Spin-Topology Dependent Brownian Diffusion of Skyrmions

Le Zhao¹*, Zidong Wang¹*, Xichao Zhang²*, Jing Xia², Keyu Wu¹, Heng-An Zhou¹, Yiqing Dong¹, Guoqiang Yu³, Kang L. Wang⁴, Xiaoxi Liu⁵, Yan Zhou²,† and Wanjun Jiang¹,†

¹State Key Laboratory of Low-Dimensional Quantum Physics and Department of Physics, Tsinghua University, Beijing 100084, China
²School of Science and Engineering, The Chinese University of Hong Kong, Shenzhen, Guangdong 518172, China
³Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China
⁴Department of Electrical Engineering, University of California, Los Angeles, California, 90095, USA
⁵Department of Electrical and Computer Engineering, Shinshu University, 4-17-1 Wakasato, Nagano 380-8553, Japan

* These authors contributed equally to this work.
† To whom correspondence should be addressed:
zhouyan@cuhk.edu.cn and jiang_lab@tsinghua.edu.cn
Abstract
Non-interacting particles exhibiting Brownian motion have been observed in many occasions of sciences, such as molecules suspended in liquids, optically trapped microbeads, and spin textures in magnetic materials. In particular, a detailed examination of Brownian motion of spin textures is important for designing thermally stable spintronic devices which motivates the present study. In this Letter, through using temporally and spatially resolved polar magneto-optic Kerr effect (MOKE) microscopy, we have experimentally observed the thermal fluctuation-induced random walk of a single isolated Néel-type magnetic skyrmion around room temperature in an interfacially asymmetric Ta/CoFeB/TaOx multilayer. More importantly, an intriguing spin-topology dependent diffusion behavior of skyrmions has been identified. The onset of Brownian dynamics of a single skyrmion induced by the random thermal noises, including a linearly temperature dependent diffusion coefficient and spin-topology dependent diffusion were further formulated based on the stochastic Thiele equation. The numerical and experimental demonstration of spin-topology dependent Brownian dynamics of skyrmions can be useful for understanding the nonequilibrium magnetization dynamics and implementing novel skyrmionic devices.

PACS: 75.60.Ch, 75.70.Kw, 75.78.-n, 12.39.Dc
Keywords: skyrmion, Brownian motion, spintronics, micromagnetics
Magnetic skyrmions are particle-like spin textures favored by a nonlinear Dzyaloshinskii-Moriya (DM) interaction \(^1\textsuperscript{-9}\), which have recently stimulated great interests in spintronics community \(^10\textsuperscript{-18}\). Specifically, the topological properties of skyrmions are governed by the skyrmion number \(Q = 1/4\pi \int \mathbf{m} \cdot (\partial \mathbf{m}/\partial x \times \partial \mathbf{m}/\partial y) \, dx \, dy\) with \(\mathbf{m} = \mathbf{M}/M_S\) being the reduced magnetization vector and \(M_S\) the saturation magnetization, respectively. This quantity is responsible for topological transport phenomena such as topological Hall effect \(^19,20\) and skyrmion Hall effect \(^21\textsuperscript{-23}\), etc. Despite many progresses being made, an experimental demonstration of Brownian dynamics of skyrmions and their connection with the accompanied spin topology in particular \(^24\textsuperscript{-28}\), are not fully addressed. These important aspects could facilitate an accurate understanding of the nonequilibrium magneto-thermodynamics, since the dynamics of skyrmions can be temporally and spatially captured \(^6,12,29\). Further, a comprehension of Brownian dynamics and their responses to temperatures could set up a vital step towards thermally stable skyrmion devices \(^30\textsuperscript{-32}\), and implementation of skyrmions for novel applications \(^33\textsuperscript{-35}\).

