

Thermal skyrmion diffusion used in a reshuffler device

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Magnetic skyrmions in thin films can be efficiently displaced with high-speed by using spin transfer torques^{1,2} and spin-orbit torques³⁻⁵ at low current densities. While this favourable combination of properties has raised expectations for using skyrmions in devices^{6,7}, only few publications have studied the thermal effects on the skyrmion dynamics⁸⁻¹⁰. However, thermally induced skyrmion dynamics can be used for applications¹¹, e.g. unconventional computing approaches¹², as they have been predicted to be useful for probabilistic computing devices¹³. In our work, we uncover thermal diffusive skyrmion

dynamics by a combined experimental and numerical study. We probe the dynamics of magnetic skyrmions in a specially tailored low pinning multilayer material. The observed thermally excited skyrmion motion dominates the dynamics. Analysing the diffusion as a function of temperature, we find an exponential dependence, which we confirm by means of numerical simulations. Diffusion of skyrmions is further employed in a signal reshuffling device as part of skyrmion-based probabilistic computing architecture. Due to its inherent 2D texture, observation of a diffusive motion of skyrmions in thin film systems may also yield insights in soft matter-like characteristics (e.g. studies of fluctuation theorems, thermally induced roughening, etc.), thus making it highly desirable to realize and study thermal effects in experimentally accessible skyrmion systems.

We investigate skyrmions in specially developed low pinning Ta(5)/Co₂₀Fe₆₀B₂₀(1)/Ta(0.08)/MgO(2)/Ta(5) stacks using magneto-optical Kerr effect (MOKE) microscopy (for details see Methods). By varying the out-of-plane field, we can tailor the skyrmion density and radius. We nucleate single skyrmions in a deterministic fashion using current injection (see Supplementary Video 1), thereby controlling the density of skyrmions precisely so as to avoid skyrmion-skyrmion interactions¹⁴.

After having injected the desired number of skyrmions into a microstructured film element, we then observe skyrmions in real-time with the Kerr microscope without any further current injection or any other external stimulus. In contrast to previous reports and our previous results in different stacks^{4,5}, here, we clearly observe skyrmion motion after the system has relaxed and no after-effect of the current pulse is present anymore (see Supplementary Video 2). At constant conditions, the skyrmions move randomly throughout the sample. To evaluate the diffusive skyrmion motion, individual skyrmions were tracked (see Supplementary Video 3) and an example of several typical skyrmion trajectories is shown in Fig. 1. Having established motion without any external excitations, the first step is to identify the origin of the dynamics. One possible origin is the presence of thermal effects that can lead to diffusion in regimes ranging from sub- to super-diffusive¹⁵ or to Brownian motion. The different regimes can be identified from the mean squared displacement

(MSD) of the measured skyrmions. For diffusive motion assuming rigid particles with no correlations (from here on denoted as pure diffusion), the average mean squared displacement is proportional to time. We therefore plot the MSD

$$\text{Eq. (1): } MSD = \left\langle (\mathbf{R}_v(t) - \mathbf{R}_v(0))^2 \right\rangle = 2\mathcal{D}t \cdot d$$

as a function of time t elapsed and calculate the diffusion coefficient \mathcal{D} . The parameter d denotes the dimension of the system and takes the value of $d = 2$ for our measurement. As the evaluation of diffusion assumes a random motion of particles, to obtain statistically sound values of the diffusion coefficient, the MSD of many particles has to be averaged and the skyrmions have to be treated as a statistical ensemble. The resulting average MSD as a function of time at room temperature is shown in the inset in Fig. 1. We find a linear dependence as in Eq. (1) and a linear fit of the evaluated data reveals the diffusion coefficient of the material stack to be $\mathcal{D} = 0.27(20) \times 10^{-12} \text{ m}^2 \cdot \text{s}^{-1}$ at a temperature $T = 296 \text{ K}$. With the MSD being linearly proportional to time and the diffusion constant above, we conclude that skyrmions exhibit diffusive dynamics which is relevant at room temperature on macroscopic time scales. To compare our results with theoretical predictions¹⁶ (details, see Supplementary Figure 1-3), we note that the central quantities are the gyrocoupling \mathcal{G} and the dissipative tensor element \mathcal{D} . The latter depends on the profile of the skyrmion, which may change with its size¹⁷. We therefore determine values of \mathcal{D} for our studied skyrmions via a numerical integration of a skyrmion that was calculated by micromagnetic simulations for the appropriate material parameters of the experimentally used system. Surprisingly, our experimentally observed diffusion coefficient at room temperature is several orders of magnitude smaller than what was predicted¹⁶ for our experimental system ($\mathcal{D} \cong 4 \times 10^{-8} \text{ m}^2 \cdot \text{s}^{-1}$). However, even for the comparatively low diffusion coefficient values, we observe skyrmion displacement at a time scale of seconds. Hence, in addition to current-induced torques that can generate skyrmion motion, also thermal diffusion can cause skyrmion dynamics and even be the dominant source of dynamics in a low-pinning system.

