nature portfolio

Peer Review File

Driving skyrmions in flow regime in synthetic ferrimagnets

Reviewers' Comments:

Reviewer #1:
Remarks to the Author:
This manuscript reports an experimental study on current-induced skyrmion motion. Utilizing Co/Tb synthetic ferrimagnets, the authors achieved a flow regime, with the velocity up to 400 m/s, for a small skyrmion (diameter < 100 nm). The design of heterostructures helps to increase the spin-orbit torque efficiency and to reduce the skyrmion Hall angle. This result can be compared with previous experiments: Ref. [38] reported ~ 150 m/s for a large skyrmion (diameter = 400-900 nm) whereas Ref. [47] reported ~ 600 m/s for an even larger skyrmion (diameter ~ 12,000 nm).
As well explained in the introduction, a (rather) straight and fast motion of magnetic skyrmion is important for the realization of skyrmion racetrack devices. This work has been carried out with careful material design, systematic measurements, and reasonable interpretations. In this respect, this work is undoubtedly a meaningful contribution to ongoing research of magnetic skyrmions. However, in comparison with previous works (Refs. [38, 47]), I do not find any breakthrough or new physics. In particular, a flow regime of current-induced skyrmion motion has already been demonstrated in Fig. 4(a) of Ref. [38]. Number-wise, the skyrmion velocity for a small skyrmion in this work is worthwhile of noting but this is an incremental advance based on already-known strategies. Therefore, I find this manuscript does not meet the standard of Nature Communications and may be suite to a more specialized journal.

Reviewer #2:
Remarks to the Author:
This paper presents a study of skyrmion (Sk) motion in synthetic ferrimagnetic multilayer whereby, varying the layer thicknesses, the magnetic interactions between transition metal and rare earth layers can be systematically varied.

A key result is observing a linear relation between Sk velocity and current density, i.e., evidence that the Sk motion is in a flow regime. Another interesting result is that Sks appears to be repelled by the wire edges, leading to a negligible Sk transverse displacement in micron-scale wires (while such a displacement is observed in wider wires). Interestingly, sample heating during the pulse does not appear to affect the Sk velocity results at high current densities, as seen in earlier studies at high current densities (e.g., their ref. 47).

The authors' claims are well supported by their results, and the manuscript is well written, with the key Sk physics well presented. In addition, the authors provide sufficient details to reproduce their studies. Of further interest would be time-resolved imaging studies to determine the reproducibility of the Sk displacements and resolve the Sk Hall angle and relaxation after pulses due to edge repulsion.

In sum, the manuscript reports significant results that merit publication in Nature Communication.

Reviewer #3:
Remarks to the Author:
In this work by Mallick et al., the authors have studied the transition from creep motion to the flow regime of skyrmions in synthetic ferrimagnets. Their results are obtained by increasing the amplitude of SOT and AFM coupling between the Co and Tb layers, by using the magnetic force microscope imaging technique. By comparing with the field-induced DW motion, the authors have proposed three different dynamical regimes of current-induced skyrmion motion: thermally activated motion, depinning motion and flow motion. Furthermore, they have achieved a relatively high-speed motion of ferrimagnetic skyrmion with a reduced skyrmion Hall effect. Through comparing the most recent developments in the field, I failed to appreciate the novelty and the robustness of their results. The present manuscript is not suitable for publication in journals such as Nature Com. I would encourage authors to revise the manuscript based on the following comments and to add additional information, after which this manuscript may be reconsidered.

(1) I noticed the value of interfacial DMI is being volume averaged. I am curious to see its comparison with similar multilayers.
(2) Its not completely clear to this referee about the advantage(s) of studying the present material system. Did authors study the damping parameters of their multilayers? It is known that Gd-based multilayers exhibit a relatively low damping parameters, as compared with the Tb based multilayers.
(3) Some of the TEM results are not very relevant. I can see clearly the layered resolved structures. Can TEM results be otherwise helpful in establishing the picture of interfacial chiral magnetism? Can they be helpful in establishing the Tb-thickness dependent DMI strength? Is Tb contributing additional source of chiral magnetism?
(4) I believe that the magnetometry data should be presented in the main text? Is there any substantial difference between them? It is clear that incorporating of Tb, introducing antiferromagnetic coupling that reduces the net magnetization. How is this helpful in reducing the size of skyrmion? What is the role of repetition number? What is the role of Al layer?
(5) The interlayers distance is approximately 6 nm. I am curious to see the

magnetization of Co/Tb of each layer is still coupled, considering a reduced magnetization. Can authors comment on this aspect?

(6) The MFM images shown in Figure 2 is not labeled. In line 150, “dotted wine line in Fig. 4” should be change to “Fig. 3”. The authors should thoroughly review the entire manuscript to avoid such errors.

(7) From Figure 4, it is clear that most of these skyrmions are still in the creep motion regime, by revealing an unequal displacement. It can also be seen from the very large error bar.

(8) It is also known that Tb element is very active, which means the nanostructure can be easily oxidized.

(9) The green solid line indicating depinning motion in the inset of Figure 3 is very unclear, the authors should make it more prominent. Additionally, the solid blue stars in Figure 3 are used to indicate the current-induced skyrmion motion, but there are no current coordinates provided.

(10) By comparing the Figures 4(a)-(d), it can be observed that some of the skyrmions annihilate during motion, and the diameter of the skyrmion is also varying. Please explain this phenomenon.

(11) The authors did not consider the effect of current-induced Joule heating on skyrmion nucleation and motion, please comments on it.

(12) In line 277, the authors mention “....1 μm narrow track without any transverse deflection....”, however, there appears to be transverse deflection of the skyrmions in Figures 4(a)-(d). The authors should quantitatively investigate the time-dependent trajectories of skyrmions.

(13) On a separate note, the so-called suppression of skyrmion Hall angle in the present synthetic ferrimagnet is most likely due to the edge repulsive force. I am looking forward to see the motion of skyrmion in a large device with a larger width. There, authors could observe free motion of skyrmion. I think a comparison of that results in those in the literature is fair.

(14) The theoretical calculation cannot be justified. Authors are using synthetic ferrimagnet, the usage of exchange stiffness should be very different with that of the Co.

(15) The skyrmion velocity is smaller than the reported value in CoGd, I suggest authors carefully rephrase the usage of their language.

Reviewer #1 (Remarks to the Author):

This manuscript reports an experimental study on current-induced skyrmion motion. Utilizing Co/Tb synthetic ferrimagnets, the authors achieved a flow regime, with the velocity up to 400 m/s, for a small skyrmion (diameter < 100 nm). The design of heterostructures helps to increase the spin-orbit torque efficiency and to reduce the skyrmion Hall angle. This result can be compared with previous experiments: Ref. [38] reported ~ 150 m/s for a large skyrmion (diameter = 400-900 nm) whereas Ref. [47] reported ~ 600 m/s for an even larger skyrmion (diameter ~ 12,000 nm). As well explained in the introduction, a (rather) straight and fast motion of magnetic skyrmion is important for the realization of skyrmion racetrack devices. This work has been carried out with careful material design, systematic measurements, and reasonable interpretations. In this respect, this work is undoubtedly a meaningful contribution to ongoing research of magnetic skyrmions. However, in comparison with previous works (Refs. [38, 47]), I do not find any breakthrough or new physics. In particular, a flow regime of current-induced skyrmion motion has already been demonstrated in Fig. 4(a) of Ref. [38]. Number-wise, the skyrmion velocity for a small skyrmion in this work is worthwhile of noting but this is an incremental advance based on already-known strategies. Therefore, I find this manuscript does not meet the standard of Nature Communications and may be suite to a more specialized journal.

Reply: We thank the referee for his/her critical reading of the manuscript and valuable opinion. However, we respectfully disagree with the reviewer that the present work is an "*incremental advance based on already-known strategies*".

It is rather well established in the skyrmion based research that the skyrmion size has to be reduced to at least in the sub-100 nm range for intended application in spintronic devices (be it in logic circuits, neuromorphic network, racetrack memories, etc.). This requirement imposes that the thermal stability of the skyrmions during electrical manipulation has to be ensured either by increasing the layer thickness or by stabilizing them in multilayers. Consequently, the skyrmions size is increased in a multi-layered structure due to the presence of a strong interlayer dipolar coupling. It is important to highlight that one of the primary motivations for utilizing antiferromagnetically coupled sub-lattice (layers) systems over conventional ferromagnetic ones, such as synthetic antiferromagnets or ferrimagnets, is their unique capability to stabilize smaller skyrmions due to the near absence of magnetization. Hence, on one hand, a fine balance needs to be present between all the different parameters to ensure thermal stability as well as smaller skyrmion size. On the other hand, driving the small compact skyrmions is challenging since the effect of pinning on them is more significant in compared to larger disordered skyrmions or domain walls [1-3].

Achieving high-speed motion of small skyrmions which to our knowledge has never been demonstrated so far, necessitates the optimization of two key factors: **maximizing spin-orbit torques (SOTs) and minimizing energy dissipation within the system**. This challenge is further compounded in multilayer structures, in which the optimization of individual layers must be harmonized across the entire thickness of the heterostructure. This inherent complexity represents a principal barrier that has, thus far, impeded the attainment of significant skyrmion velocities for these relatively small skyrmions.

In this context, we wish to draw the referees' attention to the fact that the literature mentioned also presents evidence of the existence of smaller, compact skyrmions, as anticipated in ferrimagnets. For instance**, Ref. 47 (**Quessab *et al*, Nano Letter 22, 6091, 2022) **reports skyrmions with diameters below 200 nm** as determined by STXM measurements, while **Ref. 38** (L. Berges *et al*, Phys. Rev. B 106, 144408, 2022) **indicates a diameter of 380 nm** as observed through MFM. However, it is crucial to note that despite the recognition of these smaller skyrmion structures, **the referenced studies do not provide any demonstration of their current-induced motion.** This absence of motion demonstration can be attributed to the aforementioned complexities inherent in manipulating and harnessing the mobility of such small-size textures. Hence, we posit that our work should not be regarded as a mere incremental addition to the existing literature. Instead, it signifies a substantial leap forward, characterized by comprehensive optimization efforts encompassing both the physics and materials aspects. This multifaceted optimization not only facilitates the swift linear movement of skyrmions but also guarantees their thermal stability, even in such reduced sizes.