**Micromagnetic simulations of Brownian motion of a single skyrmion.** Brownian motion is a nonequilibrium thermodynamic phenomenon that is typified by the random walk of objects, as a result of thermal fluctuations \(^36\textsuperscript{-38}\). In magnetic materials, magnetization dynamics can also be strongly influenced by the random thermal fluctuations, especially when the magnetic anisotropy energy is comparable with thermal excitations \(^39\textsuperscript{-42}\). Following a Gaussian stochastic process, the irregular thermal fluctuating field \(\mathbf{h}\) can be treated as \(\langle h_i(x, t) \rangle = 0\), \(\langle h_i(x, t)h_j(x', t') \rangle = \eta \delta_{ij} \delta(x-x') \delta(t-t')\). Here \(i\) and \(j\) are Cartesian components, \(\eta = 2\alpha k_B T/\gamma_0\mu_0 M_S\) is the variance of the effective force produced by thermal fluctuation, \(T\) is the temperature, \(k_B\) is the Boltzmann constant, \(\gamma_0\) is the absolute gyromagnetic ratio, \(\mu_0\) is the vacuum permeability, \(M_S\) is the saturation magnetization, \(\alpha\) is the Gilbert damping coefficient and \(\delta_{ij}\) and \(\delta(\ldots)\) stand for Kronecker and Dirac delta symbols, respectively \(^26,28\). This extra term results in the establishment of stochastic Landau-Lifshitz-Gilbert (LLG) equation, following which the random walk of skyrmions can be numerically simulated. The detailed description of stochastic LLG equation and micromagnetic simulation are available in the Supplementary Materials.

The micromagnetic simulation results of a single isolated Néel-type skyrmion at various temperatures are shown in Fig. 1, which clearly reveals the random-walk feature from the time-dependent trajectory. The amplitude of trajectories in the 2D \(x - y\) space increases following the increase of temperatures, suggesting an enhanced thermally induced diffusion, as shown in
Fig. 1(a). These features are phenomenologically consistent with prediction of Brownian motion. In a non-confined 2D geometry, the probability distribution \( P(x, y, t) \) of a free Brownian particle at position \((x, y)\) and at time \(t\) is described by a Gaussian function

\[
P(x, y, t) = \frac{1}{\sqrt{4\pi\mathcal{D}_{dc}t}} \cdot \exp\left[-(x^2 + y^2)/4\mathcal{D}_{dc}t\right],
\]

where \(\mathcal{D}_{dc}\) is the diffusion coefficient. The diffusion coefficient \(\mathcal{D}_{dc}\) can be obtained by calculating the mean-squared displacement (MSD) with the relation among the MSD, \(\mathcal{D}_{dc}\), and \(t^*\) being given as:

\[
\langle [r_{x,y}(t + t^*) - r_{x,y}(t)]^2 \rangle = \langle (\Delta r_{x,y})^2 \rangle = 4\mathcal{D}_{dc} t^*
\]

where \(r_{x,y}\) is the position of the skyrmion center in Cartesian coordinates \((x, y)\), \(t^*\) is the time interval between two selected data points.

Eq. (1) suggests that the MSD of a Brownian particle should be linearly proportional to the elapsed time \(t\), which agrees well with our simulation results, as shown in Fig. 1(b). Note that following the increase of temperature, the size of the skyrmion increases accordingly in our simulation. This leads to an enhanced skyrmion-edge interaction and skyrmion dissipation in a finite-size sample adopted in our simulation, which are presumably responsible for the nonlinear temperature dependence of diffusion coefficient \(\mathcal{D}_{dc}\), as shown in Fig. 1(c). We therefore limit our discussion to a relatively low temperature range of \(T = 0 \sim 120\) K. Note that the out-of-plane magnetic fields were applied in both simulations \((B_z = 1 \sim 4\) mT) and experiments \((B_z = \sim2\) mT) to maintain a fixed skyrmion size, as well as a single skyrmion state. Note that we utilized a constant value of \(M_S\) in simulation as its change in the experimentally investigated temperature range is negligible.

**A theoretical approach based on the stochastic Thiele equation.** The quasiparticle nature of skyrmion enables its thermal diffusion to be approximated by considering the motion of its center of mass, in which no distortion is considered. This assumption results in the stochastic LLG equation to be simplified as the stochastic Thiele equation. Subsequently, the diffusion dynamics of a single skyrmion and more importantly, its connection with spin topology will be established. Note that the stochastic Thiele equation also shares a similar form with the Langevin equation, the latter is often utilized to study conventional Brownian particles. The stochastic Thiele equation in the absence of pinning effect reads as:

\[
-\mathcal{G} \hat{z} \times \mathbf{v} - \alpha \mathbf{D} \cdot \mathbf{v} = \eta
\]

