

Supplementary Information for Lifetime of coexisting sub-10 nm zero-field skyrmions and antiskyrmions

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SUPPLEMENTARY NOTE 1 | PARAMETER SPACE FOR FIELD POLARISED PHASE OF FCC- AND HCP-RH/CO/IR(111).

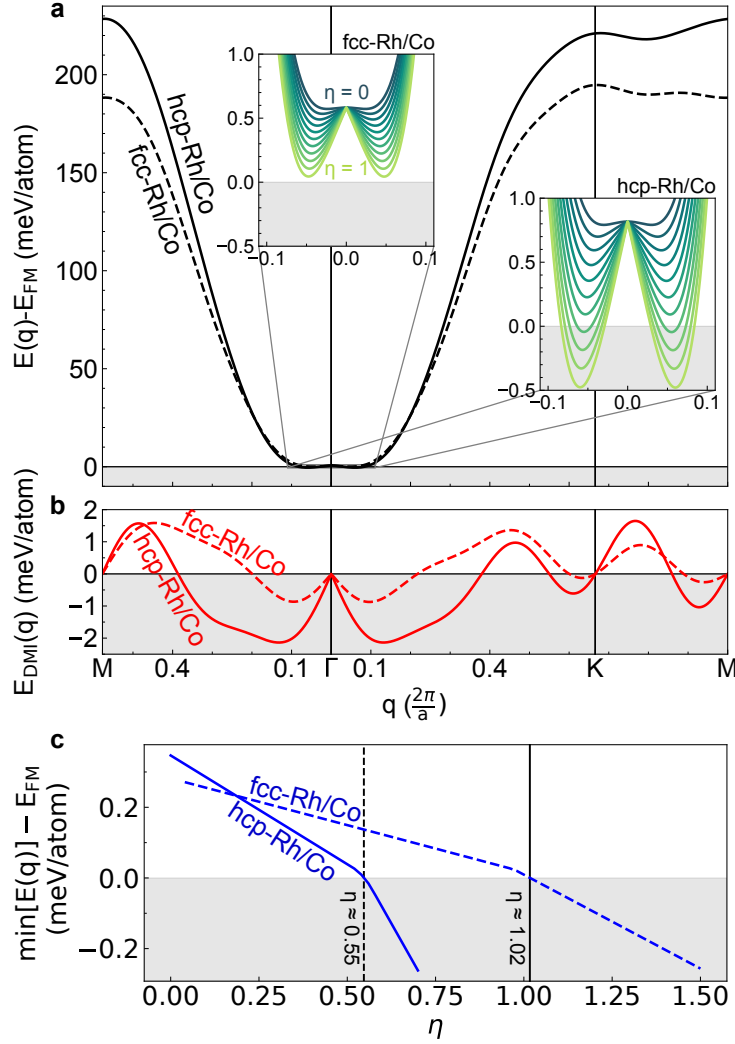
In this section we explain the choice of the parameter space of η , which scales the strength of the DMI in the spin model, for both fcc- and hcp-Rh/Co/Ir(111). Supplementary Fig. 1a displays the energy dispersion $E(\mathbf{q})$ of cycloidal spin spirals along the $\overline{\Gamma\text{M}}$ - and $\overline{\Gamma\text{KM}}$ -direction of the two-dimensional Brillouin zone as obtained from DFT for both stackings [1]. A cycloidal spin spiral is characterized by the wave vector $\mathbf{q}$ along which it propagates and the direction of a magnetic moment of an atom at lattice site $\mathbf{r}_n$ is given by $\mathbf{m}(\mathbf{r}_n) = \mathbf{e}_z \cos(\mathbf{q} \cdot \mathbf{r}_n) - \mathbf{e}_q \sin(\mathbf{q} \cdot \mathbf{r}_n)$, where $\mathbf{e}_z$ is the unit vector normal to the film, and $\mathbf{e}_q = \mathbf{q}/q$ is the unit vector along $\mathbf{q}$. The energy dispersion of such a spin spiral can be obtained analytically using the atomistic spin model given by Eq. (1) of the main text with the interaction constants calculated via DFT.