To understand this deviation of the diffusion coefficient from the prediction, we probe the diffusion behaviour of skyrmions as a function of temperature. We evaluate the MSD as a function of time for temperatures from 285 to 320 K. Pure diffusion behaviour holds for all probed temperatures. The evaluated temperature dependence of the skyrmion diffusion coefficient is shown in Fig. 2, revealing an exponential dependence.

We note that the measurement of the diffusion coefficient as a function of temperature includes a change of the skyrmion radius due to small changes in the magnetic properties with temperature. While there is a small dependence of the skyrmion diffusion on radius (see Supplementary Figure 4), this cannot explain the observation that the diffusion coefficient depends exponentially on temperature (see Fig. 2). Within the error bars, the temperature dependence can be described for the whole range including the different skyrmion sizes by a single exponential dependence, showing that any effects of the size change have a negligible effect on the temperature dependence compared to the intrinsic thermal effect that leads to the exponential dependence. We conclude that the previously used assumption of rigid skyrmions in a translationally invariant environment^{16,17} is not sufficient to describe our observations. To check the reason for this discrepancy, we further analyse the displacement as a function of time from the experiment and find that skyrmions exhibit extended dwell times at certain positions (see Supplementary Figure 5). This means that we have to consider a non-flat potential landscape of the material where skyrmions exhibit non-constant dwell times at certain positions due to the energy landscape that varies spatially. This process is to some degree analogous to thermally activated movements of particles in solids¹⁸.

To check if this explanation holds, we perform atomistic simulations using the well-studied model system¹⁴ for a Pt₉₅Ir₅/Fe/Pd ultrathin film which, without any spatially varying energy landscape, which confirms the previously published findings^{16,17}. Considering spatial variations in the energy landscape, however, we reproduce the superlinear dependence of the diffusion coefficient for temperatures below the pinning energy barrier (see inset Fig. 2); thus, we obtain qualitative agreement between simulations and

experimental findings. At higher temperatures the skyrmion motion becomes mostly unaffected by the pinning potential and we retain the theoretical solution for free skyrmion diffusion. This means that we can explain our experimental observations by simulations that consider a non-flat potential landscape. Reciprocally, we show that the dwell times can be directly used to probe the potential landscape (see Supplementary Figure 5). In our simulations, the size changes of the skyrmions are not significant and the experimental behaviour is still reproduced showing that changes in the skyrmion size do not dominate the temperature dependence of the diffusion. If disorder becomes so sizeable that the skyrmion shape changes strongly (which we do not observe in simulation, nor in the experiment), this will lead to possible additional effects that are beyond the scope of this work.

Beyond analysing the intrinsic system properties, thermal skyrmion dynamics has also been put forward as a mechanism that allows for signal decorrelation¹³, a key missing component for probabilistic computing. Logic gates used in probabilistic computing circuits are very sensitive to correlations among input signals during operation. Correlated inputs result in unwanted propagations of correlations which ultimately lead to incorrect computational results¹³. The latter has been predicted to be circumvented by a device denoted as skyrmion reshuffler and it was theoretically shown that topologically non-trivial skyrmions are suitable spin structures that have an isotropic 2D diffusion¹³. Suggestions to use skyrmions for token-based Brownian circuits have also been recently studied¹⁹.