where \(\mathcal{G} = 4\pi Q\mu_0 M_S d/\gamma_0\) is the gyrocoupling vector that is related to the spin topology \((Q)\). \(\mathbf{D}\) is the dissipative tensor that depends on the skyrmion profile. Specifically, \(\mathbf{D} = (\mu_0 M_S d/\gamma_0) \cdot (\pi^2 r_{sk}/\Delta)\) can be computed by assuming a typical skyrmion profile, where \(r_{sk}\) is the skyrmion radius, sample thickness and domain wall width,
respectively, $A$ and $K_{\text{eff}}$ are the spin-wave stiffness and effective magnetic anisotropy constants, respectively. $\mathbf{v}$ is the diffusion velocity, and $\mathbf{\eta} = (\eta_x, \eta_y)$ is the variance of effective force produced by the stochastic thermal fluctuation field $\mathbf{h}(x,t)$, where $\eta_x = \mu_0 M_s \int \frac{\partial m}{\partial x} \cdot \mathbf{h}(x,t) d^2 x$ and $\eta_y = \mu_0 M_s \int \frac{\partial m}{\partial y} \cdot \mathbf{h}(x,t) d^2 x$, respectively. The stochastic force $\mathbf{\eta}$ satisfies a Gaussian stochastic process $\langle \eta_x(t)\eta_x(t') \rangle = \langle \eta_y(t)\eta_y(t') \rangle = 2ak_B T\Delta \delta(t-t')$. The first term on the left side of Eq. (2) correlates with the spin topology ($\mathcal{G}$) and diffusion velocity ($\mathbf{v}$), the second term is related to the dissipation, and the term on the right side represents the driving force provided by the random thermal fluctuations. Solving Eq. (2), the diffusion velocity can be obtained as follows:

$$v_x = -\frac{\alpha D\eta_x + \mathcal{G}\eta_y}{g^2 + (\alpha D)^2}, \quad v_y = -\frac{\alpha D\eta_y - \mathcal{G}\eta_x}{g^2 + (\alpha D)^2} \quad (3)$$

By calculating the velocity-velocity autocorrelation functions $\langle v_{x,y}(t)v_{x,y}(t') \rangle$ and their time integrations, the MSD of an isolated skyrmion in free Brownian motion regime can be written as $\langle x^2(t) \rangle = \langle y^2(t) \rangle = \frac{2ak_B T\Delta}{g^2 + (\alpha D)^2} t$. Using the definition of MSD given in Eq. (1) –

$$\langle (\Delta r_{x,y})^2 \rangle = 4 \mathcal{D}_{dc} t, \text{ the temperature dependent } \mathcal{D}_{dc} \text{ of skyrmion is further established as:}$$

$$\mathcal{D}_{dc} = k_B T \frac{\alpha D}{g^2 + (\alpha D)^2} \quad (4)$$

Eq. (4) thus predicts a linear dependence of $\mathcal{D}_{dc}$ vs. $T$ for the fixed values of $\alpha, \mathcal{G}$ and $\mathcal{D}$. This linear dependence is acquired assumes a temperature-independent rigid skyrmion profile (i.e., $r_{sk}$ and $\Delta$ are constant as a function of temperature), which could explain the discrepancy between theoretical model and simulation in a wider temperature range ($r_{sk}$ and $\Delta$ vary as a function of temperatures). It is clear from Eq. (4) that the diffusion behaviors are different for skyrmion $Q = \pm 1, (\mathcal{G}^2 \propto (\pm Q)^2 = 1)$ and topologically trivial bubbles $Q = 0 (\mathcal{G}^2 = 0)$. Namely, $\mathcal{D}_{dc} = k_B T / \alpha D$ is found for $Q = 0$ trivial bubble, based on which one could determine $\alpha, D$ by measuring $\mathcal{D}_{dc}$ vs. $T$. This aspect has been studied in Garnets in which DMI and uniform topology are not relevant.41

Values of $\mathcal{D}_{dc}$ are however, indistinguishable for $Q = +1$ and $Q = -1$ skyrmions. Note that a $Q = -1$ skyrmion is defined as the center of skyrmion exhibiting a negative orientation ($m_z < 0$), and the positive orientation of the center corresponds to $Q = +1 (m_z > 0)$ skyrmions, respectively. For a fixed skyrmion profile and hence a fixed dissipative tensor $\mathcal{D}$, the dependence of $\mathcal{D}_{dc} - \alpha$ based on Eq. (4) is further plotted in Fig. 1(d) (red curve). The overlap between the simulation results (black square dots) and the stochastic Thiele equation
demonstrate the physics of skyrmions as Brownian particles is captured, which is also consistent with a recent numerical study \(^{28}\).