In the following, we discuss the novelty of the work in details in comparison to the existing literature:

1. Various aspects of skyrmion velocity:

We agree that the skyrmion velocity obtained by Quessab *et al*, [Ref. 47] is similar (larger) to the present work. However, while stressing on the point that the diameter of the skyrmions are 1.2 µm, we discuss in the subsequent section why it is intrinsically easier to obtain large skyrmion velocity in such systems showing large skyrmionics bubbles. In Fig. RR1, we reproduce section of a figure (Fig. 4(c)) reported in Ref. 38: L. Berges *et al*) comparing the skyrmion mobility (black dashed line) as a function of the skyrmion diameter. It should be noted that in Ref. 38, the skyrmion mobility is modelled with a domain wall width parameter $\Delta \sim 20-25$ nm (large value due to vanishing effective anisotropy to stabilize skyrmions). Ref. 47 reported a similar domain wall width parameter (20 nm), allowing us to make a comparative analysis between the results from both references 38 and 47. As depicted in Figure RR1, it appears that the increase of skyrmion mobility starts to saturate as the skyrmion diameter increases. In this saturation regime, the mobility of skyrmions should be ~0.6 ms^{-1}/GAm^{-2}. The experimentally determined mobility of skyrmions by Quessab *et al*, stands at approximately 0.55 ms^{-1}/GAm^{-2}, considering the highest observed velocity. Therefore, we can confidently assert that Fig. RR1 accurately portrays the anticipated mobility of skyrmions, accounting for minor error margins attributed to material-specific variations, as documented in various studies.

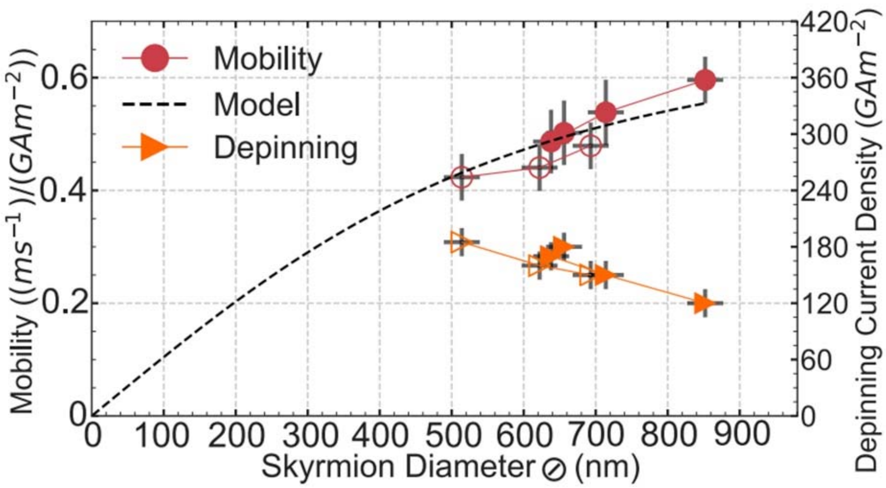


Fig. RR1: Dependence of depinning current density and mobility with respect to the skyrmion diameter.

We can also see from Fig. RR1 that the expected mobility for skyrmion diameter around 200 nm (our present work) is only ~ 0.2 ms^{-1}/GAm^{-2}. The experimental mobility in our present work is higher ~ 0.4 ms^{-1}/GAm^{-2} (from the slope of the fitting of DW flow regime) or ~ 0.5 ms^{-1}/GAm^{-2} (considering the highest skyrmion velocity at the largest current density). With this discussion this is evident that we have surpassed the expected value of mobility reported in Ref. 38 due to the fact **that our samples are more optimized for efficient SOTs combined with low pinning landscape in the films**.

Furthermore, it's important to highlight that in Ref. 38, the flow regime was theoretically modelled rather than experimentally measured, with consideration given to various values of the spin-Hall angle (θ_{SHE}). In contrast, our current study predicts the domain wall (DW) flow regime through field-induced DW dynamic measurements and subsequently overlays this data with the current-induced skyrmion motion data to demonstrate our ability to achieve the DW flow regime experimentally. We also emphasize that the first author of the present paper is a co-author of Ref. 38, where they explicitly mentioned that the apparent flow regime observed in their work is associated with larger skyrmion sizes.

Building upon the preceding discussion, it becomes evident that driving larger skyrmions (e.g., those with diameters of 500-900 nm as cited in Ref. 38, and 1.2 µm as referenced in Ref. 47) presents considerably fewer challenges compared to the task of propelling smaller skyrmions (with diameters less than 200 nm) as we demonstrated in this study.

2. Stability of the small skyrmions in the multilayers:

Maintaining the stability of skyrmions within single ferromagnetic layers often proves challenging due to various undesirable factors such as fluctuations and spontaneous creation/annihilation [4-6]. Similar challenges are highlighted in Ref. 47, where the deviation in skyrmion velocity at high current densities is attributed to skyrmion deformation and non-rigidity within the system. Further, the skyrmions reported in Ref. 47 were stable only for a few minutes, as explicitly written by the authors in their manuscript. To enhance the stability of skyrmions, one viable approach involves increasing the number of layer repetitions within multilayer structures, albeit at the expense of larger dipolar coupling, that might result into larger skyrmion sizes. Consequently, as previously discussed (as well as in the introduction of the manuscript), it becomes imperative to work with systems characterized by reduced magnetization to facilitate the stabilization of smaller skyrmions. This necessitates a comprehensive optimization process, where a myriad of material parameters must be carefully balanced in multilayer configurations to achieve both stability and smaller skyrmion sizes.

Furthermore, driving skyrmions within multilayers presents additional challenges compared to single-layer systems. This difficulty arises because it requires the optimization of spin-orbit torques for each layer while concurrently managing an overall reduction in the pinning landscape. This intricate balance is a critical obstacle preventing up to now the attainment of high skyrmion velocities in multi-layered systems so far.

3. Importance of interlayer antiferromagnetic coupling instead of ferrimagnetic alloys:

In the present work, we work with a multilayer structure of Co|Tb instead of the usual choice of CoTb alloys. The multilayers structure offers several key advantages over their alloy counterparts.

(a) The anisotropy in the CoTb based alloys are usually extremely large (even far from compensation), which makes the stabilization of skyrmions challenging even in the presence of strong interfacial DMI.
(b) The properties of the individual layers can be utilized for different aspects to facilitate large skyrmion velocity (e.g. Co|Tb interface for antiferromagnetic coupling, Pt|Co interface for improving PMA, iDMI, SOTs, Co|Al interface for giant Rashba type spin torques, etc.).
(c) It has been previously demonstrated that the synthetic ferrimagnetic multilayers are more robust than the alloys for single-pulse laser induced all optical switching [7, 8, several articles from the groups of Bert Koopmans and Reinoud Lavrijsen in Netherlands]. Recently, L. Avilés-Félix, *et al* [9], have demonstrated that Tb|Co nanolayers based MTJ can be optically controlled via helicity-dependent single shot switching. The direction of such works can lead to advancement of writing frequencies towards the THz scale.

Additionally, such multilayer stacks can handle a more robust temperature range (up to ~200°C) enabling structural integrity during the microfabrication processes. It should be noted that the present manuscript gives the first experimental demonstration (to the best of our knowledge) of hosting and driving skyrmions in such Tb|Co based multilayer synthetic ferrimagnets.

4. Robustness from ageing:

In general, one of the crucial drawbacks of using RE-TM based ferrimagnetic system is that the properties (both structural and magnetic) changes over time due to the intermixing of the RE and TM atoms. From our experience, we note that the synthetic ferrimagnetic multilayers used in this work offers a much better stability over time compared to the ferrimagnetic alloys. For example, the first measurements of the magnetization and the XMCD measurements on the samples were performed in different pieces of the same sample one and half years apart. Similarly, various dynamics measurements (notably the skyrmion motion in 1 μm and 3 μm tracks) were performed at different times. In no case, we obtained any sign of degradation/modification of the sample properties. Hence, we conclude that the synthetic ferrimagnetic multilayers are better alternatives compared to the conventionally used alloys.

5. Skyrmions propagating throughout micrometer-long length of the nanotrack:

We also note that the skyrmions in our work propagate through the whole length of the tracks from one side of the electrode to the other that is as long as 10 to 20 μm depending on the devices. This achievement is to our opinion crucial for any skyrmion based device application where the skyrmions need to be robust enough to endure the electrical nucleation, motion, and detection. This is another key achievement of this work over the ones reported in refs. [38] and [47].

To summarize, we believe that we have presented a straightforward way of simultaneously controlling various material parameters, micromagnetic energies, SOT amplitudes to demonstrate for the first-time efficient dynamics of small skyrmions (diameter < 200 nm). Moreover, we have demonstrated that these skyrmions exhibit linear trajectories, which can be attributed to the simultaneous influence of edge repulsion and a minimized skyrmion Hall effect. Another result on which it is worth to insist is to they travel substantial distances within our experimental setup, avoiding annihilation from one electrode to the other. Therefore, we contend that our work offers substantial novelties compared to the current state-of-the-art in this field, warranting its publication in Nature Communications.