For a proper operation, input signals have to be periodically reshuffled without altering the *p-value* (signal up/down ratio) used in the actual computing step²⁰. A first proof-of-concept can be performed by sampling telegraph noise signals and de-correlating them¹³. We sample an input telegraph noise signal by generating a stream of skyrmions and successively scramble it by driving the stream through a reshuffling chamber, generating an output signal that is uncorrelated to the input while preserving *p-values* with high fidelity.

To implement the skyrmion reshuffler operation, we compare independent measurements for the respective 0-bit and 1-bit channels as shown in Fig. 3. To evaluate the operation, we generate two separate skyrmions streams and consider them as an example

of an appropriately sampled input signal. Skyrmions are nucleated at the contact and then driven through the reshuffling chambers by applying a DC current. The thermal diffusion leads to a reshuffling of the order in which skyrmions enter/leave the system, thus decorrelating output and input signals. To assess the functionality of the device, we compare the *p-value* of the input and output to determine the fidelity. The deterministic generation of skyrmions as a result of input signal sampling is engineered to be an unrelated process not relevant for the reshuffling that we probe. The operation is recorded and evaluated as shown in Fig. 3. A real-time movie of the skyrmion reshuffler operation and the signal evaluation is shown in Supplementary Video 5. The *p-values* of in-/output signals are calculated according to their definition, while the correlation of the two signals is evaluated using the equation for Pearson correlation factor ρ

$$Eq. (2): \rho = \frac{cov(in,out)}{\sigma_{in} \cdot \sigma_{out}},$$

where *cov* means the covariance of the signals and σ is the standard deviation of the respective signals. Evaluating the skyrmion reshuffler operation using a current density of $j = 3 \times 10^8 \text{ A} \cdot \text{m}^{-2}$ in the wire leading to the reshuffling chamber and taking several operation runs into account, we obtain $p_{input} = 0.51 \pm 0.08$. We find a negligible change of the *p-value* between input and output signals, $\Delta p = 0.01 \pm 0.08$, showing the high fidelity of the signal retention. The calculated correlation factor is $\rho = 0.11 \pm 0.14$ which denotes a generation of an output signal that is highly uncorrelated to the device input signal. The total number of skyrmions used to represent and reshuffle the in-/output signals was approx. 100, limited by the slow optical detection process used here. Larger numbers of skyrmion streams that exhibit faster diffusion, as likely used in a real device where electronic read-out times can be orders of magnitude faster than using our optical technique, will lead to better statistics and even higher *p-value* fidelities¹³.

Our findings provide a number of statistical mechanics analysis methods for skyrmions: for example, the thermal dynamics can be utilized to quantify the energy landscape of a system and to determine the activation energy. Our results also introduce further possibilities of investigating the effects of soft matter on skyrmions systems in thin films, e.g. studying creep theorems and melting in 2D. The proof-of-concept reshuffler device demonstrates that the observed thermal skyrmion motion results in the creation of a signal highly uncorrelated to the device input while maintaining the same *p-value* of the telegraph signal, revealing the possibility of implementing skyrmion-based devices in probabilistic computing.

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Acknowledgements

The project was funded by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) - project numbers 403502522 and 49741853, SFB 767, SFB TRR173 and by grant No. EV 196/2-1. M.K., S.J. and G.J. acknowledge support of the WALL project (FP7-PEOPLE-2013-ITN 608031). L.R. acknowledges the support of the Alexander von Humboldt Foundation. P.V. would like to thank the DFG TRR146 for partial financial support. J.Z. acknowledges the help and advice of the technicians of the Kläui group, especially S. Kauschke.

Author contribution statement

M.K. and U.N. proposed and supervised the study. J.Z., S.J. and K.L. fabricated devices and characterized the multilayer samples. J.Z., D.H. prepared the measurement setup and together with N.K. and S.K. conducted the experiments using Kerr microscope. J.Z. and D.H. evaluated the experimental data with the help of P.V. and G.J. F.J. and A.D. performed the theoretical calculations and atomistic simulations of skyrmion diffusion. L.R. calculated the model parameters. J.Z. produced, measured and

analysed the skyrmion reshuffler under the supervision of D.P., K.E-S. and M.K. J.Z. drafted the manuscript with the help of M.K. and U.N. All authors commented on the manuscript.