**Spin-topology dependent skyrmion diffusion.** After examining Eq. (3) closely, it is found that the diffusion velocity \(v\) contains a linear \(\mathcal{G}\) term that is depending on the spin topology of skyrmions. Below we will reveal theoretically the spin-topology dependent Brownian diffusion dynamics. By defining a diffusion skyrmion Hall angle, it is found that:

\[
\tan \theta_{sk} = \frac{v_y}{v_x} = \frac{aD\eta_y - b\eta_x}{aD\eta_x + b\eta_y}
\]

(5)

This equation simplifies the theoretical discussion of the spin topology dependent Brownian diffusion, and suggests that \(\theta_{sk}\) can be used for identifying spin textures with varying spin topological numbers, at least as a theoretical possibility. For topological trivial bubbles (\(Q = \mathcal{G} = 0\)), \(\tan \theta_{sk} = \eta_y / \eta_x\) thus measures the time dependent directional variation of Gaussian fluctuation. For \(Q = +1\) and \(Q = -1\) skyrmions, the different values of \(\tan \theta_{sk}\) reflect the opposite spin topology, as shown in Fig. 1(e). Note that skyrmions with opposite topological number \(Q\) can be synthetized by reversing the polarity of perpendicular magnetic fields that have been previously used to reveal the skyrmion Hall effect \(^{22,46}\). The above theoretical discussion on topologically dependent diffusion will be tested experimentally.

**Experimental revelation of skyrmion Brownian motion.** The experimental demonstration of Brownian motion of a single isolated Néel-type skyrmion is provided by using a polar magneto-optic Kerr effect (MOKE) microscope around room temperature in an interfacially asymmetric Ta(5nm)/CoFeB(1nm)/TaO\(_x\)(3nm) multilayer. The film exhibits a relatively weak perpendicular magnetic anisotropy (\(K_{eff} = 30\) mT), and a temperature dependent spin reorientation transition that is accompanied with stripe to skyrmion morphological transition \(^{47,48}\). Note that thermal fluctuations in other skyrmion materials have been previously identified \(^{48,49}\), the onset Brownian dynamics in real space is however, not discussed. Due to the presence of broken interfacial inversion symmetry and strong spin-orbit interaction, an interfacial DM contribution is introduced which stabilizes Néel-type skyrmions with a left handedness in our multilayer \(^{12}\). This is the key difference with magnetic bubbles stabilized by the dipolar interaction in Garnets \(^{50}\). To study free Brownian motion, we have stabilized a single isolated skyrmion of radius \(r_{sk} \sim 1.2\) µm in a sample of size 25.5 µm (width) \(\times 74\) µm (length). Under this condition, the complication due to the repulsive skyrmion-skyrmion interaction is eliminated. The radius of the experimentally stabilized skyrmion is much larger than that of the simulated one, as a result of the strong dipole-dipole interaction \(^{51}\). The larger size of skyrmion introduces enhanced dissipation as well as skyrmion defects.
interaction. Skyrmions with opposite topological number \(Q = \pm 1\) were stabilized by reversing the polarity of fields from positive \(B_z = +2.1\) mT to negative \(B_z = -2.1\) mT\(^{22,46}\).

The acquired time-dependent random-walk trajectory of a single skyrmion \(Q = -1\) at \(T = 293.15\) K and positive field \(B_z = +2.1\) mT is shown in Fig. 2(a). The random-walk trajectory reflects the complex energy landscape settled by the randomly distributed material defects of the system\(^{52}\), in which the skyrmion constantly picks up kinetic energy from thermal fluctuation that results in escaping from the local energy minima. Time interval \(\Delta t\) is the gap between two consecutive frames that is limited by the temporal resolution of MOKE microscope. Shown on the right side of Fig. 2(a)1- (a)5 are the selected snapshots of MOKE images taken during the diffusion of skyrmion. By plotting the trajectory in a 2D plane, namely, coordinates of the skyrmion center \(R(t) = [X(t), Y(t)]\) at time \(t\), we have identified two interesting features. First, the probability of the skyrmion appearing at particular locations is larger than at other regions that can be presumably attributed to the deeper pinning potentials created by random distributed defects. Second, an anisotropic diffusion dynamics are also present which we tentatively attribute to the skyrmion-edge interaction, a point will be returned later.