Reviewer #2 (Remarks to the Author):

This paper presents a study of skyrmion (Sk) motion in synthetic ferrimagnetic multilayer whereby, varying the layer thicknesses, the magnetic interactions between transition metal and rare earth layers can be systematically varied.
A key result is observing a linear relation between Sk velocity and current density, i.e., evidence that the Sk motion is in a flow regime. Another interesting result is that Sks appears to be repelled by the wire edges, leading to a negligible Sk transverse displacement in micron-scale wires (while such a displacement is observed in wider wires). Interestingly, sample heating during the pulse does not appear to affect the Sk velocity results at high current densities, as seen in earlier studies at high current densities (e.g., their ref. 47).
The authors' claims are well supported by their results, and the manuscript is well written, with the key Sk physics well presented. In addition, the authors provide sufficient details to reproduce their studies. Of further interest would be time-resolved imaging studies to determine the reproducibility of the Sk displacements and resolve the Sk Hall angle and relaxation after pulses due to edge repulsion.
In sum, the manuscript reports significant results that merit publication in Nature Communication.

Reply: We thank the reviewer for his/her critical reading and very positive analysis of the manuscript, in particular pointed out the significance and the novelty of our results. We agree with the referee that the time-resolved imaging of the skyrmions will be useful to check the skyrmion repulsion and displacement. However, this is beyond the scope of the present work and will be considered as a future prospective study.

Reviewer #3 (Remarks to the Author):

In this work by Mallick et al., the authors have studied the transition from creep motion to the flow regime of skyrmions in synthetic ferrimagnets. Their results are obtained by increasing the amplitude of SOT and AFM coupling between the Co and Tb layers, by using the magnetic force microscope imaging technique. By comparing with the field-induced DW motion, the authors have proposed three different dynamical regimes of current-induced skyrmion motion: thermally activated motion, depinning motion and flow motion. Furthermore, they have achieved a relatively high-speed motion of ferrimagnetic skyrmion with a reduced skyrmion Hall effect. Through comparing the most recent developments in the field, I failed to appreciate the novelty and the robustness of their results. The present manuscript is not suitable for publication in journals such as Nature Com. I would encourage authors to revise the manuscript based on the following comments and to add additional information, after which this manuscript may be reconsidered.

Reply: We thank the referee for the insightful comments on the manuscript. Similarly, to the responses we have done to Reviewer #1, we disagree with the lack of novelty of our work that would impede a publication in Nature Communication. Please refer to the somehow similar remarks expressed by the first reviewer to follow the detailed response we prepared on the novelty and importance of our results. In the following, we answer to your specific queries in details.

(1) I noticed the value of interfacial DMI is being volume averaged. I am curious to see its comparison with similar multilayers.

Reply: The interfacial DMI has been measured on the single layer samples (as described in details in the supplementary information) and not on the multilayers. The main reason for this is that performing BLS measurements on the multi-layered samples lead to additional peaks in the spectra making the analysis of the data more ambiguous. This is the reason why BLS analysis is usually performed in the heterostructure with no repetitions (single layer) and assumed to be remaining constant throughout the thickness of the heterostructure due to similar layer environment. Nevertheless, in the following, we summarize a comparison of interfacial DMI obtained in various other multilayer systems that we have performed in a recent publication by F. Ajejas, *et al* [10].

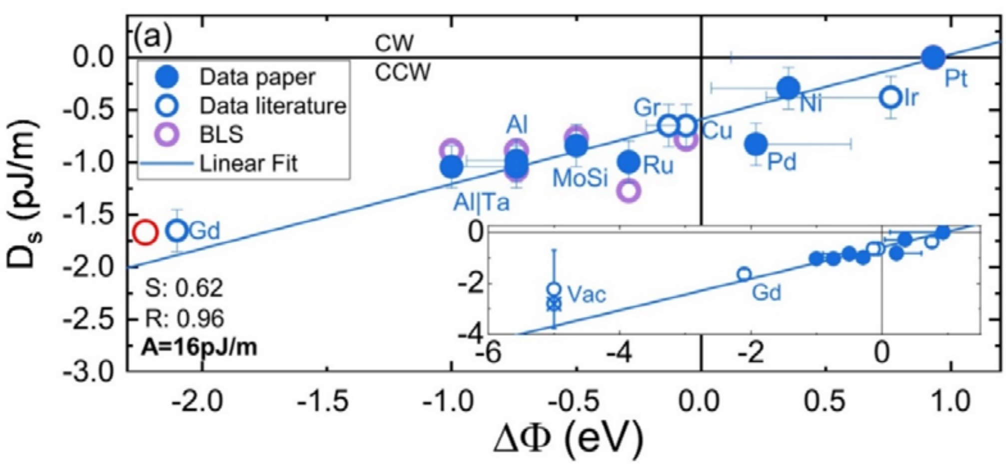


Fig. RR2: Dependence of effective DMI of Pt/Co/M interfaces with work function (ΔΦ) difference between Co and M. Figure reproduced from ref. [10]. The point corresponding to our Pt/Co/Tb system is shown in red on the left side.

The comparison of DMI of the present work with similar multilayers has been shown in Fig. RR2 in which we reproduced one figure of Ref [10]. It shows the dependence of effective DMI of Pt|Co|M (M: any metal) interfaces with work function (ΔΦ) difference between Co and M. The solid blue circles in the figure are experimental data points measured by our group whereas the open purple circles are from other literature. It should be noted that the data presented in [10] were measured in different types of samples i.e. single-layer as well as multilayers with two different techniques i.e. BLS like in the present work but also through asymmetric DW expansion for the case of multilayers. Importantly, both techniques result in similar DMI values for a given system. The open red circle (bottom left) that we have added here, corresponds to the DMI value presented in the present work for Pt|Co|Tb based multilayers. We see that the experimental value of DMI for Pt|Co|Tb agrees rather well with the prediction of [10] considering the work-function difference between Co and Tb from literature [11].

(2) It is not completely clear to this referee about the advantage(s) of studying the present material system. Did authors study the damping parameters of their multilayers? It is known that Gd-based multilayers exhibit a relatively low damping parameters, as compared with the Tb based multilayers.

Reply: We thank the reviewer for this crucial remark. As already mentioned in the manuscript, the damping parameters of our multilayers have been determined by performing BLS measurements in DE geometry. The values of the damping parameters are 0.13 ± 0.01, 0.25 ± 0.06, and 0.43 ± 0.06 for the multilayers with Tb thicknesses of 0.25, 0.5, and 1.0 nm, respectively. We agree with the referee that the damping parameter of Gd is lower than that of Tb layers. However, from our previous experience and also reports in literature [12], we believe that the damping does not play a crucial role in the motion of the skyrmions in such thin film ferrimagnets (but not in ferromagnetic systems!). On the contrary, damping plays a determinant role in reducing the topological deflection of the skyrmions, since the skyrmion Hall angle is inversely proportional to the damping of the system. Nevertheless, besides the damping, we would like to point out that there are several other advantages of Tb based multilayers in comparison to Gd based multilayers and some of them are listed below.

1. Tb based multilayers provide larger contribution to magnetic moment (both spin and orbital) in comparison to the Gd based ones due to the unfilled 4f orbital of the Tb. This leads to formation of stronger antiferromagnetic coupling with the Co sub-layers (lattices) providing more thermal stability to the textures. The only drawback so far (more profound in the alloys), was the large anisotropy of Co|Tb based systems which did not allow the stabilization of spontaneous textures due to extremely large domain wall energy. However, with the trick of using multilayer, by forming interfacial antiferromagnetic coupling, we show that it is possible to stabilize skyrmions in such Co|Tb based multilayers using both magnetic and electrical excitations.
2. It has been recently demonstrated that the orbital torque appearing from the Tb layer, is larger than that from the Gd layer [13, 14] leading to a more effective current induced magnetization switching. It has been also demonstrated that the interplay between the spin and orbital current can be strongly modified using such RE metals where they play crucial role in being orbital to spin current converters. In this paper, we obtain an enhanced $\theta_{SHE}^{eff} = 0.085$ for Pt|Co|Tb|Al based multilayers in compared to $\theta_{SHE}^{eff} = 0.06$ obtained in Pt|Co|Al based multilayers.
3. There have been several reports over last few years that reported laser induced single shot optical switching in Co|RE based multilayers with Tb being more robust in compared to Gd ones. Moreover, Gd-based materials exhibit an inherent softness, which poses challenges when attempting to reduce the size of Gd-based devices to scales below the micrometre level. In contrast, Terbium (Tb) demonstrates the capability to sustain domains with dimensions smaller than 50 nm [9].

(3) Some of the TEM results are not very relevant. I can see clearly the layered resolved structures. Can TEM results be otherwise helpful in establishing the picture of interfacial chiral magnetism? Can they be helpful in establishing the Tb-thickness dependent DMI strength? Is Tb contributing additional source of chiral magnetism?

Reply: Sorry but maybe we do not properly understand the first part of the comment. However, trying to address it, we would like to emphasize that we consider the TEM images are crucial for comprehending the multilayer structure and, consequently, gaining insights into the AFM coupling between the Co and Tb layers. From Fig. 1(b-g) of the main manuscript, it can be concluded from the TEM images and the corresponding EDX mapping that, below 1 monolayer of Tb, theoretically estimated to be around 0.35 – 0.4 nm, a significant intermixing occurs between the Tb and Pt layers. This intermixing directly impacts the AFM coupling and magnetization behaviour of the samples. Coherently, we observe gradual change in M_S and H_K in the samples up to $t_{Tb} = 0.5$ nm, beyond which the changes become pronounced. Furthermore, the TEM images provide valuable information regarding the approximate monolayer thickness of Tb. Thanks to the TEM images, we can confidently assert that the transition occurs from less than a monolayer thickness to approximately three layers of Tb across the entire thickness range of the samples discussed in this study.

From Fig. RR2 presented before, we conclude that Tb|Co interface leads to the occurrence of one of the highest iDMI reported so far when put on top of Pt|Co based systems. Hence, it is clear that Tb contributes to additional source of chiral magnetism. We also note that, as already mentioned in the supplementary information, for larger Tb thicknesses ($t_{Tb} > 0.25$ nm), the anisotropy of the samples was too large to be measured using BLS. Hence, we do not have any further information on the Tb thickness dependent DMI strength. Since the DMI is interfacial in nature, we do not expect the iDMI to vary a lot over the whole thickness range.

