Supplementary Materials for

Ultrafast Switching to Zero Field Topological Spin Textures in Ferrimagnetic TbFeCo Films

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Supplementary Text Note S1.

In centrosymmetric thin film magnets, the total energy density of isolated skyrmions state $f(\theta, r)$ can be described by the energy density difference between the isolated skyrmions state and the saturated background. The standard micromagnetic model for a centrosymmetric thin film includes the exchange stiffness energy (*A*), intrinsic uniaxial anisotropy (*K_u*), Zeeman (*H* · *M*), and stray-field energies (*w_d*). Developing and extending the micromagnetic methods allows us to reduce the problem of axisymmetric isolated states to minimization of the one-dimensional functional $F = 2\pi \int_0^\infty f(\theta, r) r dr$ with the boundary conditions $\theta(0) = \pi, \theta(\infty) = 0$ and the energy density can be written as:

$$f(\theta, r) = A\left(\theta_r^2 + \frac{1}{r^2}\sin^2\theta\right) - w_d(\theta, r) + K_u \sin^2\theta + \mu_0 HM(1 - \cos\theta)$$

Analysis of skyrmion energy *F* under scaling transformations offers a new perspective into the physics of skyrmions. We now consider a family of functions $\vartheta(r) = \vartheta(r/\eta)$ obeying the same boundary conditions. Here $\eta > 0$ is an arbitrary constant describing uniform compressions $(0 < \eta < 1)$ or expansions $(\eta > 1)$ of profile $\vartheta(r)$, and a unique soliton solution characterized by $\eta = 1$. For rescaled functions $\vartheta(r) = \vartheta(r/\eta)$, the skyrmion energy F can be expressed as a function of η :

$$\tilde{F}(\eta) = \varepsilon_e - \varepsilon_d \eta + \varepsilon_0 \eta^2$$

The values of the exchange energy ε_e , stray-field energy ε_d , and the sum of uniaxial anisotropy energy (ε_A) and Zeeman energy (ε_Z) contributions for profile $\vartheta(r)$ are given as follows:

$$\begin{split} \varepsilon_{e} &= 2\pi A \int_{0}^{\infty} \left(\vartheta_{r}^{2} + \frac{1}{r^{2}} sin^{2} \vartheta \right) r dr \\ \varepsilon_{d} &= 2\pi \int_{0}^{\infty} w_{d}(\vartheta, r) r dr \\ \varepsilon_{0} &= \varepsilon_{A} + \varepsilon_{Z} = 2\pi \int_{0}^{\infty} [K_{u} sin^{2} \vartheta + \mu_{0} H M (1 - cos \vartheta)] r dr \end{split}$$

The unique soliton solution is characterized by $\eta = \frac{\varepsilon_d}{2(\varepsilon_A + \varepsilon_Z)} = 1$. It is apparent that the presence of perpendicular anisotropy can stabilize a soliton even without the Zeeman term in low saturation magnetization (M_s), high uniaxial anisotropy (K_u) region.





Fig. S1: L-TEM images showing the generation of topological spin textures. (a) The highdensity skyrmions with tilting angle about 20 degrees along x-axis and (b) extracted and magnified image without tilting sample characterized by randomly dark and bright dot contrast, depicting versatile topological spin textures with chirality randomly assigned, as expected in this non-chiral material. Scale bar in (a) is 5 μ m, and in (b) is 2 μ m.





Fig. S2: The effect of laser illumination on the film plane. a, The multi-walled carbon nanotubes will produce two strong (002) diffraction points. When the laser irradiates, the carbon nanotubes absorb light and generate thermal expansion, resulting in a larger crystal face spacing of (002) crystal faces and a smaller distance from the diffraction point to the center. The laser illumination produce intensity distribution on the film plane. For the laser spot with Gaussian intensity distribution, the diffraction points shrinks when the laser edge reaches the carbon nanotube, and the diffraction point shrinks the most when the center is on the carbon nanotube. Therefore, the intensity distribution of the entire laser spot can be obtained by moving the laser spot along x axis and y axis, respectively. **b**, Two different laser fluence correspond to topological spin textures generation in **c** and **d**. The scale bar is 5 μ m.

Fig. S3.



Fig. S3: Enlarged topological spin textures region under zero field after laser excitation. a, The elongated laser intensity along y-direction lead to a lager topological spin textures region. b, The inhomogeneity of material lead to the enlarged topological spin textures region. The scale bar in a-b is 5 μ m.





Fig. S4: Laser-accessible skyrmions in PtCoTa multilayers. a, Hysteresis loop of the Ta(4 nm)/[Pt(3 nm)/Co(1.85 nm)/Ta(3 nm)]₆ multilayers at room temperature with labyrinth domains under zero filed (upper) and FM state under 600 Oe perpendicular field (buttom). the red star indicates the TEM observation and laser irradiation point **b**, Electronic and lattice temperature as a function of time upon 300 fs laser pulse excitation and equilibrium temperature above the threshold fluence values T_{Sky} . The insets show the LTEM images before and after laser excitation. **c**, Lattice temperature as a function time upon 10 ns laser pulse excitation and equilibrium temperature above the threshold fluence values T_{Sky} . The insets show the LTEM images before and after laser excitation and equilibrium temperature above the threshold fluence values T_{Sky} . The insets show the LTEM images before and equilibrium temperature above the threshold fluence values T_{Sky} . The insets show the LTEM images before and equilibrium temperature above the threshold fluence values T_{Sky} . The inset show the LTEM images before and equilibrium temperature above the threshold fluence values T_{Sky} . The inset show the LTEM images before and equilibrium temperature above the threshold fluence values T_{Sky} . The inset show the LTEM images before and equilibrium temperature above the threshold fluence values T_{Sky} . The inset show the LTEM images before and after laser excitation.



Fig. S5: The in-situ thermal equilibrium domain evolution. a, The M-H curves along out-ofplane direction at different temperatures for $Tb_{15}(Fe_{75}Co_{25})_{85}$ ferrimagnetic film. The deformation of rectangular hysteresis loops at 500 K corresponds to the generation of labyrinth domain, and the Q factor decreases to around 1 at 560 K, suitable for stripe domain. The spin reorientation occurs at 580 K and a complete in-plane transformation appears above 600 K. b-e, The corresponding in-situ temperature dependent domain evolution observed by L-TEM. The scale bar in b-e is 2 µm.

Fig. S6.



Fig. S6: The magnetic structure evolution via increased femtosecond laser fluence of Tb₁₅(Fe₇₅Co₂₅)₈₅ sample. For a single femtosecond laser pulse, the laser fluence should exceed 12 mJ cm⁻² to generate skyrmions and 16.6 mJ cm⁻² to generate stripe domains in the central area of laser spot from homogenous saturated ferromagnetic state. The scale bar in (a)-(f) is 5 μ m.