In order to understand the dynamical feature of the skyrmion diffusion, we define a transient velocity \(\mathbf{v}(t) = [v_x(t), v_y(t)]\) with \(v_x(t) = [X(t + \Delta t) - X(t)]/\Delta t\) and \(v_y(t) = [Y(t + \Delta t) - Y(t)]/\Delta t\), where \(\Delta t\) being the time interval between two consecutive images, \(t^* = N \cdot \Delta t\) where \(N\) is an integer number. Note that the transient velocity should not be treated as “instantaneous” velocity of a Brownian particle since the intrinsic magnetization dynamics is typically at \(10^{-9}\) s time scale\(^{53}\). Shown in Fig. 2 (b) are the experimentally determined transient velocity components along \(x\) (left) and \(y\) (right) directions. While velocities fluctuate as a function of time, they reside around zero. This can be understood as follows, while the force provided by random thermal fluctuation is dynamically varying as a function of time, which, on average is zero - \(\langle h_{t}(\mathbf{x}, t)\rangle = 0\). The distribution of the transient velocities follows a Gaussian distribution, as shown in Fig. 2 (c). This is enabled by studying the 1-D probability density function (PDF) which is defined as PDF = \(1/\sigma \sqrt{2\pi} \cdot \exp[-(x - \mu)^2/2\sigma^2]\), where \(\mu\) and \(\sigma\) are the mean and standard deviation of the measured transient velocities, respectively\(^{36,38}\). While both the magnitude and direction of the transient velocities along \(x\) (left) and \(y\) (right) directions vary as a function of time, the PDFs of which can be fitted well by a standard Gaussian distribution, as shown in Fig. 2 (c).
On the other hand, it is also noted that the full width at half maximum (FWHM) of both PDFs are different for $Q = -1$ skyrmion, namely, $v_{x,\text{FWHM}} = 20.6\, \mu\text{m/s}$, $v_{y,\text{FWHM}} = 17.5\, \mu\text{m/s}$, with an accompanied diffusion skyrmion Hall angle $\theta_{sk} = 40.2^\circ$. Such an anisotropic diffusion behaviour can be attributed to the skyrmion-edge interaction in the confined geometry, together with the spin-topology dependent skyrmion diffusion, as suggested by Eq. (5). In a larger sample in which the effect of geometry confinement (120 µm × 100 µm) is less pronounced, for $Q = -1$ skyrmion, an anisotropic diffusion behaviour is still present with $\bar{v}_{x,\text{FWHM}} = 35.8\, \mu\text{m/s}$, $\bar{v}_{y,\text{FWHM}} = 31.0\, \mu\text{m/s}$, and $\theta_{sk} = 41.0^\circ$. Thus, while geometrical confinement contributes to the anisotropic diffusion, the spin-topology of skyrmion also plays an important role. By reversing the positive field ($B_z = +2.1$ mT) to negative field ($B_z = -2.1$ mT), the diffusion dynamics of skyrmions with an opposite skyrmion number $Q = +1$ is studied. It is found that $\bar{v}_{x,\text{FWHM}} = 31.3\, \mu\text{m/s}$, $\bar{v}_{y,\text{FWHM}} = 36.3\, \mu\text{m/s}$ with $\theta_{sk} = 49.2^\circ$. The different values of $\theta_{sk}$ acquired for skyrmions with opposite skyrmion numbers ($Q = \pm1$) thus indicate the spin-topology dependent skyrmion diffusion. Note that the values of $\bar{v}_{x,\text{FWHM}}$ and $\bar{v}_{y,\text{FWHM}}$ from PDF were averaged for 3 different ($Q = \pm1$) skyrmions to minimize the statistical error. Raw data can be found in Part 4 and Part 5 of the Supplementary Materials.

**The temperature dependent Brownian motion of a single skyrmion.** The temperature dependence of Brownian motion is shown in Fig. 3. It can be seen that when $T = 282.05$ K, the skyrmion is pinned at a certain location. The reason is that the amplitude of the thermal fluctuation is not large enough to excite skyrmion to escape from the local pinning potential. When $T \geq 284.13$ K and increases further, the skyrmion collects kinetic energy from the thermal fluctuation and becomes to be thermally active, which results in the increasing amplitudes of the Brownian motion. This behavior is consistent with the simulation results given in Fig. 1(a). Note that when the temperature is larger than a certain threshold ($i.e., T > 298$ K), static skyrmion phase is unfavorable and accompanied with creation and annihilation of skyrmions by thermal fluctuations, which agrees with recent studies.²⁸