(4) I believe that the magnetometry data should be presented in the main text? Is there any substantial difference between them? It is clear that incorporating of Tb, introducing antiferromagnetic coupling that reduces the net magnetization. How is this helpful in reducing the size of skyrmion? What is the role of repetition number? What is the role of Al layer?

Reply: We thank the referee for this remark. We agree that the magnetometry data might bring additional insights to the paper. However due to restriction in pages in the main manuscript, we have decided to add in the revised version a new figure in the supplementary information file (see new figure S1). In Fig. RR3, we present the hysteresis loops for all the samples measured using AGFM in perpendicular geometry. It can be seen that the sample magnetization decreases continuously with increasing Tb thickness confirming the increase of antiferromagnetic coupling (relative increase of Tb moment opposite to Co ones) between Co and Tb. The thickness of Co (1.3 nm) in the multilayers has been chosen in such a way that the effective anisotropy of the multilayers is small enough and hence facilitates skyrmion stabilization. However, it can be seen from the hysteresis loops that the anisotropy of the systems starts increasing (near 100% remanence) beyond $t_{Tb} > 0.6$ nm. The consequence of such increase in anisotropy can be also observed during the current induced dynamics measurements in which beyond $t_{Tb} > 0.6$ nm, we have succeeded to stabilize only chiral DWs in the system instead of skyrmions.

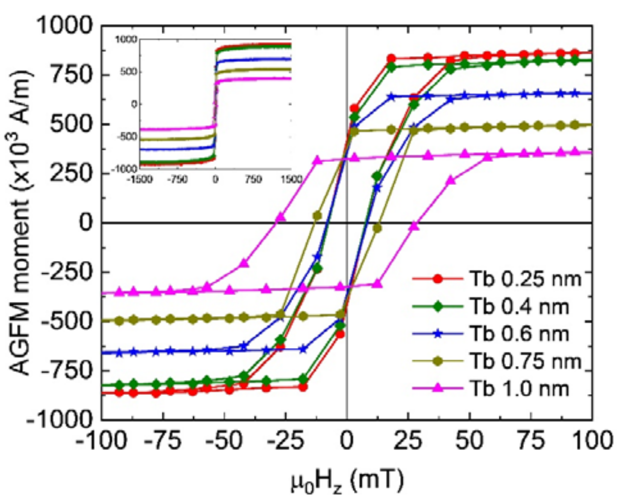


Fig. RR3: Hysteresis loops measured using AGFM in perpendicular mode for all the multilayer samples with various Tb thicknesses.

<u>Role of Tb thickness in reducing the size of skyrmions:</u> We note that the size of the skyrmions determine through MFM imaging and analysis does not vary much with the Tb thicknesses since they are mostly stabilized by applying wide electrical current pulses (20 ns) in the nanotracks near the magnetic saturation of the films. Additionally, we want to stress that the skyrmion size were maintained with such small diameter by adjusting the external field incrementally during the skyrmion motion experiment to counter further skyrmion nucleation at larger current densities, as it is explained in the main manuscript. Please note that an important effort has been done in this study to develop home-made

MFM tips in order to reduce to a minimum the perturbation of the tips while keeping a large enough signal. However, for the smallest skyrmions (around 175 nm +/- 15 nm) that we observe, the error bars on the estimated skyrmion diameter are relatively large and thus might impede to determine the exact impact of Tb thickness on skyrmion size.

Role of repetition number: As often done in literature, we have used magnetic multilayers to provide large thermal stability in the heterostructure. Co and Tb layers situated next to each other are directly exchanged coupled with antiferromagnetic coupling, whereas the Co|Tb layers from successive multilayers are dipolarly coupled. This additional dipolar interaction further reduces the domain wall energy providing additional and wider range of stability to the skyrmions. The effect of such additive chiral interactions within the multilayers are reported in ref. [6]. It is also reported that by varying the numbers of multilayers, and thus by tuning the chiral interactions along with dipolar coupling, it is possible to tune ratio, positions, as well as the chirality of the Neél DWs [4]. Therefore, following a thorough investigation and consideration of all potential scenarios, we have opted for a repetition count of 5. This choice allows us to effectively fine-tune all parameters, facilitating swift skyrmion motion.

Role of Al layer: The role of Al layer is crucial for the dynamics of the skyrmions and the discussion is already detailed in the paragraph *Optimization of SOT for skyrmion dynamics* in the manuscript. In the multilayers without any Al layer, the effective spin-Hall angles are found to be in the range of 1.9 – 3.4%, for the samples with $t_{Tb} = 0.25$, and 0.5 nm. The effective spin-Hall angle is rather small due to partial cancellation of the spin current and the related spin torques (generated by spin Hall effect) arising from the top and bottom Pt layers, as detailed in our recent publication by S. Krishnia, *et al* [15]. Application of electrical current in such low spin-Hall angle multilayers led to parasitic skyrmion nucleation within the track because the current density required for nucleation is lower than the one for skyrmion motion. This is why the Al layer has been introduced here to enhance the effective spin-orbit torques following our recent observation reported in ref. [15] in which we found that 3 nm Al layer was enough to increase the damping like torque by 30%. The introduction of a 3 nm thick Al layer between the interfaces of Tb and Pt resulted in a notable enhancement of the effective spin-Hall angle in the samples, reaching values of 8.1% to 8.6% for samples with $t_{Tb} = 0.25$, and 0.5 nm. This additional Al layer in the torque-optimized heterostructure significantly improved the control over skyrmion nucleation and motion.

(5) The interlayers distance is approximately 6 nm. I am curious to see the magnetization of Co/Tb of each layer is still coupled, considering a reduced magnetization. Can authors comment on this aspect?

Reply: It should be noted that the phase signal of the skyrmions during MFM scan was same during field scanning and current induced motion experiments while using homemade tips with same magnetic coatings. Additionally, we can see from fig. RR3 (now added in supplementary information as Fig. S1) that there is no step present in the hysteresis loops in any of the samples to suggest that there is some decoupling between the layers. Further, even at the largest current densities, the skyrmions retain their shape and contrast (no partial contrast indicative of skyrmions belonging to part of the sample thickness was observed). From these observations, we are convinced that the layers are coupled in the multilayers.

(6) The MFM images shown in Figure 2 is not labeled. In line 150, "dotted wine line in Fig. 4" should be change to "Fig. 3". The authors should thoroughly review the entire manuscript to avoid such errors.

Reply: We thank the referee for pointing out these mistakes. They have been now amended throughout the revised manuscript.

(7) From Figure 4, it is clear that most of these skyrmions are still in the creep motion regime, by revealing an unequal displacement. It can also be seen from the very large error bar.

Reply: We agree with the referee that the skyrmions are in creep regime up to $J = 3.5 \times 10^{11}$ Am^{-2}. However, beyond this current density, they go through the depinning transition in all the samples (characterized by well-defined negative curvature). In the best optimized sample (i.e. $t_{Tb} = 0.6$ nm), the skyrmions move in the DW flow regime as predicted by the field induced dynamics measurements.

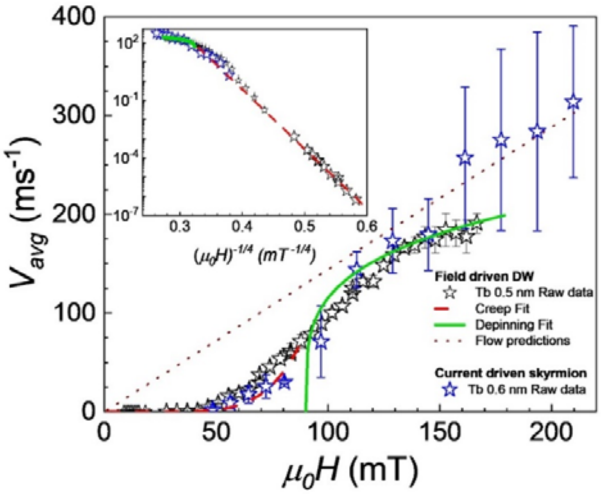
The large error bars in velocities at high current density is associated to lesser number of skyrmions present in the track due to large distance of propagation between successive pulses (larger than 2 μm).

As a consequence, only a few events inside the track can be recorded for each current pulse values, hence reducing the statistics. However, we also note that there are still scarce pinning sites in the samples which leads to unequal displacement of the skyrmions. We have discussed this effect in details in the manuscript.

(8) It is also known that Tb element is very active, which means the nanostructure can be easily oxidized.

Reply: We agree with the referee that Tb element is very active and prone to oxidation. This is the reason why all the samples studied in this work are capped with 2 nm of Pt. Additionally, there is 3 nm Al layers on top of the Tb layer in the torque optimized samples. Hence, there are 5 nm of metallic layers on top of Tb protecting it from oxidation.
We note that, the first measurements of the magnetization and the XMCD measurements on the samples were performed in different pieces of the same sample 1.5 years apart. No signature of any oxidization of Tb has been found. Similarly, various dynamics measurements (notably the skyrmion motion in 1 μm and 3 μm tracks) were performed in different times. In no such cases we obtained any sign of degradation/modification of the sample properties.

(9) The green solid line indicating depinning motion in the inset of Figure 3 is very unclear, the authors should make it more prominent. Additionally, the solid blue stars in Figure 3 are used to indicate the current-induced skyrmion motion, but there are no current coordinates provided.



Reply: We thank the referee for bringing this to our attention. We have prepared a new figure (as shown above) by showing the depinning region with more prominence. Further, we have now added the conversion factor between the current and field induced motion in the caption of the figure.

(10) By comparing the Figures 4(a)-(d), it can be observed that some of the skyrmions annihilate during motion, and the diameter of the skyrmion is also varying. Please explain this phenomenon.