At the $\overline{\Gamma}$ point, we find the ferromagnetic state which is the ground state of the fcc-stacking of Rh, but not for the hcp-stacking (c.f. insets in Supplementary Fig. 1a for $\eta = 1$). At the $\overline{\text{M}}$ point, we obtain the row-wise antiferromagnetic state and at the $\overline{\text{K}}$ point, the Néel state with angles of 120° between adjacent magnetic moments which are both much higher in energy than the ferromagnetic state. This results in a large nearest-neighbor exchange constant $J_1 > 0$. The flat energy dispersion in the vicinity of the $\overline{\Gamma}$ point, i.e. for $|\mathbf{q}| < 0.1 \times 2\pi/a$, indicates the strong frustration of exchange interactions present in Rh/Co/Ir(111).

The insets of Supplementary Fig. 1a show zooms of the region in the vicinity of the $\overline{\Gamma}$ point including the effect of spin-orbit coupling, i.e. the DMI and the magnetocrystalline anisotropy energy. The latter results in a rigid shift of the energy dispersion of spin spirals by $K/2$ with respect to the ferromagnetic state located at zero energy. The DMI contributions to the energy dispersion for the systems as obtained by DFT ($\eta = 1$) are shown separately in Supplementary Fig. 1b. From the first inset of Supplementary Fig. 1a showing the influence of the DMI on the dispersion minimum in the fcc-stacking it can be seen, that for $\eta \rightarrow 1$ the gap between the lowest spin spiral state and the FM ground state becomes very narrow with increasing DMI. Particularly the gap is given for $\eta = 1$ by a very small value of $E(q) - E_{\text{FM}} \approx 4.3 \cdot 10^{-2}$ meV/atom. For hcp-Rh/Co/Ir(111), on the other hand, we find a spin spiral ground state at $\eta = 1$. In order to determine the exact range for η , where the field polarised state gives the ground state, we generate 5 spin spiral states with wave

vectors around the minimum $\mathbf{q}_0(\eta) = \arg \min_{\mathbf{q}}[E(\mathbf{q})]$ and numerically minimise these states with respect to their energy. The choice of $\mathbf{q}$ is thereby restricted to $|\mathbf{q}|a/2\pi = p/N_{\parallel}$ by the periodic boundary conditions of our finite simulation box. Here $N_{\parallel} = 100$ is the number of atoms in our simulation box along the direction of $\mathbf{q}$ and $p \in \mathbb{Z}$ is an arbitrary integer value. The energy minima obtained in this way, shown in Supplementary Fig. 1c, differ from the minima of the analytical curves obtained for homogeneous spin spirals, because spin spirals tend to deform in the presence of magnetocrystalline anisotropy. For both stackings of Rh on Co/Ir(111) the crossing of the minimum spin spiral energy and the FM energy marks the boundary of the field polarised phase. As seen in Supplementary Fig. 1c we therefore chose to investigate fcc-Rh/Co/Ir(111) in the range $\eta \in [0, 1]$ and showed the lifetime of skyrmions in hcp-Rh/Co/Ir(111) for $\eta = 0.5$ in Fig. 2 of the main text.

Note, that experimentally isolated magnetic skyrmions have been observed in hcp-Rh/Co/Ir(111) at zero magnetic field, i.e. in the ferromagnetic ground state, using scanning tunneling microscopy [1, 2] suggesting that the DMI strength is smaller than that obtained from our DFT calculations.