Financial and Non-Financial Competing Interest statement

The authors declare no competing financial interests.

Figure captions

Fig. 1: Trajectories of selected skyrmions at 296 K. The reference position was taken from the first frame in the measurement. All skyrmions are set to start at position $[0;0]$. The timescale of the observation is in the seconds to minutes range. The inset shows the time-averaged MSD (black line) and the linear fit of the data (red dashed line).

Fig. 2: Temperature dependence of the evaluated skyrmion diffusion coefficient considering all observed skyrmions. The linear dependence in the semi-logarithmic plot shows a clear exponential dependence of the diffusion coefficient on temperature over two orders of magnitude. The error bars are calculated as standard deviations of the diffusion coefficient value over all measurements at a certain temperature. The inset shows the temperature dependence of the diffusion coefficient obtained from simulations (points) of a spin model with a non-flat energy potential landscape, normalized to the mean-field exchange constant $J_0=98$ meV (see Supplementary Video 4). The simulation results likewise show a strongly enhanced temperature dependence at low temperature, which eventually

transforms into the quasi-free diffusion expected from the theory (line) at elevated temperatures.

Fig. 3: Observation the skyrmion reshuffler device operation. a) Reshuffler operation with skyrmion nucleation by DC current. The input signal is constructed as a time frame where the skyrmion crosses the blue threshold line. The output is produced upon crossing the orange line. b) The corresponding input signal is depicted in blue colour on top and resulting output signal on the bottom. The full process of the reshuffler operation is shown in Supplementary Video 5. Radius of the reshuffling chamber is 40 μm . Reconstructed p -value for input signal is $p_{input} = 0.51 \pm 0.08$.

Methods

Sample Parameters

The samples used in this study were grown as a continuous single layer stack of Ta(5)/Co₂₀Fe₆₀B₂₀(1)/Ta(0.08)/MgO(2)/Ta(5) (thickness in parentheses is given in nm). The sample was prepared by sputtering using a Singulus Rotaris sputtering tool with a base pressure 3×10^{-8} mbar. Using a Singulus Rotaris deposition system, we can tune the thickness of the layers with high accuracy (reproducibility better than 0.01 nm). The stack was annealed at 200° C for 1 hour to optimize the properties including the perpendicular magnetic anisotropy (PMA). It has been shown that introducing a very thin metal interlayer between the CoFeB and MgO layers can tune the PMA of the material and result in a skyrmion phase nucleation at room temperature²¹. By tuning the layer thicknesses and optimizing the sputter parameters we can obtain films that exhibit skyrmions with thermal motion at room temperature and by applying

small out-of-plane (OOP) external fields, the skyrmion size can be adjusted. The hysteresis loop of the used sample shows a typical hour-glass shape, usually related to the presence of a magnetic skyrmion or stripe phase^{4,22}. We find at zero magnetic field that a stripe domain phase is present in the material. Within the resolution of the measurement, the ratio between the two magnetization states (up and down sweeps) is within the measurement precision 1:1, showing that the coercivity of the material is less than 0.05 mT. The threshold for the creation of skyrmion is at 0.25 mT, forming skyrmions with a radius of around 1 μm . Samples were then patterned by electron beam lithography (EBL) into $60 \times 120 \mu\text{m}^2$ pads with gold contacts for current injection. The sample was characterized with a superconducting quantum interference device (SQUID) and MOKE to determine the magnetic properties of the material stack.

The anisotropy field value was determined with a hard axis loop measurement using SQUID²³. The DMI of a multi-stack material can be calculated from the domain pattern periodicity based on the measured values for anisotropy and saturation magnetization^{4,24}. We measured the worm domain periodicity around zero field and calculated the DMI using the obtained data assuming an exchange stiffness²⁵ of $A = 10 \text{ pJ} \cdot \text{m}^{-1}$ and the thickness of the magnetic material of 1 nm. The domain periodicity was evaluated using a fast Fourier transformation (FFT) of the MOKE pictures obtained at zero field. The material parameters at 300 K are summed up in Supplementary Table 2.