Reply: We thank the referee for bringing this important aspect into discussion. The event of skyrmion annihilation or the variation in the diameter of the skyrmions during the experiment is a direct consequence of the magnetic tip induced perturbation of MFM measurements during the topography scans. It should be noted that, the samples that we have studied in this work are amorphous in nature and hence intrinsically offer lower pinning for the magnetic textures. Hence, the textures are seldom perturbed by the stray field arising from the magnetic tips. In order to reduce the perturbation, we have used home-made tips of magnetic bilayers CoFeB capped with Al by varying the layer thicknesses (reducing CoFeB and increasing Al). However, this can be done up-to a certain threshold because beyond that we start losing the phase signal during the measurements (this is one of the limitations of using MFM). Hence, even after rigorous tip optimization, we still have finite tip induced perturbation (specifically during the topography scan where the distance between the tip and the sample is minimum). Fig. RR4 (a) and (b) shows the MFM phase signal of the multilayer sample with $t_{Tb} = 0.25$

nm at demagnetized state and under a finite perpendicular external field, respectively, being measured with a commercially available soft magnetic tip (Co 7 nm capped with Pt 5 nm). It can be observed from the figure that the textures are significantly perturbed along the scan axis (x axis) due to the tip induced perturbation. Following this, we prepared homemade tips with the bilayer thickness selected in such a way that the tip induced perturbation is minimum (at the same time ensuring that there is finite phase signal to be measured). The consequence can be observed in the Fig. RR4 (c) and (d) where we obtain significantly less perturbation in comparison to before with the new tips. However, we note that, although the tip induced perturbation is reduced, it is not completely removed (bottom right skyrmion in (d)). This is the reason why in some places during the scans, we have annihilation as well as change in diameter of the magnetic skyrmions. To further account for this effect, we have not considered such events (skyrmions) during the calculation of the velocity.

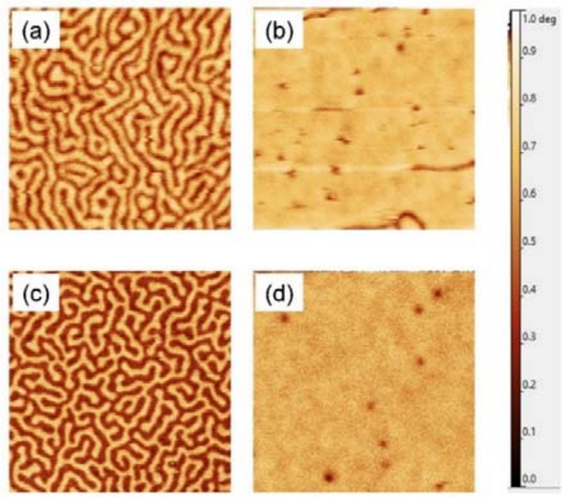


Fig. RR4: MFM phase signal for the multilayer sample with $t_{Tb} = 0.25$ nm, measured using magnetic tip with coating of Co (7 nm)/Pt (5 nm) at (a) demagnetized state, and (b) at an external field of ~ 30 mT. (c) and (d) show the same images at demagnetized state and under an external perpendicular field on the same sample, however being measured with a magnetic tip with coating of CoFeB (5 nm)/Al (40 nm). All the images are 5 µm × 5 µm.

The distribution of the skyrmion diameter in a series of 20 MFM images taken one after another after application of 10 ns wide successive current pulses with $J = 3.5 \times 10^{11}$ Am^{-2} is presented in Fig. RR5. The average skyrmion diameter from this set of measurement is ~164.7 nm (which is within the reported value of 160-190 nm). Additionally, we want to stress that the skyrmion size were maintained with such small diameter by changing the external field incrementally during the skyrmion motion experiment to counter further skyrmion nucleation at larger current densities.

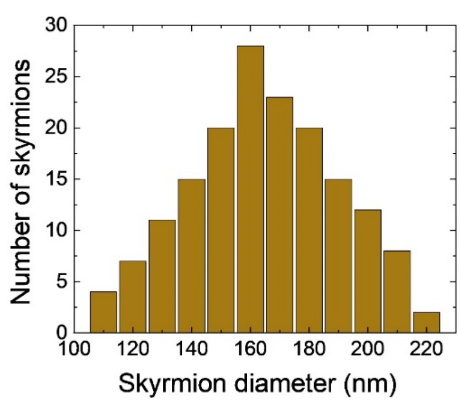


Fig. RR5: Variation of skyrmion diameter in a series of 20 MFM images while applying 10 ns wide current pulses of $J = 3.5 \times 10^{11}$ Am^{-2} after each single image in the multilayer sample with $t_{Tb} =$ 0.6 nm.

(11) The authors did not consider the effect of current-induced Joule heating on skyrmion nucleation and motion, please comments on it.

Reply: We thank the referee for this important remark. The proper estimation of Joule heating is indeed necessary in ferrimagnetic systems since the change in temperature can move the system closer or far from the magnetic and/or angular compensation. However, it is important to note (remind) that, in the thickness range of Tb studied in this work, we are far away from magnetic compensation (compensation should appear at low temperature and all the measurements are performed at room temperature). Hence, in case of the Joule heating during the current pulses, the sample will be heated up and driven even more further away from the magnetic compensation (and not closer to it). Hence, the effect of Joule heating on the dynamics of the skyrmions in our samples should not be significant.

In order to estimate the impact of the heat dissipation, we have performed the comparison of the skyrmion velocity in the same sample ($t_{Tb} = 0.6$ nm), however in two tracks with different widths (1 and 3 μm). The power dissipation in a track after applying a current pulse can be expressed as $P = J^2\rho ltw$; where ρ, l, t, w are the resistivity of the materials, length, thickness, and width of the tracks, respectively. Hence, this dissipation should vary with the track width. However, from Fig. RR6 (b) it can be seen that the skyrmion velocity is more or less similar (within the error bars) in both the tracks. This confirms the minimal effect of dissipation in the motion of the skyrmions as well as in their rigidity [16]. It should be noted that we could not extract the velocity at larger current densities in the 3 μm wide track since the heating induced by the current pulses leads to additional nucleation of skyrmions. This is why, the skyrmions are thermally nucleated by applying rather wide current pulses ($J = 3.5 \times 10^{11}$ Am^{-2}, 20 ns or more) and then are driven with smaller pulses (10 ns) to reduce the potential impact of Joule heating. We have added new discussion regarding this in the *Methods* section of the revised manuscript.

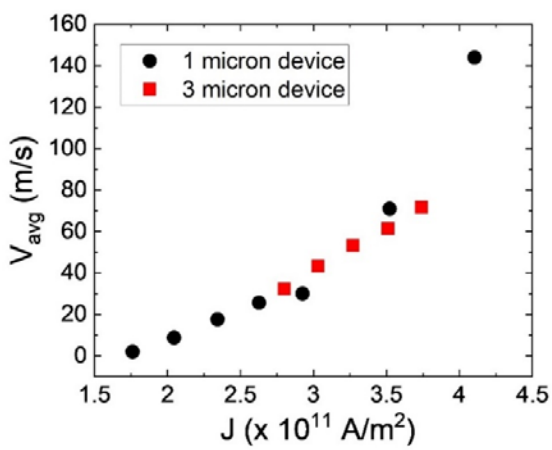


Fig. RR6: Comparison of average skyrmion velocity in two tracks of different widths (1 and 3 μm) in the multilayer sample with $t_{Tb} = 0.6$ nm while being driven by 10 ns wide current pulses with different current densities.

(12) In line 277, the authors mention "….1 μm narrow track without any transverse deflection….", however, there appears to be transverse deflection of the skyrmions in Figures 4(a)-(d). The authors should quantitatively investigate the time-dependent trajectories of skyrmions.

Reply: The time resolved measurements of the skyrmions is beyond the scope of this present work and will be considered as a future research direction. Although the pump-probe imaging would provide quantitative information on the time-dependent trajectories of the skyrmions, it will limit the statistics of data in the entire velocity range from creep to flow regime.

However, we want to stress that, in the present study, we focus on the equilibrium positions of the skyrmions in the MFM images following the application of current pulses. We note that the "apparent" straight-line motion of the skyrmions are due the combination of the repulsions from the track edges combined with the reduced skyrmion Hall effect (discussed in the next point). Importantly we note that, the combined force is enough to maintain the skyrmions close to the centre of the track and the skyrmions are robust enough to retain their stability even in the presence of such strong repulsive forces.

(13) On a separate note, the so-called suppression of skyrmion Hall angle in the present synthetic ferrimagnet is most likely due to the edge repulsive force. I am looking forward to see the motion of skyrmion in a large device with a larger width. There, authors could observe free motion of skyrmion. I think a comparison of that results in those in the literature is fair.

Reply: We believe that the properties of skyrmion Hall effect in our multilayer was already discussed in details both in the main manuscript and the supplementary information. We fully agree with the referee, as mentioned on the response to the previous comment, that the straight-line motion of the skyrmions occurs partly because of the edge repulsive force and not only due to the suppression of the skyrmion Hall angle. In order to confirm this, as discussed in the manuscript (in details in supplementary information), we have measured the skyrmion deflection in a wider track (3 µm wide). We obtain similar skyrmion velocity with a topological deflection of ~ 29° in the sample with $t_{Tb} = 0.6$ nm. We also show that the experimental obtained value of skyrmion Hall angle matches with the theoretical calculations by using various materials parameters measured using AGFM, BLS, and PPMS. Further, we show a comparison of skyrmion Hall angle in our system in comparison to the existing literature in Fig. S4 of the supplementary information. We have also provided movies of skyrmion motion in 1 µm and 3 µm wide tracks as supplementary video files.
Hence, we conclude that the linear motion of the skyrmions is associated to the combined effect of edge repulsion and reduced skyrmion Hall effect. We further note that the skyrmions retain their stability at the largest current density even in the presence of such repulsive forces due to their stability provided by the sizable effective anisotropy and the multilayers.

