the reaction coordinate. (c) Corresponding spin structures before (SP-1), after (SP+1), and at the saddle point (SP) are shown. Note that the skymion collapse occurs via radially symmetrical shrinking. (d) Calculated attempt frequencies f_0 on a logarithmic scale with respect to $B - B_c$ (top panel). Skymion lifetimes of Fe_5GeTe_2 obtained in harmonic transition state theory based on the spin model with DFT parameters as a function of magnetic field and temperature (bottom panel).

To provide insight into skymion stability in monolayer Fe_5GeTe_2 , we show in Fig. 3(b) the calculated energy barriers protecting skymions from collapsing into the FM state with respect to $B - B_c$. Remarkably, we obtain an energy barrier of more than 80 meV at the critical field $B_c = 2.2$ T. This value is comparable to state-of-the-art ultra-thin films that serve as prototype systems to host nanoscale skymions [39–41]. Note, that previous work on skymion stability in 2D magnets [28] reported smaller values than those found here for the Fe_5GeTe_2 monolayer.

From the energy decomposition of the barrier, we conclude that the DMI and MAE are mainly responsible for the skymion stability (Fig. 3(b)). We also note that the energy barrier can be enhanced if we increase the in-plane MAE, a quantity that can be easily tuned in experiments by doping or temperature [55–57]. However, if the in-plane MAE is increased larger magnetic fields are needed to obtain the field-polarized phase in which skymions can be stabilized.

To obtain information about transition mechanisms from the skymion state to the FM state via the saddle point (SP), which determines the barrier, we show in Fig. 3(c) the decomposition of the energy along the minimum energy path for skymion collapse at $B = 2.2$ T. The topological charge, calculated by $Q = \int \mathbf{m} \cdot (\frac{\partial \mathbf{m}}{\partial x} \times \frac{\partial \mathbf{m}}{\partial y}) dx dy$, changes from -1 to 0 at the SP. The skymion is annihilated via the radial symmetric collapse mechanism in which the skymion shrinks symmetrically to SP and then collapses into the FM state [52]. Again, it

is clear that the DMI and MAE prefer the skymion (Sk) state and decrease the total barrier, while the Zeeman term strongly favors the FM state. Due to frustration, the Heisenberg exchange energy gives a small positive annihilation barrier.

The stability of metastable magnetic skymions can be quantified by their mean lifetime, τ , which is given by the Arrhenius law $\tau = f_0^{-1} \exp(\frac{\Delta E}{k_B T})$, where ΔE , f_0 , and T are energy barrier, attempt frequency, and temperature, respectively. The calculated f_0 within harmonic transition state theory [50] is shown in Fig. 3(d). As expected, f_0 depends strongly on the magnetic field. This effect is similar to that observed in ultrathin transition-metal films, which can be traced back to a change of entropy with skymion radius and profile [51, 58]. From the temperature and field dependence of the skymion lifetime (Fig. 3(d)), we predict that isolated skymions in the Fe_5GeTe_2 are stable up to hours at a temperature at about 20 K and $B = 2.2$ T. Therefore, these nanoscale skymions can be probed by experiments using current state-of-the-art techniques e.g., spin-polarized scanning tunneling microscopy or Lorentz transmission electron microscopy.

We plot in Fig. 4 the comparison of atomistic spin simulation results from parameters with the full and effective models. The critical magnetic field is found to be about 1.4 T in the case of the effective model, which is 0.8 T smaller than the one for the full model. In both models, we have sub-10 nm radius skymions (Fig. 4(a)) and energy barriers of a few 10 meV

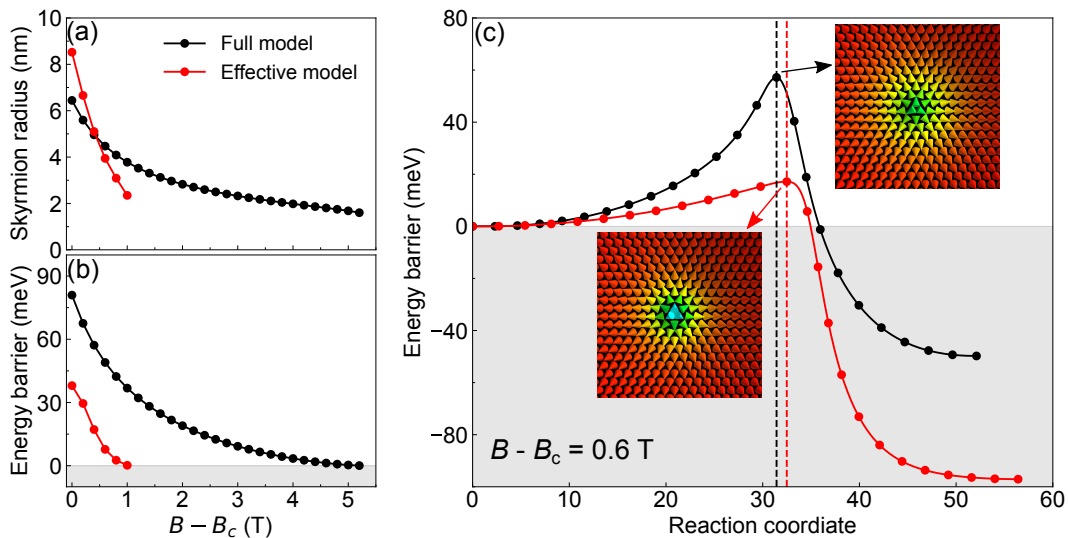


FIG. 4. Full model versus effective model comparison. (a) Skyrmion radius vs. magnetic field. (b) Skyrmion energy barriers with respect to B . The estimated critical magnetic fields B_c are 2.2 T and 1.4 T for the full model and effective model, respectively. (c) Minimum energy paths of skyrmion collapse: Energies of the spin configurations during a skyrmion collapse are shown over the reaction coordinate corresponding to the progress of the collapse at $B - B_c = 0.6$ T. Our simulations were conducted using both the effective exchange model (in red) and considering all magnetic interactions from the DFT calculation (in black).

(Fig. 4(b)). From the GNEB calculation at $B - B_c = 0.6$ T, the energy barriers for the full and effective models are rather different. We find more than 3 times larger energy barrier with the full model, indicating the importance of including full magnetic parameters for the Fe_5GeTe_2 monolayer.

In summary, we propose, based on first-principles calculations and atomistic spin simulations, that monolayer Fe_5GeTe_2 is a compelling 2D vdW magnet with skyrmionic physics. Due to the large DMI together with moderate in-plane MAE, Fe_5GeTe_2 monolayer can exhibit metastable nanoscale (sub-10 nm) skyrmions in an out-of-plane magnetic field above 2.2 T. We also highlight the importance of including beyond nearest neighbor DMI terms in the atomistic spin model. The energy barriers protecting skyrmions against collapse are up to 80 meV, which are comparable to those of state-of-the-art transition-metal ultrathin films. Using harmonic transition-state theory, we predict that nanoscale skyrmions are stable in monolayer Fe_5GeTe_2 with lifetimes of hours up to 20 K.

Acknowledgments

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