Measurement Setup

The measurement setup used was a commercial Evico GmbH MOKE microscope. The electro-magnetic coil for supplying OOP field was custom made at University in Mainz and contains a Peltier element QC-32-0.6-1.2 to change the temperature of the studied sample while on ambient air. The achievable temperature range is 280 – 350 K. The temperature is controlled by resistivity

measurement on a Pt100 resistor which is placed on top of the Peltier element next to the sample. The stability of the set temperature is found to be within 0.3 K. The coil for inducing IP field was obtained from the microscope supplier. The microscope camera is capable to record 16 frames per second, therefore the timestep in the observation and evaluation of the skyrmion diffusion is 62.5 ms. Pulses for current driven displacement were supplied by an Agilent 33250A Arbitrary Waveform Generator

Current Induced Dynamics

By current injection, we can nucleate single skyrmions in a deterministic fashion, thereby controlling the density of skyrmions precisely so as to avoid skyrmion-skyrmion interactions¹⁴. We first study the skyrmions present for a fixed perpendicular field value of 0.35 mT leading to a skyrmion radius of approx. 0.9 μm . To understand their topology, we analyse spin-orbit torque driven displacements. With applying to the sample 4 V for 5 ms, we have a 96% probability of nucleating one skyrmion from the contact pad, see Supplementary Video 1. Even for multiple pulse bursts, one pulse generates one new skyrmion in the sample when applying 4 V bias. Different origins of the nucleation have been discussed, such as heating and spin torque effects in conjunction with local defects²⁶⁻²⁹. Our results indicate that a gradient in the current at the injection spot leads to skyrmion nucleation probably from a combination of heating and spin torques. As skyrmions are always nucleated at a certain location, we conclude the presence of a hotspot, acting as a nucleation site for the skyrmions during current injection. Such a hotspot is preferential since it allows for reproducible skyrmion injection along defined paths.

Upon biasing with a current pulse, skyrmions already present in the sample are moved synchronously in a direction opposite to the current direction in line with the DMI and spin Hall angle expected for a Ta-based stack³⁰. The skyrmion velocity increases with

current density and for $6.4 \times 10^{10} \text{ A} \cdot \text{m}^{-2}$ we obtain a velocity value of $v = 5 \text{ mm} \cdot \text{s}^{-1}$, similar to what has been reported previously³. As presented by Jiang et al.³ on a stack with similar composition, we can distinguish between chiral Q=1 skyrmions and non-chiral Q=0 magnetic bubbles by applying current. Non-chiral structures either expand or shrink in size upon current injection while chiral skyrmions move parallel to the current direction. The fact that we observe that skyrmions move as a whole due to current, indicates that the skyrmion have a topological charge Q=1, which also shows that they have the appropriate spin structure that exhibits isotropic lateral diffusion being optimal for the performance of the reshuffler. The velocity values differ from the values reported previously in a Pt-based samples⁵. However, the investigated material stack has a reliable skyrmion nucleation and transport with current pulsing. At lower current densities (up to $6.4 \times 10^{10} \text{ A} \cdot \text{m}^{-2}$), we also found the skyrmion Hall angle to be relatively constant around 0° as previously identified for low velocities³¹.

Skyrmion Imaging and Tracking

We can use different approaches to nucleate skyrmions. The specific mechanism chosen for skyrmion generation is not crucial for our measurements, but we need to have a sufficiently low skyrmion density in order to prevent significant skyrmion-skyrmion interaction during the diffusion. While in our sample, skyrmions can for instance be nucleated by external field sweeps, this procedure however does not allow for the skyrmion density to be easily controlled. A good control of the skyrmion density in our case is achieved by nucleating skyrmions at the electric contacts by injecting electric current pulses. This approach allows us to fill the structure with skyrmions and due to their repulsive interaction, we find a rather homogeneous skyrmion density. The diffusion measurements are however carried out with lower skyrmion density to avoid skyrmion-skyrmion interaction, see Supplementary Video 2.