(14) The theoretical calculation cannot be justified. Authors are using synthetic ferrimagnet, the usage of exchange stiffness should be very different with that of the Co.

Reply: We agree with the referee that the theoretical calculations are not 100% accurate since we assume an exchange stiffness of Co. However, we want to bring to the attention of the referee that the exchange stiffness of Co has been measured previously to be around 14-17 pJ/m in our work involving other ferromagnetic multilayers [10]. Further, it is known that the exchange stiffness of any RE alone at room temperature is 0. Hence, when the RE (Tb) is coupled with Co, this should only decrease the effective exchange stiffness of Co. This is the reason we think that the considered exchange stiffness of 10 pJ/m in this work is a reasonable approximation. Unfortunately, a direct measurement allowing us to determine the exact value of exchange stiffness in our sample could not be done due to the large anisotropy of the samples and field limitation of the experimental set-up. However, we also want to stress that a slight modification of the exchange stiffness shall not alter the calculated value of skyrmion Hall angle a lot since $\theta_{SHA} = \arctan(\frac{1}{\alpha}\frac{2\Delta}{R})$ whereas $\Delta = \sqrt{\frac{A}{K_{eff}}}$. The change in Δ due to A, will be within the error margin of the calculated skyrmion radius (R). The value of θ_{SHA} is mainly dependent on the value of the damping parameter which have been extracted with greater care.

(15) The skyrmion velocity is smaller than the reported value in CoGd, I suggest authors carefully rephrase the usage of their language.

Reply: We thank the referee for this remark. Please consult the response of the first query by Reviewer #1 to follow the detailed discussion on why the present result is superior and brings novelty in compared to the ones reported in CoGd, or in CoFeGd alloys. In order to emphasis more on the novelty of the work, we have added additional text in the introduction and conclusion section of the revised manuscript.

References:

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2. H. Vakili *et al*, *J. Appl. Phys.* **130**, 070908 (2021)
3. J. C. Bellizotti Souza *et al*, *New J. Phys*. **25**, 053020 (2023)
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5. W. Jiang *et al*, *Phys. Rep*. **704**, 1 (2017)

6. A. Hrabec *et al*, *Nat. Commun.* **8**, 15765 (2017)
7. M. Beens *et al*, *Phys. Rev. B* **100**, 220409 (R) (2019)
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9. L. Avilés-Félix *et al*, *Sci. Rep*. **10**, 5211 (2020)
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11. H. B. Michaelson, J. Appl. Phys. **48**, 4729 (1977)
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13. G. Sala *et al*, *Phys. Rev. Res*. **4**, 033037 (2022)
14. M. Martini *et al*, *arXiv*:2206.01586v1
15. S. Krishnia *et al*, *Nano Lett*. **23**, 6785 (2023)
16. K. Litzius *et al, Nat. Elec.* **3**, 30 (2020)

Reviewers' Comments:

Reviewer #3:
Remarks to the Author:
Authors handwavingly addressed part of my comments and some comments from the other referees. Most of my comments did not reflect in the main text. In my opinion, this revised manuscript did not meet the publishing criteria of Nature Communications. And the very similar impression is shared by the first referee. Below are some general comments.
1: Tb is very active and prone to oxidize. Even caping with Pt and Al are not helpful. Especially for the patterned microstripe devices. Of course, this is based on our our experience. I suggest authors present the corresponding xas data to show if their samples are insensitive to oxidization.
2: The TEM data is again, lacking of necessary explanation. In fact, I did not see authors make necessary efforts in addressing my early comment. From this referee's perspective, the data shown in Figure 1 is just a bunch of data and could neither support either the formation of synthetic ferrimagnetic skyrmion, nor support the reduced skyrmion Hall angle.
3: Response to Comment 1 is in appropriate. I am not quite sure if the unit of IDMI is actually correct. It should be mJ/m^2.
4: Again, I am not convinced by the advantage of their multilayers. I beleive some of their responses, especially the comment related to oxidization is misleading.
5: Authors should specifically mention the method of calculating skyrmion velocity. Should we consider the inertia effect?
6: With respect to the suppression of skyrmion Hall effect (comment 12), I fully disagress with their responses. There are very obvious transverse deflection. Thus, I am not fully convinced by the advantage of this synthetic ferrimagnetic system. There are not helpful in suppression the skyrmion Hall angle. In addition, I could somewhat identify the stochastic motion of skyrmions with uneven displacement. This indicates the claim of flow motion is not being fully justified.

In short, the conclusion of "flow motion" is not being fully justified. The response and explanation to the advantage of their material system is inadequate and somewhat misleading. I strongly believe this material system is extremely sensitive to oxidize, especially in these patterned structures. I do not support its publication.

We sincerely thank the referee for bringing forward pertinent concerns that have been helpful to improve our manuscript. We are pleased to notice that we have now successfully addressed the primary concern raised in the previous round of review, specifically regarding the novelty of our work. Nevertheless, we acknowledge the existence of two prominent concerns, namely, the oxidation of Tb and the suppression of the skyrmion Hall effect. In the following sections, we respond to these concerns in a structured and pointwise manner:

Reviewer #3 (Remarks to the Author):

Authors handwavingly addressed part of my comments and some comments from the other referees. Most of my comments did not reflect in the main text. In my opinion, this revised manuscript did not meet the publishing criteria of Nature Communications. And the very similar impression is shared by the first referee. Below are some general comments.

1: Tb is very active and prone to oxidize. Even caping with Pt and Al are not helpful. Especially for the patterned microstripe devices. Of course, this is based on our our experience. I suggest authors present the corresponding xas data to show if their samples are insensitive to oxidization.

Reply: We agree that Tb is known to be an active element, but we disagree with the statement that any capping layer is useless to impede oxidization of Tb. In order to answer more factually to this comment, we have prepared new series of samples and thoroughly performed new XPS and XAS measurements on them. Nevertheless, we begin by stressing the fact that the main conclusion of our study on the large skyrmion mobility in this material system as claimed in Fig. 3 and Fig. 4 remains unaltered irrespective of the oxidation state of Tb.

In Fig. RR1, we present the XPS measurements on samples that we have grown freshly. It should be noted that since XPS is sensitive to only a few nm from the surface of the film, the samples presented in the paper did not allow us to probe Tb due to 5 nm of metallic capping layers (3 nm Al + 2 nm Pt). In Fig. RR1 (a)–(f) we show the in-situ and ex-situ XPS measurements for uncapped Tb 100 nm and Tb 100 nm films capped with Al layer with various thicknesses, respectively. We opted for a thick Tb layer in our XPS measurements to ensure ample signal strength. Given the limited penetration depth of the low-energy XPS beam (typically 3-5 nm), we anticipated probing only the surface of the films. However, we also conducted XPS spectra for Tb thicknesses as low as 1 nm, yielding comparable results albeit with lower intensity. For reference, we have marked a dotted black line at 147 eV which corresponds to the metallic peak associated to Tb 4*d*. In Fig. RR1 (a) (blue spectra), we observe the distinctive sharp peak of metallic Tb 4*d* at 147 eV for Tb 100 nm thin film (uncapped), measured in-situ in UHV immediately after the growth in the sputtering chamber. Additionally, a minor proportion of O 1*s* is detected. Notably, the Tb 4*d* peak flattens upon storing the sample in UHV for a day (yellow spectra in Fig. RR1 (b)), indicating propensity of Tb for oxidation even under UHV conditions. Subsequent exposure to ambient atmosphere results in complete oxidation of Tb, eliminating any presence of metallic Tb (Green spectra in Fig. RR1 (c)). Subsequently, we examined a new sample with the same Tb thickness, this time capped with 1.4 nm of Al in-situ in UHV (red spectra in Fig. RR1 (d)). Interestingly, the metallic Tb peak is heightened compared to the uncapped Tb sample. This suggests that the 1.4 nm Al capping layer results into a partial protection against Tb oxidation during and immediately after deposition, as confirmed by the negligible Tb oxidation within the UHV chamber during this process. However, XPS measurement on the same sample after exposure to ambient conditions demonstrates Tb oxidation once again (dark yellow spectra in Fig. RR1 (e)), indicating that although the 1.4 nm Al layer shields Tb within the UHV chamber, it fails to do so in ambient conditions. Finally, we examine another sample, this time with the Tb layer capped with a thick 3 nm Al, post-exposure to ambient conditions using XPS (violet spectra in Fig. RR1 (f)). In this case, we observe metallic Tb peaks corresponding to Tb 4*d*. Consequently, we deduce that while Tb exhibits a tendency for oxidation, this can be largely minimized by capping it with 3 nm Al. Importantly, we note that the samples discussed in the paper also feature an additional 2 nm Pt capping layer to safeguard the top Al layer from oxidation in ambient conditions as we demonstrated in our recent publication S. Krishna *et al* (Nano Letters 23, 6785 (2023)).