Based on the temperature-dependent skyrmion trajectories, the corresponding MSDs and the evolution of diffusion coefficients can be acquired, as shown in Fig. 4. It shows that the MSDs are almost linearly proportional to time $t^*$ at relatively low temperatures. However, since the motion of the skyrmion is strongly influenced by pinning, especially at lower temperatures ($e.g., T = 282.05$ K), the movements by any mechanism other than thermal diffusion will be reflected to the MSD and naturally break the linear correlation. Based on the
linear regime of MSD vs. $t^*$ (the first 15% of the result) \cite{54}, $\mathcal{D}_{dc}$ at various temperatures for skyrmions with $Q = \pm 1$ can be calculated, as shown in Fig. 4(b). It can be seen that the evolution of $\mathcal{D}_{dc} - T$ are largely overlapping for $Q = \pm 1$ skyrmion, as suggested by Eq. (4). In contrast to the exponential dependence of $\mathcal{D}_{dc} - T$ relation from micromagnetic simulation ($T < 120$ K), a linear $\mathcal{D}_{dc} - T$ relation is experimentally observed in a narrow temperature range ($283$ K $\leq T \leq 295$ K). This discrepancy could arise from the negligible size variation of skyrmion in the studied narrow temperature range, as shown in the inset to Fig. 4(b).

On the other hand, it is noted that the diffusion coefficient $\mathcal{D}_{dc}$ obtained near room temperature is about $\mathcal{D}_{dc} = \sim 0.4 \times 10^{-12}$ m$^2$ s$^{-1}$, which is much smaller than the value of $\mathcal{D}_{dc} = \sim 3.0 \times 10^{-8}$ m$^2$ s$^{-1}$ obtained from simulations. We note that the experimentally observed skyrmion radius is approximately 100 times larger than the simulated skyrmion radius, and the diffusion coefficient $\mathcal{D}_{dc}$ given by Eq. (4) thus rapidly decreases with the increase of skyrmion radius since it contributes significantly to the dissipative tensor $\mathcal{D}$. Namely, the diffusion coefficient $\mathcal{D}_{dc}$ approaches zero as the skyrmion radius approaches infinity. In addition, the random distribution of defects could also influence the diffusion dynamics.

In conventional Brownian motion, the surrounding media provides both driving and friction forces. The friction force is the second term on the left side of the stochastic Thiele equation, namely, $\alpha \mathcal{D} \cdot \mathbf{v}$. The standard deviation of friction forces can be calculated as $\sigma_{F_\alpha} = \alpha \mathcal{D} \sqrt{\langle v_x^2 \rangle}$, $\sigma_{F_\beta} = \alpha \mathcal{D} \sqrt{\langle v_y^2 \rangle}$, respectively. By incorporating the material specific parameters (given in the Supplementary Materials), the friction forces $\sigma_{F_\alpha} = 2.5 \times 10^{-19}$ N and $\sigma_{F_\beta} = 1.9 \times 10^{-19}$ N were estimated, respectively. The amplitude of these forces is at the same level as the skyrmion-edge repulsive interaction that is consistent with theoretical predictions \cite{55}.

In summary, we have experimentally and numerically studied the diffusion dynamics of a single isolated skyrmion driven by the random thermal fluctuations. The diffusion characteristics of skyrmion, including random trajectory, Gaussian distribution of probability density distribution and their linearly temperature dependent diffusion coefficient are consistent with conventional Brownian particles. Moreover, by studying skyrmions with opposite skyrmion numbers ($Q = \pm 1$), we have observed a spin-topology dependent diffusion behavior which is previous absent in other bubble materials. Our results thus revealed the intriguing topological aspects of nonequilibrium thermodynamics of skyrmions, which could also provide some insights for developing thermally stable skyrmion-based spintronic devices and potentially enable novel applications of skyrmions in reservoir/probability computing schemes, for examples \cite{33-35}.
Acknowledgements. Work carried out at Tsinghua was supported by the Basic Science Center Project of NSFC (Grant No. 51788104), National Key R&D Program of China (Grant Nos. 2017YFA0206200 and 2016YFA0302300), the National Natural Science Foundation of China (Grant No. 11774194, 51831005, 1181101082), and the Beijing Advanced Innovation Center for Future Chip (ICFC). X.Z. was supported by the Presidential Postdoctoral Fellowship of the Chinese University of Hong Kong, Shenzhen (CUHKSZ). X.Z. was supported by JSPS RONPAKU (Dissertation Ph.D.) Program. Y.Z. was supported by the President’s Fund of CUHKSZ, the National Natural Science Foundation of China (Grant No. 11574137), and Shenzhen Fundamental Research Fund (Grant Nos. JCYJ20160331164412545 and JCYJ20170410171958839).