Finding skyrmions and the determination of the skyrmion radius is done using a custom Wolfram Mathematica script. The program binarizes the frame with observed skyrmions and finds structures with one direction of magnetization. The binarization threshold is obtained by comparing the Fast Fourier Transformation (FFT) of the original picture with the binarized picture using various thresholds. The threshold that yields the same FFT of the binarized picture as the FFT of the original one is then taken for further evaluation.

The spatial resolution of our MOKE microscope limits the determination of the magnetic structure profile, as the domain wall width of the skyrmion falls into less than one pixel of the measurement camera. Therefore, we see a rapid intensity change within one pixel. All observed skyrmions have a round shape with some deformations that fall into the resolution limit of the measurement technique. To determine the skyrmion radius, we fit the structures using the function “EquivalentDiskRadius” in Wolfram Mathematica, which fits a disk that covers the same area as the observed structures. The errors of the skyrmion radius determination are then largely determined by the resolution of the microscope.

For the tracking of skyrmion motion, the original images are evaluated instead of the binarized. We track the skyrmion motion with a 62.5 ms time resolution using the IMAGEJ³² software with the TRACKMATE plugin^{33,34}, see Supplementary Video 3. The identification of skyrmions is based on their contrast and intensity and the validity of the tracking algorithm was confirmed by manual analysis and comparison for several samples. In order to apply the diffusion theory and obtain a reliable diffusion coefficient, we average over the measurement of several skyrmions.

To make use of the full statistics, the time-averaged MSD³⁵ is employed in the MSD evaluation using the data of all skyrmions observed. In this method, the skyrmion position at every timeframe is used as a reference point (starting point) for that particular skyrmion tracking. The obtained MSDs for every starting point are then averaged over the whole time of the measurement video. This

effectively means that we have many data points for shorter time differences, whereas only a few data points for longer timescales. Therefore, the statistical error increases largely considering longer measurement times. To compensate for this, we only consider the timescales of half of the measurement time for the evaluation of the diffusion coefficient. The diffusion coefficient is obtained as linear fit of the average MSD depending on time, as described in the Eq. 1. The error of the evaluation is calculated as the standard deviation of the MSDs of the observed skyrmions.

For the observation of skyrmion dwell times at different locations and deducing the effect of the non-flat potential landscape, the time-averaged MSD was not used in the evaluation. Here only the skyrmion positions in the first frame of the observation video are taken as reference for the calculation of the MSD and the evaluation of the dwell times.

The lowest temperature that we can analyse for this sample stack is 290 K due to the limitation of the total measurement time and the necessary displacement distances to obtain sufficient statistics for the case of low diffusion. Note that the temperature range is limited by the measurements in ambient atmosphere and above 305 K more and more skyrmions are nucleated leading to a skyrmion lattice formation, where then the assumption of non-interacting skyrmions breaks down.

For the reshuffler measurement, the current density is set as $j = 3 \times 10^8 \text{ A} \cdot \text{m}^{-2}$, calculated at the moving pad before the reshuffling chamber. The current density inside the chamber is up to a factor of four lower due to the larger dimensions, enhancing the competition of skyrmion diffusion to current induced motion. The dependence of the interplay between diffusion and current-induced displacement for different current densities is beyond the scope of this work, but was theoretically investigated¹³. Tracking of skyrmions was used to reconstruct the signal in the reshuffler operation. To evaluate the operation, we generate two separate skyrmions streams and assume they are the result of an appropriately sampled input signal. By continuously generating skyrmions at the contact by DC current and driving them through the reshuffler chamber we measure input and output streams by detecting each skyrmion that crosses

the blue and orange threshold lines (Fig. 3). Upon crossing the blue threshold line in front of the reshuffling chambers, the corresponding bit information (0 or 1, depending on which chamber the skyrmion enters) is triggered allowing reconstruction of the equivalent input signal that would have generated this specific sequence of skyrmions. The output signal is obtained using the same principle, where the order of skyrmion crossing the orange threshold line allows for the reconstruction of an output telegraph signal (depicted in Fig. 3).

Data availability

The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

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Additional Information

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