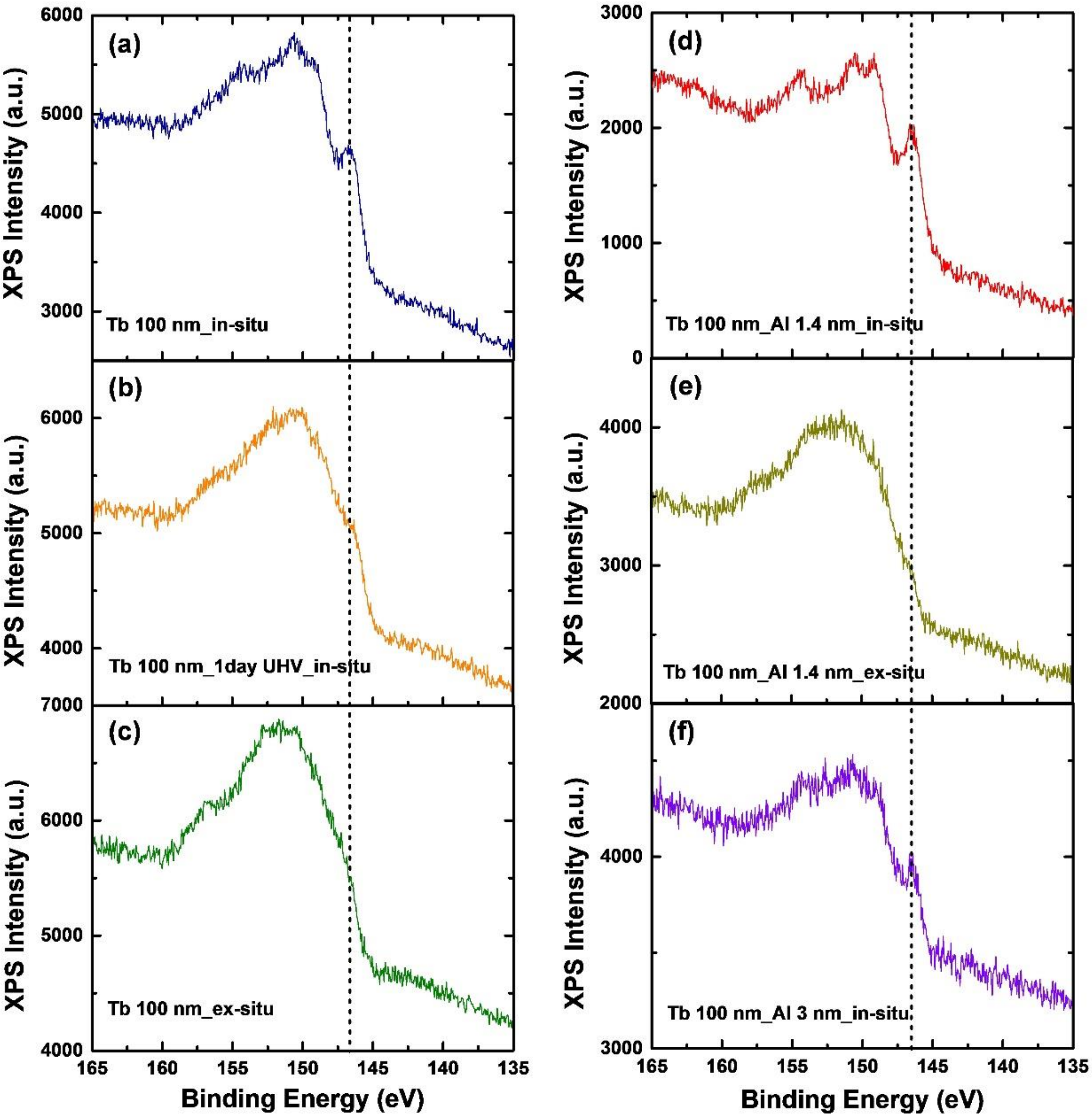

Fig. RR1: XPS measurements of the following: (a) Tb 100 nm (no capping layer) measured in-situ after deposition, (b) Tb 100 nm (no capping layer) measured in-situ after 1 day in UHV after deposition, (c) Tb 100 nm (no capping layer) measured ex-situ, (d) Tb 100 nm (Al 1.4nm capping layer) measured in-situ, (e) Tb 100 nm (Al 1.4nm capping layer) measured ex-situ, (f) Tb 100 nm (Al 3nm capping layer) measured ex-situ. The dotted line at ~147 eV corresponds to peak associated to metallic Tb 4d.

Additionally, we have been able to perform a few XMCD experiments in house measurements based on urgent access to beamline in synchrotron SOLEIL in order to further answer the reviewer's comment. This study is only partial and the analysis is not complete but we would like to share the preliminary results with the editor and the reviewers.

The XMCD measurements have been performed for 2 multilayer samples from the main manuscript with Tb thickness of $t_{Tb} =$ 0.5 nm and 1.0 nm. In Fig. RR2 we show the XMCD and corresponding XAS spectra for (a) $t_{Tb} =$ 0.5 nm at Tb *M* edge, (b) $t_{Tb} =$ 0.5 nm at Co *L* edge, and (c) $t_{Tb} =$ 1.0 nm at Tb *M* edge. We obtain comparable XMCD spectra of the Tb *M* edge, consistent with findings from previous studies [G.v.d Laan *et al, Phys. Rev. B* **59**, 8835 (1999); M. Taguchi *et al, Phys. Rev. B* **68**, 104408 (2003); T. Ueno *et al, J. Phys. D: Appl. Phys.* **49**, 205001 (2016); D. H. Suzuki *et al, Phys. Rev. B* **107**, 134430 (2023)]. The distinct peak in the Tb XMCD spectra for both samples (denoted by *) substantiates the presence of metallic Tb within the multilayers [G.v.d Laan *et al, Phys. Rev. B* **59**, 8835 (1999); M. Taguchi *et al, Phys. Rev. B* **68**, 104408 (2003); T. Ueno *et al, J. Phys. D: Appl. Phys.* **49**, 205001 (2016)]. Further scrutiny of the Co XMCD spectra reveals no indication of Co oxidation. Although quantifying the precise oxygen content within Tb in our samples necessitates dedicated beamline access, a task exceeding the scope of our current work, we can offer a qualitative estimate. Drawing upon the insights presented in Suzuki et al., we tentatively assess approximately 10% oxidation of Tb during the thin film growth within the UHV chamber [D. H. Suzuki *et al, Phys. Rev. B* **107**, 134430 (2023)]. In summary, aligning with the results from our XPS and XMCD measurements, we affirm the presence of metallic Tb in our samples. Furthermore, we acknowledge the finite oxidation of Tb during thin film growth, albeit occurring at a minute proportion. In the revised version, we have now included the aforementioned discussion in the supplementary information.

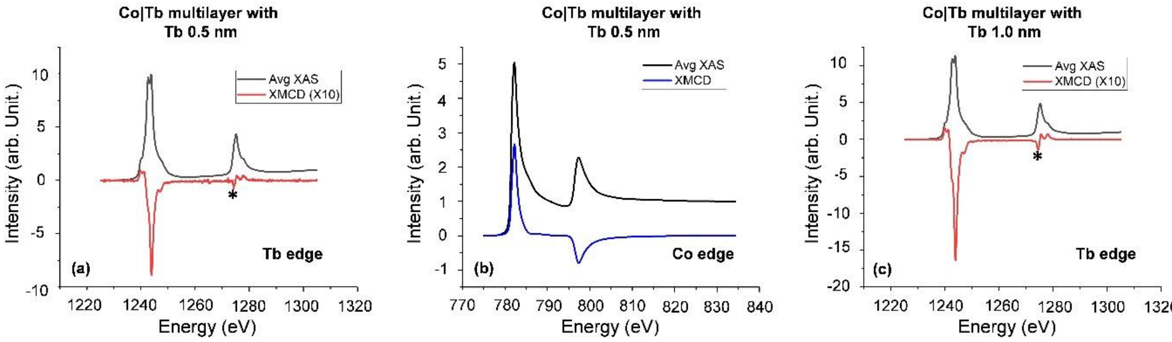

Fig. RR2: XAS and XMCD spectra of Co|Tb multilayer samples with (a) t_{Tb} = 0.5 nm at Tb M edge, (b) t_{Tb} = 0.5 nm at Co L edge, and (c) t_{Tb} = 1.0 nm at Tb M edge. The '' mark in the XMCD spectra represent the peak associated to metallic Tb.*

Additionally, it is crucial to emphasize that all samples analysed in this study were prepared in a consistent manner, following identical deposition conditions. Any potential variations occurring within the sputtering chamber during the growth process should remain uniform across all samples. Moreover, we conducted both field-induced DW motion and current-induced skyrmion motion experiments on distinct pieces of the same samples. Remarkably, even after 18-24 months, these samples demonstrated consistent properties. This longevity underscores the absence of significant alterations in the samples over time, reaffirming the effectiveness of the 5 nm thick metallic capping layer.

2: The TEM data is again, lacking of necessary explanation. In fact, I did not see authors make necessary efforts in addressing my early comment. From this referee's perspective, the data shown in Figure 1 is just a bunch of data and could neither support either the formation of synthetic ferrimagnetic skyrmion, nor support the reduced skyrmion Hall angle.

Reply: We acknowledge the referee's concerns regarding the interpretation of Fig. 1 in the manuscript. It is imperative to clarify that the primary objective of Fig. 1 is not to substantiate either the existence of synthetic ferrimagnetic skyrmions or the assertion of a reduced skyrmion Hall angle. Instead, its purpose is to delineate the sample structure and illustrate the formation of synthetic ferrimagnetic architecture contingent upon the thickness of Tb.

In Fig. 1, we present the TEM cross-section of three distinct samples with varying thicknesses of Tb: t_{Tb} = 0.25, 0.5, and 1.0 nm, while the monolayer thickness of Tb is ~0.35 nm. Consequently, within this thickness range, the transition occurs from less than a monolayer of Tb to a few layers of Tb in the samples. This transition is validated by the EDX spectra, wherein discernible peaks associated with Tb emerge solely for samples with $t_{Tb} > 0.5$ nm, indicating the formation of a uniform Tb layer.

Our message elaborated from Fig.1 is that this elucidation is pivotal for comprehending the subsequent magnetization and anisotropy behaviors, wherein the antiferromagnetic coupling between Co and Tb layers assumes significance for $t_{Tb} > 0.25$ nm. Therefore, we respectfully dissent with the referee's assertion regarding the perceived insignificance of Fig. 1. From our standpoint, this figure holds paramount importance as it establishes the groundwork for understanding the AFM coupling, which constitutes the essence of the manuscript.

3: Response to Comment 1 is in appropriate. I am not quite sure if the unit of IDMI is actually correct. It should be mJ/m^2.