References
1 Rossler, U. K., Bogdanov, A. N. & Pfleiderer, C. Nature 442, 797-801 (2006).
2 Mühlbauer, S. et al. Science 323, 915-919 (2009).
3 Yu, X. Z. et al. Nature 465, 901-904 (2010).
4 Nagaosa, N. & Tokura, Y. Nature Nanotechnology 8, 899-911 (2013).
5 Romming, N. et al. Science 341, 636-639 (2013).
6 Chen, G., Mascaraque, A., N'Diaye, A. T. & Schmid, A. K. Appl Phys Lett 106, 242404 (2015).
7 Wiesendanger, R. Nat Rev Mater 1, 16044 (2016).
8 Fert, A., Reyren, N. & Cros, V. Nat Rev Mater 2, 17031 (2017).
9 Jiang, W. et al. Physics Reports-Review Section of Physics Letters 704, 1-49 (2017).
10 Fert, A., Cros, V. & Sampaio, J. Nature Nanotechnology 8, 152-156 (2013).
11 Sampaio, J., Cros, V., Rohart, S., Thiaville, A. & Fert, A. Nat. Nanotech. 8, 839-844 (2013).
12 Jiang, W. et al. Science 349, 283-286 (2015).
13 Du, H. et al. Nat. Commun. 6, 8504-8504 (2015).
14 Zhang, X. C., Ezawa, M. & Zhou, Y. Sci. Rep. 5, 9400 (2015).
15 Woo, S. et al. Nat. Mater. 15, 501-506 (2016).
16 Boulle, O. et al. Nature Nanotechnology 11, 449-454 (2016).
17 Kang, W., Huang, Y., Zhang, X., Zhou, Y. & Zhao, W. Proc. IEEE 104, 2040-2061 (2016).
18 Yu, G. et al. Nano Lett. 17, 261-268 (2017).
19 Schulz, T. et al. Nat. Phys. 8, 301 (2012).
20 Neubauer, A. et al. Physical Review Letters 102, 186602 (2009).
21 Zang, J., Mostovoy, M., Han, J. H. & Nagaosa, N. Phys. Rev. Lett. 107, 136804 (2011).
22 Jiang, W. et al. Nature Physics 13, 162-169 (2017).
23 Litzius, K. et al. Nature Physics 13, 170-175 (2017).
24 Troncoso, R. E. & Núñez, Á. S. *Annals of Physics* **351**, 850-856 (2014).
25 Troncoso, R. E. & Núñez, Á. S. *Phys. Rev. B* **89**, 224403 (2014).
26 Schütte, C., Iwasaki, J., Rosch, A. & Nagaosa, N. *Phys. Rev. B* **90**, 174434 (2014).
27 Barker, J. & Tretiakov, O. A. *Phys. Rev. Lett.* **116**, 147203 (2016).
28 Miltat, J., Rohart, S. & Thiaville, A. *Phys. Rev. B* **97** (2018).
29 Moreau-Luchaire, C. *et al.* *Nature Nanotechnology* **11**, 444-448 (2016).
30 Sonntag, A., Hermenau, J., Krause, S. & Wiesendanger, R. *Phys. Rev. Lett.* **113**, 077202 (2014).
31 Hagemeister, J., Romming, N., von Bergmann, K., Vedmedenko, E. Y. & Wiesendanger, R. *Nat. Commun.* **6**, 8455-8455 (2015).
32 Zhang, X. C., Ezawa, M. & Zhou, Y. *Physical Review B* **94**, 064406 (2016).
33 Prychynenko, D. *et al.* *Physical Review Applied* **9**, 014034 (2018).
34 George Bourianoff, Daniele Pinna, Matthias Sitte & Everschor-Sitte, K. *AIP Advances* **8**, 055602 (2018).
35 Jakub Zázvorka *et al.* (Preprint: [https://arxiv.org/abs/1805.05924](https://arxiv.org/abs/1805.05924), 2018).
36 Uhlenbeck, G. E. & Ornstein, L. S. *Physical Review* **36**, 0823-0841 (1930).
37 Wang, M. C. & Uhlenbeck, G. E. *Rev. Mod. Phys.* **17**, 323 (1945).
38 Mori, H. *Prog Theor Phys* **33**, 423 (1965).
39 Kotera, T. & Toda, M. *Journal of the Physical Society of Japan* **14**, 1475-1490 (1959).
40 William Fuller Brown, J. *Physical Review* **130**, 1677 (1963).
41 Garcia-Palacios, J. L. & Lazaro, F. J. *Physical Review B* **58**, 14937-14958 (1998).
42 Reitz, D., Ghosh, A. & Tchernyshyov, O. *Physical Review B* **97**, 054424 (2018).
43 Thiele, A. A. *Physical Review Letters* **30**, 230 (1973).
44 Mochizuki, M. *et al.* *Nature Materials* **13**, 241-246 (2014).
45 Aurore Finco *et al.* *Physical Review Letters* **139**, 037202 (2017).
46 Everschor-Sitte, K. & Sitte, M. *Journal of Applied Physics* **115**, 172602 (2014).
47 Choi, J. *et al.* *Physical Review Letters* **98**, 207205 (2007).
48 Tolley, R., Montoya, S. A. & Fullerton, E. E. *Phys Rev Mater* **2**, 044404 (2018).
49 Seaberg, M. H. *et al.* *Physical Review Letters* **119**, 067403 (2017).
50 Malozemoff, A. P. & Slonczewski, J. C. *Magnetic domain walls in bubble materials*. (Academic Press, 1979).
51 Ezawa, M. *Physical Review Letters* **105**, 197202 (2010).
52 Reichhardt, C. & Reichhardt, C. J. O. *Reports on Progress in Physics* **80**, 026501 (2017).
53 Li, T. C., Kheifets, S., Medellin, D. & Raizen, M. G. *Science* **328**, 1673-1675 (2010).
54 Saxton, M. J. *Biophys J* **72**, 1744-1753 (1997).
55 Navau, C., Del-Valle, N. & Sanchez, A. *Physical Review B* **94**, 184104 (2016).
Figure Captions