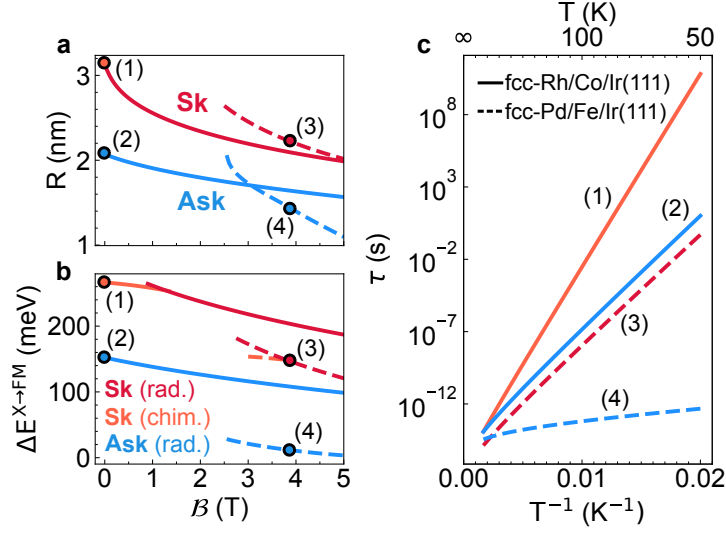
Supplementary Figure 1 | Energy dispersion of spin spirals in hcp- and

fcc-Rh/Co/Ir(111). **a** Energy dispersions $E(\mathbf{q})$ of homogenous spin spirals obtained based on the DFT parameterized atomistic spin model including the effect of spin-orbit coupling with respect to the energy of the ferromagnetic state, E_{FM} , along $\overline{\Gamma\text{M}}$ - and $\overline{\Gamma\text{KM}}$ -direction of the two-dimensional Brillouin zone. The insets show a zoom around the $\overline{\Gamma}$ -point, illustrating the effect of DMI on the energy minimum in both systems by showing dispersions for 11 values of $\eta \in [0, 1]$. **b** DMI contribution to the energy dispersions, corresponding to $\eta = 1$. **c** Energy minima of $E(q)$ for $q \neq 0$ with respect to the energy of the FM state. For each η a set of spin spirals around the dispersion minimum is minimised in energy and the lowest in energy is determined. The vertical lines denote the systems transition from FM groundstate to spin spiral groundstate.

SUPPLEMENTARY NOTE 2 | SK/ASK LIFETIME IN PD/FE/IR(111) VS. RH/CO/IR(111).

In the main text, we have compared the lifetimes of skyrmions and antiskyrmions in Rh/Co/Ir(111) to those obtained in Pd/Fe/Ir(111). Since Pd/Fe/Ir(111) exhibits a spin spiral ground state a finite magnetic field has to be applied in order to obtain isolated skyrmions [3–8]. We chose a value of $\mathcal{B} = 3.9$ T, slightly above the transition to the field-polarized phase of Pd/Fe/Ir(111), since at this value the energy barrier of skyrmions is similar to that of antiskyrmions in fcc-Rh/Co/Ir(111) at zero magnetic field (see Supplementary Fig. 2b, points denoted by (2) and (3)). Note, that at these field values the radii of antiskyrmions in fcc-Rh/Co/Ir(111) are smaller than those of skyrmions in Pd/Fe/Ir(111) (see Supplementary Fig. 2a, points denoted by (2) and (3)).

However, the energy barriers protecting antiskyrmions from collapsing into the ferromagnetic ground state is larger than that of skyrmions in Pd/Fe/Ir(111) at this field value (see Supplementary Fig. 2c). Skyrmions in fcc-Rh/Co/Ir(111) exhibit much larger lifetimes due to the higher energy barriers resulting from the effect of the DMI (cf. Supplementary Fig. 2b and Fig. 1 of the main text). We have also calculated the lifetimes of antiskyrmion in Pd/Fe/Ir(111) here. As seen in Supplementary Fig. 2c, these lifetimes are extremely small.