Reply: In the manuscript, we have mentioned the surface contribution of iDMI, $D_s = -1.62 \pm 0.20$ pJ/m. Since the thickness of the Co+Tb in the same sample is 1.25 nm, hence the volume average of the iDMI is $D = \frac{D_s}{t} = -1.28 \pm 0.16$ mJ/m². We have now provided this information in the revised manuscript.

4: Again, I am not convinced by the advantage of their multilayers. I believe some of their responses, especially the comment related to oxidization is misleading.

Reply: Since more than a decade and the first observation of skyrmions stabilized at room temperature by interfacial DM interaction, it is admitted that the use of magnetic multilayers presents several notable advantages compared to single films. Firstly, they facilitate enhanced dipolar coupling, thereby easing the stabilization of skyrmions by reducing domain wall energy. Secondly, they contribute to greater thermal stability for skyrmions, as they are coupled throughout the sample's thickness. Thirdly, they yield increased signal strength during MFM measurements. These advantages also hold in case of ferrimagnetically coupled layers.

However, due to their coupling across the entire sample thickness, careful optimization of each layer repetition is essential for swift electrical manipulation. To achieve this, a Pt thickness of 3 nm has been chosen for each layer, ensuring efficient introduction of PMA, iDMI, and sufficient spin current injection into the magnetic layer. Additionally, maintaining a 3 nm top layer of Al serves to prevent electron backflow and simultaneously enhance damping-like torque as we demonstrated recently in S. Krishnia *et al, Nano Letters* **23**, 6785 (2023). In essence, the selection of multilayers and the repetition of layers has been meticulously executed to both stabilize skyrmions and manipulate them efficiently.

In the first comment of the referee, we have now addressed the concerns regarding the oxidization of Tb in our samples.

5: Authors should specifically mention the method of calculating skyrmion velocity. Should we consider the inertia effect?

Reply: As outlined in the manuscript, we determine the velocity of skyrmions by tracking their displacement within the nanotrack over successive current pulses. This involves measuring the distance travelled by individual skyrmions and averaging this over numerous data points spanning 5-20 pulses. The resultant average propagation distance per pulse is then divided by the FWHM time of the applied current pulses to obtain the skyrmion velocity. Notably, these measurements are conducted under conditions for which skyrmions are in equilibrium, thus disregarding any possible effects of inertia. In this context, we have now added a brief discussion elaborating the method to calculate the skyrmion velocity in the revised manuscript.

Exploring skyrmion motion in the transient regime would necessitate time-resolved synchrotron-based measurements, which is beyond our capabilities and beyond the scope of our current investigation. Furthermore, such an approach would lack the statistical robustness provided by MFM measurements, which offer comprehensive data analysis capabilities.

6a: With respect to the suppression of skyrmion Hall effect (comment 12), I fully disagress with their responses. There are very obvious transverse deflection. Thus, I am not fully convinced by the advantage of this synthetic ferrimagnetic system. There are not helpful in suppression the skyrmion Hall angle.

Reply: We have no difficulties to acknowledge here again the substantial influence exerted by the device boundary in our observation of an almost straight skyrmion motion. As already explicitly stated in the previous version of our manuscript (Fig. S5 of supplementary information), we contend that the overarching straight-line motion of skyrmions in our sample is a result of the combined impact of both a reduced skyrmion Hall effect and the edge repulsion inherent in the device, a point also emphasized by the Editor. However, it is imperative to underscore that relying solely on edge repulsion does not facilitate the efficient motion of skyrmions from one side of the contact to the opposite end at such elevated velocities. To substantiate this claim, we conducted a comparative analysis of skyrmion velocity and motion in our sample against a scenario where no Tb layer was present. As an experimental protocol, the skyrmion Hall angle has been always experimentally estimated in wider tracks (3 or 5 μm) since the narrow track (~1 μm) introduces strong edge repulsion leading to wrong estimation of the angles.

In Pt|Co|Al multilayers (ferromagnetic multilayers without any Tb layer), the skyrmions are deflected at an angle of ~45° when driven in a 5 μm wide wire. It should be noted that, up to a threshold current density ~2.2×10^{11}A/m^2 (skyrmion velocity ~ 15 ms^{-1}), the skyrmions deflect towards one edge of the track and henceforth follows the track to move in the longitudinal direction. However, beyond this threshold current density, the skyrmions move towards the edge of the track and then annihilate. Hence, in such systems, the skyrmions cannot be driven the whole length of the wire at larger velocity without getting annihilated (see videos V1-V3 to track the movement of the skyrmions). The calculated skyrmion Hall angle ($\theta_{SHA} = \arctan(\frac{1}{\alpha}\frac{2\Delta}{R})$) in this sample is ~ 57°, where α, Δ, R are experimentally measured.

Nevertheless, in the best optimized sample reported in this work (Pt|Co|Tb|Al multilayer with Tb = 0.6 nm), we calculate the skyrmion Hall angle to be $\theta_{SHA} =$ 27.5°. This proves that there is a clear expected reduction in skyrmion Hall angle due to increase in α which is a consequence of the formation of antiferromagnetic coupling between the Co and Tb layers as explained in the manuscript. The experimentally observed skyrmion Hall angle in this sample in a 3 μm wide wire is ~29°, which matches rather well with the calculated value.

Further, we reduce the width of the track to 1 μm to confine the skyrmions more profoundly. In this scenario, the reduction in skyrmion Hall angle helps the skyrmions to move the whole length of the narrow 1 μm wide device aided by the edge repulsion. In a nutshell, the reduced skyrmion Hall angle assists the skyrmion to reduce the gyrotropic transverse force due to SOT which balances with the force appearing from the edge of the track to keep the skyrmions stable even at largest current densities. The following table shows the calculated and the experimentally observed skyrmion Hall angles for various samples:

Sample	**Damping (α)**	**Skyr. radius (R)**	**Calculated $\theta_{SHA} = \arctan(\frac{1}{\alpha}\frac{2\Delta}{R})$**	**Measured θ_{SHA}**
Tb 0 nm	0.05	>100 nm	56.6°	45°
Tb 0.25 nm	0.13	~100 nm	45.1°	Not calculated
Tb 0.6 nm	0.25	~70-90 nm	27.5°	29°
Tb 1 nm	>0.43	No skyrmion	18.1°	Not applicable

For comprehensive understanding, we present 3 separate videos (from new measurements) related to the motion of skyrmions in Pt|Co|Al multilayers (ferromagnetic multilayers without any Tb layer). The description of the videos is the following:

V1: Skyrmion motion in a 5 μm wide wire at $J = 1.9 \times 10^{11}$A/m^2: The skyrmions reach the edge of the track and move along the edge with a velocity ~ 15 ms^{-1}.

V2: Skyrmion motion in a 5 μm wide wire at $J = 2.2 \times 10^{11}$A/m^2: The skyrmions reach the edge of the track and annihilate.

V3: Skyrmion motion in a 1.5 µm wide wire at $J = 2.2 \times 10^{11}$ A/m^2: The skyrmions reach the edge of the track, face a defect and annihilate.

The preceding discussion clarifies that achieving a straight-line motion of skyrmions necessitates the combined actions of edge repulsion and a reduced skyrmion Hall effect. Devoid of edge repulsion, our system would exhibit a finite skyrmion Hall angle, as we experimentally evidence in wider tracks. Conversely, in the absence of a reduced skyrmion Hall angle, relying solely on edge repulsion would prove insufficient in guiding skyrmions at high velocities, ultimately resulting in their annihilation. Consequently, we posit that these two forces complement each other, culminating in the efficient and unhindered motion of skyrmions in our system. In this context, we have now added a short discussion in the revised manuscript as well as in the supplementary information.

6b: In addition, I could somewhat identify the stochastic motion of skyrmions with uneven displacement. This indicates the claim of flow motion is not being fully justified.

Reply: In the previous round of response to the referees, we extensively addressed this aspect. The irregular displacement of skyrmions is not attributable to pinning effects within the sample; rather, it stems directly from skyrmion hopping behavior.

Additionally, we have quantified the pinning length (ξ) within our samples through field-induced domain wall (DW) velocity measurements, following the methodology outlined by Jeudy *et al.* [*Phys. Rev. B* **98**, 054406 (2018)]. The calculated ξ values across the entire thickness range of Tb fall within the range of 4 – 7 nm (data are not shown here). Contrastingly, the displacement of individual skyrmions between successive current pulses spans from hundreds of nanometres to a few microns which is orders of magnitude larger than the pinning length. Consequently, it can be inferred that skyrmions do not become immobilized by pinning sites.

Rather, we believe that their motion might appear as stochastic or influenced by defects mostly because of inter-skyrmion interactions or due to strong repulsive forces from the sides of the track. As previously mentioned, at larger current densities, our synthetic ferrimagnetic skyrmions traverse significant distances, thereby limiting the number of motion events to 2-3 before traversing the entire length (~10 µm) of the nanotrack. This reduction in the number of data points diminishes statistical robustness, consequently increasing the standard deviation of individual data points that we include in the graph.

However, it is important to emphasize that this study represents the first experimental study of skyrmion motion in the three different mobility regimes i.e. creep, depinning and flow motion. We believe that the experimental results shown in Fig. 4 (together with the fits in the depinning regime) are conclusive enough to support our conclusion that, at least in the sample with Tb = 0.6 nm, the current-induced motion at large current densities correspond to a flow regime.

In short, the conclusion of "flow motion" is not being fully justified. The response and explanation to the advantage of their material system is inadequate and somewhat misleading. I strongly believe this material system is extremely sensitive to oxidize, especially in these patterned structures. I do not support its publication.

Reply: We have thoroughly addressed all the inquiries mentioned in our previous discussion. Furthermore, in response to the two principal concerns raised by the referee and underscored by the editor, we conducted supplementary measurements. Additionally, we have made revisions to the relevant sections of the manuscript, which are now highlighted in yellow. We believe that these modifications effectively demonstrate the significance and timeliness of our work, thereby satisfying both the referee and the editor, and warranting its publication in Nature Communications.