FIG. 1 (color online). Micromagnetic simulation study of skyrmion Brownian motion. (a) Simulated trajectories of a single isolated skyrmion ($Q = -1$) with its radius $r_{sk} < 100$ nm at selected temperatures. The total simulation time is 20 ns with a step of 20 ps. (b) The mean squared displacements (MSDs) and their linear fits at selected temperatures. (c) Diffusion coefficient ($D_{dc}$) as a function of temperature for skyrmion with opposite skyrmion numbers ($Q = \pm 1$). (d) Simulated $D_{dc}$ as a function of damping coefficient ($\alpha$) for skyrmions with $Q = \pm 1$ at $T = 100$ K and $B_z = 3$ mT. The solid curve represents the analytical solution based on Eq. (2) with a dissipation parameter $\mathcal{D} = 6.11 \times 10^{-14}$ J s m$^{-2}$ and a gyrocoupling parameter $\mathcal{G} = -4.83 \times 10^{-14}$ J s m$^{-2}$. (e) Evolution of diffusion skyrmion Hall angle $\theta_{sk}$ as a function of time $t$ for $Q = \pm 1$ skyrmions.

FIG. 2 (color online). Brownian motion of a single isolated skyrmion ($Q = -1$) in a Ta/CoFeB/TaO$_x$ thin film of 25.5 µm × 74 µm at $T = 293.15$ K and $B_z = +2.1$ mT. (a) Trajectory of the skyrmion captured from the video tracking results with 18 frames per second over 157.62 s (i.e., 2837 frames). The color scale denotes the evolution of time. Selected top-views of the skyrmion captured by using MOKE microscope at $t_1 = 19.5$ s, $t_2 = 40$ s, $t_3 = 78$ s, $t_4 = 133$ s and $t_5 = 156.5$ s are given. Dark contrast corresponds to the magnetization pointing along the $-z$ direction. (b) Measured transient skyrmion velocities ($v_x$, $v_y$) as a function of time. (c) Probability density function (PDF) of the skyrmion velocity along $x$ and $y$ axes, respectively. The associated fittings yield estimation of full width at half maximum (FWHM), red line.

FIG. 3 (color online). Evolution of trajectories of a single isolated ($Q = -1$) skyrmion at various temperatures, (a) $T = 282.05$ K, (b) $T = 284.13$ K, (c) $T = 287.02$ K, (d) $T = 291.17$ K, (e) $T = 293.45$ K, and (f) $T = 294.03$ K.

Fig. 4 (color online). (a) The calculated MSDs of a single isolated ($Q = -1$) skyrmion measured at selected temperatures. (b) The diffusion coefficient $D_{dc}$ as a function of temperature for $Q = \pm 1$ skyrmions. Values of $D_{dc}$ at different temperatures were calculated based on the MSD data by using Eq. (1), which can be linearly fitted by using Eq. (5) (red line). The inset to Fig. (b) corresponds to the experimentally measured $r_{sk}$ as a function of temperature.
Figure 1

Figure 2