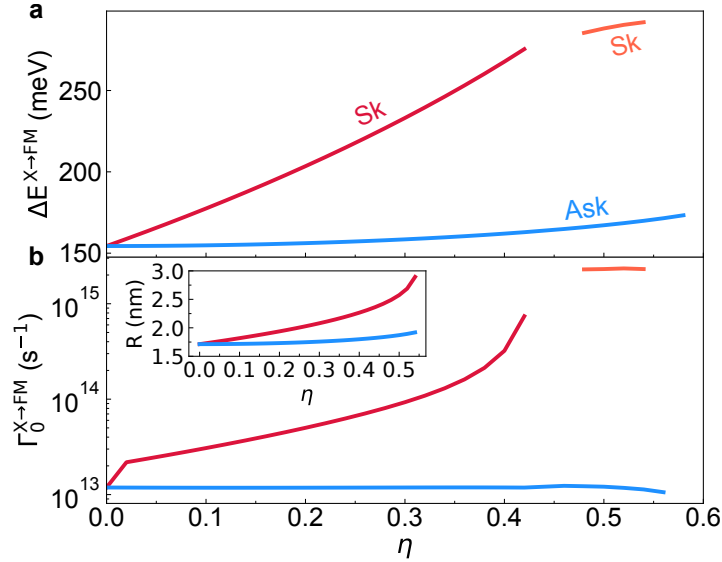


Supplementary Figure 2 | Skyrmions and antiskyrmions in Rh/Co/Ir(111)

vs. Pd/Fe/Ir(111). **a** Radii of isolated skyrmions (Sk, red) and antiskyrmions (Ask, blue) as a function of the applied magnetic field in Pd/Fe/Ir(111) (dashed lines) and in fcc-Rh/Co/Ir(111) (solid lines). Numbers in brackets indicate the values at the specific magnetic fields at which the lifetimes shown in panel **c** have been calculated. **b** Energy barriers for annihilation of skyrmions and antiskyrmions in Pd/Fe/Ir(111) and in fcc-Rh/Co/Ir(111). The colors and lines are chosen as in panel **a**. In addition, the collapse of the skyrmion via the chimera transition is indicated in orange, while the collapse via the radial transition is given in red. **c** Calculated lifetimes of skyrmions and antiskyrmions vs. temperature (same curves as Fig. 2 of the main text). Numbers in brackets correspond to those in **a** and **b** and denote the values at which the lifetimes have been obtained.

SUPPLEMENTARY NOTE 3 | LIFETIME OF SK/ASK IN HCP-RH/CO/IR(111).

In Fig. 2 of the main text we presented the lifetime of skyrmions and antiskyrmions in hcp-Rh/Co/Ir(111) but with reduced DMI by 50% ($\eta = 0.5$). Here we show the respective energy barriers (Supplementary Fig. 3a) and pre-exponential factors for $T = 1$ K (Supplementary Fig. 3b). For both skyrmions and antiskyrmions these quantities are again computed versus DMI strength η in the range discussed in Supplementary Note 1. We find, that the energy barriers and the pre-exponential factors for skyrmion and antiskyrmion annihilation show qualitatively the same behavior in hcp-Rh/Co/Ir(111) upon varying the DMI strength as in fcc-Rh/Co/Ir(111). The inset of Supplementary Fig. 3b additionally shows the radius of skyrmions and antiskyrmions, analogously to Fig. 3c in the main text.



Supplementary Figure 3 | Energy barrier and pre-exponential factor for skyrmion/antiskyrmion annihilation in hcp-Rh/Co/Ir(111). **a** Energy barrier for annihilation of skyrmions (red) and antiskyrmions (blue). Energy barriers corresponding to the chimera transition are colored in orange. **b** Pre-exponential factor for annihilation of skyrmions and antiskyrmions for $T = 1$ K. The coloring is the same as in **b**. Additionally the radius of both states is given in the inset.

SUPPLEMENTARY NOTE 4 | HARMONIC VS. ZERO MODE APPROXIMATION FOR HELICITY OF SKYRMIONS.

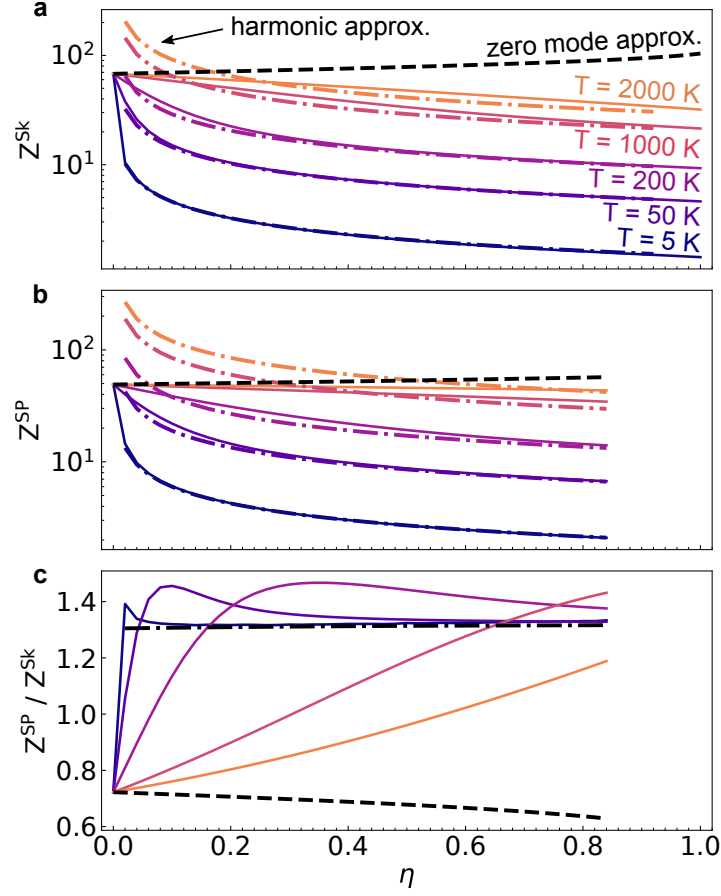
For the pre-exponential factors of the radial annihilation/nucleation of skyrmions presented in the main text the partition function for the skyrmion helicity have been computed numerically. Here we explain, why neither the harmonic nor the zero mode approximation for this mode can be used for the full parameter space of DMI strength η and temperatures T .

In the method section of the main text we explain by means of the micromagnetic model, why for skyrmions the change in helicity can be described as a zero mode without DMI ($\eta = 0$) and as a harmonic mode otherwise ($\eta > 0$). Both approximations are found to be i.g. not continuous, as shown in Supplementary Fig. 4. We therefore compare them with numerically computed partition functions, obtained as described in the method section. Since a states Boltzmann partition function is expected to be smooth on a continuous parameterisation of the energy model, studying the transition between both approximations is interesting from a fundamental point of view. But it is also necessary in order to assure a correct treatment of this mode for the calculation of the transition rates and lifetimes presented in this paper.

In Supplementary Fig. 4a we see, that the harmonic approximation is valid for low temperatures and $\eta > 0$, as predicted in the method section of the main text. The skyrmion helicity for $\eta = 0$ can always be described as a zero mode, independent on the temperature. For rising temperatures we find, that the the deviations from the harmonic approximation increase mostly for low values of η . For $\eta \rightarrow 1$ the harmonic approximation still describes the partition functions very well. For the radial skyrmion saddle point (Supplementary Fig. 4b) we find the same effect. The ratio of partition functions of saddle point state and initial state, as it goes into the pre-exponential factor for the radial skyrmion annihilation shown in Fig. 3c in the main text, is shown in Supplementary Fig. 4c. Here not only the ratio of partition functions in the zero mode approximation are independent on temperature, but also in the harmonic approximation. Here it becomes particularly distinct that the partition functions deviation from the approximations is an interplay of DMI strength and temperature.

The reason for the kink-like rising of the skyrmions pre-exponential factor between $\eta = 0$ and $\eta = 0.02$ in Fig. 3c of the main text can be seen from Supplementary Fig. 4c. For

higher temperatures the transition between zero mode and harmonic approximation becomes smother.



Supplementary Figure 4 | Numerically obtained helicity partition functions vs. harmonic/zero mode approximation. Comparison of harmonic (dotted lines), zero mode approximation (dashed lines) for the skyrmions helicity mode partition function with direct numerical computation (solid lines). Partition functions are shown for **a** the skyrmion state and **b** the radial skyrmion saddle point state. **c** shows the ratio of both as it is relevant for the radial skyrmion annihilation rate.

Supplementary References

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